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nuclear science and technology

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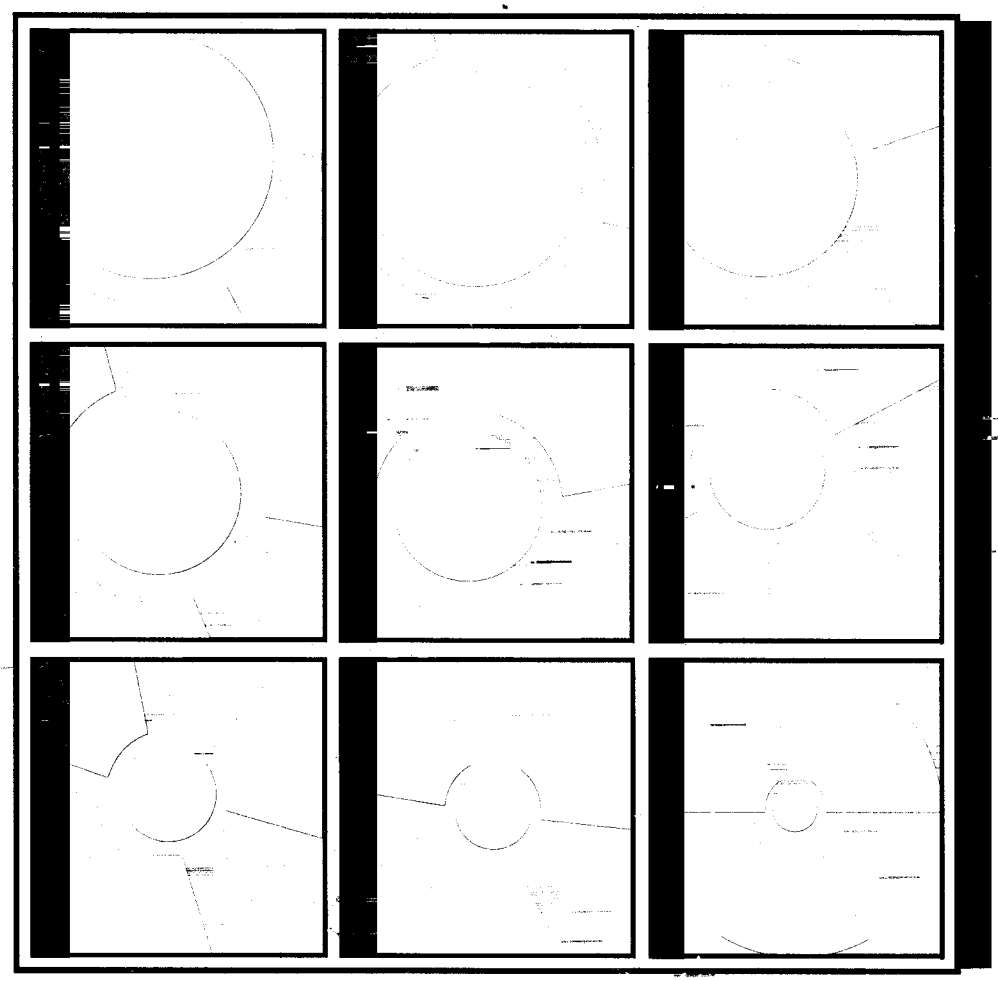

Report

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European Commission

nuclear science and technology

A present review of the thorium nuclear fuel cycles

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FOREWORD

To discourse on thorium these days when uranium is cheap and plentiful may be taken as a provocation. Besides, thorium may be considered as a "parent pauvre" compared to uranium, as it has no readily fissile component, and needs a "spark" to produce energy.

In a sense, it is a provocation indeed, which has come out from simple considerations :

- the vast amount of work done on thorium in the past thirty years must be preserved for the coming generations,

- the ideas revived by Carlo Rubbia and others, that the thorium cycle may present advantages over uranium from the point of view of waste management.

Besides, other parameters should be kept in mind :

- the finite uranium reserves in the world which can be usefully complemented by thorium : the case of India, with huge energy needs and relatively small uranium reserves, but rich in thorium, is exemplary;

- the refractory properties and neutronic stability of thorium oxide may be useful for certain types of reactors;

- by-product U-233 is a very versatile fissile material which compares favourably with U-235 or Pu-239 in many reactor applications.

In this study, we have tried to summarise in a simple way, the state of the art for thorium. It is difficult to give a true account of the huge sum of research work and accomplishments in the field, and we beg for understanding of the critical reader for any unintentional oversight.

We hope, however, to have contributed to a renewed comprehension of this interesting fuel cycle, which could play an important role in our energy future, together with uranium.

I wish to thank the European Commission for sponsoring this study, and also all who, in the past and present, have helped me in this work, as recalled further on.

ACKNOWLEDGEMENTS

I am particularly indebted for the redaction of this Report to a number of friends, former colleagues and specialists whom I wish to recall here. Those whom I may have forgotten will, I hope, forgive me :

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The reviewers of this Report, Mrs. Sue E. ION, P.D. WILSON, K. AISWORTH of BNFL, L. PATARIN of Cogema, M. SALVATORES of CEA, E. TEUCHERT formerly with KFA, Jülich, G. SCHLOSSER of Siemens, L. KOCH, TUI, who took the pains to make useful remarks, most of which have been included in this last edition. They will forgive me, I hope, if (rarely) I was not fully of their opinion.

All of those, cited in the References, who in their publications contributed to the knowledge on the thorium fuel cycle, that I felt worthwhile to quote.

Sometimes, for simplicity and clearness' sake, I have taken the liberty to reproduce excerpts, tables and charts from those, feeling that I could do so in a document which is not intended for commercial use, but as a contribution to an important scientific sector in the nuclear fuel cycle.

Thanks to my wife, this Report came into readable form.

I will appreciate further comments to this work, being conscious that there are, most probably, unwanted errors or omissions.

Michel Lung

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INTRODUCTION

Thorium was discovered by Berzelius in 1828. For a long time, production of thorium has been limited to very specific uses (special glass fabrication, gas lighting candles, special alloys) and it is so far a by-product of rare earth preparation. Its production is of some hundred tonnes per year only. It has reached about 1 000 tonnes in the 70s, to decrease thereafter due to some loss of interest.

The known reserves (RAR - Reasonably Assured Resources) are of 1-2 million tonnes (See Chapter 5). Those of uranium are of about 1,5 million tonnes of low cost and about 3 million tonnes if a price of up to $ 130/kg U could be paid. (it is now around $ 20-30/kg).

Thorium half-life (alpha decay) is $1.4\ 10^{10}$ years (U-238 : $4.5\ 10^{9}$ years).

Thorium is almost uniquely composed of the isotope Th-232. Th-230 having a half-life of $7,5\ 10^{4}$ years is present as traces.

Theoretically, the fact that thorium has a longer half-life (by a factor of 3) than Uranium would suggest that its natural occurrence is higher than that of Uranium in the earth's crust. Indeed, very large deposits have been found in India (360 000 t), in Canada, USA, Russia, China (380 000 t), Brazil. Turkey alone may have 800 000 t of thorium.

Thus thorium, besides uranium, could present a sizeable potential source for nuclear energy applications.

We shall see in the following chapters how this potential could be put to use when the need arises, and, indeed, how major experiments have shown this ability already in a number of countries.

We shall show how this important interest for thorium has dwindled for the past 20 years, except in a few countries, and we would like, in conclusion, to make some proposals for the future, based on the new technologies which found their way since the pioneering stage thirty years ago.

1. EXECUTIVE SUMMARY

Interest in thorium

During the pioneering years of nuclear energy, 1950-1970, with great enthusiasm and regardless of the costs, a large number of possible avenues for energy production with thorium have been investigated, not only in the USA or USSR, but also in Europe and, to some extent, in Asia. Thorium as a source of energy which could supplement the limited uranium reserves, has been widely investigated in those years, including in countries like Australia, Italy, Norway, Netherlands where nuclear energy has been virtually put to a rest today. For example, it is remarkable that the thorium-based Elk River (1963) and Peach Bottom (1967) reactors were started only a few years after the "founding fathers" of the two main reactor families of today, based on uranium fuel, PWR Shippingport (1957) and BWR Dresden (1960).

Now, thorium continues to generate interest in specialised circles, but more at an academic level, for long term prospects, especially in Japan. Only is thorium considered an industrial fuel for a not too distant future in India which has large thorium deposits compared to its limited uranium resources, and which wants to take advantage of some positive characteristics of thorium.

Recently, however, renewed interest was vested in thorium, not for its abundance as raw material, but because it may generate less long-lived minor actinides than with the traditional uranium fuel cycle. We will try to summarise those different findings and viewpoints and give a balanced view of the advantages and weaknesses of the thorium as a starting material for nuclear (fission) energy.

Thorium as nuclear fuel

Thorium-232, the naturally-occurring isotope, cannot alone sustain a neutronic chain reaction, which is the case for natural uranium in well-moderated, well-reflected, little neutron absorbing conditions, thanks to the presence of 0.7 % U-235 isotope.

Thorium presents good fertile material properties. It can breed U-233 in a similar way as U-238 breeds Pu-239. Uranium-233 is a particularly good fissile material on which a complete line of energy producing systems can be based, so that one can use indifferently U-235, Pu-239 or U-233 to generate fission power; each one of these isotopes, in combination with fertile isotopes U-238 or Th-232, is able to be "burnt" in specific reactor types with specific fuel cycles. These fuel cycles can be "open", or "closed", depending on the degree of self-sustainability envisaged for the fissile materials.

Conclusion :
Thorium is essentially a fertile material, similar to U-238.

Thorium abundance and reserves

Thorium is reputedly more abundant than uranium although its limited uses so far have not led to prospect it very extensively. Hence the world's reasonably assured reserves (RAR) are known to be at least as important as those of uranium, and quite probably, higher.

As it is demonstrated that, using breeder-type reactors, the known RAR uranium reserves can provide an energy source about equivalent to those of the coal's reserves, it can be assured that the thorium reserves can also double the potential available from uranium.

Most of the time, so far, thorium has been obtained as a by-product of uranium or rare-earth industry.

Preparation of thorium, similar to that of the rare-earths, entails its separation from many other (valuable) compounds, hence it is not too straightforward, necessitates many manipulations and chemical steps (possibility of corrosion by highly aggressive chemicals). The high melting points of thorium (1750°C) and thorium oxide (3300°C) give them remarkable resilient properties (stability, refractory and radiation resisting characteristics) which are paid by added difficulties of preparation compared with uranium.

Conclusion :
- Thorium is at least as abundant as uranium, quite probably more.

- Thorium and thorium oxide have excellent stability characteristics, but their preparation is somewhat more complex and costly than for uranium.

Neutronic properties of thorium and uranium-233 for energy production in fission reactors

Thorium, as a fertile material, will absorb readily incoming neutrons, preferably thermal, to produce Pa-233 which decomposes into fissile U-233. Hence thorium in a reactor needs a neutron "driver" which can be U-235, Pu-239, U-233 if some is already available. Recent proposals advocate also an external neutron source from spallation neutrons generated by some type of a high flux accelerator using high energy heavy particles. A higher energy neutron flux not only will generate U-233, but also, by n, 2n reactions, will generate some U-232. This U-232 has strong gamma emitters in its decay chain, especially Tl-208 (2.6 MeV), which creates a real problem of handling the purified U-233 for a fuel fabrication, for example. Conversely, it is a radioactive "tag" attached to U-233 which can help prevent proliferation.

Indeed, U-233 can be used as a weapon material with about 5 kg of it (not very different from plutonium). Like for U-235, a coarse bomb with U-233 is simpler to fabricate than with plutonium, because there are few spontaneous neutrons emitted unlike with plutonium. (In the latter case, the neutrons are being emitted by the higher isotopes Pu-240 and Pu-242, always present in some quantities with Pu-239, which obliges to manufacture a more intricate implosion weapon).

The neutronic fissile properties of U-233 otherwise compare very favourably with U-235 and Pu-239.

This is because it has the best ratio of neutrons emitted by fission over neutrons captured. These excellent properties will permit, not only to have a sustainable chain (i.e. the breeding of U-233 will replenish the loss by fission) in well-thermalized, low-loss heavy water reactors (Candu type), but also in epithermal light water reactors like PWRs or BWRs, or high temperature *in* gas cooled reactors with a graphite reflector, or *in* molten-salt reactors.

This "sustainability" has been a coveted property aimed at in the 1960s. Today with cheap uranium prices and excess amounts of plutonium, this is more considered as a long-term objective.

The couple thorium-oxide/uranium-oxide is particularly interesting in high temperature reactors due to the high melting-point of the oxides and the remarkable neutronic damage resistance of thorium oxide, due to its chemical and metallographic stability. Long residence times and burn-ups of almost 200 000 MWd/ton H.M. of bare or clad material have been demonstrated.

The fact that the intermediate product Pa-233 has a relatively long half-life (27 days) can raise difficulties after shutting down a reactor, due to the reactivity increase by the slow and steady transformation of Pa-233 into U-233, although less spontaneous neutrons might be present than with plutonium.

Another interesting feature of the thorium fuel cycle is that, due to the lower position of thorium on the Mendeleiev's Table, the production of "long-lived minor actinides" will be noticeably lower, and if uranium + protactinium are removed by reprocessing, the fission products plus minor actinides left as waste will have a long-term (up to 10 000 years) radiotoxicity considerably decreased compared to a U-Pu fuel cycle (if not for the very long term of 10^5 + years). Grossly speaking, it is claimed that the radiotoxicity presented by a High Level Waste repository would be quite small much sooner than for a corresponding U-Pu system (for example in about 10 000 years against 100 000 years).

Whether this is a really important issue, as it is presented these days, remains to be seen.

From all this, it is clear that to take full advantage of the thorium fuel cycle, it is mandatory to retrieve the fissile U-233 formed, by reprocessing, and to regenerate it, as much as possible, in a "self-sustained" type of reactor in which thorium will breed at least as much fissile U-233 material as will be consumed. Which means that reprocessing is a necessary step, with a risk to divert the pure U-233 produced for proliferation purposes.

To avoid this, some would recommend to add U-238 to the thorium fuel in order to somewhat dilute U-233 in non-fissile depleted uranium. This would be at the cost of a loss of reactivity in the reactor, and of a higher production of long-lived minor actinides from U-238, with the presence of Pu isotopes. This could be considered as a draw-off between a socio-political demand and the cost of a physical complication.

Conclusion :

- Use of thorium is linked with reprocessing to isolate valuable fissile U-233, for sustained operation using thorium/U-233.

- Uranium-233 is a remarkable fissile material, its neutronic properties permit to use it efficiently in thermal, epithermal, high temperature and homogeneous reactors. This feature, which could permit to save the natural uranium resources, was originally the main reason to have started thorium research in the 1950s and 1960s.

- The thorium-U-233 system will generate less long-lived minor actinides, the overall radiotoxicity of the high level wastes, after reprocessing, is claimed to be substantially lower than that of U-Pu waste. This ecological aspect is put forward today by some researchers. Besides, absence of plutonium can be a selling advantage nowadays !

- U-233 is a powerful fissile material with which rather simple weapons can be manufactured, because no spontaneous neutrons are produced compared with plutonium, in which case more sophisticated implosion-type weapons are to be made. However, the presence of highly gamma active daughter products of U-232 which is present along with U-233, will permit to localise U-233 more easily than in the case of Pu-239, and hence, deter proliferation.

- This "contamination" with U-232, the higher the fuel burn-up, will also considerably complicate U-233 fuel fabrication, which must be made remotely behind shielding to comply with the modern radiological protection standards, contributing to a high fuel fabrication price, somewhat higher than MOX fuel fabrication.

New concepts of accelerator-driven, thorium-based reactors

In order to generate energy, a nuclear reactor must be the seat of a sustained chain reaction by fissions. The density of fissions conditions the power level.

Decreasing number of fissions, and the reaction has a tendency to diverge. Too less, and it dies away. Thus, a subtle neutron balance must be kept between neutrons produced and neutrons lost (by fission, by absorption, by losses to the environment...) which is translated by saying that the reactivity coefficient k eff should be temporarily very slightly positive. In traditional reactors, k eff would be around i.e. 0.997, if it were not for a small supplement of 0.3 % neutrons called delayed neutrons which are used to drive the reactor, a somewhat subtle process.

It could also be possible to drive a "subcritical" reactor where the neutron balance is in itself lower than 1 by a larger margin (i.e. k eff = 0.95), by a massive, controlled addition of external neutrons. This is the idea of accelerator-driven reactors, which could be safer as they need this extra source of neutrons to become critical. The power level would be a function of the extra neutron flux.

These extra neutrons would be obtained by spallation ("tearing off") of fast protons or other heavy particles impinging on a heavy metal target (for example a liquid lead-bismuth target or otherwise). These particles are to be accelerated by a powerful accelerator (linac or cyclotron).

The proponents of this system claim that it will bring added safety to the fission reactor which can be shut down more easily. It will also reduce the fissile hold-up in the reactor, another safety feature.

In some configurations, it could also be used favourably to transmute the long-lived radioelements into shorter-lived or stable isotopes. Theoretically, the surplus of extra neutrons could help compensate the poisoning by some fission products, hence avoid reprocessing and consequently avoid one of the possible causes of proliferation.

A thorium reactor will add to the overall safety by the lesser radioactivity of the filiation products, as seen in § 4 above, inasmuch as the extra neutrons generated from the accelerator beam could make up to some extent for the genuine inertness of the fertile thorium fuel, acting as a neutronic "driver". A molten-metal or molten-salt thorium-U-233 reactor, or a pebble-bed thorium/U-233, due to their homogeneous configuration, will give even better results.

Conclusion :
the accelerator-driven reactor techniques are an interesting proposition which merit further consideration.

However, there are many questions which should be addressed :

- the total cost of a reactor (even simplified) plus a powerful accelerator,
- the technical aspects of coupling two rather different machines,
- the fact that, today, driving a reactor is well mastered by "traditional" means and that the problems, nowadays, are not necessarily in this area,
- with a thorium reactor, the delayed reactivity due to Pa-233 decay after shut-down should be carefully investigated, as this effect is more pronounced with thorium.

Practical experience with thorium reactors

- Thermal heavy-water prototype reactors with Th/U-233 have been operated in the US and are now experimented in India. They demonstrate that they can be run as "self-sustained' reactors where U-233 burnt can be replaced by the neutron captures in the fertile thorium.

- The light-water PWR Shippingport experience in the US (1977-1982) has demonstrated that an experimental U-233/Th light water reactor can also be self-sustained. The BWR Elk River reactor (1963) and PWR Indian Point experiment (1962), both with U/Th fuel elements, show that such modern type reactors can run on a thorium cycle.

- Different high temperature reactor prototypes in USA, Germany and UK (the Dragon OECD-EURATOM project) have shown similar capacities with an excellent behaviour at very high coolant temperatures (more than 1 000°C).

- A molten salt reactor experiment has been operated rather successfully at Oak Ridge for some years.

To start with, mixed cycles with usually U-235/U-238 together with Th-232, have been organised to bring the necessary missing neutrons at the beginning. This was followed by separation of uranium and thorium (and possibly plutonium) in a reprocessing plant.

These cycles have been piloted and the experience exists.

The separation of U-233 and thorium is usually done by wet liquid-liquid extraction using the THOREX process for which a pilot plant has been operated during many years at Oak-Ridge.

The dissolution of thorium metal and thorium oxide is not as straightforward as with uranium and needs some addition of hydrofluoric acid, which is well known to be aggressive for chemical equipment, needing appropriate buffering agents to prevent corrosion. The reprocessing of High Temperature Reactor's carbide fuels adds extra difficulties due to the impervious coatings of the carbide or oxide fuel. Although the operations have been demonstrated at pilot plant stage, these are not straightforward and present a real challenge, but this is also true for uranium oxycarbide coated fuels.

Besides, as mentioned before, handling of purified U-233 cannot usually avoid the presence of some U-232 and adequate gamma protection. The consequence is that remote or semi-remote fabrication of U-233 fuel is mandatory, which complicates handling and increases the costs. Reprocessed thorium also contains Th-228 and Th-234 which prevent direct handling for many years.

Conclusion :

- The feasibility of different types reactors based on U-233/Th has been successfully demonstrated and experience is available. However, to put it simply, U-233 breeding needs an extra neutron while the uranium cycle does not.

- The thorium fuel cycle has been extensively investigated.

- Reprocessing of U/Th is somewhat more expensive than with the traditional uranium cycle but should not pose *too* great problems to the reprocessors today once the proper flowsheets have been selected, except for the head-end of pyrocarbon/silicium-coated carbide or oxide fuels.

- U-233 fuel fabrication by remote means is a challenge at an extra cost.

Thorium experience in the world (Main achievements)

Extensive experiments have been conducted in **the USA** following an initial development for the defence programme. A number of national laboratories have been involved, as well as the largest US companies.

Three extended reactor programmes have been undertaken in the USA :

- the High Temperature Gas Cooled Reactors with Peach Bottom, Fort St Vrain,
- the Light Water Reactors with Elk River (BWR), Indian Point (PWR) and
- the "seed-blanket" Shippingport PWR, a light water breeder.

All these reactors have produced electricity.

The prototype Molten Salt Reactor Experiment (MSRE) which has operated in Oak Ridge until 1976 has demonstrated the feasibility of such a machine with a good breeding gain, and a continuous "milking out" of the fission gaseous products.

In Germany, besides much theoretical research and tests at a pilot scale, the High Temperature Pebble-Bed Reactor has been successfully demonstrated with the very reliable AVR (15 MWe) which has operated for 21 years, and a larger prototype of 300 MWe, called THTR, which was built and operated for some time. However, unforeseen technological difficulties and the political climate led to the decision to stop its operation prematurely in 1989.

In India, the Th/U-233 fuel cycle development is part of the present programme, with a critical facility using U-233/Al alloy fuel elements PURNIMA 3 and a similar test reactor of 30 KW KAMINI under start-up.

Some member countries within **OECD NEA** have set up, in association with EURATOM, the HTR prototype DRAGON for some of its members. This small 30 MWt reactor has made a successful demonstration in the UK between 1967 and 1973.

Research has been conducted in many other countries, especially on the back-end of the fuel cycle, in **Italy** where a prototype reprocessing plant and remote fuel fabrication plant (PCUT) has shortly been operated. **Russia, Canada and France** have also extracted thorium, conducted studies and tests. Some research goes on in **Canada** for application of the thorium cycle to the Candu reactors, and theoretical research has been going on, for a number of years, and continuing in **Japan** with limited experiments. The Japanese High Temperature Test Reactor and the HTR-10 Chinese prototype under construction, could well be, in the future, used with thorium.

Conclusion :
Extensive experience has been accumulated so far, theoretical as well as practical and engineering-wise. Many neutronic data are available, although not as many as with the U/Pu cycle, leaving some uncertainties in multigroup calculations in special cases.

A corpus of experience exists today for any country with nuclear engineers and a good technological background, to consider a nuclear energy programme using thorium. India is having a try at it.

The thorium fuel cycle : Advantages ?

Why have so many expensive projects been stopped between 1975 and 1980 without further continuation ? Even after recognising some of the interesting properties of the particular Th/U-233 fuel cycle ?

- Just because these projects were somewhat more complicated and more expensive compared to a somewhat cheaper and simpler uranium fuel cycle,

- Also because Presidents Ford and Carter have enacted a ban on reprocessing under the fear of proliferation. Without reprocessing, the thorium fuel cycle is not viable if one wants to use U-233 in a long-range project,

- Also because the initial objectives to find enough nuclear raw material have changed since : uranium is plentiful and it is the cheapest available raw material per unit of energy today. The by-product plutonium must be taken care of as a priority for non-proliferation reasons and it is reasonable to burn it as a fuel,

- It is not sure today that even with no ban on reprocessing in the US the many projects involving thorium would have been continued, for the other reasons indicated,

- It is not sure that the recent interest in thorium raised due to its somewhat lower radiotoxicity of the spent fuel or high-level waste compared with the uranium fuel cycle, is a sufficient incentive to start it all again, with the very moderate demand for reactors these days. India continues, rightly so, for national independence, in view of its huge energy needs, limited uranium reserves and large thorium resources.

The time does not seem ripe yet to relaunch a flourishing thorium nuclear industry. However, as time passes, modern technological breakthroughs make their way and prepare for a thorium come back.

The main one, in our view, is the progressive introduction of remote fuel fabrication techniques due to MOX fuels, burning actinides, etc... These techniques will, on one hand, bring a simplification of the reprocessing processes, and they will be ready for the remote fabrication of U-233 fuel elements. They will also be a bonus for antiproliferation systems, by favourizing "spiking" by leaving some radioactivity in the fuel and "coprocessing" by leaving some thorium with U-233.

Another incentive will be the trend to achieve higher thermodynamic yields with high temperature reactors which may compete some day with high temperature thermal combustion generators or which may permit interesting high-temperature cogeneration experiments, like chemical syntheses or hydrogen production. Thorium/U-233 reactors are especially well-suited for high-temperature reactors, but high temperature reactors can be made with uranium fuel too.

However, thorium utilisation, whatever its interest, is dictated by long-term views of the uses of nuclear energy, preferably in a period of nuclear expansion when thorium will supplement the uranium reserves. Then it will be possible to take advantage of the specificities of U-233 and thorium compared to other fuels.

The Reader is invited to complete these general considerations by the details given in the next chapters, and by the Concluding Remarks.

2. THORIUM AND CONCERNED ISOTOPES: NUCLEAR REACTIONS AND POSSIBLE APPLICATIONS

2.1. Thorium Radioactive Decay

The decay chain of thorium is given in Fig.1. (Ref.2).

Note : it can be seen that Th-232 and U-232 have a decay chain in common, in which can be found semi-strong to strong gamma emitters (Pa-233, 27 days, 40 % with 0,3 MeV; Bi-212, 60.6 minutes, a fraction with strong gamma up to 1.8 MeV; Tl-208, 3.1 minutes, with strong gamma emissions up to 2.6 MeV). Were it not for the very slow decay of Th-232 ("4 n" series) (Fig.2) due to its very long half-life, natural thorium would be quite radioactive. The minute fraction of Th-230 is on the way of the U-238 ("4 n + 2" series). It decays into Ra-226 which itself decays into short-lived nuclides, among which gamma emitter Bi-214 (0,6 MeV), to end up also with Pb-206.

Compared with the uranium decay chain, either from U-238 or U-235 ("4 n + 3" series), it is the thorium chain which has the strongest gamma emitters, even if they are not abundant. Hence, an easy way to detect even small amounts of material contaminated with thorium or U-232. We will see later that it is, from this viewpoint, a plus for the proliferation resistance due to its rather easy detectability, but an inconvenience for U-233 fuel fabrication.

For example, the activities per gram U-233 are as follows : (Table 1 and Fig.3)

	1 hour	1 day	1 month	1 year	Responsible nuclides
Activities in disintregations/sec (becquerel)	10 E 8	10 E 9			Th-228
	10 E 6	10 E 8			Ra-224 Rn-220 Po-216
	10 E 5	10 E 7			Pb-212 Bi-212 Tl-208
	~ 10 E 8	~ 10 E 9	10 E 10	10 E 11	All nuclides

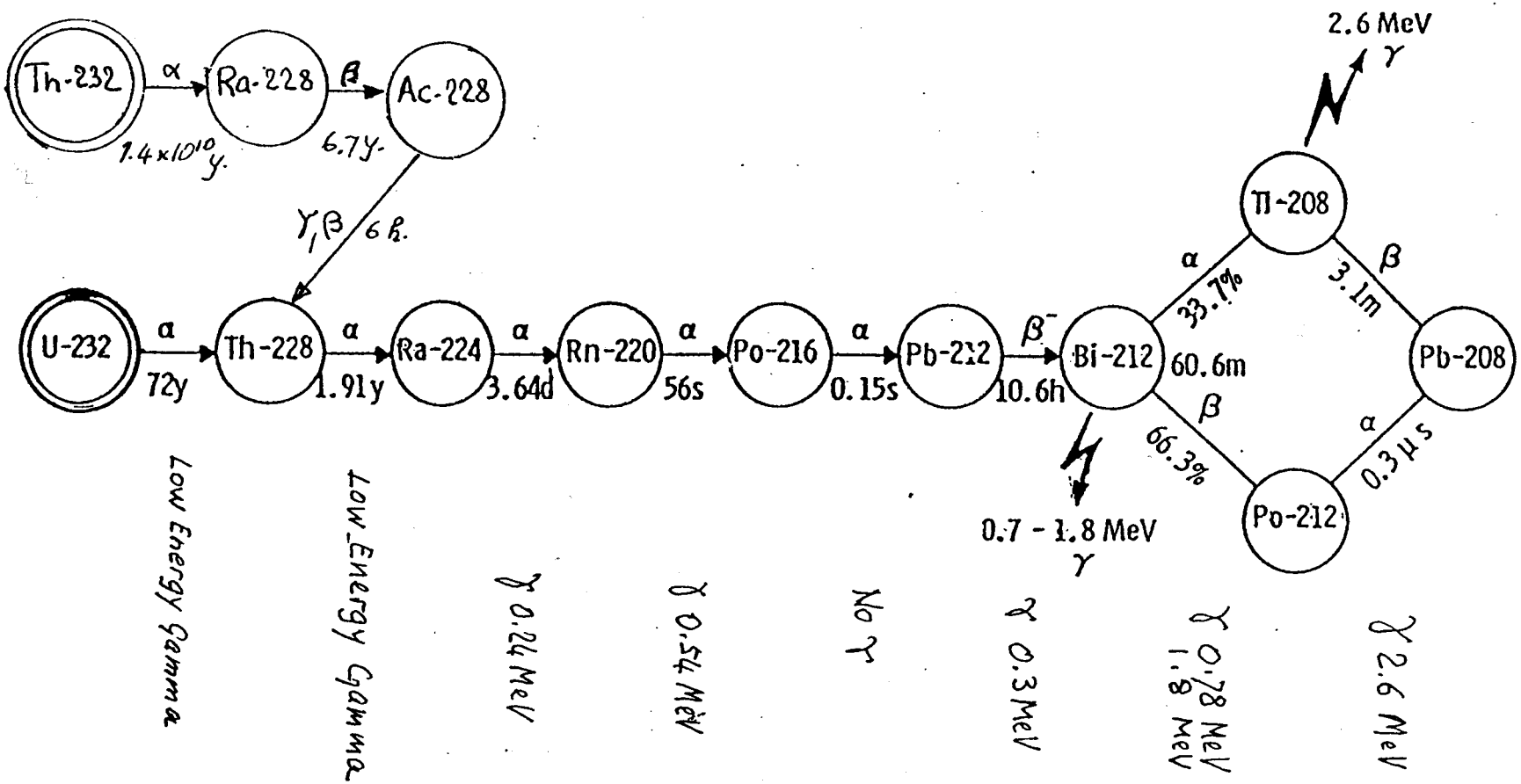


Figure 1: Th-232, U-232 decay chain (2)

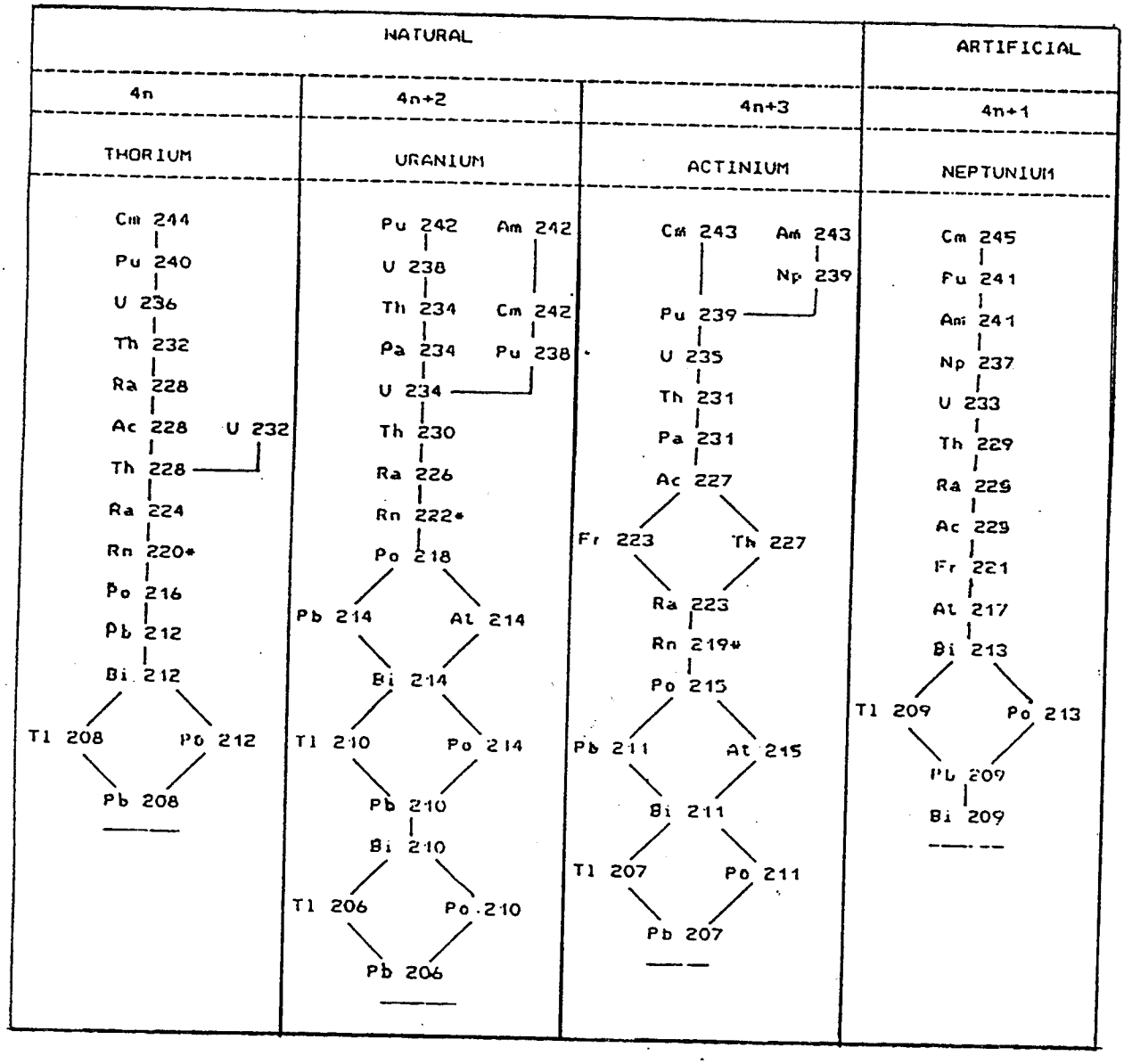


Figure 2: Four main decay series

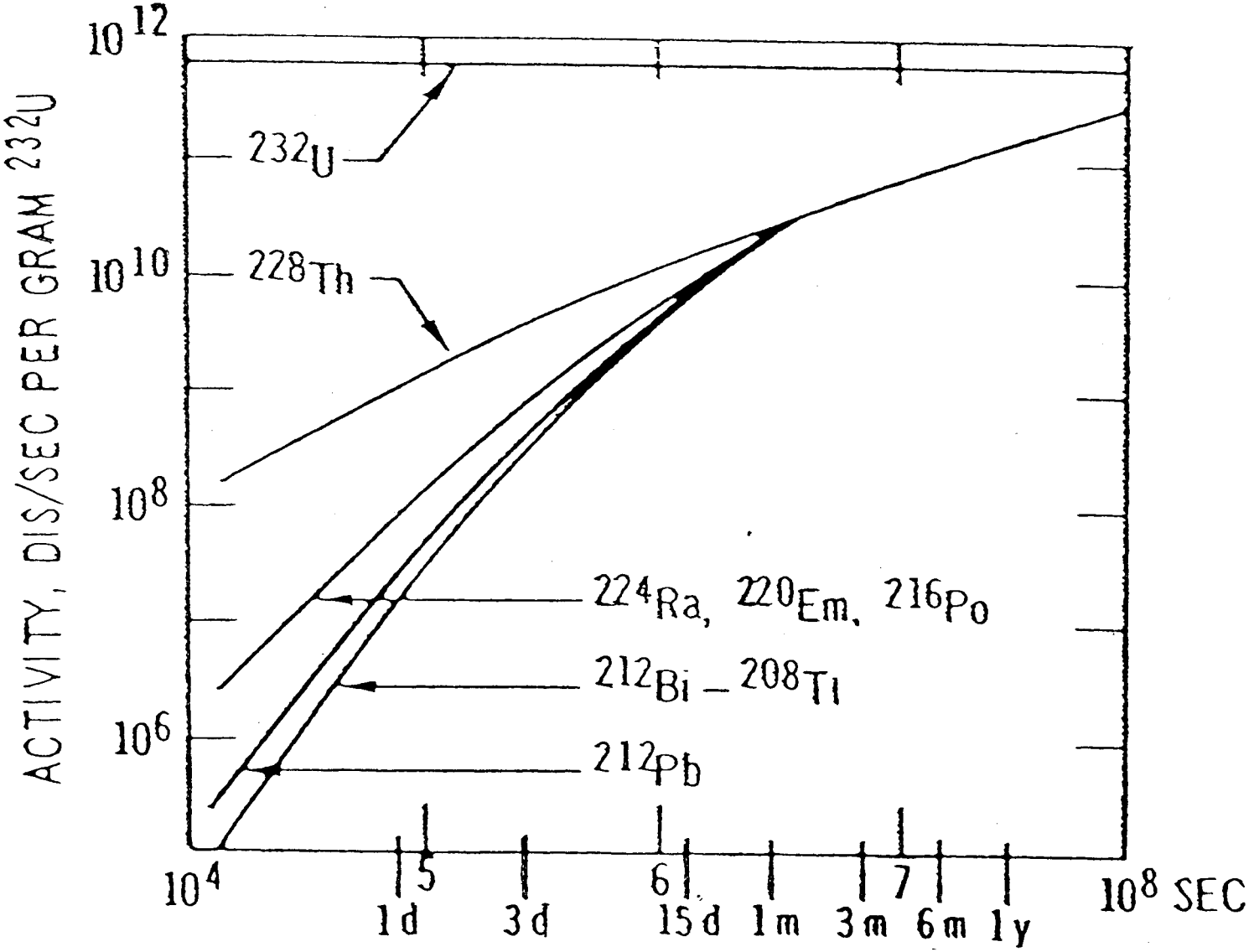


Figure 3: Decay of U-232 and build-up of daughters vs. time (99)

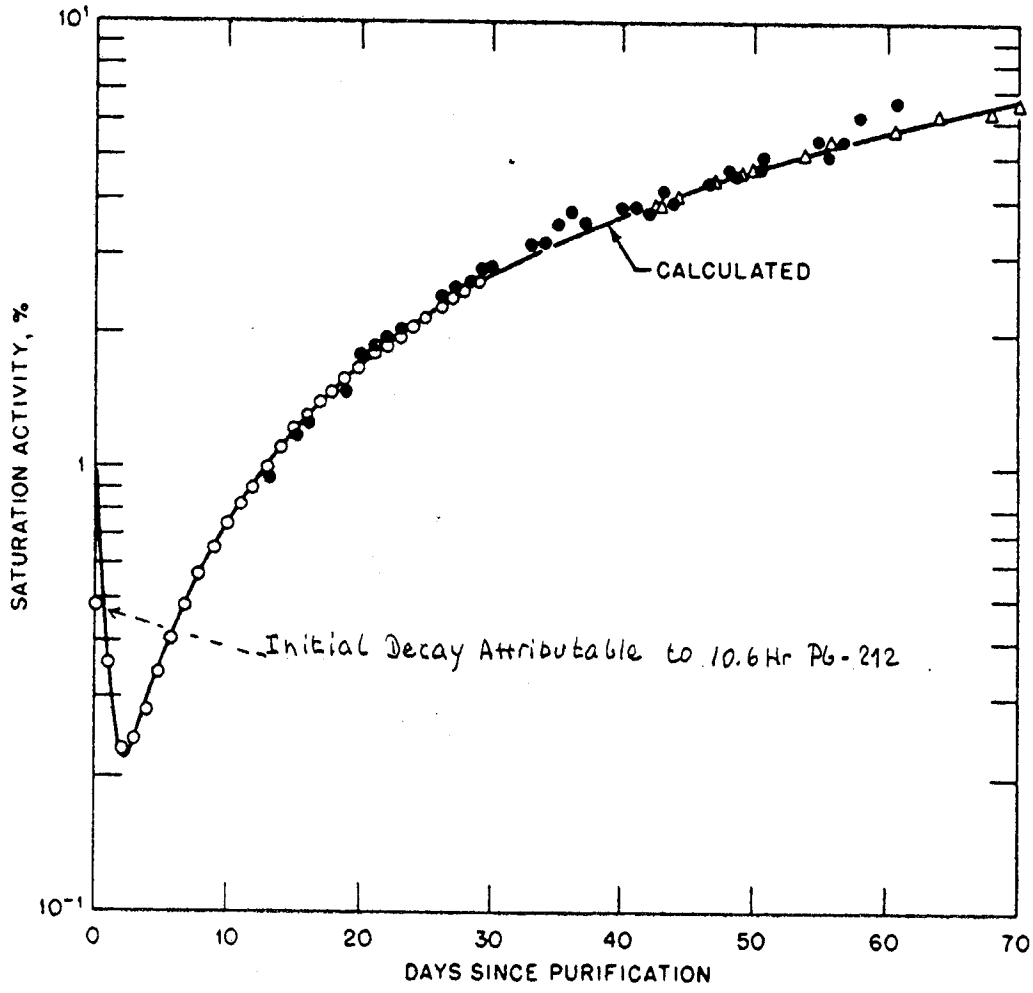


Figure 4: Calculated and observed growth of gamma activity of ^{233}U material containing about 42 ppm of ^{232}U. O, ORNL-368 (run HJ-3). •B&W total gamma activity. △, B&W 2.6-Mev ^{208}Tl. (99)

Therefore U-233 which contains traces of U-232 should be manipulated quickly after reprocessing if one wants to avoid remote manipulation. (Fig.4) (See also Chapters 6.4.2. and 7.3.2. where the case of reprocessed thorium is also discussed).

Figure 5 shows the shielding thicknesses required for fabrication of U-233 High Temperature Reactor fuels. Above 2 inches, remote handling is recommended. It is clear that with today's norms remote fuel fabrication is compulsory for U-233.

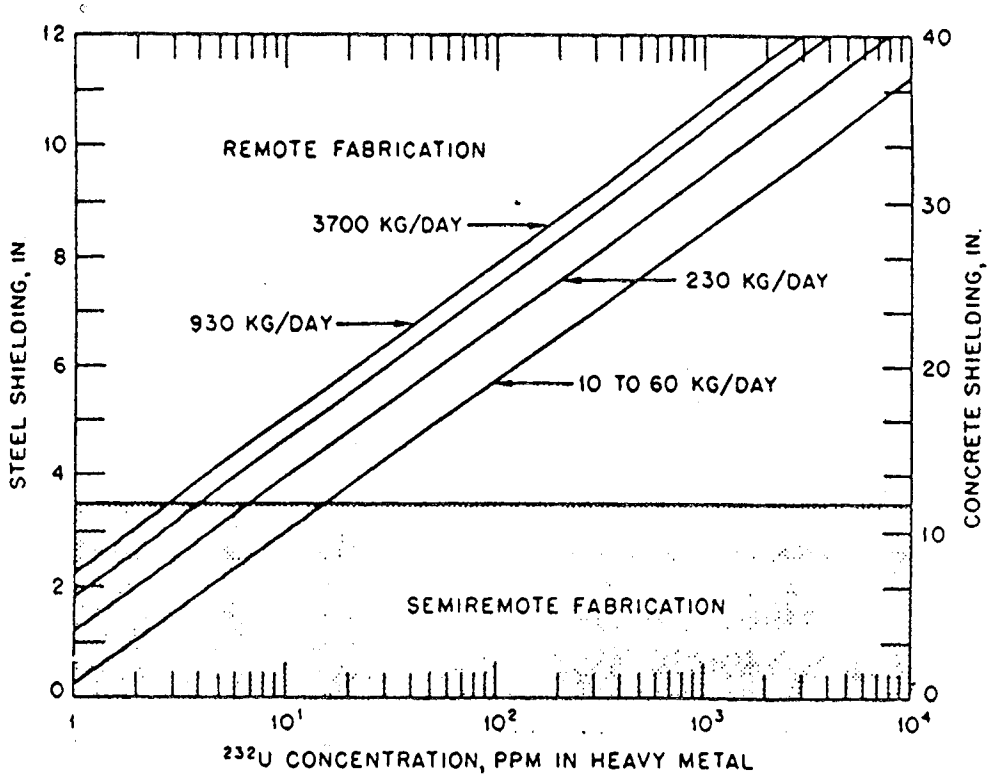


Figure 5: Shielding thicknesses required for fabrication of U-233 High Temperature Reactor fuels (93).

This has an important industrial bearing on the possible development of the thorium fuel cycle, and has been so far one of the most serious obstacles to this development.

Table 2. (11) gives some characteristics of the thorium isotopes. For connected isotopes, one should better refer to the more recent Segré isotope tables.

Table 2: Survey of thorium isotopes and their formation (11)

ε = electron capture.

mass number	half-life	mode of decay; energy	mode of formation	mode of detection	application
212 to 222	≤ 1.2 s	α, ε	heavy beam reaction with ^{206}Pb, ^{208}Pb, and ^{209}Bi	recoil technique	investigations on nuclear properties
223	0.66 s	α: 7.29 MeV	^{227}U decay	α-spectrometry	
224	1.0 s	α: 7.17; 7.00 MeV γ: 177 keV	^{228}U decay		
225	8 min	ε α: 6.48; 6.44 MeV γ: 322 keV; ...	^{229}U decay	α-spectrometry; half-life determination	
226	31 min	α: 6.34; 6.23 MeV γ: 111 keV	^{230}U decay from artificial ^{226}Ac		
227	18.7 d	α: 6.04; 5.98; 5.76 MeV γ: 236 keV; ...	natural	α-spectrometry	heat source; n-source material; radiochemical education; source for Np-decay members
228	1.91 a	α: 5.42; 5.34 MeV γ: 84 keV	natural ^{232}Th(d, 2n) ^{232}Pa $\xrightarrow{\beta^-, \alpha}$ ^{228}Ra(2n)		
229	7340 a	α: 4.90; 4.85; 4.82 MeV γ: 94 keV	^{233}U decay		
230	7.54 × 10^4 a	α: 4.69; 4.62 MeV	natural		tracer (long-lived), starting nuclide for ^{231}Pa and ^{232}U production
231	25.5 h	β$^-$: 0.3; 0.4 MeV (max) γ: 26; 84 keV; ...	natural ^{232}Th(n, 2n)	α-spectrometry	tracer
232	1.405 × 10^{10} a	α: 4.01; 3.95 MeV	natural	XRF, classical methods	nuclear fuel
233	23.3 min	β$^-$: 1.23 MeV (max) γ: 87; 29; 459 keV	^{232}Th(n, γ) ^{232}Th(d, p)	γ-spectrometry; GM-counter	tracer
234	24.1 d	β$^-$: 0.2 MeV (max) γ: 92; 62 keV	natural	γ-spectrometry β-detection (liquid scintillation counter, GM-counter)	tracer; radiochemical education
235	6.9 min	β$^-$ γ: 416 to 932 keV	^{234}Th(n, γ)	β-detection (GM-counter); half-life determination	investigations on nuclear properties
236	37.1 min	β$^-$: 1.0; 1.1 MeV (max) γ: 111; 113 keV	^{238}U(γ, 2p) ^{238}U(p, 3p)		

References for 2.1 on pp. 20/1

2.2. Neutronic Reactions

2.2.1. Preliminary remarks

Very soon it appeared that thorium-232 and uranium-238 had very parallel behaviours when submitted to a flux of thermal neutrons as observed in a "conventional" reactor. (Fig.1.) (19)

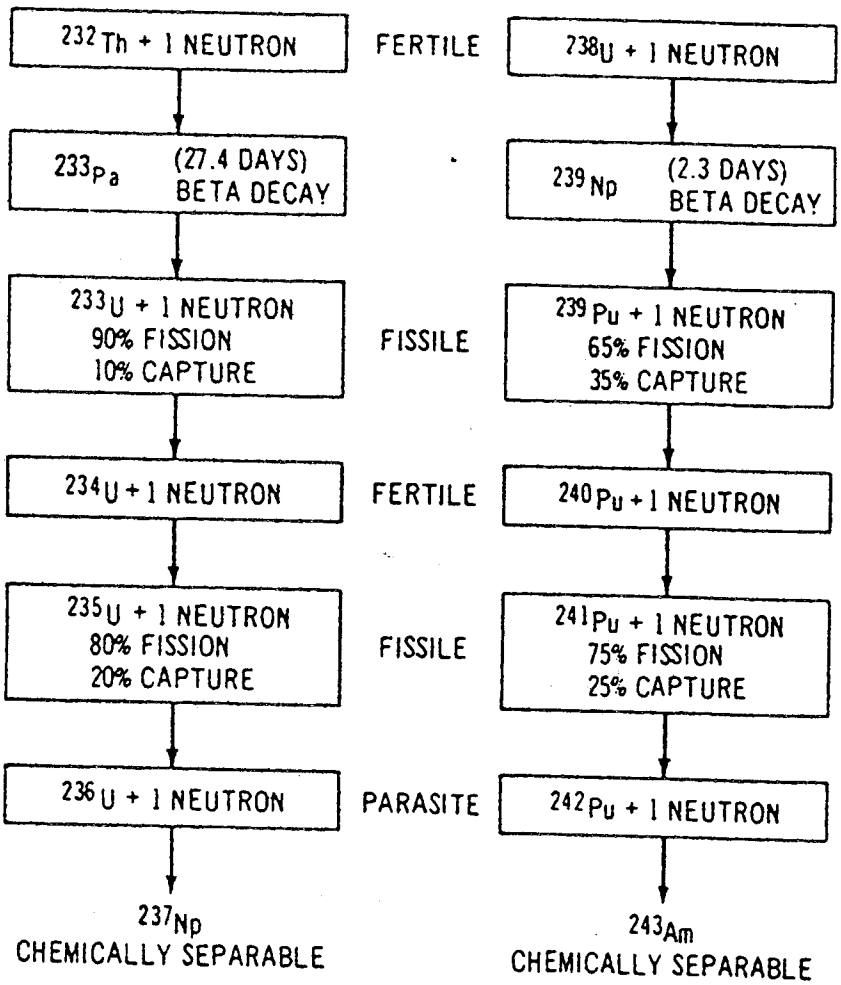


Figure 1: Isotopic build-up in thorium and U-238 systems (19)

For example, the approximate thermal neutron capture cross sections and fission cross sections are given in Table 1. below :

Table 1

	Th-232	Th-230	U-238	U-235	U-234
Abundance	100 %	ε	99,275 %	0,720 %	0,005 %
σ c barns	7.4	23,2	2.3	98,4	100,2
σ f barns	+ε	0,0012	-	**584**	0,6

It is clear that uranium, thanks to U-235, can be fissioned and entertain a chain reaction, under proper conditions of neutron concentration, neutron energy and U-235 concentrations. Uranium-235 is the only natural "match" capable of starting a chain reaction.

The same is not possible with thorium, which can only absorb the neutrons by (reactions n, γ or n, 2n).

The resulting nuclides are shown on Figures 2 and 3. Thorium is transformed into Th-233, a beta emitter (T = 22 min), then into Pa-233, a beta emitter with a gamma (0,3 MeV) fraction, which decays (T = 27 d) into Uranium-233.

Uranium 233 thus bred has a number of interesting properties seen from the standpoint of energy production by fission.

In fact, were it not for the extra neutron used to produce U-233, it is probable that the nuclear industry would prefer use U-233 to plutonium, because its qualities more than counterbalance its drawbacks.

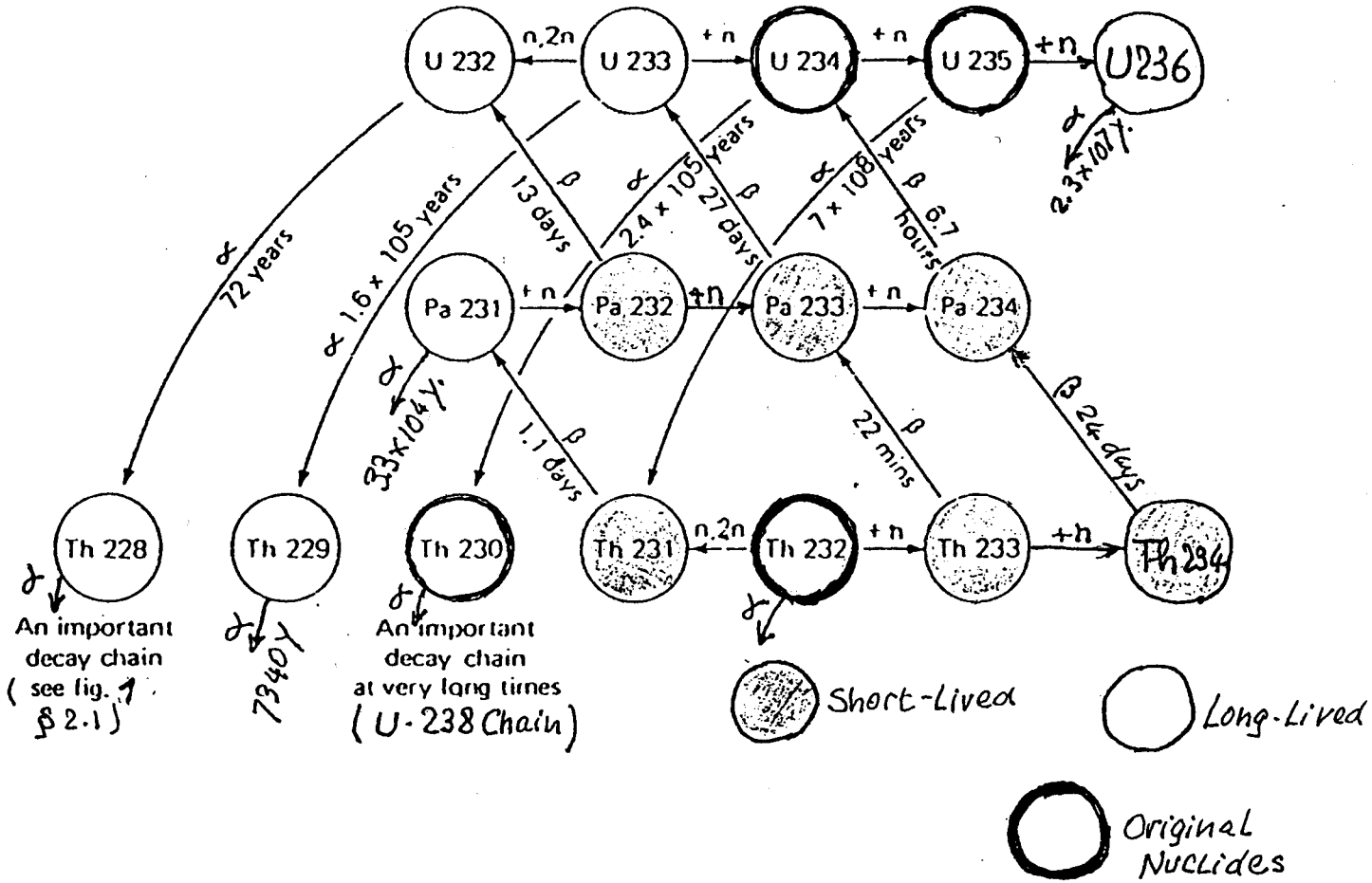


Figure 2: Main isotopes in Th-232/U-233 fuel (176)

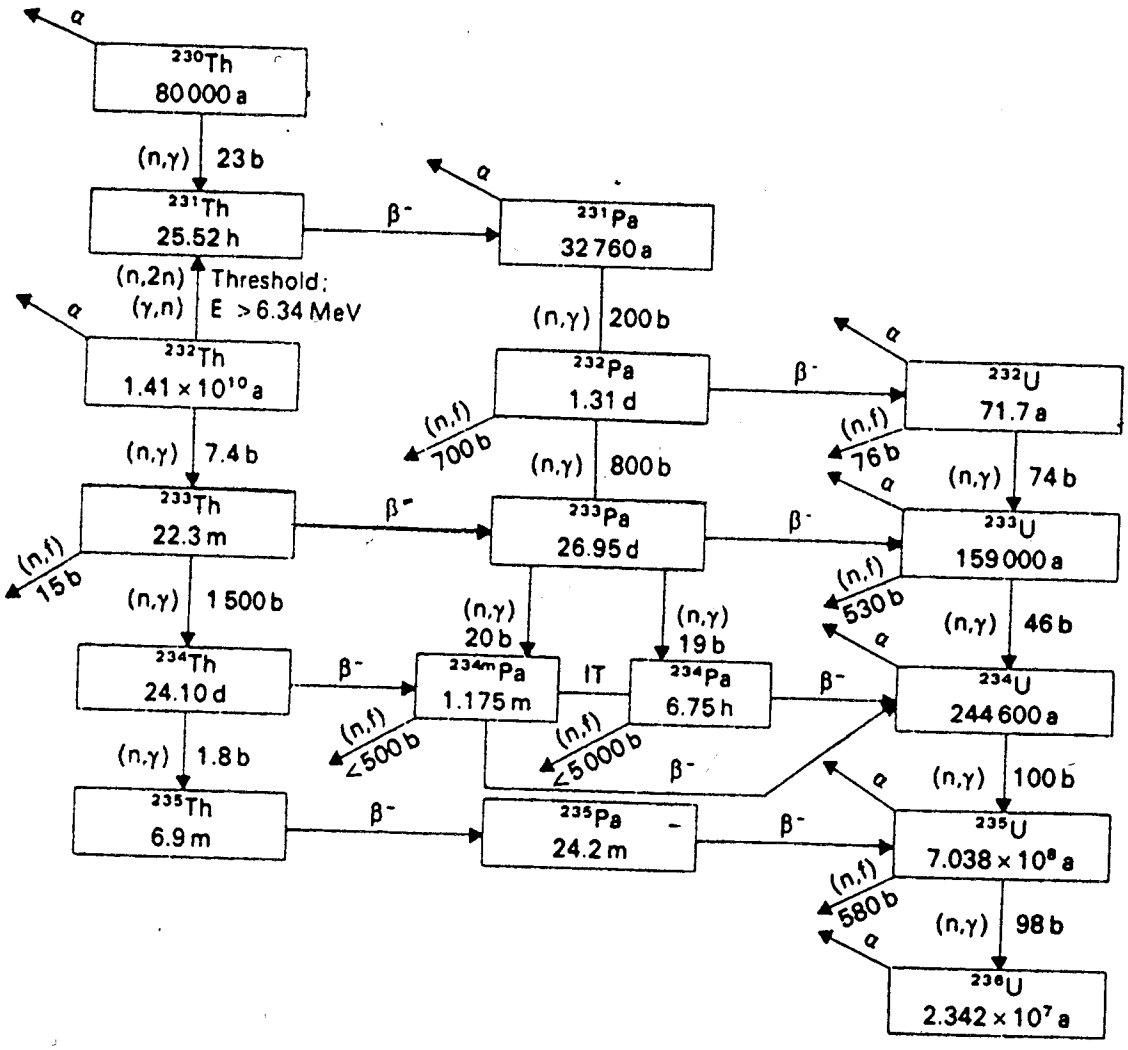


Figure 3: Nuclear reactions in the reactor neutron irradiation of thorium-232 (15)

2.2.2. **Considerations on the most relevant individual isotopes**

NOTE : many of the remarks below have been quoted from Thorn and al (176) who have treated the subject with great clarity.

Thorium-232

The nuclear property of Th-232 which has the greatest importance in determining its behaviour in a reactor, is its neutron capture cross-section. Plots of the variation of this cross-section versus neutron energy are given in Fig.1 and 2. Figure 1 gives the variation up to an energy of 1 keV, while Fig.2. gives the variation above 1 keV. Fig.2 also shows the variation of other cross-sections of interest.

It is a general rule that the neutron capture cross-section of an isotope varies inversely as the neutron velocity (or square root of the energy). This rule applies fairly well over much of the lower energy range of Fig.1. At higher energies (the "resonance region") the cross-section rises to peaks at certain energies. Depending on the neutron flux in the resonance region, many neutrons may be absorbed at the energies of resonance.

Figure 2 plots the U-238 capture cross-section for comparison. It will be seen that the U-238 value is lower than the Th-232 value throughout the energy range, typically by a factor of about 1.5.

Figure 2 also plots the fission cross-section of Th-232. Below 1 MeV the cross-section is zero, and only above about 1.4 MeV does the cross-section rise rapidly. Even in a fast reactor only 10-15 % of the neutron flux is typically in the energy range above 1 MeV, and this is the reason why fission in Th-232 is of minor importance. In contrast, the fission cross-section of U-238, also shown in Fig.2, rises quite rapidly from below 1 MeV, and since there are many more neutrons at these slightly lower energies. Fast fission of U-238 is of significance in all reactors and is particularly important in fast reactors.

For the purposes of reactor calculations, it is necessary to divide the energy range of Fig.1 and 2 into intervals within which the material cross-sections are represented by average values. These energy intervals or groups are chosen to give adequate representation of the cross-section variations of the different neutron reactions and the choice of the number of energy groups depends on the type of calculation. For accurate analysis it is almost always necessary to divide the range into many groups, but for the purposes of qualitative comparison it is convenient to use an approximately equivalent cross-section applicable over a wider range of energies. Where this covers the whole range of interest, it is

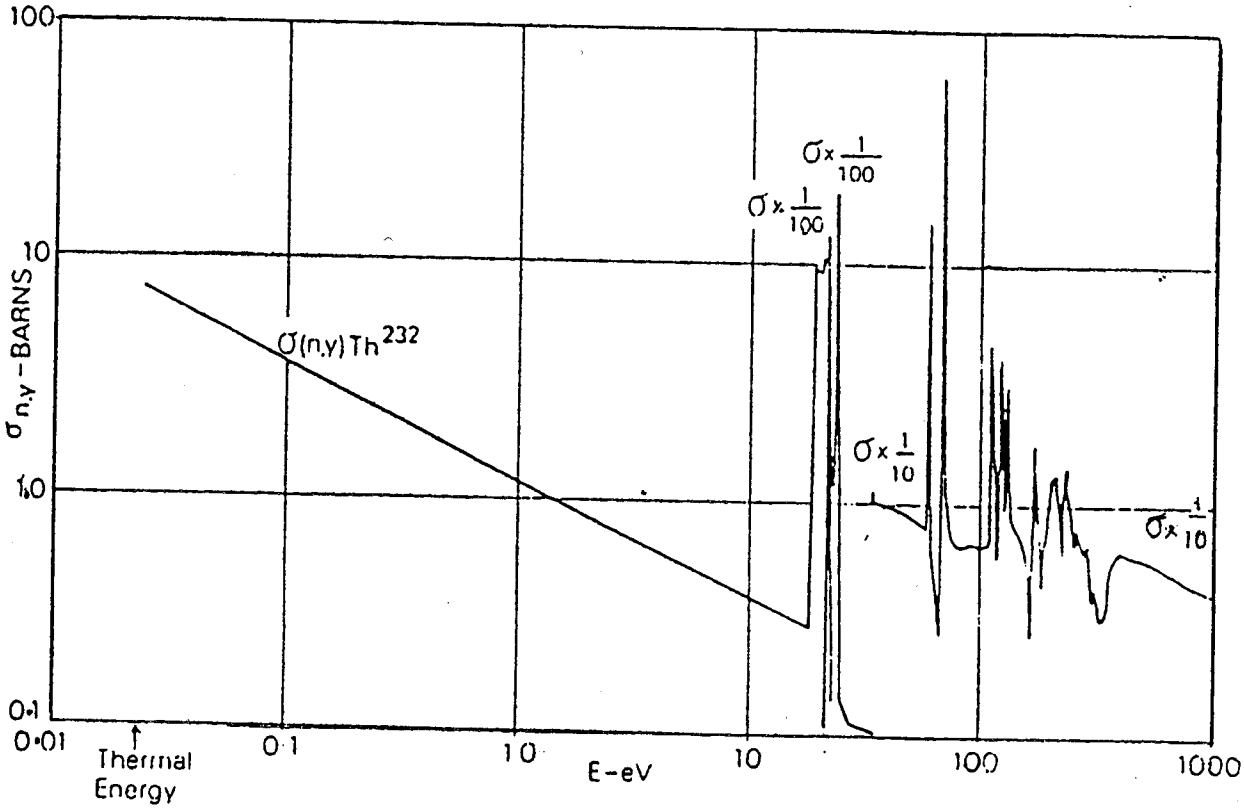


Figure 1: Th-232 capture cross-section (176)

termed a "one-group cross-section". There is a variety of thermal reactors, each having different neutron energy spectra and therefore also different one-group cross-sections. It is customary for these systems to quote two parameters, the cross-section at a neutron velocity of 2200 m/sec (an energy of 0.025 eV) which is most probably velocity at room temperature, and the resonance integral, which is the integral over the energies of the resonance region of the additional cross-section due to resonances weighted by the reciprocal of the energy.

Table 2 gives these parameters for the main fertile isotopes, together with one group cross-sections or a fast reactor. (Those are typical values).

Table 2

CONDENSED NUCLEAR DATA FOR FERTILE ISOTOPES (176)

	Thermal reactor				Fast reactor	
	2200 m/sec value (0.025 eV)		Typical resonance integral		One group	
	Th-232	U-238	Th-232	U-238	Th-232	U-238
Cross-section (barns) (σ)						
Fission (σ f)	0	0	0	0	0.01	0.05
Capture (σ c)	7.6	2.7	85	275	0.35	0.3
Neutrons per Fission (ν) (on average)	-	-	-	-	2.3	2.75

For the thermal reactor data, the higher 2200m/sec capture cross-section of Th-232 is compensated by the lower resonance integral. The importance in any particular reactor of the resonances which occur at neutron energies above thermal, will depend on the degree of neutron moderation in the reactor core. The effect on reactor performance of this property of the various fertile isotopes is therefore least in well moderated reactors having a "soft" spectrum, (i.e. more neutrons at the low energy end), as typified by current Heavy Water Moderated Reactors. It is greatest in reactors such as the High Temperature Reactor (HTR) which depend on some, but less, moderation. The degree to which the fuel is segregated from the moderator also influences the outcome, because a discrete

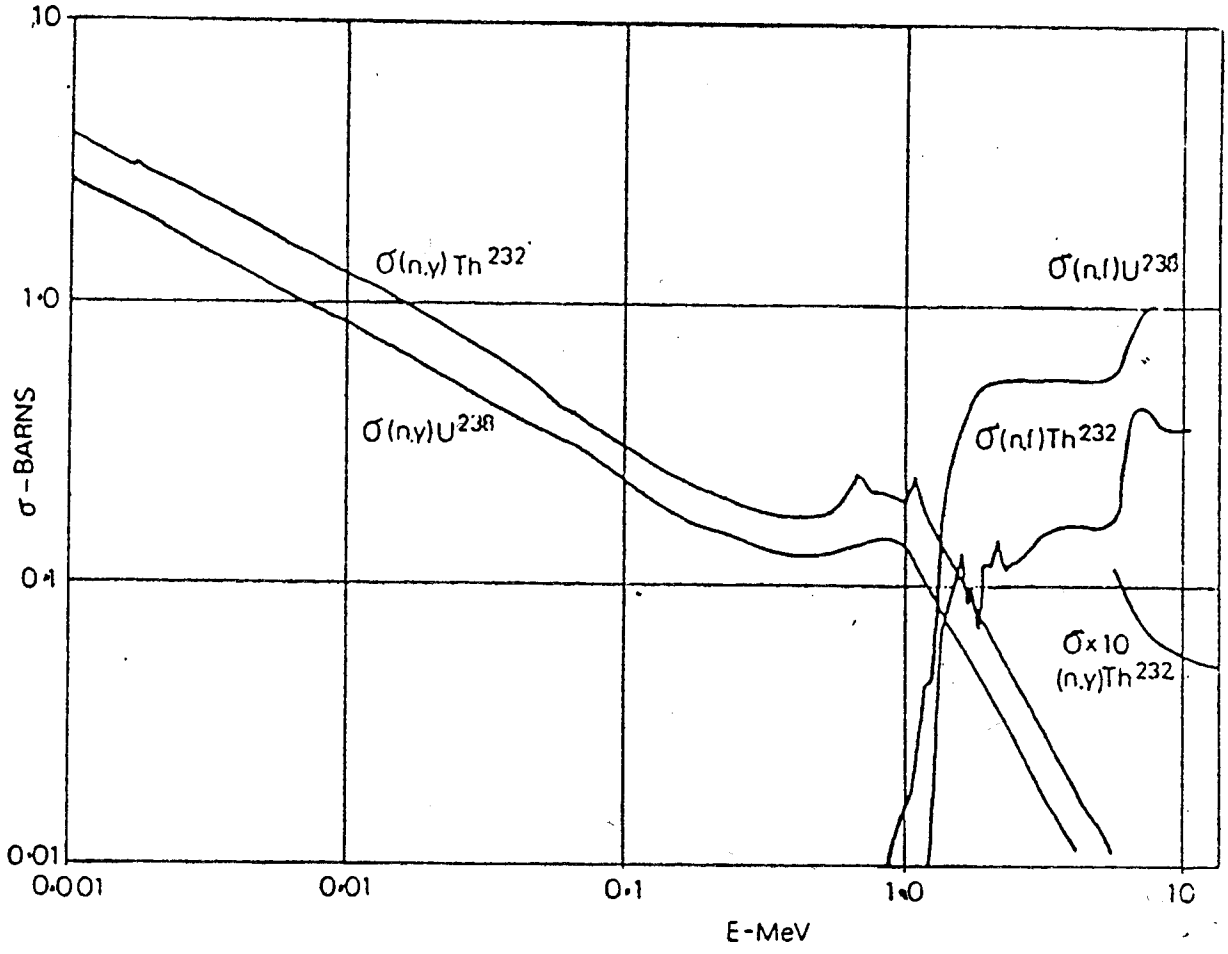


Figure 2: U-238 and Th-232 capture and fission cross-sections (176)

mass of fuel allows neutrons to slow down through the resonance energy region with reduced probability of capture. The more thoroughly the fuel is dispersed in the moderator, the more resonance capture takes place, and the higher the concentration of fissile material ("enrichment") which is necessary to maintain the chain reaction. Because the resonance integral of Th-232 is smaller than that of U-238, as shown in Table 3, this effect is less marked, and from this point of view the use of Thorium is more likely to be favoured.

There are two main differences in the integral properties for fast reactors. Firstly, the fission cross-section of Th-232 is much lower than that of U-238, and the number of neutrons produced per fission is also lower. Secondly, the neutron capture cross-section of Th-232 is higher, by about 15 %.

The importance of the first effect, that of the fission cross-section, is that in uranium/plutonium fuelled fast reactors, a significant proportion (about 15 %) of the fissions take place in U-238. This effect is a double bonus, because each fission which takes place in a fertile material means a corresponding reduction in the destruction of the valuable fissile material and the extra fission neutrons produced contribute towards the formation of fresh fissile atoms. In a Th-232 / U-233 fuelled fast reactor, only about 2 % of the fissions take place in Th-232, and the breeding gain suffers accordingly. In thermal reactors fission in fertile materials is very small in Th-232 but is still quite significant in U-238.

The importance of the second effect, the higher capture cross-section, is in terms of the fuel enrichment. A high capture rate in the fertile material implies a need for a high feed enrichment, which increases the fissile material doubling time resulting from a given breeding gain.

Uranium-233

The cross-section of U-233 of most importance is usually that of fission, with a relatively small capture cross-section, when compared with other fissile atoms. Figure 3 gives the variation of the fission cross-section with energy up to 1 keV. Figure 4 gives the variation of some other cross-sections above 1 keV.

Figure 3 shows that there are many resonances in the U-233 fission cross-section, in the range 1 to 200 eV. Fig. 4 gives a plot of the fission cross-sections of the main alternatives to U-233, i.e. U-235 and Pu-239. Overall, it may be seen that the fission cross-sections of the latter two isotopes are lower than that of U-233. Fig. 5 summarizes the fission neutron yields ν for different incident neutron energies.

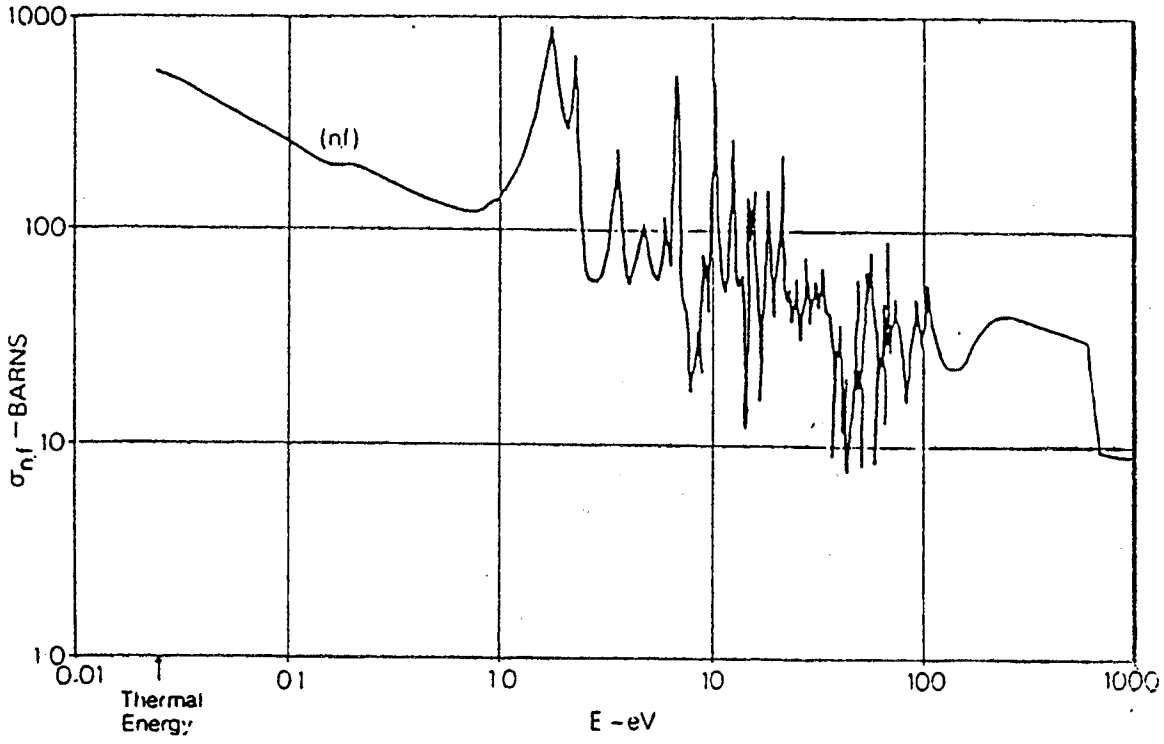


Figure 3: U-233 fission cross-sections

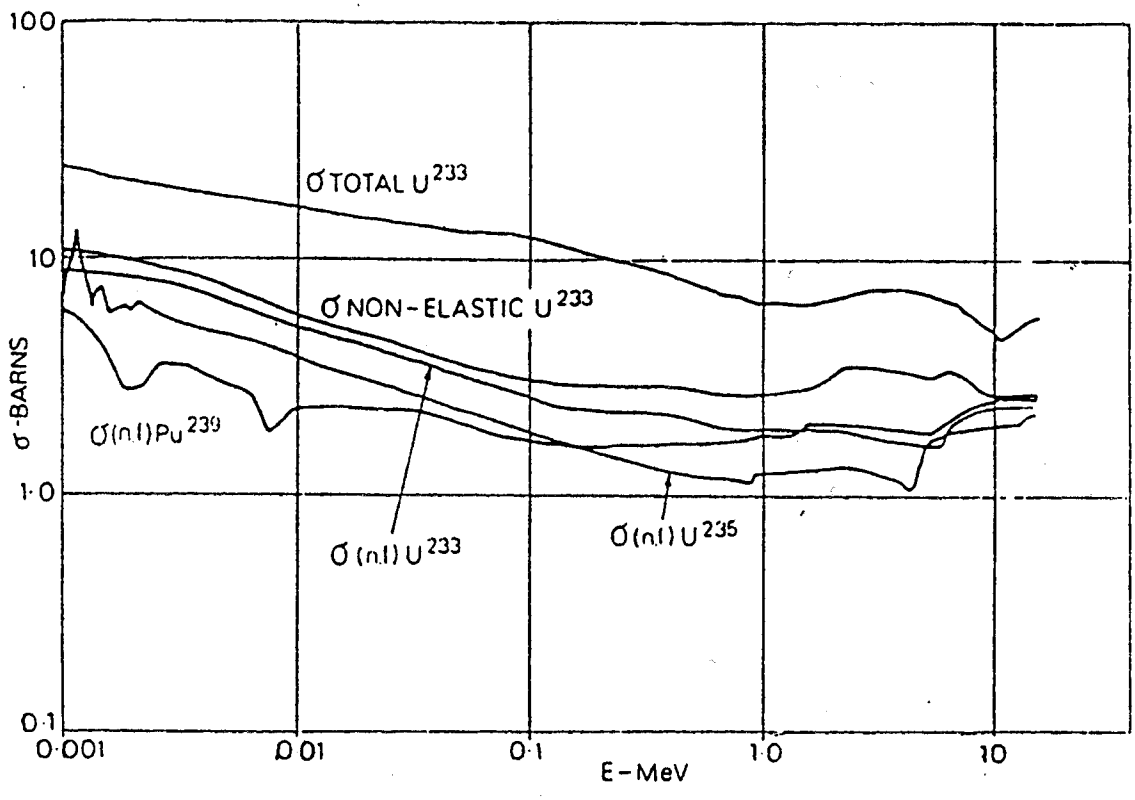


Figure 4: U-233, U-235 and Pu-239 cross sections

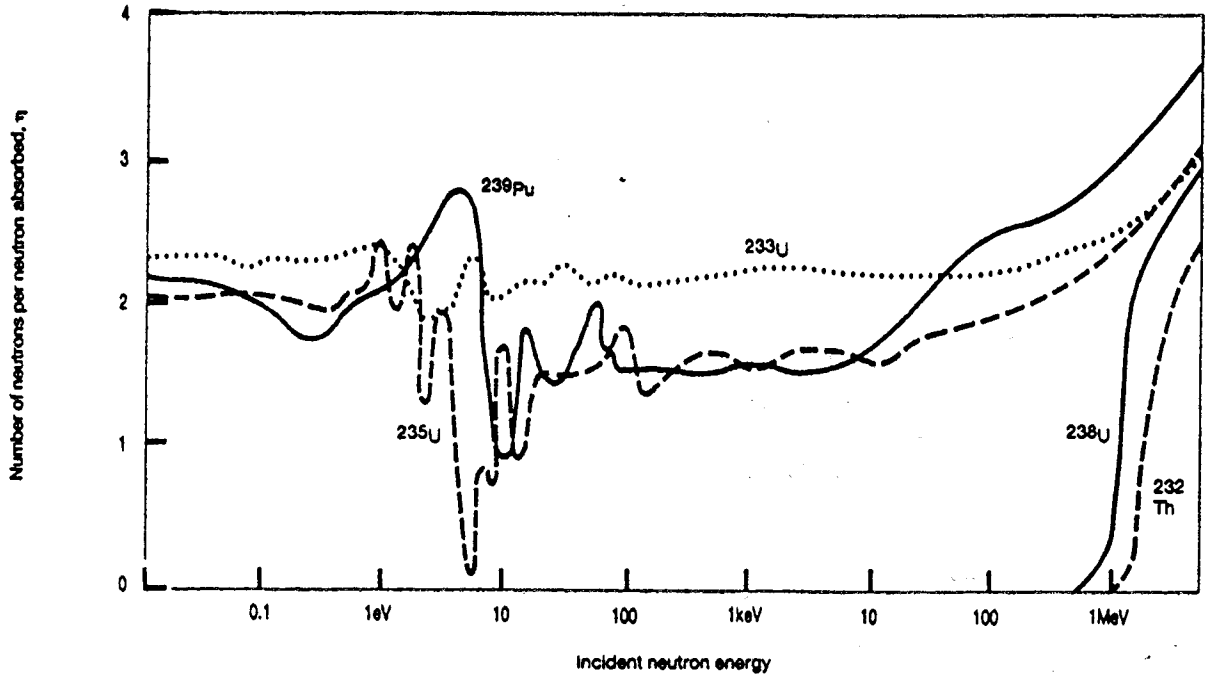


Figure 5: Neutron yield per neutron absorbed (NEA-10)

The neutronic properties of U-233 for fission energy can be summarized as follows :

- U-233 is the best neutronic fuel compared with U-235, Pu-239, Pu-241, due mainly to its low neutron capture.

- U-233 has the property of keeping a good fission capability with thermalized, but also with epithermal neutrons, which gives excellent characteristics for epithermal or fast reactors and thermal breeders.

- The minor actinides produced in a thorium/U-233 fuel cycle have much shorter decay chains than with U-238/U-235 or plutonium fuel cycles; this is an interesting feature from the waste management point of view.

- The good fissile properties of U-233 are such that only a small amount of the metal (about 5 kg) can be used to prepare a nuclear weapon. The small amount of spontaneous neutrons (compared to "industrial" or "reactor-grade" plutonium) would permit to manufacture rather simple "gun-type" weapons (cf. Chapter 7, Table 2 and Figure 18).

- The Th-232 and U-232 by-product decay chain is accompanied by some hard short-lived gamma emitters. This drawback is a quality from the non-proliferation standpoint, because it is difficult to distract even minor quantities of fissile material unnoticed.

Protactinium-233

The importance of Pa-233 lies in its half-life of 27 days. This time is long enough to be significant as a delay in the production of U-233 from Th-232. There are three disadvantages :

1) Capture in Pa-233 robs the cycle of a neutron.
2) Subsequently it robs the cycle of a fissile atom.
3) The delay in the production of fresh fissile atom has important effects on reactivity after shutdown of the reactor.

When a Pa-233 atom captures a neutron, the effective result is that a potential U-233 atom is no longer produced. The capture cross-section is not small : around 40 barns for thermal neutrons. As may be seen from Figure 2, Chapter 2.2.1., neutron capture in Pa-233 leads to Pa-234 which decays to U-234, both being neutron absorbers, giving fissile U-235. The losses to fissile isotope production that are incurred in this way depend on the relative rates of beta decay of Pa-233 (to U-233) and the rate of neutron absorption in Pa-233. While the former process is determined solely by the natural decay constant of the isotope,

the latter is directly dependent on the neutron flux level of the reactor. Fortunately the natural radioactive decay rate is generally much higher than the rate of neutron capture so that at typical fast reactor and thermal reactor (PWR) flux levels, only a few per cent of the potential U-233 production is lost. However, the higher losses associated with higher flux levels *may* detract from and ultimately limit the economic advantages which might be expected from designing for a compact core and high fuel rating (i.e. high power per unit mass of fuel).

The other important feature of Pa-233 is its effect after the reactor is shut down. In the days immediately following, shut-down of the decay of Pa-233 provides the main source of radioactivity from the actinide group, and this activity is a significant proportion (up to 30 %) of the total. The isotope decays with the emission of a beta particle and of gamma rays. A further effect is that after the reactor is shut down the U-233 content increases and the capture probability decreases as the proto-actinium decays. This effect means that the fuel becomes more reactive which may have implications for the control of the reactor.

Uranium-232

The main importance of the isotope U-232 is that it is at the head of a decay chain, some of whose members emit gamma rays of high energy. The decay chain is shown in Figure 1 of Chapter 2.1. All the members of the chain have very short half-lives except for the first two, and Th-228 has a much shorter half-life than U-232. The most important of the gamma ray emitters is Tl-208, with 2.6 MeV gamma energy, and this energy is in a range for which it is particularly difficult to reduce the dose to acceptable levels. The gamma ray emission, coupled with the decay times involved and the impossibility of separating U-232 from U-233 by chemical processes, makes the presence of the U-232 a serious drawback to the use of the Th-232/U-233 fuel (See also Chapters 6.4.2. and 7.3.2.). U-232, besides, is a spontaneous neutron emitter (cf. Table 2, Chap. 7). It is fortunate, however, that the production of U-232 depends on (n,2n) reactions, which are of low probability, but it is cumbersome enough.

In a typical fast reactor, the (n,2n) cross-section of U-233 is lower than the neutron capture cross-section by a factor of about 400. In a thermal reactor the (n,2n) cross-section is relatively smaller still. The net result is that in a Th-232/U233 fuelled fast reactor, the discharge fuel contains less than 0.1 % (1000 ppm) of U-232, though if the U-233 is recycled, then the U-232 content increases.

The cross-sections for the different isotopes mentioned, for thermal neutrons (0.025 eV, 2200 m/sec) and for an average in the resonance region ("resonance integral"), are given in Table 3 (2).

Table 3: Comparative cross-section values (2)

		ISOTOPE											
		Th-232	Pa-233	U-233	U-234	U-235	U-236	U-238	Np-239	Pu-239	Pu-240	Pu-241	Pu-242
	THERMAL DATA												
Absorption	σ_a (0.025 eV)	7.40	41.46	571.01	95.77	678.40	6.00	2.73	80.00	1013.04	290.08	1375.37	30.00
Capture	σ_c (0.025 eV)	7.40	41.46	45.99	95.77	101.30	6.00	2.73	80.00	271.19	290.02	367.81	30.00
Fission	σ_f (0.025 eV)	0.00	0.00	525.11	0.00	577.10	0.00	0.00	0.00	741.85	0.06	1007.56	0.00
	α	--	--	0.0874	--	0.1755	--	--	--	0.3656	--	0.3651	--
	ν	--	--	2.498	--	2.442	--	--	--	2.880	--	2.936	--
	η (ETA)	--	--	2.300	--	2.077	--	--	--	2.109	--	2.151	--
	INFINITELY DILUTE RI (barns) 0.625 eV-10 MeV (Resonance Integral)												
	ABSORPTION	85.78	858.83	883.73	632.16	380.13	348.82	273.57	0.00	445.15	8494.02	686.76	1118.65
	CAPTURE	85.20	857.00	135.10	627.96	130.22	346.55	272.37	0.00	168.58	8486.17	112.41	1115.00
	FISSION	0.58	1.83	748.63	4.20	249.91	2.27	1.20	0.00	276.57	7.85	574.35	3.65
	α	--	--	0.1805	--	0.5210	--	--	--	0.6096	--	0.1957	--

LEGEND : $\alpha = \frac{\sigma c}{\sigma f}$ ν = average neutrons per fission ETA value $\eta = \frac{\nu}{1 + \alpha}$

2.2.3. **Theoretical analysis of the Th-derived isotopes behaviour in a reactor: a parallel with uranium** (Remarks by ESCHBACH and Al) (19)

NOTE : this part can be skipped if interest in the neutron physics is limited, but we have kept it because it gives a somewhat additional lighting on the subject.

In Fig.1, we notice the striking parallel that exists between thorium and U-238 series. Both series begin with an abundant fertile material, and, when Th-232 and U-238 capture a neutron, they form a new isotope that beta decays to form a fissile isotope (U-233 or Pu-239). Thereafter, each series has the same rhythm of fertile isotope, fissile isotope, and parasite, which terminates the chain when it captures a neutron to form a chemically separable isotope.

If we consider more closely the isotopic properties in each series, we find some significant differences. Uranium-238, for example, has a much larger fast fission cross-section, or fast effect, than thorium. The fast effect is important because it produces both neutrons and heat from an inexpensive material. Now when U-238 captures a neutron, it forms Np-239; and, when thorium captures a neutron, it forms Pa-233. Here again we observe a difference : Pa-233 has a much longer half-life than Np-239 and also has a larger absorption cross-section, which includes a sizeable resonance integral. So the probability of Pa-233 absorbing a neutron to form U-234 instead of U-233 is much greater than the probability of Np-239 absorbing a neutron to form Pu-240 instead of Pu-239. This probability of Pa-233 forming non-fissile U-234 instead of fissile U-233 becomes a serious problem if the flux level is very high or if the neutron spectrum is very hard.

The next isotopes in the series, U-233 and Pu-239, are both fissile, but their characteristics are also distinctly different. In the thermal-neutron spectrum, U-233 has a much lower alpha ($\alpha = \sigma c/\sigma f$) and therefore a higher eta [$\eta = \nu/(1+\alpha)$] than Pu-239. This difference in eta, however, is not as large as the difference in alpha might suggest because Pu-239 has a larger ν, or a larger number of neutrons produced per fission. Our next observation is that Pu-239, because it has a higher alpha, produces 3.5 times more of the fertile isotope Pu-240 than U-233 produces of the fertile isotope U-234. Also, when comparing the fertile materials Pu-240 and U-234, we find that Pu-240 has a much larger cross-section, possibly making it the more valuable of the two fertile isotopes.

After each of these two fertile materials absorbs a neutron, two new fissile isotopes are formed, namely, U-235 and Pu-241. Here the difference in alpha is small, and it seems to favour the thorium series. The difference in ν, however, causes Pu-241 to have the larger eta; and this larger eta of Pu-241 over U-235

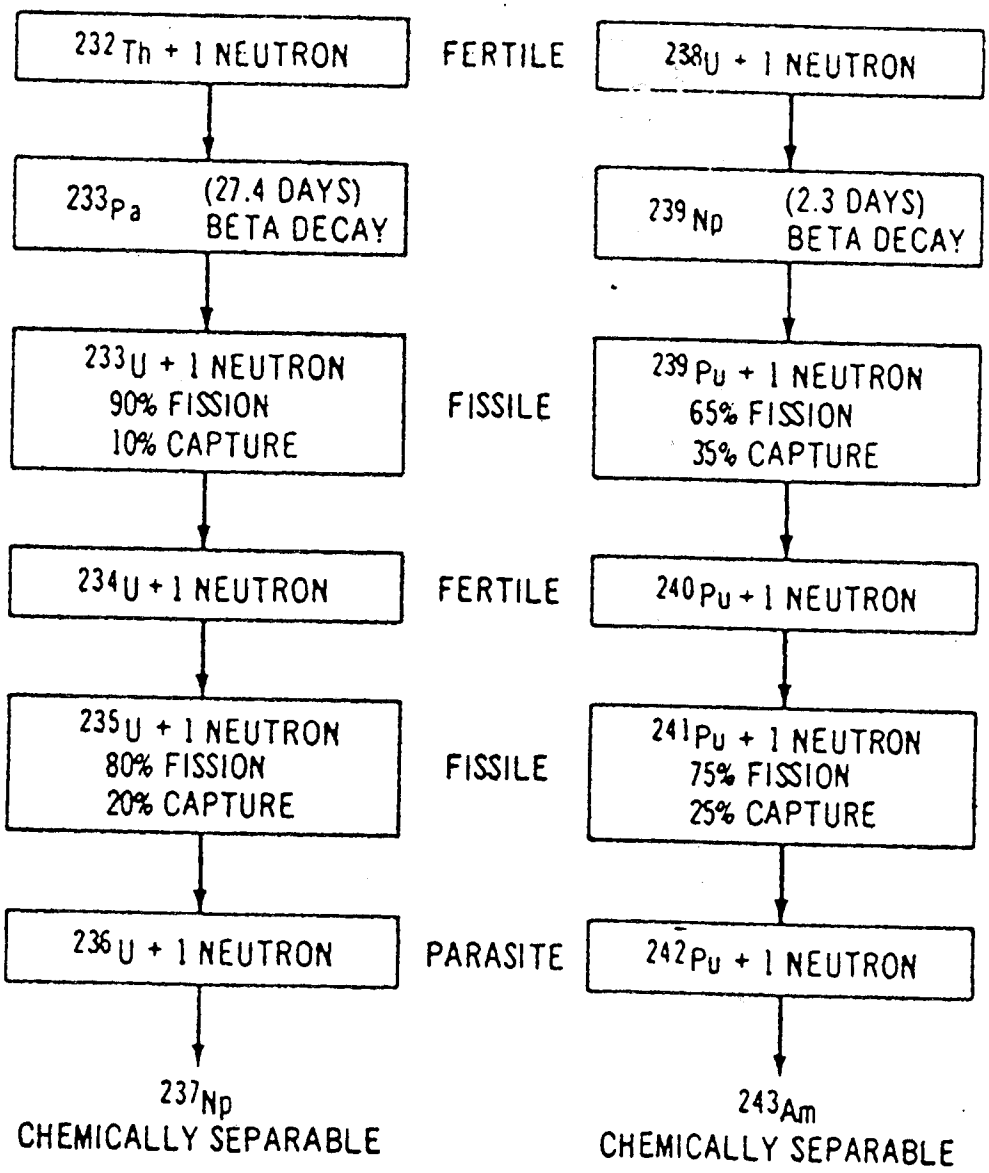


Figure 1: Isotopic build-up in Th-232 and U-238 systems (19)

tends to reduce the net effect of the larger eta of U-233 over Pu-239. Moreover, Pu-241 has a larger cross-section than U-235. However, the 13-year half-life of Pu-241 means that some Pu-241 will be lost by decay. As for the percentage of fissions to the percentage of captures in thermal reactors, Pu-241 forms Pu-242 about 25 % of the time, whereas U-235 forms U-236 about 20 % of the time. Both U-236 and Pu-242 are parasites because they require the absorption of still another neutron to produce another non-fissile, nonfertile isotope. These non-fissile, nonfertile isotopes, Np-237 and Am-243, are both chemically separable and terminate their respective series. Even though we have described U-236 and Pu-242 as parasites, it should be noted that both of them would become valuable if a market ever arose for transuranium heat-source isotopes. In that event Np-237 and Am-243 targets would be irradiated to form Pu-238 and Cm-244, respectively.

Now, let us take an even more detailed look at the two sets of fissile isotopes as shown in Table 4.

Table 4

ETA OF FISSILE ISOTOPES FOR VARIOUS REACTOR SPECTRA

Neutron temperature, °C	Spectral index r	^{233}U	^{235}U	^{239}Pu	^{241}Pu
100	0.05	2.27	2.06	2.06	2.24
	0.15	2.26	2.01	2.00	2.24
	0.25	2.23	1.97	1.96	2.23
	0.35	2.22	1.94	1.93	2.22
400	0.05	2.26	2.04	1.95	2.20
	0.15	2.24	1.98	1.92	2.20
	0.25	2.22	1.94	1.90	2.20
	0.35	2.20	1.89	1.88	2.20
1000	0.05	2.25	2.01	1.87	2.12
	0.15	2.23	1.95	1.87	2.15
	0.25	2.20	1.89	1.87	2.17
	0.35	2.19	1.83	1.86	2.20

In particular, let us consider the effect of the neutron spectrum on eta, which is the ratio of the neutrons produced to the neutrons absorbed. Note that the table shows the computed eta values for four spectral indexes and three neutron temperatures. The spectral index of 0.05 represents a well-moderated reactor; the spectral indexes of 0.15 and 0.25, typical power reactors; and the spectral index of 0.35, a power reactor using a highly enriched fuel. When considering temperatures, we should keep in mind that the neutron temperature of 100°C relates to a heavy-water-moderated reactor (HWR), the 400°C neutron temperature relates to a light-water-moderated reactor (LWR), and the 1000°C temperature relates to a high-temperature gas-cooled reactor (HTGCR).

Looking at the isotope U-233, we see that its eta changes little when either the spectral index or the neutron temperature increases. This is in marked contrast with Pu-239, whose eta changes considerably when either the temperature or the spectral index increases. As for the eta of U-235, we see that it has only a small reduction when the temperature increases but a significant reduction when the spectrum hardens. It should be pointed out, however, that the concentrations of U-235 used in these calculations are typical of the dilute quantities produced from U-234 captures. These dilute U-235 quantities are only one-tenth of the U-235 concentrations in light-water moderated reactor fuels. Thus the self-shielding of the U-235 resonance here is very small when compared to the self-shielding in U-235 enriched fuel.

Plutonium-241 is the second fissile plutonium isotope shown in Table 4. Although some uncertainties still exist, our data for Pu-241 indicate a very small eta variation when the temperature or the spectrum changes. The one exception is in the 1000°C neutron-temperature range. The reason the Pu-241 eta increases in this range is that a higher percentage of the reactions are taking place above the 0.3-ev resonance.

From what has been shown thus far, we can conclude, without considering special fuel configurations, that plutonium performs best in a soft spectrum and at a low neutron temperature, two characteristics that are typical of HWR designs. We can conclude further that, since spectrum changes do not affect remarkably the eta of U-233, the reactor in which U-233 is used could maximize some other fuel or reactor characteristic. For example, U-233 used in an HTGCR would allow the thermal-to-electrical efficiency of that reactor to be maximized, or U-233 used in a tight-lattice LWR would allow the U-238 fast effect to be maximized. Additional comments : (19)

Table 5 (19)

FATE OF 100 ATOMS OF ^{233}U OR ^{239}Pu IN OPTIMIZED REACTOR SYSTEMS WITHOUT NEUTRON LOSSES

	$^{233}U-Th$ in an optimized HWR (ϵ = 1.01)					$Pu-^{238}U$ in an optimized LWR (ϵ = 1.05)			
	Atoms	Neutrons in	Neutrons out	Heat, %		Atoms	Neutrons in	Neutrons out	Heat, %
^{232}Th	120	120	2(net)	2	^{238}U	99	99	13(net)	8
^{233}Pa	120	2			^{239}Np	99	0		
^{233}U formed	118*				^{239}Pu formed	99*			
^{233}U	100	100	223	89	^{239}Pu	100	100	190	67
^{234}U	11	11			^{240}Pu	34	34		
^{235}U	11	11	19	9	^{241}Pu	33	33	72	25
^{236}U	2	2			^{242}Pu	9	9		
		124	242				176	262	

*Initial conversion ratio is 1.18 for $^{233}U-Th$ and 0.99 for $Pu-^{238}U$.

$$\eta\epsilon = \left(\frac{242}{100 + 11}\right)(1.01) = 2.20 \text{ for } ^{233}U-Th$$

$$= \left(\frac{262}{100 + 33}\right)(1.05) = 2.07 \text{ for } Pu-^{238}U$$

Table 5 shows both the fate of 100 atoms of U-233 enriching thorium in an idealized HWR and the fate of 100 atoms of Pu-239 enriching uranium in an idealized LWR. If we examine the box on the left, we see that 100 neutrons absorbed in U-233 produce 223 neutrons, forming at the same time 11 atoms of U-234. In its turn U-234 absorbs 11 neutrons that produce 11 atoms of U-235. These U-235 atoms absorb an additional 11 neutrons, 9 of which cause fission, producing 19 more neutrons. The two U-236 atoms formed from the two U-235 captures absorb two more neutrons to terminate the series. Thus there is a total of 124 neutrons absorbed and 242 neutrons gained, leaving an excess of 118 neutrons for absorption in Th-232, which, as shown at the top of the left-hand column above the box, also absorbs two neutrons from the thorium fast effect. Since Pa-233 captures two neutrons, the potential conversion ratio of a U-233-Th fuel when there are no neutron losses is 118/100, or 1.18.

Turning now to the other box, in which the fate of 100 Pu-239 atoms is shown, we obtain 262 neutrons from 176 neutrons absorbed; leaving an excess of 86 neutrons to be absorbed in U-238. Because of the U-238 fast effect, 13 more neutrons become available to form 99 atoms of new Pu-239. Thus the potential conversion ratio for the idealized Pu-U-238 system is 0.99.

When the two idealized conversion ratios are compared, it is evident that the U-233-Th system, in a HWR, has a potential conversion ratio 20 % higher than that of the Pu-U-238 system in a LWR. As for the percentage of heat produced, we see that U-233 produces nearly 90 % of the heat in U-233-Th fuel, whereas Pu-239 produces only 67 % of the U-235-U-238 fuel.

2.2.4. **Other remarks** (A. RADKOWSKI) (2)

Uranium-238 has a much higher fast effect (direct fission by fast neutrons) than thorium. This is offset somewhat by a higher (n,2n) cross-section for thorium. Uranium-233 has a lower fission product absorption than U-235, which also must contend with the high parasitic capture of U-236. All these factors must be taken into consideration in an accurate calculation.

As a result of a composite of these effects, it turns out that for a given neutron input the energy obtained from thorium is less than from uranium at short irradiations but much higher for long irradiations (Fig. 7). Fortunately, thorium

oxide can withstand much longer irradiations than uranium oxide. Thus the key to obtaining energy gains from thorium is a core arrangement in which the thorium can be utilized for very long periods of time.

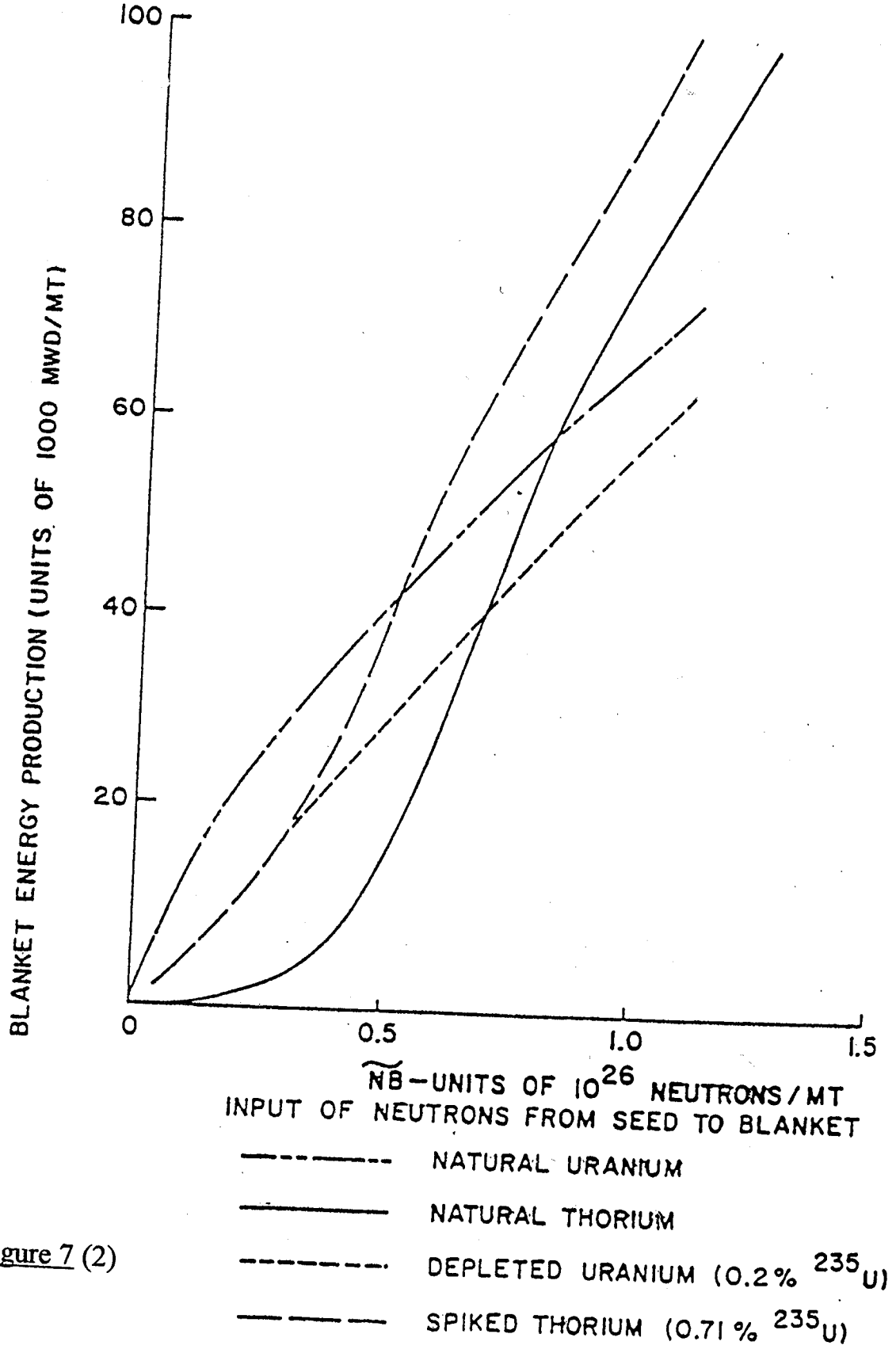


Figure 7 (2)

2.3. Theoretical Application to Nuclear Reactors

NOTE : we shall again revert to THORN and al (176) for the clarity of this Chapter.

2.3.1. **Thermal reactors**

As already mentioned, the neutron flux spectra differ significantly for the various thermal reactors. However, in comparing the thorium-based fuel cycles with the uranium/plutonium cycles, the physics parameters differ in the same basic way, though to a varying degree, for all the reactors. Table 1 gives the 2200 m/sec cross-sections and the resonance integrals of the main fissionable isotopes for thermal reactors.

Table 1: CONDENSED NUCLEAR DATA FOR FISSILE ISOTOPES IN THERMAL REACTORS

	U-233	U-235	Pu-239
Cross-section (barns)			
Fission			
(1) 2200 m/sec value	527	579	741
(2) Resonance integral	764	275	301
Capture			
(1) 2200 m/sec value	54	100	267
(2) Resonance integral	140	144	200
Neutron/fission (on average)	2.5	2.4	2.9
Eta (1)	2.27	2.04	2.12
Eta (2)	2.11	1.56	1.74

If we compare the nuclear data of Table 1 it may be seen that the advantage of U-233 over the other two lies in its low neutron capture cross-section. Thus for a given number of fissions (which equates with energy production) there is a much

smaller total destruction of primary fissile material for this isotope than for either of the other two. After account is taken of the average number of neutrons per fission this same feature leads to a greater number of neutrons being contributed to the chain reaction from the destruction of each fissile atom and this is so even as compared with the Pu-239 isotope for which the number of neutrons yielded per fission is the greatest.

The presence of Pa-233 as an intermediate product between the fertile Th-232 and fissile U-233 detracts from this advantage by an amount depending on the compactness of the core design adopted. This is of particular importance in thermal reactor conditions in which the economics and logistics of the thorium fuel cycle push reactor optimisation towards high ratings where the loss of neutrons and the subsequent loss of new fissile material begin to increase rapidly. This same feature leads to marked transients in core reactivity during sustained shutdown or part-load operation and thus to problems of fissile and control rod inventory at the high ratings that are desirable.

The advantages of Pu-239 are its high fission cross-section and its high release of neutrons per fission. The high fission cross-section means that fewer neutrons are lost in unproductive capture in structural materials and other non-fissionable materials. The high neutron release of Pu-239 is important because it means that, as far as this effect is concerned, fewer atoms of Pu-239 are needed to sustain the chain reaction.

A further factor which is important in comparing Th-232/U-233 systems with uranium/plutonium systems is that the higher plutonium isotopes build up a greater extent than do the higher uranium isotopes from U-233. The cause is the higher capture cross-section of Pu-239 as may be seen from Table 1. Neutron capture in Pu-239 leads to the new isotope Pu-240 which itself has a high capture cross-section in thermal reactors. Thus as irradiation proceeds the probability of neutron capture increases rapidly and the amount of fissile material necessary to provide sufficient reactivity to sustain long irradiations is correspondingly increased.

As far as U-235 is concerned, it may be seen from Table 1 that this isotope is in an intermediate position between the other fissile isotopes since, although it has a higher fission cross-section than U-233, it also has a higher capture cross-section. Unfortunately, unlike U-233 and Pu-239, successive neutron capture in U-235 does not lead eventually to further isotopes which are readily fissile. Thus

neutrons and fissile atoms lost in capture by U-235 can be regarded as permanently lost.

A comparison involving U-233 and Pu-239 is of course incomplete without consideration of their original production, and the properties of their parent materials. There is no unique combination of the fuel parameters in Table 2 of Chapter 2.2.2. and Table 1 of this Chapter which can be taken as a universal figure of merit. Even within the subject matter of this section, at least two separate considerations arise of economic significance : the inventory of fissile material held in the reactor, and the production rate of new fuel compared with fuel consumption. Furthermore, both of these are dependent on the reactor design as well as on the properties of the fuel itself. As a summing up of the situation for thermal reactors, it is nevertheless broadly true to say that the thorium/U-233 cycle may well require a higher fissile inventory but is likely to consume less fissile material, net, per fission.

This is shown in a Canadian study (147) adapting the thorium fuel cycle to the CANDU, Fig.1. Similar results are found in Indian studies (7).

2.3.2. **Fast reactors**

Table 2: FAST REACTOR ONE-GROUP NUCLEAR DATA FOR FISSILE ISOTOPES

	U-233	U-235	Pu-239
Cross-section (barns)			
Fission	2.8	2.0	1.9
Capture	0.3	0.5	0.6
Neutron/fission (on average)	2.5	2.5	2.9
Eta	2.26	2.0	2.20

Table 2 gives one-group data for a typical fast reactor. The fission cross-section of U-233 is considerably higher than the corresponding data for U-235 and Pu-239, and the capture cross-section is lower. Of the capture cross-sections,

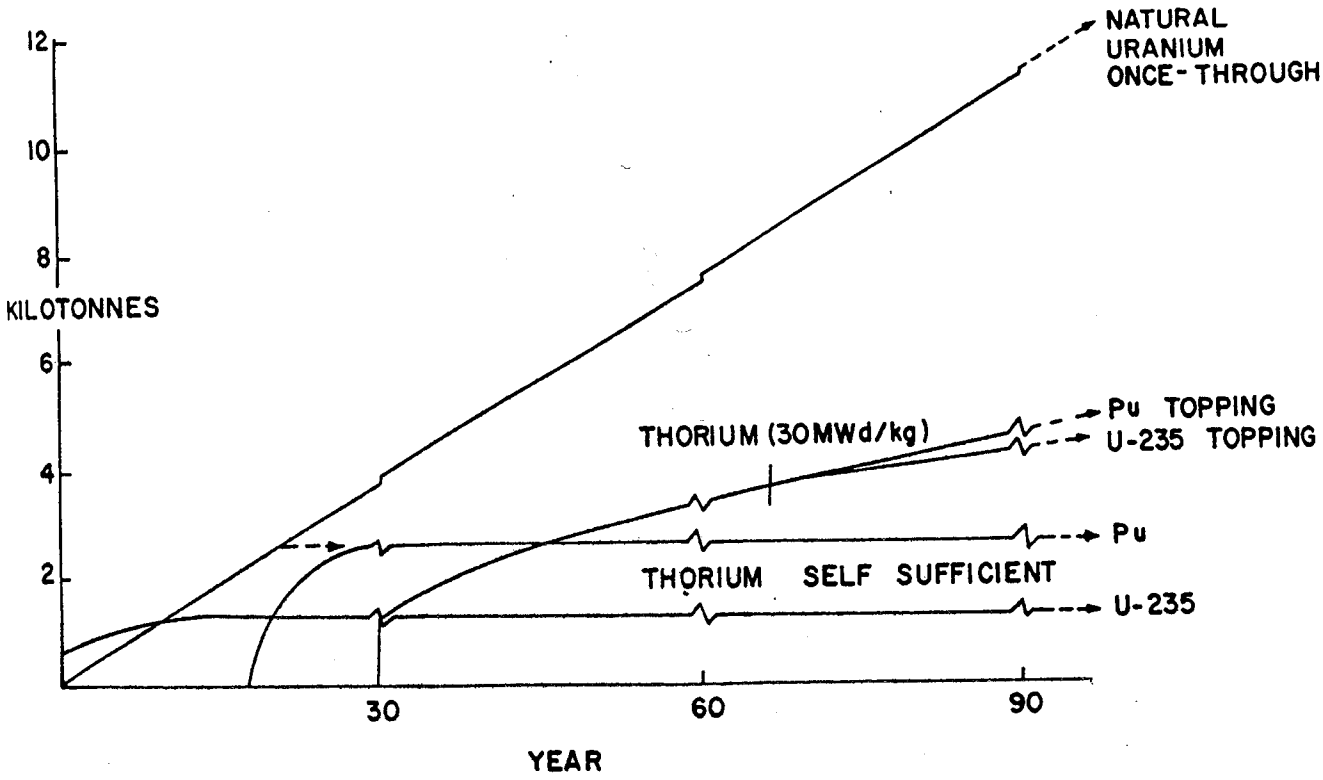


Figure 1: Cumulative uranium requirements - CANDU fuel cycles - Per Gwe at 72% capacity factor (147, Garvey).

Pu-239 has the highest value, but the difference is much reduced in comparison with the thermal reactor values. This change makes Pu-239 relatively more attractive as a fuel for use in fast reactors than in thermal reactors. U-233 again appears the most attractive from the data of Table 2. In the case of fast breeders, however, the overall production of new fuel compared with fuel consumption depends on contributions to the neutron balance from other sources. Unfortunately the apparent attractiveness of the U-233 isotope itself is more than offset by the low fission contribution from the fertile Th-232 isotope as compared with the much larger contribution from U-238 in fuel cycles which use that fertile isotope. Thus the Th-232/U-233 fuel cycle proves to be relatively unattractive and fast reactors studies suggest that a thorium/U-233 cycle would give about 20 % less bred fissile material than a U-238/plutonium cycle on a reasonably comparable basis.

2.3.3. **Conclusions from the above**

Among natural fuel cycles, the thorium cycle is the poorest in neutrons . Once U-233 has been bred, and separated, however, the thorium/U-233 has interesting features.

In comparing the fissile material utilisation of the Th-232/U-233 and the U-238/ plutonium fuel cycles in thermal reactors, a balance has to be struck between a number of opposing effects. The important advantageous feature of the thorium cycle is the low capture cross-section of U-233 compared with U-235 or Pu. The beneficial effect of this is countered by :

1) losses of neutrons in Pa-233 which is present as an intermediate product between the fertile Th-232 and the fissile U-233,

2) the relatively higher neutron capture cross-section of Th-232, from which the U-233 must be bred, compared with U-238,

3) the fact that the contribution to fission by the build up of higher isotopes resulting from neutron captures starting from U-233, is less useful for energy production than the equivalent process starting from plutonium.

The individual significance of these factors depends on the reactor type and on even more specific design features. (Please refer to Tables 2 and 3 above). For example, the Pa-233 effect is intensified by the use of compact core designs with high power density and could preponderate if this design objective is greatly emphasised. On the other hand, the use of a low degree of neutron moderation, whilst increasing the neutron capture in either Th-232 or U-238, has relatively less effect with the former. (cf. Table 2, resonance integral)

It can be concluded that if good fissile material utilisation is given top priority in selecting a reactor type and in determining its design, and that if that selection is confined to the range of thermal reactors, a thorium cycle is likely to provide the lowest consumption of fissile material though it may well require a relatively higher inventory, as said earlier.

If fast reactors are brought into the comparison however, at least from a breeder point of view, the disadvantages of Th-232 outweigh all that might be gained from the use of U-233, one consequence being that the achievable breeding gain is at best very small : in the uranium/plutonium cycle direct fission in U-238

makes a significant contribution (15-20 %) to the total number of fissions, whereas in the thorium cycle the contribution from direct fission in Th-232 is negligible.

2.4. Production of Long-lived Actinides

The problem of waste as a by-product of human activities has recently become a paramount factor in strategic decisions.

Spent nuclear fuel elements in a once-through cycle still contain more than 95 % of the long-lived heavy metal in a concentrated form compared to the original ore, plus about 3 % fission products, most of which are relatively short lived, plus about 0.1-0.3 % of higher "minor" actinides created by neutron absorptions within the reactor : Pa, Np, Pu, Am, Cm. Many of those have very long half-lives :

Pa-231	$3.2 \cdot 10^4$ y
Np-237	$2.1 \cdot 10^6$ y
Pu-239	24 000 y
Pu-240	6 600 y
Am-241	458 y
Am-243	7 900 y
Cm-245	9 000 y

Besides, some higher actinides, usually alpha emitters, are also neutron emitters by spontaneous fission.

By reprocessing, fertile and fissile material are recovered, i.e. Th, U, Pu are recovered. The quality of reprocessing processes today warrants very high recovery yields, i.e. the "waste" will contain only 0.1-0.2 % of these heavy metals, i.e. 1 or 2 kilograms H M per ton, compared with the original spent fuel.

It is appropriate to note here that the thorium fuel cycle is justified by reprocessing, so that the total minor actinide content of the "waste" will, in all cases, be very limited.

So far, the waste in a reprocessing plant, lumps together the fission products and the minor actinides. Usually, today, all of those are concentrated and vitrified, and the intention is to store these glass blocks deep underground in specially-built repositories with surrounding clay to absorb any leaking material.

Table 1 below shows the fate of some of these longer-lived nuclides in the waste.

Table 1

ISOTOPE	Half-life (y)	Emission	Fraction % remaining after years			
			500	1 000	10 000	100 000
Fission products						
Technetium-99	$2.1\text{-}10^5$	0.3 MeV-β	100	100	97	72
Iodine-129	$1.7\text{-}10^7$	0.1 MeV-β	100	100	100	100
Actinides						
Plutonium (all)	$10^3\text{-}10^5$	α,β, low γ, n	88	85	58	8
Neptunium-237	$2.1\text{-}10^6$	α, 0,09 MeV γ	100	100	100	97

It has been proposed to isolate the minor actinides to shorten their residual life, and "incinerate" them by transmutation in appropriate devices (accelerators, fast reactors, fusion reactors).

The long-lived fission products pose the same problems with thorium as with the uranium fuel, but possibly less than the minor actinides in the longer term, in spite of the higher environment mobility of Tc-99, I-129. This is due to their small relative concentration, for example compared to naturally occurring K-40 (β emitter 1.3 MeV, $1.2\text{-}10^9$ years). (cf. Table 1, and also Chap. 4.4. (50)).

However, it would seem from a comparison of the fission products yields from U-233 compared with U-235, U-238 and Pu-239, that the production of Kr-85 is noticeably enhanced in the thorium fuel cycle, compared with that of the uranium cycle, by a factor of about two.

It is worthwhile mentioning that the thorium fuel cycles, due to their being 2 neutrons below the uranium cycles, are producing considerably less long-lived minor actinides (conversely, Pu fuels are producing more, for the reverse reason)

The results of preliminary studies, carried out in Japan and India for different uranium and thorium-based fuel combinations and for a 1300 MWe PWR, are presented in the following Table 2.

It can be observed that the Th-232/U-233 ("U3+Th2") cycle produces less neptunium than the U-238/U-235 ("U5+U8") cycle, but also much less other minor long-lived actinides.

This property to produce less long-lived actinides is certainly a plus in favour of the thorium cycle. How much this "plus" amounts to, remains to be evaluated.

Table 2: Relative production of minor actinides in diverse cycles (174 - IAEA 1990)

MINOR ACTINIDES :	DIVERSE FUEL TYPES			
	U5+U8 (1) ***	U5+Th2 %	U3+U8 %	U3+Th2 %
Np237	3.6E-2 * 9.0E-2 **	92 107	20 13	1 3
Am (241+242M+243)	1.6E-2 * 4.7E-2 **	0.04 0.28	106 117	6.3E-5 1.8E-3
Cm (243 TO 246)	3.6E-1 * 2.2E-2 **	0.01 0.14	111 132	1.67E-5 6.36E-4

* Discharge burn up = 30 GWd/t
** Discharge burn up = 60 GWd/t
*** Reference cycle used to normalize the results of other cycles

(1) g/t HM

This table also shows that Np isolation might be a useful step in a more "advanced" reprocessing technique, although its biotoxicity might be rather small due to its very long half-life.

In order to permit useful comparisons, however, Figure 1 recalls the relative activities of U-238/U-235 spent fuel with time compared to usual uranium ores (47). Figure 3, Chapter 4, shows a comparison of radiotoxicities of spent fuel between uranium and thorium fuel cycles : there would be an advantage for thorium only before 10 000 years, after the effect is reversed, because of the

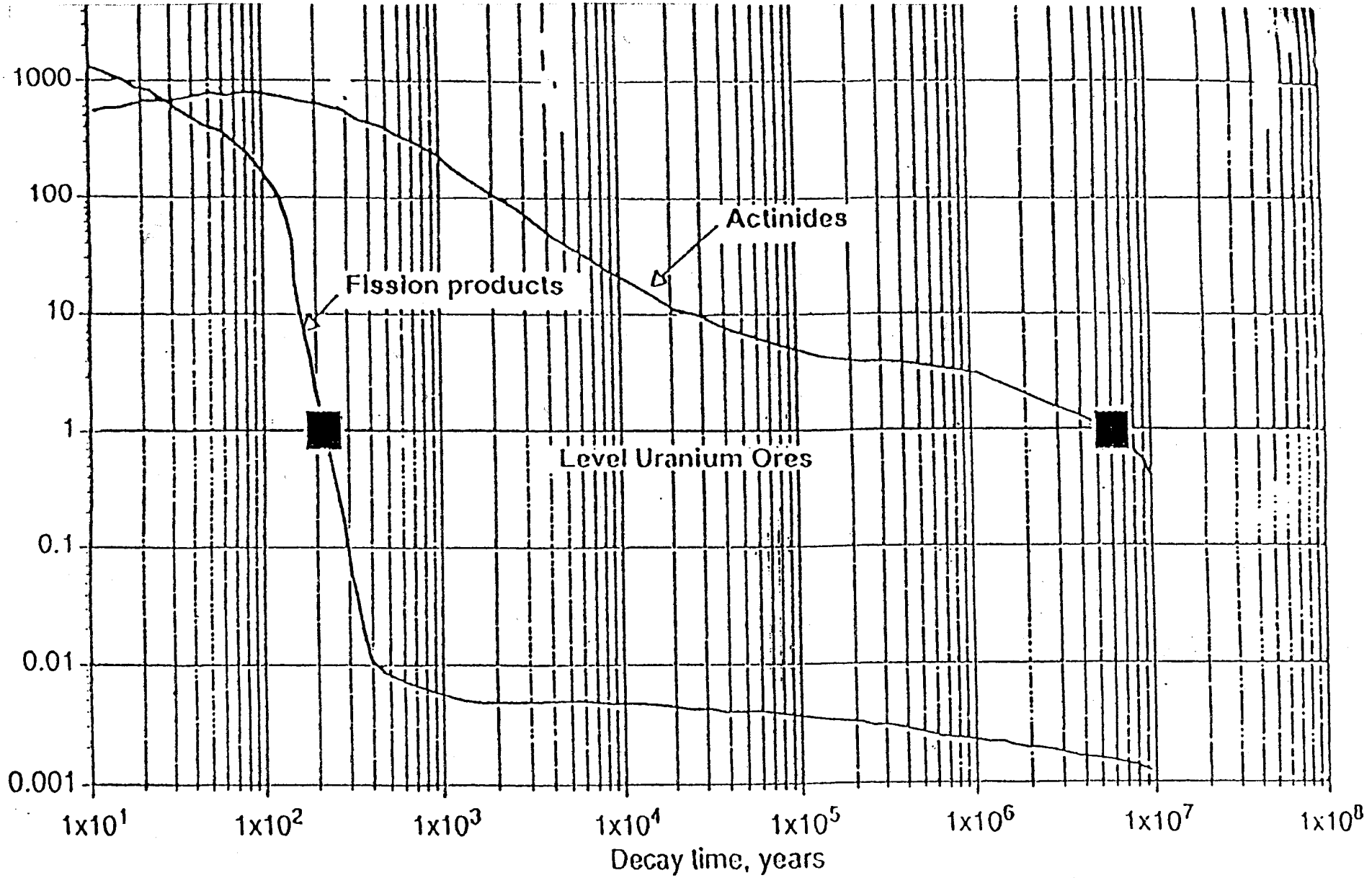


Figure 1: Typical PWR spent fuel radioactivity evolution with time compared to uranium ores radioactivity level

daughter products of U-233, but this might not be very important, in view of the low activities remaining.

Notes

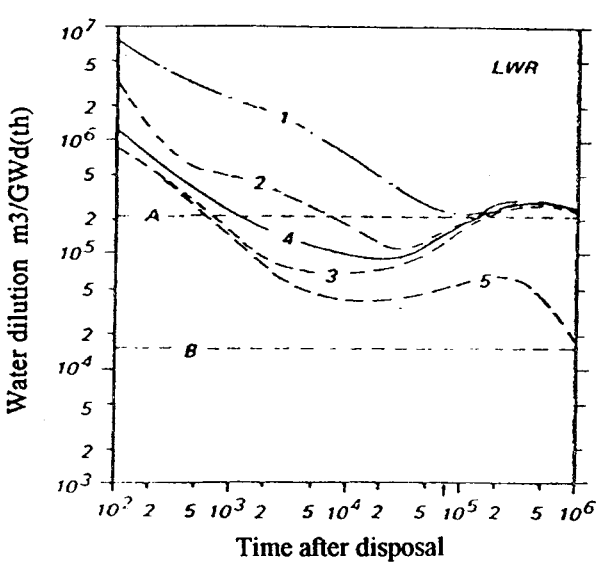
(1) It will be useful to draw the same curve, after reprocessing, of the residual activity of the vitrified glasses for different cycle types, once only the fission products and the minor actinides are left: then the picture changes, as can be seen also in Fig. 3, Ch. 4. (See further discussion on the subject in Chapters 4 (42) and 7 (130, 132). We give herewith the conclusions arrived at by H.J. Rütten in 1980 (Fig. 2) (136), which show indeed some advantage for thorium cycles, especially if supplementary separations of actinides could be performed, which is not yet proven.

(2) Indeed, the relevance of the threat posed by the long-lived minor actinides in the High Level Waste repositories is disputable. According to the EC PAGIS Study, there would be no radiological consequence whatsoever from the repository before 400 000 years, and after 1 million years the maximum exposure above the repository would be $2.2 - 10^{-7}$ Sv/yr (to be compared to the 1.10^{-3} Sv/yr authorized by the ICRP for the populations). By removing the actinides, a factor 2 would be achieved : $1.2 - 10^{-7}$ Sv/yr.

(3) To enter into more detail, please refer to the following figures and tables (T. Naka and T. Takeda) (142) :

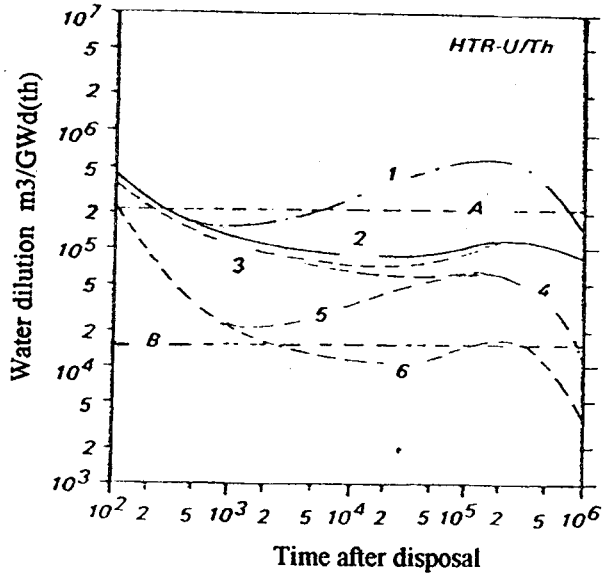
Isotope formation and decay. (Fig.3)
Actinides production. (Table 3)
Nuclide build-up as burnup in the reactor. (Figs. 4 and 5)

(Comparison is made with natural uranium ore : A) 70% Pechblende, B) 7% Pechblende).



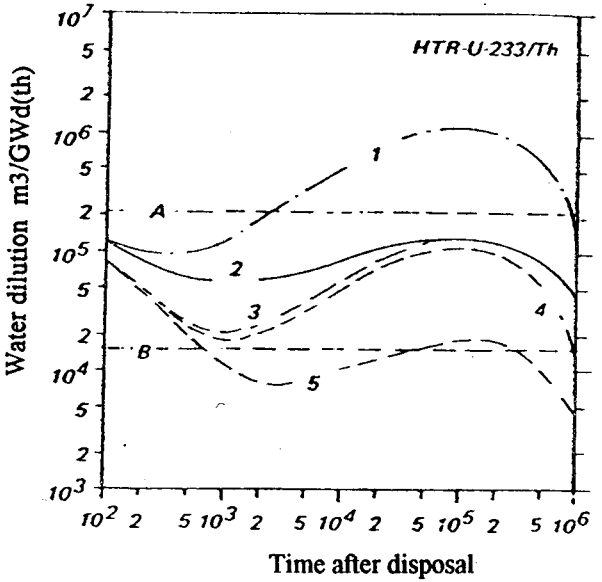
2.1. LWR U-Pu cycle according to different reprocessing strategies:

1. Recovery of 99% U and 99% Pu
2. Additional separation of 99%Am
3. Additional separation of 99%Cm
4. Simult. separation of Am and Cm
5. Additional separation of 99% Np



2.2. HTR U-Th cycle according to different reprocessing strategies:

1. Recovery of 99% U+Pu+Th
2. Recovery of 99.9% U, 99% Pu+Th
3. Additional separation of 99% Pa
4. Additional separation of 99% Np
5. Additional separation of 99% Am
6. Additional separation of 99.99%U



2.3 HTR U233-Th cycle according to different reprocessing strategies:

1. Recovery of 99% U+Pu+Th
2. Recovery of 99% U, 99% Pu+Th
3. Additional separation of 99% Pa
4. Additional separation of 99% Np
5. Additional separation of 99.99% U

Figure 2: Comparative toxicities (water dilution volumes) for traditional and HTR fuel cycles (136)

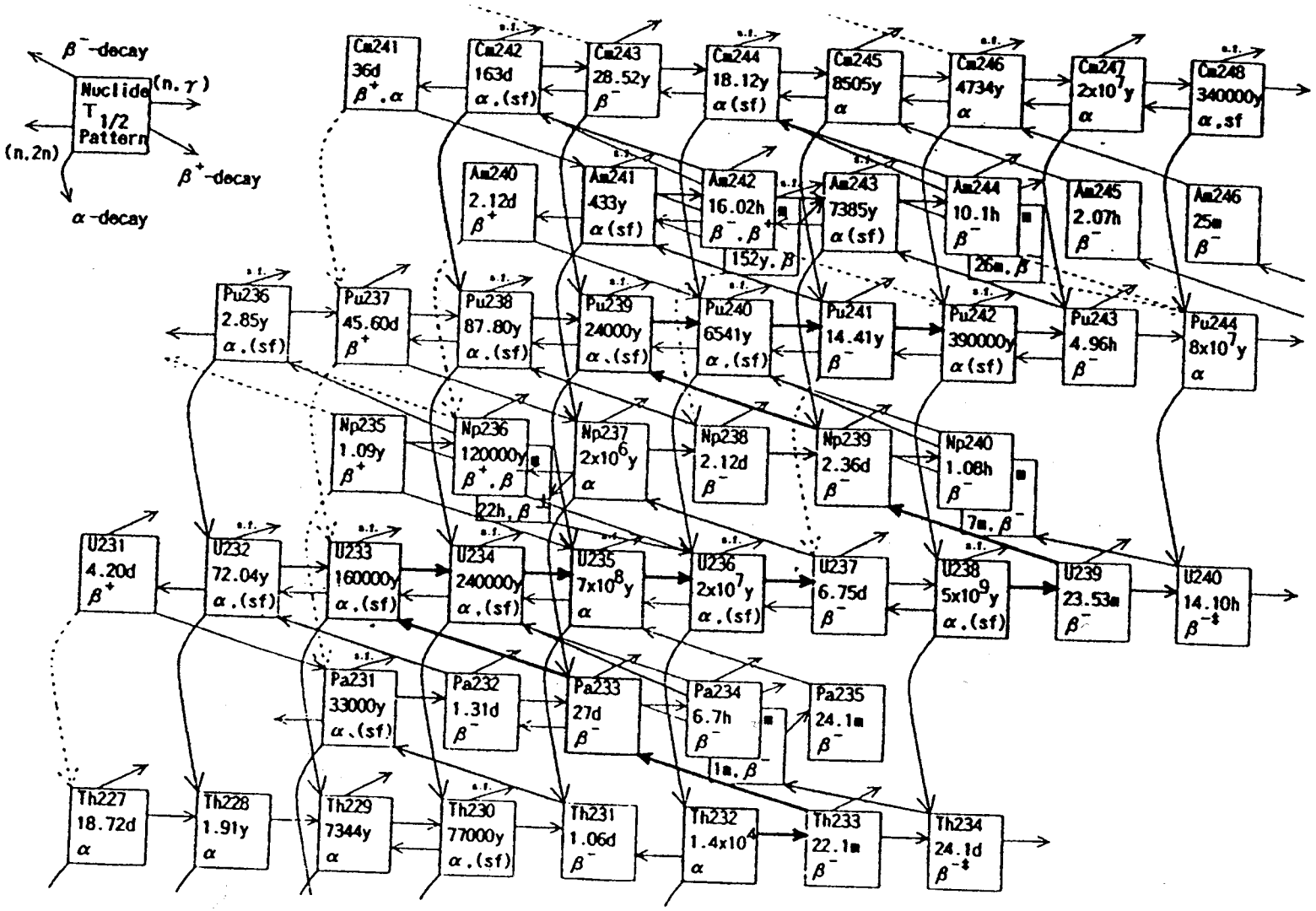


Figure 3: Actinides burn-up chain in the ORIGEN2 (142)

Table 3: Production of actinides (g/t) during burn-up (142)

Fuel type		Discharge burnup of 30GWd/t U235+U238	U235+Th232	U233+U238	U233+Th232	Discharge burnup of 60GWd/t U235+U238	U235+Th232	U233+U238	U233+Th232
Nuclide	Th232	2.1E-04	-2.7E+04	8.8E-02	-3.0E+04	7.3E-04	-4.7E+04	1.7E-01	-5.2E+04
	Pa231	1.7E-05	5.6E+01	6.0E-03	5.1E+01	2.3E-05	6.1E+01	2.7E-02	6.1E+01
	Pa233	1.2E-05	2.2E+01	2.4E-06	2.5E+01	3.0E-05	2.1E+01	4.1E-06	2.3E+01
	U232	2.8E-04	4.5E+01	2.8E-01	4.4E+01	1.5E-03	8.6E+01	4.8E-01	8.9E+01
	U233	2.8E-04	1.4E+04	-1.7E+04	-5.6E+03	7.8E-04	1.6E+04	-2.9E+04	-1.7E+04
	U234	1.6E+00	1.6E+03	1.2E+03	3.6E+03	7.1E+00	3.3E+03	1.5E+03	5.2E+03
	U235	-2.0E+04	-2.5E+04	2.8E+02	4.8E+02	-3.8E+04	-4.2E+04	5.9E+02	1.6E+03
	U236	3.2E+03	4.4E+03	3.6E+01	4.8E+01	5.8E+03	6.6E+03	1.6E+02	3.4E+02
	U238	-2.4E+04	4.0E+00	-2.5E+04	3.0E-02	-4.3E+04	1.5E+01	-4.8E+04	4.0E-01
	Np237	3.6E+02	3.3E+02	7.1E+01	2.4E+00	9.0E+02	9.6E+02	1.2E+02	2.7E+01
	Pu238	1.2E+02	8.7E+01	4.4E+01	3.7E-01	4.7E+02	4.4E+02	1.5E+02	7.4E+00
	Pu239	5.0E+03	1.0E+01	5.1E+03	3.6E-02	5.9E+03	7.3E+01	5.9E+03	1.0E+00
	Pu240	2.2E+03	2.0E+00	2.3E+03	5.9E-03	2.9E+03	2.5E+01	2.9E+03	2.8E-01
	Pu241	1.3E+03	1.0E+00	1.3E+03	2.3E-03	1.9E+03	1.2E+01	2.0E+03	1.1E-01
	Pu242	4.9E+02	1.6E-01	5.3E+02	2.9E-04	1.2E+03	3.8E+00	1.3E+03	2.6E-02
	Am241	6.1E+01	3.9E-02	6.1E+01	7.5E-05	1.2E+02	5.6E-01	1.2E+02	4.5E-03
	Am242M	7.8E-01	2.9E-04	7.6E-01	4.4E-07	2.0E+00	7.5E-03	1.9E+00	5.1E-05
	Am243	9.6E+01	1.8E-02	1.1E+02	2.6E-05	3.5E+02	7.1E-01	4.3E+02	3.9E-03
	Cm243	3.8E-01	5.6E-05	4.1E-01	6.9E-08	5.2E-01	1.0E-03	5.9E-01	5.1E-06
	Cm244	2.7E+01	2.8E-03	3.1E+01	3.1E-06	1.9E+02	2.3E-01	2.5E+02	1.0E-03
	Cm245	9.9E-01	8.7E-05	1.2E+00	7.9E-08	1.0E+01	1.1E-02	1.4E+01	4.1E-05
	Cm246	1.2E-01	5.8E-06	1.5E-01	4.9E-09	2.1E+00	1.6E-03	3.4E+00	5.4E-06
Fuel materials	Th	2.1E-04	-2.7E+04	8.8E-02	-3.0E+04	7.3E-04	-4.7E+04	1.7E-01	-5.2E+04
	U	-4.1E+04	-5.0E+03	-4.0E+04	-1.5E+03	-7.5E+04	-1.7E+04	-7.4E+04	-9.9E+03
	Pu	9.2E+03	1.0E+02	9.3E+03	4.1E-01	1.2E+04	5.5E+02	1.2E+04	8.9E+00
	TOTAL	-3.1E+04	-3.2E+04	-3.1E+04	-3.1E+04	-6.3E+04	-6.3E+04	-6.2E+04	-6.2E+04
Minor Actinides	Pa	3.0E-05(0%)	7.8E+01(19)	6.0E-03(0)	7.6E+01(96)	5.3E-05(0)	8.2E+01(8)	2.7E-02(0)	8.4E+01(76)
	Np	3.6E+02(64)	3.3E+02(81)	7.1E+01(25)	2.4E+00(4)	9.0E+02(56)	9.6E+02(92)	1.2E+02(13)	2.7E+01(24)
	Am	1.6E+02(29)	5.7E-02(0)	1.7E+02(61)	1.0E-04(0)	4.7E+02(29)	1.3E+00(0)	5.5E+02(57)	8.5E-03(0)
	Cm	3.6E+01(6)	4.8E-03(0)	4.0E+01(14)	6.0E-06(0)	2.2E+02(14)	3.0E-01(0)	2.9E+02(30)	1.4E-03(0)
	TOTAL	5.6E+02(100)	4.0E+02(100)	2.8E+02(100)	7.9E+01(100)	1.6E+03(100)	1.0E+03(100)	9.6E+02(100)	1.1E+02(100)
ratio to (U235+U238)*		-	0.7	0.5	0.1	2.8	1.9	1.7	0.2
ratio to (30GWd/t)**		-	-	-	-	2.8	2.6	3.4	1.4

* ;ratio of minor actinides total to that of U235+U238 cycle 30Gwd/t burnup.
** ;ratio of minor actinides total of 60GWd/t burnup to that of 30GWd/t burnup.

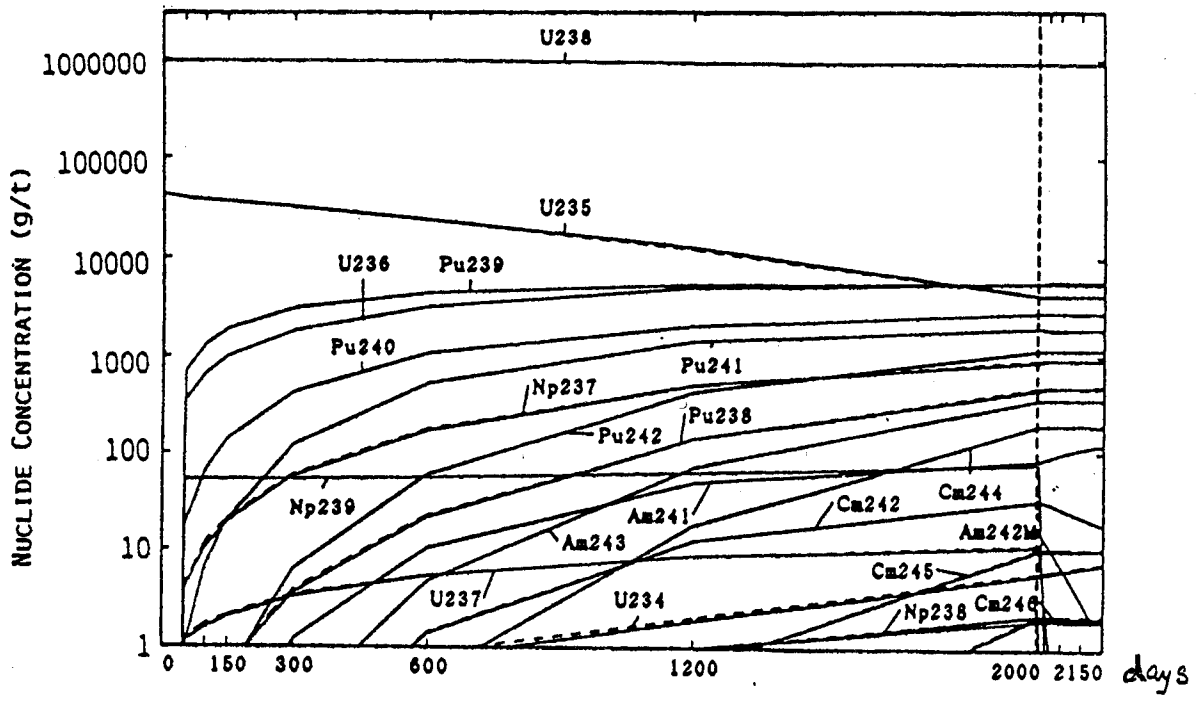


Figure 4: Build-up of heavy nuclides as a function of irradiation time [U-235 + U-238 fuel; burn-up at discharge = 60GW d/t; irradiation time of 2,000 days and cooling during 150 days] (142)

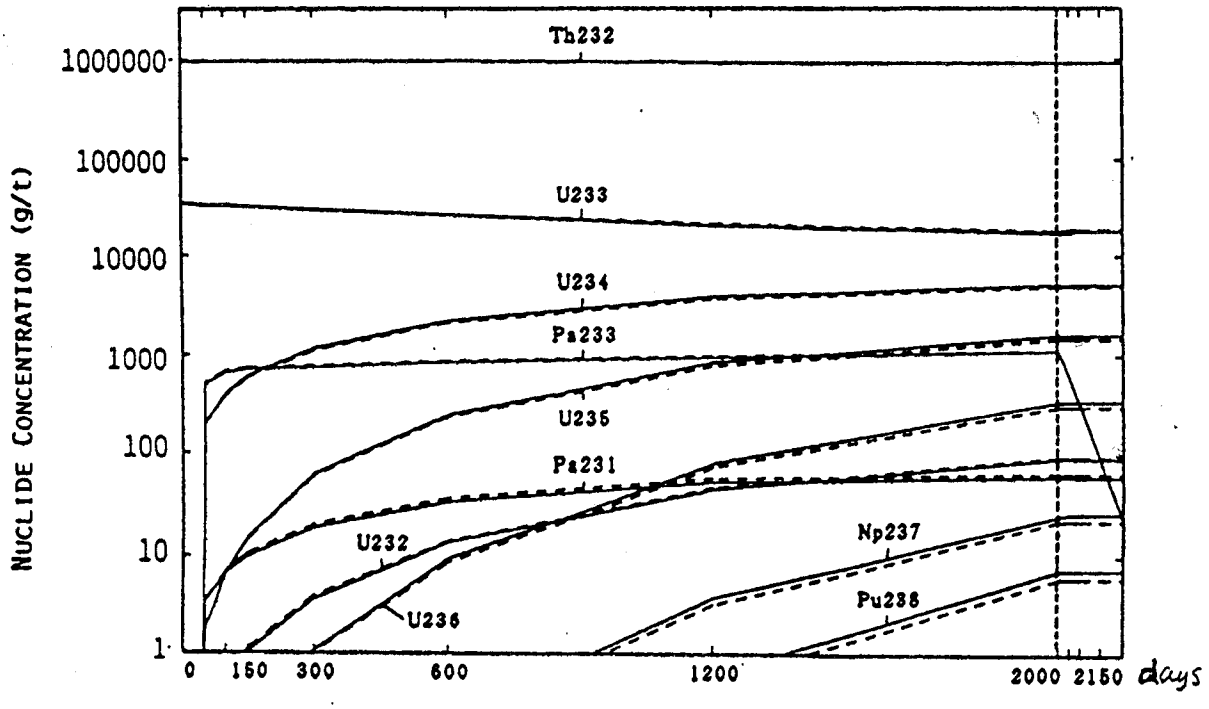


Figure 5: Build-up of heavy nuclides as a function of irradiation [U-233 + Th-232 fuel; burn-up, irradiation and cooling time as in fig. 4] (142)

3. THORIUM-BASED REACTORS, MIXED CYCLES, EXPERIENCE GAINED

3.1. U-233 Breeding

From the preceding considerations, it is clear that, unlike a U-235/U-238 reactor, a thorium reactor cannot achieve sustainable fission if no extra neutron source is available. However, once sufficiently U-233 has been bred within the thorium, sustained fission is theoretically achievable in properly designed thermal, epithermal, high temperature and fast breeder reactors.

But thorium can also be used with another fissile material, like U-235 or plutonium.

The simplest ways to accumulate sufficient U-233 is in a thermal or fast breeder using a U-235/U-238 core with a thorium blanket, which, upon reprocessing, will produce U-233. Any reactor type is suitable for this, a heavy water reactor (CANDU type, HWR), a light water reactor (PWR or BWR), or a gas-cooled high temperature reactor (HTR). A fast breeder reactor is, of course, suitable, fed with U-235 or Pu-239. Those reactors fitted to produce U-233 have been called "pre-breeders".

Most nuclear countries have thus bred U-233 in their programme at a point in time. In the US, more than 3 tons of U-233 were elaborated in the 1950s until 1975, and part of this stockpile may still be available. (cf. Chapter 7).

India is also producing U-233 at the present time for their nuclear programme. Thorium or thorium oxide (or thoria) rods are introduced in the Indian PHWRs where they flatten the flux. India is also breeding *some* U-233 in the Fast-Breeder Prototype Reactor.

3.2. Using Thorium in Power Reactors, Introductory Remarks (176)

3.2.1. Virtually every type of reactor has been associated at one time or another with some proposal to utilise a thorium fuel cycle. The purposes behind such proposals have varied and have influenced the related designs to some extent. Few have been the subject of major development work. Perhaps the earliest and certainly the *more* thoroughly studied application is the high temperature reactors. But some interest has been shown in the possibilities of thorium cycles for light water and heavy water reactors also.

It is self evident that the best utilisation of fissile material, expressed as the power capable of being produced from available resources of fissile and fertile material, will be provided by reactors which breed. The rate at which fertile material can be brought into use in this way depends on the breeding ratio (the breeding ratio quoted refers to fuel cycle equilibrium and is defined as : Atoms of fissile isotopes produced/Atoms of fissile isotopes destroyed) and on the fissile inventory required by the fuel cycle. This leads to the concept of "doubling time", the time over which the net production of fissile material enables the breeding power system to be doubled.

A simple consideration of fissile material utilisation leads directly to a preference for fast reactors of which the paramount characteristic is their ability to provide shorter doubling times. (See also Chapter 4 : Accelerator-driven reactors).

However, various reasons have been put forward for having an interest in the possible use of thorium in thermal reactors. Some of the arguments against fast reactors are based on fears of the possible proliferation of nuclear weapons. A different argument stems from the fact that fast reactors might not be available as the dominant source of nuclear power for a number of years to come, and it may be considered that meanwhile quite radical departures from current thermal reactor practice with uranium should receive attention.

3.2.2. The principal cases made for the thorium fuel cycle postulate an extension of the fissile material reserves, or use of readily available thorium compared to uranium, as in India, or other considerations such as high temperature or high irradiation resistance of thorium-base fuels, lower production of minor long-lived actinides in the nuclear waste, etc...

The following deals mainly with the fuel economy. But in confining one's attention to fuel utilisation, however, it is important no to lose sight of other factors such as design feasibility and costs affecting the selection of performance parameters and optimisation.

3.2.3. All reactors require an initial inventory of fissile material. All reactors other than breeders also require a make-up of fissile material in periodic refuelling. If this fissile material is U-235, it has to be derived from natural uranium, and the required amount of the latter can be quantified if an assumption is made about the enrichment of the separation "tails" which are rejected. The possibilities exist of using either U-233 or plutonium instead of U-235 as the required fissile material, but that would only correspond to a demand for U-235

elsewhere in the system and can therefore also be equated, though more approximately, to a requirement for natural uranium.

If a scheme does not include reprocessing and recycle of material (a "once-through" cycle), then the make-up required is, of course, the full amount of fissile material to be loaded into the reactor. In all cases, material becoming unrecoverable in the course of fuel cycle operations such as fuel fabrication, has to be taken into account. Similarly, the hold-up of material in these operations has to be included in the total inventory requirement.

Table 1: URANIUM REQUIREMENTS OF VARIOUS THERMAL REACTOR SYSTEMS (176)

	Tonnes Nat (a)
PWR U Once-Through	4 700
" Improved U-Once-Through	4 000
" Thorium/U-233 recycle "denatured" (c)	3 300
" Uranium/plutonium recycle	3 000
HWR Nat U Once-through	4 000
" 1-2 % U-235 Once-through	2 800
" Thorium/U-233 recycle	1 800
" Uranium/plutonium recycle	1 900
HTR (b) U Once-Through	3 400
" Thorium Once-Through	3 200
" Thorium/U-233 recycle ("denatured") (c)	2 500
" Thorium/U-233 recycle	1 800

Note :

(a) Tonnes Natural U per GW(e) capacity operated for 30 years at 75 % load factor, assuming 0.2 % tails from enrichment process.

(b) Batch refuelling. Continuous refuelling is better by a few hundred tonnes.

(c) "Denatured" by mixing U-233 with low enriched uranium.

The discussion which follows will bear at the same time on pure U-233/thorium fuelled reactors, but also on hybrid (or "crossed-progeny") cycles involving other fissile or fertile isotopes (U-235, Pu-239, U-238), for two main reasons :

1) The unavailability of U-233 at the beginning obliges to breed it first, then burn it.

2) Playing with the neutronic properties of each isotope - theoretically, but not always economically - could permit to achieve optimized results in such areas as :

- energy yields per ton HM (U-235, Pu-239, U-233),
- breeding (U-238, Th-232),
- lengthening of cycles and cutting outages (fuel stability, enrichment),
- avoiding proliferation (by dilution of fissile material or by presence of hard gamma isotopes),
- reducing the production of long-lived actinides (Th-232/U-233).

Among the interesting features of these isotopes, we can mention :

For thorium as fertile material :

- metallurgical stability, ability to withstand high levels of irradiation as a breeding material,
- production of less long-lived minor actinides compared to uranium,
- good antiproliferation characteristics (detectability), due to the gamma-emitting daughter-products).
- But a fuel cycle somewhat more expensive than for uranium due to the necessity of reprocessing to take advantage of U-233.

For uranium-238 as fertile material :

- contribution to energy production by fission with fast neutrons.

For uranium-233 as fissile material :

- low neutron capture, best eta values even at high temperatures,
- excellent neutronic properties at all neutron energies,
- production of shorter-lived actinides,
- good antiproliferation characteristics (detectability).

- But a more expensive fuel cycle compared to U-235 due in major part to fuel fabrication which has to be done remotely and can be compared to that of recent Pu-MOX fabrication, and to some extent also to somewhat more difficult steps in reprocessing, as will be seen.

For uranium-235 :

- good overall properties, naturally occurring isotope, the nuclear energy "workhorse",
- easy to handle, reprocess, fabricate into fuel, at least at low dilutions with U-238.
- But : - expensive enrichment, when necessary,
  - production of long-lived minor actinides.

For plutonium :

We should theoretically distinguish Pu-239, Pu-240 and Pu-241. However, in modern reactors these will usually come together in the usual proportions, with much Pu-240, 241, 242. Plutonium is an excellent fuel, due to the properties of Pu-239 and Pu-241, and is especially suited for fast reactors. However, the presence of Pu-240, Am-241, complicates things for neutronic and handling reasons, and the production of long-lived actinides to be found in the waste is appreciably enhanced; also, fuel fabrication with plutonium is more complicated and expensive than with U-235-238, even if it is mastered.

Very roughly, the following could be said according to the aims :

- <u>For industrial simplicity</u>

A once-through uranium 235-238 cycle could be preferred for its rather good proliferation resistance.

But : - problem of spent fuel disposal containing all long-lived actinides,
- poor overall economy : can be done with cheap uranium prices. The spent fuel should be retrievable for the future, for economical reasons when uranium prices will go up, and for "sustainability" reasons, not to waste valuable energy potentialities.

- For industrial efficiency (today)

A uranium 235-238 thermal cycle with reprocessing, plutonium recycling, would be preferred, assuring some sustainability.

However, for the longer term, it may be wise to spare the only naturally available fission "match", U-235, and burn also other fissile material, U-233 by using thorium. The presence of long-lived actinides in the waste might be considered a problem, as well as that of proliferation resistance when extracting U-233.

- For long-term resources sustainability :

- Breeding is the answer. The couple plutonium/U-238 is the best answer with fast reactors having a reasonable doubling time. However, the problems associated with plutonium handling are enhanced, as well as those of proliferation resistance.

- The couple U-233/thorium permits a limited breeding ("conversion") in thermal and epithermal reactors, high temperature reactors as well. Liquid-metal coolant of fast reactors can be avoided. The production of long-lived minor actinides is greatly diminished. Proliferation detectability is enhanced due to the U-232 filiation of gamma emitters.

However, the neutron balance in Th-232/U-233 converters does not permit an appreciable excess production of U-233, due to the "neutron sink" played by thorium as we have seen before (Chapter 2). The so-called "doubling effect times" permitting to breed enough U-233 to fuel another similar reactor, are very long; besides the fuel burn-ups in such case would be limited to uneconomically low values.

Hence, the realistic use of thorium is that it permits to tap a huge energy potential, along with that of uranium. This is in itself an extremely important asset which has to be kept in mind in long-term energy strategies.

3.2.4. **Mixed fuel cycles with thorium, U-233** (cf. ESCHBACH and al, ref. 19)

We call mixed cycles (or "crossed-progeny" cycles) those fuel cycles involving simultaneously or in parallel in the same reactor, fissile and fertile material from the uranium and thorium families. Although mixed fuel cycles will complicate the system, especially when the spent fuel will have to be reprocessed, it is

interesting to analyse some theoretical considerations which could be useful when planning thorium utilization.

The following remarks have been made by the Battelle Institute (19).

We will show how crossing the progeny of the U-233/Th and Pu/U-238 systems, as represented by preceding Table 5, Chapter 2.2.3., and Table 1 below, will maximize the virtues of both the fertile and the fissile materials. The first thing to notice is that the HWR spectrum has been slightly softened to better accommodate the plutonium spiking.

Table 1

FATE OF 100 ATOMS OF ^{239}Pu OR ^{233}U IN OPTIMIZED REACTOR SYSTEMS WITHOUT NEUTRON LOSSES

	Pu–Th in an optimized HWR (ϵ = 1.01)					^{233}U–^{238}U in an optimized LWR (ϵ = 1.10)				
	Atoms	Neutrons in	Neutrons out	Heat, %		Atoms	Neutrons in	Neutrons out	Heat, %
^{232}Th	108	108	3(net)	3	^{238}U	142	142	24(net)	15
^{233}Pa	108	1			^{239}Np	142	0		
^{233}U formed	107*				^{239}Pu formed	142*			
^{239}Pu	100	100	205	75	^{233}U	100	100	223	77
^{240}Pu	29	29			^{234}U	11	11		
^{241}Pu	28	28	64	22	^{235}U	11	11	19	8
^{242}Pu	7	7			^{236}U	2	2		
		164	269				124	242	

*Initial conversion ratio is 1.07 for Pu–Th and 1.42 for ^{233}U–^{238}U.

$$\eta\epsilon = \left(\frac{269}{100 + 28}\right)(1.01) = 2.12 \text{ for Pu–Th}$$

$$= \left(\frac{242}{100 + 11}\right)(1.10) = 2.40 \text{ for } ^{233}U\text{–}^{238}U$$

Furthermore, the lattice spacing of the LWR has been tightened to maximize the fast effect of U-238. Tightening the lattice will reduce the water volume and hence increase thermal utilization. Moreover, the reactor power per unit volume will be significantly increased if the lattice is tightened.

Considering the box to the left of the table, we see that plutonium in the HWR spectrum produces 269 neutrons for 163 neutrons absorbed, leaving an excess of 105 neutrons to be absorbed by Th-232. Three more neutrons are gained from the thorium fast effect, but one of these is lost to Pa-233 absorption. Hence the

number of U-233 atoms formed from 100 atoms of Pu-239 is 107, which is equal to a conversion ratio of 1.07. Thus the conversion ratio of Pu/Th is 10 % less than that of U-233/Th, (Table 5, Chapter 2.2.3.), and the eta epsilon is 4 % less, or 2.12 for Pu/Th and 2.20 for U-233/Th. These comparisons show that a small reduction in neutron availability occurs when plutonium instead of U-233 is used to enrich thorium.

However, this small reduction would be more than offset by the gain in the conversion ratio when using U-233/U-238, as the box to the right of the table shows. This box indicates that the number of neutrons available for U-238 absorption is equal to 242 neutrons produced, minus 124 neutrons absorbed, plus 24 neutrons gained from the U-238 fast effect.

The conversion ratio of U-233/U-238 therefore is 1.42, as compared to 0.99 with Pu/U-238 when no neutrons are lost, or a gain of 42 %. A significant increase is also seen in the percent of heat derived from the larger U-238 fast effect of U-233/U-238. In fact, it nearly doubles, going from 8 % in Pu/U-238 to 15 % in U-233/U-238.

The authors of this analysis come to the following conclusions :

1) Mixed cycles could be theoretically competitive with slightly enriched uranium fuel cycles;

2) Mixed fuel cycles would upgrade present LWR performance, making it a true advanced converter. (Note : this was demonstrated in the Shippingport experiment);

3) Heavy-water-moderated reactors fuelled with Pu/Th could economically supply the U-233 for the mixed fuel cycles;

4) Unlike U-235/thorium, Pu/thorium fuel would not contaminate the new U-233 with large amounts of U-235 and U-236;

5) The high enrichment required by thorium fuels could be decreased by reducing the thorium density;

6) Very high specific power and attractive reductions in reactivity swing could be obtained by using thorium, U-238, and plutonium as a ternary fuel.

The advantages of good neutron and overall material economy might be outweighed, however, by the complexity of having two separate fuel cycles, different fuelling times, possible complex reactivity problems at reactor halt (Pa-233, Pu-240, etc); these mixed cycles must rest on a well established, well equipped nuclear energy industry to minimize the costs. However, they could be considered for the future.

Many neutronic experiments to substantiate these views have been carried out in the 1960s on critical facilities in the US, especially the Advanced Epithermal Thorium Reactor (AETR) at Oak Ridge, the Mecca of thorium applied research in its time.

3.3. Application to High Temperature Reactors (HTR)

Here the thorium cycle was the original choice and development has reached a stage close to the commercial initiation of at least the reactor part of the cycle. That choice was related to the objectives of seeking a high outlet temperature and a compact reactor core compared to the traditional gas-cooled reactors.

The HTR is a graphite moderated helium cooled system in which the fuel is relatively dispersed in the moderator in the form of separate particles (instead of being segregated as stacks of fuel pellets in metallic cans, in the manner adopted in most reactor types). One result is that the core is composed entirely of ceramic materials capable of withstanding relatively high temperatures. The good neutronic yields of Th/U-233 even at high temperatures have been reported earlier. Thorium also permits the use of only relatively low neutron moderation and is compatible with the concept of a relatively compact core design. From simple theoretical studies, it seemed that conversion factors approaching unity might be achieved.

As designs were worked up in more detail, and with more attention to particular considerations such as fission product behaviour and fuel manufacturing costs, coating and some lumping of the fuel was adopted, with adverse effects on nuclear economy. Attention to an HTR concept using the U-238 plutonium fuel cycle was encouraged by these and other considerations, including the lack of an

established reprocessing route, limitations to power density set by neutron capture in protactinium (Pa-233) and the readier availability of U-235 at moderate enrichment levels. The plutonium produced could be recycled in the HTR, or more effectively, in fast reactors. However, although the HTR is no longer associated exclusively with the thorium/U-333 cycle, it is closer than any other reactor system to being applied in that role. (176)

Because there is no established thorium ceramic fuel reprocessing route, commercial attention to the thorium HTR has so far been directed to a "once-through" mode of operation, at least as a temporary regime, though this requires nearly double the amount of U-235 over a reactor lifetime compared with the U-233 recycle case. Even so, the U-235 requirement is much less than that of the current light water reactors operating in the once-through mode. (See Table 1) (6)

The ceramic-type fuel, being impervious to fission-product release into the helium coolant, has the great advantages to permit easy access for maintenance to reactor parts not containing the fuel directly, hence a very low personnel irradiation, and it produces almost no liquid operating waste.

Table 1

		HTR-Th/U	HTR-NB*	LWR	FBR
Conversion ratio		0.84	1.01	0.58	1.21
Fissile inventory (Core + fuel cycle with 1.5 y out of pile)	Kg/GWe	2400	4500	2600	5400
Net fissile requirement per year	Kg/GWe-y	153	16	575	- 125

* HTR net breeding cycle (Th/U-233)

A rough comparison of fuel types and functional parameters between HTRs and LWRs is given next page (Table 2) (11).

Among the industrial prototypes which have been built, the more important are :

- in the USA:
  - the General Dynamics Peach Bottom HTGR of 30 MWe (1966-1974) for Philadelphia Electric Co.
  - the Fort St Vrain HTGR of 330 MWe of General Dynamics (1976-1989) for Public Service of Colorado.

- In Winfrith UK, for OECD-EURATOM, the DRAGON HTGR of 20 MWth. (1966-1973)

- In Germany, the HTGRs pebble-bed reactors :
  - AVR (13 MWe, 1967-1988) at Jülich, of BB-Krupp,
  - THTR (300 MWe, 1985-1989) at Schmehausen (Hochtemperatur-Reactor-Bau GmbH - HRB).

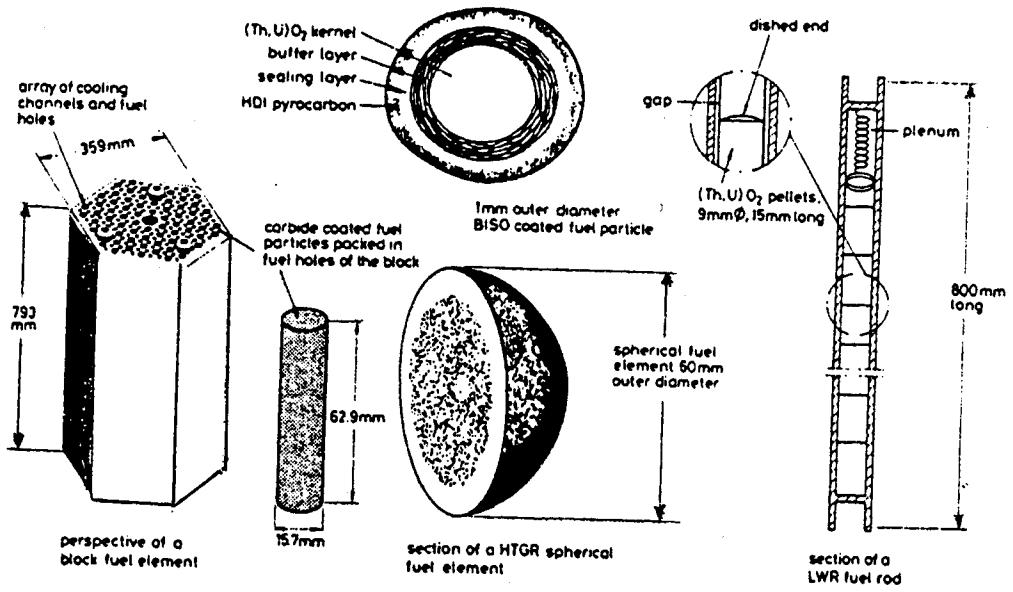
All these reactors were cooled with helium gas.

The Peach Bottom HTR (22) core consisted of a graphite core with graphite sleeves within which 682 cylindrical fuel elements of about 3.6 m length and 6 cm diameter were placed. The fuel element itself is almost made of graphite, save for a 2 300 mm long annular portion (Fig.1) of enriched U-235/thorium "compacts" that contain coated Th/U particles in a graphite matrix. Bottom-driven control rods contain burnable poison as zirconium diboride in a graphite matrix. As U-235 burns, U-233 is being bred into the thorium.

The reactor has delivered some 1 400 GWh, giving an honourable 70 % availability factor in about 7 years.

Table 2

The Behavior of Thorium Fuels in Nuclear Reactors Source : Gmelin (11)



Comparison of converter fuel geometries, drawn by the authors of this article, based on [13, 14].

Comparison of the Functional Parameters of the High-Temperature Gas-Cooled Reactor (HTGR) and the Light Water Reactor (LWR) [7].

	HTGR	LWR
Operation Parameters		
fuel	MO_2 or MC_2	MO_2, MC or MN, M = (Th, U)
core size in m	6 Ø and 6 long	2 Ø and 2.5 long
fuel temperature in °C	800 to 1400	650 to 2000
coolant pressure in bar	50	60 to 150
specific power in MW/kg	0.3 to 0.5	1 to 2
power density in MW/m^3	6	20 to 80
Φ_F in n/m^2	4 to 7×10^{25}	0.6×10^{25}
fuel residence time in y	2 to 4	0.5 to 2
burnup in % FIMA	10 to 14 (78% separated fissile)	0.5 to 5
Fuel Behavior		
fast neutron damage	PyC shrinkage	cladding and fuel swelling
fission fragment damage	unimportant	cladding embrittlement and fuel swelling
swelling	unimportant	influences heat transfer and fuel temperature, life, and creep
temperature gradients	Amoeba effect	fuel restructuring and redistribution
corrosion	fission product-coating reactions	fission product-cladding reactions
fission gas release	strains coatings (pressure vessel failure)	clad distension, and higher fuel temperatures

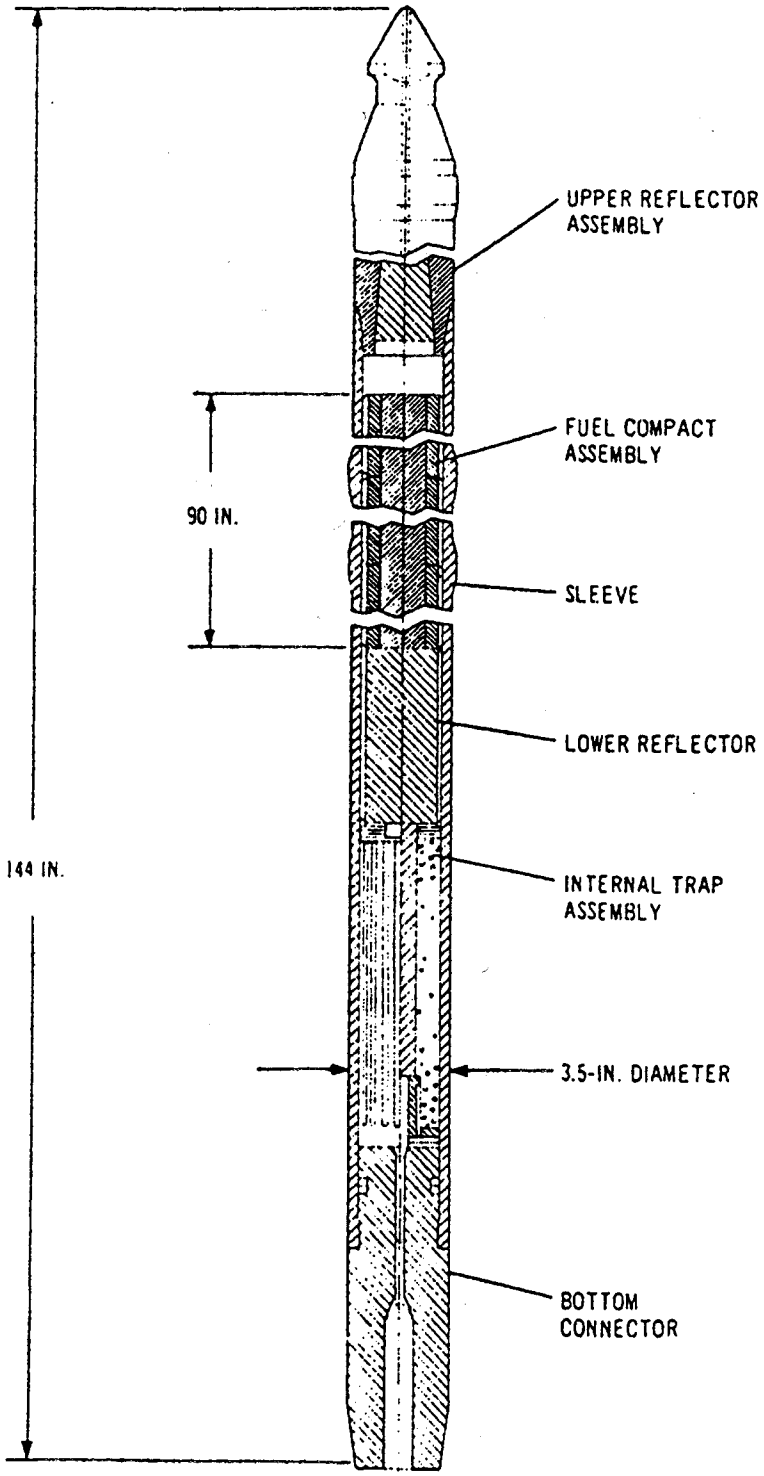


Figure 1: Cutaway view of a single fuel element of Peach Bottom Reactor (22)

The Fort St Vrain (11) a larger version, was probably too quickly extrapolated, as it has met with many structural problems. Among those, the reactor used water-lubricated helium circulator bearings which resulted in frequent water ingress into the reactor system and caused significant down time. It is now under dismantling, and it is reported that the graphite blocks are quite radioactive (due probably to fission product leaks). The electricity production in 12 years was of about 6 000 GWh, meaning that the plant has been down most of the time, (1976-1989).

The small DRAGON OECD project (21) (150) has operated successfully between 1966 and 1973, sometimes at 13 % higher rates than nominal. (Fig. 2)

The coolant enters the core at 350°C and leaves at 750°C at 20 atm pressure. It has been shown that technology exists to reach temperatures of 1 000°C, and 1 500°C in the core. Views of the reactor and of the first batch of U-235/U-238 and U-235/Th fuel elements are given. The fuel clusters (Fig. 3) are particularly complicated. 70 rods were (Th/U) C fuel, with Th to U-235 ratio greater than 3. Those elements were left during many 240 day cycles when the "driver" rods had to be replaced; they could reach irradiations of 100 000 MWD/t without problem. It was observed that around 1 000°C the graphite shrank notably under the fast neutronic flux, and had to be replaced in 1971.

Later in the course of experiments, less Th/U fuel elements were used, to concentrate on U-C and UO2 fuel elements. The clusters were simplified ("telephone dial" arrangement), with possibility to test "integrated bloc" fuel elements, the General Dynamics model. (Fig. 4)

A wide variety of fuel types were tested with success. Also the helium contamination has always remained very low, due to good fuel performance. (Coatings were mainly of the composite type : an inner pyrocarbon coating, a layer of silicon carbide and a thick outer layer of pyrocarbon). The kernel size was about 400 microns in diameter, the coating thickness about 120 microns.

1, Arrangement of reactor pit
2, Specimen facility coolant circuit
3, Fission-chamber assembly
4, Neutron source drive
5, Thermocouple branch
6, Reentrant tube
7, Purge-gas pre-cooler
8, Bottom biological shield
9, Thermal column
10, Ion chamber
11, Specimen facility
12, Reactor pressure vessel
13, Support structure inside pressure vessel
14, Core and static graphite bedplate
15, Heat sink and dump tank installation
16, Fuel elements
17, Reflector assemblies
18, Thermal shield
19, Core unclamp mechanism
20, Core seal
21, Support structure inside pressure vessel
22, Concrete biological shield
23, Retractable TV viewing facility
24, Main shield plug
25, Valve floor
26, Control-rod mechanism
27, Charge/discharge machine installation
28, Nonretracting TV viewing assembly
29, Heat exchanger shielding
30, Primary heat exchanger and circulator
31, Main entry valve
32, Load facility
33, 25-ton crane

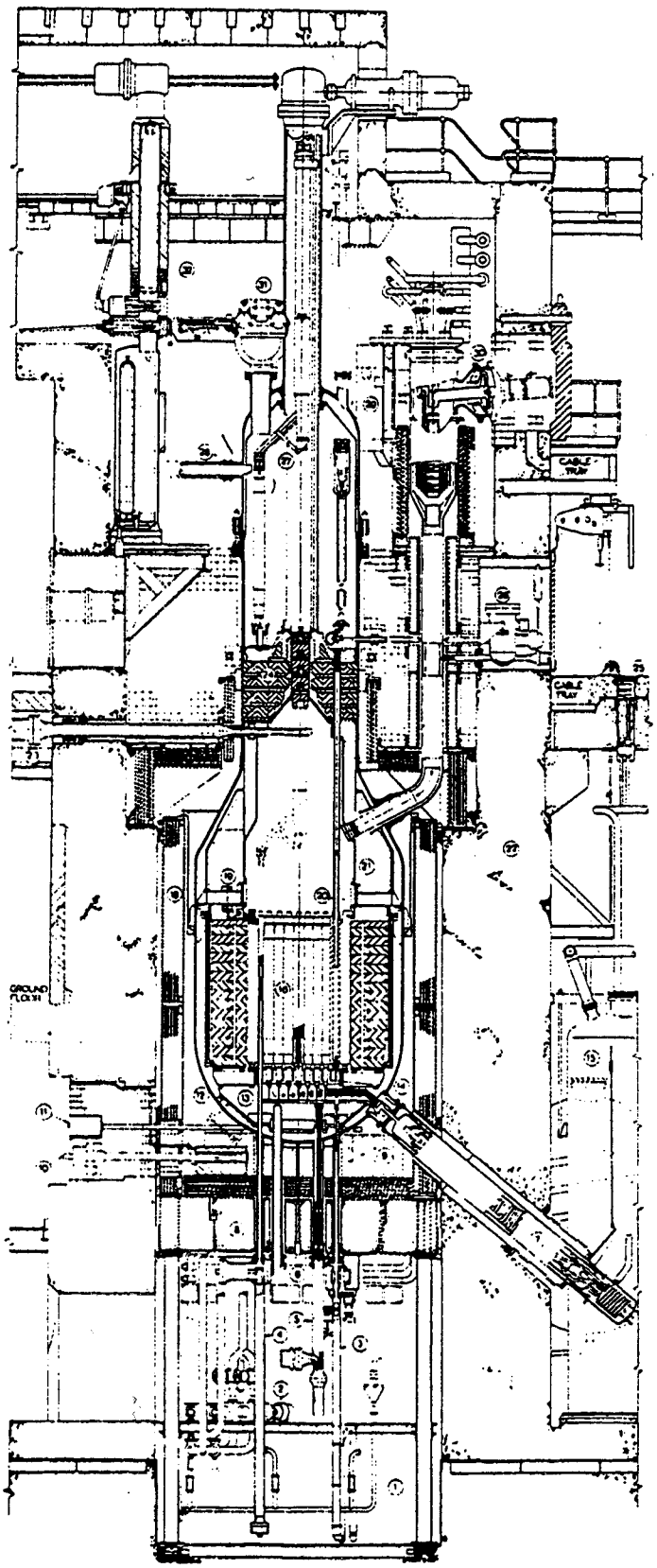


Figure 2: DRAGON - General assembly of reactor composite section

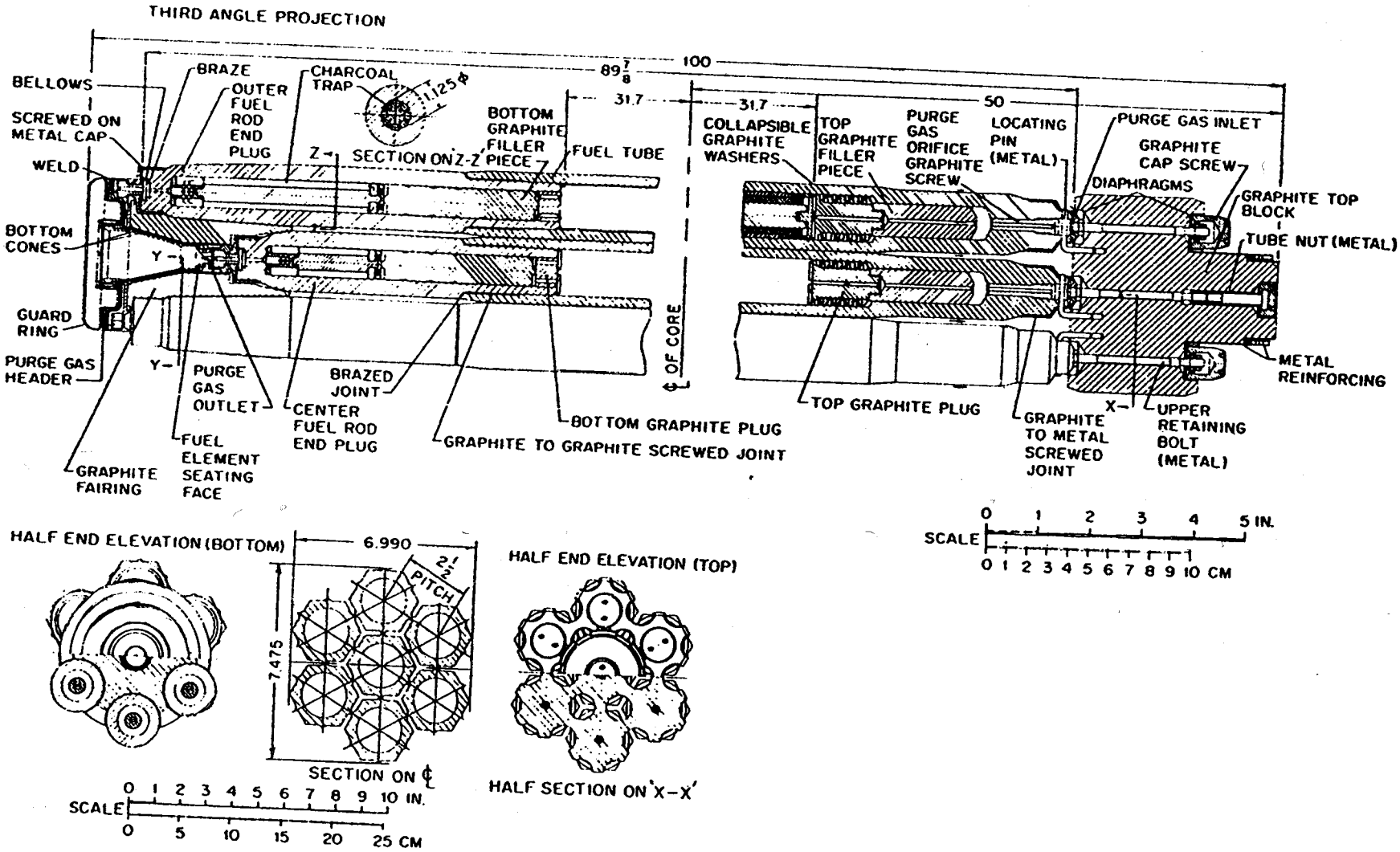


Figure 3: DRAGON - Fuel element MK IX

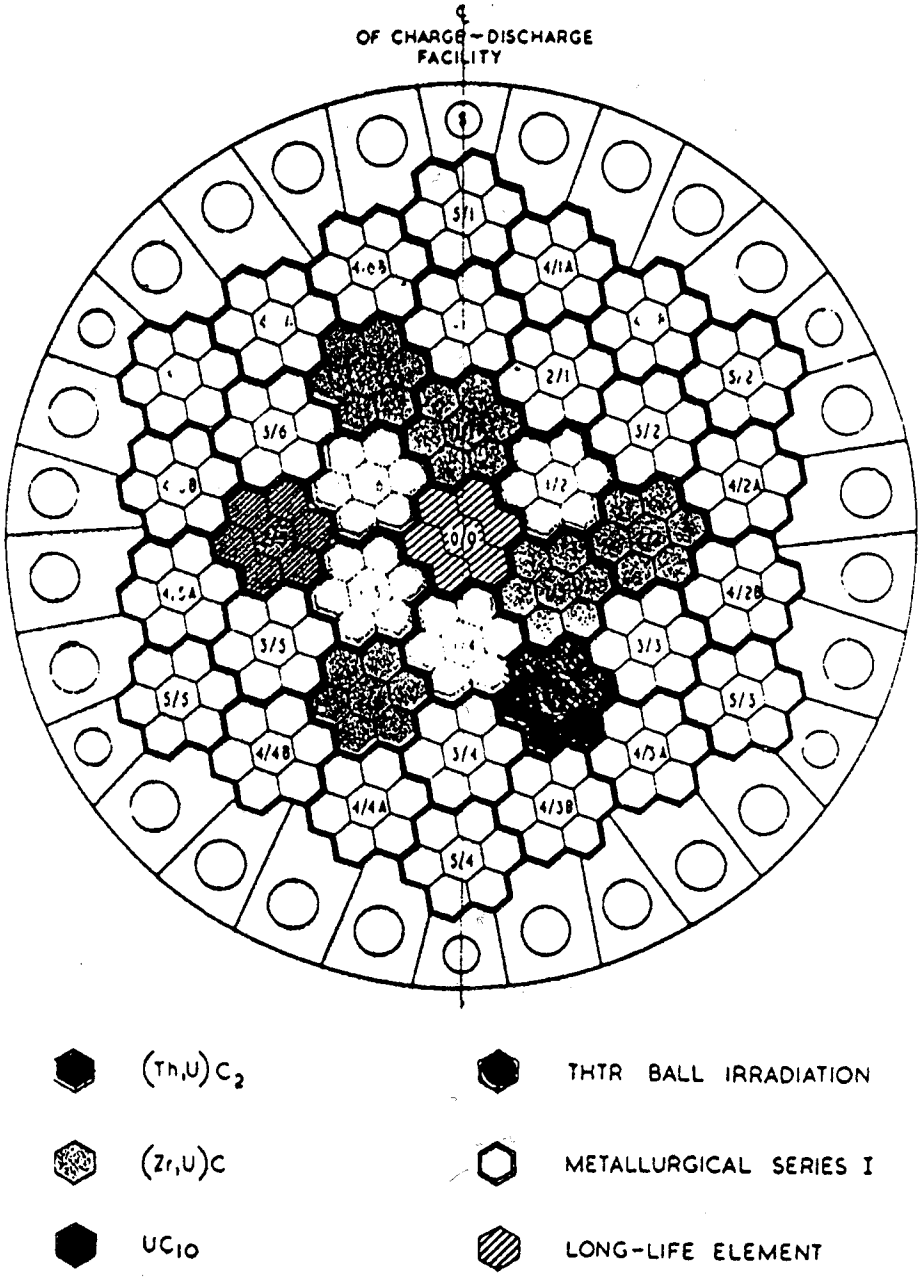


Figure 4: DRAGON - Reactor core plan

We quote some of the conclusions of the 1974 Report :

"As far as the fuel is concerned, the Dragon model can be adapted to either of the two fundamental cycles, one using highly enriched U + thorium which breeds U-233, the other using low enriched U in which the U-238 progressively breeds plutonium. Each one of these two cycles presents advantages, but until recently, the low enriched U cycle has been preferred in general, due in part to the fact that it can incorporate a configuration of fuel called "block with incorporated pins", on which the Dragon Project has mostly been oriented in the recent research work.

"All the signatory organizations which are interested in developing HTRs with a prismatic core have decided to adopt the type of fuel element with "integrated bloc". This in part results of agreements with General Atomic of USA (Division of General Dynamics), which has an important program of construction of high temperature reactors. Hence, the French and German programs are clearly oriented towards the thorium/highly enriched U cycle, on which originally all the work of the Dragon Project was centred."

However, after 1974 the project has been stopped, mostly for political/economic reasons between the signatories.

The AVR and THTR pebble-bed reactors in Germany

Those are quite special reactors in that the fuel is contained in 6 cm diameter graphite pebbles. One plays on the (low) speed of flow of these pebbles to regulate the neutronic reaction, by action on the temperature. Coolant is helium. (Table 3).

A standard graphite ball of 6 cm diameter contains about 35 000 pyro-carbon coated , 500 microns diameter microspheres (See Fig. 5). These particles in a "feed" ball contain 1 g of U-235 and 10 g of thorium. A "breed" ball contains 0.5 g of uranium and 13 g of thorium in the form of oxides. A fuel free zone of 0.8 cm covers the outside of the ball.

The residence time is 2-6 years in the reactor, the maximum power 2.4 kW for a ball at 1 350°C maximum temperature and the burn-up 100 000 MWd/ton.

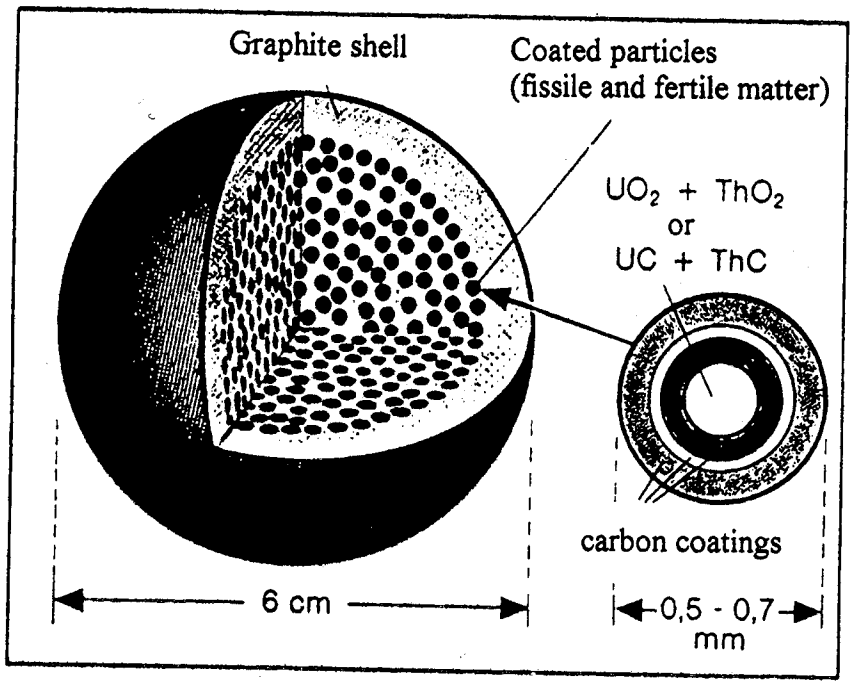


Figure 5: Pebble-shaped fuel element

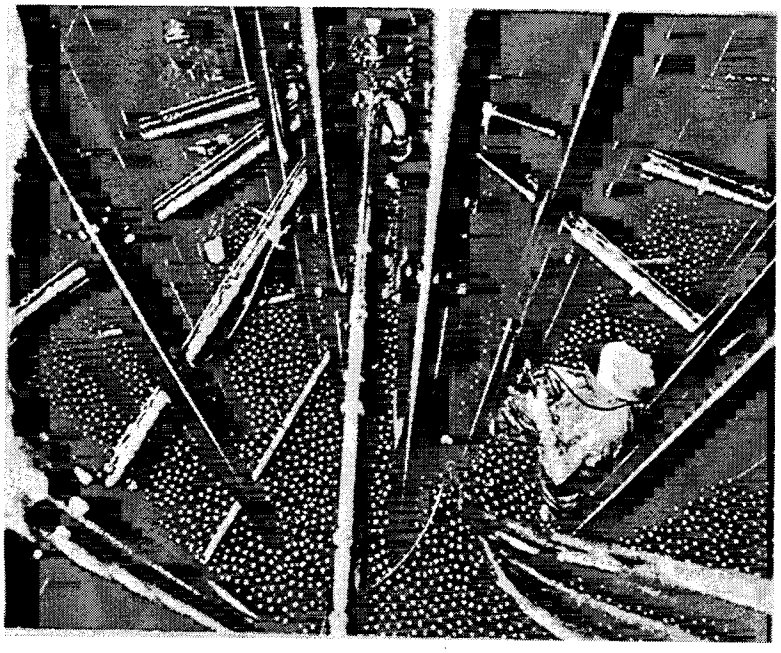


Figure 7: View of the THTR core being filled with pebbles

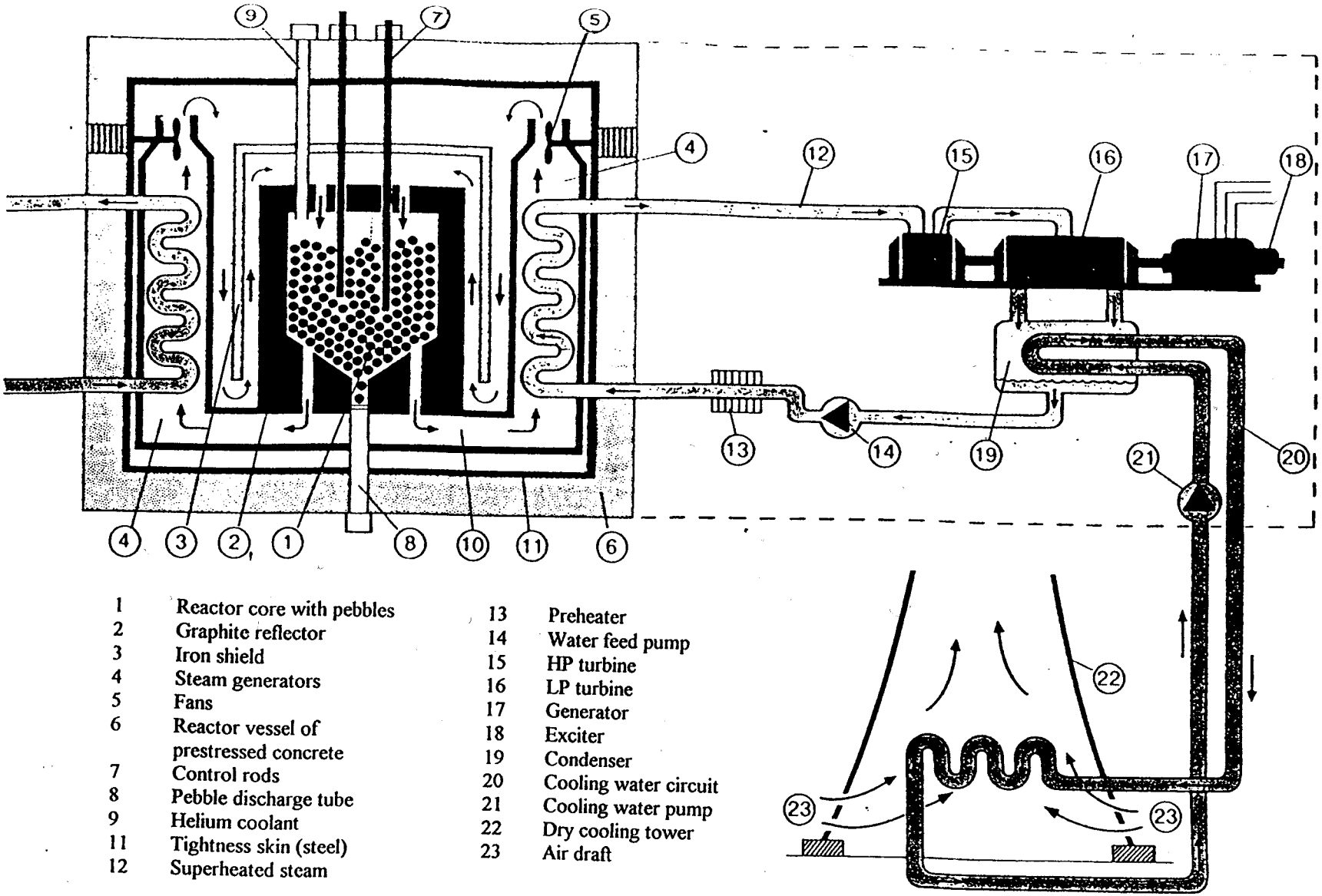


Figure 6: Operating principle of the thorium High Temperature Reactor (THTR)

Figure 6 shows the principle of operation of such pebble-bed reactors, and Figure 7 shows the reactor THTR being filled with the balls.

The AVR reactor at Jülich has been a great success and operation lasted more than 20 years between 1967 and 1988. (Fig. 8, Table 4) . Its core contained 80 000 pebbles. Almost over the whole period the fuel was thorium and highly enriched uranium as mixed oxides, carbide coated. The maximum burn-up achieved was beyond 200 000 MWd/t. Over many years the average helium outlet temperature was as high as 950°C. The reactor showed remarkable stability and "forgiveness".

In view of the excellent results obtained with AVR, it was decided to build a 300 MWe industrial prototype, the **THTR Reactor** at Hamm-Uentrop / Schmehausen, which operated between 1985 and 1989 (Fig. 8) (Tables 3, 4 and Table 5). Its core contained 280 000 fissile and moderator pebbles, and 35 000 absorber pebbles containing burnable boron.

Unfortunately, like for the Fort St Vrain, extrapolation from a 14 MWe machine to a 300 MWe reactor generated problems with the reactor internals; the cost of repairs was estimated too high and the reactor was abandoned, in a difficult political climate.

The HTR development was originally based on the thorium cycle. Both pebble-bed HTR reactors, AVR and THTR, were operated in that cycle and closure of the cycle was under development. Due to the objection against the use of highly enriched uranium (HEU), the thorium cycle development was stopped in 1980.

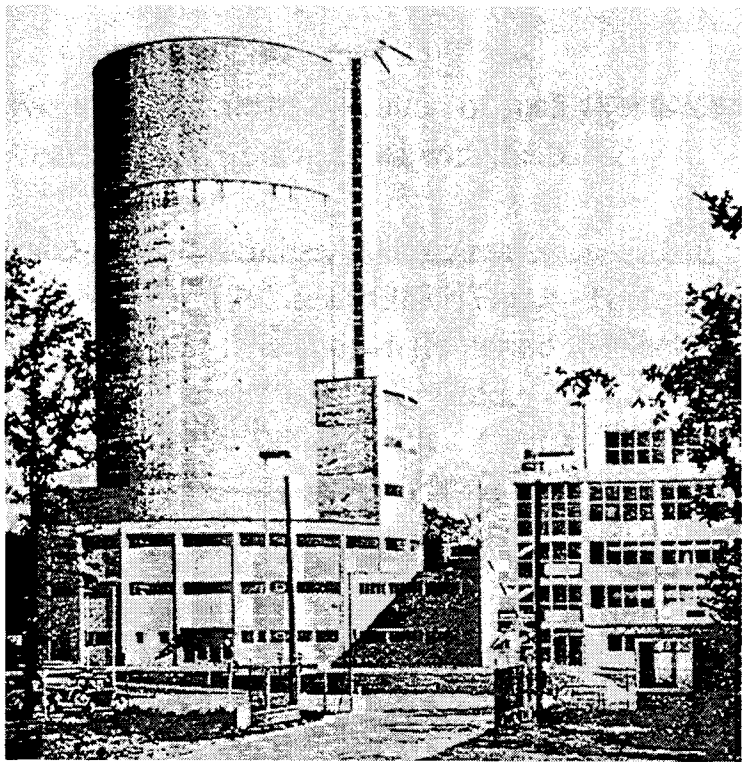
Computational research shows that the thorium cycle is also possible with medium enriched uranium (MEU) as feed fuel, to enhance non-proliferation, and closure of this cycle could bring similar advantages in uranium savings as with HEU. For example (6) it can be shown that a HTR could operate on a 20 % enriched U-235 cycle or a corresponding 12 % U-233 cycle.

The HTR were then converted to the low enriched uranium (LEU) cycle with direct final disposal of the fuel elements. The feasibility of the LEU cycle was then demonstrated in the AVR.

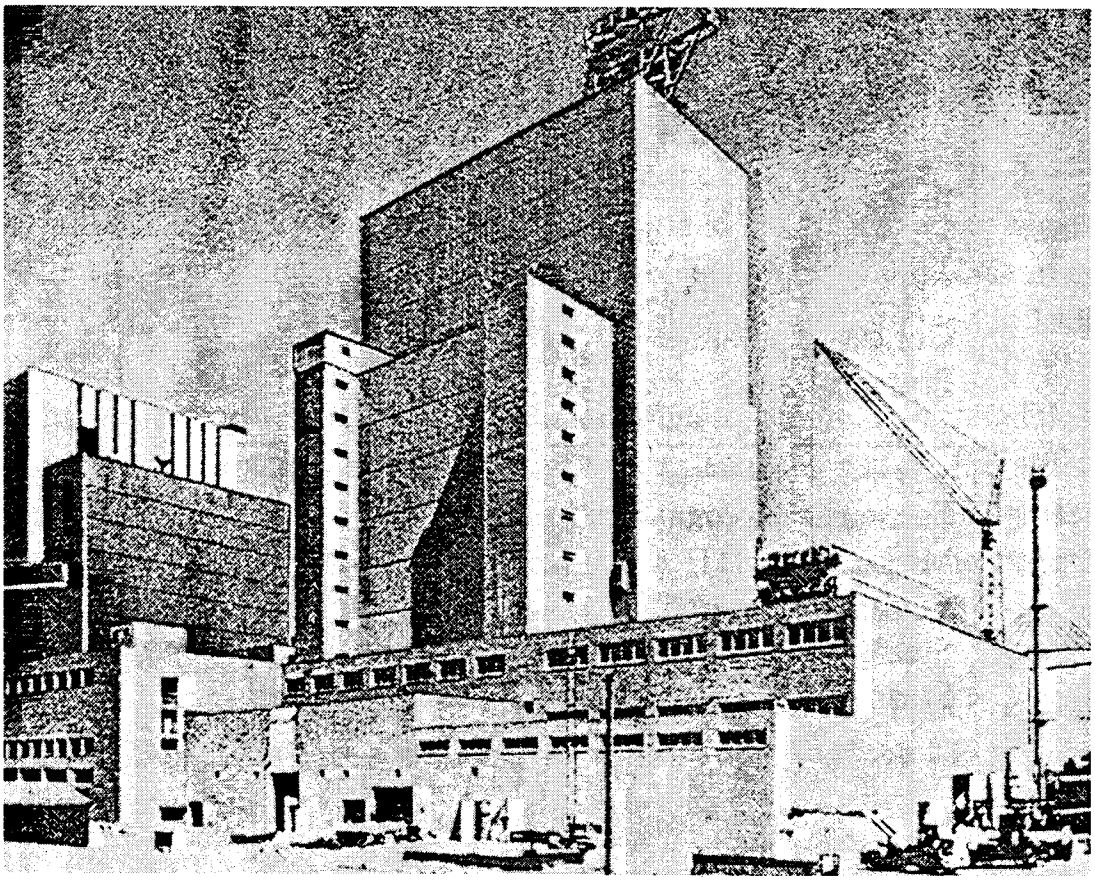
Over the last years the HTR development was preferably directed towards the modular HTR German pebble-bed type of reactors (MODUL), using passive

Table 3: THTR characteristics

Thermal power	759.5 MW
Electrical power	307.5 MW
Thermal efficiency	40.49 %
Fissile material	U-235
U-235 enrichment	93 %
Mass of U-235 at criticality	344 kg
Fertile material	Th-232
Mass of Th-232 at criticality	6400 kg
Fraction of fissile material to total heavy material (thorium, uranium, plutonium)	5.4 %
Nb of control and scram rods	42
Nb of reflector rods (out of which 12 for control)	36
Absorber material	B4C
Primary circuit :	
Coolant	Helium
Entrance temperature	250°C
Outlet temperature	750°C
Pressure	39.2 bar
Secundary circuit :	
Medium	Water
Entrance temperature	180°C
Steam outlet temperature	530°C
Steam pressure	177.5 bar



AVR, JÜLICH



THTR, SCHMEHAUSEN

Figure 8: The reactors AVR-Jülich and THTR-SCHMEHAUSEN

Table 4: Experimental and HTR prototype characteristics (16 - IAEA/INFCE 1980)

	Experimental Reactors			Prototype Reactors	
Name	Dragon	Peach Bottom	AVR	Fort St. Vrain	THTR
Location	Winfrith, U.K.	Peach Bottom, USA	KFA Julich, Germany F.R.	Colorado, USA	Schmehausen Germany, F.R.
Power (MW(th)/MW(e))	20 / -	115 / 40	46 / 15	837 / 330	750 / 300
Fuel elements	Cylindrical/ PRISMATIC	Cylindrical	Spherical	Hexagonal *	Spherical
He temperature, inlet/outlet (^{o}C)	350 / 750	377 / 750	270 / 950	400 / 785	270 / 750
He pressure (bar)	20	25	11	48	40
Core power density (MW/m^3)	14	8.3	2.3	6.3	6
Fuel coating	TRISO PyC, SiC	BISO-BISO PyC	BISO PyC	TRISO-TRISO PyC, SiC	BISO PyC
Fuel kernel composition	Thorium, uranium dicarbides	Thorium, uranium dicarbides	Thorium, uranium mixed oxide	Thorium, uranium dicarbides	Thorium, uranium mixed oxide
Reactor vessel	Steel	Steel	Steel	PCRV	PCRV
Start/end of power operation	1966/1975	1967/1974	1968	1977	(1981)

* It had 5 tonnes ThUC & 15 tonnes ThC2 (6)

safety features for potential siting in industrial areas and with applications other than electricity production (Fig. 9)

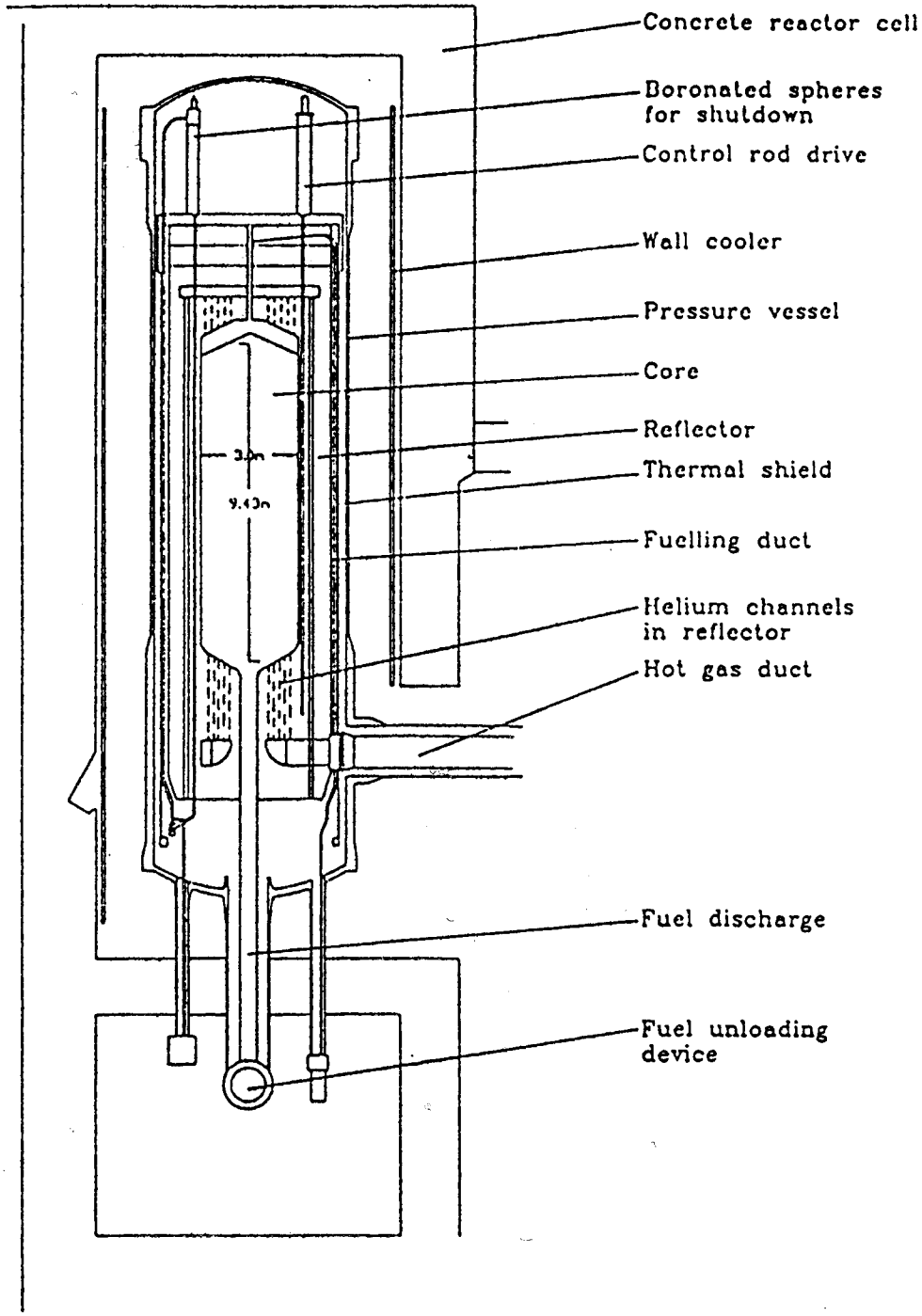


Figure 9: MODUL reactor scheme (6-E Teuchert)

Advantageous use of the thorium cycle in MODUL would be possible. However, it has been postponed to a later commercial phase of this reactor.

The tables 4, 5 and 6 give some more information on those reactor types and their anticipated fuel cycle (IAEA, INFCE Study, 1980).

At present, all HTR prototypes have been shut down and most research has been stopped, to the exception of Russia, China and Japan, with a limited renewed interest in professional circles in the USA (General Atomics) and France (Framatome).

Table 5

Technical Data on Th-Fuelled Reactors (compiled by the authors of this article, using the publications of the International Atomic Energy Agency, IAEA, Vienna.

reactor	Indian Point Buchanan, New York [15, Vol. III, p. 59]	Elk River Minnesota [15, Vol. IV, p. 103]	Fort St. Vrain Colorado [15, Vol. IX, p. 185]	Uentrop, Hamm Nordrhein-Westfalen (Germany) [15, Vol. X, p. 305]
reactor type	PWR	BWR	HTGR	HTGR
power				
MW(th)	585	58+15(fossil)	842	750
MW(e)	165	22	330	296
efficiency	25.8	30(net)	39.2	39.5
coolant	H_2O	H_2O	He	He
temp. in °C	271	280	770	750
pressure in bar	104	63	47.6	39
core				
height in m	2.5	1.52	4.75	6.0
diameter in m	1.97	1.45	5.94	5.6
temp. in °C	318 to 2677	507 to 1548	830(1350max)	1050(1350max)
fuel				
composition	$(Th_{0.93}U_{0.07})O_2$	$(Th_{0.96}U_{0.04})O_2$	$(Th_{0.81}U_{0.19})C_2$ and ThC_2	$(Th_{0.90}U_{0.10})O_2$
^{235}U enrichm. in %	93	93	93	93
loading in kg				
U-235	1100	170	870	344
U as UX_2	1300	183	936	369
Th as ThX_2	17207	3779	19500	3444
power density in MW/m^3	76	23.1	6.4	6
specific power (thermal) in MW/kg ^{235}U	0.53	0.34	0.97	2.2
fuel elements				
type	clad rods	clad rods	prism	spheres
No.	23400	3700	1482	358000
cladding	304 SS	304 SS	TRISO coatings	BISO coatings
burnup in % FIMA*	1.9 to 4 (max)	0.9	10(75% fissile and 7% fertile)	12 to 14
residency in a flux in $n \cdot m^{-2} \cdot s^{-1}$	1 to 3	ca. 1 (2 max)	4	4
average $\Phi_{th} \times 10^{-14}$	0.15	0.15	0.42	1.3
average $\Phi_f \times 10^{-14}$	1.7	0.48	0.32	0.53
conversion ratio	0.5	0.37	0.6	0.53

* 1% FIMA=2.23×10^3 MWd/kg HM. (For ThO2, ThC, 1% FIMA= 9610 MWd/t) (Hence, 14% FIMA is about 135 000 MWd/t).

Table 6

Fuel Cycle Information - HTR (Recycle) (16-IAEA INFCE)

	France	Germany, F.R. HRM	USA	USA
	1	2	4	5
Fuel cycle	LEU	HEU-235/Th	MEU-235/Th	MEU-233/Th
Topping isotope	235U	235U	235U	233U
Special feature	-	-	Denature in-situ	Denature in-situ
Conversion ratio (av. Eq^m)	0.48	0.74	0 6	0.77
Refueling interval (years)	1	Continuous	1	1
Fraction of core replaced	0.33	0.21	0.25	0.33
Eq^m reload enrichment (%)	10.9	93	18.3	11.7
Out-of-core delay time (years)	1	2	1	1
Out-of-core fissile loss (%)	-	2	2	2
Average discharge exposure (MW.d/t)	120000	80000	96000	48000
Moderation ratio C/H.M.	495	160	290	190
Fissile requirements[a,b] (t/GW(e))				
Initial core	1.022	1.91	1.26	1.26
Annual Eq^m, recycled	0.064	0.27	0.157[c]	0.322[e]
Annual Eq^m makeup	0.518	0.18	0.371[d]	0.226[f]
30-y cumulative gross	15.1	8.51	12.31	7.81
30-y cumulative net	14.55	5.10	11.24	6.69
Natural uranium requirements[a,b] (t/GW(e))				
Initial core	196.11	350	235	None
Annual Eq^m	114.24	35	72	None
30-y cumulative, gross	2993[g]	1680	2386	None
30-y cumulative, net	2892[g]	1008	2177	None
Th requirements[a,b] (t/GW(e))				
Initial core		48	23.85	31.0
Annual Eq^m		7.1	4.19	9.5
30-y cumulative, gross		261	145	307
30-y cumulative, net		18	11.74	18.7
SWU requirements[a,b] (t/GW(e))				
Intial core	193	454	297	None
Annual Eq^m	121	45	87	None
30-y cumulative, gross	3471[h]	2184	2884	None
30-y cumulative, net	3346[h]	1302	2651	None
Fissile material in spent fuel[a,b]				
Enrichment (Eq^m) (%)				
U-fiss./U	1 41	60	6.9	7.9
Pu-fiss./Pu	57	-	53	68
Annual Eq^m discharge (kg/GW(e))				
U-fiss.	65	245	152	317
Pu-fiss.	44	1		47
30-y fissile material discharged (kg/GW(e))				
U-fiss.	1926	9390	5376	10437
30-y fissile material discharge (kg/GW(e))				
Pu-fiss.	1311	30	859	1456

[a] Normalized to 70% capacity factor and 0.2% tails enrichment.

[b] 30-year net cumulative requirements are 30-year cumulative gross requirements less in-core and ex-core inventory at the end of 30 years.

[c] Of this total, 0.126 t/GW(e) is recycled in the HTGR and 0.042 t/GW(e) is discharged at an enrichment of approximately 3% and is credited in the net consumption.

[d] Net annual makeup. Includes a credit of 0.042 t/GW(e) as described in ftnt. d. Gross annual makeup = 0.397 + 0.042 = 0.439 t/GW(e).

[e] Of this total 0.205 t/GW(e) is recycled in the HTGR and 0.140 t/GW(e) is credited in the net consumption.

[f] Net annual makeup. Includes a credit of 0.140 t/GW(e) as described in f. Gross annual makeup = 0.242 + 0.140 = 0.382 t/GW(e).

[g] Uranium recycle only (symbiosis with FPR would give much lower requirements).

In Japan, the Japan Atomic Research Institute (JAERI) is building at its O-Arai Center a 30 MWt prototype **High Temperature Test Reactor (HTTR)** which should attain criticality before year 2000. (Figs. 10 and 11). The reactor core is made up of 30 columns of graphite fuel elements in a cylindrical array, each column having a stack of 5 fuel elements. It is planned to unload each column every 2 years (660 days). Each fuel element is of the "pin-in-block" design, with a massive graphite block in which are installed concentrically about 35-37 fuel rods. These fuel rods are made of fuel "compacts" stacked in a graphite sleeve. The fuel compacts are themselves made of Triso-coated *UO2* kernels of about 1 mm diameter (Fig. 12). The design helium coolant outlet temperature is 950°C.

This interesting prototype will timely revive the past techniques with recent technologies and will pave the way for other developments. (For the time being, the fuel will be uranium.)

China is also building a prototype HTR of 10 MWth called HTR 10, of a design similar with the AVR, using uranium-base coated "pebbles". The outlet temperature of 700 °C could later be increased to 950°C, and the steam heat exchanger will later be replaced by a gas heat exchanger to drive a gas turbine (Fig. 13). HTR 10 should be completed by about 2000.

General Atomics has also a project for a 300 MWe modular HTR (MTR) integrated with a gas turbine (thermal yield 55 %). This novel design may lead to a US- French-Russian cooperation (Fig. 14).

It is our opinion, as was said in the case of DRAGON, that one day a prosperous high temperature model will emerge, based either on uranium or thorium cycles. The high temperatures could be used for chemical reactions (e.g. H2 production from water) and/or electricity production with excellent thermodynamic yields.

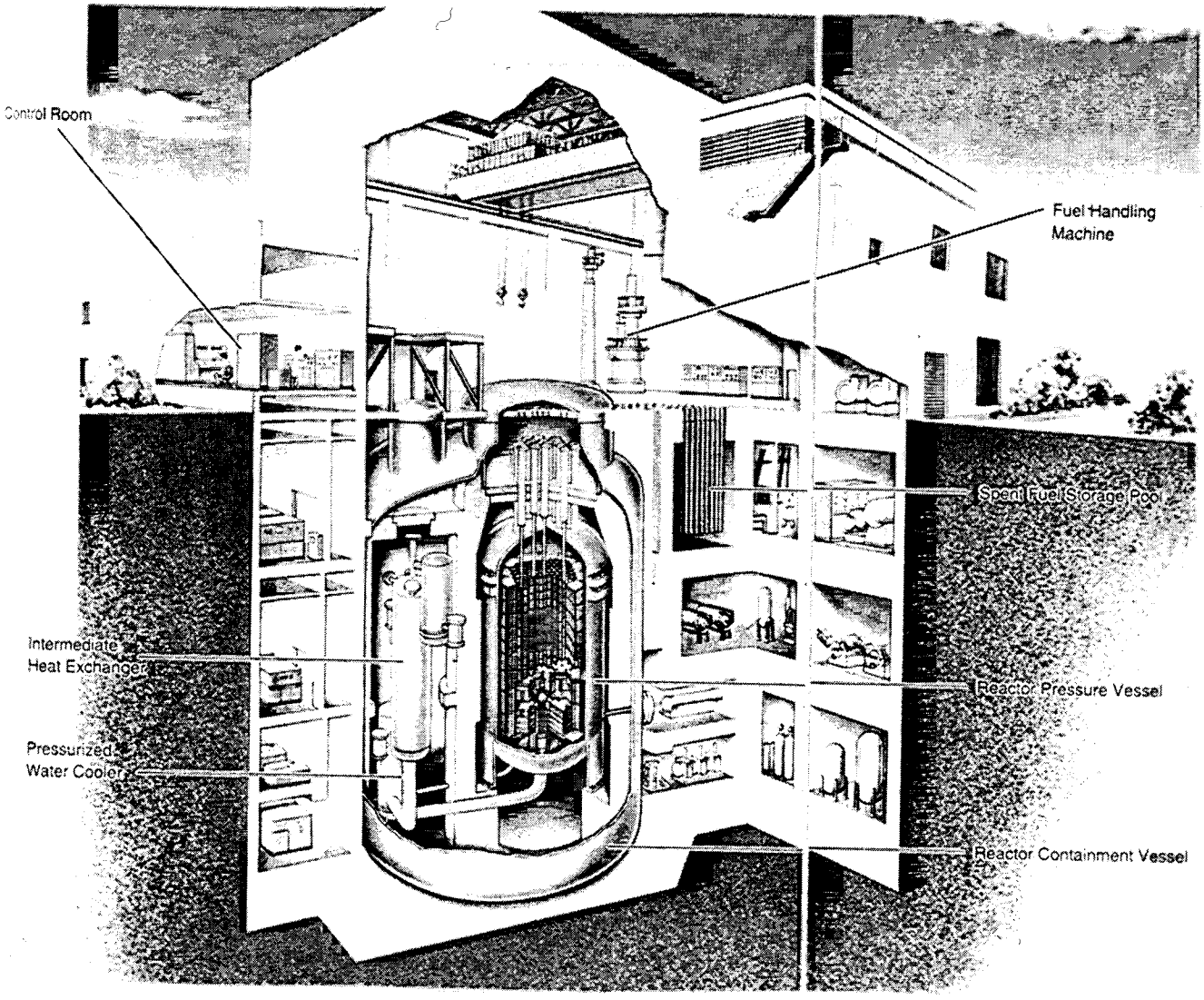
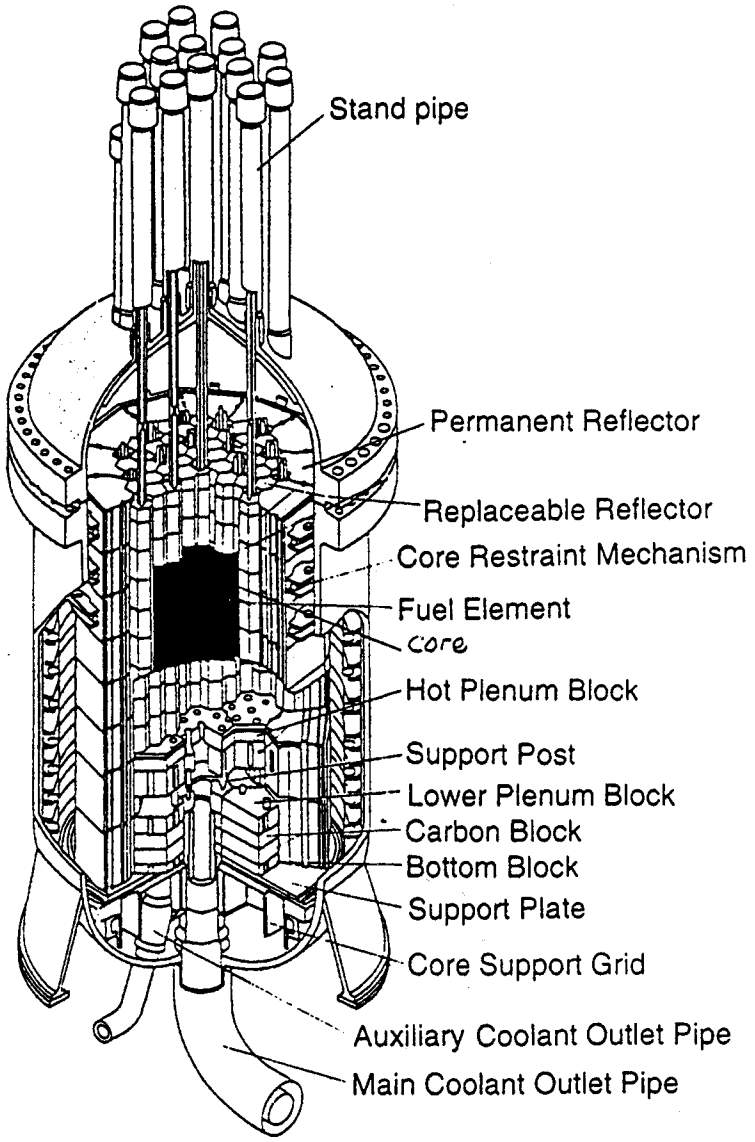


Figure 10: The reactor HTTR - JAERI



Specification of HTTR

Reactor Thermal Power	30MW
Reactor Coolant	Helium Gas
Reactor Outlet Coolant Temperature	850°C/950°C
Reactor Inlet Coolant Temperature	395°C
Primary Coolant Pressure	4MPa
Core Material	Graphite
Core Height	290cm
Core Diameter	230cm
Average Power Density	2.5W/cc
Fuel	Low Enriched UO_2
Fuel Element Type	Prismatic Block
Uranium Enrichment	3~10%
Average	6%
Reactor Pressure Vessel	Steel (2 1/4Cr-1Mo)
Number of Main Cooling Loop	1

Figure 11: Specification of HTTR

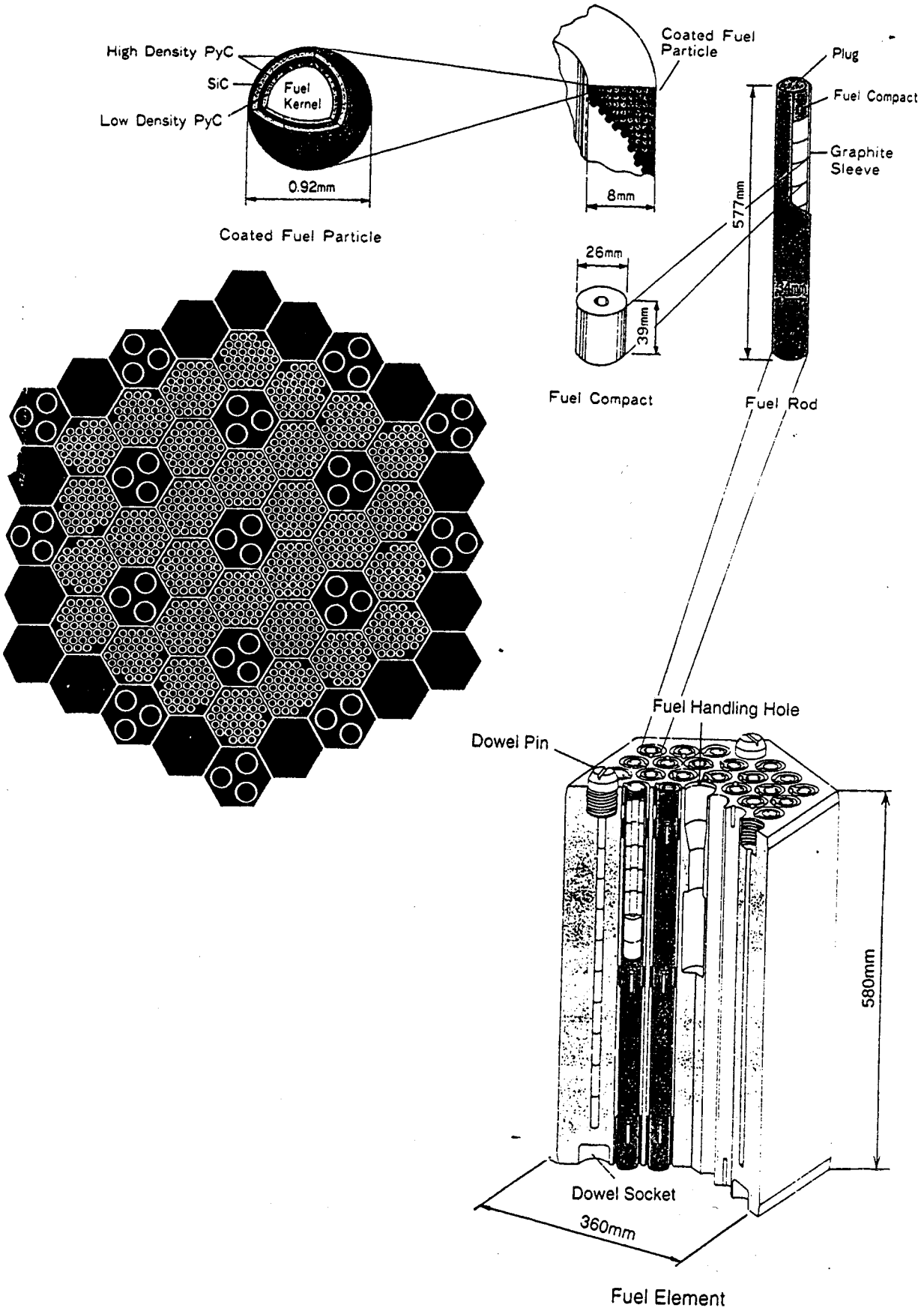
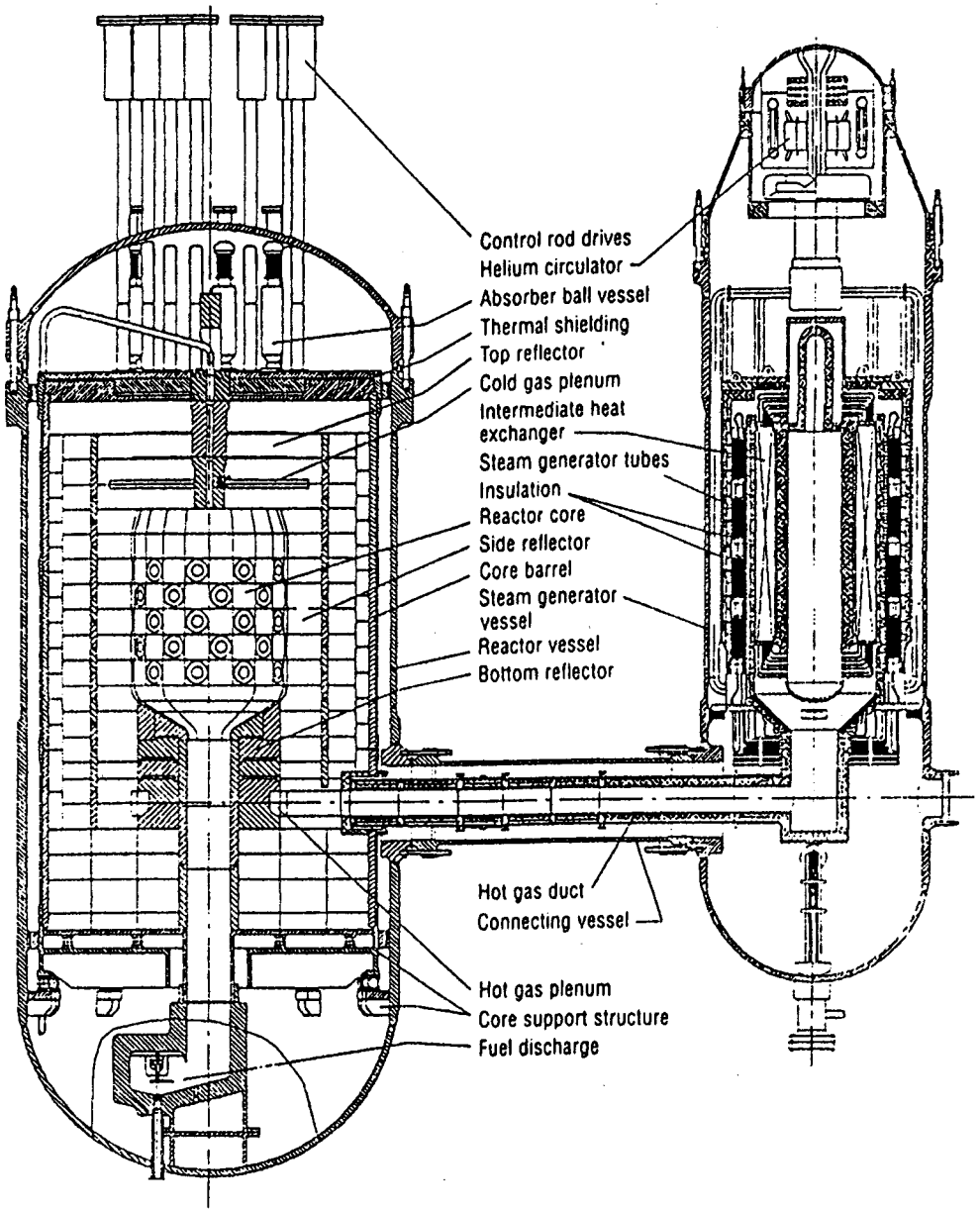


Figure 12: Fuel and core arrangement of the JAERI's HTTR



Main design parameters of HTR-10

Reactor thermal power	10 MW
Primary helium pressure	3.0 MPa
Reactor core diameter	180 cm
Average core height	197 cm
Average helium temperature at reactor outlet	700°C
Average helium temperature at reactor inlet	250°C
Helium mass flow rate at full power	4.3 kg/s
Main steam pressure at steam generator outlet	4.0 MPa
Main steam temperature at steam generator outlet	440°C
Feed water temperature	104°C
Main steam flow rate	3.47 kg/s
Number of control rods in side reflector	10
Number of absorber ball units in side reflector	7
Nuclear fuel	UO_2
Heavy metal loading per fuel element	5 g
Enrichment of fresh fuel element	17%
Number of fuel elements in reactor core	27 000
Fuel loading mode	multi-pass

Figure 13: The HTR-10 design

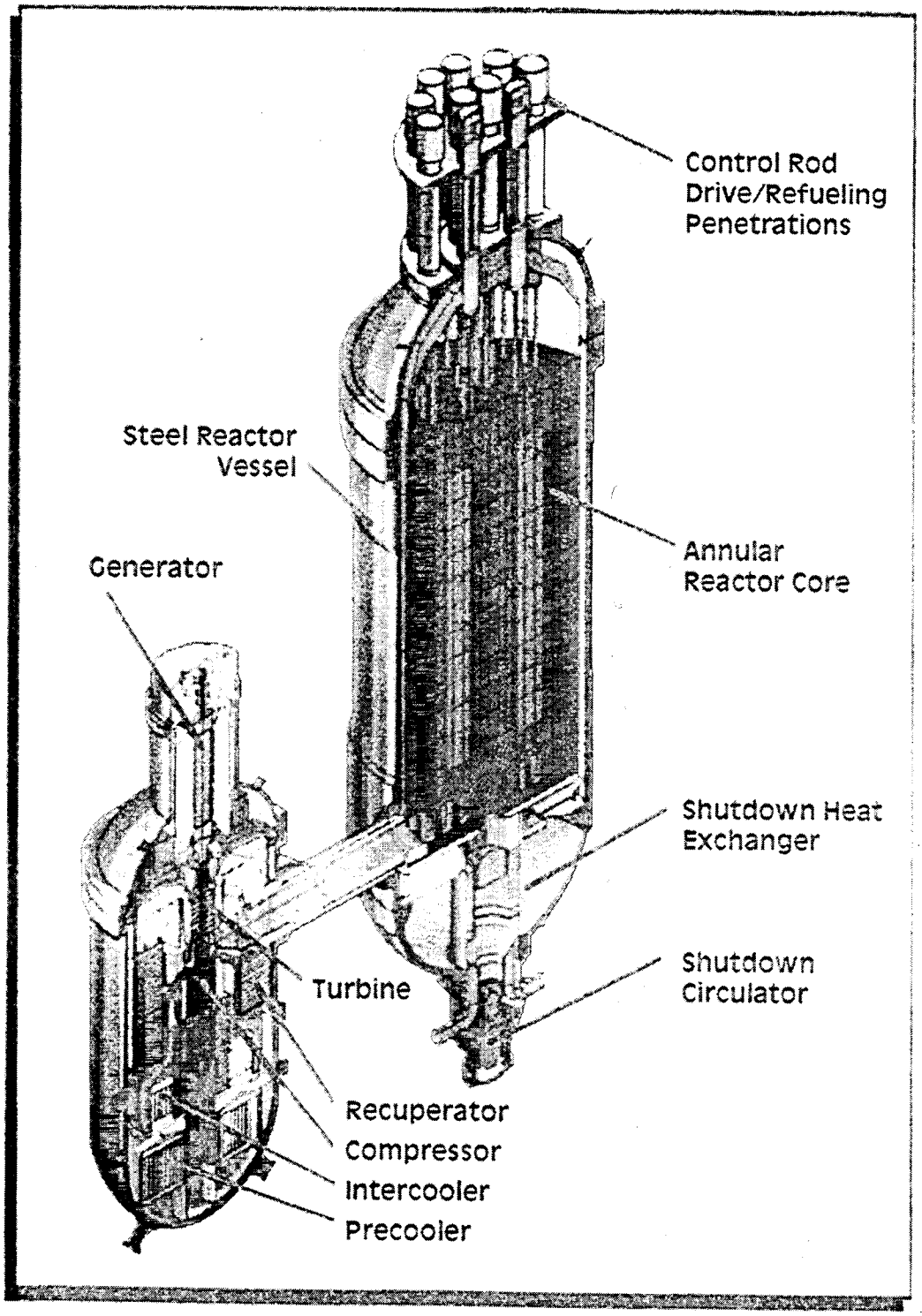


Figure 14: The very compact concept MHR of General Atomic (USA)

3.4. Heavy Water Moderated Reactors (HWR)

Heavy water is an excellent moderator material, particularly where economy is sought in the use of fissile material, because of its relatively low capture of neutrons. In the best known form of Heavy Water Reactor (CANDU), the heavy water is contained in a vessel, "calandria", through which pass a number of tubes providing channels to contain the fuel and the flowing coolant. In CANDU, heavy water is also used as the coolant, under pressure. The neutron economy with this type of reactor is so good that in many commercial applications the fuel supplied can be natural uranium.

Introducing thorium and U-233 recycle requires the introduction of reprocessing and also more expensive, because remote, fuel fabrication, the cost of which must be offset against any other benefit. This, in turn, is likely to require extended fuel irradiation to reduce the impact of those costs, though as a result some of the neutronic benefit will be lost due to increased neutron capture in fission products. Nevertheless, it appears that CANDU would be a relatively promising candidate for the application of the thorium cycle.

A fuel cycle can be conceived which is self-supporting in that enough U-233 is created to replace that which is fissioned, though it remains necessary to provide fissile material from elsewhere (U-235 or plutonium) to provide inventories for new construction. To achieve this self-support, fuel burn-up has to be limited to about 13 000 MWd/t HM. This is called SSET (Self Sustaining Equilibrium Thorium Cycle). Depending on the various costs involved, however, it will be more economic to aim for higher fuel burn-up and accept the need for a continuing supply of fissile material "driver" for topping up. Taking a "high burn-up" case of 40 000 MWd/t HM and assuming that its plutonium requirements are to be supplied by natural uranium CANDUs, the thorium-using reactors would use only about 30 tonnes Th, plus 140 Kg Pu per year, (generated by 45 tonnes of natural uranium) compared to 140 tonnes of natural uranium per GW-year for a once-through natural uranium cycle. (176) (7)

It would appear that if uranium prices rise sufficiently more than the other costs involved, a thorium cycle would become preferable, or at least on a par with the existing natural uranium once-through cycle.

This reasoning is actively pursued by India which has considerable thorium reserves and relatively much less uranium. The utilisation of thorium for power production has been recognized to be the main long-term objective of the Indian

Nuclear Power Programme right from its inception. A sustained R&D programme is thus being pursued on all aspects related to thorium fuel cycle. (142).

A critical facility using uranyl (U-233) nitrate solution (PURNIMA-2) was commissioned in the eighties. PURNIMA-3, which went critical in November 1990, is another unique critical facility based on U-233/Al alloy fuel. Work is in an advanced stage for a 30 kW reactor at Kalpakkam, **KAMINI**, which would also be based on U-233/al alloy fuel. (9). Figure 1 shows one of the 9 20 % U-233/Al fuel assemblies used for PURNIMA 3 and now for KAMINI (142).

Irradiation of ThO2 and (Th-Pu)O2 clusters, development of advanced fuel fabrication and reprocessing techniques for thorium/uranium fuels are also being pursued in India.

Studies have been carried out there to use the thorium/uranium fuel cycle in the existing PHWR System. Short-term possibilities on the irradiation of thorium in power reactors for purpose of initial power flattening in PHWR as well as once-through mode of thorium irradiation in PHWR cores, along with plutonium recycle, have also be examined.

A design of a new reactor based on thorium, the **Advanced Heavy Water Reactor** (AHWR) has also been initiated in India (8) where the Safety Report is underway. This reactor, of 750 MWt thermal capacity would use MOX (U-Pu) fuel as driver and ThO2 as blanket fuel in the same fuel element. The pressure tube design is maintained, with improvements to be able to remotely change the tubes if necessary during the reactor lifetime. The calandria would be vertical. The MOX driver fuel permits to use ordinary water as a coolant, which could permit to operate a steam turbine in line. An overall negative reactivity void coefficient is obtained, as well as other passive safety systems. Figure 2 gives a concept of AHWR fuel.

Research goes on also in Canada but, so far, no CANDU has been converted to a thorium fuel cycle. (140) (146) 148)

The following Table 1 gives theoretical characteristics of U/Th HWRs (IAEA INFCE 1980).

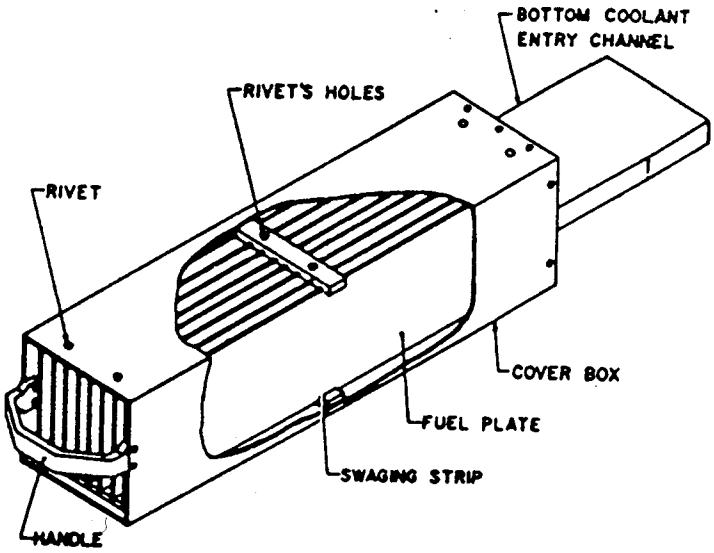


Figure 1: Schematic fuel subassembly of PURNIMA3/KAMINI (142)

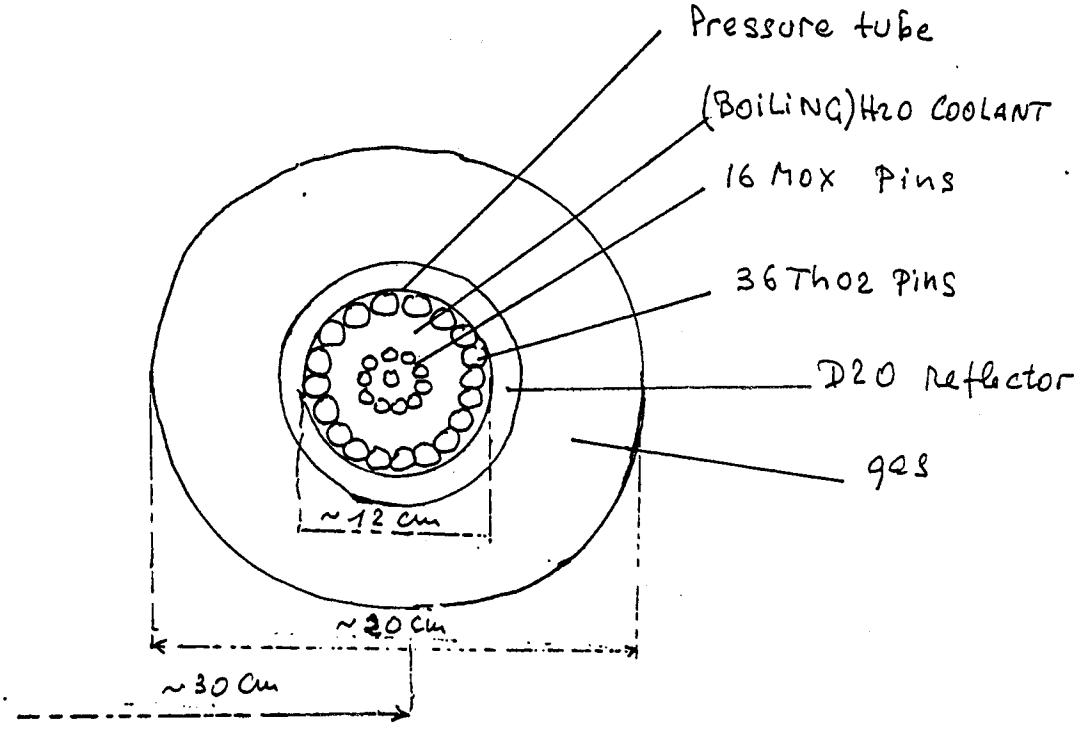


Figure 2: Concept of AHWR fuel element

Table 1: Fuel cycle information - HWRs (16)

	Canada				Germany, F.R.	India		Romania			USA
	1a	1b	2a	2b	3	4	5	6	7a	7b	8
Fuel cycle	HEU-235/Th	Pu/Th	HEU-235/Th	Pu/Th	HEU-235/Th	HEU-235/Th	HEU-233/Th	HEU-235/Th	HEU-233/Th	HEU-233/Th	HEU-233/Th
Topping isotope	^{235}U	Pu-fiss.	^{235}U	Pu-fiss.	^{235}U	^{235}U	^{233}U	^{235}U	^{233}U	^{233}U	^{233}U
Special feature	–	–	SSET	SSET	–	SSET	SSET	–	SSET	–	–
Conversion ratio (av.Eq^m)	0.88	–	1.008	1.008	–	1.0	1.0	1.007	1.002	1.973	–
Fuel residence time (efpd)	1029	1052	380	380	1033	605	1135	404	555	555	525
Annual H.E. replacement (t/GW(e)) (100% cap. factor)	42.7	41.5	125	125	25.2	84.2	41.9	114.4	83.9	83.9	83.3
Eq^m reload enrichment (wt% H.E.)	2.11	2.17	1.64	1.64	3.75	1.46	–	1.71	1.71	1.56	1.65
Out-of-core delay time (y)	2	2	2	2	2	2	2	2	2	2	
Out-of-core fissile loss (%)	1	1	0.5	0.5	1	0.5	–	0.5	0.2	0.2	2.5
Average discharge exposure (MW.d/t)	29 300	30 100	10 000	10 000	45 000	15 000	27 000	11 000	15 000	15 000	14 000
Fissile requirements[a,b] (t/GW(e))											
Initial core	2.760	3.029	2.530	2.780	2.170	2.940	2.015	2.540	2.110	2.000	1.859
Annual Eq^m, recycled	0.5	.472	1.44	1.44	.397	0.885	–	1.37	.974	.895	.856
Annual Eq^m, makeup	0.129	.162	0	0	.231	0	–	0	0	.027	.106
30-y cumulative, gross	7.54	8.78	5.93	6.52	9.663	–	–	5.74	4.058	4.566	6.52
30-y cumulative, net	4.25	4.81	0.47	0.72	6.797	–	–	0	0	.755	3.17
Natural U requirements[a,b] (t/GW(e))											
Initial core	543	N.A.	494	N.A.	430	574	N.A.	500	N.A.	N.A.	N.A.
Annual Eq^m	24.7	N.A.	0	N.A.	52.5	0	N.A.	0	N.A.	N.A.	N.A.
30-y cumulative, gross	1489	N.A.	1158	N.A.	2110	–	N.A.	1136	N.A.	N.A.	N.A.
30-y cumulative, net	–	N.A.	–	N.A.	1529	–	N.A.	0	N.A.	N.A.	N.A.
Th requirements[a,b] (t/GW(e))											
Initial core	117	115.1	128.0	127.4	69.0	392	–	124.4	125.2	104.7	105.4
Annual Eq^m makeup	28.7	28.2	82.1	82.1	22.3	59	–	79	57.9	50.7	50.4
30-y cumulative, makeup	207	205	336	334	716	1770	–	2445	1820	1590	–
30-y consumptive, net	32	32	36	36	–	27	–	40	32	28	–
SWU requirements[a,b] (t/GW(e))											
Initial core	704	N.A.	641	N.A.	544	–	N.A.	648	N.A	N.A.	N.A.
Annual Eq^m	32	N.A.	0	N.A.	67.9	–	N.A.	0	N.A.	N.A.	N.A.
30-y cumulative, gross	1932	N.A.	1496	N.A.	2716	–	N.A.	1476	N.A.	N.A.	N.A.
30-y cumulative, net	–	N.A.	–	N.A.	1964	–	N.A.		N.A.	N.A.	N.A.
Fissile material in spent fuel[a]											
Enrichment (Eq^m) (%)											
U-fiss./U	85	–	–	–	57.0	89.6	–	68.6	68.6	10.4	11.2
Pu-fiss./Pu	75.0	–	–	–	–	–	–	N.A.	N.A.	45.5	67.7
Annual Eq^m discharge (kg/GW(e))											
U-fiss.	505	–	–	–	397	866	–	1360	1000	882	848
Pu-fiss.	1.8	–	–	–	–	0	–	0	0	52	22.8
30-y fissile material discharge (kg/GW(e))											
U-fiss.	15000	–	–	–	11900	25980	–	42200	31400	27800	26.02
Pu-fiss.	55	–	–	–	–	0	–	0	0	1600	706

[a] Normalized to 70% capacity factor and 0.2% tails enrichment.

[b] 30-year net cumulative requirements are 30-year cumulative gross requirements less in-core and ex-core inventory at the end of 30-years of operation which can be used in other reactors.

3.5. Light Water Reactors (LWR)

Similar remarks as those for HWR apply to the use of thorium in conventional Light Water Reactors (LWR). In these, the moderator is "ordinary" water, and serves at the same time as the coolant. Hydrogen has a relatively high neutron capture, but as it is of the same mass as the neutron, the energy of the latter is reduced, per collision, by the greatest possible amount. This leads to a compact core arrangement, at the cost of neutron economy and a need for fuel enrichment. Most reactors currently operating or under construction are of this type. There being two sub-varieties, the Pressurised Water Reactor (PWR) in which the heat is transferred from the coolant in a steam generator to a separate steam/water circuit driving the turbine, and the Boiling Water Reactor (BWR) in which the coolant is made to boil in passing through the reactor core and the steam formed is taken directly to the turbine. More attention has been given to the use of thorium in the PWR than in the BWR.

The BWR Elk River Reactor (ERR) of 24 MWe (20) was built by Allis-Chalmers under contract to the USAEC for the Rural Cooperative Power Co. It operated between 1963 and 1968 and achieved an honourable career with 65 % availability. (Fig.1).

It was fuelled with "standard" box and pin fuel elements, with cruciform control rods in between. It operated on a U-235 - U-238/Th once-through fuel cycle, with U-235 content of 4.3 % and 5.2 % in weight of total ThO2 + UO2 pellets. (Fig. 1).

Experimental reprocessing of the spent fuel elements and experimental fuel refabrication, has been done in the Rotondella plant built by Allis-Chalmers for the CNEN in Southern Italy (PCUT Project). (153)

The PWR at Indian Point was a 285 MWe reactor built by Babcock and Wilcox for Consolidated Edison, and was operated between 1962 and 1980 very successfully with an excellent availability factor. (179)

It has also used UO2/ThO2 fuel elements for a time, but this was rather quickly abandoned on economic grounds.

Although U-235 driver requirements would be greater than in HWRs or HTRs, there could be interest in the possibility of back-fitting to a thorium cycle, should there be a sharp change in uranium prices. Over the period required for the

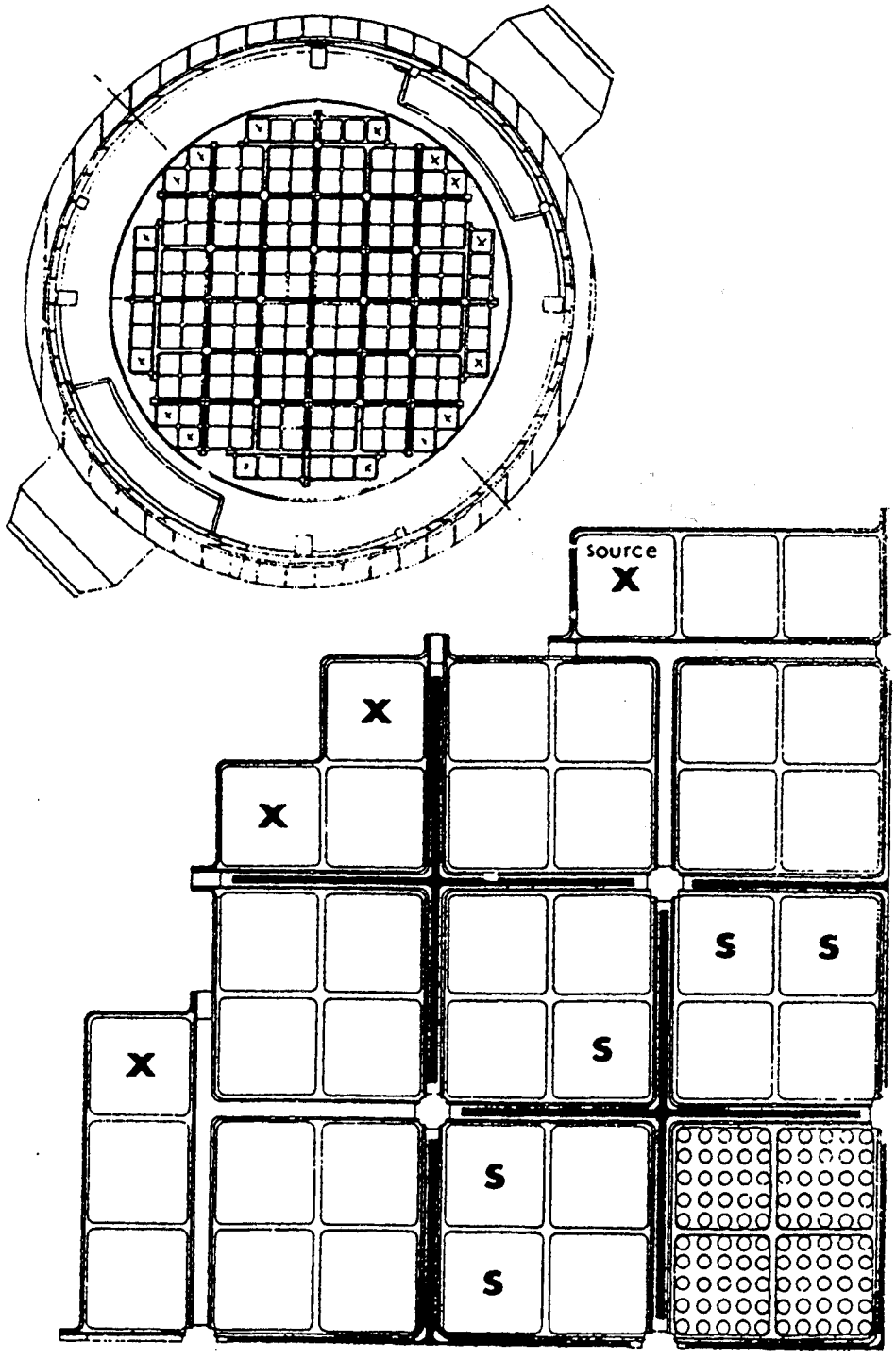


Figure 1: Characteristics of the Elk River reactor: core loaded with 128 regular elements enriched 4.3%, 20 spiked elements (S) enriched 5.2% and 16 dummy elements (X)

development and introduction of a thorium cycle, however, improvements would be carried out in the performance of LWRs on the U/plutonium cycle and this must be allowed for in making a comparison. The adoption of thorium with U-233 recycle might further reduce the U-235 requirement by 20-25 %. The increase in fuel fabrication costs due to the gamma-activity caused by the associated U-232, and the incidence of thorium/U-233 reprocessing costs, though greatly uncertain, seem unlikely today to allow this course to become decisively economic, especially when with plutonium recycle (MOX fuels), a similar level of U-235 requirement can be achieved on the U-238 cycle. (After Thorn and al, 1976)

The important **German-Brazilian contribution** (12) to thorium utilization in PWRs has to be mentioned here (1979-1988). It should be remembered for the future as a reference work. A large amount of theoretical and some practical research with experimental fuel pins, has concluded to the technical feasibility of backfitting existing PWRs for a thorium cycle with U-235 - U-238 or Pu spiking, as mixed oxides. (Fig. 2 and 3). Considerable work has been performed on the metallurgical and irradiation behaviour of such fuels, with a good degree of confidence. The conclusions were that for once-through operation, the U/Th cycle was competitive with a U/U cycle. Similar conclusions were achieved by a French EDF study (175).

More theoretical, the possibility to play on the hardness of the neutron spectrum to increase the breeding power, and still take advantage of the good epithermal characteristics of U-233, has been investigated. This is the so-called Spectral-Shift Controlled Reactor (SSCR) whereby reactivity control could be obtained by introducing variable quantities of heavy water which would harden the neutron spectrum. But separating D2O from H20 is not a simple proposition !

A radically new version aiming at using thorium with great emphasis on neutron economy has been tested in the reconverted **Shippingport PWR reactor** in the USA, in what is called the Light Water Breeder Reactor (LWBR). The old 100 MWe Shippingport reactor, built by Westinghouse for the USAEC, has been a faithful reactor from 1957 until 1974. Development studies were carried out in the 1960s to adapt it to a U-233/Th cycle. Practical tests were carried out from the end of the 60s until 1974 when the reactor was shut down and has since been fully dismantled. The core was fitted with about 40 t U-233/Th fuel elements.

In the LWBR concept, the aim is to achieve high U-233 production, perhaps sufficient to give a small breeding gain, and the project was successful indeed in

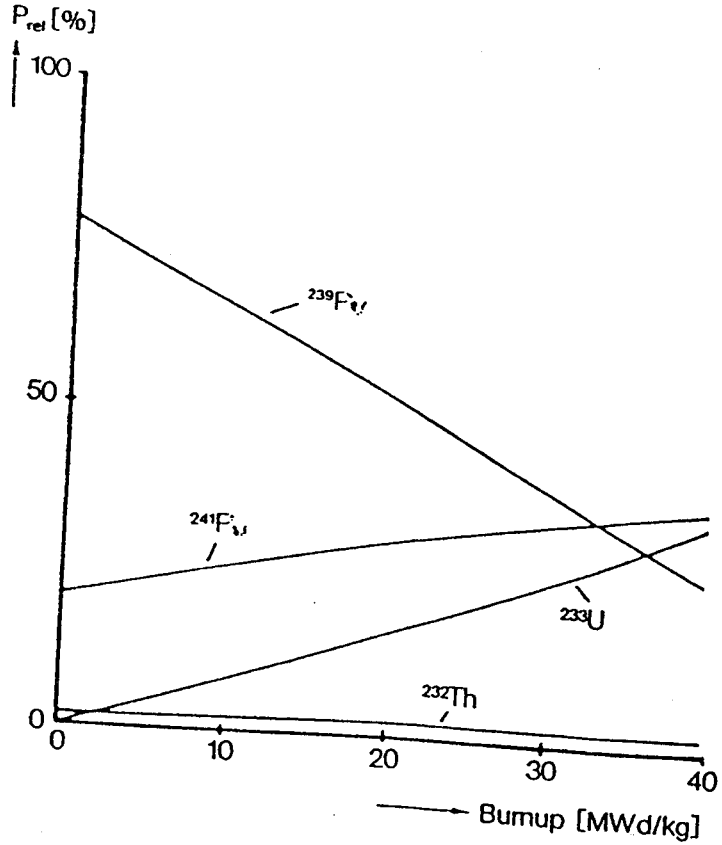


Figure 2: Relative energy contribution of the isotopes of Th/Pu fuel (12)

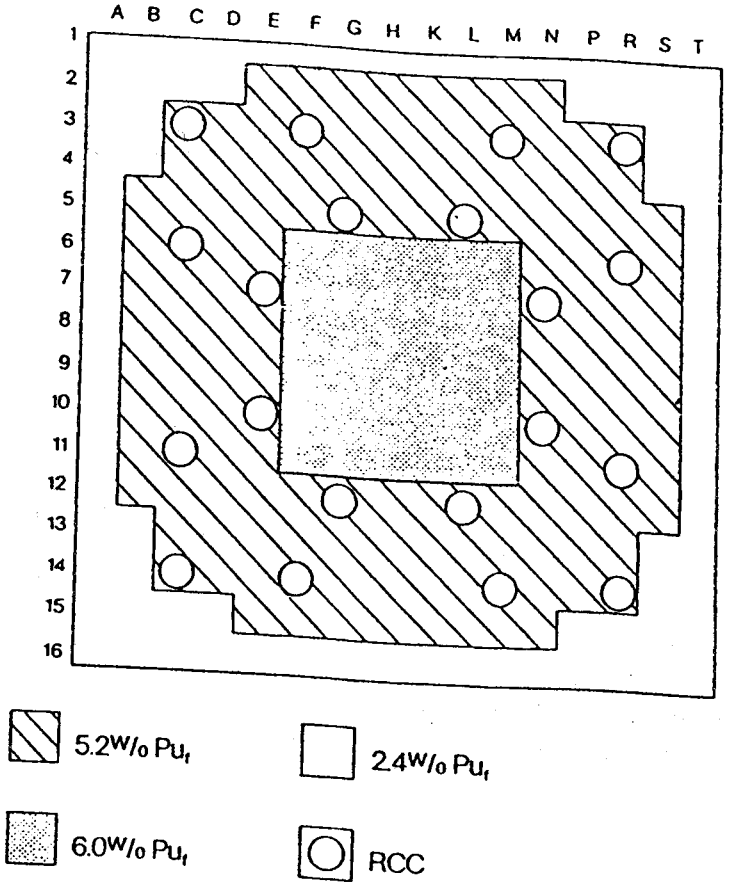
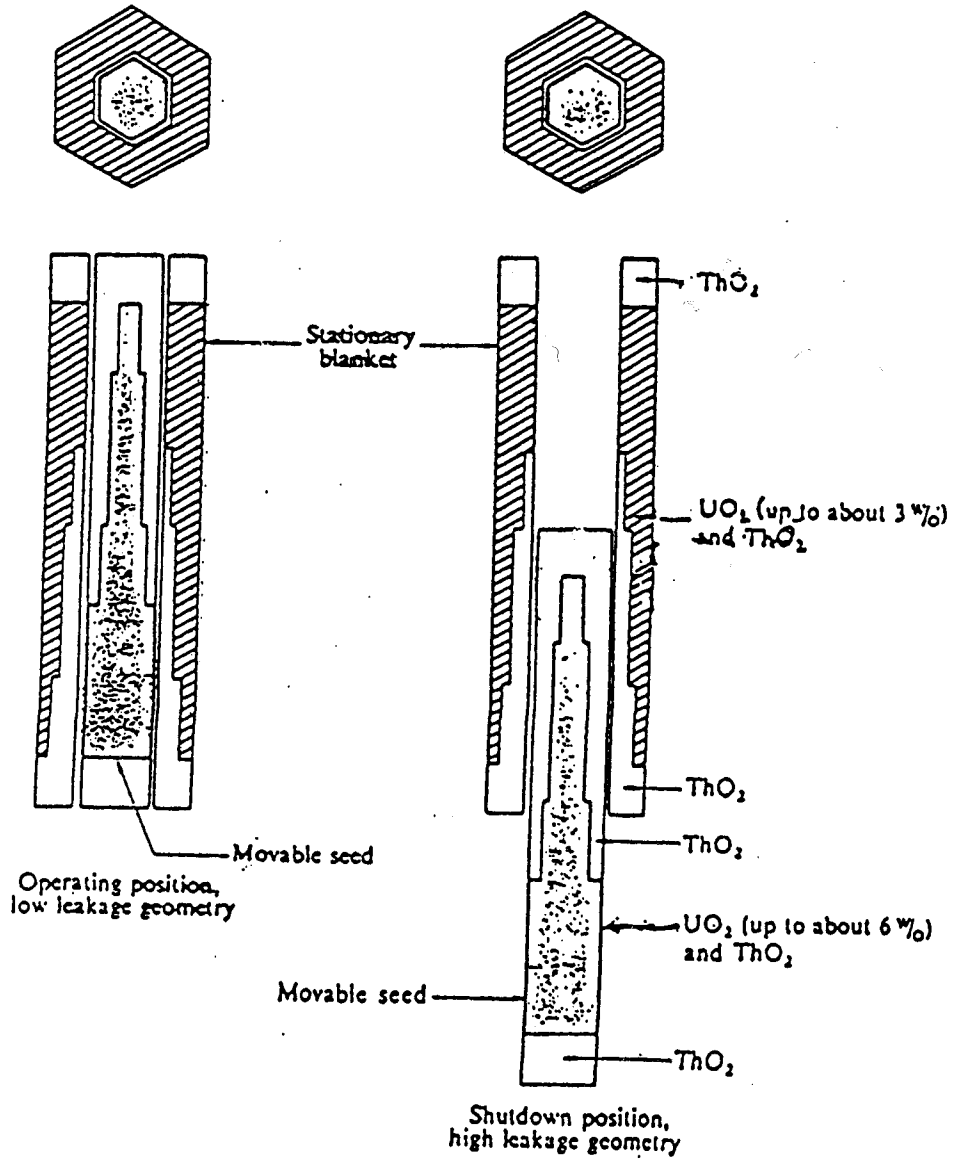


Figure 3: Th/Pu fuel assembly with 3 enrichment zones (12)

demonstrating a small breeding gain. To do this, the design adopts a lower moderator ratio, that is to say a lower ratio of water to fuel volume of about half that used in conventional PWR. There is therefore a reduced area available for coolant flow requiring either greater pumping power or a longer core with a lower power density. A lower power density also contributes further to improving the conversion ratio but requires a higher initial fissile loading. The Shippingport core has two other special features to increase the possibility of breeding being achieved. The fuel charge is of the "seed-blanket" type.(Fig. 4). That is to say, there are two types of fuel pins in a module. One, the seed, has a higher U-233 content, and is the primary producer of neutrons, and the blanket is the primary fertile component. The other special feature is that reactivity control is provided by moving the seed fuel, so avoiding the loss of neutrons that would occur in conventional absorber control rods. Back-fitting of a complete core to existing reactors might be theoretically possible but would require derating in addition to the rejection of the previous fuel charge and an appreciable outage time.

The level of conversion ratio aimed for is similar to that of the "self-sufficient" CANDU or Indian SSET versions already mentioned and requires the initial fissile charge to be U-233 which must be supplied from other reactors, called pre-breeders, using thorium and either U-235 or plutonium. The "special tricks" necessary with the LWR are symptomatic of the lesser neutron economy compared with HWRs.

Table 1 (16) summarizes the characteristics of "standard" LWRs with thorium, extrapolated from the experience mentioned above (IAEA, INFCE 1980).



– Variable geometry control concept. (Courtesy USAEC.)

Figure 4: "Seed-Blanket" system

Table 1: Fuel cycle information - LWR's (16)

	GERMANY, F. R.		USA		USA
	1a	1b	2a	2b	3
Fuel cycle	HEU-^{235}U/Th	MEU-^{235}U/Th	MEU-^{235}U/Th	MEU-^{233}U/Th	MEU-^{233}U/Th
Topping isotope					
Special feature	PWR current design				SSCR
Conversion ratio (av.Eq[m])					0.83
Refuelling interval (years)	1	1	1.07	1.07	1.07
Fraction of core replaced/refuelling	0.33	0.33	0.33	0.33	0.33
Annual H.E. replacement (100% cap. factor), t/GW(e)	32.9	33.4	32.7	32.7	32.9
Eq[m] reload enrichment (%)	3.91	3.95	4.43	3.37	2.89
Out-of-core delay time (years)	2	2	2	2	2
Out-of-core fissile loss (%)	1	1	1.5	1.5	1.5
Average discharge exposure (MW.d/t)	34 000	33 500	33 400	33 400	33 400
Peak pellet exposure (MW.d/t)	-	-	55 000	55 000	-
Fissile requirements[a,b] (t/GW(e))					
Initial core	2.544	2.544	2.598	1.904	1.704
Annual Eq[m], recycled	0.558	0.558	0.610	0.478	0.512
Annual Eq[m], makeup	0.337	0.375	0.404	0.295	0.229
30-y cumulative, gross	13.433	14.535	15.315	11.163	9.454
30-y cumulative, net	10.306	11.370	12.045	9.575	6.726
Natural U requirements[a,b] (t/GW(e))					
Initial core	497	493	512	N.A.	N.A.
Annual Eq[m]	66	73	81	N.A.	N.A.
30-y cumulative, gross	2630	2827	3040	N.A.	N.A.
30-y cumulative, net	2015	2211	2531	N.A.	N.A.
Th requirements[a,b] (t/GW(e))					
Initial core	73	63	78.9	57.9	69
Annual Eq[m]	28.6	21.3	22.1	16.2	19.6
30 y cumulative, gross	902	681	773	539	641
30 y cumulative, net	-	-	-	-	641
SWU requirements[a,b] (t/GW(e))					
Initial core	644	715	604	N.A.	N.A.
Annual Eq[m]	86	86	100	N.A.	N.A.
30-y cumulative, gross	3422	3465	3788	N.A.	N.A.
30-y cumulative, net	2625	2739	3291	N.A.	N.A.
Fissile material in spent fuel[a,b]					
Enrichment (Eq[m]) (%)					
U-fiss./U	63	9.3	11.7	7.9	9.2
Pu-fiss./Pu	73	75	69.4	74.7	76.5
Annual Eq[m] discharge (kg/GW(e))					
U-fiss.	541	530	594	478	446
Pu-fiss.	5	68	60	65	71
30-y fissile material discharge (kg/GW(e))					
U-fiss.	16230	15900	17462	13705	13380
30-y cumulative material discharge (kg/GW(e))					
Pu-fiss.	140	2040	1691	1789	2656

[a] Normalized to 70% capacity factor and 0.2% tails enrichment

[b] 30-year net cumulative requirements are 30-year cumulative gross requirements less in-core and ex-core inventory at the end of 30 years of operation which can be used in other reactors.

3.6. Once-through Cycles (176)

All the thermal reactor types discussed above (HTR, HWR, LWR) are capable of operating on one-through cycles with uranium fuelling. There is an inevitable loss of fissile material in such a scheme, though reprocessing costs are saved, and if the fissile material is cheap enough such cycles can be economic. The only major answer is to utilise more of the fissile material in one pass through the reactor. In an idealised situation, a reactor which was self-sustaining in reactivity could operate indefinitely, and the loss of fissile material on discharge could be confined to the last charge on decommissioning the reactor. Technology is still a long way from achieving a fuel life comparable with the life of the reactor plant, and indeed it is hardly to be expected that this will be possible, in view of the relatively intense irradiation damage experienced by fuel element materials. However, if fuel could be developed to withstand a sufficiently high irradiation dose, then benefit could be taken from a fuel cycle in which there was sufficient breeding to compensate for the loss of reactivity caused by neutron absorption in fission products so that the reactivity of the core was maintained. In thermal reactors, this can be most closely approached with a Th/U-233 cycle.

Theoretical studies of such cycles have been carried out but alternative ways of meeting fissile material supply limitations by adopting reprocessing and recycle appear much more practicable. The nearest to being a possible exception to this is the HTR, and especially the pebble-bed reactor type, but even in this case, because of the higher inventory of fissile material which is required for such a Th/U-233 cycle, with current technology it proves possible to do only slightly better than with the once-through uranium/plutonium cycle. (176).

3.7. Fast Reactors

From the fuel economy standpoint, fast reactors are expected to be breeders. With thorium, the breeding gain is less good because, although U-233 has some advantages over U-235 and Pu-239, they are insufficient to compensate for the very low fission probability in Th-232. Indeed, with the sole use of thorium and U-233, the breeding gain (i.e. breeding ratio minus unity) in a study of representative reactor designs was only 0.04 compared with 0.28 when using the uranium/plutonium cycle. Furthermore, because of the negative reactivity effect of thorium in the core, a higher enrichment and a larger fissile inventory are required, so that the doubling time became 112 years compared with 16 years with plutonium.

Thus, it would seem that a fast reactor using a Th/U-233 cycle presents no interest over the Light Water Breeder mentioned above, even if the sodium void coefficient is lower for the thorium fast reactor than with uranium.

However, to breed U-233 efficiently, a plutonium fuelled fast reactor with a thorium oxide blanket will be the best choice.

This principle is put to application in the Indian Fast Breeder Test Reactor at Kalpakkam, being fitted with thoria radial and axial blanket (Figs. 1 and 2) (141)

Table 1 gives characteristics of some fast breeder designs where it can be seen that the Pu/Th arrangement gives a good fissile gain in U-233.

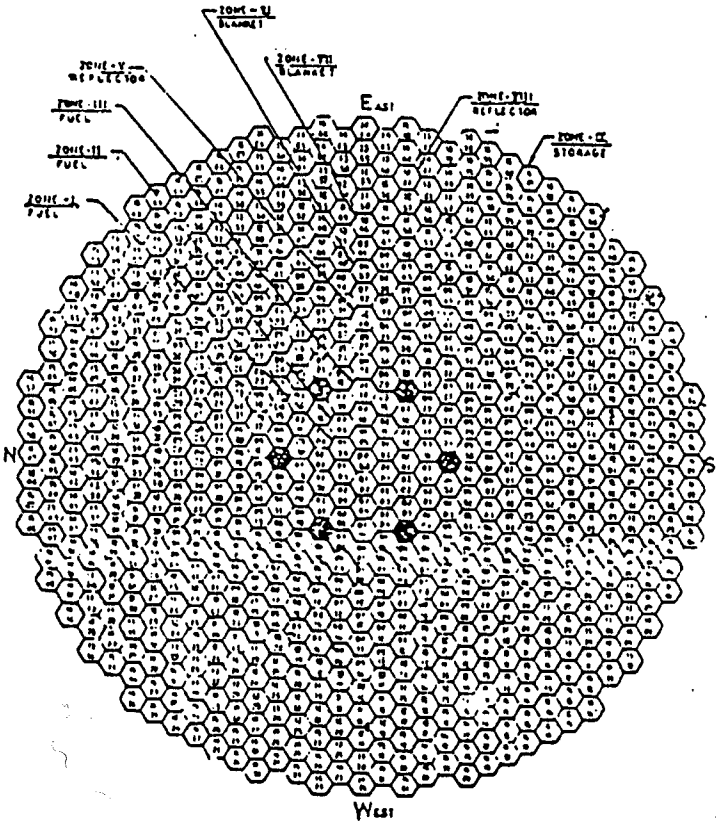


Figure 1: FBTR (India) core configuration

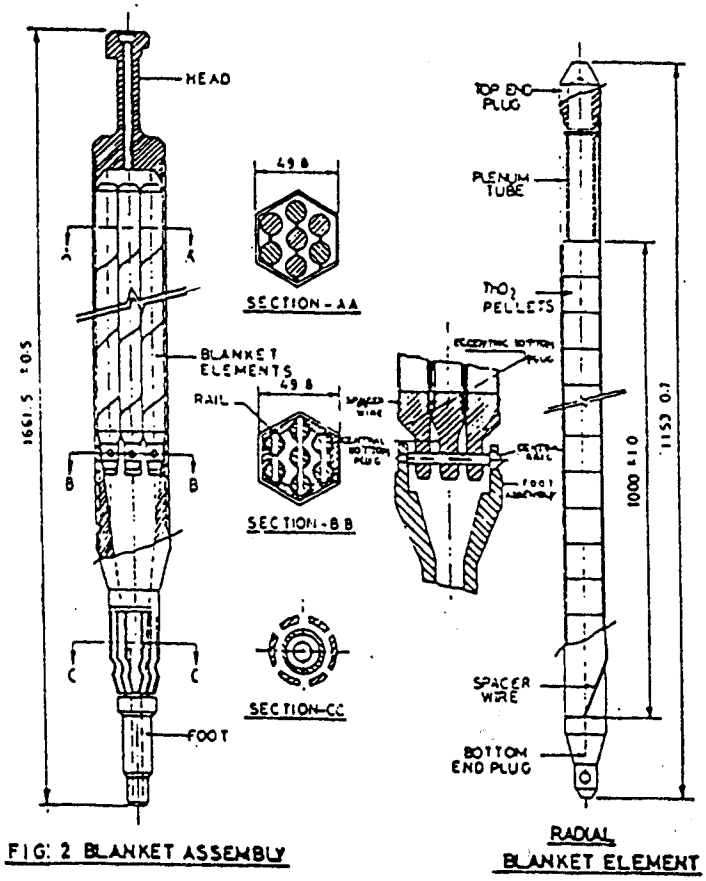


Figure 2: Blanket element and assembly of the FBTR (India)

Table 1: Summary of Breeding Performance and Mass Flow Data for Alternative Fast Breeder Fuel Cycles (16)

TABLE 1. SUMMARY OF BREEDING PERFORMANCE AND MASS FLOW DATA FOR ALTERNATIVE FAST BREEDER FUEL CYCLES (16)
(1 GW(e) generating capacity, 0.75 capacity factor)

Main characteristics	Homogeneous core							Heterogeneous core		
Core material	Pu-U	Pu-U	Pu-U	Pu-U	Pu-Th	U-Th	U-U	Pu-U	Pu-U	Pu-U
Blanket material										
Axial	U	U	U	Th	Th	Th	Th	U	U	Th
Radial	U	U	Th	Th	Th	Th	Th	U	U	Th
Fuel cycle residue (%)	1	1	2	2	2	2	2	1	1	2
External cycle time (years)	2	1	2	2	2	2	2	2	1	2
Oxide fuel LMFBR										
Fissile inventory (kg)										
Core (initial load)	3158	3158	3184	3198	3659	3304	2900	3926	3926	4064
Cycle	6315	4736	6367	6396	7317	6608	5799	7852	5889	8128
Annual net fissile gain (kg/a)										
Uranium	0	0	133	292	740	43	-301	0	0	531
Plutonium	245	252	81	-93	-669	0	457	282	290	-335
Total	245	252	214	199	72	43	156	282	290	196
Annual consumption (kg/a)										
^{238}U	1420	1420	1359	962	0	0	876	1532	1532	607
^{232}Th	0	0	324	687	1496	1374	684	0	0	1171
Total	1420	1420	1683	1649	1496	1374	1560	1532	1532	1778
Breeding gain[a]	0.325	0.325	0.314	0.305	0.184	0.099	0.240	0.382	0.382	0.331
System doubling time (years)	17.8	13.1	20.6	22.3	70.8	108	25.9	19.3	14.1	28.8
Carbide fuel LMFBR										
Fissile inventory (kg)										
Core (initial load)	2615	2615	2643	2655	3075	2903	2511	3800	3800	4034
Cycle	5229	3922	5286	5311	6150	5807	5021	7600	5700	8069
Annual net fissile gain (kg/a)										
Uranium	0	0	154	318	734	58	-237	0	0	575
Plutonium	354	359	158	-32	-610	0	454	401	409	-279
Total	354	359	312	286	124	58	217	401	409	296
Annual consumption (kg/a)										
^{238}U	1523	1523	1417	984	0	0	897	1724	1724	568
^{232}Th	0	0	342	701	1457	1326	657	0	0	1267
Total	1523	1523	1759	1685	1457	1326	1554	1724	1724	1835
Breeding gain[a]	0.479	0.479	0.450	0.426	0.223	0.114	0.330	0.561	0.561	0.479
System doubling time (years)	10.2	7.6	11.7	12.9	34.5	70.0	16.0	13.1	9.7	18.9

[a] The different fissile isotopes are equally weighted. Therefore the breeding gain was calculated from the relation: (breeding gain) = (breeding ratio) − 1.

3.8. Advanced or Special Concepts

3.8.1. The Suspension Reactor

An experimental 1 MWth Pressurized Suspension Reactor (KSTR or "SUS POP") was operated in the Netherlands at Kema, Arnhem, by the staff of Dr. J.J. Went between 1974 and 1977. (29)

The underlying ideas were that the suspension of UO2/ThO2 particles in water is as near as possible to a homogeneous reactor with a good neutronic efficiency, a high degree of safety (temperature control, boiling if necessary), breeding capacity, and on-line purification of most of the fission products which are ejected from the fuel microspheres (diameter 5 microns), especially Xe-135 gas which constitutes one of the high-yield and neutrons absorbing fission products; these could be removed on line in the liquid. U-233 would eventually replace U-235.

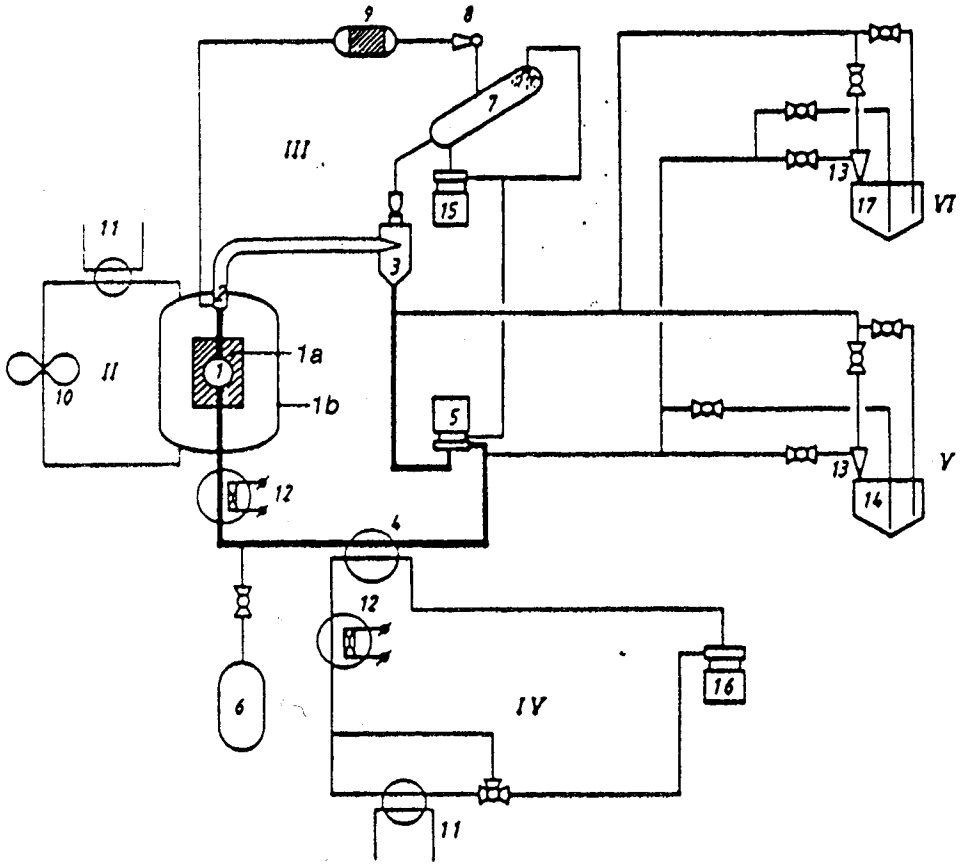
This unusual prototype has been operated successfully, but no extrapolation has been envisaged. There were expected problems with equipment erosion by the suspension, and particle attrition.

Table 1 gives the reactor basic data. Figure 1 gives the operating diagram, Figure 2 shows the reactor vessel arrangement.

The whole reactor has now been dismantled.

Table 1: Basic data (29) KSTR Reactor "SUSPOP", KEMA (29)

Fuel	(22.5% ^{235}U, 2.5% ^{238}U, 75% Th)O_2
Concentration	O-400g/l H_2O
Paritricle size	5 μm
Moderator	H_2O
Reflector	BeO + graphite
Nominal thermal power	1,000 kW
Specific power	50 kW/l
Max. operating temperature	255°C
Max. operating pressure	60 bar
Pressurizing gas	H_2
Reactor vessel configuration	Spherical with conical inlet and outlet sections.
Volume reactor vessel	18.3 l
Reactor vessel diameter	(max.) 310 mm
Mass flow	15 kg/s
Flow speed in pipes	5 m/s



I	main suspension circuit (MS)	5	suspension pump (MP-1)
II	reflector cooling system (RCS)	6	dump vessel (MV-3)
III	gas purification system (GPS)	7	spray condensor
IV	primary cooling water system (PCSA)	8	ejector
V	concentration control system	9	recombinator
VI	fuel supply and discharge system	10	gas pump
		11	heat exchanger
1	reactor vessel (MR-1)	12	electr. heating
1a	reflector	13	hydroclone
1b	pressure vessel	14	storage vessel (MV-10)
2	gas-liquid contactor (MJ-2)	15	auxiliary pump
3	gas separator (MJ-1)	16	cooling water pump
4	main heat exchanger (ME-1)	17	transfer vessel (GV-60)

Figure 1: Diagram of KSTR (29)

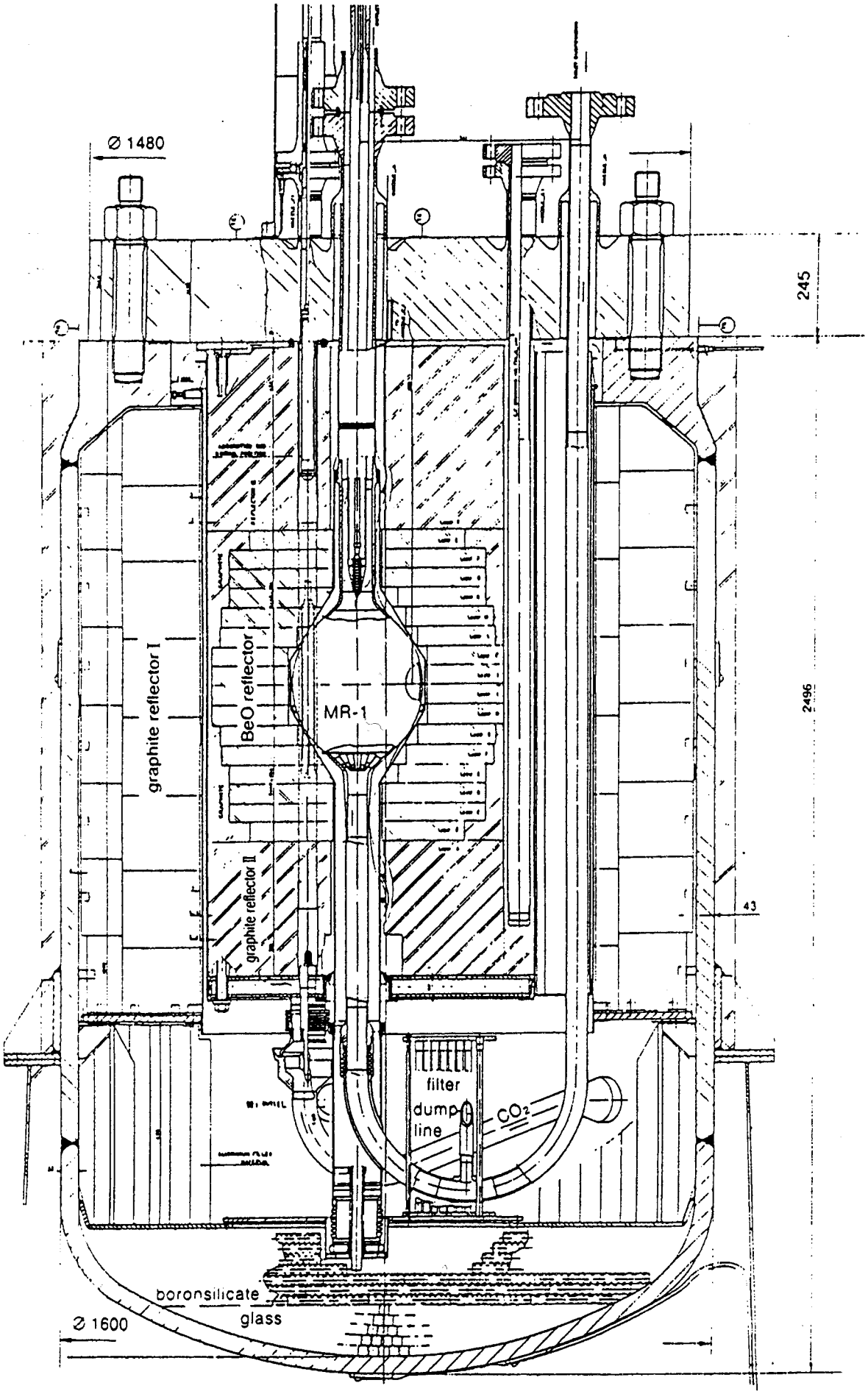


Figure 2: Pressure vessel MV-1 with core vessel MR-1 and reflector (29)

3.8.2. **The Molten Salt Reactor Experiments and Homogeneous Breeder concepts**

The Molten Salt Breeder Reactor (MSBR) is, from the theoretical neutronic standpoint, the most efficient reactor type, as far as fissile material holdup, epithermal reactions with U-233 and thermal breeding into the thorium atoms are concerned.

From a practical standpoint, it should be possible to process the salt continuously to "milk out" the neutron poisons and some extra U-233, leaving the other actinides to be gradually transmuted or being "burnt" by contributing to the fission reaction.

As disadvantages, one can cite possible corrosion problems and the high contamination of the primary circuits by direct contact with the fuel and fission products.

Experience of power generation with such a type of fluid is very limited.

However, this type of reactor has the benefit of significant R&D work carried out in the USA as a part of the aircraft reactor development programme and later in the 8-MW(th) Molten Salt Reactor Experiment **(MSRE)** at Oak Ridge National Laboratory in the US, in the second part of the 1960s. (17) (26)

MSRE is a one-region reactor with a 1.5 m diameter core (Figs. 1 and 2). The fuel is a mixture of Li-7 F, Be F2 and Zr2 F4 with less than 1 mole % U-235 F4. This fuel at about 650°C circulates through a graphite reflector core (Fig. 3). It is circulated at about 300 m^3/h by a centrifugal pump located at the high point of the system, which permits, thanks to a small helium sweep, to purge the volatile fission products noble gases and I-129 which are trapped in a charcoal bed. Feed make-up takes place by introducing small capsules of fresh salt with UF4-LiF eutectic in the system.

The reactor can be drained through a freeze valve and the temperature is adjusted with control rods.

The primary loop is cooled by means of a primary heat exchanger with a secondary salt loop, cooled by an air radiator.

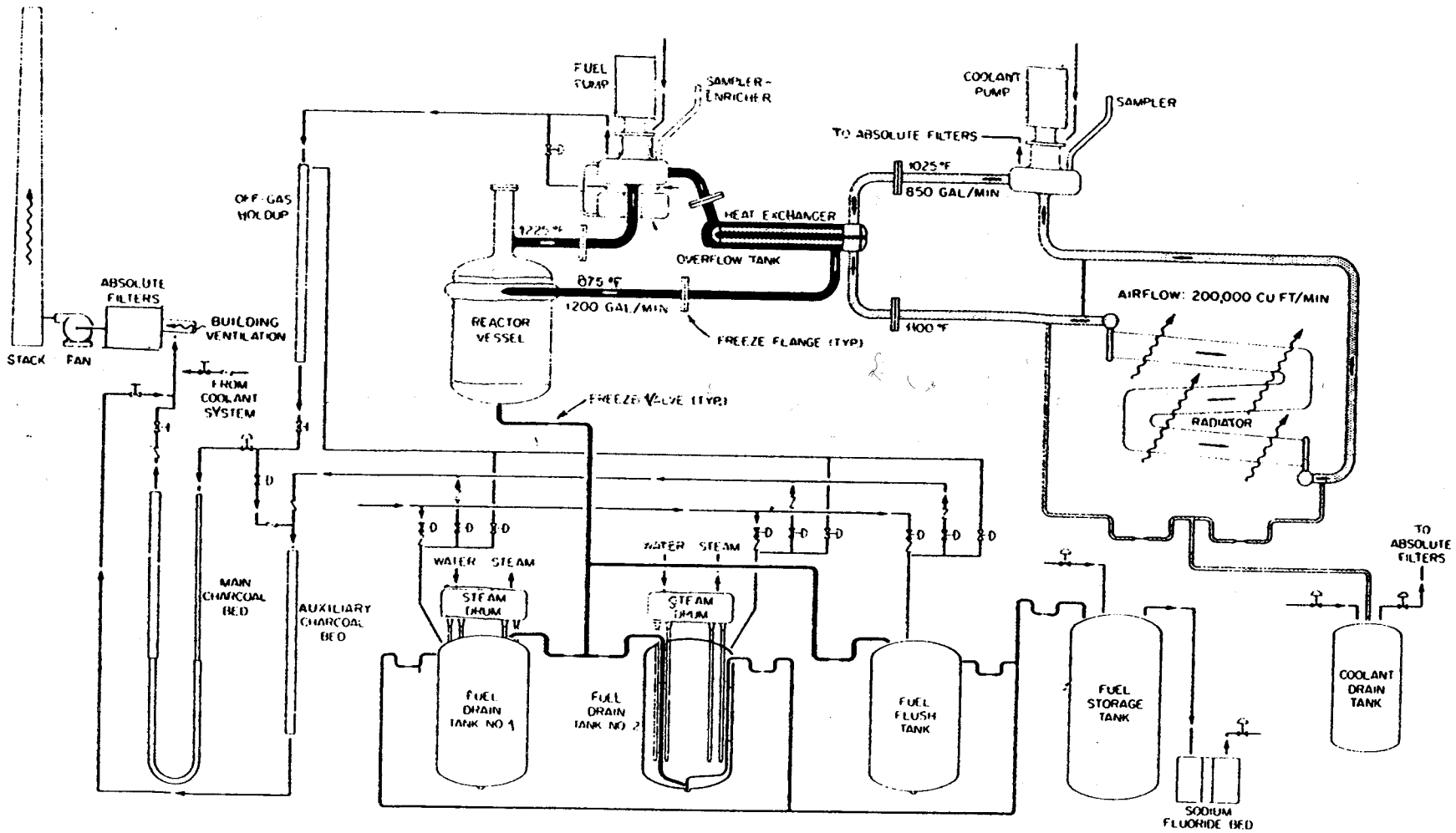


Figure 1: MSRE flow sheet (26)

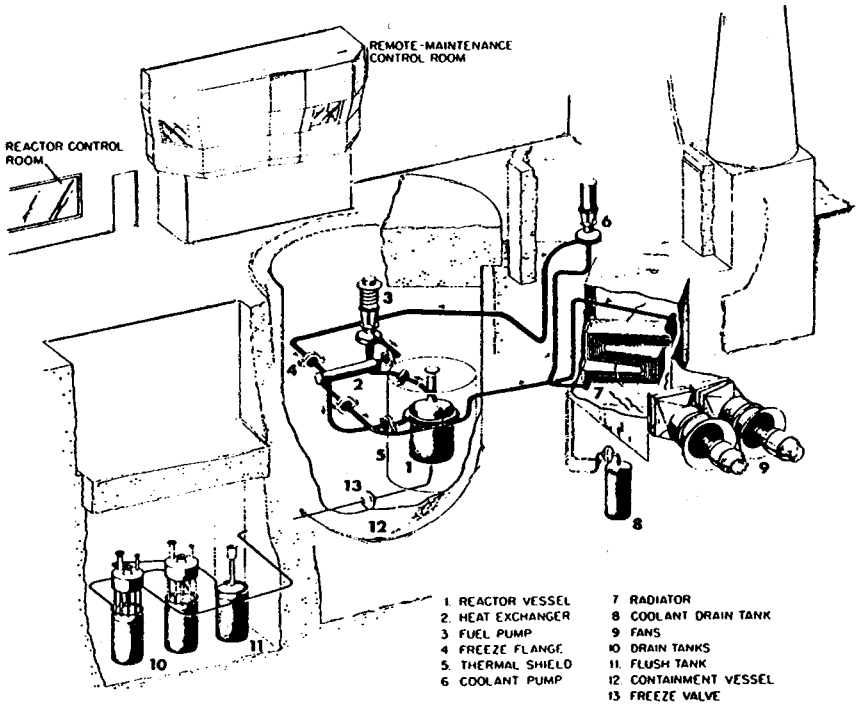


Figure 2: Physical layout, MSRE (26)

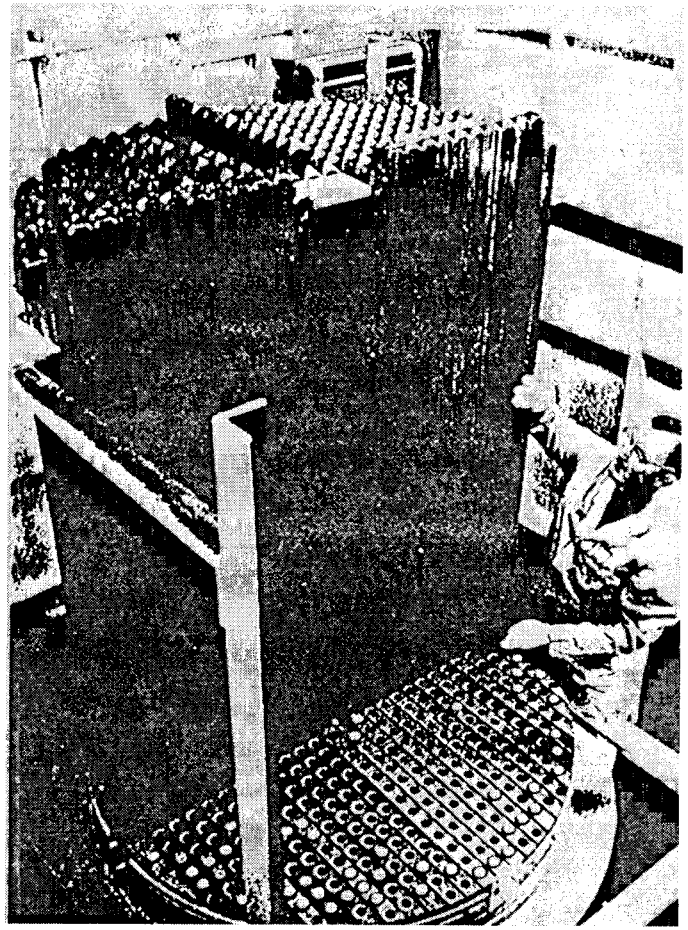


Figure 3: View of MSRE Core (26)

The metal of the primary and secondary systems is Hastelloy N. As long as the salt system has been correctly cleaned and oxygen free, no corrosion of the metal occurs. The report after 3000 hours operation was very satisfactory (26). Fuel stability is excellent. Some xenon diffuses in the graphite matrix, but an equilibrium is to be reached.

Thus, it has been demonstrated that, at least in a simple form, such a concept is feasible. Of course, the MSRE is still a rather long way from power reactor applications.

As a matter of fact, the ORNL team has worked extensively on **Molten Salt Thermal Breeder Reactor Concepts**. (27) (124).

The molten salt thermal breeder reactor uses graphite as moderator and fluorine salt solutions of fissile and fertile materials as fuel. A mixture of lithium and beryllium fluoride is used as the carrier salt. The fuel salt solution passes through holes in graphite blocks. The heat passes through a secondary coolant system containing sodium salts to the steam generator. The conceptual design considered has an overall thermodynamic efficiency of about 44 %.

The fuel-salt solution is processed in an on-site "simple" fuel reprocessing plant, based on the volatilization of the fission products fluorides, thus avoiding the expensive recycle fuel fabrication operations necessary for conventional reactor concepts. This also helps remove the Pa-233 from the core and thus reduces the parasitic absorption in Pa-233. (123)

The reactor core, with top, bottom and radial graphite reflectors, is housed in a reactor vessel made of Hastelloy N. The graphite blocks may have to be changed every two to six years because of irradiation damage. (Figures 4 and 5).

The molten-salt thermal breeder reactor is started with U-233 and thorium. The breeding ratio is very small (of the order of 1.07); however, because of a comparatively small fissile material inventory needed (about 1.5 kg of U-233 per MWe), the reported system doubling time is the same as that expected from oxide-fuelled, sodium-cooled FBRs. (16)

A fast molten-salt breeder reactor can be designed in the same way but with no graphite and with a fuel as molten-chlorides diluted in sodium chloride. The theoretical breeding ratio has been calculated to be as high as 1.70 in the optimal configuration.

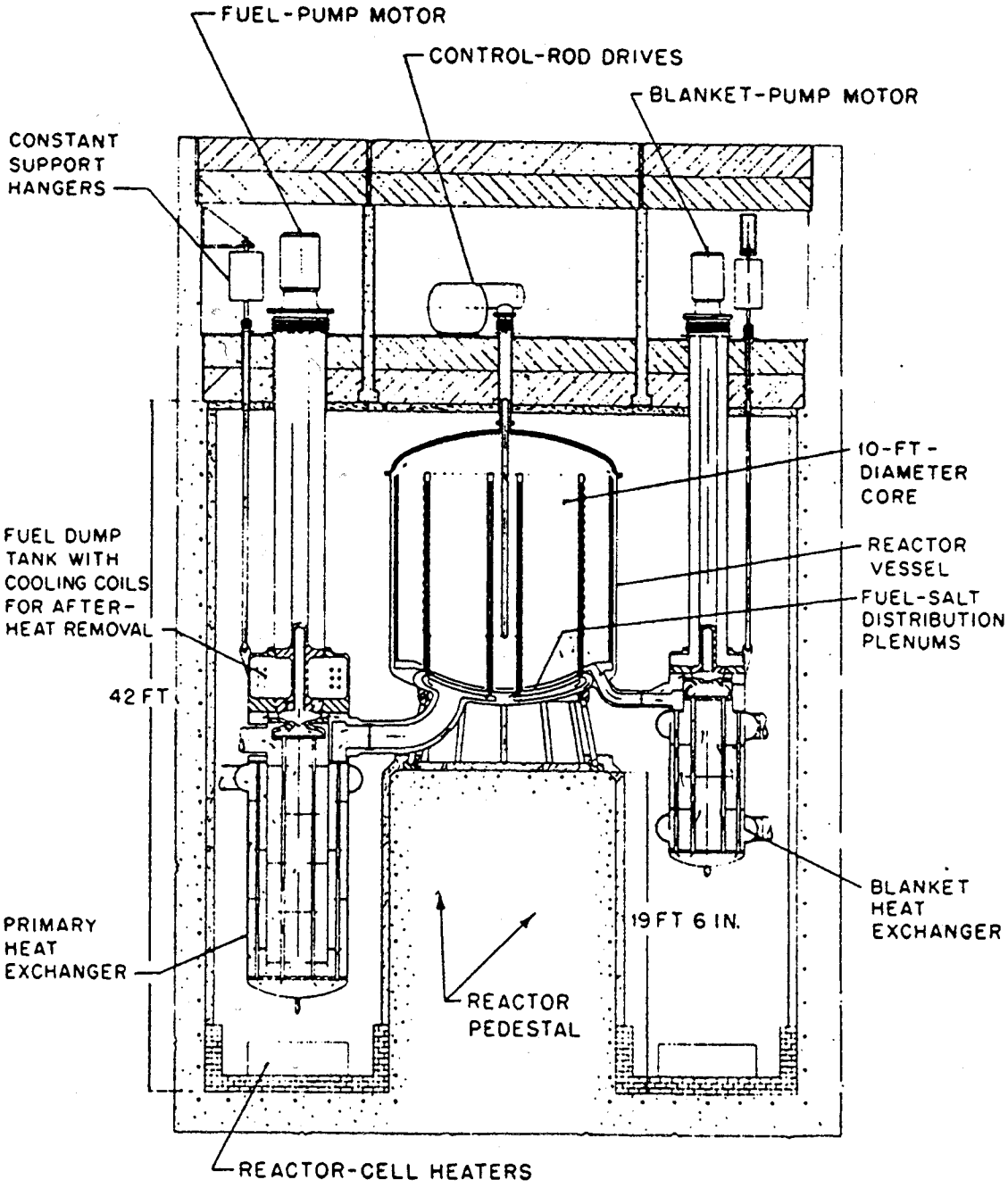


Figure 4: Elevation view of reactor cell (MSBR) (27)

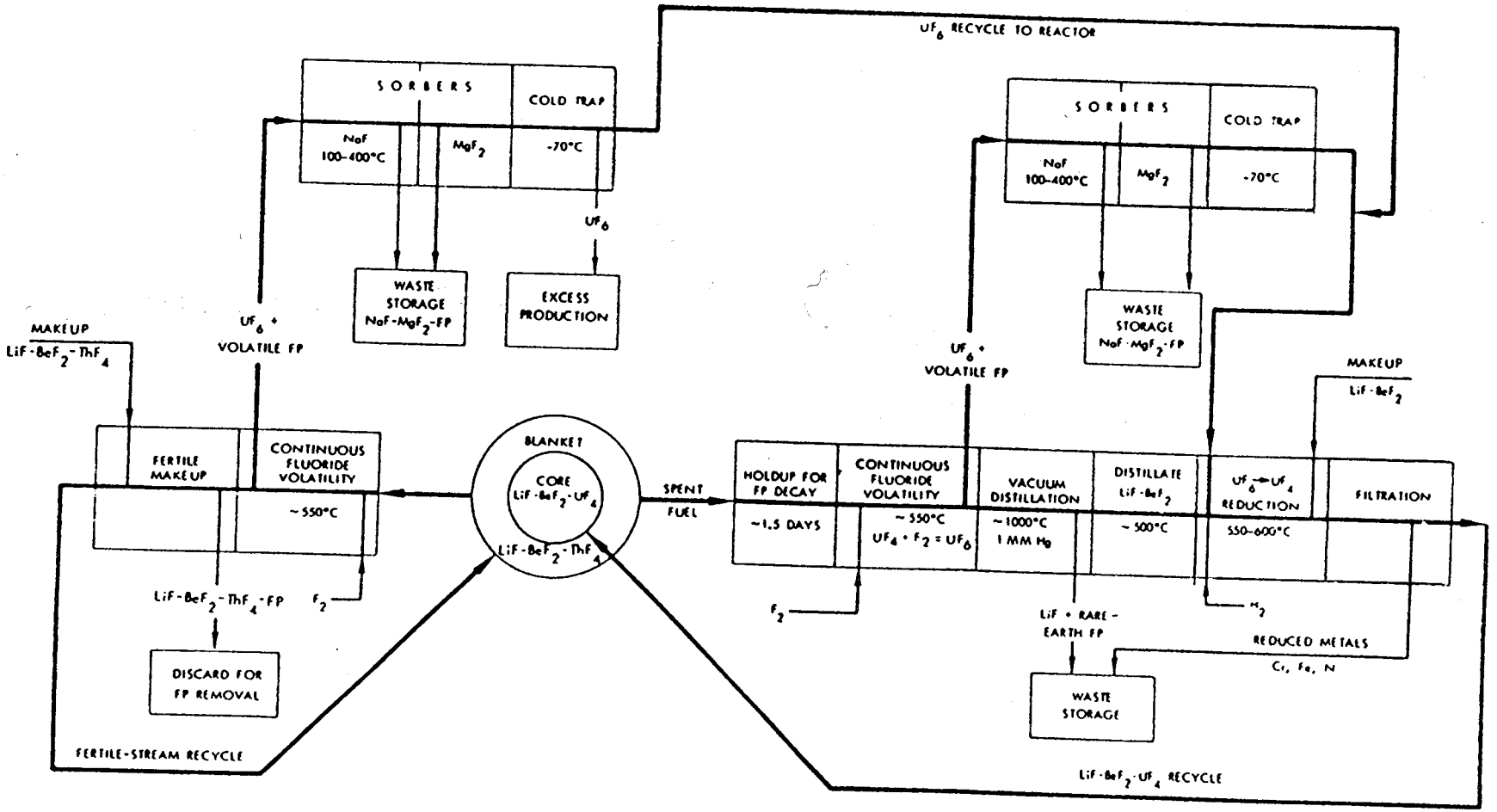


Figure 5: Fuel and fertile-stream processing (MSBR PROJECT) (27)

From Oak Ridge experience and further studies, it appears that additional work in the development of material for the primary circuit to contain the molten salts is needed, tritium containment, components, reliability, etc...

Clearly, these are advanced designs which have been pioneered but are far from industrial realization.

Theoretical work continues on Molten Salt Breeder Reactors driven by an accelerator, with a Japanese-French-Russian team. (55) (142). See also Chapter 4.

3.9. Recent Developments in Advanced Mixed-cycles Reactor Studies

(Please refer also to Chapter 3.2.4.).

Recent studies are in progress for a "Non proliferative Light Water Thorium Reactor Core" at the Tel Aviv University (Prof. A. Radkovsky), supported by the Raytheon Corporation of USA, in cooperation with the Kurtchatov Institute and the Brookhaven National Laboratory. (2)

The underlying motivations are principally :

- eliminate as far as possible "proliferative" materials by U-238 "denaturation" of the fissile components,
- eliminate as much as possible the production of long-lived minor actinides in the waste products, by recourse to the thorium fuel cycle,
- contribute to a sustainable fission energy production at reasonable cost by tapping vast thorium resources,
- use the reactor to burn the weapon's grade plutonium stockpile,
- contribute to safety in an economical way.

These ultimate aims might seem difficult to reconcile but the project is certainly worth of interest and investigation, even if in the end some industrial simplifications will oblige to mitigate the expected results (especially in the case of burning Pu-239).

The idea starts with the Shippingport reactor experiments with a seed/blanket reactor* which uses a movable seed within the blanket to adjust the reactivity without having to use control rods or burnable poisons, detrimental to the neutron economy.

We would like to quote some excerpts of Prof. Radkovsky's arguments :

"In the LWBR Shippingport Project, U-233 created in a production reactor, was added to thorium in appropriate quantities to create the seed-blanket units.* The project was successful in demonstrating a small breeding gain, as predicted. Of course like all breeders the LWBR was highly proliferative and the expense of producing U-233 fuel elements was very great.

"In our project it was felt that breeding was unnecessary, and that non-proliferation was of the utmost importance. It then follows that the U-233 must be burned in place as it is formed in the thorium. The only way to accomplish this is to utilize the seed-blanket principle in which the extraordinary ability of thorium oxide to withstand radiation of over 100 000 MWD/t is exploited. In the seed regions we use the highest enrichment of uranium (20 % U-235 and 80 % U-238) that is nonproliferative, because it is necessary that the multiplication factor of the seed be as high as possible. In a conventional uniform core the addition of 20 % enriched uranium to thorium in sufficient quantities to produce such long burn-ups would not be feasible, since so much of the uranium would be required that there would be insufficient room for the thorium. This is the essential reason why the seed-blanket arrangement is necessary, because we can replace the seeds after about one year or 18 months of operation, while leaving the blankets in the core for ten to twelve years.

"In the seed regions we utilize a very high water to fuel volume ratio, about 3.5/1, in order to create a very thermal spectrum. This has two purposes : to minimize the absorption of the U-238, thereby making the multiplication factor of the seed as high as possible and also minimizing the production of plutonium. For a given seed power the high multiplication factor of the seed increases the fraction generated by the thorium. Fortunately the seed is not "overmoderated", since that all the coefficients of reactivity are negative and of about the same magnitudes as in conventional uranium reactors. The reason for this is the high leakage of neutrons from the seed regions into the subcritical blanket regions. We use metallic fuel in the seed because of the reduced space available for fuel elements.

(*) See Chapter 3.5, Fig. 4

"The blanket consists primarily of thorium oxide rods. However, a small amount of 20 % enriched uranium is also added in the blanket and uniformly mixed with the thorium. Here again there are two reasons. First of all, a pure thorium oxide blanket would have zero multiplication factor, except for a very small fast effect, at the beginning of blanket life, so that almost no power would be produced in the blanket and the seed would have to generate nearly all of the core power. This would necessitate a large derating of the core power output at the beginning of core life, and of course every ten to twelve years when a new blanket is installed. By adding a small quantity of 20 % enriched uranium to the blanket, the blanket fraction of core power is raised to a sufficient amount, about equal to that which is obtained when the blanket U-233 content is built up. (This results in a blanket multiplication factor of about k = 0.90).

"The second reason is to denature the residual U-233 content of the blanket so that it could not conceivably be used for weapons. The U-233 will be uniformly mixed with nonfissionable isotopes, U-232, U-234, U-236 and U-238. U-236 is probably even more deleterious for weapons usage than the U-238. In any case isotopic separation of U-233 would be impossible for all practical purposes, considering the high gamma activity of U-232. Besides any nation with isotope separation facilities would find it much easier to separate U-235 from natural uranium.

"We are now making a major effort to design a nonparasitic control system. One of the schemes being considered is similar to that used in the LWBR, except that zirconium takes the place of the natural thorium used in the LWBR. One advantage we have over the LWBR is that we have room for shutdown rods, while in the LWBR the tightness of both the seed and blanket lattices left no room for shutdown rod. We envisage a shutdown rod as being made up of hafnium plates surrounding the seed. A movement of about half the core height will probably be adequate. The nonparasitic control by moving seed fuel will not have to include shutdown functions as in the LWBR, which made it necessary for the moving fuel to go down below the core boundary by half the core height. In our case, a smaller movement below the core should be adequate, which is also important from safety and economic standpoints.

"Our control elements are about half as long as in a conventional core, have good clearances, and can scram the core twice as fast as in a conventional core. The pressure vessel height can be reduced, leading to a reduction in primary plant components, and thus a saving in plant costs.

"It is anticipated that the use of non parasitic control will reduce the uranium fuel loading in our design, since less power will be developed in the seed and more in the blanket. We expect that the annual production of plutonium will be about 5 % of that from a standard uranium core. The density of plutonium in the discharged seed fuel will be so low that it will be difficult and expensive to separate. (*Note :* this may be exaggerated in view of past experience). The blanket plutonium is not included because it will contain so much Pu-238 that it will not be usable for weapons.

"A fundamental difficulty with seed-blanket cores is that the seed fuel must be replaced when the multiplication factor of the seed becomes too low to drive the blanket adequately. This usually occurs when only half the seed fuel has been depleted. Originally it was thought that the seed fuel could be reprocessed and reused, but from a nonproliferation standpoint any reprocessing of fuel is objectionable. (*Note :* this may be a wonderful wish, but difficult to fulfil !).

"We are investigating various schemes to solve this problem.

"Besides the savings in plant costs from the reduction in height of the pressure vessel, there is a considerable saving in plant costs due to the elimination of the soluble boron control system (except for a simple system for emergency shutdown and refueling when the core is depressurized). Much piping and equipment can be eliminated. Safety is enhanced since small boron leaks corrode high strength steel parts and can prevent the operation of vital valves due to accumulation of boron crystals. Load following is much easier without boron control, particularly at the beginning of a cycle when there is so much boron in the coolant that the temperature coefficient of reactivity is very small in magnitude.

"In a conventional core, after each cycle, all the fuel assemblies must be relocated and a fuel management group is necessary. In our core, replacement seed and blanket assemblies always go in the same locations, thus saving refueling time and personnel.

"Core manufacturing costs will be reduced. The manufacture of thorium oxide rods is very similar to that of uranium oxide rods, but since our blanket is replaced only once in ten to twelve years, while the uranium oxide rods in a conventional core are replaced every three years, our manufacturing costs will be much less. We have not considered 18 months refueling of conventional cores, because any savings there are offset by the much higher costs of burnable poison

elements. It is also probable that the cost of thorium will eventually be much less than that of uranium. Our enrichment costs are anticipated to be lower than that of conventional cores because most of our energy is derived from thorium."

As we can see, this design is still in a theoretical development stage, but it shows the way to some practical ideas for the future. We are not sure that the obsession to avoid reprocessing by all means for proliferation reasons is justified, for an elaborate reactor which will need a strongly established nuclear energy industry in what should be a stable country anyway. It is hoped that practical R&D experiments could be performed in the future in the same way as the US DOE have done for the Shippingport reactor, or the Belgian-EC partners have done to take advantage of the BR3 PWR prototype to test Pu recycle schemes.

Comment : it is refreshing to observe such new ideas which, some day, might bear practical fruit. As the author says, the current line of thought preventing proliferation by all means, including reprocessing, might be overdone. The presence of plutonium along with U-233 in the blanket might complicate the process. But the idea of separate U-235 or Pu-239 replaceable seeds with a long-lived Th-232/U-233 blanket seems to us an interesting idea worth of further investigation.

3.10. The Long-term : Accelerator-driven reactors
(with thorium to minimize the long-lived actinides waste).

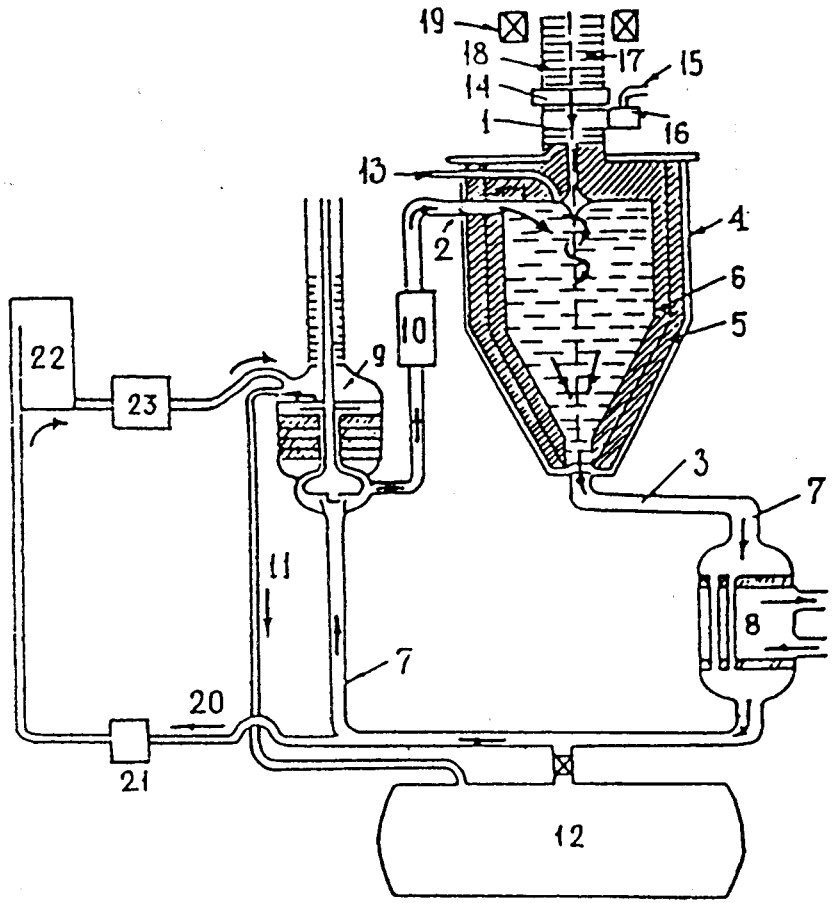
The subject will be treated in more detail in the next chapter.

It may be interesting to quote remarks of the UK AEA on the subject in 1979, (176) :

"Taking a long term view, theoretical possibilities exist of manufacturing fissile from fertile materials by irradiation, not in nuclear reactors as we know them, but by the use of neutrons derived from either particle accelerators or fusion devices. These materials could well be U-233 and thorium and several studies make this assumption. For the accelerator/breeder, to have any significant impact on the world's energy supply, it appears that accelerators would be required which are very much larger than any that have so far been built. For example, with a Linear Accelerator Breeder as discussed by Steinberg of Brookhaven, it has been suggested that a 1 GeV proton accelerator delivering 300 MW of beam power would be required to produce between 1 and 2 tonnes of fissile material per

annum. There would also be a need to dissipate heat generated in the target with consequent design and cooling problems which are quite novel to accelerator targets. If this line were followed, the engineering outcome could well be more like a reactor with an accelerator as a neutron booster, than an accelerator/target assembly."

Figure 1 (55) shows the Japanese concept of such an accelerator-boosted molten salt reactor. In a self-sustaining nuclear program, a few accelerator-boosted, dedicated reactors would breed enough U-233 to feed a park of "burners" for energy generation. (59)



Set-up of single-fluid type
Acc. Molten Salt Breeder

1- Proton beam,
2- Salt inlet,
3- Salt outlet,
4- Reactor vessel,
5-6- Graphite,
7- Primary loop,
8- Heat exchanger,
9- Main Salt pump,
10- Throttle valve,
11- Overflow line,
12- Storage tank,
13- Pressured salt inlet,
14- Gate valve,
15- Vacuum line,
16- Vapor trap,
17- Duct,
18- Orifice,
19- Focussing magnet,
20- Bypath for salt purification,
21- Pump for bypath line,
22- Process facility,
23- Filter.

Figure 1: Scheme of single-fluid accelerated molten salt breeder (55)

4. THE ACCELERATOR-DRIVEN SUBCRITICAL SYSTEMS BASED ON THE THORIUM FUEL CYCLE

4.1. General Remarks

A traditional fission reactor is essentially a critical facility in the neutronic sense. This presents advantages, but not without problems associated with reactivity, which has to be checked with great care and precision.

As a matter of fact a fuel containing a substantial part of actinides, produces in a critical assembly only a limited fraction of delayed neutrons. The shorter neutron life-time and a modest Doppler coefficient render the control of the reactor more difficult. It is consequently interesting to modulate the reactivity through an injection of particles, generating neutrons, by an external accelerator to a subcritical assembly.

Carlo Rubbia recently (57) has shed a new light with much lustre on the hybrid systems of a subcritical fission reactor driven by an accelerator. (46) (47)

In a traditional reactor, the neutron absorption at thermal or low energy by a fissile nucleus of, usually, uranium-233, 235 or plutonium-239, results in a fission with emission of ν neutrons (2.2 to 2.9 neutrons according to the isotope). The chain reaction will be sustained when the balance k of neutrons causing the next generation of fissions is at least equal to 1. There is divergence when $k > 1$. Practically, the operation of a reactor is obtained with the help of a small fraction (*0.3* - 0.7 %) of "delayed" neutrons which permit to correct the differences between k being very slightly below 1 (called "multiplying" media) (e.g. 0.993) and k being very slightly above 1 (called "diverging" media) (e.g. 1.00 +). If for some reason the neutron multiplication is not checked as, for example, by temperature effect, negative void coefficient, control rods insertion, etc..., the reaction can become explosive, as happened at Chernobyl.

The hybrid accelerator/reactor system relies on a reactor with a multiplying system where k is clearly below 1 (e.g. 0.95) and where the chain reaction is brought to a steady state by means of a strong addition of "foreign" neutrons created by an accelerator.

These neutrons are spallation neutrons generated by impingement of particles of high energy (e.g. 200 MeV) usually protons or deuterons on heavy nuclides (e.g. Pb), which create a primary hadronic cascade, in which are many neutrons. When sufficiently high energy protons impinge on a heavy metal target, they can create a flux of 40-70 neutrons per proton.

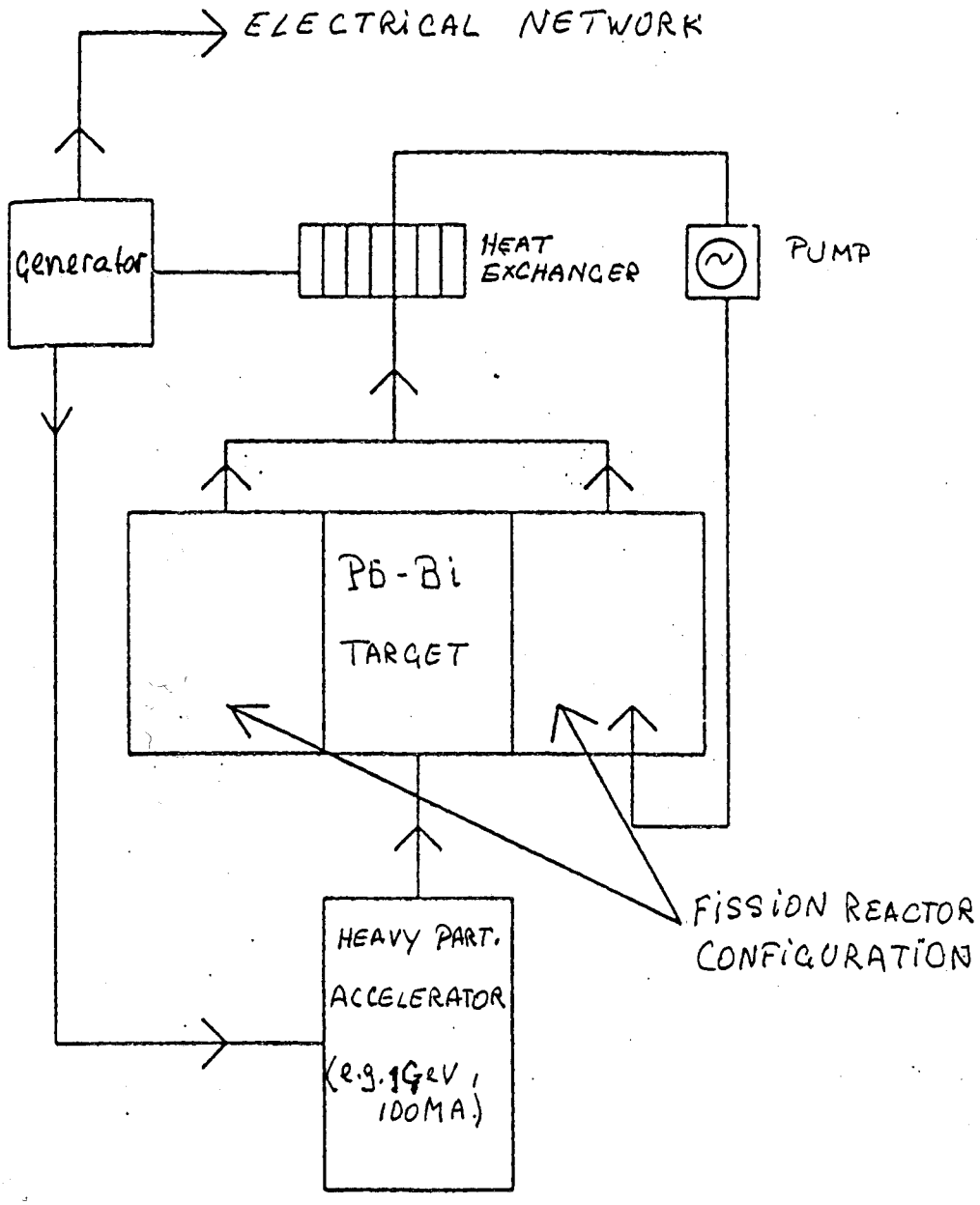


Figure 1: Scheme of an accelerator-driven system

It has been calculated that a 1.6 GeV linear accelerator with beam intensities of 100-250 mA (which is at the top of what can be built today), with a 50 % electric yield and a thermodynamic yield of the reactor electricity production of 42 %, would enhance artificially the neutron yield ν per fission from an average 2.5 to an average 3.9. However, lately, proponents of the hybrid systems are proposing more modestly to use a 1 GeV, 10-15 mA cyclotrons which are easier and cheaper to construct.

Thus the reactor operation would depend on the outside neutron flux and the reactor could be largely subcritical ($k \geq 0.95$), which would enhance its inherent safety.

Fig. 1 gives the scheme of such a hybrid accelerator-reactor system.

This idea has been around since the 1960s (Bennett Lewis, Chalk River (62), taken over by Steinberg at Brookhaven and others in the 70s (61), but at that time the accelerator power was too small (Los Alamos LAMPF 800 MeV, 20 mA peak) (47). As early as 1981, K. Furukawa and al. had the idea to combine a molten salt breeder thorium reactor with an accelerator (60).

The idea to use these hybrid systems is twofold :

- produce energy as safely and cleanly as possible,

- transmute isotopes, either to produce valuable isotopes, to produce new fissile nuclei, or to burn unwanted waste (long-lived fission products and minor actinides).

Depending upon the end-use, these hybrid systems can be quite different. But they have in common an outside accelerator which provides an extra source of neutrons by spallation.

The principle of an accelerator-driven reactor will apply indistinctly to any type of reactor. In particular, it can be used with a uranium-fed reactor, but also with a **thorium-based reactor**. In fact, use of thorium may present some advantages, and in particular :

- The inconvenience of thorium as a "neutron sink" compared with, for example, natural uranium, can be alleviated by using an adjustable extra source of neutrons. Hence this hybrid system might be an elegant means to tap the huge thorium potential energy resources more easily than with a traditional reactor;

- As has been seen in other chapters, the production of transuranium elements, or minor actinides, in reactors based on the couple U-233/Th, will produce substantially less long-lived actinides than the couples U-234/U-238 or Pu-239/U-238.

This shows in the curves, Figs 2 and 3, (42) where it can be seen that thorium spent fuels and thorium "waste" after reprocessing are less toxic by a factor of about 30 than their counterparts from uranium (case of a thermal amplifier). The radiotoxicity is defined as the yearly limit of 20 mSv compared to the nuclide activity :

$$\text{Radiotoxicity} = 0.02\ \text{Sv} \times \frac{\text{A : Activity of the nuclides Bq}}{\text{ALI : Annual Limit of Intake}}$$

It can be seen, however, that after 10^5 years, due principally to transformation of U-233 ($1.6\ 10^5$y) into Th-229 ($7.3\ 10^3$ y), the radiotoxicity of the thorium waste would increase compared to uranium waste. This comes from the ALI values and is probably not very significant from a health standpoint, considering the conservative waste management precautions taken (cf. the EC PAGIS study).

At this point, we wish to quote remarks by Dr. M. Salvatores of CEA on this issue of radiotoxicity (May 1996) :

"When dealing with the issue of radiotoxicity, one has to specify what "type" of radiotoxicity. If one is speaking of "potential toxicity" (i.e. related essentially to the activity of a isotope "father" of its progeny), the case of an irradiated fuel based on Th, has been compared to a U-based fuel, after normalization to the same energy produced. The comparison indicates that, if the same hypotheses are made for both types of fuels (e.g. both directly stored in an open cycle strategy; or both reprocessed in closed cycle strategy and using the same hypotheses on the losses during reprocessing) :

- At "short" storage times (i.e. $t \leq 10^4$ y), the potential toxicity of the irradiated Th-fuel is lower by a factor ~ 50 with respect to the corresponding U-fuel.

- At "long" storage times (i.e. $10^6 \geq t \geq 10^4$ y), the Th-fuel or the U-fuel exhibit approximately the same toxicity. This is due to the effect of U-233, Pu-231 etc... in the overall toxicity.

As far as toxicity from mill-tailings, the thorium case is better than the uranium case (see for example "Impact Radiologique à Long Terme de l'Extraction du Thorium", S. Menard et J.P. Shapira. (40)

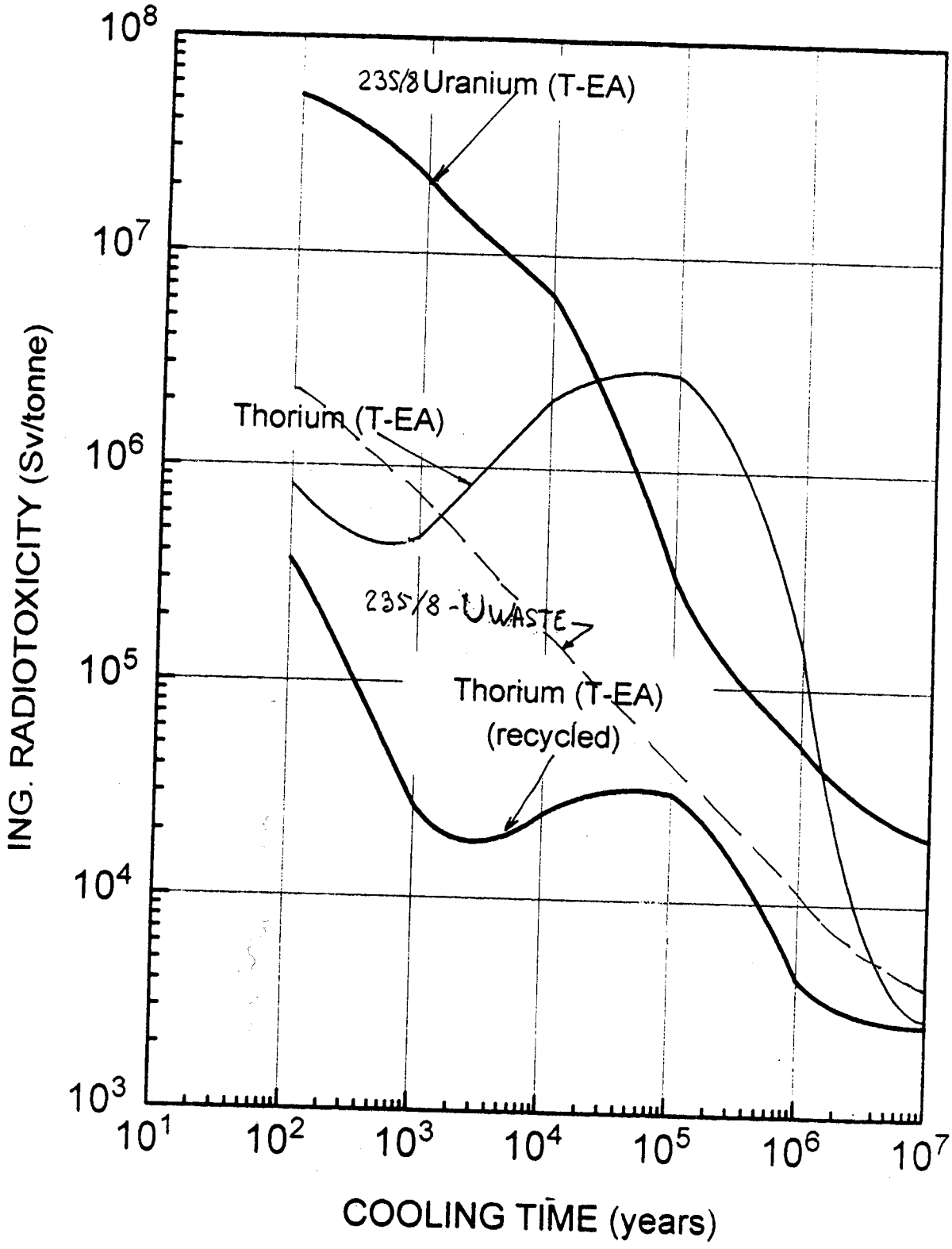


Figure 2: Comparison of the ingestion radiotoxicities of spent uranium and thorium fuels and thorium waste from a T-EA (burnup 40GWd/t). The thorium waste results from the removal of 99% uranium and protactinium from the spent fuel. (42)

NOTE 1 : T-EA : Thermalized neutrons reactor-energy amplifier.
NOTE 2 : The radioactivity curve for reprocessed uranium 235/238 Spent fuel waste has been given as a comparison.

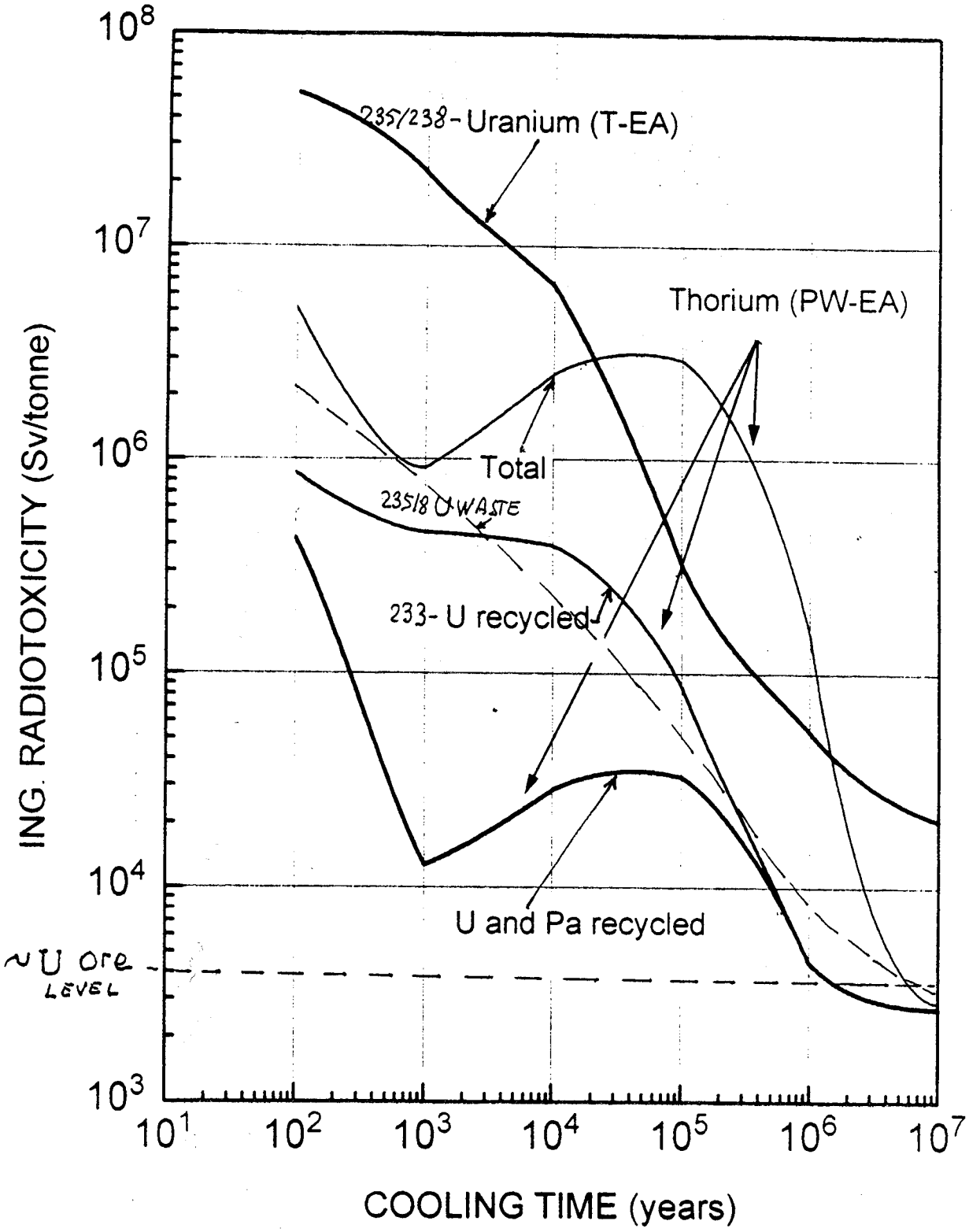


Figure 3: Comparison of the ingestion radiotoxicities of spent thorium fuel and waste from a PW-EA (burnup 40GWd/t). The thorium waste results from the removal of 99% uranium and protactinium from the spent fuel. The radiotoxicity of spent T-EA uranium fuel is shown for reference. (42)

NOTE 1 : T-EA : Thermalized neutrons reactor with energy amplifier, PW : Press. Reactor.
NOTE 2 : The approximative radiotoxicity curve for U-238-235 reactor fuel reprocessing waste has been given as a comparison.

A sometime misleading picture is given when open-cycle uranium-fuelled PWRs are compared with Th-based systems (as in the case of the CERN Energy Amplifier), for which reprocessing is implicitly considered with extra-high decontamination factors (losses of 0.01 %, compared to present day values of around 0.1 %.

As a conclusion, the use of thorium in a hybrid system, as in any reactor, presents advantages from the standpoint of radiotoxicity.

The drawback of thorium, that upon average it needs one more neutron compared to uranium to sustain reaction, would become less when cheap extra neutrons could be artificially produced with an accelerator source.

Whereas a thermal breeder based on the couple U-233/Th as had been tested at Oak Ridge with the Molten Salt Reactor Experiment, would have a doubling time of about 20 years (against about 5 years for a fast Pu/U breeder), a breeder assisted by extra neutrons from a very powerful accelerator could theoretically have a very short doubling time, a few months. (58)

These features of enhanced thermal breeding, combined with the interesting characteristics of the thorium fuel cycles in terms of optimal fuel and lower toxicity, with molten salt reactors, in terms of safety and flexibility, have been combined by K. Furukawa, A. Lecocq and al. (59) in the THORIMS concept (Thorium Molten-Salt Nuclear Energy Synergetics). In this concept, some accelerator-enhanced thorium molten salt "breeders" would supply U-233 fuel for a park of energy-producing "burners", also molten salt reactors. This configuration, besides its inherent safety and stability makes possible the "milking out" of neutron poisons with an in-line system.

However, problems of material resistance under corrosion and irradiation would need to be assessed at prototype scale, following the Molten Salt Reactor Experiment at Oak Ridge (1965-1969) and work on MSBR (1963-1976). The authors would see such developments in the middle of the next century.

It has been shown that to reach a reasonable breeding ratio in a thermal reactor with a Th-based fuel, one needs to be fairly subcritical. In that case, the contribution needed from an external source is fairly substantial, and in the case of spallation neutrons obtained with a high energy proton beam, the accelerator current should be relatively high (i.e. 50 mA) (M. Salvatores).(39)

4.2. The Energy Amplifier

Note : whereas the remarks below could apply indistinctly to any reactor type, we have tried to put the thorium-fuelled reactor into perspective.

The Energy Amplifier is the project proposed by Carlo Rubbia, for which he claims the advantages of :

- great reactor safety,
- enhanced safety due to presence of less long-lived actinides and proliferation resistance,
- these actinides can be recycled as seeds for another fuel load. (48) (57).

The accelerator would be a double cyclotron of 1 GeV total energy, with a 10 mA mean intensity. Such an accelerator is a rather modest extrapolation of the Paul Scherrer Institute (PSI) Zürich cyclotrons. The multiplying medium can be either a PWR, or a high temperature, helium cooled reactor, or even a liquid-metal cooled reactor.

Neutron multiplication can be realised by a high energy, high intensity proton beam striking a heavy metal target located in the central region of the core.

The neutron yield from high energy protons on high density material is quite large. The beam energy fraction required to produce one neutron with the help of a high energy cascade is rather proton energy independent and of the order of 35 MeV for Pb or Bi, and of 22 MeV for Th. As far as the core is concerned, the number of neutrons produced by N injected neutrons in a lattice having an effective criticality factor keff is equal to : $N_{tot} = N / 1 - k$.

Of course keff should be not too far below 1 to get a good multiplication factor.

The actual energetic gain of the system depends then from different factors, namely the actual energy spectrum of the neutrons and the energy dependence of the cross sections, particularly complicated in the resonance region.

For a value of keff in the range of 0.9 - 0.95, the number of neutrons in the core region is 10 to 20 times the number injected by the target; let us assume that 40 % of all neutrons end up in fissions, considering that the fission energy yield is about 200 MeV, the energetic gain could be, for example, of the order of 40.

(Whereas in the case of a chain reaction, g would be unlimited, but with other constraints).

With a U-233/Th fuel cycle, it is necessary to maintain the neutron flux around or below 10^{14} n/cm^2/sec, to enhance production of U-233 from Th-232 :

Th-232 + n $\Rightarrow$ Th-233 (12 m) (β) $\Rightarrow$ Pa-233 (27 d) (β) $\Rightarrow$ U-233

If the flux is higher than 10^{14} n/cm^2/sec, there is a risk of neutron capture by the protactinium whose capture cross-section (55 barn) is far from negligible, conducing to production of by-product U-232, the decay chain of which, as we have seen, is very counter-productive.

With the neutron flux of 10^{14} n/cm^2/sec, one reaches an equilibrium state where the U-233 concentration is around 1.3 %.

Concerning the energy amplification system, as well as the control of reactivity, it has to be noted that in an operating core the fission fragments play also an important role. They may capture some of the neutrons, thus affecting the neutron inventory; on the other hand they decay in different times. There is an interplay between neutron capture and decays which leads to a complicated composition of the operating core, depending from the actual operating conditions (mainly the neutron spectrum) as well as from the past history.

Fission fragments are generally radioactive and produce additional heat, even if the proton beam is switched off. Immediately after turn-off of the proton beam the power produced by this residual activity is about 7 % of the steady condition. Activity decreases slowly with time, approximately as t-exp -0.20, where t is in seconds, leading to a reduction of a factor 10 in about one day.

An efficient core cooling must then be maintained after the shut-down of the energy amplifier, like in a nuclear power reactor, in order to avoid core melt-down.

If the medium works on a U-233/Th cycle, one has to take care of the progressive transformation of Pa-233 (t 1/2 - 27 days) which, by giving more U-233, will increase the reactivity in the reactor. If one uses "standard" flux levels, the effect can be as high as 2 % in Keff, which can be a relatively severe impediment when deterministic safety is looked for.

The type of lattices which can be envisaged for the multiplication zone of the energy amplifier is as large as the number of different reactor systems, using thermal or epithermal or fast neutron spectra, with different moderation, different types of fuels and perhaps of blankets (U, Th, Pu), different cooling systems (H20, D20, Gas, Na, Pb, molten salts). (cf. Fig. 6, Fig. 7).

The merits of each one of these systems and, before that, its practical feasibility, must be analysed deeply, before emitting a judgement. It can be stated in general that the introduction of a driver accelerator increases the complexity of the system, hence its cost, but increases probably the degrees of freedom for choosing fuel cycles (i.e. Th) which may open larger perspectives for a suitable and accepted nuclear energy production.

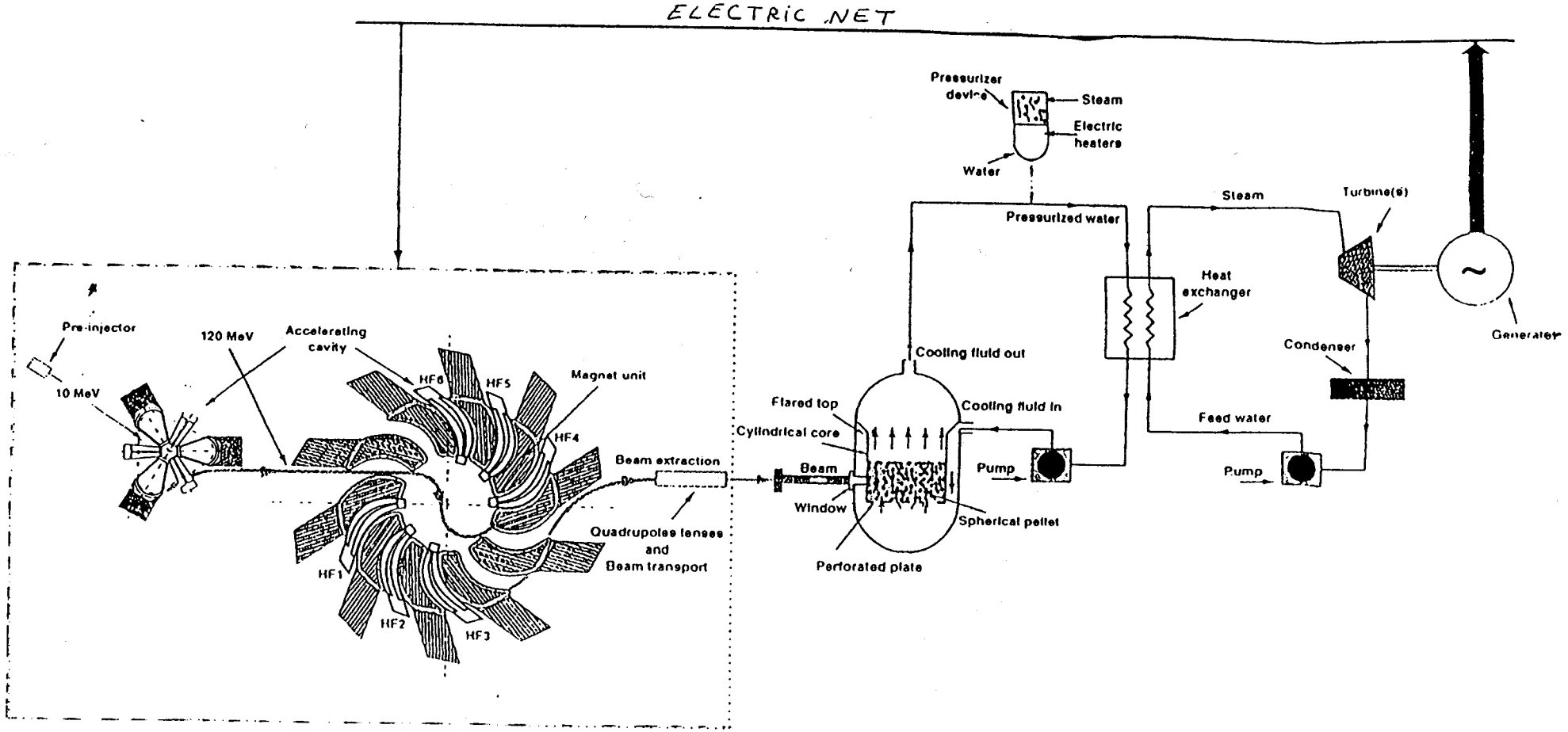


Figure 4: Scheme of the Energy Amplifier (57)

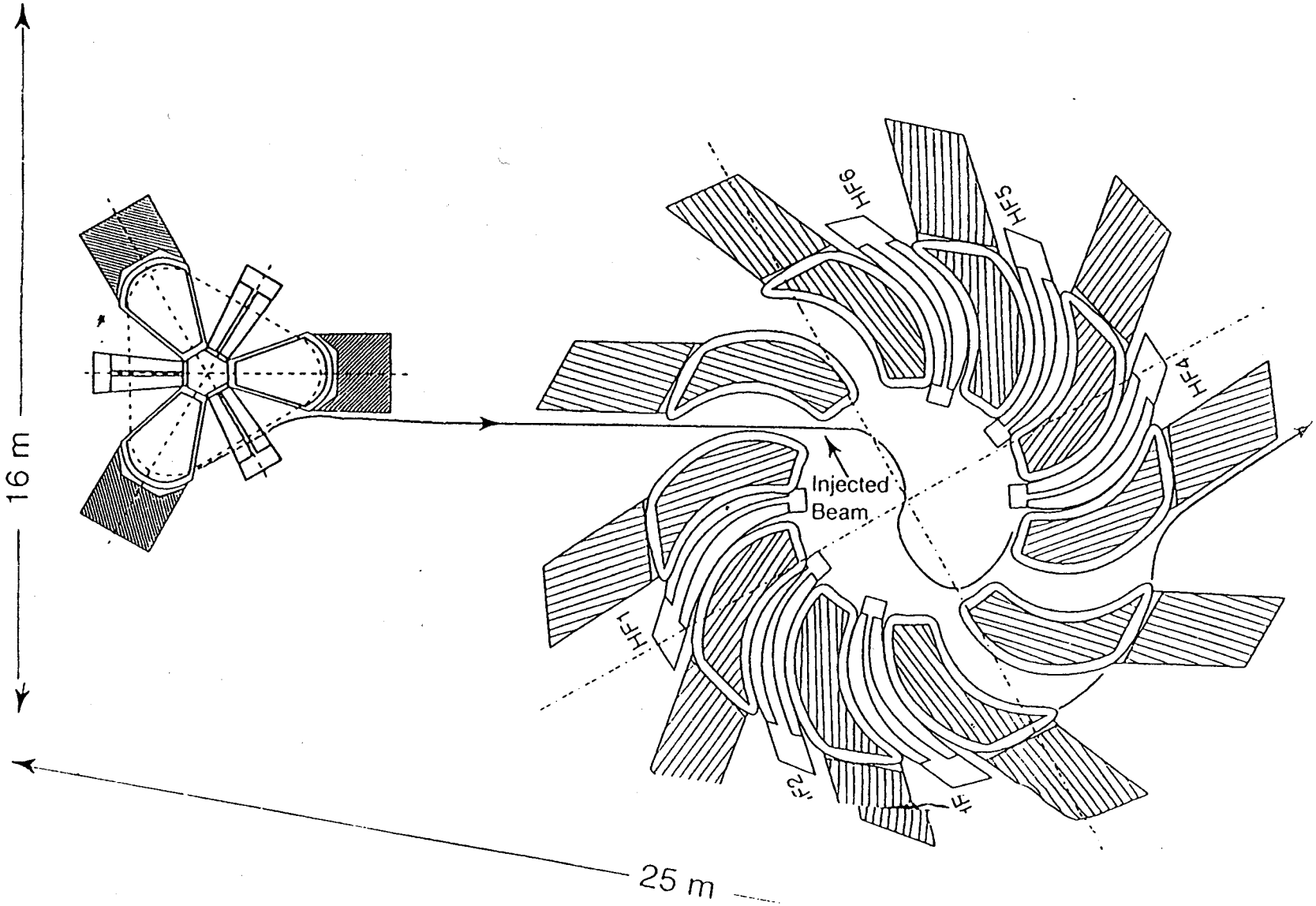


Figure 5: Accelerator size (57)

It is important to note that the generation of scientists specifically skilled in the field of nuclear reactor physics, which was particularly active in the years 50s and 60s, is now dwindling, either by age or by diversification, and the risk of "rediscovering the wheel" is rather high. It is a fact nevertheless that a rethinking of the past tentatives may be useful for looking at different approaches.

One of the main technological problems to tackle in this project is the proton "window" or windows to the reactor and the durability in time. Another may be the dispersion of the spallation neutron beam(s) to be directed homogeneously within a large reactor.

Figures 4, 5, 6, 7, give some preliminary idea of how an energy amplifier would look like.

It is necessary to test these concepts on prototypes...

Recently, C. Rubbia and co-workers from CERN and French IN2 P3 laboratories have physically tested their idea, using a water moderated lattice of 36 tonnes of 270 uranium rods lent by the Madrid Polytechnic University. The keff of this lattice has been measured to be 0.9. Artificial addition of 5.10^{10} n/s has shown that the gain was of about 30. Calculations show that with a keff = 0.95 and spallation neutrons from protons of a GeV cyclotron and 10 mA, the gain would have been of 60 and it could be possible to produce about 200 MW of electricity. It was also shown that this gain will not increase practically for beam energies higher than 1 GeV. (41)

Studies show that by replacing water coolant and moderator by molten lead, the neutrons will remain fast neutrons, which, inter alia, would decrease poisoning by the fission products and increase transmutation of actinides, and still increase the energy gain (up to 100).

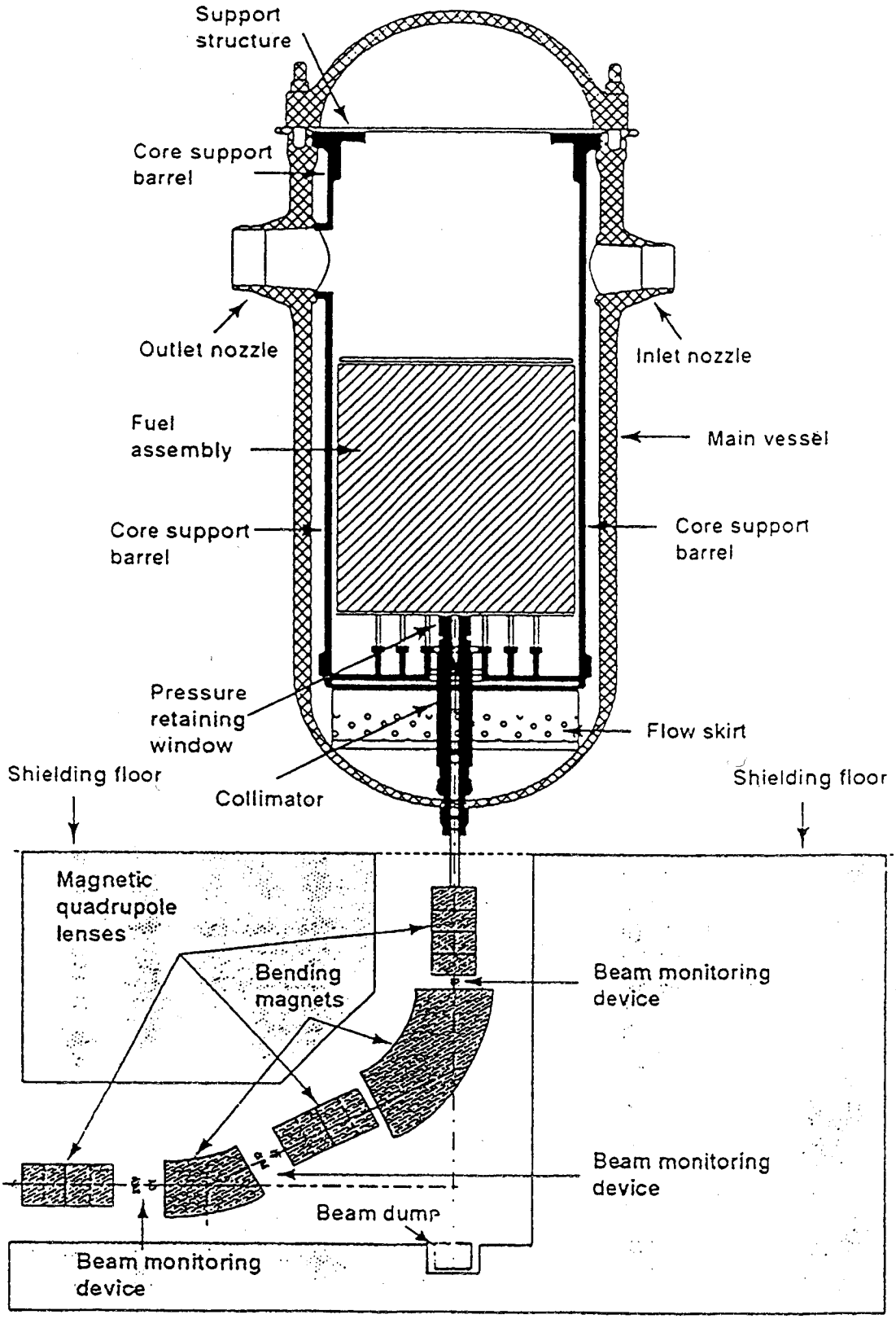


Figure 6: Accelerator-driven PWR (57)

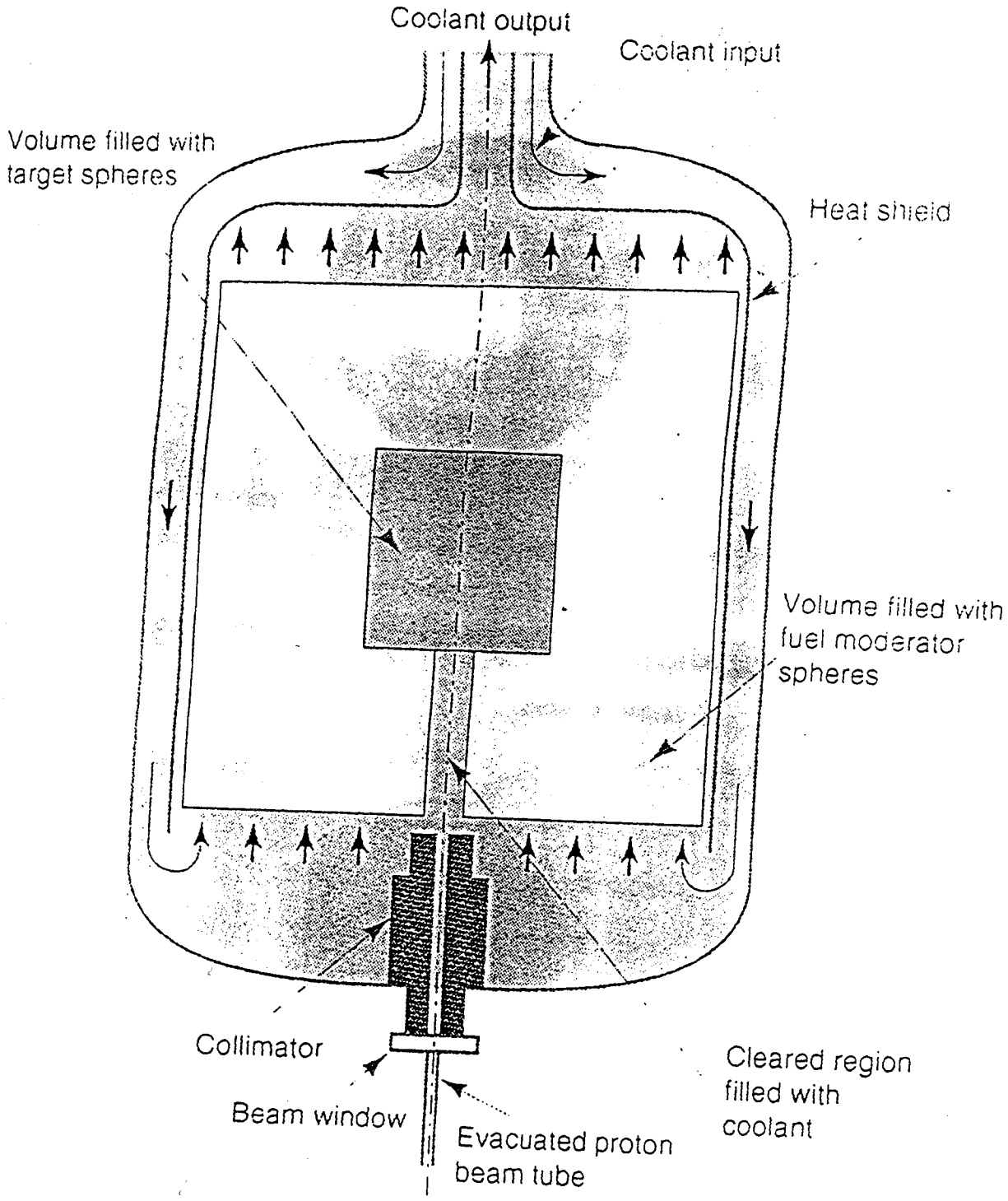


Figure 7: Pebble-bed configuration (47)

4.3. Isotopic Incineration by Actinides Transmutation

There, efficient burning of the actinides requires the highest fluxes possible, and the geometry of the "burner" is certainly of importance.

Studies on these systems are at present conducted in different laboratories. One of the earliest and "boldest" projects is without doubt the C.D. Bowman project.

The C.D. Bowman Project - Accelerator Based Conversion ("ABC") and High Level Waste Transmutation (ATW) - (58) (50)

This project is also based primarily on the thorium fuel cycle, but, besides producing energy, it is clearly oriented towards transmutation of long-lived actinides and some fission products.

High thermal neutron fluxes are called for (10^{16} n/cm²/sec). Neutron multiplication is obtained by U-233 fission or by fission of the actinides to be incinerated, Pu, Np, Am or Cm. The reactor is a molten salt reactor like has operated at Oak Ridge (cf. Chapter 3.8.), using fused fluoride salts with a graphite reflector. U-233 is bred from thorium in a low-flux zone (10^{14} n/sec/cm²) and Pa-233 formed is extracted continuously to avoid neutron capture and let to decay into U-233. The actinides are "burnt" in the high neutron flux zone, with epithermal neutron fluxes to take advantage of absorption resonances. (cf. Figs 8 and 9).

This system would - at least theoretically - present many advantages :

1) This type of reactor "ABC" is particularly suitable to burn the weapon's excess plutonium, according to Bowman (50), and all minor actinides.

For example, for Np-237 (Fig. 10) :

- In low neutron flux :

$$\text{Np-237} + n \Rightarrow \text{Np-238} \Rightarrow \text{Pu-238} + n \Rightarrow \text{Pu-239} + n \Rightarrow \text{fission}$$

- In high neutron flux :

$$\text{Np-237} + n \Rightarrow \text{Np-238} + n \Rightarrow \text{fission}$$

The first reaction will consume 3 neutrons and release 2.9 neutrons from Pu fission. The second will consume *two* neutrons and release 2.7 neutrons from Np fission, thus it will be a net energy producer. (Fig. 11)

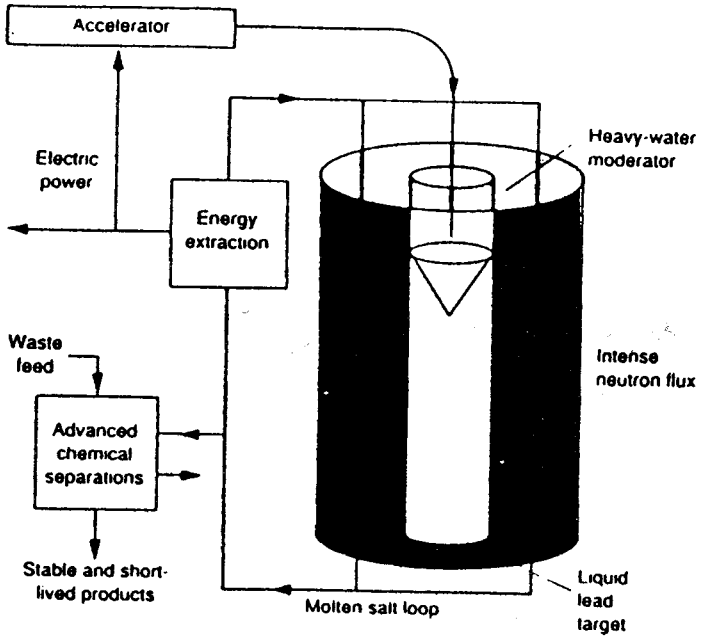


Fig. 8 The proton beam strikes the lead target generating neutrons which are moderated in the surrounding heavy water blanket. Molten salt carrying fissile material for heat generation and electric power production circulates in the heavy water blanket through double-walled pipes. Some of this power drives the accelerator. Nuclear waste including that produced in the molten salt is also circulated through the blanket in a separate loop and transmuted to stable and short-lived nuclides which are extracted and stored.

C.D. BOWMAN (58)

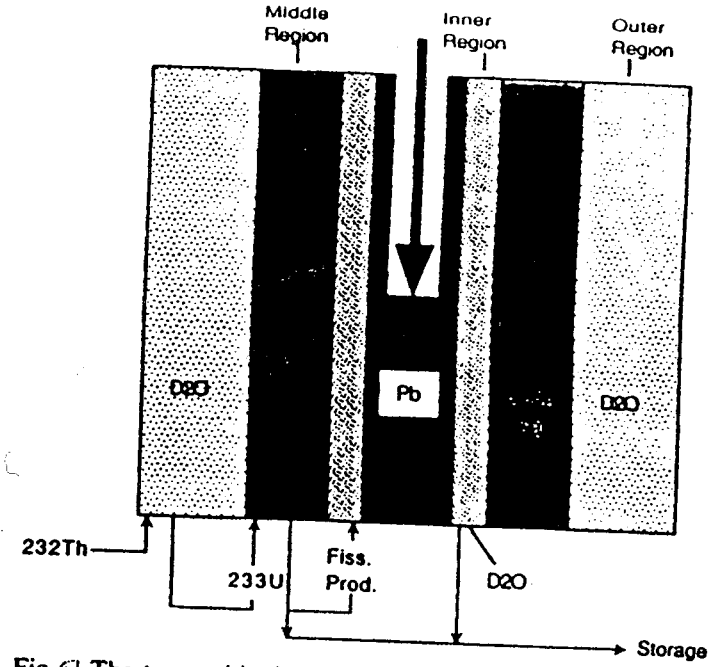


Fig. 9 The target–blanket system. The beam strikes the liquid Pb target from above producing an intense neutron flux in the surrounding moderator. The blanket is divided into three sectors. For energy production from ^{232}Th, the ^{232}Th is fed into the outer sector where it is converted to ^{233}Pa which is immediately extracted. After it has decayed to ^{233}U, the ^{233}U is fed into the middle sector where fission energy is generated. The fission products are removed and returned to the inner sector for transmutation.

C.D. BOWMAN (58)

Figures 8 and 9: Two aspects of the C.D. Bowman "ABC" and "ATW" systems

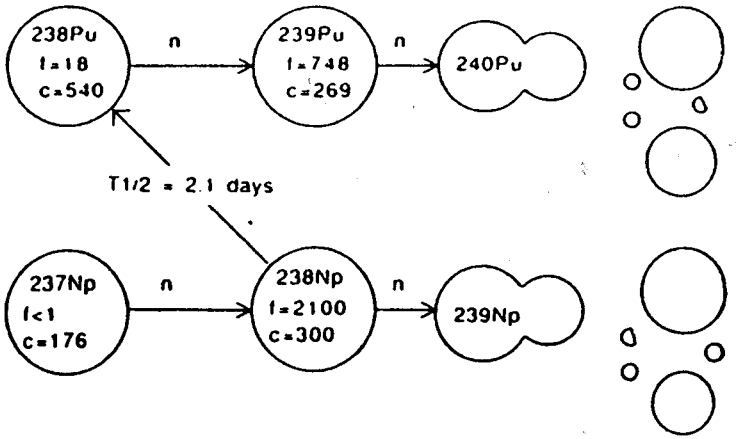


Fig.10 The two-step capture process for the higher actinide waste. In a high neutron flux, two neutrons are captured in succession on the target nucleus ^{237}Np causing fission as ^{239}Np with the production of about 2.7 fission neutrons so that ^{237}Np behaves as a fuel in a high flux. In a lower flux the nucleus ^{238}Np decays to non-fissile ^{238}Pu before the second neutron can be captured. The nucleus may be destroyed by fission of ^{239}Pu with the release of 2.9 neutrons. On average about four neutrons are needed for destruction in the lower flux. Therefore the ^{237}Np waste is a poison in the low flux but a fuel in the high flux.

C.D. BOWMAN (57)

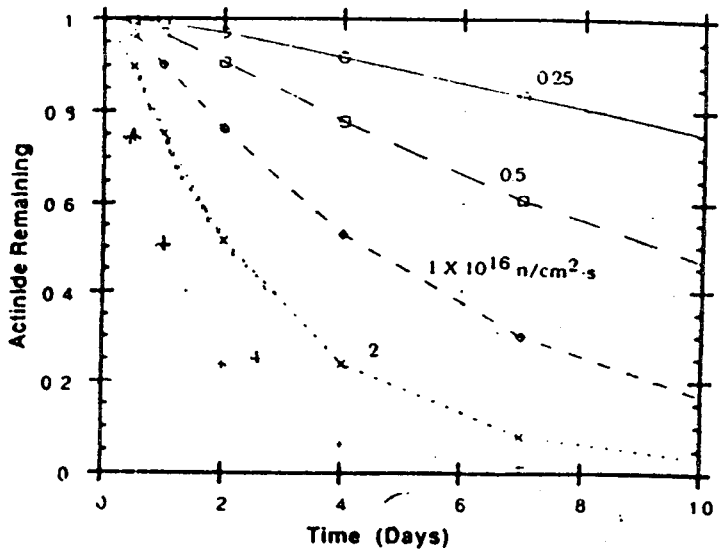


Fig.11 Transmutation of ^{237}Np in a high thermal neutron flux. The curves show the amount of actinide remaining as a function of irradiation time. Each curve differs from its neighbor by a factor of two in flux.

C.D. BOWMAN (57)

Figures 10 and 11: The fate of Neptunium in a high energy converter

2) Besides, the high neutron flux can be used to transmute the long-lived fission products ("ATW" function). Out of the seven long-lived fission products, according to Bowman, only Technetium-99, Iodine-129 and Caesium-135 have dose indices some 10 000 times greater than the four others. (50).

The "effective half-life" of major actinides and fission products can be counted in days in a high neutron flux (Fig. 12).

3) Bowman claims that in case of an accident, there is a very small inventory of fissile matter which may be dispersed. A 500 MWe reactor would need only 20 kg Pu inventory, and burn 1.8 kg of it each day in a 10^{16} neutron flux, compared to a 4-ton inventory in Superphenix. In case of a major accident, the quantity of fissile material released in the atmosphere would be minimal.

The principle of operation is shown in Figure 9, and the necessary chemical separations (volatilization of I, noble gases, fission products separations by aqueous chemistry from the salts) are schematised on Figure 13.

The problems to be solved industrially, however, are rather formidable :

- continuous operation of a molten salt reactor, with the inherent problems (corrosion, heat exchangers, etc...),
- availability of an advanced accelerator capable of 1-2 GeV with intensities high enough to take care of the fission products (100 mA ?),
- and not the least, an in-line separation of Pa-233 and of the fission products + actinides to be transmuted, to avoid that stable or short-lived compounds be transformed into radioactive or longer-lived compounds.

C.D. Bowman believes that such a system could be operative within about 20 years.

On the base of these ideas, studies and limited experiments are being conducted in different institutes around the world :

- At JAERI, in Japan, in the frame of the OMEGA project (Options Making Extra Gain from Actinides), a fast neutron transmutation proposal corresponds to a subcritical fissile region including metallic fuel in the form of Np-Pu-Zr and Am-Cm-Pu-Y, cooled by Na or Pb-Bi (Fig. 14). An alternative project proposes a molten salt continuous transmutation facility where the minor actinides salts are in solution in NaCl or other solutes and continuous reprocessing is envisaged.(51)

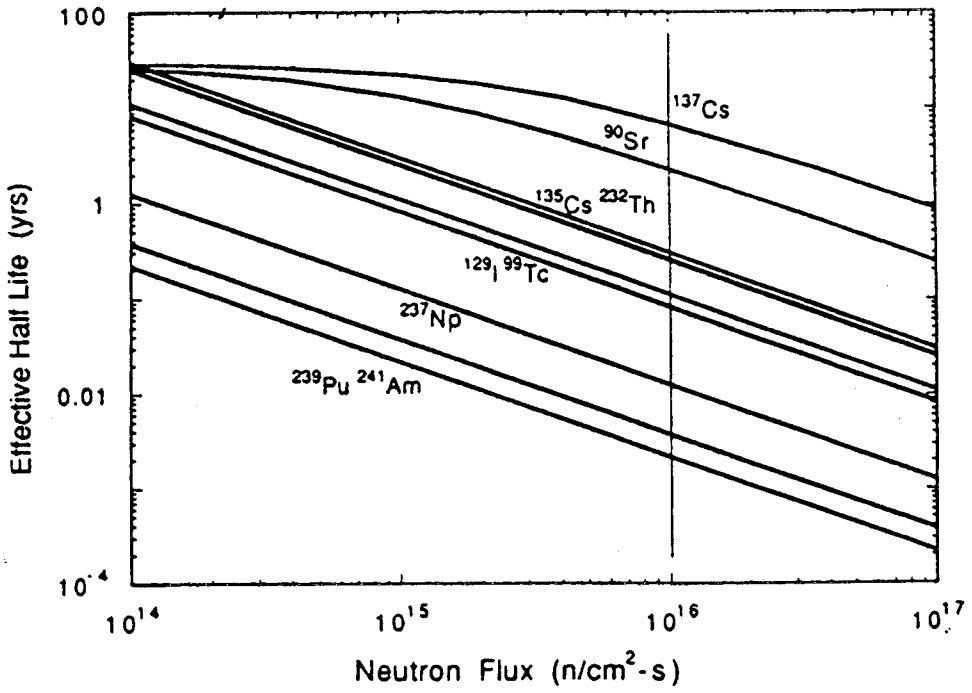


Figure 12: The dependence of effective half-life on the neutron flux for several radioactive nuclides of interest (58)

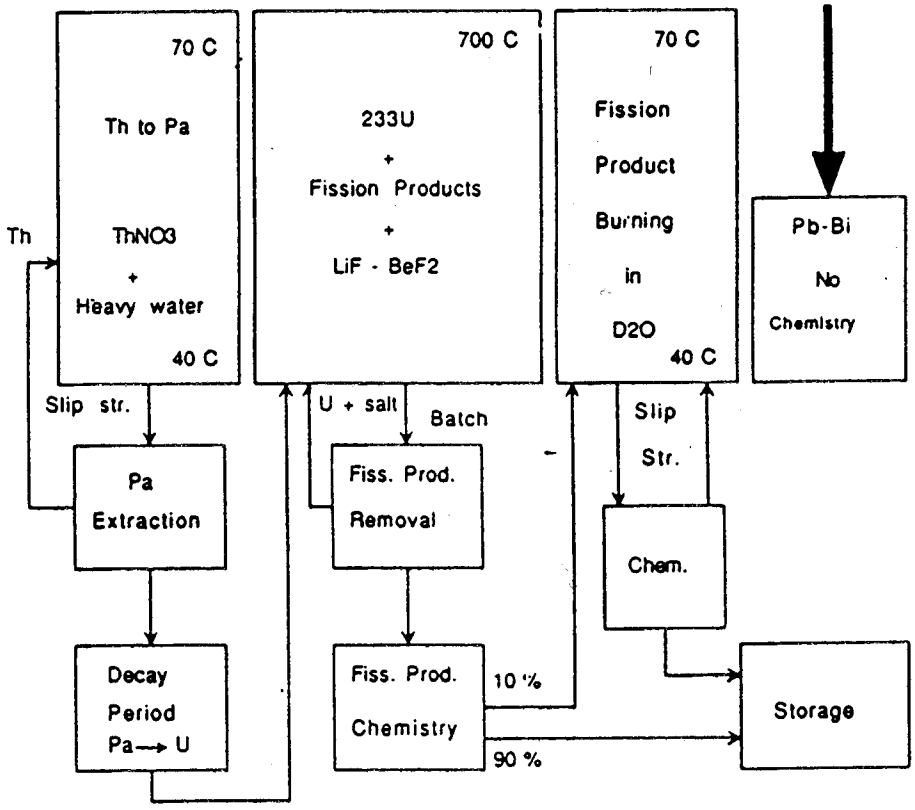
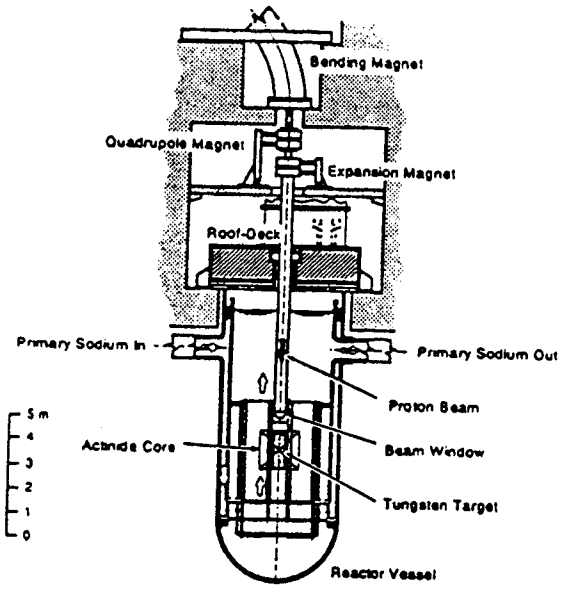
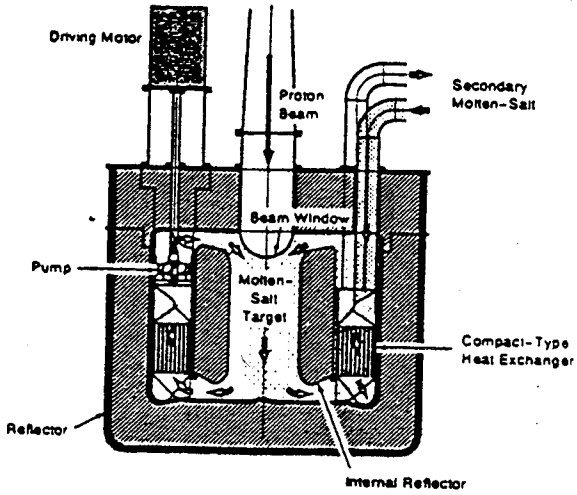


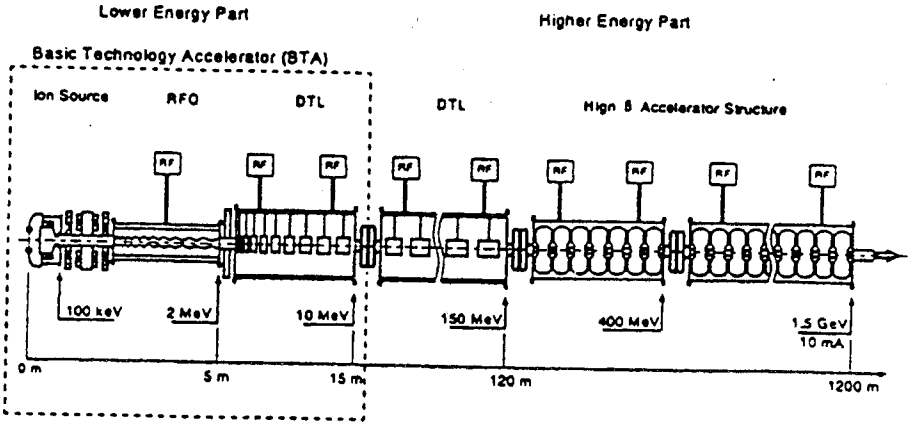
Figure 13: Chemistry requirements for the blankets three sectors (58)



Solid Target/Core System



Concept of Molten-Salt Target/Core System



Conceptual Layout of Engineering Test Accelerator (ETA)

Figure 14: Transmutation system concepts at JAERI (51)

- At BNL, in USA, the PHOENIX concept consists of subcritical accelerator driven modules, resembling the FFTF facility with a keff = 0.9. (52)

- At LANL, in USA, an intense thermal neutron source is studied. The transmutation system is composed of a proton accelerator, a heavy metal target surrounded by heavy water moderator and a molten salt blanket containing actinides and fission products to be transmuted, following Bowman's ideas. (50)

- Other projects are being studied in Russia also (53) (54) (Fig. 15) . As is pointed out by some (E.V. GAI, IPPE), extra work must be done to ascertain the equilibrium rates of production of isotopes like U-232 and U, Th lower isotopes which can be generated by (n,2n) reactions.

- In France, the SPIN Project (Separation and Transmutation) favours transmutation in an "ordinary" fast reactor or in a dedicated accelerator, so far (M. Salvatores).

However, K. Furukawa and A. Lecocq are pursuing the concept of a molten-salt breeder, able to transmute minor actinides (cf. § 3.10, Fig.1). (59)

Although only some of these projects are based on the thorium fuel cycle, they could at a given stage, and we felt useful to recall them in this Chapter.

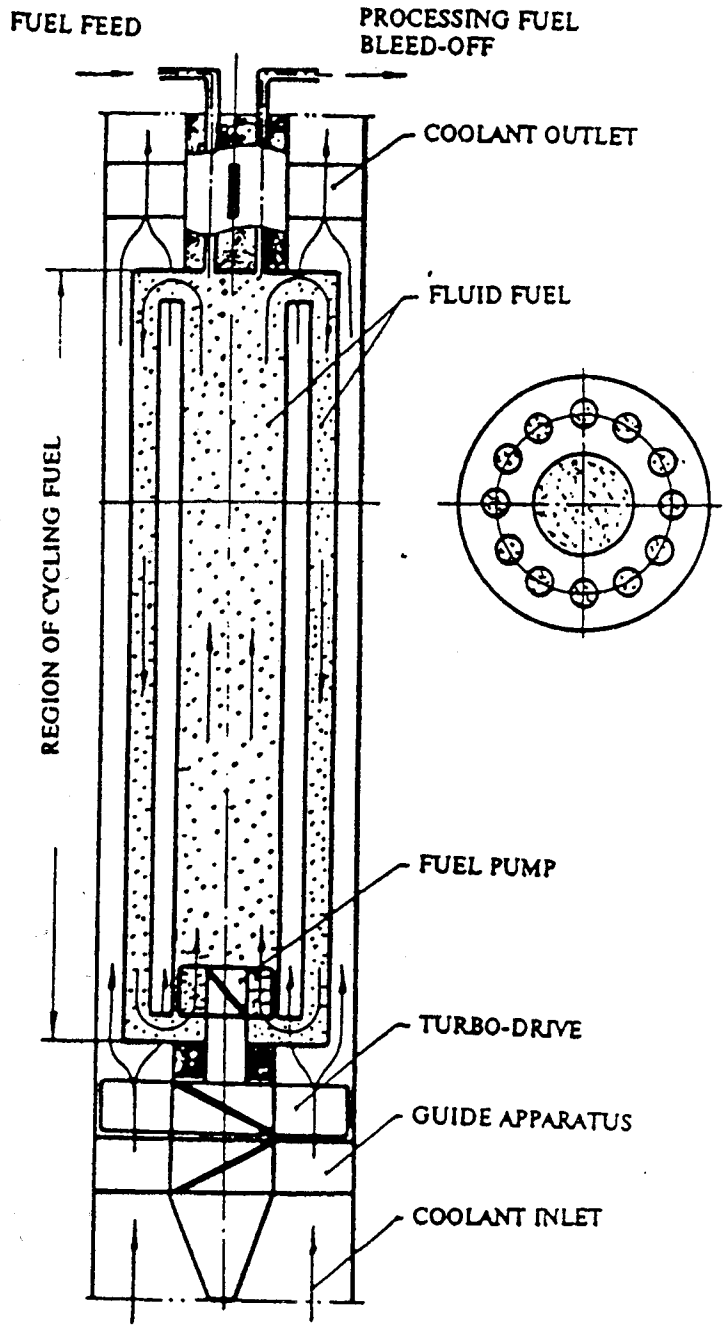


Fig. 15 CONCEPTUAL DESIGN OF CYCLING FUEL
V.D. KAZARITSKY (53)

4.4. Isotopes Production via an Accelerator-driven Subcritical Reactor

Although not really related with thorium, we felt that some remarks could be devoted to this application, because it may prove an interesting and useful testing ground for the accelerator-driven systems.

A practical application of the accelerator-driven system can be used to produce Technetium-99 ^{m}, the most frequently used radioisotope in nuclear medicine, starting from its mother fission product Mo-99.

Instead of using an expensive reactor to generate fission products, hence Mo-99 which will be "milked" to give Tc-99 ^{m}, it is possible to rely on a small hybrid system.

The advantages of such an application are considered to be :

- a rather simple machine compared to reactor irradiation;
- avoiding the reprocessing of large amounts of uranium;
- obtaining good yields of the radioisotopes.

The idea is sponsored by Ion Beam Applications S.A. (IBA) of Belgium (44), which manufactures accelerators for production of isotopes, especially mini-cyclotrons. The system is based upon :

- A proton-cyclotron : 1.5 mA, 150 MeV.

- A beam transport system to a neutron source, which is composed of a molten lead target, where the accelerated protons strike the target and produce neutrons by spallation (typical yield : ca 0.4 neutrons/incident proton at an energy of the proton of 100 MeV).

- A number of secondary targets made of highly enriched U-235 (about 120 g in total), surrounded by water as moderator (which surrounds also the primary target). These targets with the moderator constitute a subcritical assembly with a keff of about 0.8, resulting in a 5-fold multiplication factor of neutrons by fission neutrons.

- A thick graphite reflector.

- The whole system is encased in a 1.5 m thick concrete "box" for shielding. (Fig. 16).

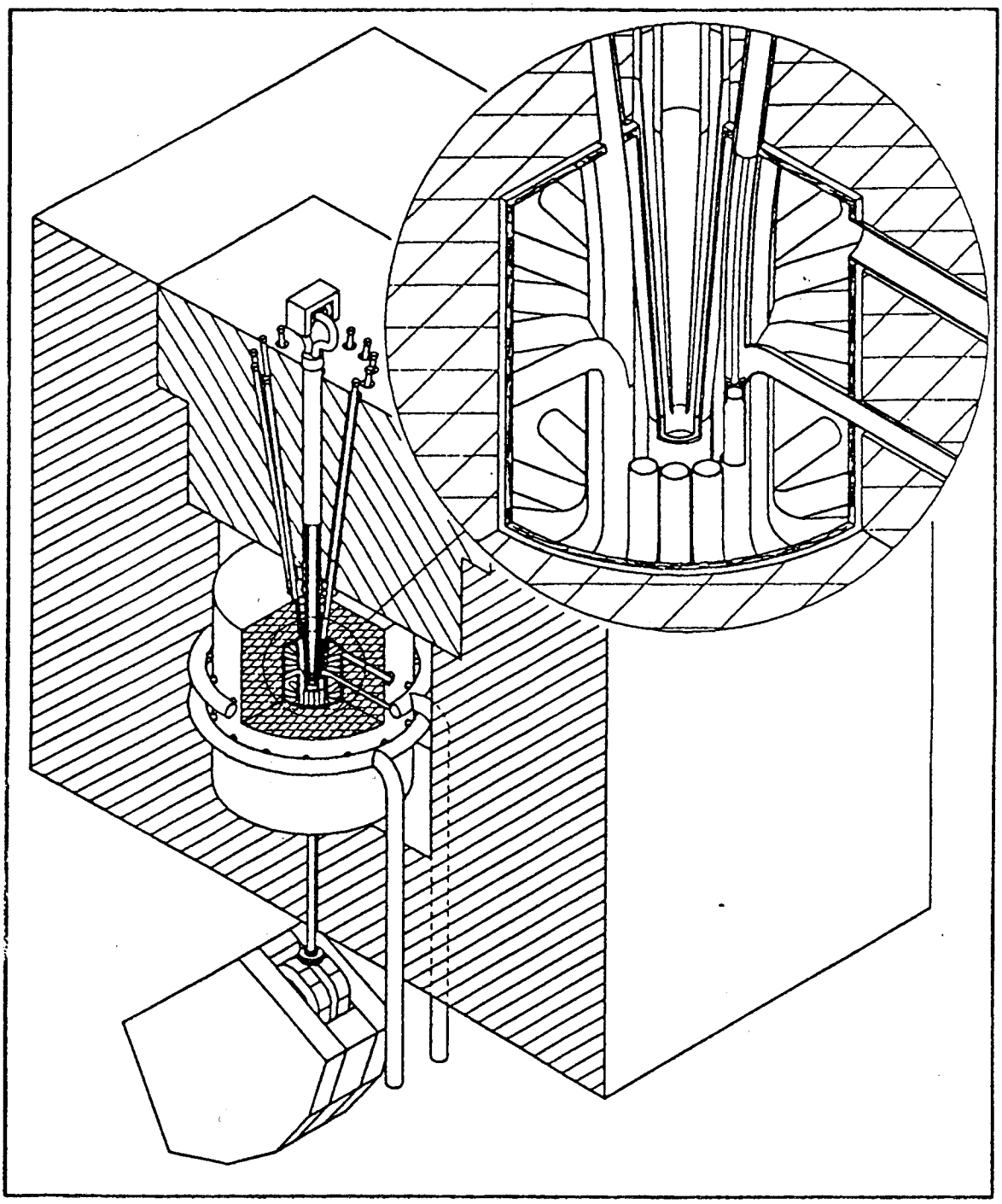


Figure 16: Axonometric view of the proposed system with a blow-up of the core region, showing the coaxial feed for the molten lead target, the surrounding cooling tubes containing the ^{235}U targets and the graphite refector. The concrete walls are 1.5 m thick. (JONGEN-44)

- Once per week, the uranium targets are replaced and reprocessed to separate the Mo-99. Most of the U-235 which is unused, will be recycled.

- The separation of the Mo-99 which is produced as a fission product is done with standard methods, and the typical values are the following : the saturation yield of Mo-99 for pure U-235 irradiated in a 2.10^{14} n/cm² is about 335 Ci/gr. The world consumption requires about $2.7.10^{4}$ Ci/week, and consequently, a total consumption of about 100 gr per week of irradiated U would be needed : a very reasonable amount for the world.

- Tc-99 distribution to the hospitals is done as Mo-99, in view of the very short half-life of Tc : 6 hours (Mo-99 has a half-life of 66 hours, 11 times higher, and then the logistics plays in favour of the distribution of Mo-99).

This rather simple system would present the advantage to permit a good decentralization of the Mo-99 production (e.g. one unit per country or per region) and would be more efficient than either the reactor production or the accelerator irradiation of Mo-100 into "instant Tc", whereby the short-half-life creates distribution and storage problems.

This application, for the time being, does not require Th or U-233, although it could. We thought it was interesting to mention it when dealing with hybrid accelerator/reactor systems.

4.5. Tentative Evaluation of the Hybrid Systems

It is today somewhat difficult to fully appreciate the impact of the hybrid systems on our future energy needs. Some have tried to put these in perspective (45) (49) (54) but a truly objective view is not yet available, in our opinion.

One must, before all, realize that all these systems are concepts, putting together some known pieces in a scientific/technological puzzle.

The state of advancement of those projects is far from that of the power systems described in Chapter 3 where industrial prototypes have been built, operated, and practical experience has been gained.

We feel that simple prototypes should be developed to get a better grasp on the physics, on the technological problems and on the economics of these systems compared to the "traditional" fission reactors. No doubt that useful findings will bring practical experience.

Some are today trying to kill many birds with one stone, viz.: :

- energy production,
- long-lived actinides transmutation,
- long-lived fission products transmutation.

The relative complexity of these different targets might blur the resulting image.

a) - If we decide that **energy production** is the main target, and that nuclear energy strikes anyway a definite positive balance between advantages and drawbacks compared to traditional fossile fuels in the longer term, as far as reserves/safety/environment/economics, then in order to meet the immense energy needs of the world, recourse to breeding is mandatory.

It is then quite possible that accelerator-enhanced breeding may bring advantages in matters of safety, doubling time, use of thorium as an extra source.

Then, the safety, the energy balance and the resulting economics must be compared with the more conventional breeder reactors, being granted that one will tend towards higher temperatures in order to obtain better thermodynamic yields.

b) - For the time being, **burning actinides** is certainly a valuable proposition in matters of safety, but we feel that this is a secondary aim, knowing that there are today technical ways developed already, albeit possibly not the best, to tackle the problems of long-lived actinides such as isolation in a reprocessing plant and trends towards "advanced" reprocessing.

It seems that hybrid systems could improve the overall economy of the long-lived actinides in some configurations, but this will have to be further tested. It should not be at the cost of too great technological complications nor too heavy expenses.

Similar remarks would apply, in our view, towards the claim of possibly reducing proliferation risks; more convincing arguments should be needed.

c) - As far as the **long-lived fission products** (LLFPs), of which 3 seem to be more toxic than the others : Tc-99, I-129, Cs-135, (due in part to their environmental mobility), according to C.D. Bowman (50), more theoretical work is definitely necessary to find out if separation/transmutation is decidedly the best solution, also considering the costs and comparing the safety of the radioactive waste disposal practised today with that of other industrial toxic waste.

Coming back to the central question, which is how to provide the 15-18 Gtoe of energy the world will need in 30-50 years, from today's 9 Gtoe, it seems well worth trying a number of avenues, among them the hybrid system.

It has been queried that the economics of an ADR could be competitive (Fig. 17) (44). These optimistic and preliminary calculations would have to be seriously verified.

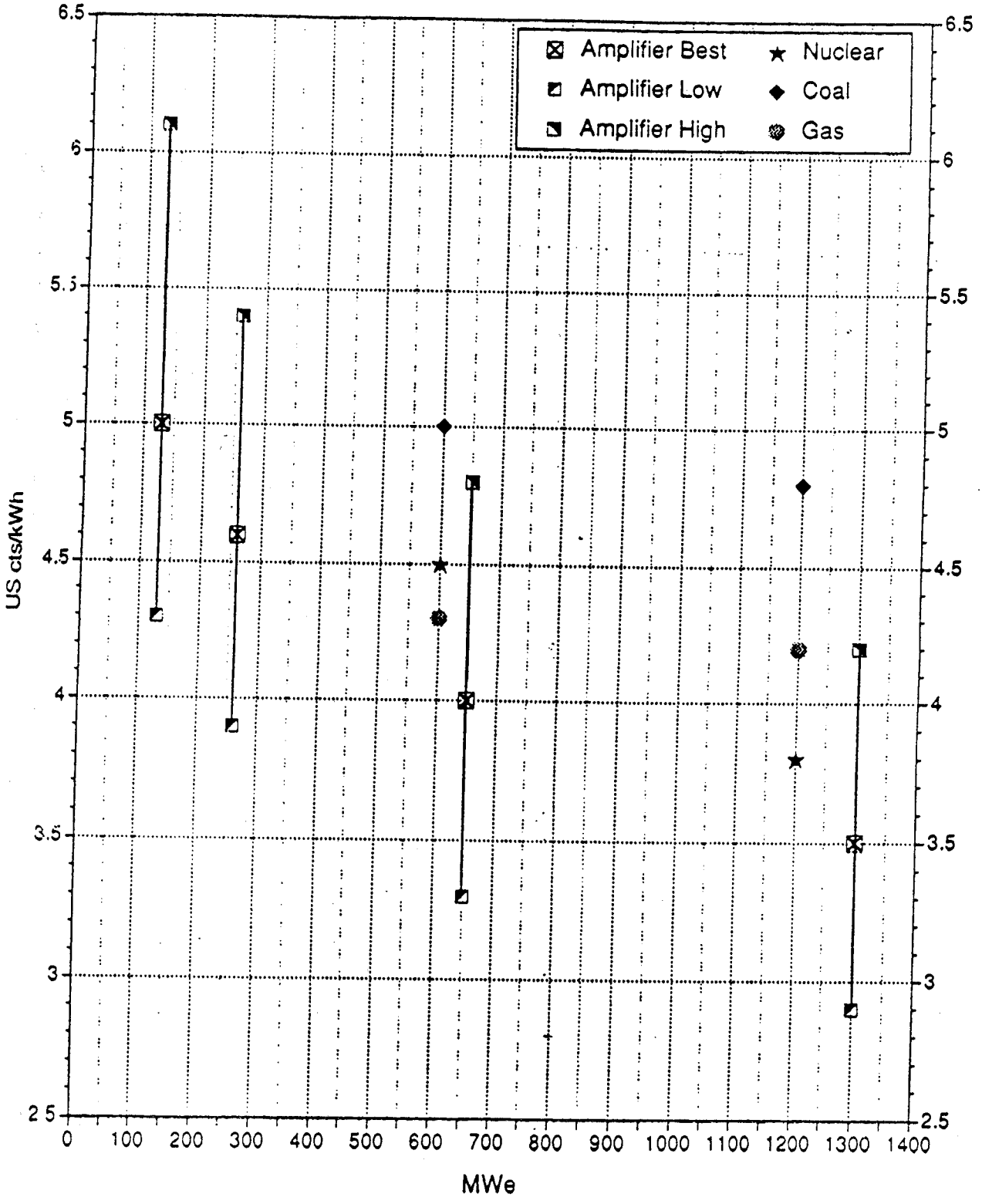


Figure 17: Evaluated cost of electrical Kwh for different options of energy transformation (Geles, Rubbia - 44)

5. THORIUM RESOURCES, THORIUM COMPOUNDS EXTRACTION AND PREPARATION

5.1. Thorium Resources

5.1.1 General remarks

Thorium was discovered by Berzelius in 1828. For a long time, production of thorium has been limited to very specific uses (special glass fabrication, gas lighting candles, special alloys) and it is so far a by-product of rare earths preparation. Its production is of some hundred tonnes per year only. It has reached about 1 000 tonnes in the 70s (85), to decrease thereafter due to some loss of interest. Besides nuclear applications, thorium is mainly used in alloys (i.e. with tungsten in electric filament lamps, and in electronics).

The known reserves (RAR - Reasonably Assured Resources) are of 1-2 million tonnes. Those of uranium are of about 1.5 million tonnes of low cost and 3 million tonnes if a price of up to $130/kg U could be paid. (74, 73, 175). (It is now around $ 20-30/kg). According to the same sources, the Estimated Additional Resources (EAR) are around 3 million tonnes.

Thorium life (alpha decay) is $1,4\ 10^{10}$ years (U-238 : $4,5\ 10^{9}$ years).

Thorium is almost uniquely composed of the isotope Th-232. Th-230 having a half-life of $7.5\ 10^{4}$ years is present as traces.

Natural thorium has a mass of 232.05, a density of 12 (uranium 18,7) and melts at 1 750° (uranium at 1 130°). Thus thorium is a highly refractory metal. Thorium oxide melts at 3 300° (U02 : 2 800°). Thorium carbide melts around 2500°C.

Theoretically, the fact that thorium has a longer half-life (by a factor of 3) than uranium could contribute to the fact that its natural occurrence is higher than that of uranium in the earth's crust. The earth's crust contains nearly 12 ppm Th, about 4 ppm U (139). Indeed, very large deposits have been found in India (360 000 tonnes), in Canada, USA, Russia, China (380 000 t), Brazil. Turkey alone may have 800 000 t of thorium. (175) The present knowledge of the thorium reserves is poor due to the relatively small efforts made so far, arising out of insignificant demand.

Thorium toxicity

Thorium is reputed to have a "similar chemical toxicity to that of uranium". Its radiotoxicity is different, however, from that of uranium. If it emits somewhat less alphas due to its longer half-life, the daughter products are notably more aggressive, being often beta emitters (Ra-228, beta, half-life 6.7 years) and gamma emitters (cf. Chap. 2.1., Fig. 1).

If one gram of thorium emits only a few hundred becquerels and only the handling of sizeable thorium quantities requires shielding, ingestion of thorium compounds is more dangerous than that of uranium, due to the filiation of natural or aged thorium.

5.1.2. **Thorium deposits**

The following table gives an idea where substantial thorium deposits have been detected.

Table 1: WORLD THORIUM RESERVES AND PRODUCTION
(Based on USBM Mineral Industry Survey, May 16, 1985)

In thousands of tonnes

	Name of the country	Reserve Base RAR
1.	USA	200 + 520 *
2.	Australia	40
3.	Brazil	70
4.	Canada	240
5.	India	360
6.	Malaysia	10
7.	Norway	150
8.	OMEC	55
9.	OPEC	35
10.	China	380 **
11.	USSR	120 **
	TOTAL	1 160

* Bastnaesite
** Excluded in the total

(AIEA TECDOC 412-1985)

It is interesting to note that in the USA alone, the ashes accumulated yearly from the coal-fired power plants contain 700 t U and 1700 t Th (1 % of which are released to the atmosphere (W.A. Gabbard, ORNL, 1995).

5.2. Nature of Thorium Resources (73)

Thorium is found in beach sands, stream placers, vein deposits, precambrian conglomerates and carbonatites.

Beach sands and stream placers. Extensive resources of monazite containing 4.6-7.0 % thorium are found in beach sands in Australia, India, Brazil and Egypt. Thorium is also present in stream placer deposits in Idaho and in the south-eastern part of the USA. Uranothorianite sands are found in Madagascar.

Veins. In the USA substantial resources of thorium are contained in vein deposits at Lemhi Pass on the Montana-Idaho border. Individual veins are up to 6 m wide and several thousand metres long. Veins have also been found in the USA in the Wet Mountains, Colorado, and at Bokan Mountain, Alaska.

In Canada, the pegmatite veins at the Bancroft, Ontario, area contain thorium. At the Madawaska uranium mine the acid-soluble thorium content is about half the soluble uranium content. In South Africa, about 54 000 t of monazite was produced from a vein at Steenkrampskraal (Cape Province) during the 1950s and early 1960s. An additional 25 000 t containing 5.3 % thorium remain in the deposit. Vein deposits also occur in Turkey.

Precambrian conglomerates. In the Elliot Lake and Agnew Lake areas in Ontario, Canada, thorium is associated mineralogically with uranium in precambrian quartz pebble conglomerates. Thorium was produced as a by-product of uranium at Elliot Lake from 1959 to 1968 and could be recovered again with favourable market conditions. The acid-soluble thorium in the conglomerates at Elliot Lake ranges from one-half to one-third the acid-soluble uranium content. However, in the conglomerates of the Agnew Lake area the ratio can be as high as four parts of acid-soluble thorium to one part of uranium. These resources are currently classified as EAR in view of uncertainty about the thorium grade and the variability of the thorium/uranium ratios.

Carbonatites and alkaline rocks. In the Fen carbonatite complex in southern Norway and other carbonatitic complexes, there are interesting concentrations of thorium, in part found in refractory minerals. The Ilimaussaq alkaline complex in South Greenland is rich in thorium-bearing minerals, first of all steen-strupine. The Th : U ratio of the rocks varies from about 1 : 1 to more than 3 : 1. Reasonably Assured Resources amount to 54 000 t Th and Estimated Additional Resources are 32 000 t Th. The cost of production is not evaluated. Speculative

Resources of uranium in this complex are estimated at some 600 000 t in rocks having more than 150 ppm U. These rocks generally have Th : U ratios of 3 : 1 to 1 : 1.

Note : Granite rock usually contains from 20 to 70 ppm of U + Th minerals.

As an example, some typical composition of monazites are given below. (11)

Table 2: Composition of Monazite concentrates in wt%

constituent	India	Brazil	Florida beach sand [1]	South Africa monazite rock	Malagasy Republic	Korea
ThO_2	8 88	6 5	3 1	5.9	8 75	5.47
U_3O_8	0 35	0 17	0 47	0.12	0 41	0.34
$(RE)_2O_3$ [2]	59 37	59.2	40.7	46.41	46 2	65.0
Ce_2O_3	(28 46)	(26.8)	--	(24.9)	(23 2)	24.7
P_2O_5	27.03	26.0	19.3	27.0	20.0	-
Fe_2O_3	0 32	0.51	4.47	4.5	-	0.35
TiO_2	0.36	1.75	-	0.42	2.2	0.19
SiO_2	1 00	2 2	8.3	3.3	6 7	4.08

[1] Florida beach sand contains about 70 wt % monazite -- [2] Rare earth oxides, including Ce_2O_3

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Table 3: Principal Thorium-Containing Minerals (11)

mineral	nominal composition [1]	examples of where found
monazite	$(La,Ce,Th)PO_4$	Brazil, India, Sri Lanka, Australia, South Africa, United States
brockite	$(Ca,Ce,Th)[PO_4,CO_3]\cdot H_2O$ [2]	United States
thorianite	ThO_2	Sri Lanka, Canada
uranothorianite	$(U,Th)O_2$	Malagasy Republic
thorogummite	$(Th,U)[(OH)_4|SiO_4]$	Brazil
thorite	$ThSiO_4$	Idaho and Montana
auerlith	$ThSiO_4\cdot YPO_4$	Idaho and Montana
uranothorite	$(U,Th)SiO_4$	Blind River, Ontario
brannerite	$(U,Ca,Th)(Ti,Fe)_2O_6$	Blind River, Ontario
bastnaesite	$(La,Ce,Th)FCO_3$	California
pyrochlore	$(Na,Ca,U,Th)(Nb,Ta)_4O_{12}$	Colorado
allanite	$(Ca,Ce,Th)_2(Al,Fe,Mn,Mg)_3(SiO_4)_3OH$	Idaho and Montana

[1] Some mineral formulae are changed according to "Uranium" Erg -Bd A 1. 1979, pp 267/73 [2] Sometimes formulated without CO_3

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The figure below gives an idea where the main deposits detected today are to be found in the world. (175)

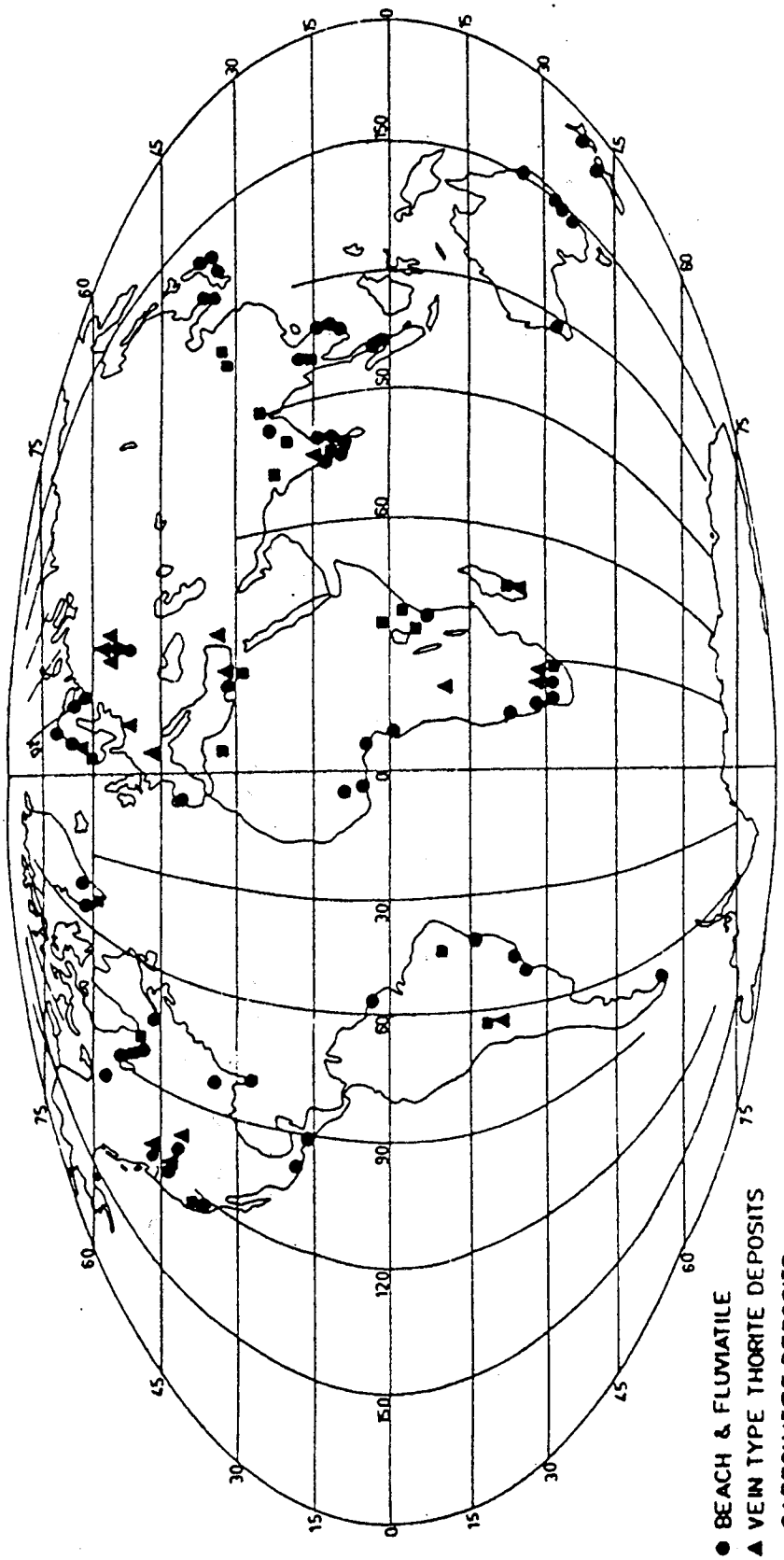

Figure 1: Thorium deposits in the world

5.3. Thorium Compounds Extraction and Preparation

5.3.1. Exploration

Thorium ore had been found by geologists looking also for other minerals. The rare earth industry and uranium industry of which thorium is a by-product, have developed the knowledge of thorium placers. Research for thorium is similar to that of uranium, with which it is very often associated.

The natural radioactivity of thorium minerals forms the basis of most prospecting instruments, though these do not measure the thorium directly but rather the intensity of gamma and beta activity (although some are based on alpha activity) from the natural isotopes formed by the decay of thorium. This gives the relative concentration of deposits.

Geological base maps of a proposed prospecting area are essential to a systematic exploration survey and these can be enhanced by aerial photography and aeromagnetic surveys. Airborne gamma-ray detection and gamma spectrometry are also good mapping aids. The gamma radiation surveys are made with Geiger and/or scintillation counters which are either carried by hand, ground vehicles, aircraft, lowered into bore holes, or trailed on lake or sea beds. Gamma spectrometer surveys are particularly advantageous for thorium prospecting as they can discriminate between the three principal radioactive elements, potassium, uranium and thorium. Interpretation of spectrometer data needs special knowledge but combined with photogeology the method can provide very good initial evaluation of a chosen area.

In geochemical surveys, properly controlled sampling and analytical procedures of lake or stream sediments can provide useful information to a prospector, but geobotanical surveys of trees and deep rooted shrubs only indicate any underlying uranium or thorium deposits which may or may not be worth exploiting.

An emanometer or radon monitor is useful for detecting in rocks or sub-soils irregular concentrations of the noble gas isotopes, radon, and thoron and their immediate decay products. The technique can distinguish between thoron (Rn-220, T = 55s, γ 0.5 MeV) from the thorium decay, or other radons from U-238 and U-235 decay (respectively Rn-222, T = 3.8 d, some γ and Rn-219, T = 3.9 s, γ 0.2 - 0.4 MeV).

Mechanical drilling is still a major method of exploration for the evaluation of underlying ores.

5.3.2. **Mining and extraction**

Deep and shallow mining are both used, and also the open-cast mining for rich shallow seams.

Processing for thorium recovery

There is a number of minerals in which thorium is found. Therefore there is a number of basic process flow sheets with modifications that can be considered for metal recovery. The thorium mineral is often associated with others, and may be treated as a byproduct of another operation. Examples of this are the beach sands concentrates in which titanium and zirconium may be the metals of primary interest, or the processing of uranium ores such as those found in the Elliot Lake area of Canada where associated thorium can be recovered as a by-product. Hence there are several process alternatives.

The case of monazite ore sands is a typical and most common one, for which physical treatments are first performed.

Physical and magnetic concentration

The beach sands that are mined in various locations in the world contain zirconium, titanium, thorium, and rare earth elements. Such a sand is that of Travancore, India, with a composition given in Table 4. These sands are amenable to physical concentration techniques to produce individual concentrates of the constituent minerals.

Table 4: Composition of Travancore (India) beach sands concentrate (1)

Composition of Travancore Beach Sands Concentrate, India

	mineral	content in wt %
monazite	$(RE.Th.U)PO_4$	0.5 to 1.0
ilmenite	$FeTiO_3$	65 to 80
garnet	$(Fe.Mg.Ca)_3Al_2SiO_4$	1 to 5
rutile	TiO_2	3 to 6
zircon	$ZrSiO_4$	4 to 6
sillimanite	Al_2SiO_5	2 to 5

Pinched sluices and trays, cone concentrators, spirals and wet tables are typical of the plants used for the primary concentration of the heavy minerals found in combination with monazite. Usually the bearing sands contain 0.5 - 2 % monazite, but some contain as much as 40 %.

Individual minerals - ilmenite, rutile, monazite, zircon, sillimanite and garnet - are separated by utilizing the differences in their physical properties, i.e. specific gravity, magnetic susceptibility, electric conductivity and surface properties. The wet concentrate is passed through rotary dryers at up to 150°C, the dried feed then being electrostatically or electromagnetically treated. The electrically conducting ilmenite and rutile constituents are first separated. The non-conducting monazite, being heavy and moderately magnetic, is isolated by high intensity magnetic separators and air or wet tables. The resultant concentrate contains 98 % monazite.

Usually, feeds containing 1 - 2 % of heavy minerals found in combination with monazite can be concentrated to 90 % heavy minerals with an overall recovery of 85-90 %.

Two typical separation/concentration flowsheets are given below, one for Indian beach sands (175) (Fig. 2), the other for Australian beach sands (11) (Fig. 3.).

5.3.3. **Heavy metal chemical extraction**

Chemical treatment

The chemical properties of thorium and the rare earth elements associated with it are closely similar, so their separation is difficult and time consuming. Concentrates of the rare earths must be chemically processed to separate them from the other components forming the mineral and from impurities.

Monazite, the chief commercial ore from which thorium is extracted, is chemically inert and any chemical treatment for extracting thorium must initially be very severe to achieve the complete dissolution necessary for the separation of the rare earth elements (a valuable by-product), uranium (which is frequently present) and phosphates. Many processes are available, having their own variations and techniques and they are often proprietary.

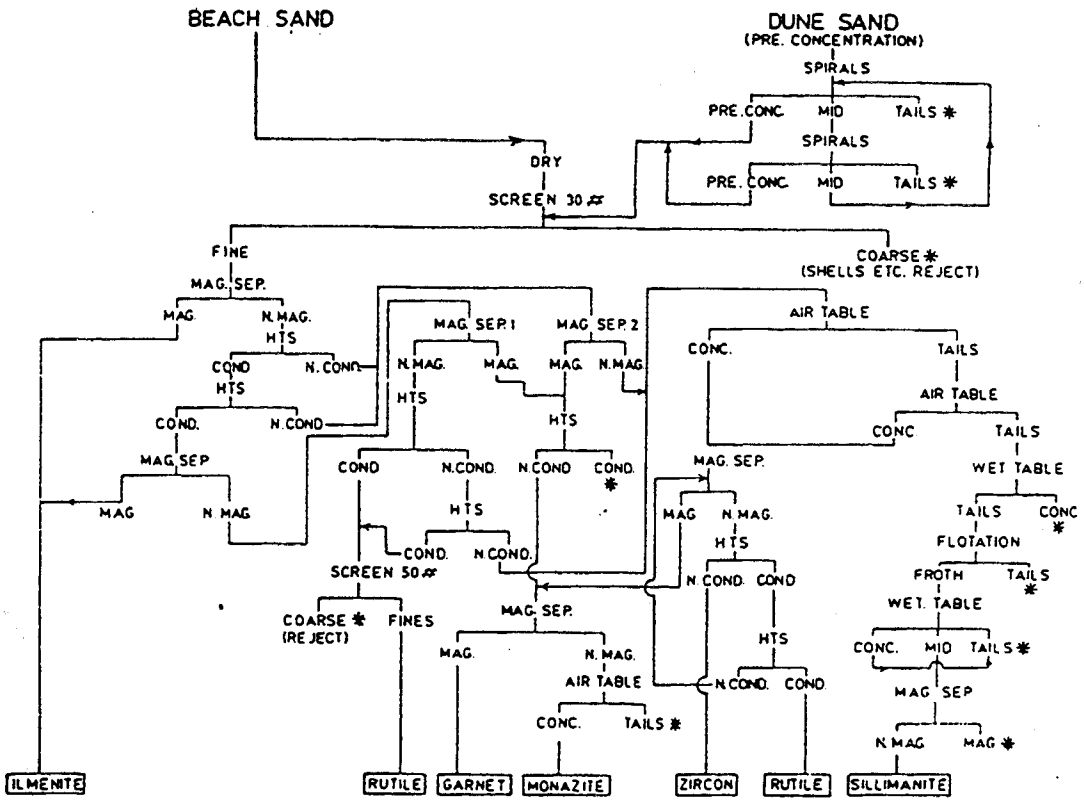

Figure 2: General flow sheet for treatment of west coast beach sand (India) (H.T.S. = High Tension Seperator)

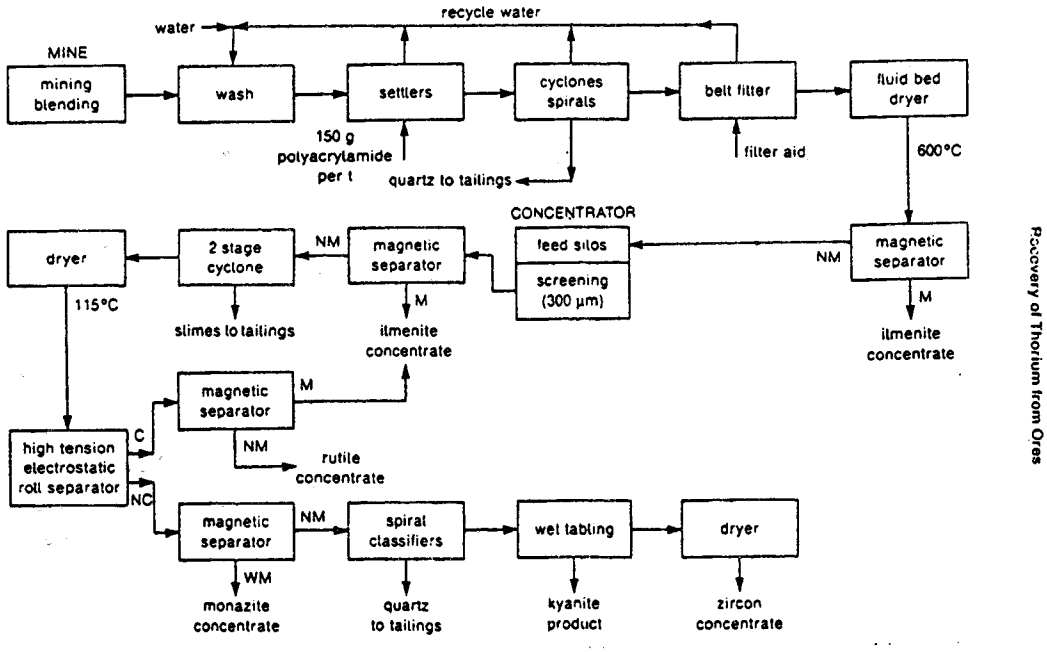

Figure 3: Schematic folow sheet for treatment of Australian beach sands. From (5). Magnetic and electric properties of the ore minerals: ilmenite (M.C), rutile (NM, C) monazite (WM, NC), zircon (NM, NC), quartz etc (NM, NC). M = magnetic, WM = weakly magnetic, NM = non-magnetic, C = conducting, NC = non-conducting

The most common dissolution processes are :

(1) acidic - using highly concentrated sulphuric acid,
(2) alkaline - using highly concentrated sodium hydroxide.

These are described here but other processes such as sodium carbonate roasting, chlorination and carburisation have been adopted. The two more common processes are rather straightforward, but quite tedious, very similar to rare earth extractions which have similar reactions.

The acid process

There are several possible acid processes of which just one will be described here. Monazite, ground to below 100 mesh, is reacted above about 200°C in highly concentrated sulphuric acid in agitated heated pots for about 24 hours before the slurry is cooled and added to cold water in leach tanks fitted with agitators. The rare earth and thorium sulphates dissolve leaving silica, rutile, ilmenite and zircon residues which are filtered off. Sodium pyrophosphate is added to the solution and thorium pyrophosphate is precipitated and recovered by filtering, leaving a rare earth sulphate solution.

A typical flowsheet is also given for a sulphuric acid digestion process for monazite. (70) (V.S. Keni) (Fig. 4).

The alkali process

The alkaline process requires the monazite to be finely dry ground before treating it in 50-70 % sodium hydroxide for about 4 hours at about 140°C in a cast-iron pot equipped with an agitator. The slurry is settled for several hours and the solution containing water-soluble trisodium phosphate and excess sodium hydroxide is decanted and filtered leaving the rare earth elements and thorium as insoluble hydroxides. Treating the hydroxides with hydrochloric acid (pH 3 - 5) dissolves the rare earth hydroxide and leaves solid thorium hydrate only slightly contaminated with rare earth hydroxides. The thorium is recovered by a stronger acid solution.

A typical flowsheet is given for the caustic soda process (11) (Fig. 5).

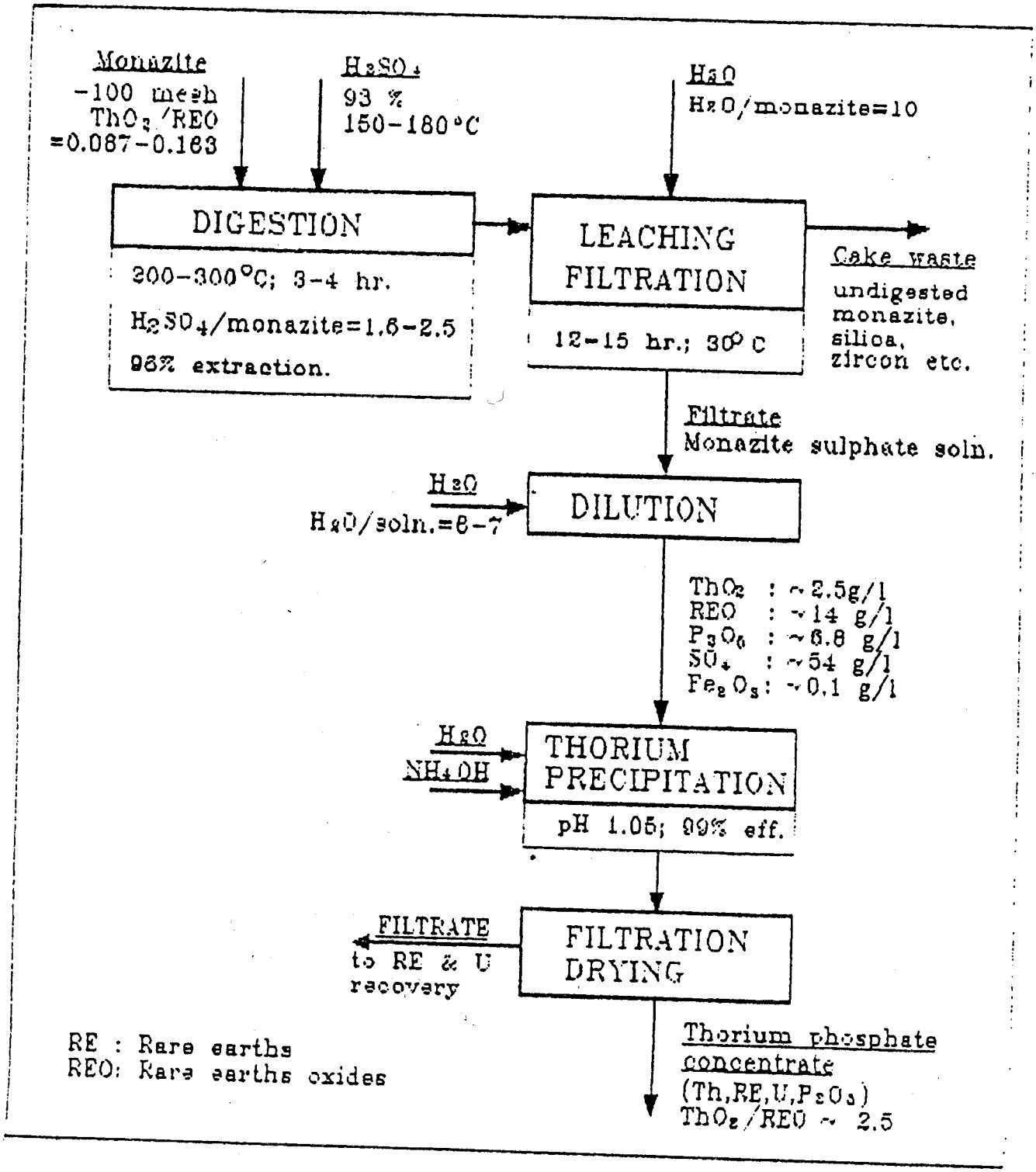

Figure 4: Sulphuric acid digestion process for preparing thorium concentrates from Monazite (70)

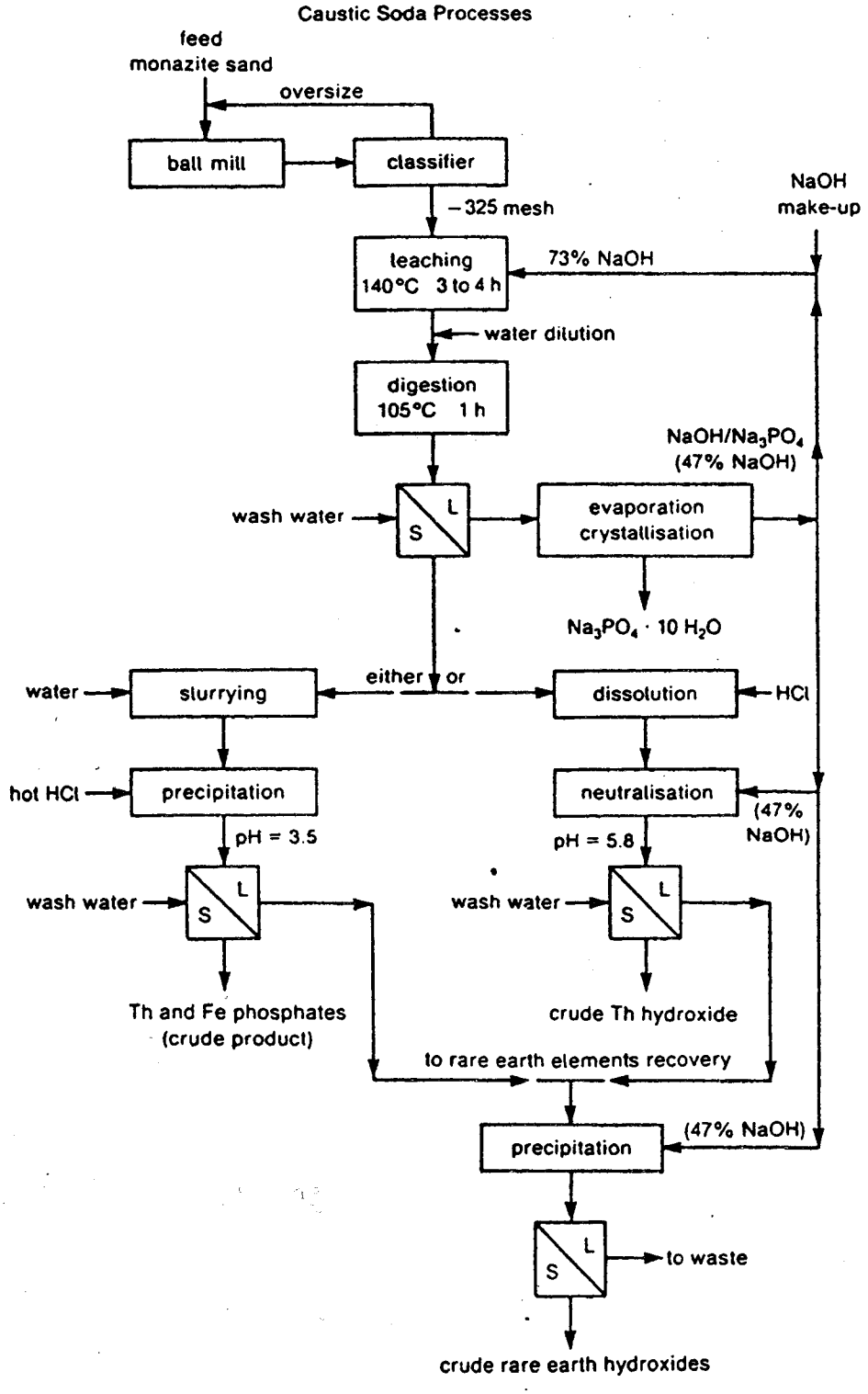

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Figure 5: Schematic flow sheet of the caustic process

5.3.4. **Refining**

For nuclear applications thorium has to meet stringent requirements of purity, particularly concerning neutron absorbing elements. This purity is obtained industrially by solvent extraction, by ion exchange, or by direct chemical precipitation.

Solvent extraction makes use of the different solubilities of some heavy metal compounds in organic solvents which are immiscible with water. Successive transfer operations between the aqueous and organic phases thus lead to a relative concentration of one heavy metal in the aqueous phase and another in the organic phase. The transfer coefficients can be varied, e. g. by variation of nitric acid concentration so that the method can be made very flexible and recovery of material from the solvent phase can be achieved by back-transfer at a different acidity level.

In purifying thorium by this process, the crude thorium produced by a chemical treatment as discussed above, is converted to nitrate in which form it exists in an aqueous phase. Contact between the aqueous carrier and the organic solvent (tributyl phosphate in kerosene or hexane) transfers thorium to the solvent from which it is stripped by scrubbing with dilute nitric acid. Nuclear-grade thorium nitrate is obtained with more than 99 % thorium recovery by successive operations.

Amines as extractants from sulphate solutions are also very efficient. (11)

Ion exchange is an effective way of separating individual lanthanides in a pure state. Basically, it is a chromatographic technique for separating ions by their sorption from solutions on a suitable ion exchange medium followed by differential displacement of the individual absorbed ions with an eluting solution.

Direct precipitation processes use either the addition of foreign ions to precipitate insoluble salts, or else exploit variations in solubility with pH of complex salts. Repeated precipitations are needed to ensure adequate purity.

A typical separation flowsheet for Th and rare earths nitrates is given below (1) (Fig. 6), in the case when there is little uranium present initially (category "A" concentrates). If there is uranium present with Th and R.Es (category "B" concentrates), a more sophisticated extraction flowsheet, with two TBP concentrations and appropriate scrubbing and stripping stages in liquid-liquid

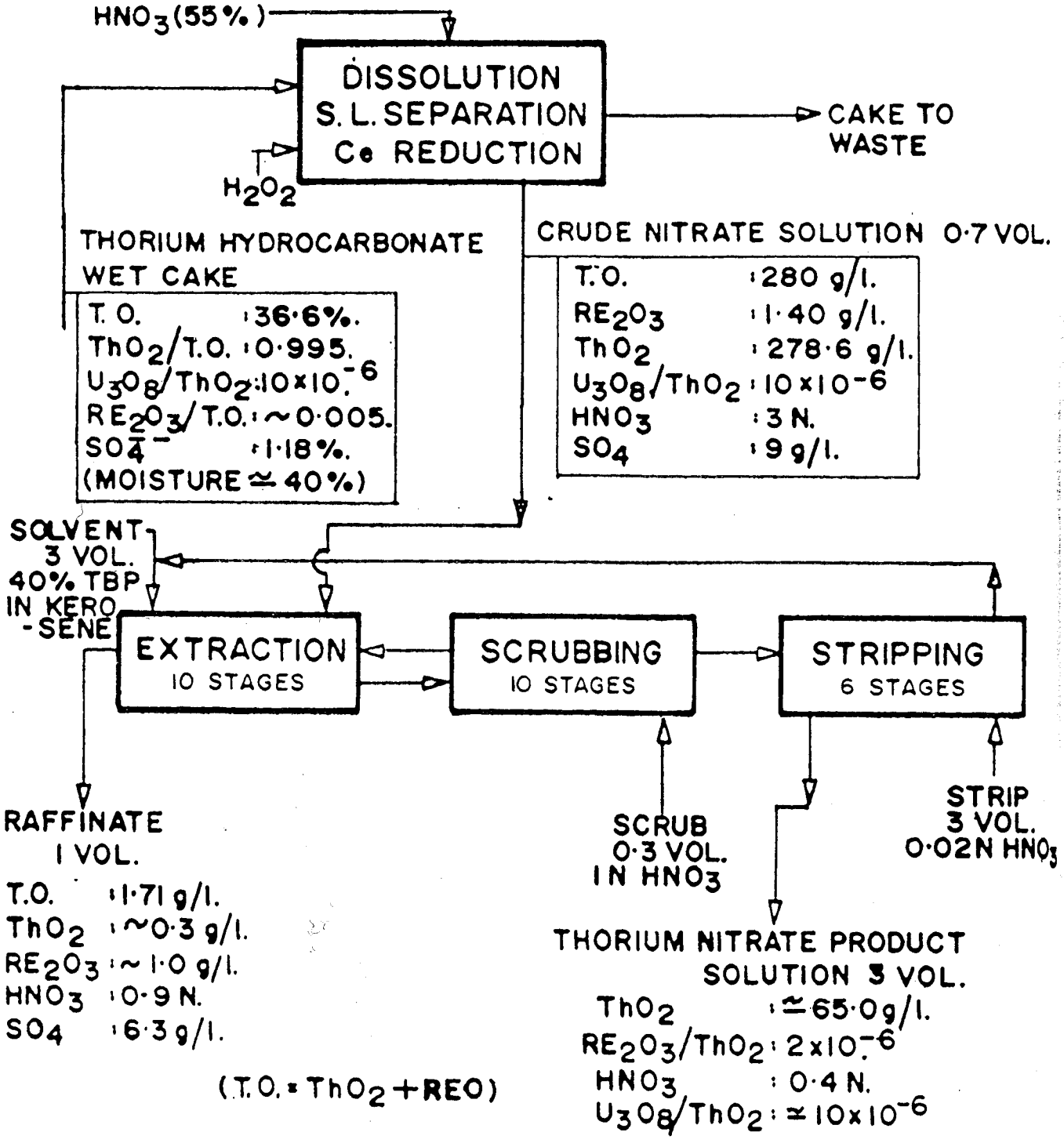


Figure 6: Flow sheet for solvent-extraction-purification process to prepare category "A" concentrates (70)

contactors, would separate uranium from thorium nitrates, if necessary. The purity obtained is U in Th : 10 ppm, rare earths in thorium, about 3 ppm.

5.3.5. **Thorium production estimates**

During the period from 1960 to 1970, thorium was mentioned as both an alternative and a successor to uranium for nuclear power reactors. Therefore the possibility of geological exploration for new occurrences and methods to treat the various thorium-containing minerals became important. However, although the demand has not as yet materialized, considerable research was carried out in the application of various separation techniques for the recovery and isolation of thorium from various minerals.

In Table 5. are shown some production data for thorium for 1973. Also given are the tonnages of monazite concentrates for 1976, 1977 and 1978.

Table 5: Production rates of thorium and monazite concentrates (11)

Thorium Production Rate
NA = not available.

	monazite concentrates, in tons per year, for			tons thorium per year for
	1976	1977	1978 (estimated)	1973
United States	NA	NA	NA	[1]
Canada	0	0	0	0
Brazil	1628	1814	1814	82
Soviet Union	NA	NA	NA	[1]
Zaire	[1]	[1]	[1]	12
Other African	[1]	[1]	[1]	6
India	3027	3027	3118	266
Malaysia	1899	2018	2201	138
Thailand	[1]	[1]	[1]	14
Australia	4601	8847	8621	275
Other	262	120	92	138
total	11417	15846	15867	930
tonnes thorium at 6%	657	951	952	

[1] included in other

In 1979 the current thorium production estimate was of 200 tonnes per year. (176)

The total amount of thorium which has been prepared in the US is estimated at some 2 000 tonnes, as well as 2 000 tonnes in the Federal Republic of Germany, until 1988. Some quantities are produced in other countries, Australia, Canada, and especially India, to provide for the ongoing programme. France had produced about 2000 tonnes of purified thorium nitrate, sold in majority to the USA.

5.3.6. **Reduction to thorium metal or thorium oxide**

Purified thorium nitrate can be used as feed material for producing thorium.

The reduction of thorium compounds to the pure metal thorium is not easy because at its high melting point of about 1 700°C thorium reacts readily with hydrogen, oxygen, nitrogen, carbon and many oxides. The metal is usually produced as a sponge or powder by one of the following methods :

(1) Thorium tetrafluoride is prepared by heating thorium oxide to 325°C and exposing it to anhydrous hydrofluoric acid gas. The thorium tetrafluoride is then reduced with calcium at 800°C in the presence of a zinc chloride booster. The booster reacts with calcium in an exothermic reaction thereby aiding in liquefying the thorium compound and fluxing the slag. The zinc is subsequently removed by pyrovacuum treatment at 1 360°C.

(2) Thorium oxide is obtained by precipitating thorium oxalate from a thorium nitrate solution with oxalic acid and calcining the thorium oxalate at 650°C or above. The thorium oxide is then reduced to metal with calcium at 1 000 - 1 100°C, using calcium chloride as a flux.

(3) Electrolysis of tetrachloride or tetrafluoride : thorium tetrachloride may be obtained by chlorinating a mixture of thorium oxide and carbon at 600°C and purifying the first distillate by redistillation. Alternatively, thorium tetrafluoride may be obtained as described above. Molten salt electrolysis of thorium chloride or fluoride in graphite crucibles which act as anode with molybdenum as the cathode, results in thorium metal being deposited on the cathode.

6. THORIUM-BASED FUEL ELEMENT FABRICATION

6.1. Introduction

Thorium fuel fabrication is used to breed U-233 in thermal or fast reactors, under the form of thorium metal rods or ThO2 pellets in tubes, or ThO2, ThC microspheres generally associated with fissile material (UO2, PuO2, UC, PuC). In the latter case, the fuel will present more homogeneous properties, and the fabrication will be more complex, from a physical point of view as well as a radiological point of view, as it often entails a rather radioactive "driver" fissile component.

This chapter has been rather extensively developed, as in our view, it represents *a* crucial point of the development of the thorium fuel cycle.

6.2. Thorium Metal Fuel Elements

(We are indebted to R. Vijarayaghavan, BARC, DAE, India (82) for the elements of this paragraph).

Thorium metal pellets have, inter alia, been prepared to fabricate "J-rods" (Fig.3) to be inserted into the CIRUS reactor for irradiation and U-233 extraction.

Thorium metal powder is obtained by reduction of ThO2 by calciothermy of other means (cf. Chapter 5) (Fig.1).

Thorium metal powder is quite ductile and can easily be consolidated by conventional powder metallurgical techniques. Thorium pellets of sintered density of about 98 % TD can be produced by compacting at a pressure of 300 MPa followed by sintering in vacuum at 1300° C for 1 hour. Proper lubrication of die walls is essential. Sintered pellets have been cold rolled to more than 90 % reduction in thickness without any intermediate annealing. Because of its tendency to seize to steel dies and oxidise in air, copper jacketing of thorium is resorted to during hot extrusion and cold drawing operations.

Thorium has been converted into various shapes such as strips, blocks, rods, tubes, wires and foils, etc... by using conventional fabrication techniques (Fig 2).

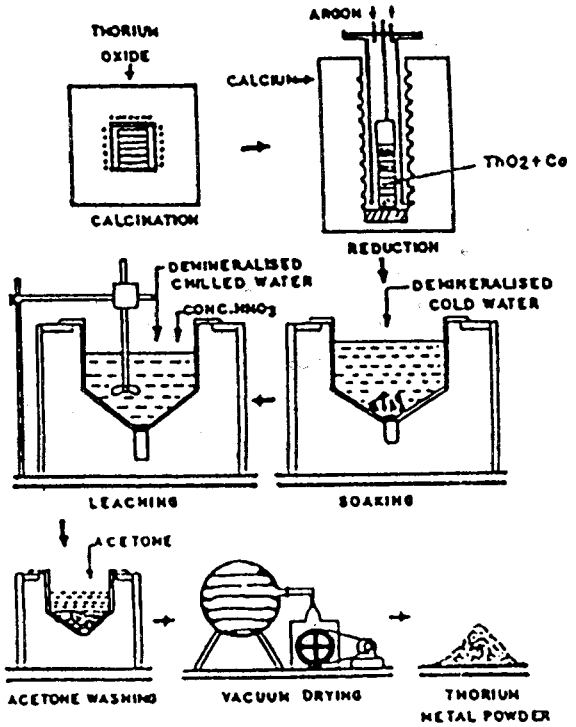


FIG.1- PRODUCTION OF THORIUM METAL POWDER BY CALCIO-THERMIC REDUCTION OF THORIUM OXIDE

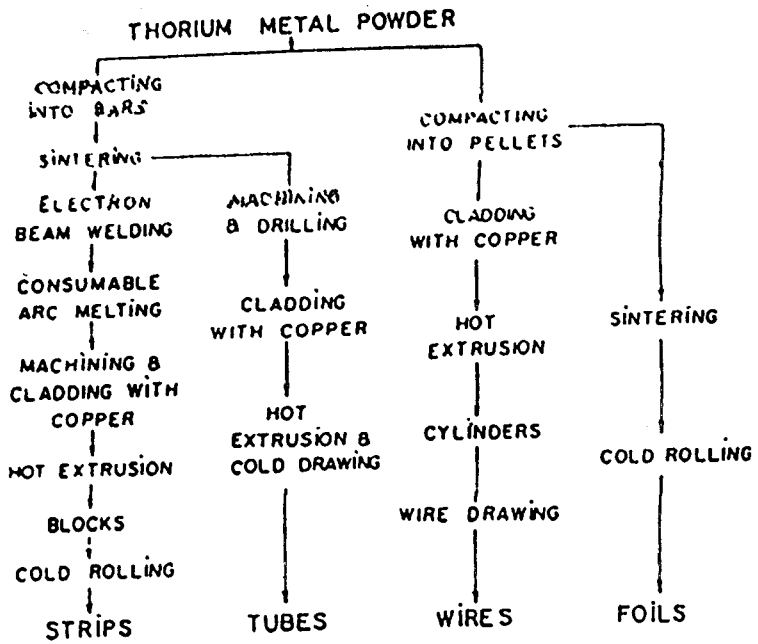


FIG. 2- FABRICATION OF THORIUM METAL SHAPES

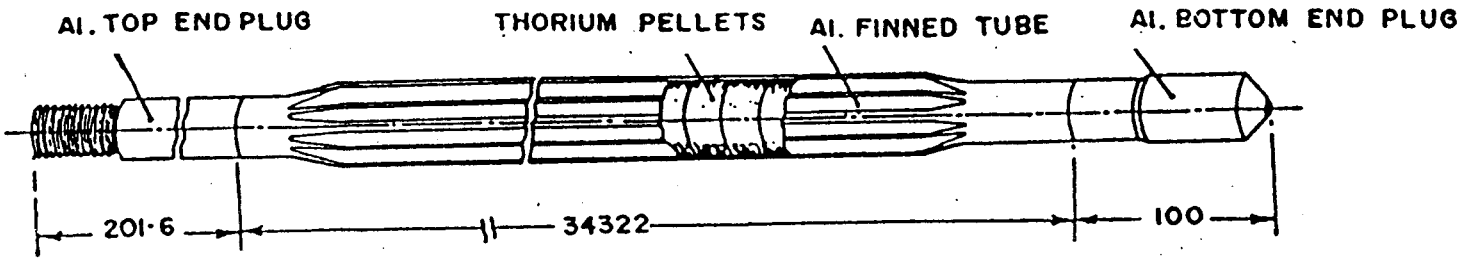


FIG.3-THORIUM J-ROD

Thorium is a very soft metal and can be scratched with a knife. A fresh surface of thorium exhibits a bright silvery finish which tarnishes or darkens with prolonged exposure to air. Oxygen has virtually zero solubility in thorium at room temperature. The oxide inclusions are dispersed at random and do not seriously impair the mechanical fabrication even in amounts up to 3 % by weight. Nitrogen has a solid solubility of 0.17 wt % at room temperature. Carbon, even present in small quantities, greatly impairs the fabricability of thorium by markedly increasing the tensile strength and reducing the ductility. The metal work hardens rapidly during the initial stages of deformation. Additional working increases the hardness only slightly and hence extreme degrees of cold work are possible with thorium.

Extensive experimental investigations were carried out on the compatibility of thorium with various materials such as stainless steel, zirconium, chromium, vanadium, etc... It has been found that iron and nickel from stainless steel diffuse into thorium at 500° C forming brittle phases such as ThNi,x. Thorium diffuses into zirconium at about 800° C. Chromium and vanadium are both compatible with thorium up to 1000° C.

Safety precautions :

Owing to the toxicity, radioactivity and pyrophoricity, adequate precautions are required to be taken in handling and processing of thorium. Pure or fresh thorium is a weak alpha-emitter but old thorium with accumulated decay products also emits beta-particles and penetrating gamma rays (cf. Fig.1, Chap. 2.1.). However, as a result of the very long half-life of thorium decay, these emissions do not present a real danger for direct handling of limited thorium amounts (some hundreds Bq/g). Some shielding is required for large amounts. Thorium is a very active metal and in finely divided form it can be pyrophoric and in dust form it may be explosive. The tendency of the metal powder to ignite spontaneously depends to a large extent on the fineness of the powder. The presence of moisture and hydride as well as residual calcium may render thorium metal pyrophoric. The pyrophoricity can be minimized by giving thorium metal powder a protective treatment. This treatment usually consists in immersing the powder in an aqueous solution of a salt of a metal that is less electro-positive than thorium.

6.3. Thorium Oxide Fuel Elements

6.3.1. **Indian Experience** (81) (83)

The use of thorium in reactors has been mainly envisaged in the form of thorium oxide (thoria) in view of the greater experience and proven performance of the oxide fuels. The conventional dry powder metallurgy techniques of compacting and sintering are adopted for the fabrication of both low and high density thoria pellets. The conditions of compaction of thoria are similar to those employed for UOx. However, unlike the sintering of UOx, where the sintering atmosphere and O/U ratio play an important role, ThO2 being the most stable oxide known, can be sintered in any atmosphere such as air, hydrogen or vacuum. Pressing and sintering characteristics of thoria powders calcined from different compounds such as oxalate, nitrate, hydroxide and hydro-carbonate and the effect of additives such as CaO, CaF, MgO and Nb2O5 on the sinterability of thoria have been extensively studied. Thorium oxide is a difficult material to sinter and even at temperature of 2000° C the achievable density is lower than 80 % TD. Densities 94 % of TD can be obtained by sintering at about 1600° C in H2 with 0.02 % MgO additive, while with 0.25 % Nb2O5 additive thorium oxide could be sintered at 1150°C-1200°C in air to get the same density. In case of MgO additive with reducing atmosphere like H2, the activation is considered to be due to metal interstitials or vacant oxygen sites created in the structure by substitution of Th by Mg which has a lower valency. The remarkable effect of doping with Nb2O5 is considered to be due to oxygen interstitials or vacant thorium sites created in the structure by the substitution of Th by Nb which has a higher valency. The oxidative furnace atmosphere of air further reinforces the formation of oxygen interstitials and hence sintering rates are enhanced at low temperature of 1150-1200°C.

Figures 4 and 5 give the flowsheets for fabrication of high density thoria pellets. Such pellets have been fabricated for use in the CIRUS and DHRUVA thermal Indian research reactors, and experimental fission/fusion hybrid systems.

Figure 6 shows a typical thoria fuel element clad in zircalloy, for the Dhruva reactor.

Similar fabrication methods have been used for the blanket fuel pins and fuel elements of the Indian Fast Breeder Test Reactor (FBTR).

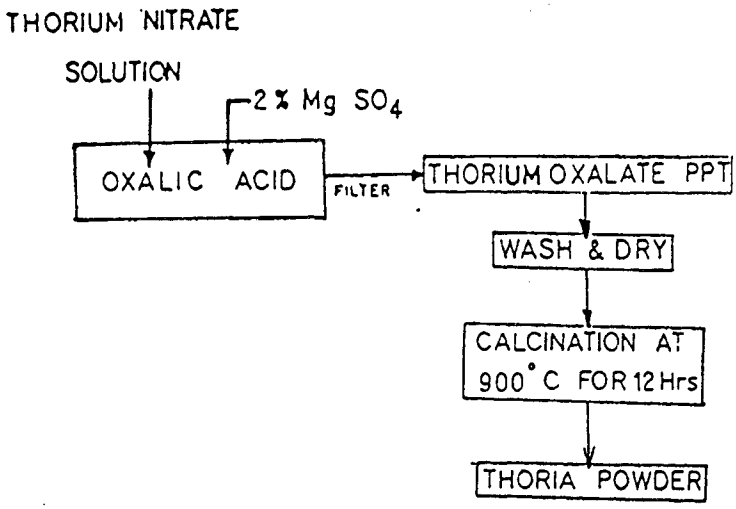


Fig. 4 Flow sheet for production of thoria powder

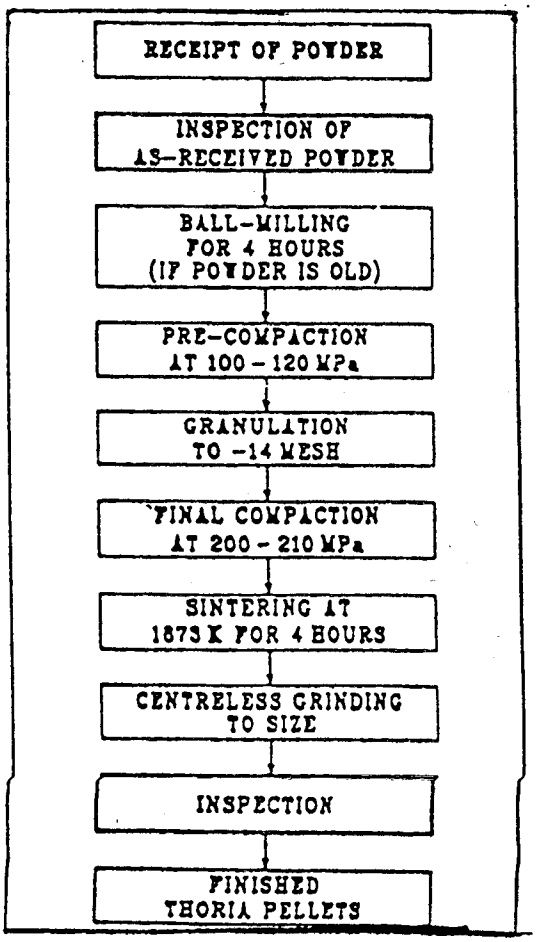


FIG-5. THORIA PELLET FABRICATION PROCESS FLOW-SHEET

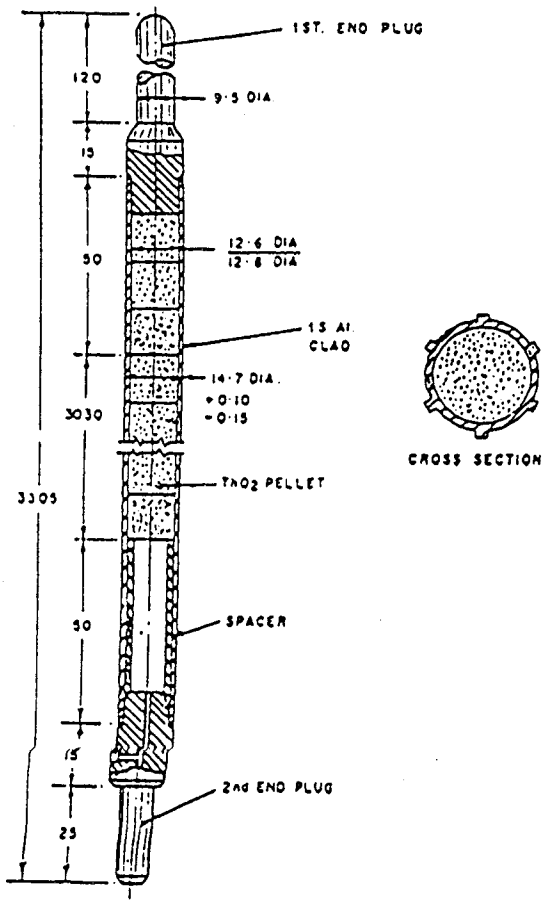
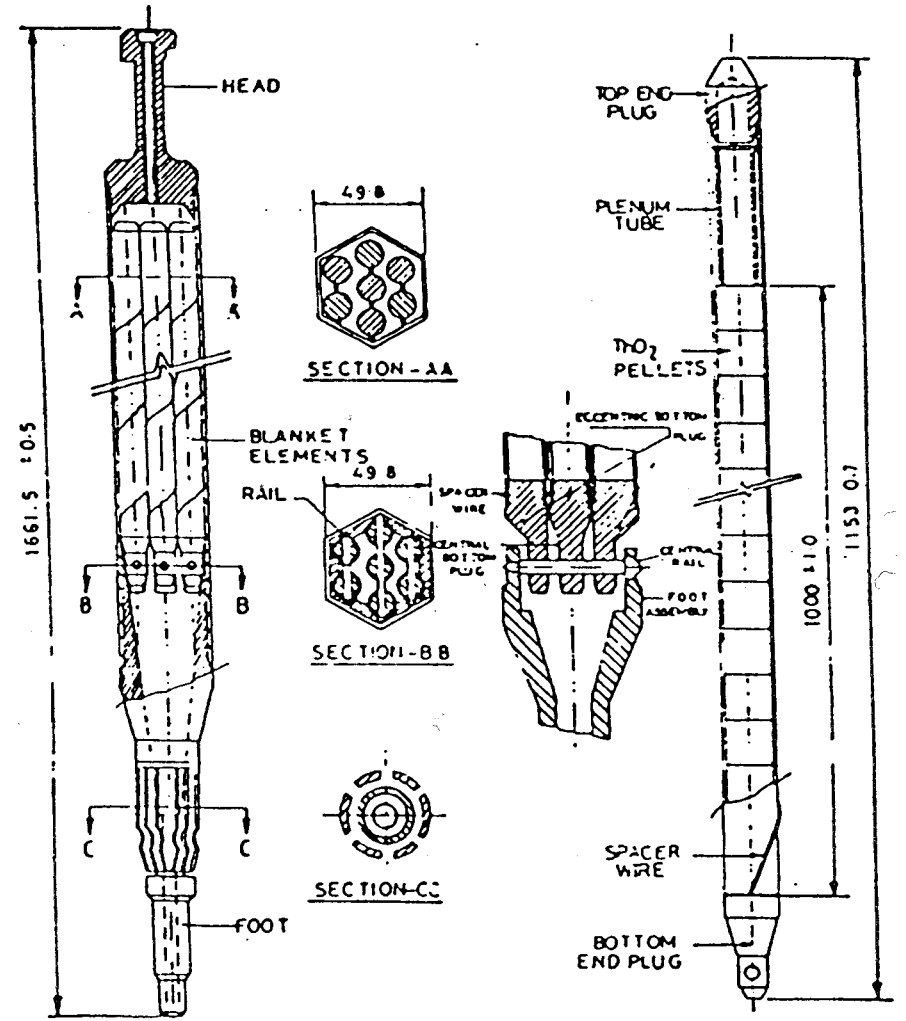


FIG.6 - THORIA ELEMENT FOR DHRUVA CLUSTER



RADIAL

FIG: 7 BLANKET ASSEMBLY

F.B.T.R.

RADIAL

BLANKET ELEMENT

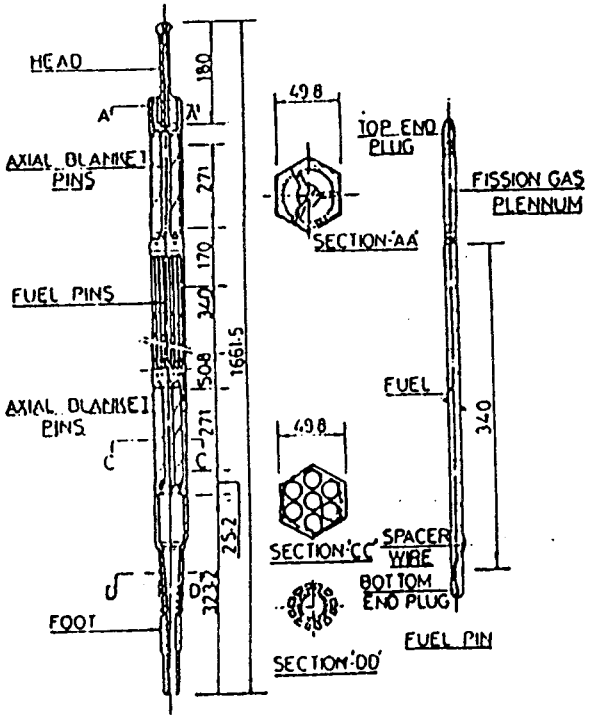


FIG:8 FUEL SUB-ASSEMBLY

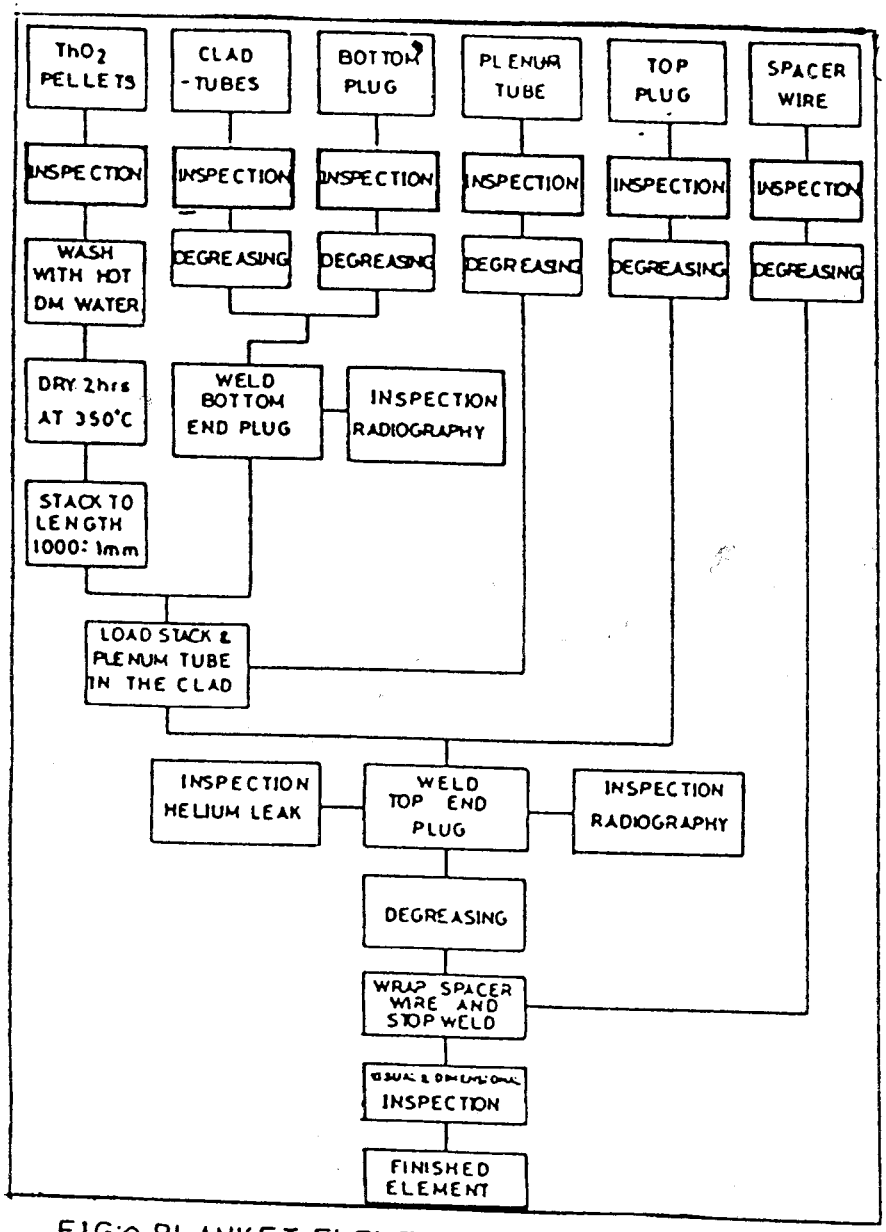


FIG:9. BLANKET ELEMENT FABRICATION PROCESS FLOW-SHEET

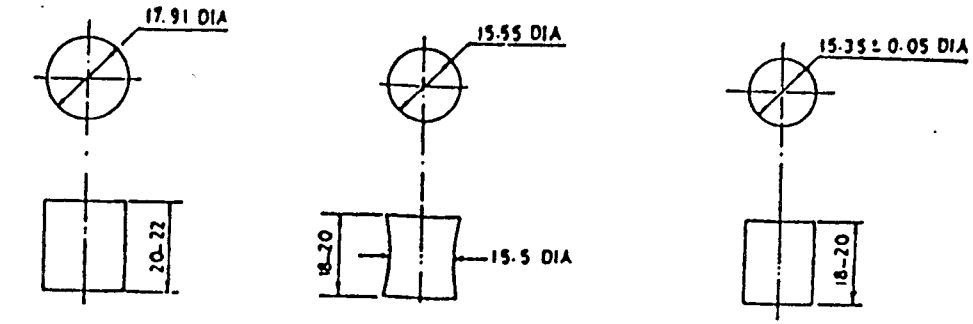


FIG:10. THORIA PELLETS NOTE :-ALL DIMENSIONS IN MM.

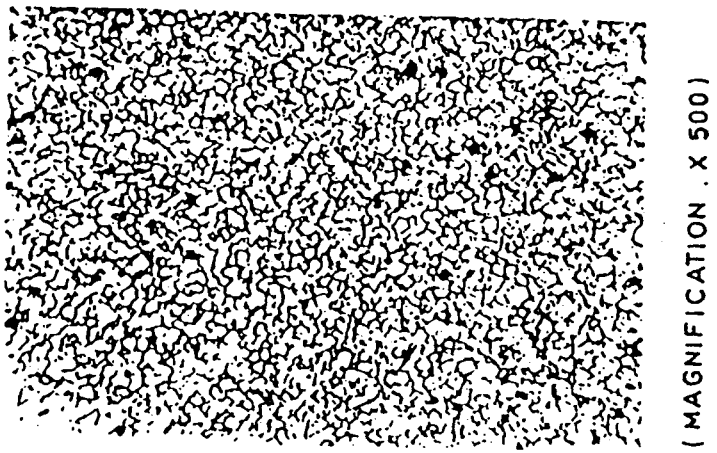


Fig. 11. Microstructure of sintered thoria pellet

The following figures 7 and 8 show a radial blanket, axial blanket fuel elements and the fuel fabrication and assembly processes. (83) (Fig. 9,10, 11).

Now in India also, thoria fuel elements have been manufactured and are being inserted in the heavy water power reactors, to flatten the flux and, at the same time, to breed U-233.

6.3.2. **US Experience** (89)

A thoria fabrication has been running in the US in the 1960s-1970s.

Thoria target fuels, Al-clad, have been irradiated in the Hanford and Savannah River reactors (Figs.13 and 14). Thoria was obtained from purified thorium nitrate, denitrated as "light" thoria, transformed into a sol and dried/sintered at about 1060-1500°C into dense thoria (Fig. 12). Thoria powder was compacted into the aluminium cladding by vibrocompaction. The thoria specifications are found on Table 1.

The flow diagram for the unit operations for the Hanford thoria elements is given on Figure 15. (About the sol-gel techniques, see next Chapter).

At Hanford, the fabrication line (about 100 m^2) was taking place in fume hoods. Where larger quantities were handled (reception, blending), some lead protection (1.2 cm thick) was used. The operating personnel received between 60 to 440 m Rems per individual and per month (average 200 mR/month or 2 mSv/month) for 13 operators per shift. This could not be accepted nowadays and this shows that "aged" thoria, like thorium, produces beta and gamma radiation (Ra-228, Ac-228 etc...) which must be taken care of.

It was found in Hanford that imperfectly sealed elements showed hydration of thoria in the hot water of the reactor (30-80° C), causing sometimes swelling of the fuel elements, due probably to aluminium hydroxide and thoria hydrate. When the powder was sintered at 1200° C instead of 1050° C, it became non reactive.

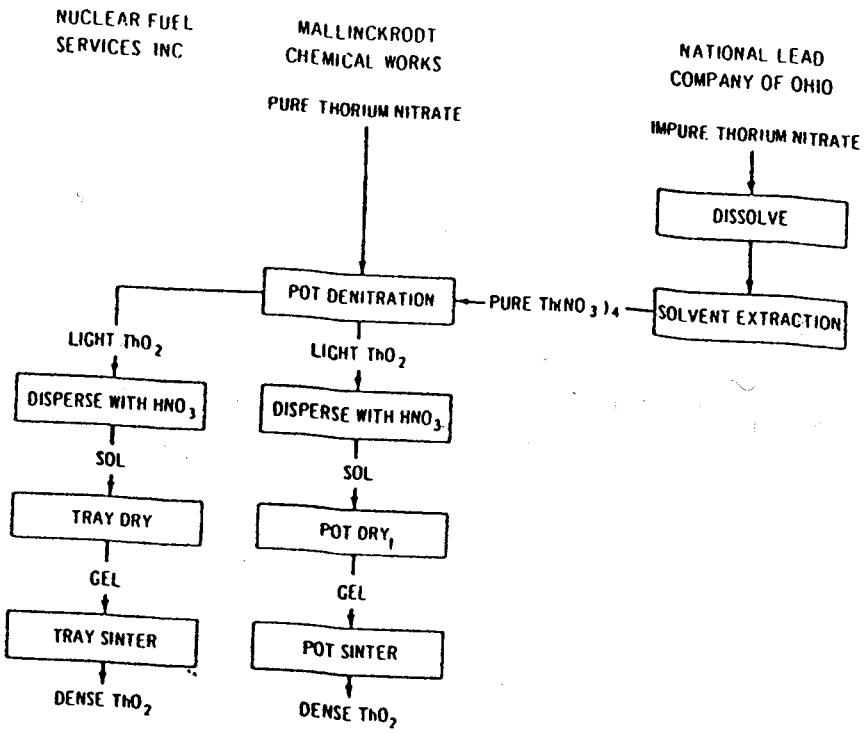


Figure 12: Thoria production flow sheets from different suppliers (89)

Table 1: Thoria specifications

Item	Specification	Basis
Thorium content	87.4% minimum	Control on total impurity levels; theoretical content, 87.9%
Ionium content (^{230}Th : ^{232}Th)	$1:1 \times 10^6$ maximum	Based on ability of the supplier to produce; ^{230}Th, which converts to ^{232}U upon irradiation, should be held to a minimum
Total impurities	2000 ppm maximum	Control of total impurities; analysis performed for Al, B, Be, Bi, C, Cd, Cl, Co, Cr, Cu, Dy, Eu, Fe, K, Li, Mg, Mn, Na, Ni, P, Pb, Si, Sn, Sm, U, Zn
Specific impurities		
Uranium	10 ppm maximum	Undesirable isotopic diluent in ^{233}U product
Boron	TNT* content plus 3 ppm maximum	Neutron absorber, high cross-section impurity
Li, Cd, Sm, Gd, Dy, Eu	TNT content plus 2 ppm maximum	Neutron absorber, high cross-section impurity
Chlorine	100 ppm maximum	Accelerates corrosion in separations equipment
Silicon	300 ppm maximum	Tends to form emulsions during separation by solvent extraction
Loss-on-Ignition (fired at 1050°C for 6 hr)	1000 ppm maximum	Ability of supplier and requirement that material will not off-gas sufficiently to produce enough internal pressure to distort aluminum jacket
Dissolution	At least 95% of thoria sample shall dissolve in 6 hr on refluxing with 12.3 *M* HNO_3 combined with 0.025*M* HF and 0.1 *M* $Al(NO_3)_3$	Required for economical separation of ^{233}U from thoria
Tap density	7.0 g/cm^3 minimum	To attain compacted bulk densities above 70% of theoretical in core of the target element to minimize displacement of fuel in the reactor
Particle-size distribution		
Mesh:		
+6	0	To attain high compacted bulk densities
−6 +20	10 to 60	
−20 +70	15 to 50	
−70 +200	15 to 50	
−200	0 to 50	

*Thorium Nitrate Tetrahydrate.

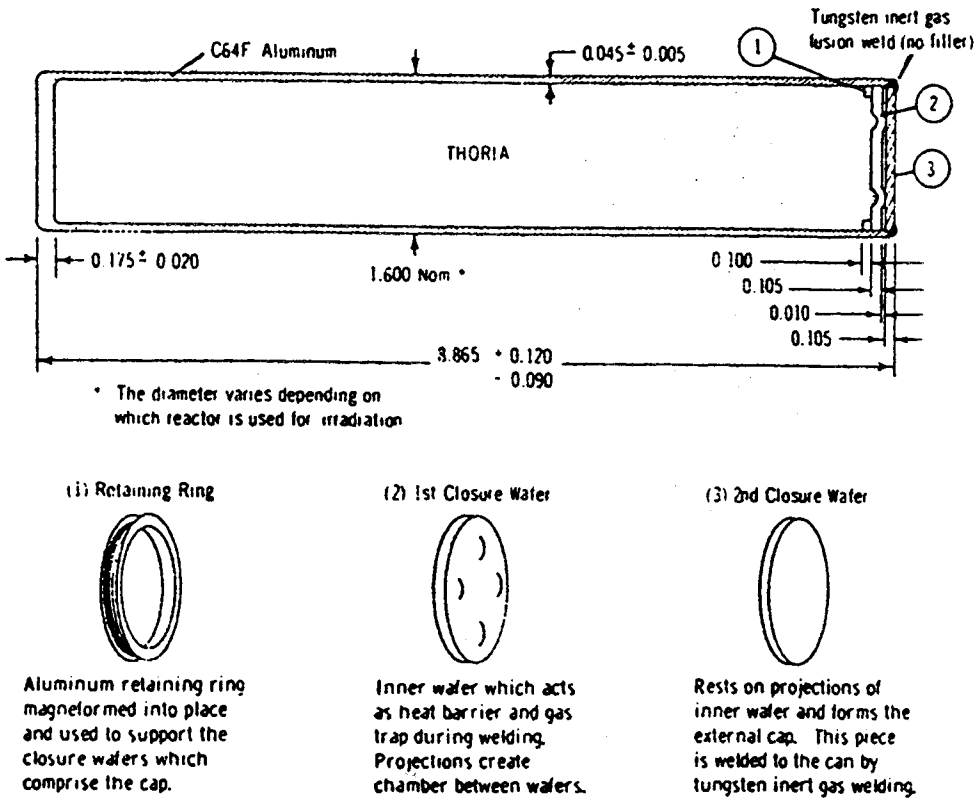


Figure 13: Thoria element used at Hanford

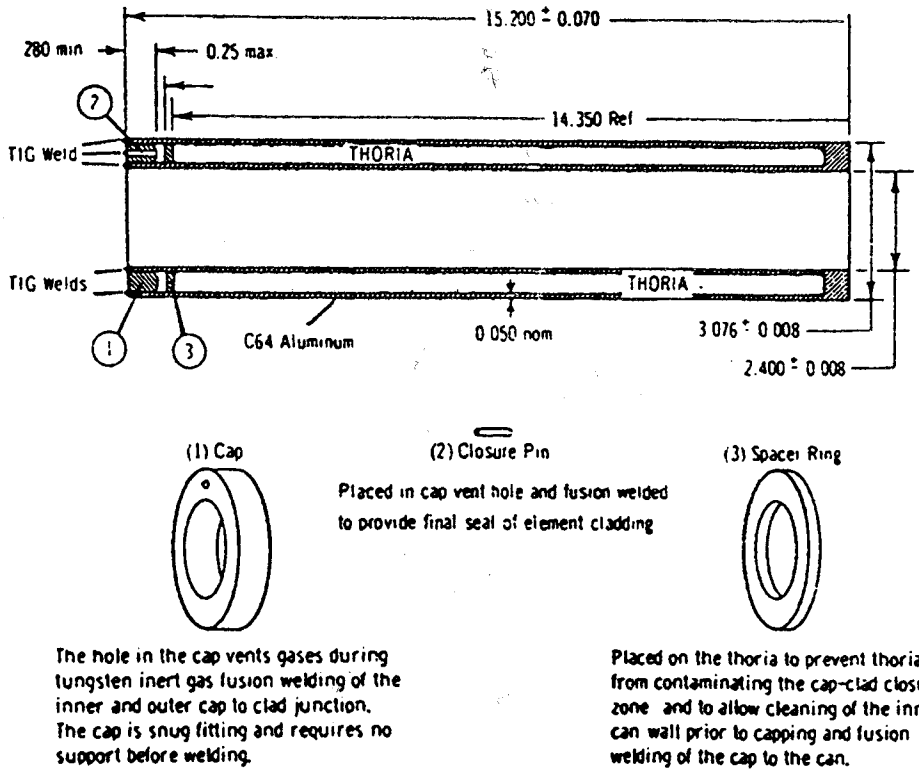


Figure 14: Thoria element used at Savannah River

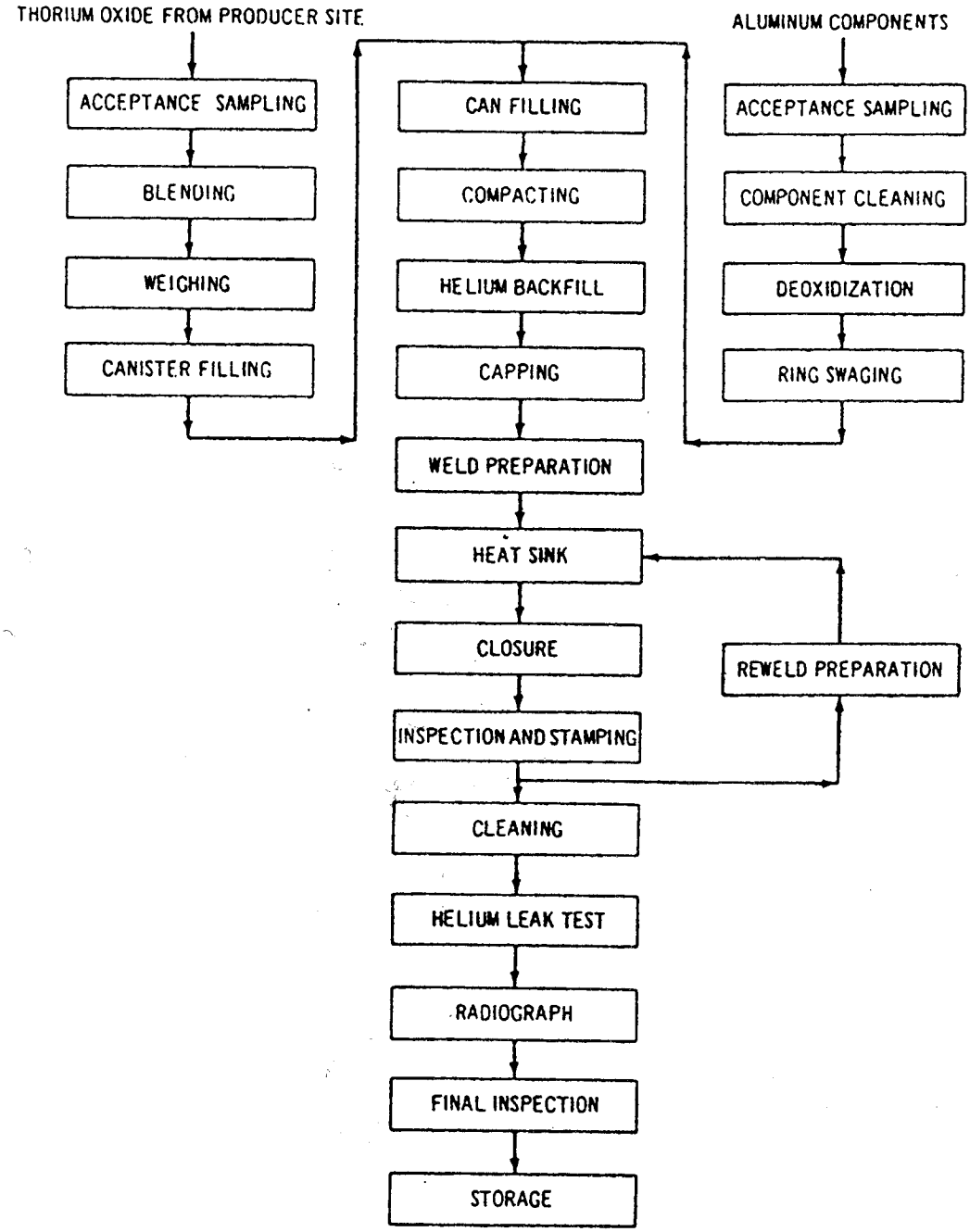


Figure 15: Flow diagram for thoria-element fabrication at Hanford (89)

6.4. Mixed Fuels

Thorium being a purely fertile material, a reactor containing thorium must also contain fertile material. The "driver" or "seed" material can be separated from the fertile fuel elements, but it is also frequently a mixed fuel : UO2-ThO2, PuO2-ThO2; U could be U-235 or U-233. This configuration permits to approach as nearly as possible the homogeneous reactor state, the optimum for neutron conservation and breeding with thermal or epithermal neutrons.

Hence, the fabrication of such mixed fuels.

There are basically two types of fuels :

- The oxides, usually in shape of pellets/rods as sintered microspheres, or coated with pyrocarbon and included in a graphite matrix ("compacts"), or shaped into fuel "balls" as in the case of the pebble-bed reactors.

- The carbides, which can be microspheres aggregated together in graphite (as above).

More exotic fuels have been tested, such as nitrides, other compounds, and molten salts.

6.4.1. Mixed oxides : General Remarks

6.4.1.1. Indian Experience (After C. Ganguly, Radiometallurgy, BARC, India) (81)

UO2, ThO2 and PuO2 are isostructural (CaF2 type FCC), completely solid soluble and have very similar thermodynamic and thermophysical properties. Hence, the fabrication processes of these oxide or mixed oxides are similar.

One of the major problems of Th-232/U-233 fuel cycle is the penetrating gamma radiation of certain daughter products of U-232, which is always associated with U-233. Hence, handling of U-233 bearing materials requires remotisation and automation in shielded glove boxes or hot cells. Likewise, the Pu-239 obtained by reprocessing spent uranium fuel from power reactors would contain significant quantities of Pu-240, Pu-241 and Pu-238 which are associated with neutron and gamma radiations and hence the necessity of glove box and remote automated handling for keeping the personnel exposure to radiation within permissible

limits. The fabrication processes for U-233 and plutonium bearing fuels should, therefore, be amenable to automation and remotisation.

The three fabrication methods that have been explored by the author for (Th,U)O2 and (Th,Pu)O2 fuel pellets are as follows :

a) conventional "powder-pellet" route for both (Th,Pu)O2 and (Th,U)O2,
b) pellet-impregnation technique for (Th,U)O2 only,
c) sol-gel microsphere pelletisation (SGMP) process for (Th,U)O2 and (Th,Ce)O2 (Ce is for simulating Pu).

a) Powder-Pellet Route

The conventional powder-pellet route involves simultaneous mixing-grinding of ThO2 powder with UO2 or PuO2 powders, granulation, cold-pelletisation of granules at ~350 MPa and high temperature (~1973K) sintering of pellets in Argon + 8 % H2 atmosphere. ThO2 and PuO2 powders are produced by air-calcination of the oxalates of thorium and plutonium respectively. These powders are extremely fine ($\leq$ 1 μm) and have poor flowability. Likewise, the UO2 powder obtained via the ammonium diuranate (ADU) route is also fine and not free-flowing. Further, the platelet morphology of oxalate derived ThO2 powder causes problems in achieving homogeneity while mixing with UO2 or PuO2 powders.

The following modifications were made in the conventional "powder-pellet" route in order to prepare ThO2-UO2 and ThO2-PuO2 pellets of high density and good microhomogeneity :

- doping thorium nitrate feed solution with -1 w/o magnesium sulphate or nitrate before precipitation of thorium oxalate in order to get -0.4 w/o MgO as "sintering aid" in the calcined ThO2 powder,

- pre-milling oxalate-derived ThO2 powder in order to break the platelet morphology before co-milling with UO2 or PuO2 for proper homogenisation of oxide powder mixtures,

- admixing -0.25 w/o Nb2O5 powder for improving sinterability of ThO2-UO2 and ThO2-PuO2 powders.

With MgO doped ThO2 powder, it was possible to achieve high pellet density in case of ThO2-UO2 and ThO2-PuO2-4 %PuO2 pellets at relatively low sintering temperature ($\leq$ 1773 K). Nb2O5 was found to have a more pronounced effect on densification of these ThO2 based mixed oxides, achieving nearly 98 % TD at low sintering temperature of 1473 K.

b) Pellet Impregnation Technique

The pellet-impregnation concept essentially consists of preparation of relatively less radioactive ThO2 pellet of low density by conventional "powder-pellet" route in an unshielded area, impregnating these pellets in uranyl nitrate solution (U-233) in a shielded facility followed by high temperature ($\geq$ 1973 K) sintering for densification and ThO2-UO2 solid solution formation. Thus, fine U-233 bearing powders are avoided, handling of U-233 is restricted only in certain parts of the fuel fabrication plants and process steps like calcination of uranyl nitrate and mixing-grinding of ThO2-UO2 powders are eliminated.

ThO2-UO2 pellets of high density ($\geq$ 96 % TD) and homogeneous uranium distribution could be reproducibly fabricated by this technique. However, the limitation of the pellet-impregnation technique is that a maximum of ~2 % U-233 could be introduced in the ThO2 pellets.

Note : this technique has been tested in Canada also.

c) Sol-Gel Microsphere Pelletisation Process

The SGMP process for fabrication of high density ThO2-2 % UO2 and ThO2-4 % CeO2 pellets was developed in collaboration with the Institute of Chemical Technology (ICT), KFA, Jülich, FRG. (See further, § 6.4.3). The standard external gelation of thorium (EGT) process of KFA, for preparation of spherical fuel particles for high temperature gas-cooled reactor was significantly modified to tailor-make gel microspheres of thorium-uranium oxide and thorium-cerium oxide, which after controlled calcination were found suitable for direct pelletisation and sintering to high density. The process flowsheet developed (Fig.1) incorporated the following three modifications in the EGT route :

- use of heavy metal nitrate feed solution of lower molarity (M $\leq$ 1.2),

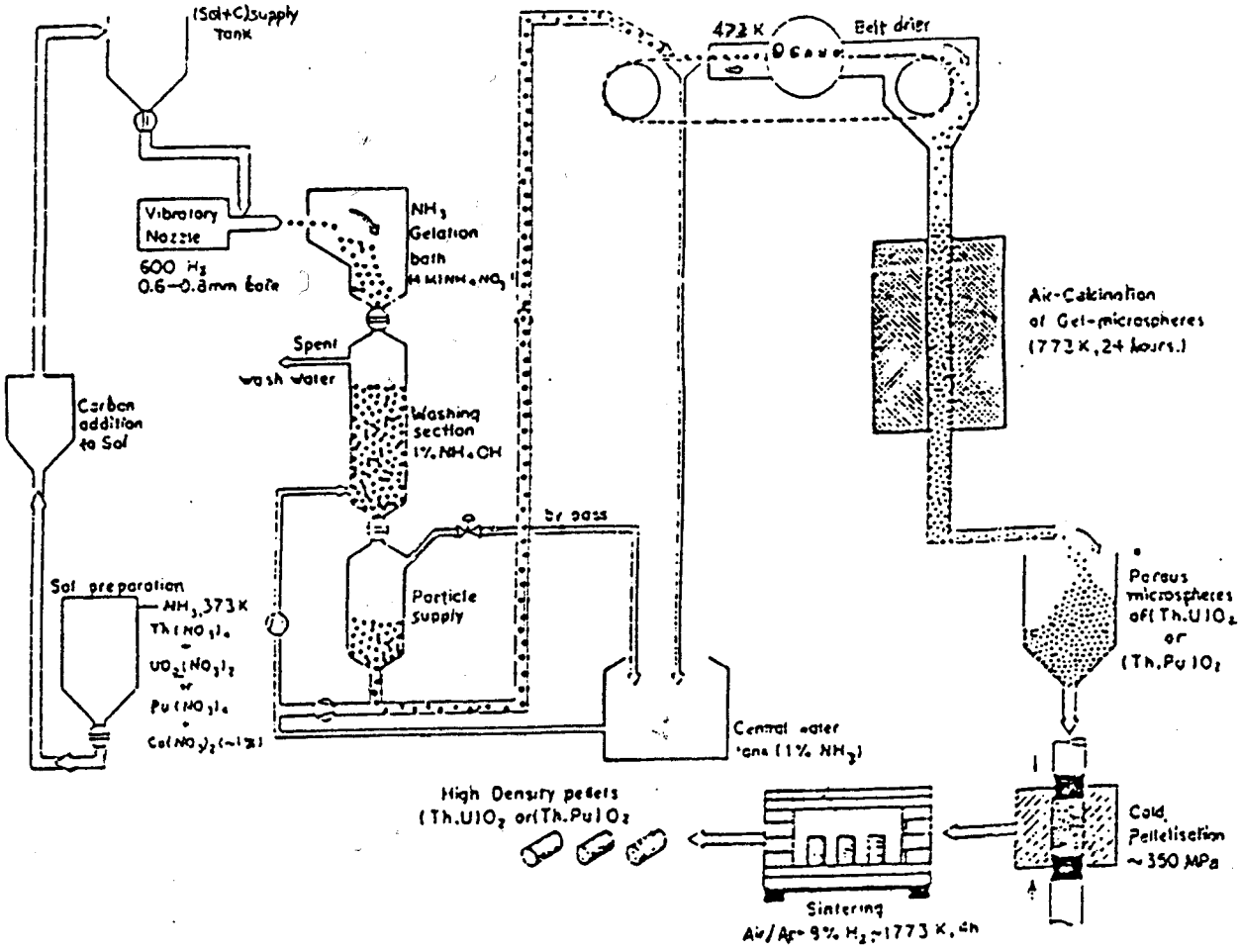


Figure 1: Sol-gel microsphere pelletisation of ThO_2-based mixed oxide fuel (81)

- addition of 1 w/o calcium nitrate in feed solution in order to have 0.4 % CaO as "sintering aid" in calcined microspheres,

- addition of carbon black "pore former" to the sol prior to gelation in order to produce "porous" microspheres upon air-calcination.

Microsphere boundaries were found to be totally absent in case of sintered pellets prepared from "porous" microspheres. The etched microstructure and SEM picture of fractured surface of sintered pellets prepared from "porous" microspheres showed equiaxed grains in the size range of 15-20 μm and spherical pores in the pellets were in the optimum diameter range of 2-5 μm.

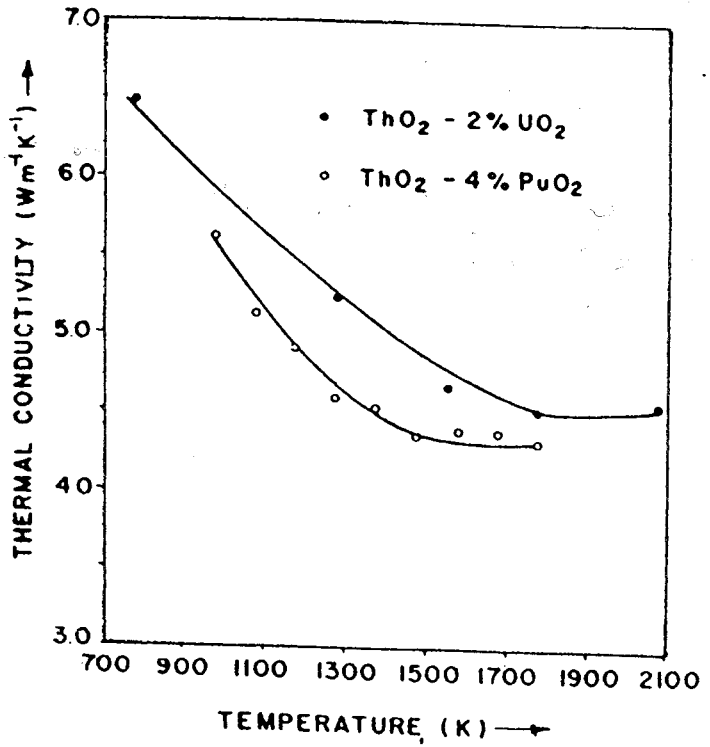
The thermal conductivity and hardness properties of typical ThO2-UO2 and ThO2-PuO2 sintered pellets thus obtained are given in Figure 2 as a function of temperature.

Conclusion

The sol-gel microsphere pelletisation process minimises the radiotoxic dust hazard as it uses dust-free and freeflowing microspheres as "press-feed" material. It ensures excellent microhomogeneity, microstructure and high density, and is amenable for automated and remote fabrication of highly radiotoxic ThO2-PuO2 and ThO2-UO2 fuel pellets. "Porous" microspheres should be used as starting material. These are prepared by adding carbon black pore former to the sol prior to gelation and later removing the same by controlled air calcination of gel microspheres at around 973 K for 24 hours.

Zircalloy clad ThO2-4 % PuO2 fuel pins could be successfully irradiated to 18 000 MWD/T without any pin failure in pressurised water loop of CIRUS reactor, simulating the operating conditions of an Indian pressurised heavy water reactor.

Other data on irradiated ThO2-PuO2 fuels are given by M.D. Freshley and H.M. Mattys, Battelle PNL (96), among many others.



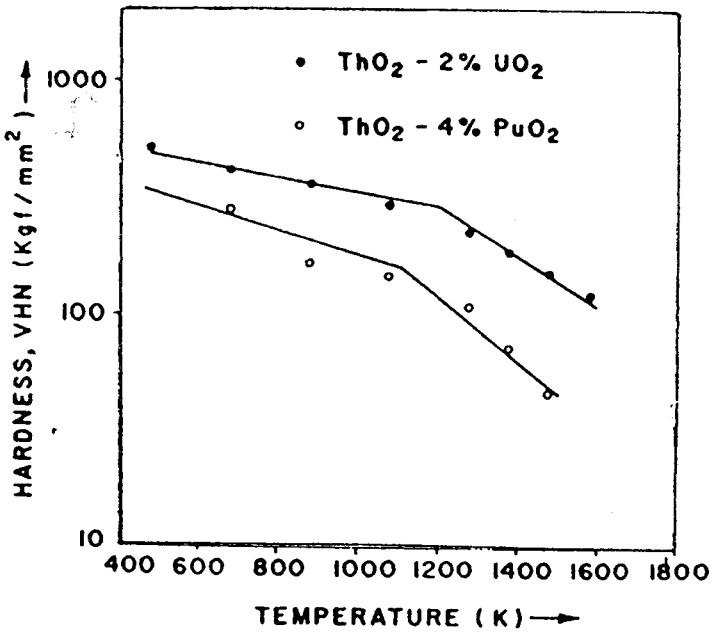


Figure 2: Thermal conductivity and hardness of ThO_2 - 2%OU_2 and ThO_2 - 4%PuO_2 sintered pellets as a function of temperature (85)

6.4.1.2. Remarks on the sol-gel techniques

The obtention of the desired microspheres by the sol-gelation is the delicate part of the process, where many parameters interplay; it is an "art" where much empirical research has been devoted. It seems, indeed, as is claimed by many, that some of the processes do work and are reproducible.

a) We quote the comments from a **Canadian team** here (1985) (86) :

"Numerous sol-gel processes for the production of ceramic nuclear fuel have been developed over the past 25 years. The sols have been prepared by the forced base hydrolysis of solutions of the heavy metal salts using aqueous ammonium hydroxide or an amine dissolved in an organic phase. An alternative approach, applied only to ThO2, is to prepare the oxide from a solution of the nitrate salt by hydrothermal or thermal denitration and disperse the resulting powder to a sol. The sols prepared as above can be gelled by contacting them with a hydrating organic solvent or by raising the pH through : contacting the aqueous sol with an organic solvent containing ammonia or an amine dissolving an ammonia donor in the sol which releases ammonia when heated, or by contacting the sol with aqueous ammonium hydroxide.

"When the final product is to be in the shape of a microsphere, gelation is preceded by the formation of spherical drops of the sol. If the gelation medium is an organic solvent, the drops can be formed within the gelation bath where the spherical shape of the drop is maintained by the aqueous-organic interfacial tension. When ammonium hydroxide is used for gelation, the drops must be formed in air and passed through an ammonia gas blanket to form a gelled skin on the exterior of the drops to prevent them from deforming upon impact with the gelation bath. Despite the mechanical integrity provided by the gelled skin, some workers still add an organic thickener to increase the viscosity of the sol and produce larger microspheres.

"The viscosity of the sol can also be increased by taking it close to the point of gelation. Microspheres of about 500 μm in size have been produced by the latter technique which also incorporates horizontal jetting, while microspheres of about 1200 μm have been produced with the use of organic thickeners."

AECL has developed a process to prepare good quality ThO2 microspheres made by a sol-gel process where neither thickeners nor high levels of sol neutralization are employed. (Fig.3).

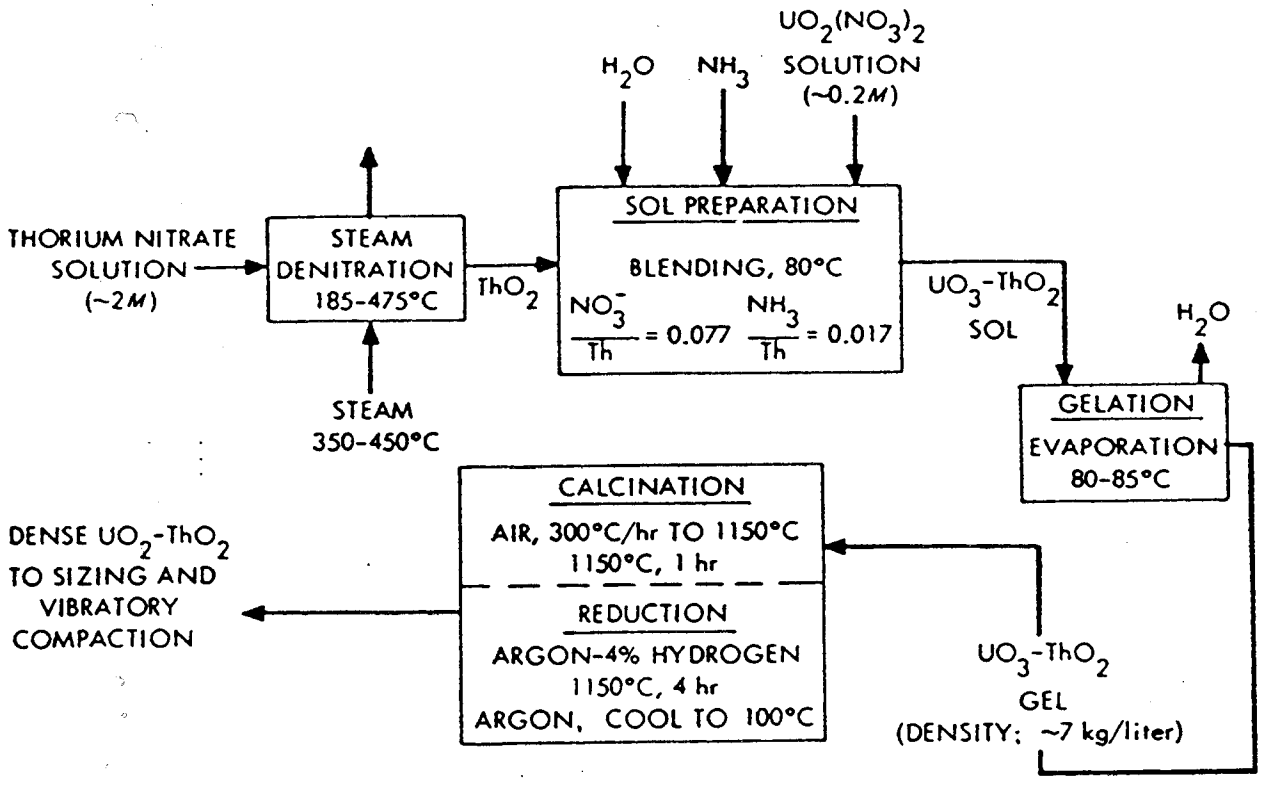


Figure 3: *Original sol-gel process flow sheet AECL (86)*

It is likely that a similar process could be used in the case of mixed fuels too.

b) Much research has been done, in particular in the USA, in the 1960s, on the sol-gel preparation of ThO2-UO2 and ThO2-PuO2 fuels. The processes were piloted at ORNL (95), and applied semi-industrially by private companies (B & W) (94) Mallinckrodt.

The ORNL process calls for a solid solution of ThO2 obtained by denitration of Th nitrate, in uranyl nitrate, sol formation with ammonia addition and gel formation by evaporation (Fig.4).

The density of PuO2-ThO2 products after calcination at 1150° C varied from 95 % to 99 % TD (Fig.5).

c) Many other laboratories have tested the sol-gel process; among them **KFA-Jülich and NUKEM in Germany** merit a special mention for their intensive research and accomplishments. Remarks about these processes are to be found further in § 6.4.3.

6.4.2. Industrial prototype fabrication plants for mixed oxide fuels containing thorium

a) General Remarks

Among the more representative fabrication lines we should mention the semi-shielded KILOROD facility at ORNL, and the development of a remote thorium-based fuel fabrication plant by the ORNL team, named TURF. (90, 93).

Based in part on R&D pioneered by ORNL and other laboratories, private companies, especially Du Pont at Savannah River and Battelle PNL at Hanford, Babcock and Wilcox, etc..., under USAEC contracts, have installed pilot-to-industrial size facilities to produce the different types of fuel used in the prototype reactors, BWR, PWR, HTR (See Chapter 3 above). Fuel-based product could be obtained from National Lead, Mallinckrodt, Nuclear Fuel Services, among others (89).

Complete fuel element fabrication on 10 kg scale was available at Babcock and Wilcox (92, 99), and we remind that Allis- Chalmers had built a remotized fuel refabrication pilot facility at Rotondella, Italy, for ELK River BWR fuel (153).

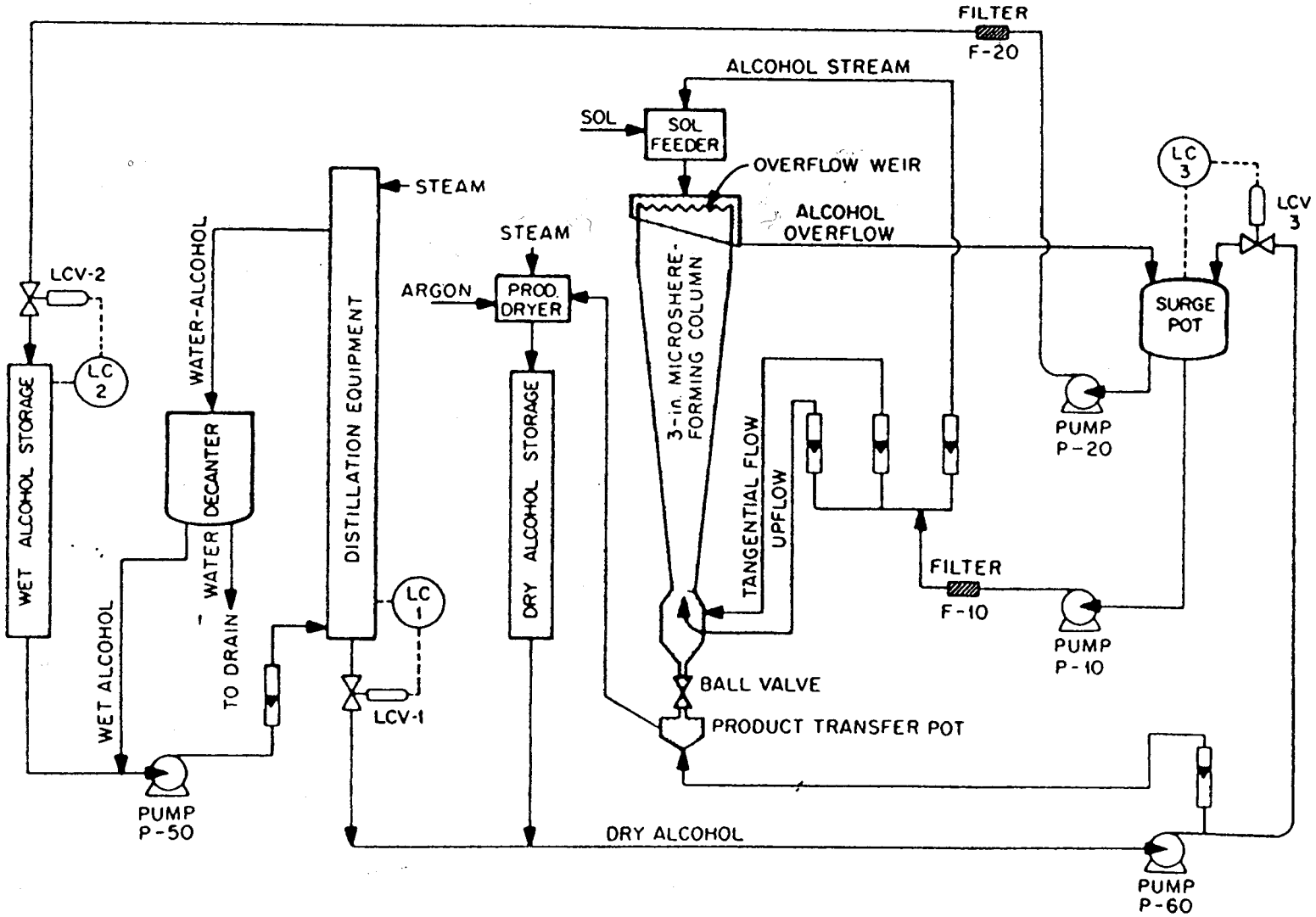


Figure 4: Sol-gel microsphere process flow diagram for the coaled Particle Development Laboratory. [-LCV = level control valve; LC = level controller] - (ORNL, 95)

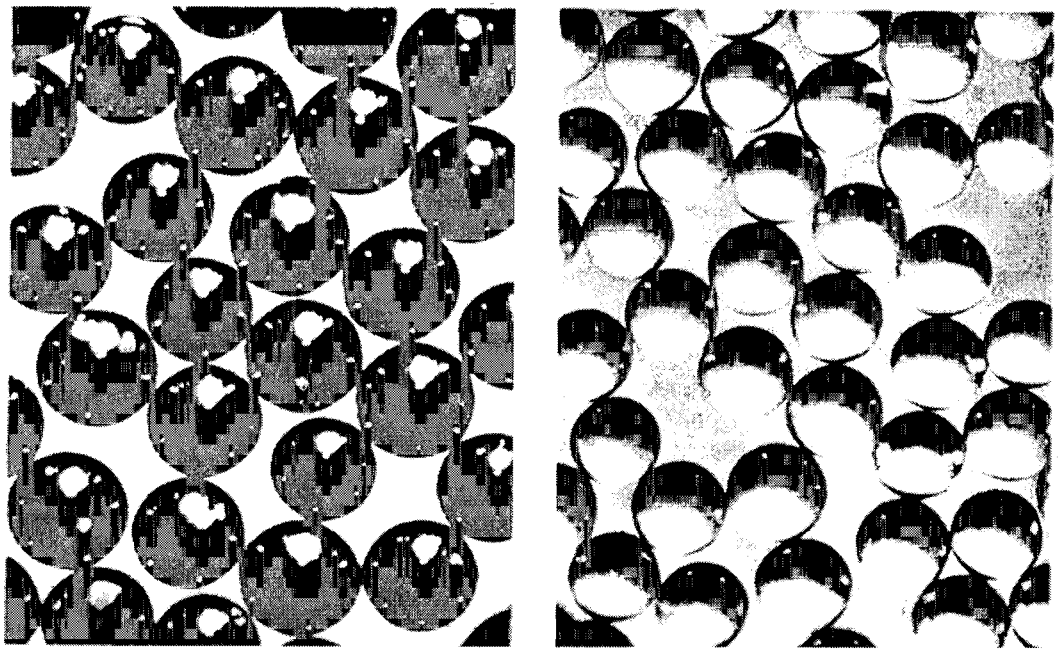


Figure 5: Microspheres of 75% urania - 25% plutonia before and after calcining (ORNL, 95)

It was soon found out that under certain conditions the sol-gel process was the optimum process to avoid fine, sticking powders, a drawback with irradiating material clinging to the equipment, and that the process had to be as streamlined and as simple as possible to take advantage of a time window of a few days to a few weeks after purification by solvent extraction or ion exchange of the daughters of U-232 and Th-228 and Th-234 (from freshly recycled thorium), during which the gamma radioactivity was minimum.

The process conditions were validated by irradiation and inspection of the fuel.

As a conclusion, it can be said that the know-how exists to manufacture reliable UO2-ThO2 and PuO2-ThO2 fuels, semi remotely or fully remotely.

b) **The KILOROD fuel fabrication at Oak Ridge** was operated since 1964 (90) on a 10 kg/day scale. The process scheme, using a sol-gel process and vibratory compaction of the powders into the pin cladding, is given in Figure 6.

A view of the installation (in a former reprocessing cell) is given Figure 7.

The amounts processed in a 8-month campaign are given in Table 1.

From the experience gained, ORNL had prepared the design parameters for U-233/Th fuel fabricating plants having capacities from 60 to 3700 (!) kg/day of heavy metal. Such data might be useful some day. However, the simple vibratory compaction process of the mixed oxide powders would not probably be sufficient for the high burn-up fuels considered today.

Radiation Safety Problems

The main problem comes from the presence of U-232 daughters which deliver irradiation doses to the operators. We shall insist somewhat on this crucial point.

We quote (90) :

"In this program U-233 solution (containing 38 ppm U-232), which was purified by solvent extraction and which was approximately 5 days old, was furnished to the Kilorod facility at about 40-day intervals. In each of these intervals (called campaigns), the postpurification time in sol-gel processing varied from about 5 to 25 days and in rod fabrication from about 9 to 28 days. In each portion of the

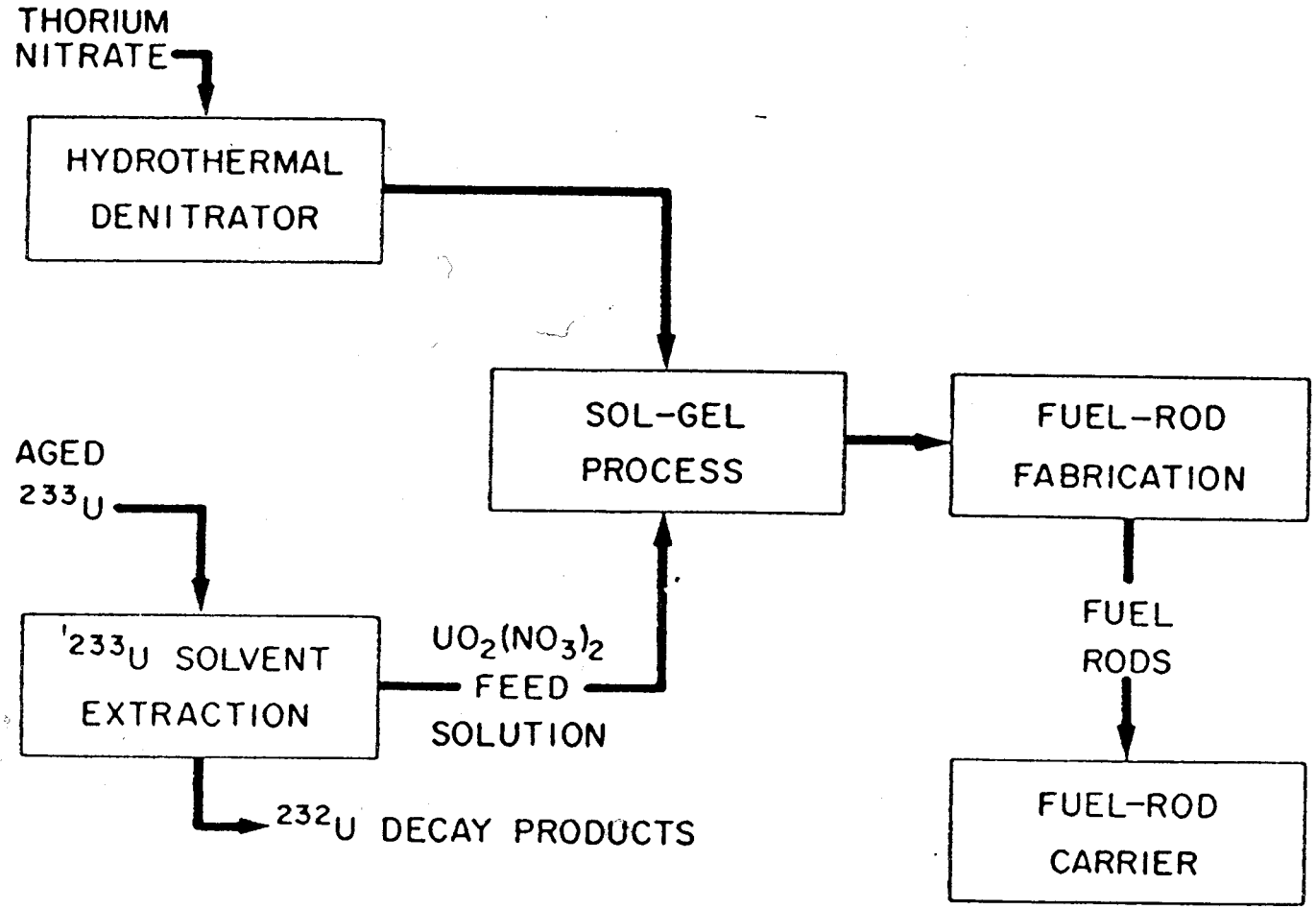


Figure 6: KILOROD facility flow sheet

Table 1: Summary of KILOROD program results

Operational phase	Material processed, kg	Material utilization, %	Losses or recycle, %	On-stream time, %
ThO_2 preparation	1277	99.0	1.0	100
^{233}U purification	50	100.0	0.1	100
Sol–gel process	994	99.8	0.1	90
Rod fabrication	980	94.3	0.3 (7.6)*	80

*Recycle, including start-up phase.

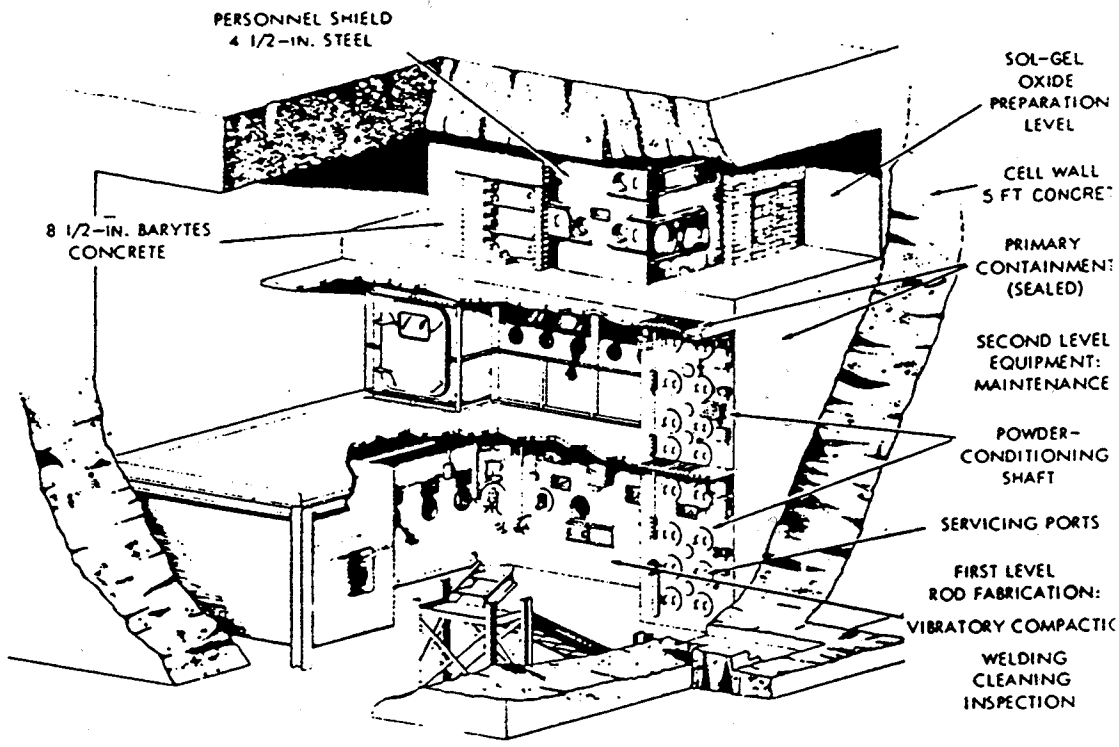


Figure 7: KILOROD facility

line, there was an incremental time after processing of about 7 days for postcampaign cleanout of equipment. In the Kilorod facility there was also some lag time between campaigns for data analysis. Because of fixed contamination of equipment, it was not possible to clean out thoroughly after each campaign; a more extensive cleanout and decontamination was necessary after the seventh campaign.

"Approximately 3 to 4 years had passed since the last purification of the U-233 used in the Kilorod program. Consequently the daughters of U-232 were nearly in equilibrium. The solvent-extraction process removed the Th-228, Ra-224 and Pb-212 daughters by factors of about 2500, 5000, and 100, respectively. Because of the relatively low efficiency for removal of Pb-212, presumably caused by extraction of its Rn-220 parent, the hard gamma activity in the product decreased for approximately the first 2.5 days and then began to increase, approaching the activity from initially pure material after 5 to 10 days. (Figures 8-A, B, C, D).

"The average radiation exposures in the Kilorod program were 19.5 and 19.1 mrems per man per week (0.19 mSv) for the sol-gel process and the rod fabrication, respectively. Hand exposures were 62 and 113 mrems per man per week for the sol-gel process and the rod fabrication. These operator doses are significantly higher than those experienced by the supervisors, who may be thought of as "controls" for the program. The dose incurred in process operations from material actually being processed was no more than 20 % of the total. The dose during processing operations from material held up in the equipment was 36 % of the total. The dose incurred during postcampaign cleanout operations was 34 % of the total. The dose from in-cubicle maintenance operations averaged 10 % of the total.

"The radiation level from holdup of material in the Kilorod process was estimated on the basis of the radiation dose rates to film packs placed in strategic areas inside the Kilorod cubicles. In the sol-gel process, the radiation background from material holdup was relatively constant in campaigns 4 through 7. These data can be explained by assuming a steady-state mass of about 2 kg of oxide and exponential removal of old material with a rate constant of about 0.15 per campaign.

"The background levels in the rod-fabrication equipment continued to increase from campaign to campaign. The data are best explained by assuming that about 1.5 kg of oxide of the nearly 100 processed in the first campaign was irretrievably lodged in the equipment.

"Separate records were maintained of the radiation dose rates during post-campaign cleanout operations. The dose from these operations, accumulated over seven campaigns, represented about 34 % of the total dose. Approximately 60 % of the total dose in the seventh campaign was incurred in the postcampaign cleanout period, during which "complete" decontamination was attempted.

"Doses were also accumulated for maintenance operations performed inside the cubicles. These doses were 20, 10, and 2 % of the quarterly exposures during the first, second, and third yearly quarters of operation, respectively. These exposures do not reflect the actual weight of maintenance operations. The predominant effect of maintenance is delay of the process, which adds the requirement that materials be processed at longer postpurification times. A similar effect occurs from other delays in the process, caused by holding materials for analysis, holding off-specification materials for blending, etc...".

From this experience, predicted dose rates in a stream-lined fabrication plant of 10 kg/day capacity with eight operators, has been derived (Table 2), based on the radiation buildup after purification (Fig. 8-A, B, C, D).

Considering the accepted practical dose rates at the time (1968), which were of 2-5 Rem per year per operator (20-50 mSv), it was concluded that "direct fabrication is feasible with 3 % UO2-233/97 % ThO2 fuels that contain less than 20 ppm U-232. With shadow shielding, this limit can be increased to 200 ppm. With shadow shielding and frequent cleaning and recycling, this limit is 600 ppm. Plants with larger capacities will require shielding when operating under sustained conditions".

c) Similar results were obtained at a **pilot fabrication plant of about 10 kg HM/batch operated at Babcock-Wilcox** on the same ORNL process, from 1965. (91, 92, 99) . Its cost was 1 million dollars in 1965. It consisted of a sol-gel preparation plant and a fuel fabrication plant (Figs. 9 and 10). Both plants, like Kilorod, were partly shielded only.

The total production of the plant in 1968 is shown on Table 3.

Table 2. PREDICTED DOSE RATE IN GLOVE-BOX FACILITIES FOR FABRICATING (U233)O2/ThO2 FUELS AT 10Kg/DAY (EIGHT OPERATORS AND 5 DAYS PER WEEK) (90).

Concept	Shield factor	^{233}U, %	^{232}U, ppm	Post purification time, days				Time between solvent extraction, days	No. of campaigns	Dose from fresh material,† %	Weekly dose, mrems
				Sol–gel		Rod fabrication					
				Begin	End*	Begin	End*				
Batch	1	3	20	3	12	7	16	11	5	40‡	37
Continuous	1	3	200	≥1	≤3	4	4.8	1	10	68§	40
SBR line° (Sol–gel)	0.1	25	250	3	12	7	16	11	5	29‡	600
HTGR line (microspheres)	0.1	25	250	3	12	7	16	11	5	56§	250

*Assumes 20% of processing time for cleanout.
†Dose contribution from maintenance is 10% in shielded line, 1% in unshielded line.
‡Assumes holdup equivalent to that of the Kilorod Facility.
§Assumes holdup equivalent to one-fourth that of the Kilorod Facility.
°SBR: Seed-Blanket Reactor

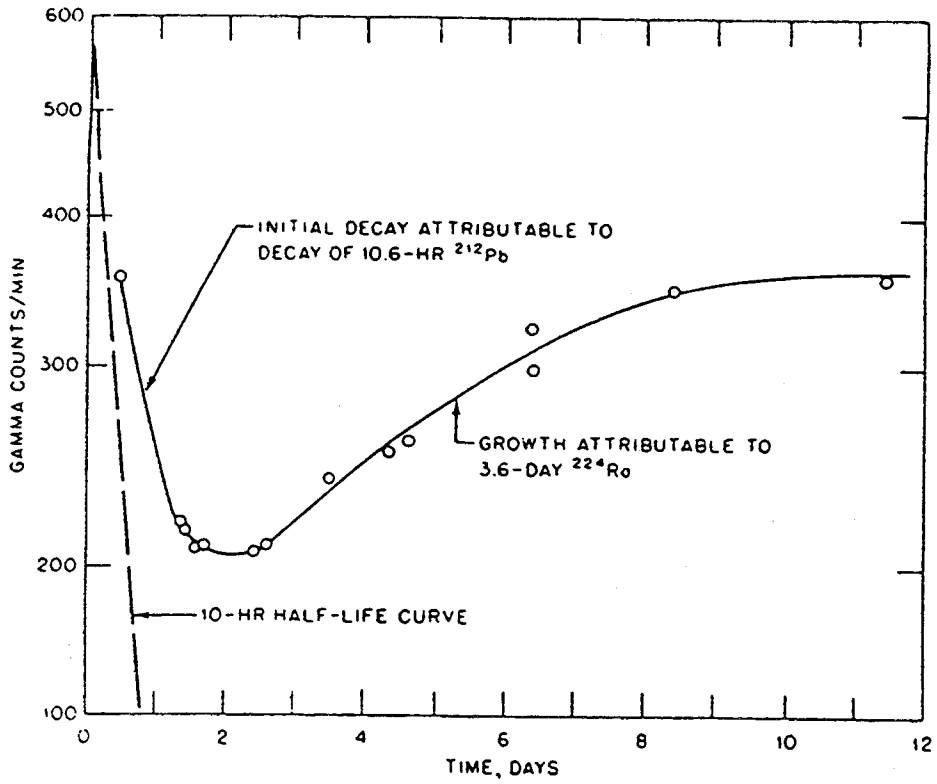


Figure 8A: Gamma activity of a ThO_2 gel containing ^{233}U (93)

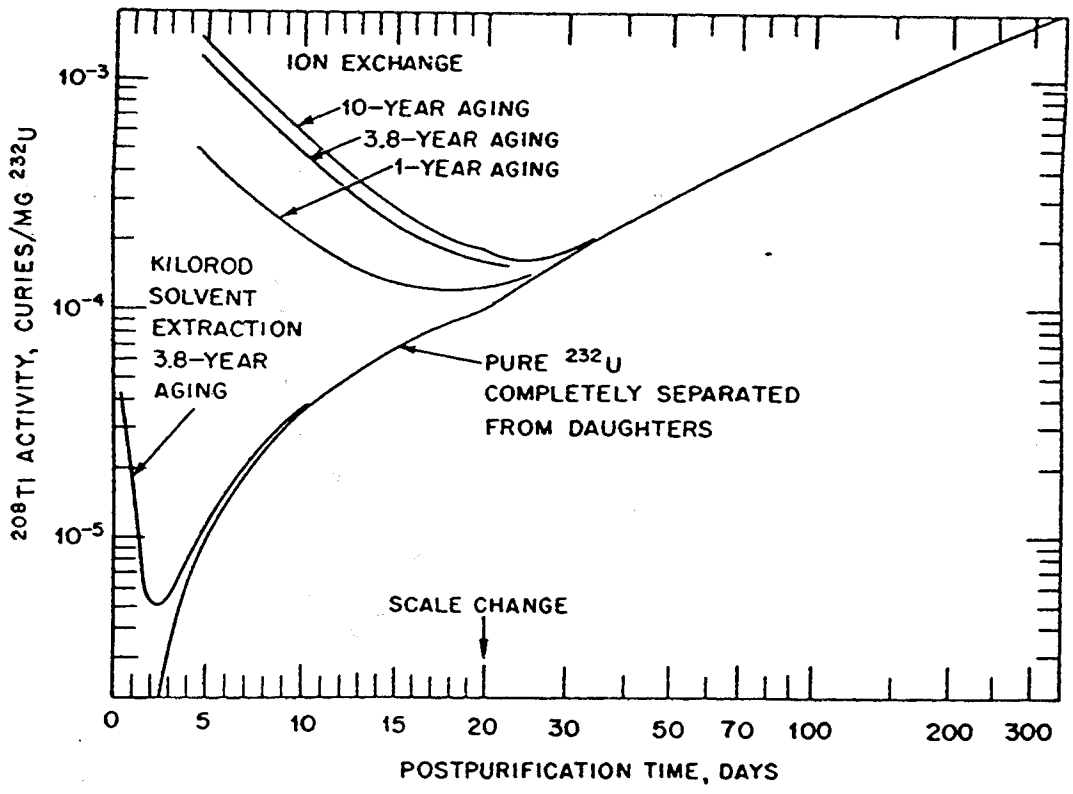


Figure 8B: The ^{208}Tl activity from ^{232}U decay as a function of post-purifications time, method of purification, and aging time before purification (90)

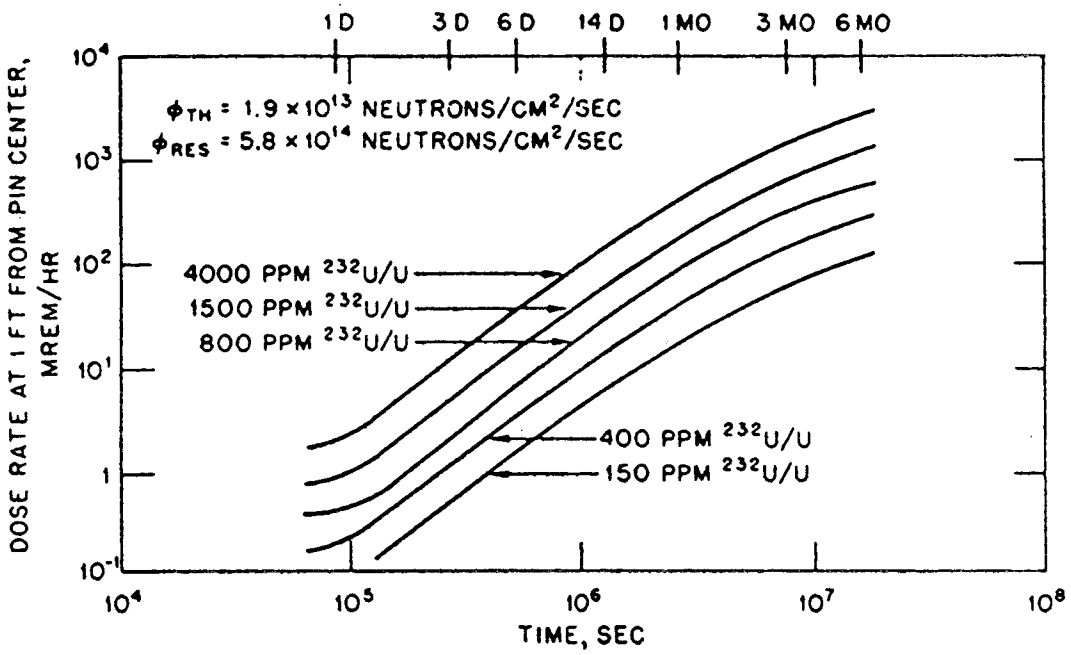


Figure 8C: Estimated gamma activity vs. time from a single fuel pin with various levels of ^{232}U contamination (93)

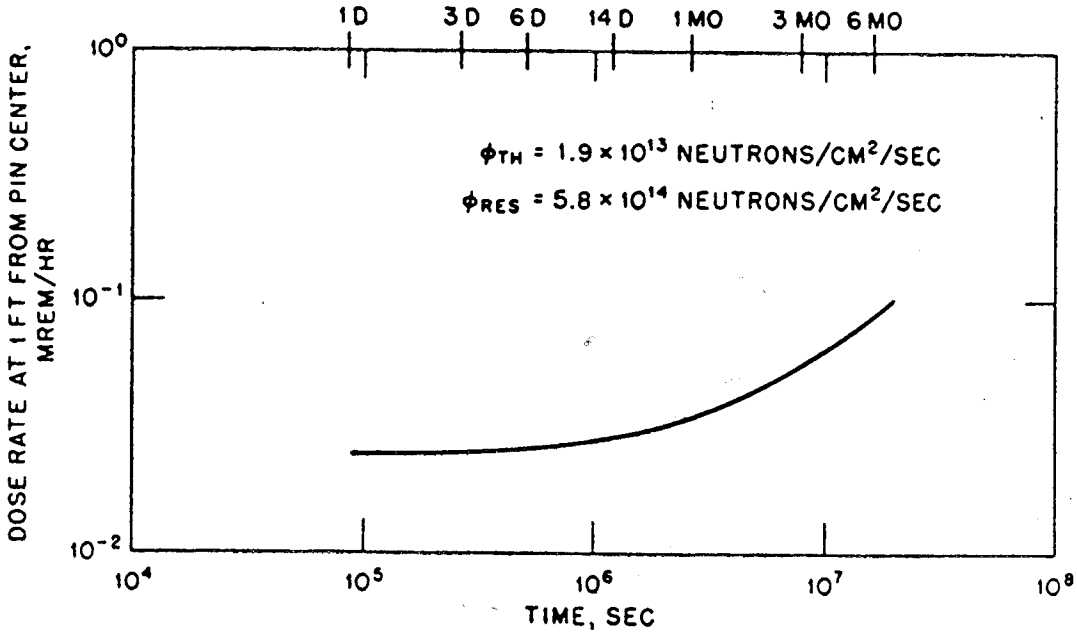
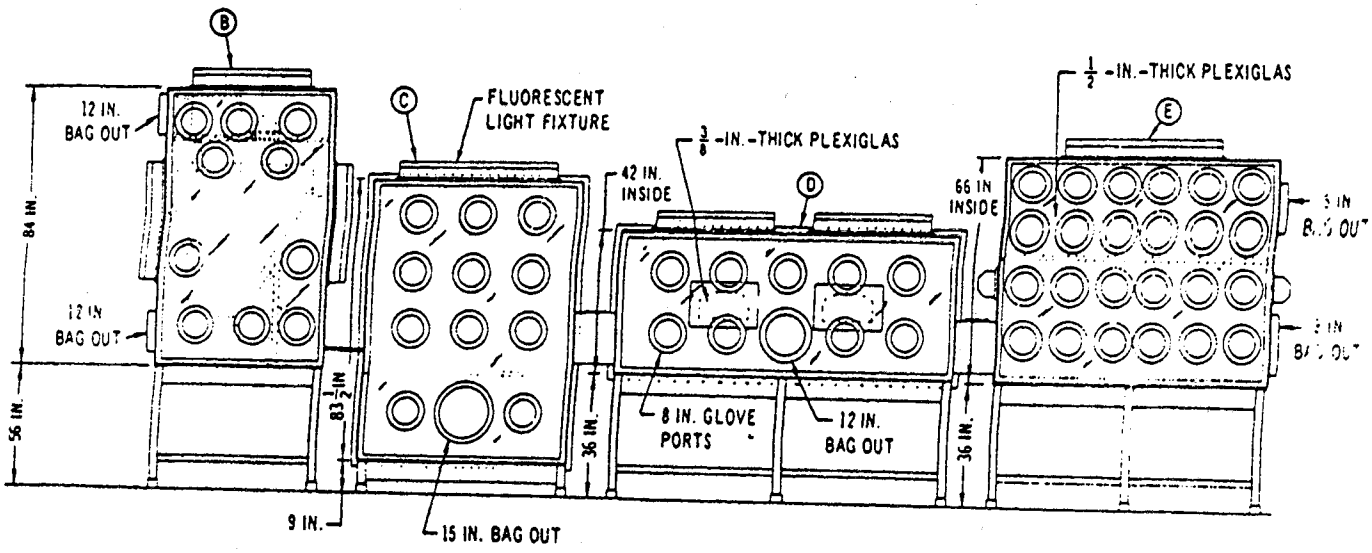


Figure 8D: Estimated neutron activity vs. time from a single fuel pin at equilibrium cycle (1.6 x 10^{-4} ^{232}U/^{232} Th) (93)

Table 3: Summary of the materials prepared in the sol-gel pilot line (92).

Material produced	No. of runs	Total production, kg
Sol–gel thoria	7	62
Sol–gel thoria–urania	45	473
Steam-denitrated thoria	61	632



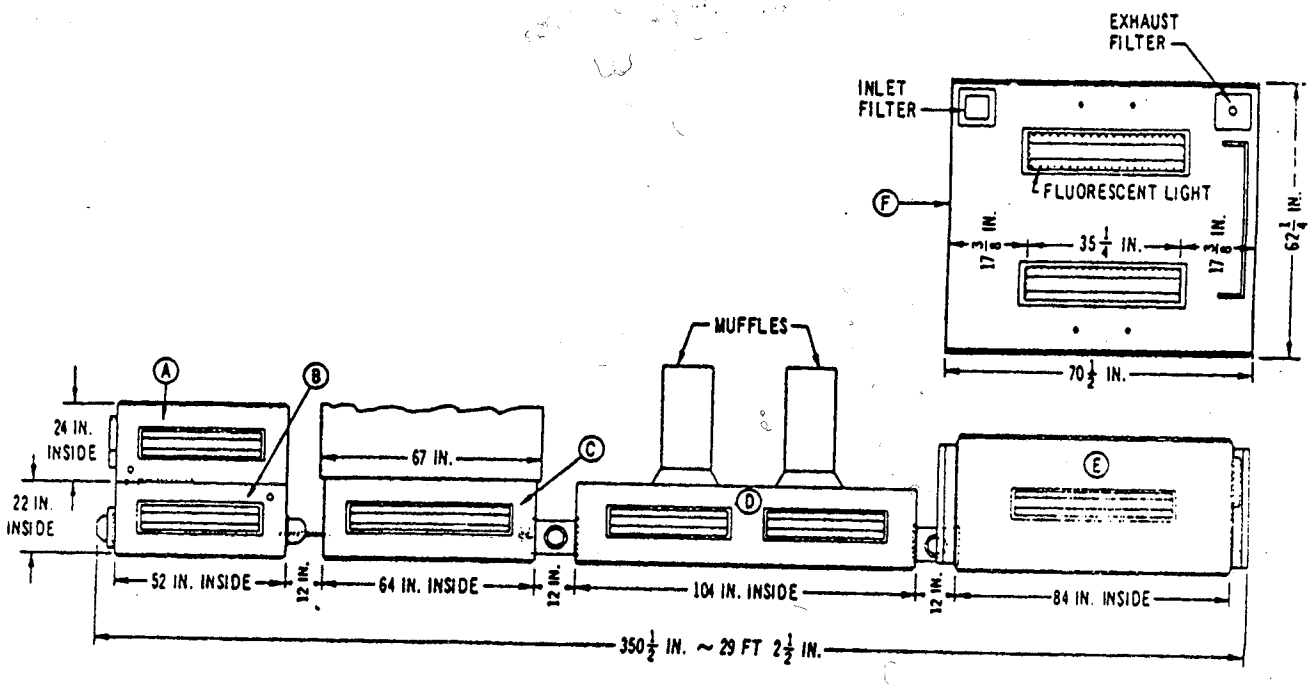


Figure 9: Fuel preparation glove-box line, plan view and front elevation. A, measuring box, B, blend tank box, C, evaporator box, D, furnace box, E, powder preparation box, F, denitrator box (92)

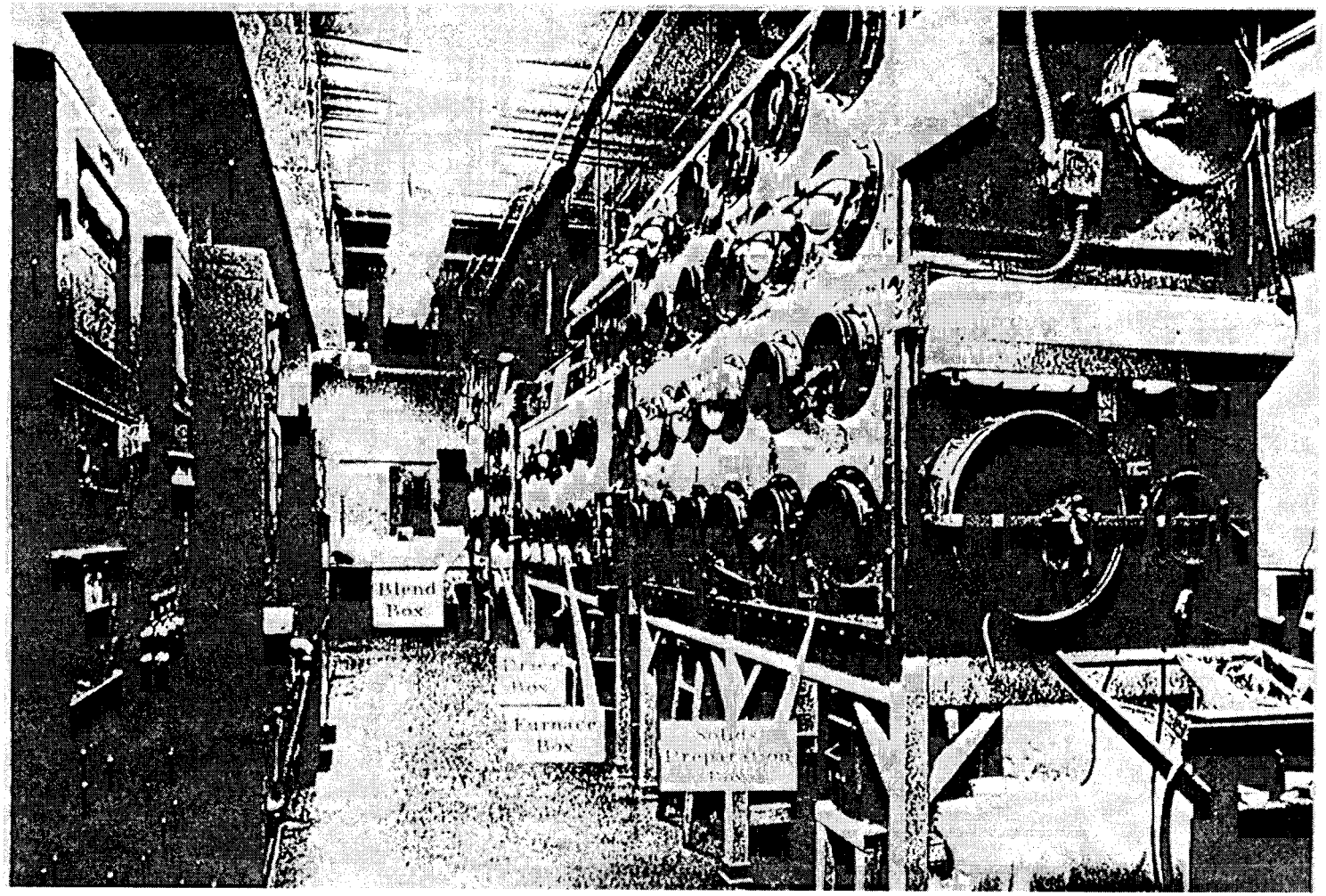


Figure 10: Glove-box line (Babcock & Wilcox, 91)

c.1) The dose rates **in the sol-gel plant** after some runs are given on the figures 11, 12, 13. U-233 had been purified/"deactivated" by ion exchange at Babcock-Wilcox for the first run. U-233 was 48-70 days old from the second to the seventh process run, coming from ORNL. A typical run time was 2 days.

Although it is not said, we infer that the U-232 content in U-233 was similar to the ORNL runs (around 40 ppm). Babcock-Wilcox concluded that, "with freshly deactivated U-233, the plant could process up to 1000 ppm U-232 product." But at that time doses rates of 300 mR/week were considered acceptable.

It is clear that today, when practical accepted dose rates are of the order of 100-500 mR/year/operator (1-5 mSv), remote fabrication is mandatory in all practical cases.

c.2) After preparing the ThO2/UO2-233 reactor fuel, **Babcok and Wilcox** had a complete line of **fabrication of the ThO2/UO2-233 fuel elements** in an unshielded glove-box line (92, 99).

Figure 14 gives a schematic diagram of the operations and Figures 15 and 16 give a view of the overall rod fabrication line.

The zircalloy fuel pins are filled with the fuel powder and vibrocompacted to about 87 % TD. Once filled, plugged and leak tested, the rods are assembled underwater into a fuel element. (Figs. 17, 18).

Up to 1968, 119 rods (232 kg of ThO2 /3 % UO2-233) were fabricated. The U-233 contained 42 ppm U-232. The fabrication of 37 fuel rods contributed to 160 mR to the operator (500 mR on the hands). It was felt at the time that "with some streamlining and freshly decontaminated U-233 a direct method of fuel fabrication with higher U-232 content could be quite feasible."

d) A similar plant but with more remotized fabrication was built by **Allis Chalmers at Rotondella in Italy** to manufacture Elk River fuel elements. This plant has been dismantled now. (153)

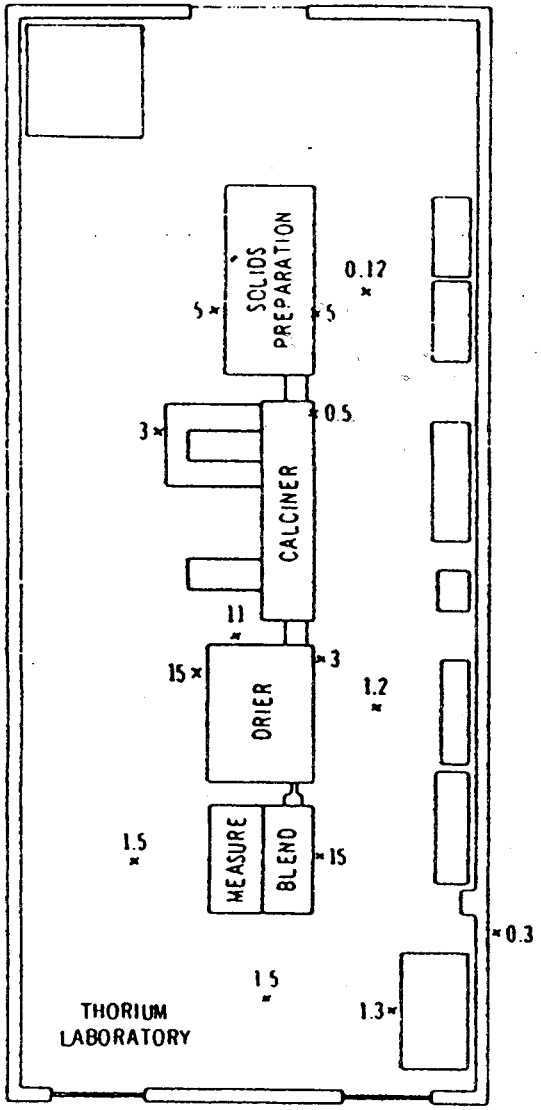


Figure 11

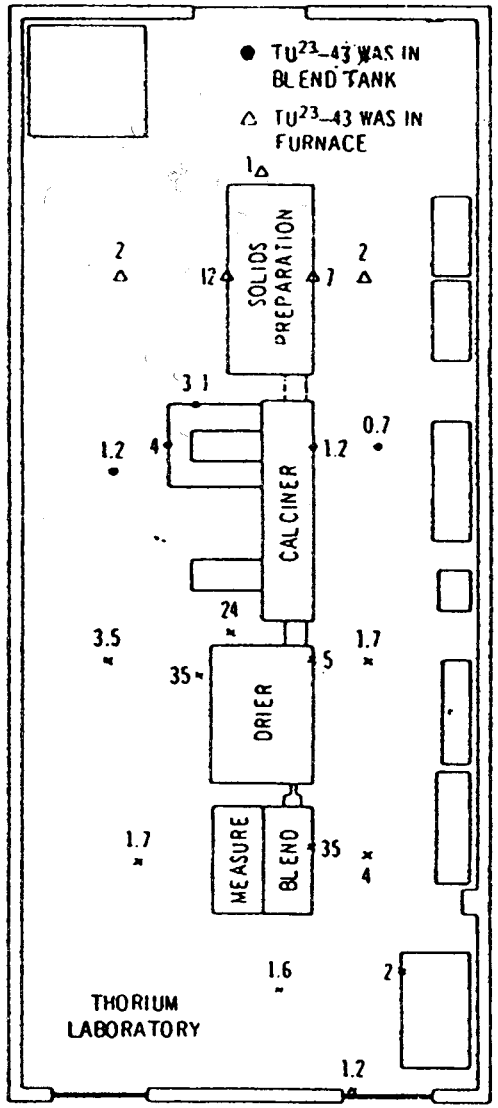


Figure 12

Fig. 11 - Sol-gel area survey during first run. Readings taken batch when was at each station. All values are in milliroentgens per hour. (U nitrate deactivated by ion exchange shortly before).

Fig. 12 - Sol-gel area survey during fourth run. All values are in milliroentgens per hour. (U nitrate 48-60 days old, from ORNL)

(91)

(Thoria/UO2-233 runs).

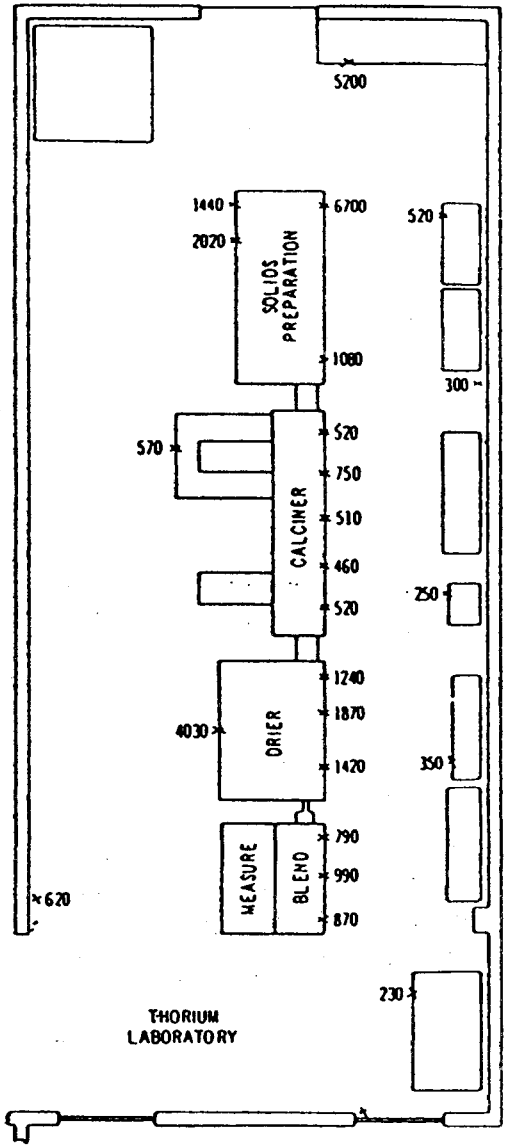


Fig. 13 - Cumulative dose in sol-gel line during U-233 run. Plan view. Dose monitored by film packs. Readings taken 40 to 50 in. above floor. All values are in milliroentgens.
(20 workdays - Aug. 9 - Sept. 3, 1965)
(4 workers, integrated dose : 673 mR)

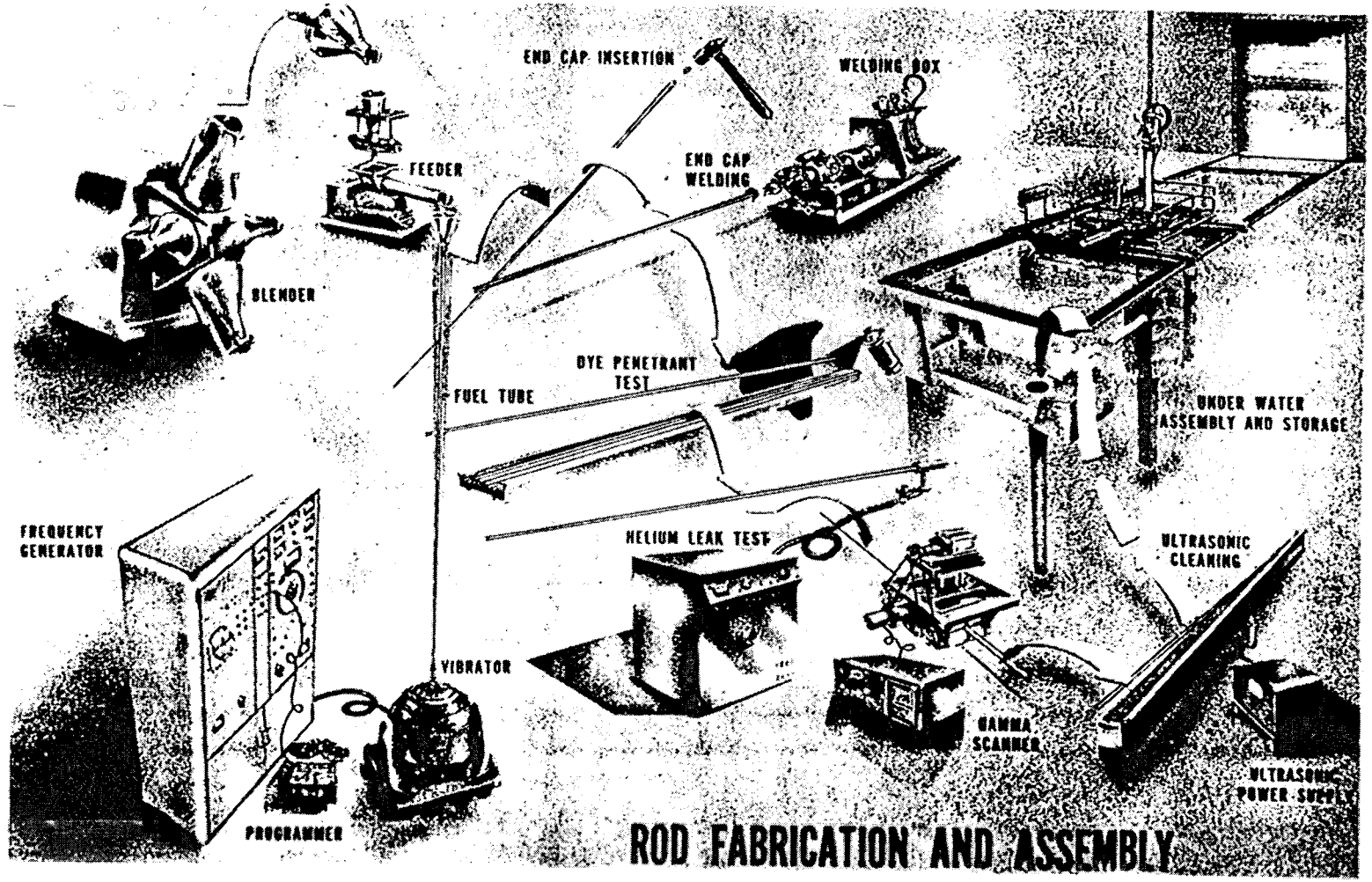


Figure 14: Schematic view of the rod fabrication and assembly line

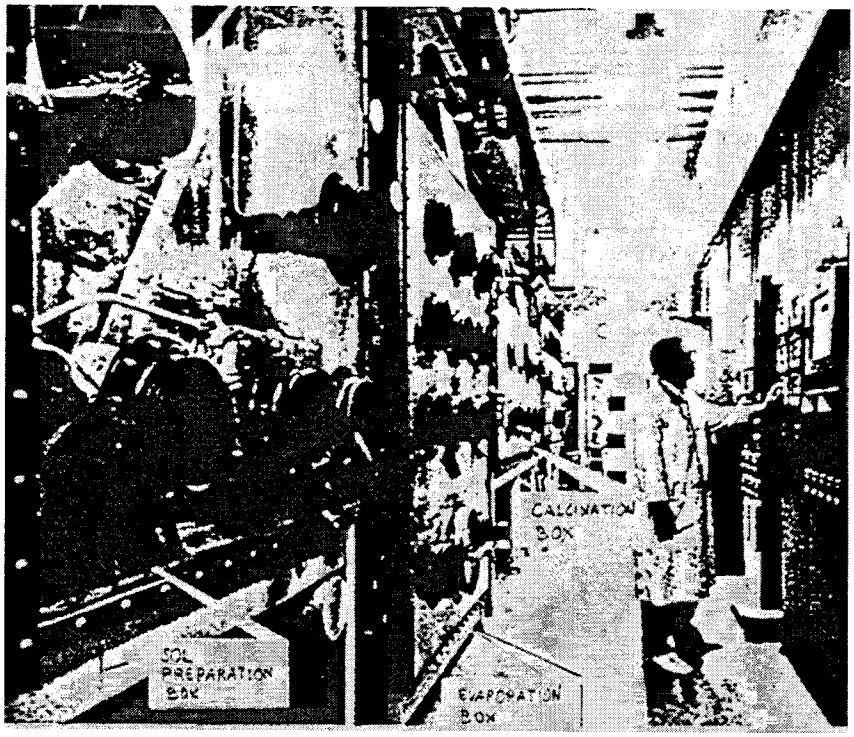


Figure 15: View of fuel preparation glove-box line (99)

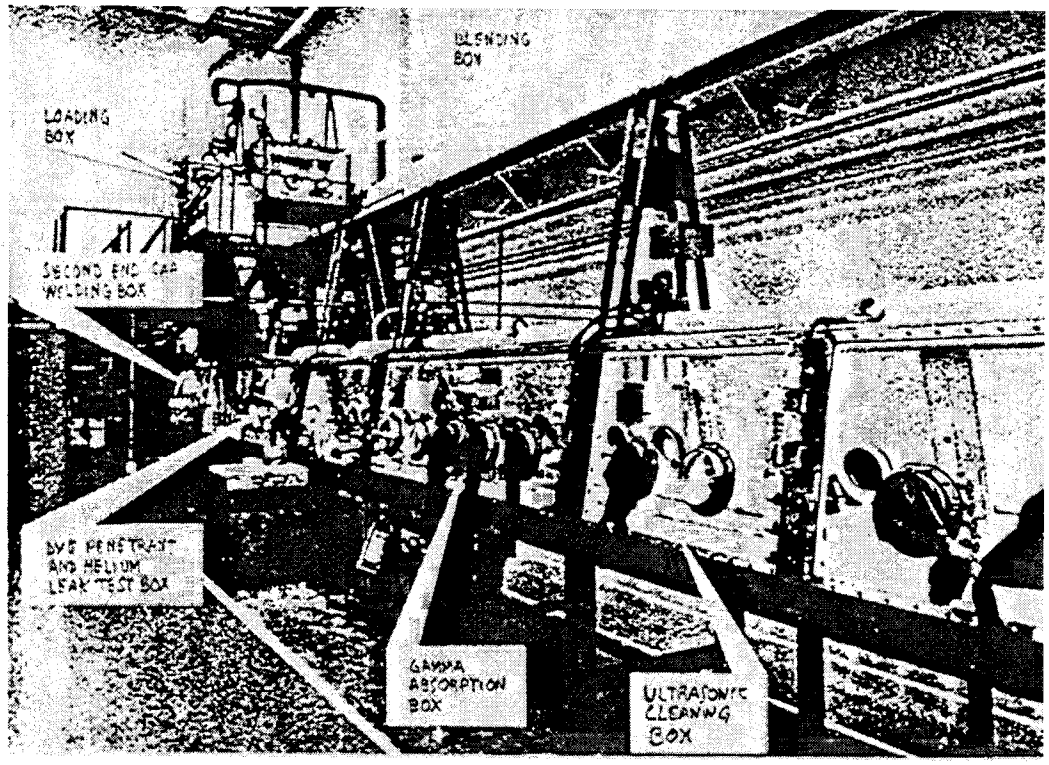


Figure 16: View of the rod fabrication glove-box line (99)

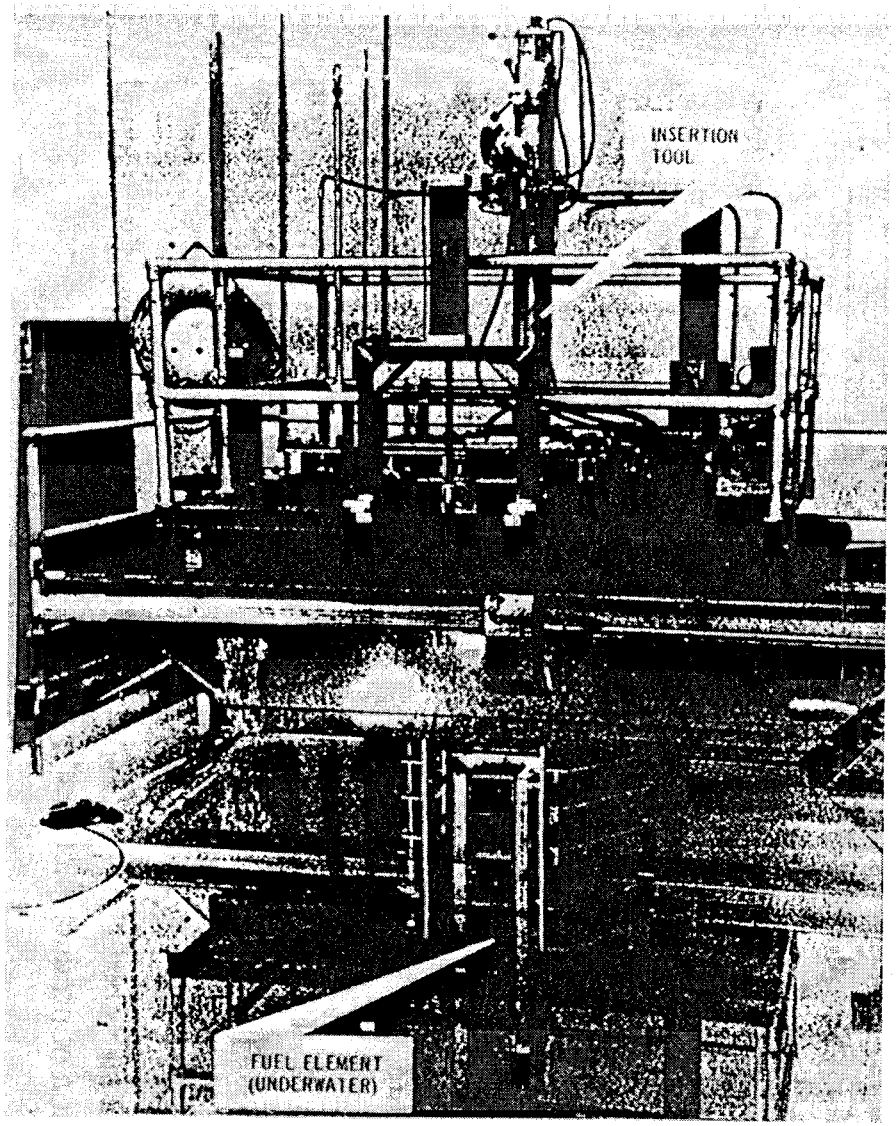


Figure 17: Assembly machine after completion of underwater operations (92)

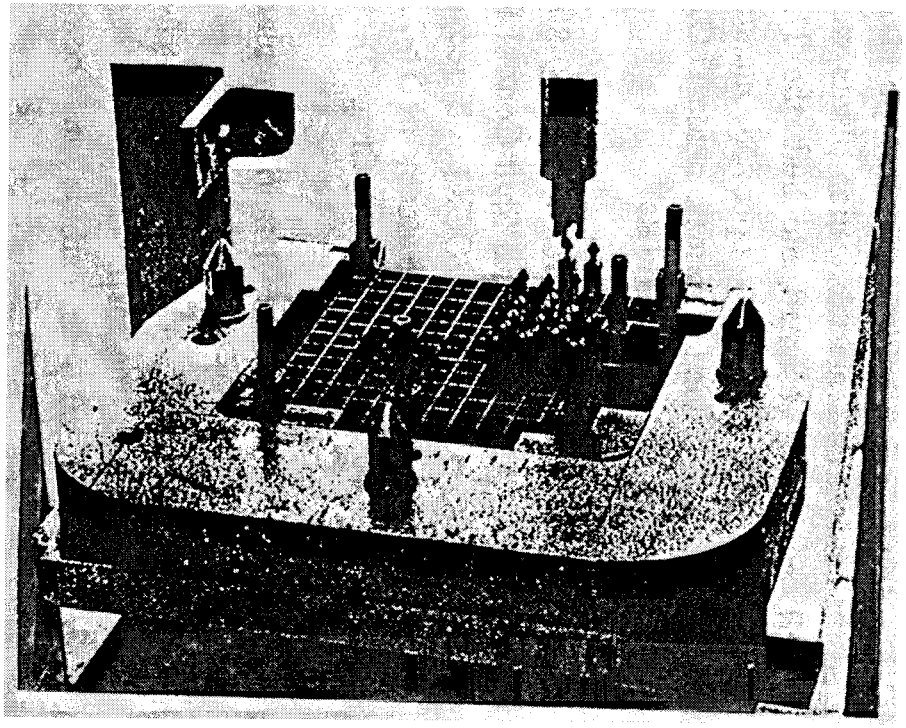


Figure 18: Fuel rod grasping device and fuel rods in element (92)

e) **The TURF ORNL remote fuel fabrication prototype plant.** As early as 1965, it soon appeared that industrial-scale fabrication plants would have to be totally remotized. ORNL engineers designed and built the TURF (Thorium-Uranium Recycle Facility) plant with remotely operated, manually maintained equipment (93). The TURF plant was due to fabricate oxide ThO2/UO2-233 fuel for water-cooled reactors and carbide fuel for HGTRs.

The U-232 content in total uranium was expected to be 500-800 ppm in total uranium for reactors under consideration (Spectral Shift Control Reactor, SSCR, or Heavy Water Organic-Cooled Reactor, HWOCR). If recycling irradiated thorium is practised, gamma radiation from the decay of Th-228 and Th-234 makes remote fabrication mandatory also. (cf. Fig. 20, Chap. 7).

Hence, the remotization of the TURF oxide fuel fabrication plant as shown on Figure 19 (plan view) and Figure 20 (view of a vibrating compactor). It can be seen that the machines become quite complex when remote handling, remote cleaning are mandatory. The same is true for HGTR fuel "compacts" prepared from oxide microspheres transformed into carbide and coating them with pyrolytic carbon. A line for making HGTR fuels in TURF was under design at ORNL in 1968.

Figure 21 shows the shielding requirements in inches of steel or concrete for such plants, based on the following assumptions :

1) The time between solvent extraction and receipt of material at the fuel-element fabrication plant is 5 days.

2) A major cleanup of the equipment and enclosures is performed after 5 working days.

3) No substantial quantity of the material is located closer than 1 ft to the enclosure wall.

4) The amount of material retained in the equipment (from the walls, in crevices, etc...) during processing is 3 kg, but the material is released and continues in the process at 5-day cleanup intervals.

5) The plant processes 110 % of quantity shipped to allow for internal rejection of products not meeting specifications.

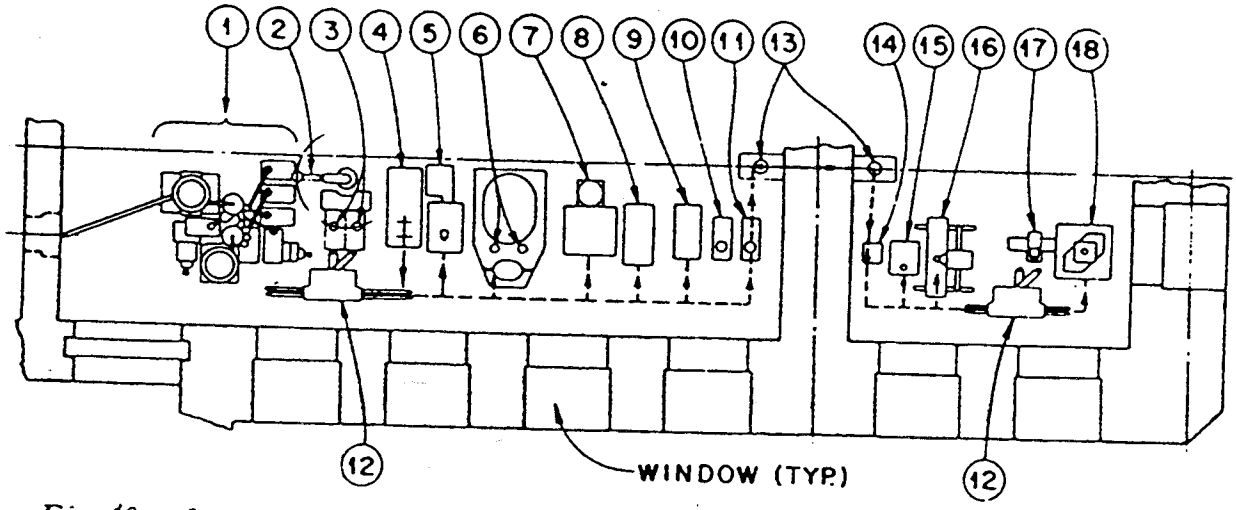


Fig. 19—Layout of equipment for the TURF oxide fabrication line. 1, Powder-preparation equipment. 2, Fuel transfer machine. 3, Vibratory compactor. 4, Tube magazine. 5, Check weigher. 6, Gamma scanner. 7, Capping machine. 8, Welding machine. 9, Leak-check machine. 10, Reject rack. 11, Cleaning and transfer basket. 12, Fuel-rod transfer machine. 13, Cleaner transfer machine. 14, Fuel-rod basket. 15, Contamination monitor. 16, Weld radiography machine. 17, Fuel-rod dimensional check. 18, Fuel-element assembly and inspection machine. (93)

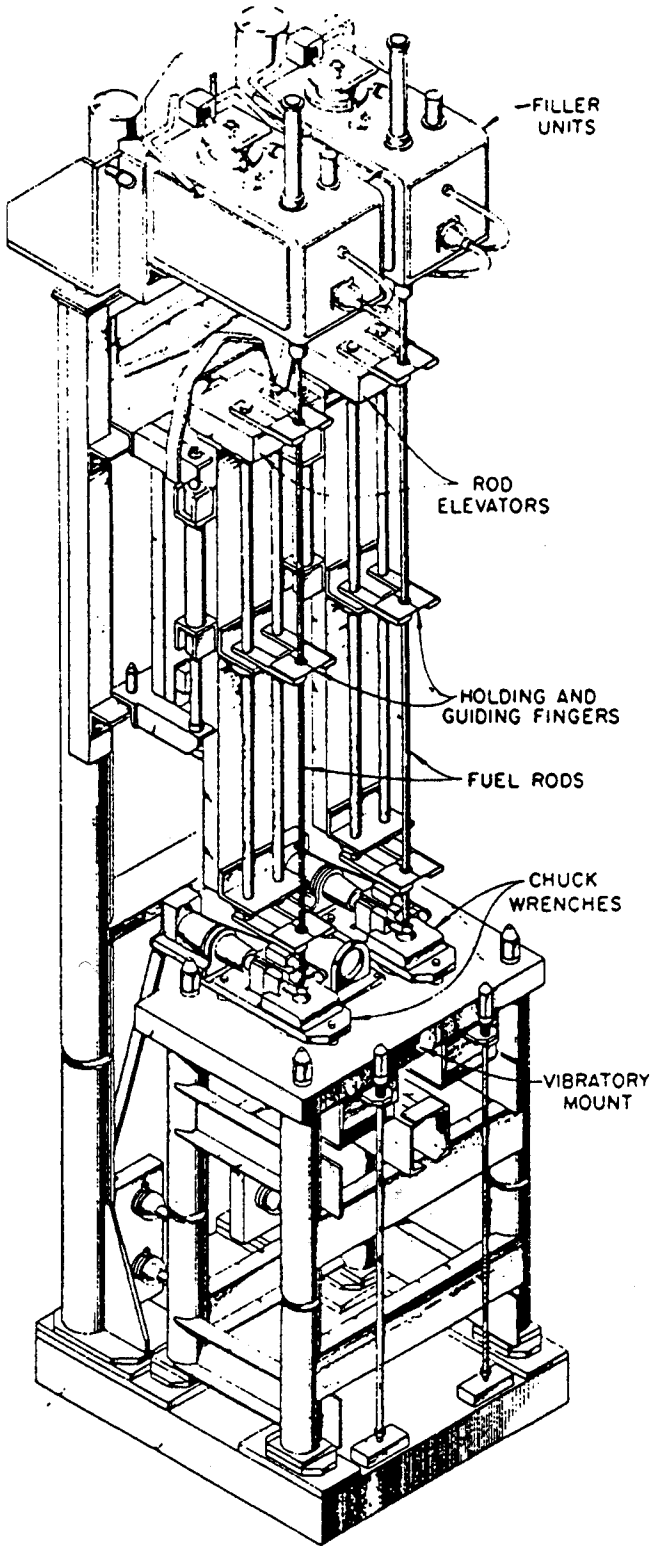


Figure 20: Vibratory compactor - "TURF Plant" (93)

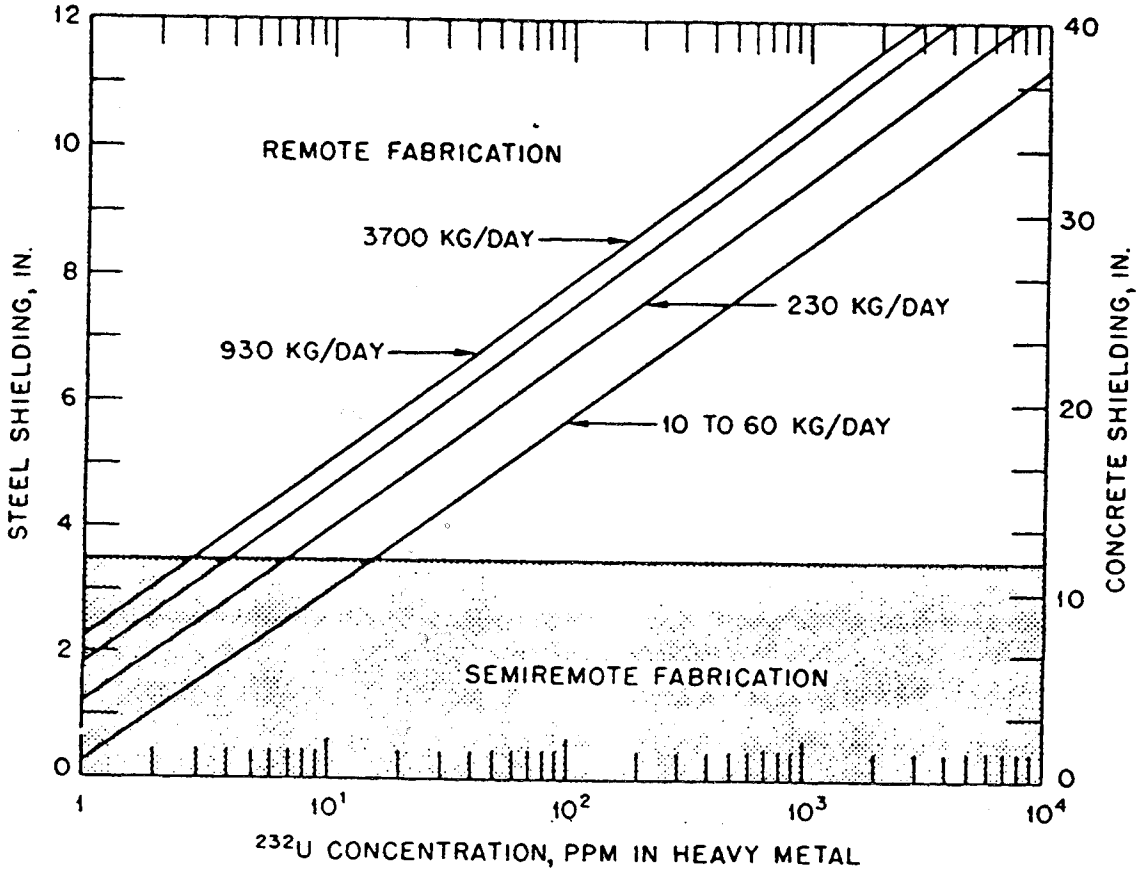


Fig. 21 - Shielding requirements for plants fabricating U-233-bearing HTGR fuel - (93) - (Permissible limits : 5 Rem or 50 mSv/yr)

6) The limit of personnel exposure to radiation is 40 mr/week (0.4 mSv/week).

In the case of HGTR fuel exposed to approximately 100 000 MWd/t, one might expect from 40 to 70 ppm of U-232 in total heavy metal (10 Wt % UO2/90 Wt % virgin ThO2) or 400-700 ppm U-232 in total uranium.

A shielding of 3.5 inches steel was chosen as a practical limit for semi-remote fabrication, because of the difficulty of working through a greater distance with gloved hands or tongs for contact maintenance of the equipment.

A cost analysis based on these designs showed that (Tables 4 and 5 and Fig. 22) it would cost almost 1.5 and 2 times more, for oxide fuels and for carbide fuels, respectively, to fabricate such fuels remotely. As can be seen in this 1968 analysis, the assumptions taken above do not seem conservative enough to us anymore today, and heavier shielding would be necessary (something like 1,2 m), hence possibly higher costs.

Remarks

Although such a plant could be built and operated, the fuel fabrication step has been the main roadblock to the development of the thorium fuel cycle so far. These technical complexities, especially penalizing 30 years ago, and the added costs, combined with other factors, have led to a provisory abandonment of this fuel cycle (except in India for strategic reasons).

The enormous technical progress accomplished since the 1960s, in mechanics, electrical motors, materials, electronics and computers, and practical examples of remotized fuel fabrication (e.g. Melox in France) seem to indicate that time has come to revisit these concepts and start to take advantage of potentialities needed in a not-too-distant future.

Table 4: Cost ratio for comparison of remote and hooded plants fabricating oxide-cluster fuel elements for HWOCR (93)

	Ratio of remote to hooded cost			
	Plant capacity, kg heavy metal/day			
	60	230	930	3700
Capital	1.67	1.60	1.53	1.42
Operating	1.28	1.23	1.19	1.15
Total (including hardware)	1.35	1.30	1.22	1.16

Table 5: Cost ratios for comparison of remote and hooded plants fabricating (Th, U)C_2 particles (93)

	Ratio of remote to hooded cost			
	Plant capacity, kg heavy metal/day			
	60	230	930	3700
Capital	1.87	1.79	1.73	1.75
Operating	1.47	1.43	1.37	1.37
Total (including hardware)	1.53	1.42	1.34	1.35

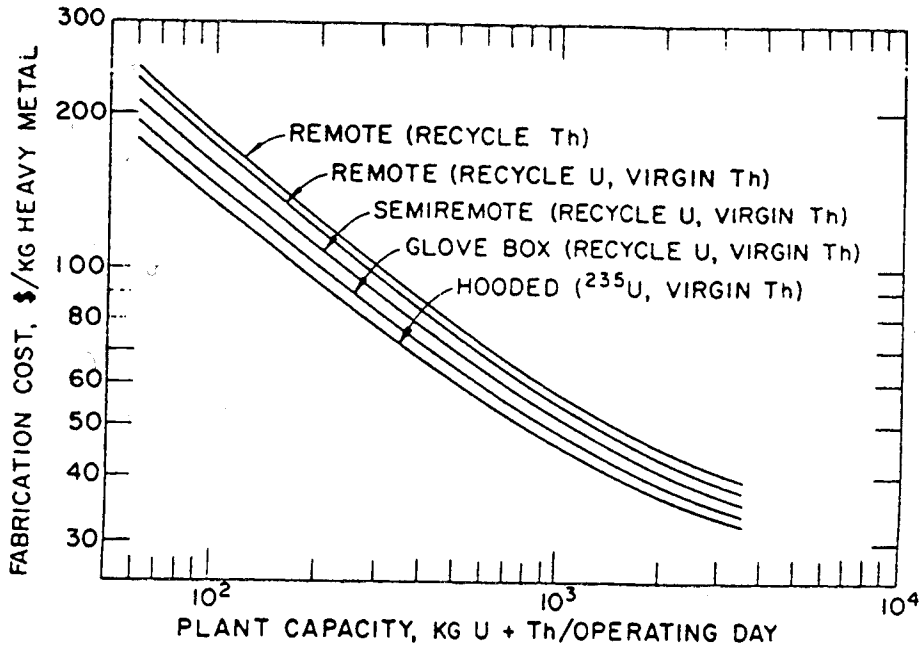


Figure 22: Effect of production rate and mode of fabrication on the cost of fabricating SSCR (spectral shift) fuel elements in a single-purpose plant. Amortization rate 22%. Plant operating days per year, 260.

6.4.3. **Thorium Oxides and Thorium Carbides as base fuel for HTRs**

6.4.3.1. General Remarks

These fuels have been developed mainly for the high temperature, gas-cooled reactors to obtain an optimum heat transfer with excellent neutron slowing-down and reflecting properties.

Usually coated particles will be used to provide fission product containment at the high fuel temperatures of a gas-cooled reactor, so that the gas is relatively free from radioactivity, which may present many advantages, especially during maintenance.

Coated particle is a generic term applied to miniature spherical fuel elements usually 0.5 to 1 mm in diameter. The fuel "kernel" is commonly an oxide or carbide of U (with various enrichments), Th or Pu. Coatings in most general use are pyrolytic carbon (PyC) and silicon carbide (SiC), although a variety of other materials have been examined. With the variety of kernel and coating materials available, coated particles can be optimized for a particular application.

6.4.3.2. Production of kernels (85)

Powder agglomeration processes and wet-chemical processes are developed for kernel production. Both are described :

a) - Powder Agglomeration Processes

The powder agglomeration processes use finely ground powders such as UO2, U3O8, or ThO2 which, after the addition of carbon black and slight quantities of binders and water of organic solvents, are formed into spherical particles by mechanical movements. After drying, carbonizing the binder, and burning out the carbon, the kernels are sintered to oxide. In order to produce carbide kernels, a mixture of oxide powder and carbon black is reacted to carbide and melted. Agglomeration processes were developed, inter alia, under the Dragon Project.

For example, the granulation process used by NUKEM for the production of $(Th,U)C_2$ kernels for the AVR reactor has gained technical importance. In this process each particle in the granular oxide powder-carbon black material is first enclosed by a carbon black layer about 100 µm thick. The particles are then melted in bulk at 2500°C in graphite crucibles. In other processes the kernels

which have first been converted to carbide and sintered at about 2000° C are allowed to fall through a vertical graphite tube furnace heated to a temperature of 2500 to 2700° C or through a plasma, and are thereby melted.

Depending on the excess of C, kernel densities of 80 to 90 % of the theoretical density and higher are attained. Carbide kernels have been used *so far* in the Dragon, Peach Bottom, AVR, and Fort St Vrain reactors.

The disadvantages of powder agglomeration processes are that generally high throughputs and yields cannot be achieved and that a wide grain spectrum is often produced. Moreover, the quality of the powder must meet stringent requirements with regard to fineness and sinterability. In conjunction with these disadvantages, with oxide kernels, relatively low kernel densities of about 80 % of the theoretical density are attained.

Also, as we have seen for oxides, the powders have a tendency to stick to the equipment and hence create problems of contamination and radiation hazard.

In the fabrication of kernels from highly enriched U, powder agglomeration processes offer the advantages that the converted U3O8 or UO2 powder can be processed directly and additional nitrate waste is not produced.

b) - Wet-Chemical Processes

Wet-chemical kernel production processes can be divided into the following four groups :

1) Sol-gel processes with internal or external gelation of the sol droplets.

2) Hydrolysis processes with internal gelation of the droplets from an uranium solution in the presence of hexamethylene tetramine by heat treatment.

3) Gel precipitation processes with gelling of the sol or emulsion or solution droplets with ammonia.

4) Polycondensation processes with solidification of solutions of two organic compounds containing uranium, thorium, or boron in liquid state (boron for absorber particles).

5) A special wet-chemical process is the resin process originally developed by ORNL, in which cation exchange resin beads of suitable size are loaded with uranium from a uranyl nitrate solution.

Wet-chemical processes have acquired increasing importance because they are highly flexible for use in the production of kernels of different size and chemical composition with a high throughput and narrow particle size distribution. Moreover, wet-chemical processes have been developed which permit direct use of the raw materials UO2(NO3)2 from the reprocessing/refabrication stage and Th(NO3)4. In fresh fuel fabrication it is necessary to produce the uranyl nitrate solution from intermediate U3O8.

A characteristic which all wet-chemical processes have in common is that aqueous solutions containing U and/or Th are transformed into droplets and then gelled to spheres. The droplets are formed in liquids which are either not miscible with water or miscible with water to only a slight extent, or they are formed by free fall through air and are collected in aqueous solutions. Gelation of the droplets is accomplished either through a chemical reaction (usually with ammonia at room temperature or with materials such as hexamethylene-tetramine with separation of the ammonia by heat treatment) or by removal of water in case of aquasols. During chemical reactions with ammonia, the uranyl or thorium ions are precipitated as so-called ammonium diuranate (ADU) or thorium hydroxide, whereby NH4NO3 is formed as a by-product in the wet spheres.

Gel-Precipitation Process of NUKEM

Nukem has developed a gel-precipitation process to the point where it can be used for production. With only a slight change in composition, this process can be employed to produce all types of reactor fuels and fertile materials in current use. Up to 1985, about 10 t of (Th,U)O2 kernels have been produced in a mixture ratio of ThO2-UO2 ranging between 4 : 1 and 40 : 1, and with diameters of 200, 400, 500, 600 and 800 µm. In addition, about 2 t of ThO2 kernels with diameters of 500 and 600 µm, as well as about 100 kg of melted UC2 kernels with a diameter of 200 µm, have been produced. Moreover, using this process during the development program, laboratory quantities of UO2 kernels as well as sintered UC2 and U(C,O) kernels with a diameter of 200 µm were produced.

The gel-precipitation schema for (Th,U)O2 kernels is illustrated in Fig.1. Figure 2 (12) gives one process used at Nukem for sol-gel preparation of kernels.

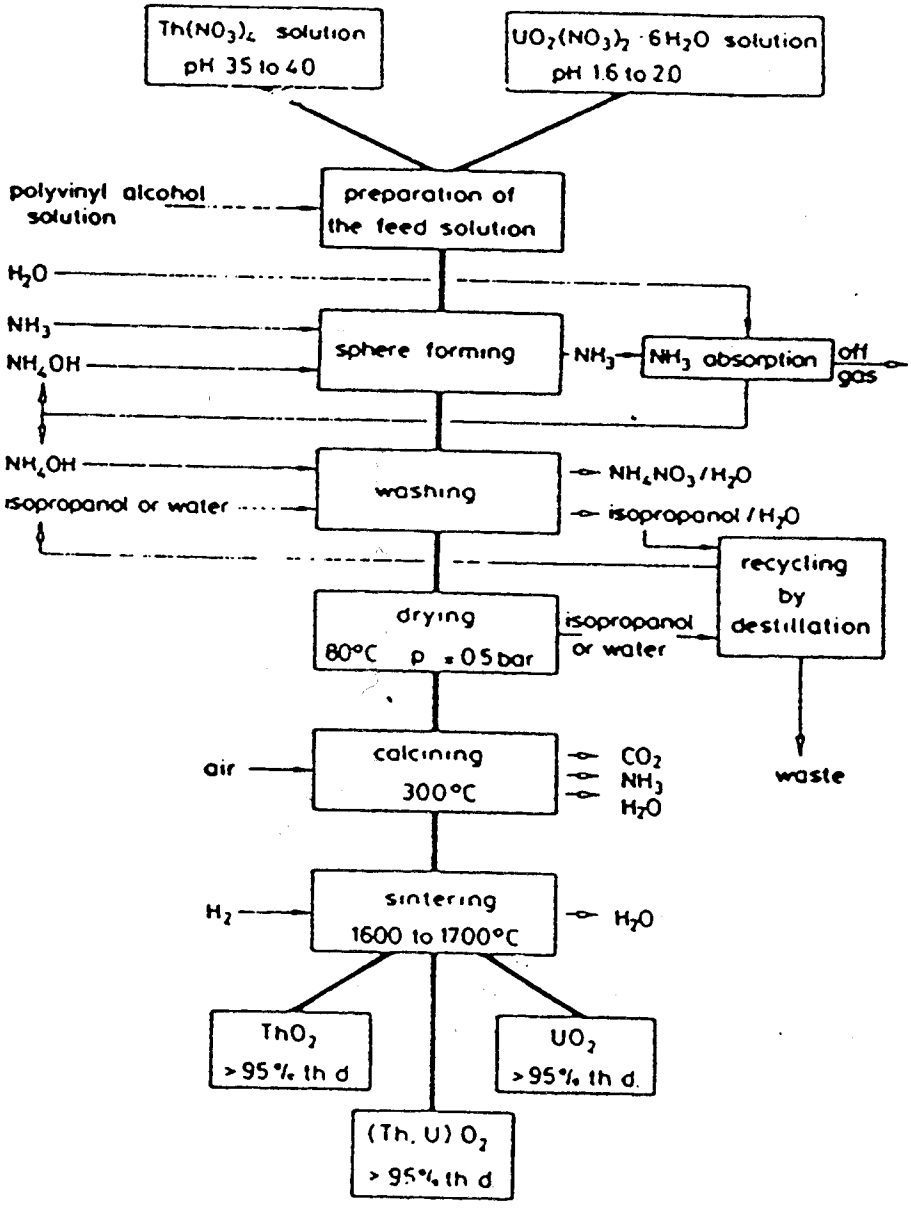


Figure 1: Flow sheet of the gel precipitation process for the fabrication of ThO_2, UO_2 and mixed oxide kernels

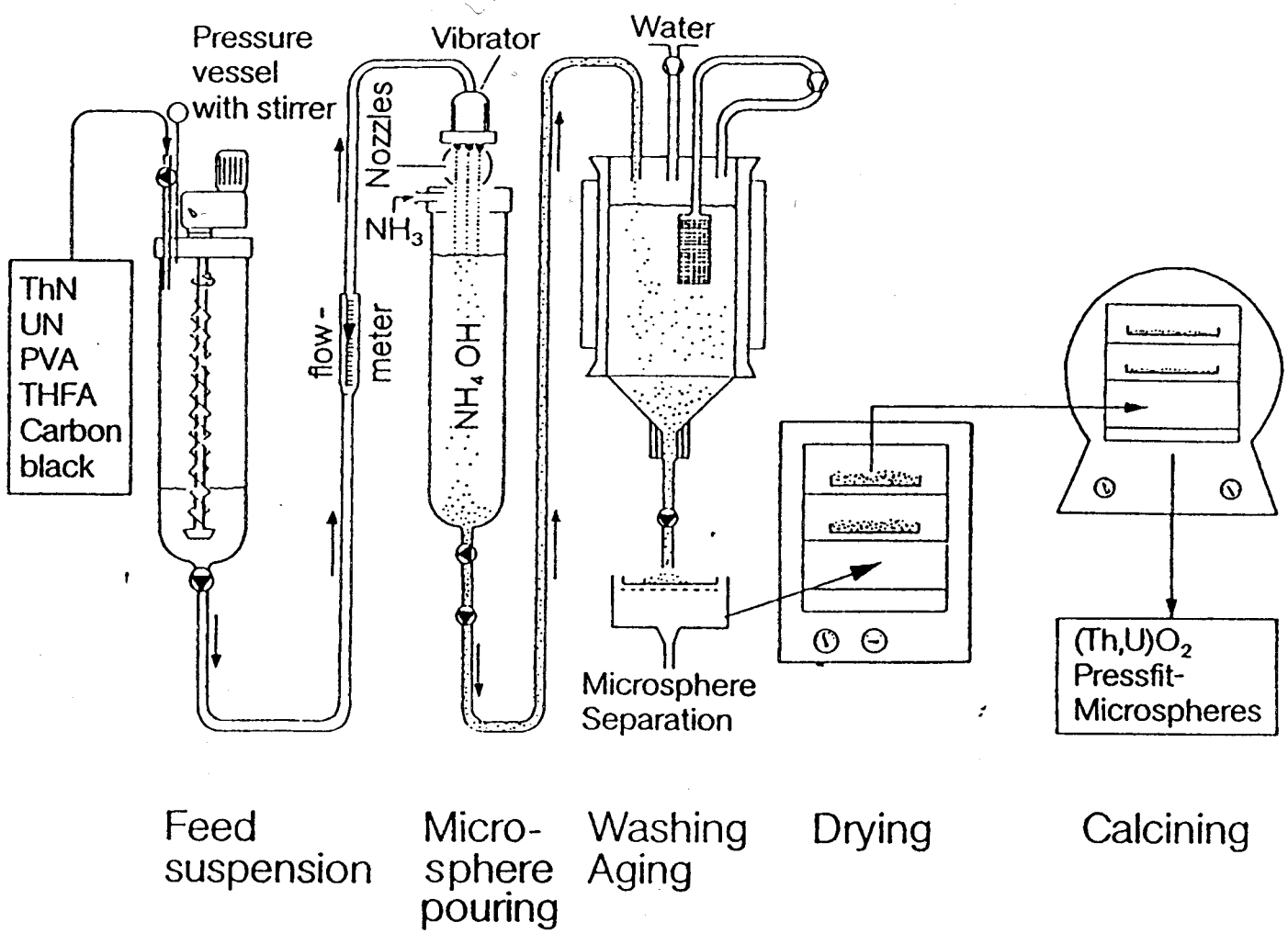


Figure 2: Flow sheet of the microsphere production process (12)

In subsequent processing stages the spherical particles are washed, dried and calcined. These processing stages are followed by sintering to produce oxide kernels.

To produce carbide kernels, the oxide particles are mixed with carbon black and reacted to carbide by sintering or melting. The kernel size depends only on the concentration of U and/or Th in the droplet and on the final density of the particular chemical compound which can be attained.

6.4.3.3. Kernel Coating

Two principal types of coating designs used on the HTGR fuel kernels are called *BISO and TRISO. The BISO is an all-C sequence of two or more PyC* pyrocarbon coatings used for low-burnup ThO2 kernels. It consists of a porous buffer layer enclosed by one or two high-density PyC coatings. The TRISO design is used especially for fissile material intended for high burnup and differs from the BISO in having a SiC coating sandwiched between 2 high-density PyC coatings.

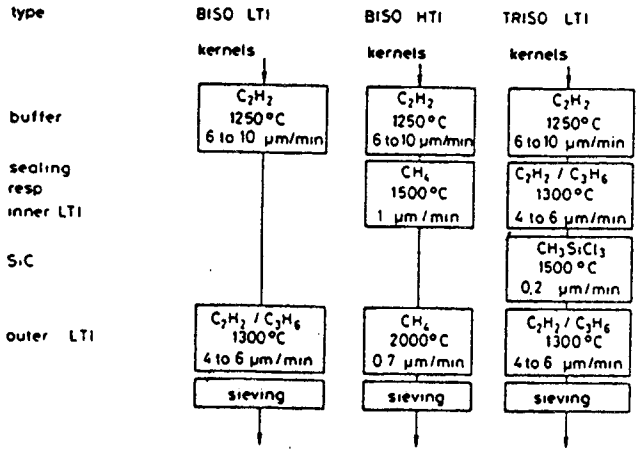
The so-called LTI coating is a low-temperature isotropic PyC deposited near 1350° C (about 86 % theoretical density). HTI is a high temperature isotropic PyC deposited near 2000° C.

The process flowsheets for obtaining these coatings are represented on Figure 3 and their physical characteristics on Table 1.

The BISO HTI-type coating has been applied in 2500 coating batches of 3 kg each for the THTR and the AVR reactors in Germany. The BISO and TRISO LTI types are applied in 3 to 10 kg batches onto 400 to 500 μm kernels, while the TRISO LTI type is also deposited in 0.5 kg batches onto 200 μm fissile kernels.

A modification of the described coating technique has recently been developed. In this technique, the fluidizing gas is injected through the bottom and the coating gas from top. It is expected that the so-called top-coating improves the process and gives a more homogeneous product.

Typical coating designs that have been investigated extensively are described by Gulden and Nickel (87). Some specifications on the particles employed in the Fort St Vrain and THTR Uentrop reactors are listed in Table 2.



The BISO HTI-type coating has been applied in 2500 coating batches of 3 kg each for the THTR and the AVR. The BISO LTI and TRISO LTI types are applied in 3 to 10 kg batches onto 400 to 500 µm kernels, while the TRISO LTI type is also deposited in 0.5 kg batches onto 200 µm fissile kernels.

Figure 3: Flowsheet for BISO LTI, BISO HTI, and TRISO LTI coating processes (85)

Table 1 (85):

	BISO HTI	BISO LTI	TRISO LTI	TRISO LTI
outer LTI				
thickness				
$\bar{x}$[b] (in µm)	75	80	35	35
v[c] (in %)	10	10	10	10
density (in g/cm³)	1.85	1.95	1.90	1.90
anisotropy BAF_{opt}[d]	1.15	1.03	1.03	1.03
coated particle				
OPyC contamination (HM/HM_{tot})	10×10^{-4}	1×10^{-4}	5×10^{-6}	5×10^{-6}
def. SiC (HM/HM_{tot})	—	—	1×10^{-4}	1×10^{-3}

[a] HMO = heavy metal oxide. – [b] Mean value. – [c] Relative standard deviation. – [d] BAF_{opt} = Bacon anisotropy factor (optical).

Table 2 (87):

Specifications of Coated MX_2 Fuel Particles in Technical High-Temperature Graphite-Moderated Gas-Cooled Reactors (HTGR), where M Denotes U, Th or (Th,U) and X is C or O [7].

reactor	Fort St. Vrain fissile	Fort St. Vrain fertile	THTGR-300[f], Uentrop fissile-fertile mixed	LHTGR[g] fissile	LHTGR[g] fertile
kernel composition[a]	$Th_{0.81}U_{0.19}C_2$	ThC_2	$Th_{0.91}U_{0.09}O_2$	UC_2	ThO_2
diameter in µm	200	350	400		
coating design	TRISO	TRISO	BISO	TRISO	BISO
buffer[b] thickness in µm	50	70	75(C_2H_2, 1450°C)		
inner PyC thickness in µm	—	—	60(1500°C)		
SiC thickness[c] in µm	15	15	—		
outer PyC thickness in µm	50[d]	50[d]	80(HTI, CH_4, 2000°C)[e]		

[a] HEU (= high-enriched uranium), 93% ^{235}U + 7% ^{238}U. – [b] Buffer, a porous PyC with density D = 1.1 g/cm³. – [c] LTI, low temperature isotropic PyC deposited near 1350°C. D = 1.85 to 1.95 g/cm³ (theoretical density D_{TD} 2.2 g/cm³). – [d] HTI, high temperature isotropic PyC deposited near 2000°C. D = 1.85 to 1.90 g/cm³. – [e] SiC, high density with values 3.21 to 3.22 g/cm³ (D_{TD} = 3.215 g/cm³). – [f] THTGR, thorium HTGR. – [g] LHTGR, large HTGR (proposed for the future).

6.4.4. **HTGR Fuel Element Manufacture**

6.4.4.1. The manufacturing process established at Oak Ridge (93) and designed for the remotized TURF facility, is given on Fig. 4.

The facility has been divided into 3 zones :

Zone 1. The oxide microspheres are received from the fuel reconstitution facility, converted to carbide, and inspected.

Zone 2. The fuel particles are coated with pyrolytic carbon and inspected.

Zone 3. "Compacts" are prepared, inspected, and loaded into the graphite sleeves. Finally, two fuel segments are assembled together, the end-pieces are inspected, and the assembly is shipped to the reactor site. The so-called "compacts" are those cylindrical blocks made of the sintered kernels, packed in the fuel holes of the graphite sleeves (Fig. 5).

6.4.4.2. The manufacturing process established in Germany, Jülich and Nukem (85), to make the reactor "pebbles" of about 60 mm diameter for the ATR and THTR reactors, is described in Fig. 6. About 650 000 such pebbles were manufactured. (87).

Regarding the key problem of optimum retention of fission products and the related requirements on the tolerable defect rates of SiC coating layers within the fuel element fabrication process, the following crucial points have been identified :

1) The defect rate due to operation is chiefly controlled by precise coated-particle design and by sufficiently narrow bandwidths of the coating layer properties, especially thickness.

2) The defect rate due to fabrication is controlled by the sphere-molding step and the precautions taken to exclude coated-particle failures during molding.

All efforts to reduce the fabrication particle defect fraction below the design value of 60×10^{-6} are concerned by these precautions.

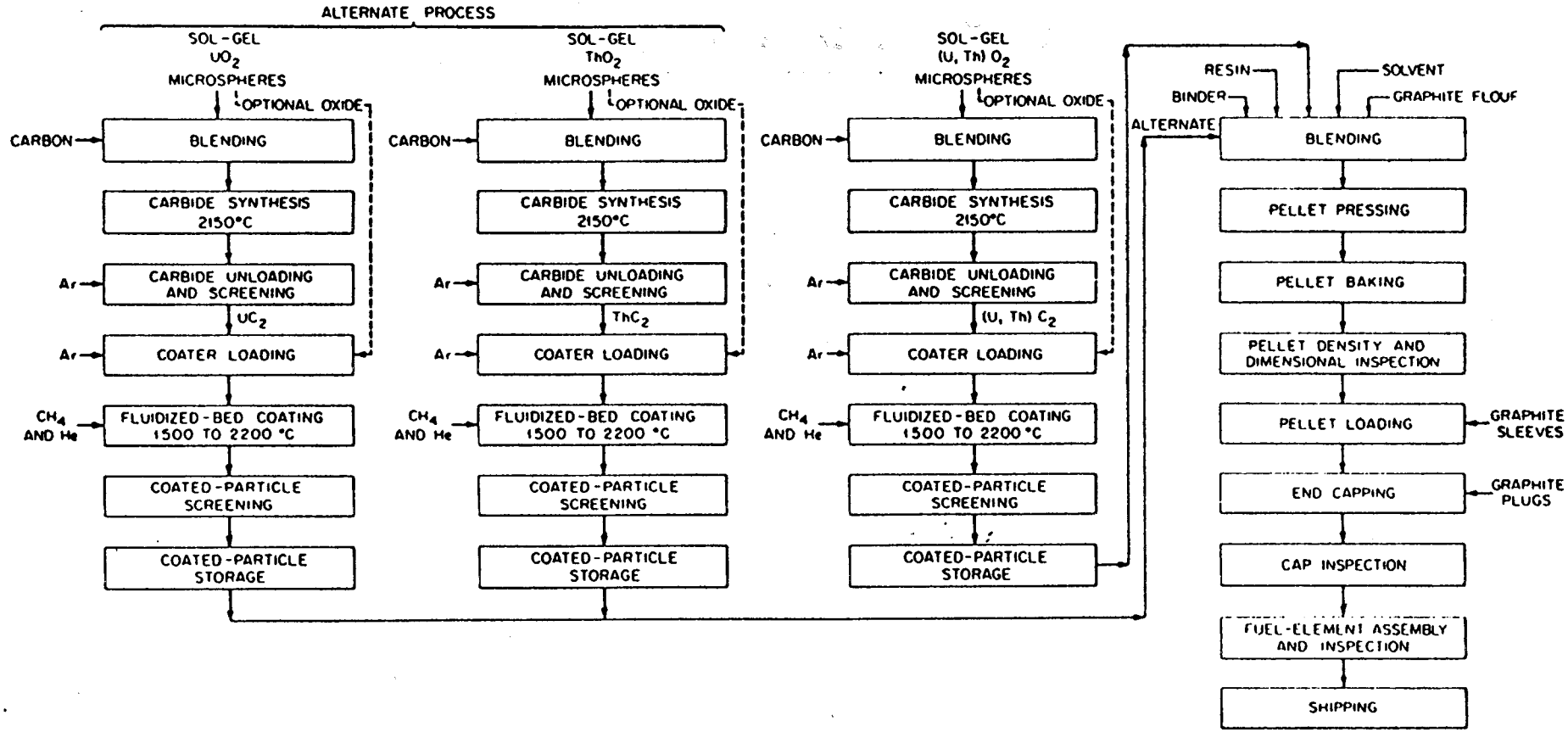


Figure 4: Flow sheet for fabrication of HTGR fuel elements (93)

6.4.5. Some Remarks about LWR and HTGR Fuel

The cladding on LWR fuel, by virtue of its good thermal conductivity, operates at temperatures controlled largely by the coolant ranging from 300 to about 650°C. Under influence of the coolant pressure, the cladding serves to stabilize the fuel geometry and restrain fuel swelling.

The coatings on the HTGR fuel particles operate at very nearly the same temperature as the fuel, 800 to 1350° C. These high temperatures intensify effects from irradiation-induced dimensional changes, fission gas pressure buildup, and temperature-controlled diffusional processes. The effectiveness by which the fission products are retained in the different fuels is governed by a complex interrelation between many factors including radiation damage, swelling, chemical corrosion and composition changes at elevated temperatures

Figure 5 gives some of the characteristics of HGTR fuel "compacts" and "pebbles" compared to LWR fuel pins.

The following Table 3 compares further the two main types of fuel.

From this comparison and from the above discussions, one can deduct that, whereas HTGR carbide fuel can withstand very high temperatures for a long time and high burn-up, compared to LWR oxide fuel, it is much more complex, and expensive, to prepare.

6.4.6. Other Mixed Fuel Types

For completeness' sake we shall mention the nitrides (97), berillides (155) and of course the molten salts, which would need a development of their own, outside the scope of this Study.

Figure 5: Comparison of converter fuel geometries (Gmelin, 85)

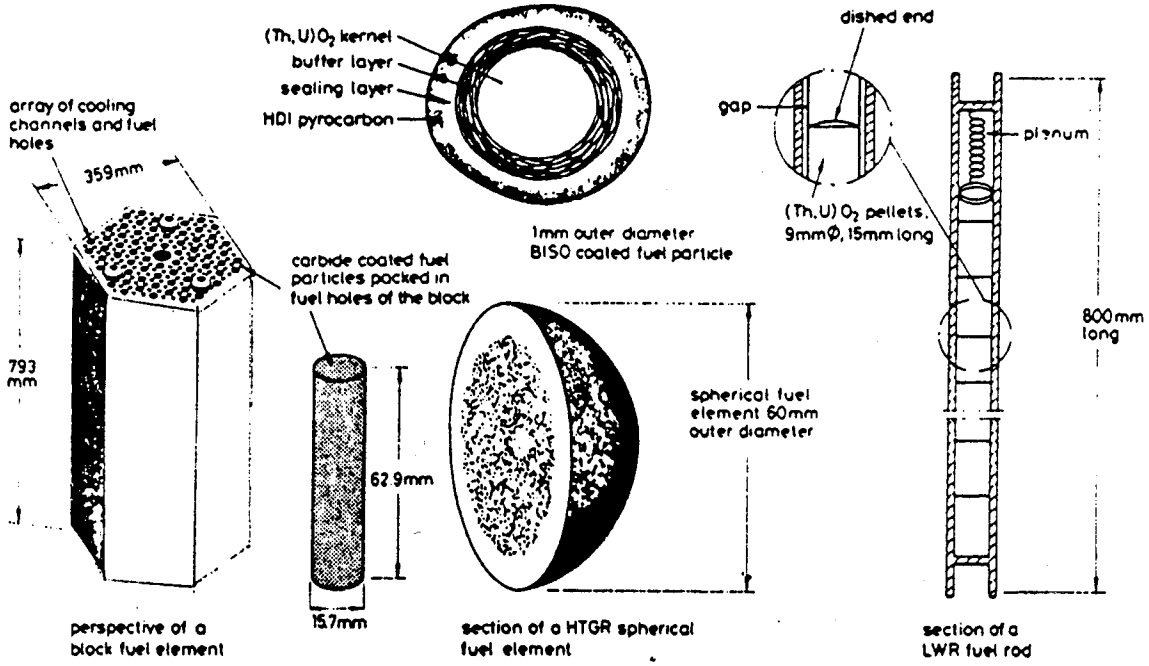
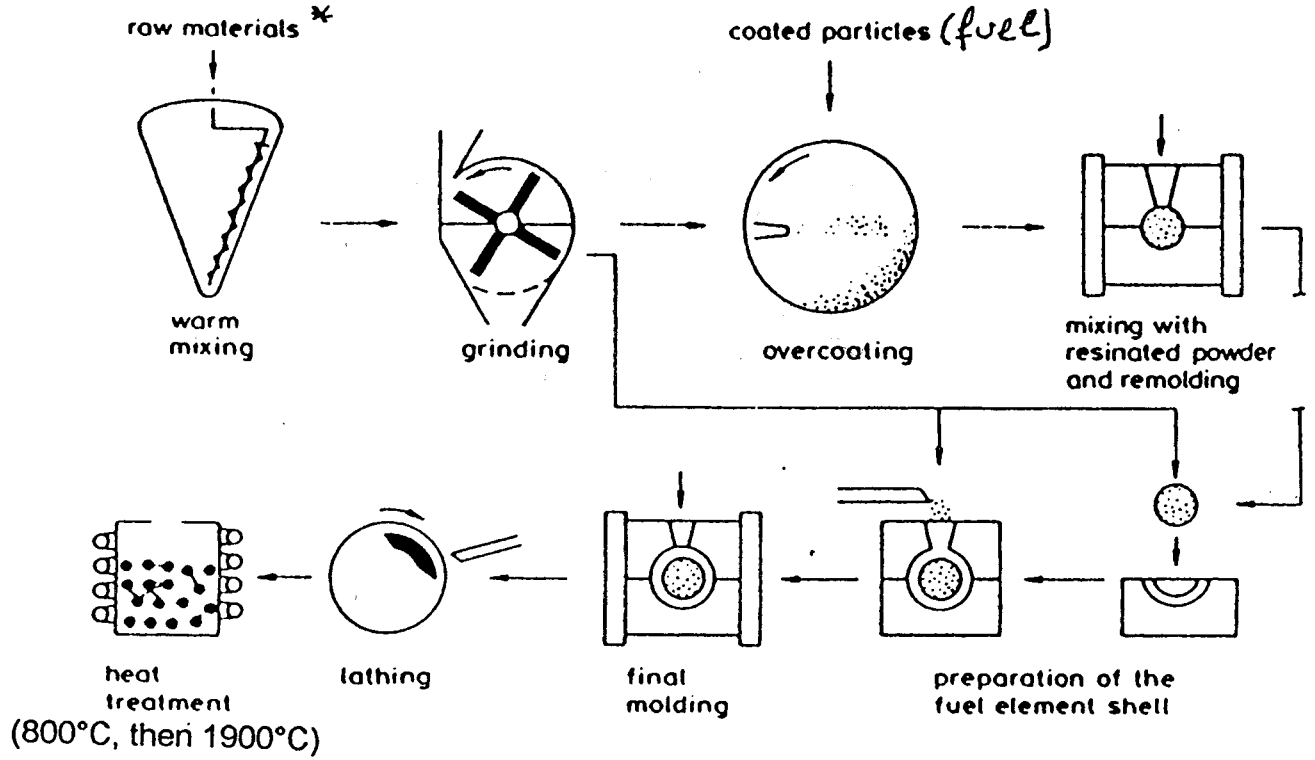


Table 3 (85): Comparison of the Functional Parameters of the High-Temperature Gas-Cooled Reactor (HTGR) and the Light Water Reactor (LWR) [7].

	HTGR	LWR
Operation Parameters		
fuel	MO_2 or MC_2	MO_2, MC or MN, M = (Th, U)
core size in m	6 Ø and 6 long	2 Ø and 2.5 long
fuel temperature in °C	800 to 1400	650 to 2000
coolant pressure in bar	50	60 to 150
specific power in MW/kg	0.3 to 0.5	1 to 2
power density in MW/m³	6	20 to 80
Φ_F in n/m²	4 to 7×10^{25}	0.6×10^{25}
fuel residence time in y	2 to 4	0.5 to 2
burnup in % FIMA [a)]	10 to 14 (78% separated fissile)	0.5 to 5
Fuel Behavior		
fast neutron damage	PyC shrinkage	cladding and fuel swelling
fission fragment damage	unimportant	cladding embrittlement and fuel swelling
swelling	unimportant	influences heat transfer and fuel temperature, life, and creep

[a)] 1% FIMA = 2.23×10^3 MWd/kg M, where M is the gram atom weight of fissionable metal and FIMA denotes the fissionable initial heavy metal atom in which the fission energy assumed is 1 f = 200 MeV.

(For ThO2, ThC, 1 % FIMA = 9610 MWd/t - PuO2 = 9330 - UO2 (HEU) = 9480)



* Phenol, graphite powder, hexamethylene tetramine (85)

Figure 6: Fabrication of spherical fuel elements (85)

6.5. Fabrication of U-233 MTR-Type Fuel Elements

Although logically one should first deal with spent thorium fuel reprocessing, we have preferred to mention U-233 fuel fabrication with the other types of fuels. G.J. Prasad and C. Ganguly (84) have reported the manufacture of U-233 MTR-type fuel elements which are fuelling the materials testing reactor KAMINI at the Indira Gandhi Institute, Kalpakkam. To our knowledge, this is the first time that such fuel elements have been manufactured with U-233.

The Al clad Al-20 % U-233 plate fuel elements were fabricated by suitable modification of the conventional "melting, casting, picture-framing, roll-bonding" route. A total of 72 fuel plates were fabricated. Each plate contained approximately 8.5 g U-233.

A small zirconium addition (1 %) improved the morphology of Al-20 % U alloys and facilitated subsequent rolling. Since zirconium has a lower parasitic neutron absorption cross section, it is definitely a better additive compared to that of silicon which has been used by earlier workers.

Use of aluminium moulds for casting and silicone oil for lubricating the roll surfaces also contributed significantly in achieving yield and quality of the final product.

The fabrication campaign has paved the way for taking up large scale production of Al clad aluminium matrix plate fuel elements for research reactor.

The small amount of U-232 (a few ppm) did not impair the fabrication of the "meat" which was carried out in unshielded glove-boxes. Table 1 gives the salient specifications of the fuel elements, while:

Fig.1 shows the fabrication flowsheet;
Fig.2 is a view of one Al-clad plate;
Fig.3 shows the fuel assembly construction and
Fig.4 shows the fuel elements inside the PURNIMA III critical facility, to be used later in the KAMINI reactor.

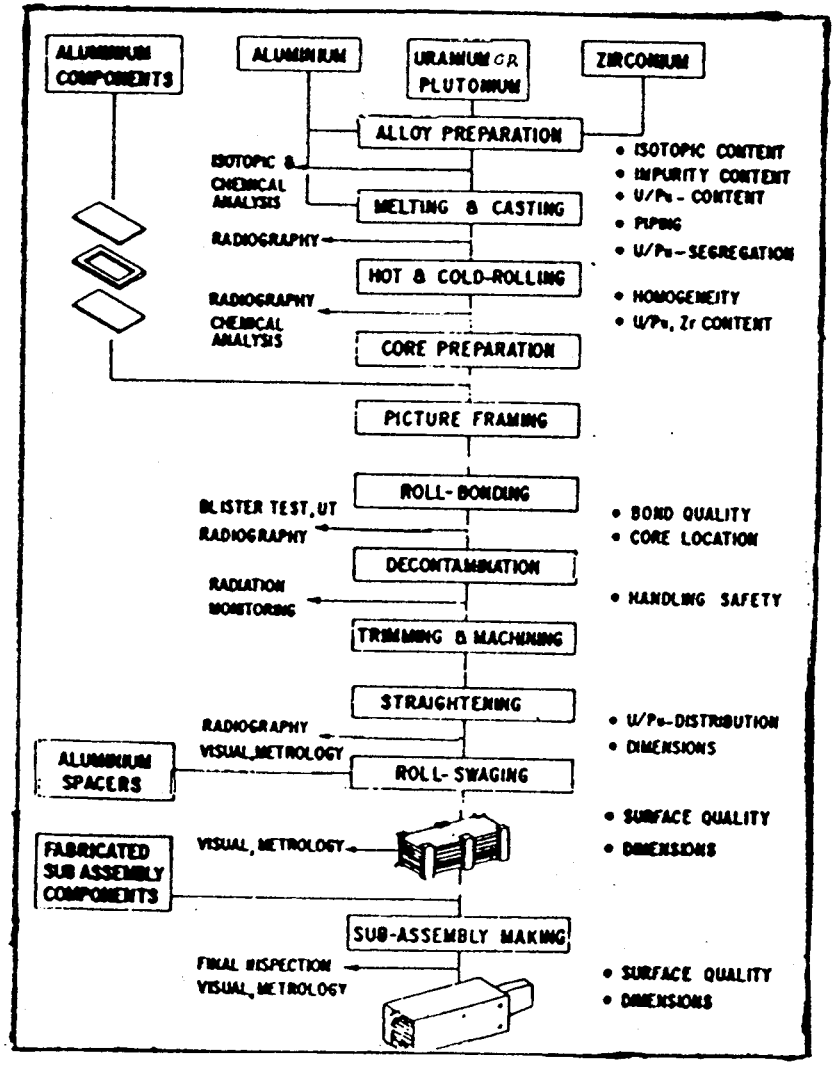


Fig. 1 Flowsheet for fabrication of fuel plate and fuel subassembly

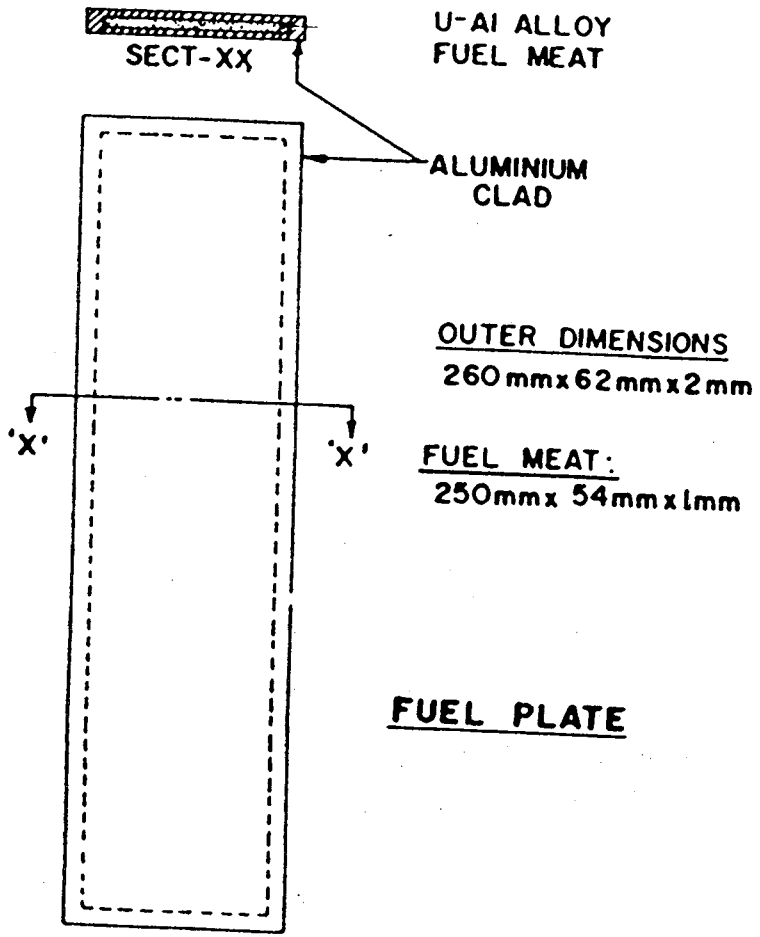
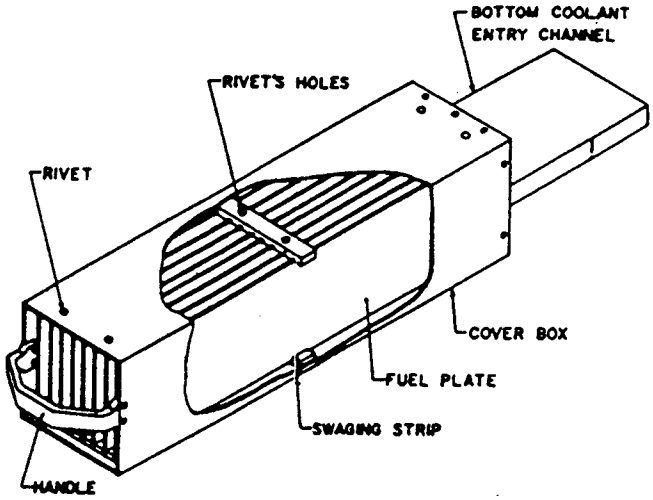


Fig. 2 Schematic of Al clad, Al-20%U - 1%Zr alloy fuel plane (34)

Table I : Salient specifications

Fuel plate dimension (mm)	:	2 x 62 x 260
Fuel alloy meat dimension (mm)	:	1 x 55 x 250
Aluminium clad thickness (mm)	:	0.5
Fuel alloy	:	Al-20%U-1%Zr
Fissile isotope	:	U233
Fissile isotope content of uranium	:	98%
Uranium content per fuel plate (g)	:	8.5
No. of fuel plates per subassembly	:	8
No. of fuel subassembly	:	9
Water gap between fuel plates (mm)	:	6

Figure 3:



FUEL SUB-ASSEMBLY

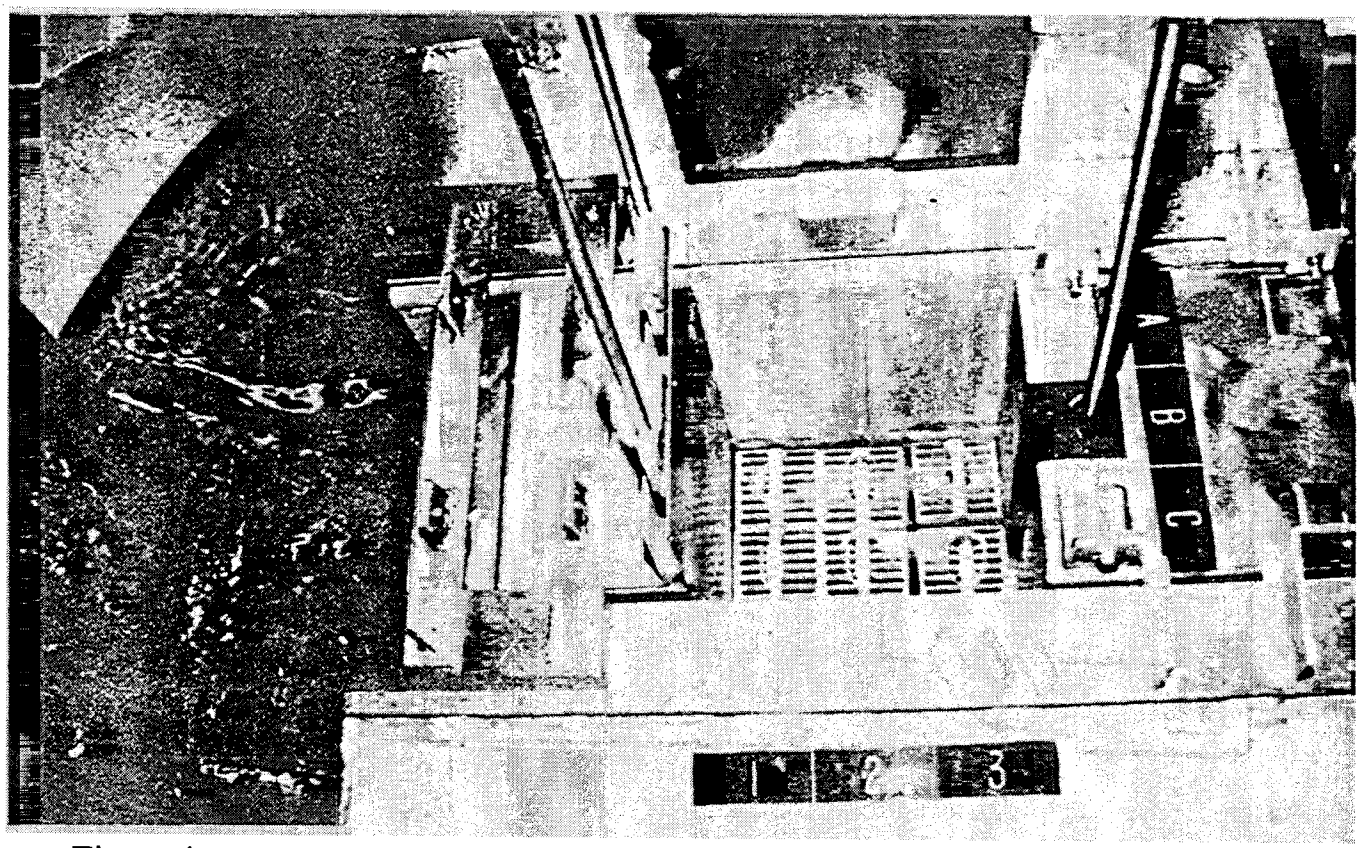


Figure 4:

7. REPROCESSING OF THORIUM-BASE IRRADIATED FUELS AND WASTE MANAGEMENT

7.1. Introduction

Reprocessing is an obligatory step in an economical Thorium-based Fuel Cycle, due to the fact that 1) U-233 bred from thorium has to be separated in order to be used efficiently for energy production, and 2) the used fuels discharged from the reactors need to be freed from nuclear poisons in order to be recycled eventually.

Thus, if there is no need of an enrichment step in the thorium fuel cycle as for U-235/238 LWRs, there is a need for U-233 isolation by chemical means, and possible recycling. There is a great parallel in that respect between the U-238/Pu cycle and the Th/U-233 cycle, each with its peculiarities of course, due to the physical and chemical properties of the isotopes concerned.

In that respect, it might seem that some advantage is to be put to the credit of the Th/U-233 against U-238/Pu, in that the former presents more flexibility in terms of reactors (thermal breeders, high temperature reactors), in terms of fuel stability, and of the allegedly lower toxicity of the ultimate nuclear waste, compared to the uranium-plutonium fuel cycle.

These advantages, however, have an extra cost due to the great stability of the thorium oxide, which is difficult to dissolve, and the necessity to have recourse to remotized and shielded fuel fabrication.

On the other hand, these contingencies might pave the way for integrated reprocessing fuel refabrication plants of a new generation.

In this Chapter we will consider four main aspects :

- The different preparation operations leading to a classical solution of thorium and uranium nitrates, starting from oxide or carbide fuels.
- The usual solvent extraction/purification treatments leading to thorium and uranium salts ready for fuel refabrication.
- The particularities of waste management in the case of a thorium fuel cycle.
- A few words will be devoted to the molten-salt processes, and we shall try to summarize by concluding remarks.

7.2. The "Head-End" Operations

7.2.1. Preliminary Remarks

One has to consider different cases depending of the type of fuel to be treated :

- fertile thorium fuels to be milked out of their U-233,
- UO2-ThO2 fuels to be reprocessed for fuel refabrication,
- coated carbide fuels and oxide fuels for high temperature reactors which present a more complicated case.

The aim of all these operations is to obtain, after preliminary operations leading to dissolution, a solution of U-Th nitrates together with a small quantity of minor actinides and fission products. Ternary fuels such as U-Pu-Th fuels, are a particular case of this general case.

Extraction of U-233 from irradiated thorium rods has been conducted extensively in the USA in the 1950s until 1970s.

Table 1 gives a summary of the thorium-U-233 processing in the US, showing that almost 700 tons Th have been irradiated, delivering more than 1.5 tons of U-233. The separation operations were piloted at ORNL in the Thorex plant, and also at the Knolls Atomic Power Laboratory, and the process was later adapted to the Great "canyon" plants at Hanford and Savannah River (SRP), and the commercial direct maintenance plant of Nuclear Fuel Services at West Valley near Buffalo, now being dismantled, which reprocessed the first core of Indian Point 1 (95 % ThO2, 5 % UO2-235), but without recovering the thorium, left in the waste.

At SRP and Hanford, thorium oxide was the main starting material, compacted in aluminium-clad fuel elements (cf. Ch. 6), although some thorium metal was processed too.

The processes are very well explained in an interesting paper by H.C. Rathvon, A.G.Blasewitz, and colleagues (119). This paper is interesting not only because it gives details on the processes, the adaptation of these large Purex-type plants to the THOREX (or parent) processes, the avoidance of cross contamination with residual U-235/U-238 in the system by flushing, but also because it is one of the early documents (1966) to give some details on these mysterious canyon-type

Table 1. Summary of thorium-^{233}U processing in the United States

Table 1. Summary of thorium-^{233}U processing in the United States

Site	Date	Thorium processed (tons)	^{233}U recovered (kg)	^{232}U content (ppm U)	Flowsheet employed	Remarks
			Irradiated fuel reprocessing			
ORNL	1954 and 1958	5	8	10–40	Interim-23	Pilot-scale development
	1955–1958	30	47	10–40	Thorex	Pilot-scale development up to 4,000 Mwd/ton, cooled 30 d
	Total	35	55			
SRP	1964–1965	14	107	225	Interim-23	Th discarded
	1965	9	19	38	Interim-23	Th discarded
	1966, 1968,	193	412	6–9	Thorex	Th recovered, 1.5 *M* HNO_3, 0.25 *M* $Th(NO_3)_4$, AF, 30% TBP
	Total	216	538			
Hanford	1965	4			Interim-23	Th discarded, flowsheet test
	1966	250	270	6–10	Acid thorex	Th recovered, acid-deficient feed
	1970	400	589	6–10	Acid thorex	HNO3 added below HA column feed plate
	Total	654	859			
Nuclear Fuel Services	1969	17	103(1,019)[a]	125	Interim-23	15,800 MWd/ton, Th discarded, 4.3 *M* HNO_3, 112 g/L Th used as salting agent
			Unirradiated processing			
ORNL	1962	2	50	40	2.5% DSBPP	Rod fabrication, Th added to process; recycled
	1973, 1974, 1975	30	1100	10	5.0% DSBPP-IX	Th added to process; discharged
	1957–continuing	9	225	3–250	2.5% DSBPP	Th added to process
	1957–continuing	9	1,000	3–250		
	Total	41				

[a]Mixture of ^{235}U and ^{233}U; Total ^{235}U and ^{233}U is in parentheses.

Source : R. BROOKSBANK, 1995.

defence reprocessing plants and their huge equipment (Fig. 1 : the Hanford annular dissolver, Fig. 2, the SRP feed centrifuge).

At the beginning, thorium metal rods clad in aluminium ("targets") were dissolved, later thoria rods, as described by Boswell (89). The process used was a simplified THOREX process called INTERIM-23, whereby only U-233 was purified and recovered, thorium being discarded as waste.

Later the THOREX process was used in both plants, U-233 and thorium being both separated and purified (Fig. 3). THOREX is in its principle very similar to PUREX for processing U-Pu fuels.

7.2.2. **Handling and head-end operations for fertile fuels to extract U-233**

In both Hanford and SRP plants, decanning of the aluminium cans was performed before ThO2 dissolution took place. At Hanford, the usual caustic decanning with NaOH was performed, and after rinsing and centrifugation to recycle the thoria fines, dissolution took place. At SRP, decanning was done by low molar HNO3 (ab. 1 M) catalyzed by mercury nitrate, another well-known method.

Due to its great stability, dissolution of Th or ThO2 is not as straightforward as that of U, and especially of UO2: it needs strong nitric acid, aided with some HF, and it takes a rather long time, especially for high temperature sintered compact ThO2. Dissolution can take up to 35 hours.

Dissolution of the thoria powder was done with strong nitric acid (10-12 M), aided by some hydrofluoric ions (0.02 - 0.05 M). To prevent corrosion of the equipment by HF, aluminium nitrate is added as a buffer, another well-known process (0.1 - 0.5 M).

The problem was to avoid settling and "concreting" of the dense thoria powder at the dissolver bottom, blocking the reaction, etc..., in spite of the small powder size (ab. 0.1 mm). Agitation through air-lift circulation was an answer, but excess abrasion by hard thoria powder must be minimized.

Dissolution in this case would take 10-12 hours per batch.

The general head-end schema is given on Figure 4 for SRP.

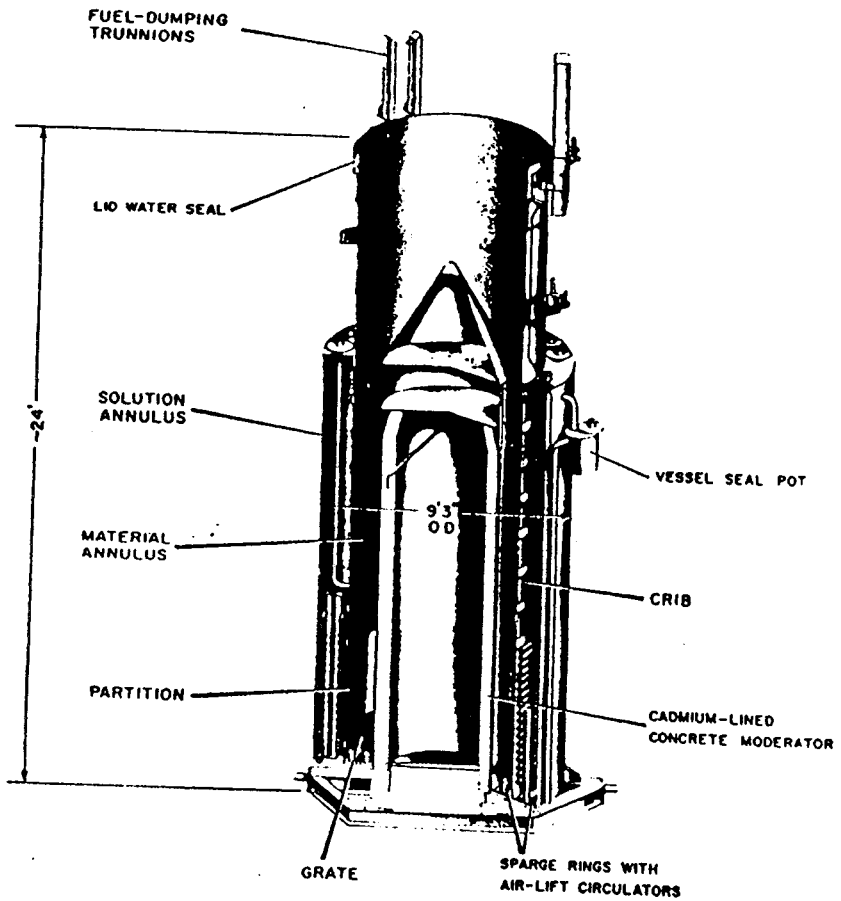


Figure 1: Hanford Purex annular dissolver (119)

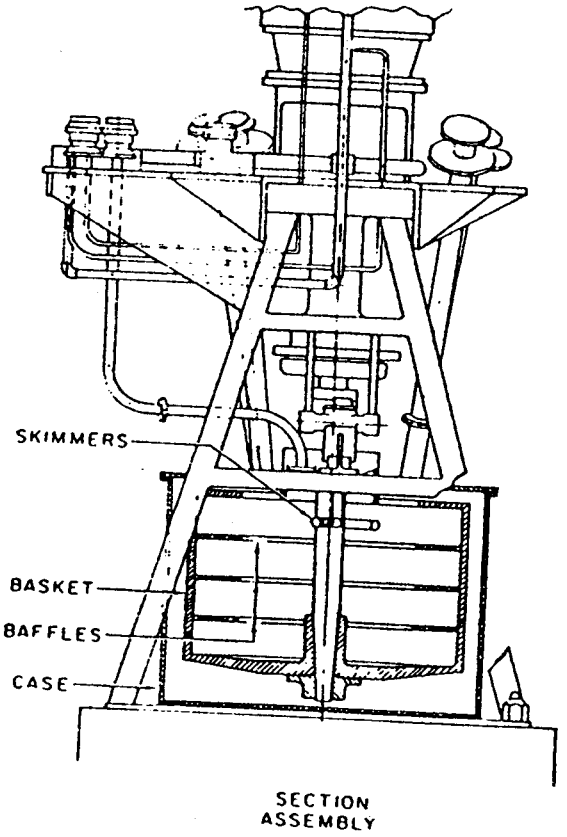
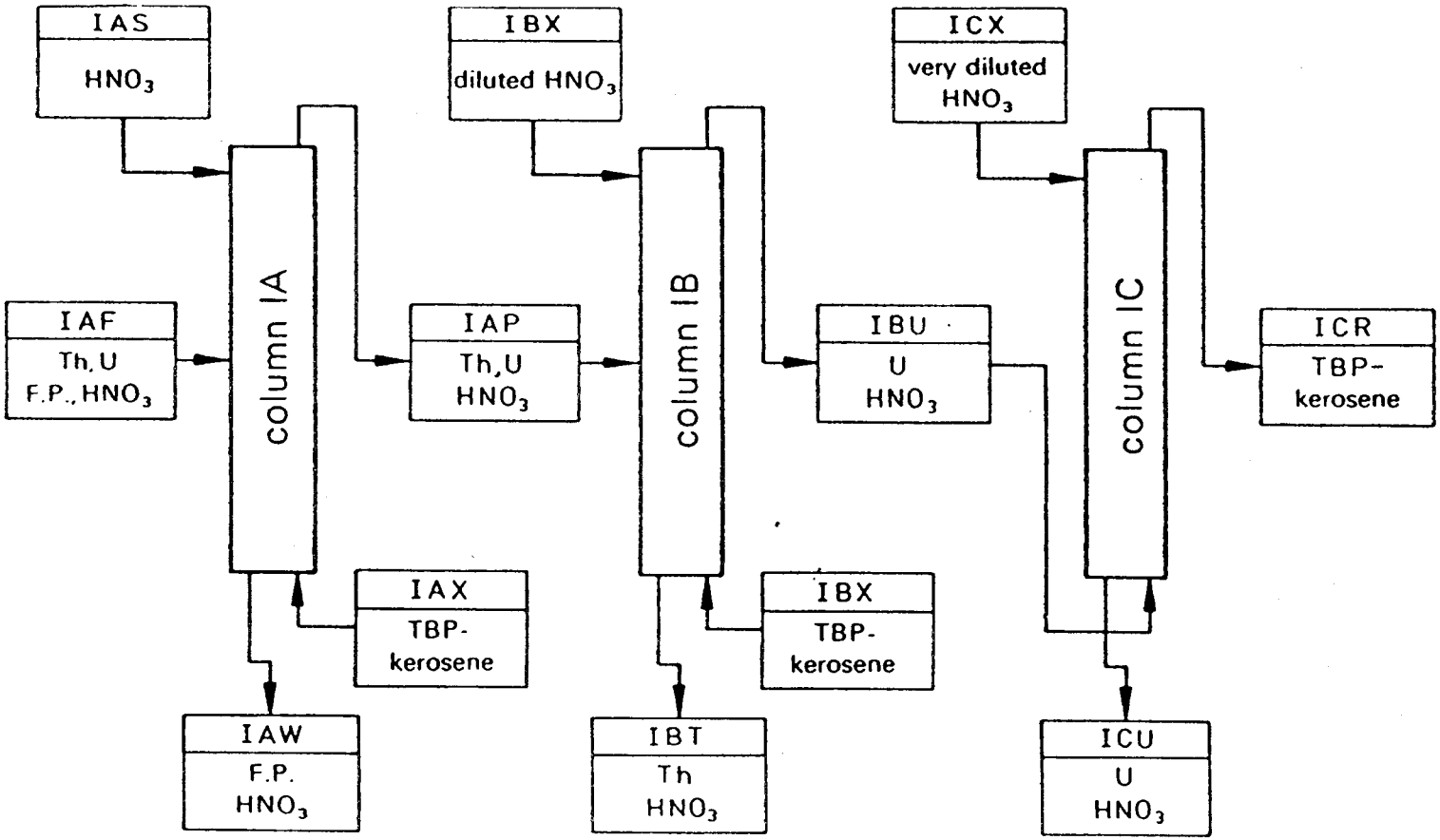


Figure 2: Savannah River Plant centrifuge (119)



Simplified flow diagram of the THOREX process. F.P. = fission products.

Figure 3: Simplified flow diagram of the THOREX process (116)

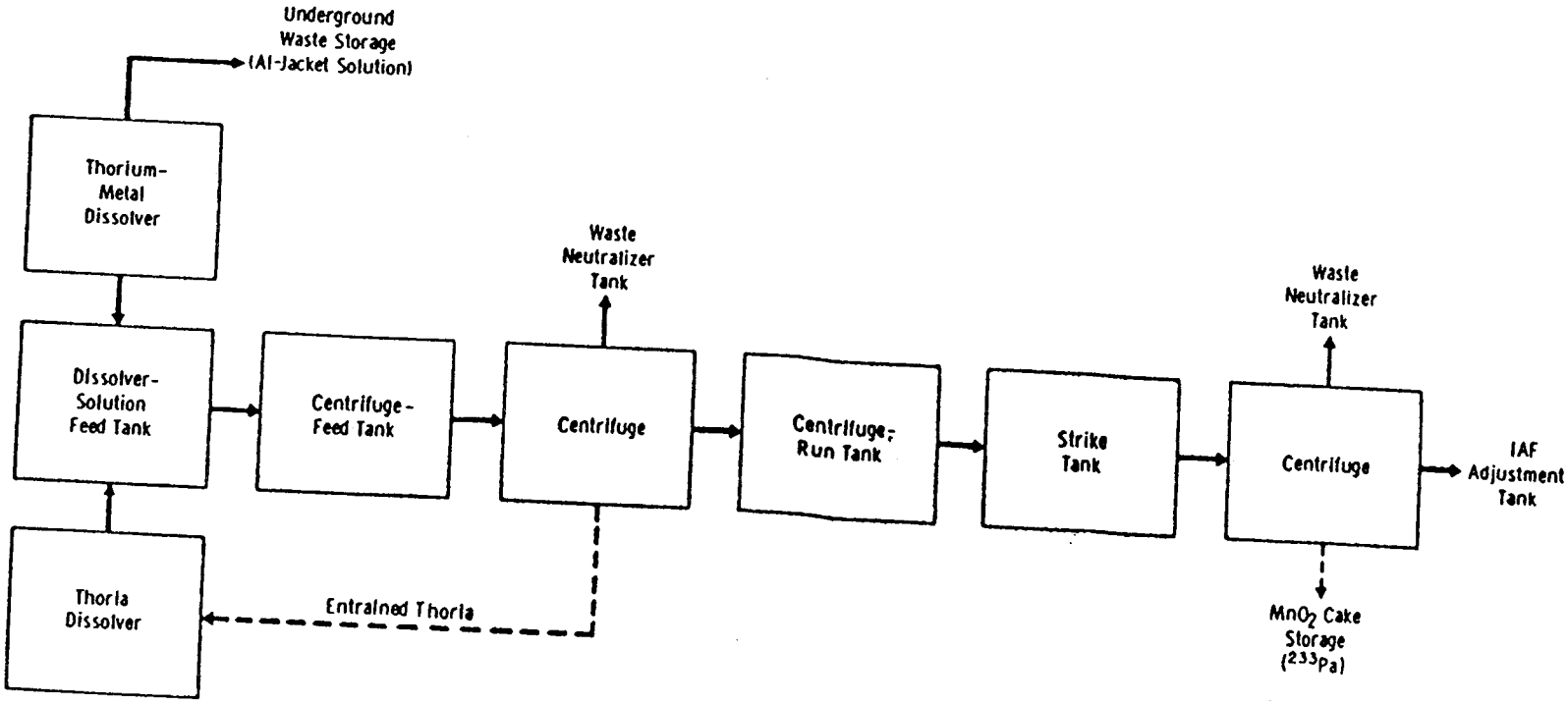


Figure 4: Savannah River Plant thorium process. Head-end flow diagram (119)

Flowsheets showing in more detail the operations of decladding, dissolution, clarification and feed adjustment in the case of Hanford, are shown on Figures 5 and 6 (119). The quality of the products was excellent, due to good previous flushings, to avoid cross-contamination from previous runs with uranium and plutonium.

Similar processes have been used in India on a small scale for Th and ThO2 rods (G.R. Subramanian), (110, 111), and in other countries (England at Dounreay, Canada at Whiteshell, Germany at Jülich and Karlsruhe).

In India, the engineering pilot scale laboratory is soon to be replaced by a~50 ton/year reprocessing plant at IGCAR, Kalpakkam, now under construction.

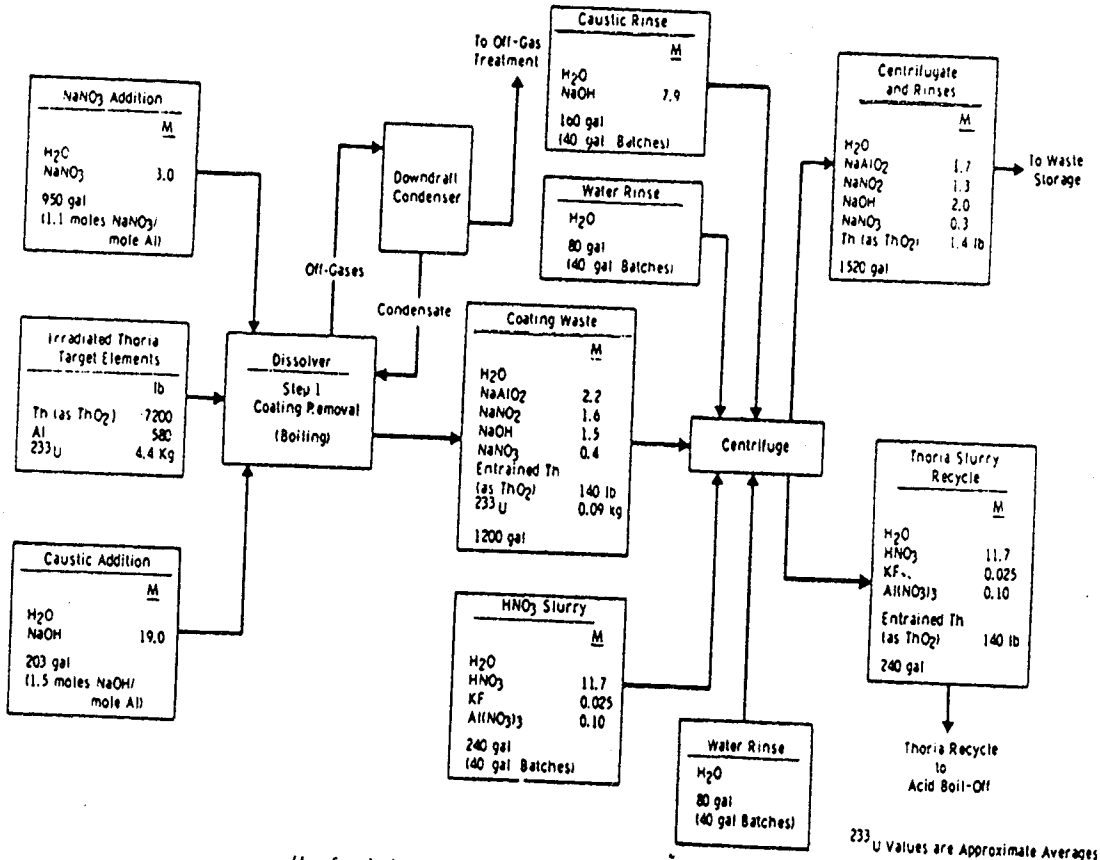
The reprocessing operations in general concerned low irradiated fuel elements. Table 1, for the USA, shows that the U-232 content was not high usually. However, as will be seen, the concentrated thorium nitrate parent products can cause a radiation problem.

As a rule, we can say that a rather "traditional" reprocessing plant can take care of irradiated thoria fuels, even if the dissolution is more difficult and precautions about HF corrosion must be carefully taken, but this is not new. (The Marcoule UP1 plant in France has partly operated with HF addition too).

7.2.3. **Handling and head-end operations for oxide LWR and *oxide or* carbide HTR fuels**

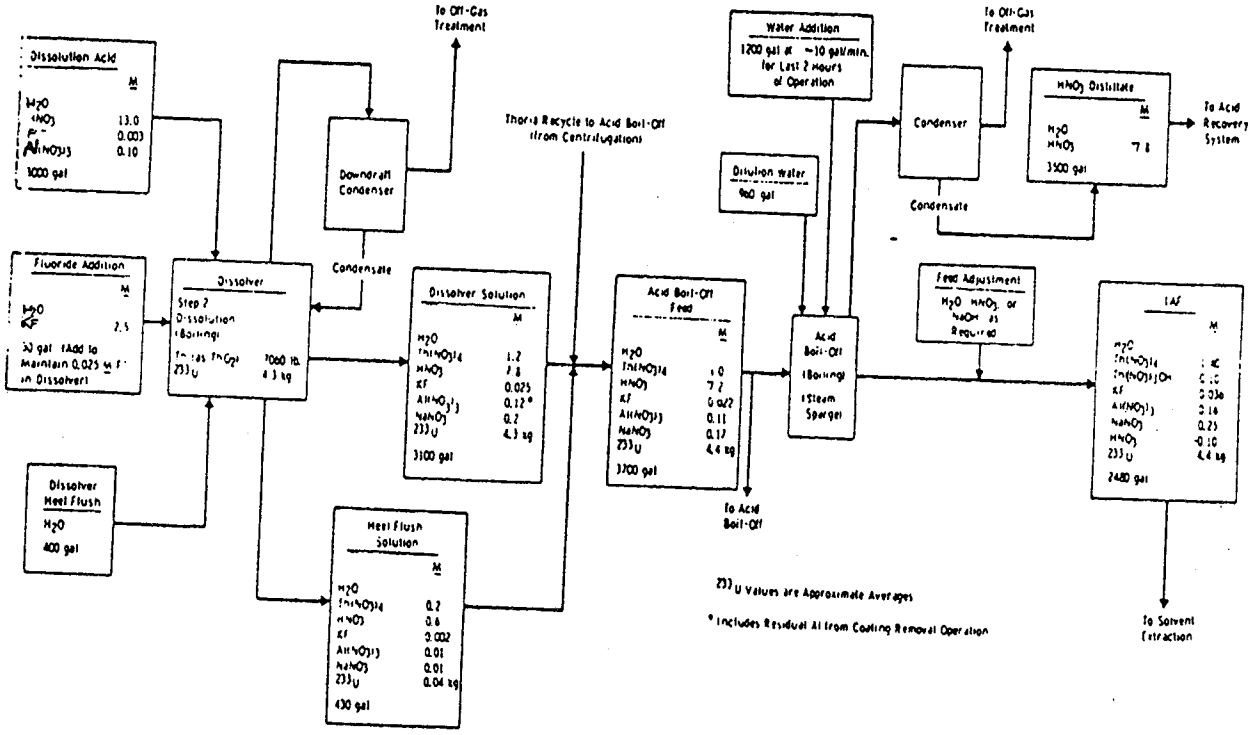
The case of oxide LWR fuels is being discussed among others by E.L. Nicholson (120) at ORNL. The pilot tests made at Oak Ridge were confirmed by "commercial runs" of the PWR Indian Point first ThO2-UO2 core in the West Valley plant, for sizeable irradiation rates (16 000 MWd/t).

The head-end process is shown Figure 7. The PWR fuel bundles clad in zircalloy are sheared in a bundle shear and the resulting fuel + cladding pieces (Fig. 8) are received in the perforated basket of the dissolver for dissolution, as explained in § 7.2.2. above. It was found that, although irradiated UO2-ThO2 dissolves quicker than UO2-ThO2 unirradiated, the dissolution is much slower than for UO2 alone and could take much more than 10 hours. The dissolution duration is anyway much longer than with UO2, and will be longer, as the thorium oxide is more compact and sintered; concentrated nitric acid is needed, with the resulting corrosion problems. Cf. Figs 9 and 10. The dissolution, as previously said, takes



Hanford thorium-process flow sheet. Target decladding.

Figure 5:



Hanford thorium-process flow sheet. Thoria dissolution and feed adjustment.

Figure 6:

(119)

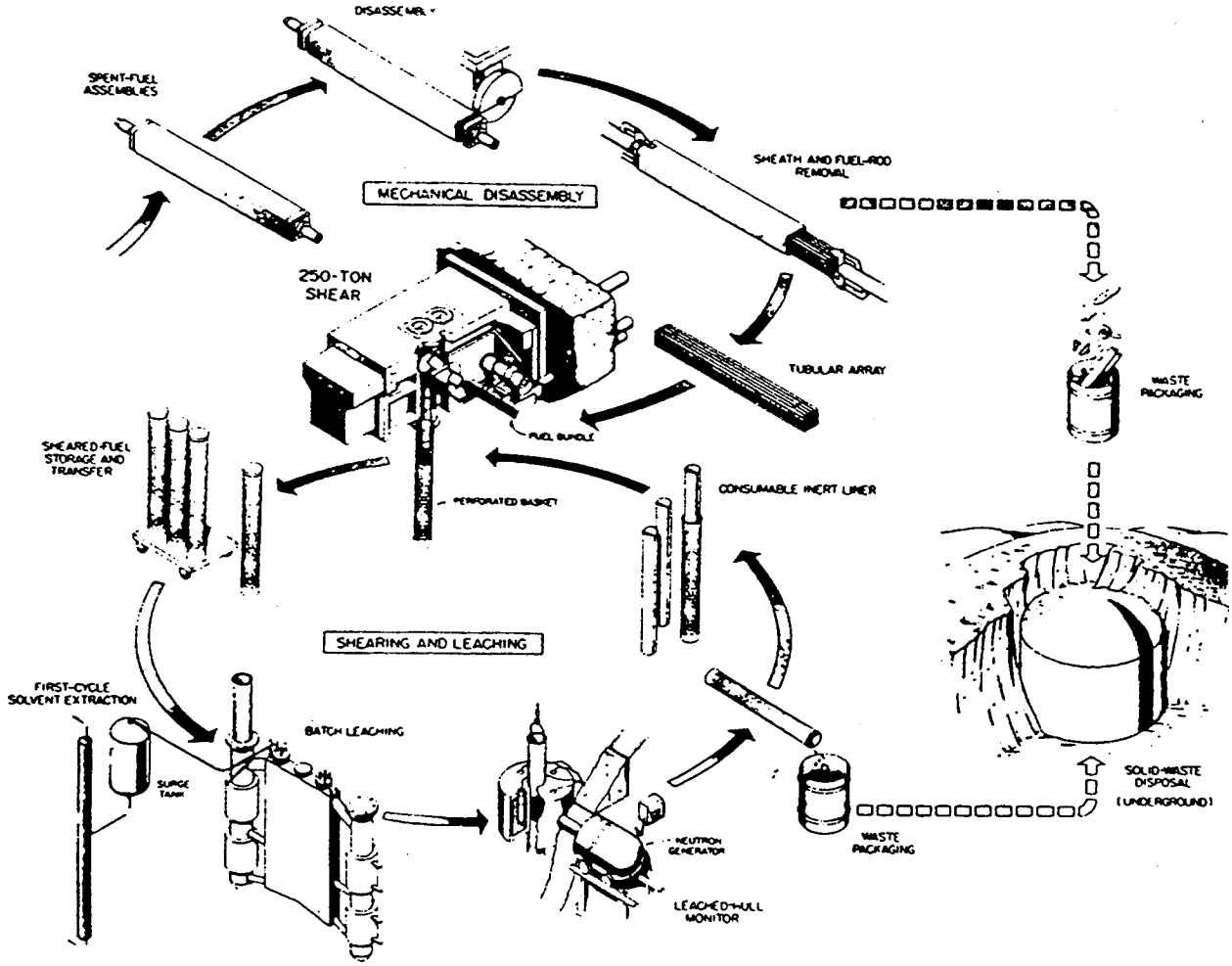


Figure 7: *Shear–leach process for spent reactor fuels.* (NFS) (120)

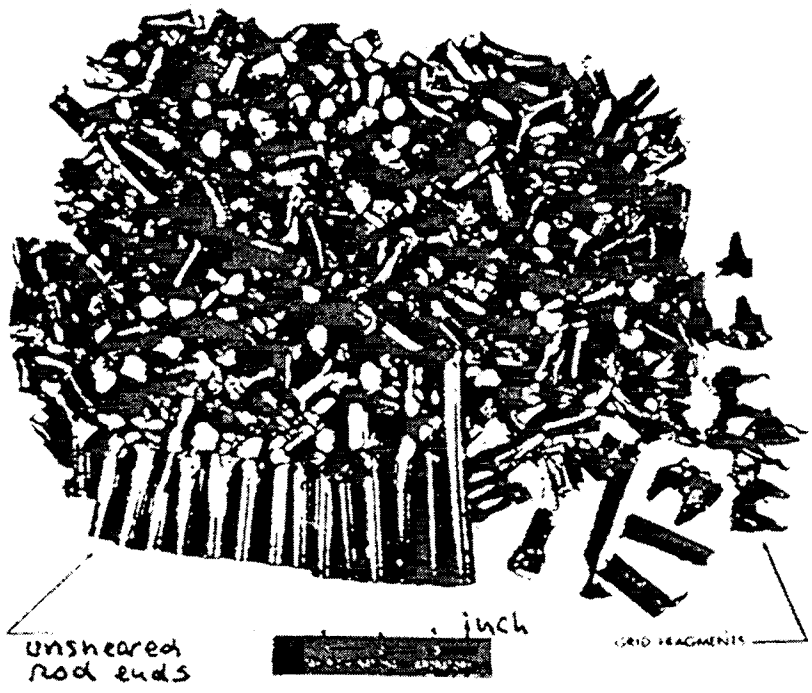


Figure 8: Sheared Elk River fuel bundle

place with a small amount of HF, which itself is buffered by aluminium nitrate. However, some of the zircalloy from the hulls will be dissolved by HF and will be carried into the feed solution to the extractions. Data (120) show that up to 5 % of the Zr could be dissolved, which could about double the Zr from the fission products. This has to be taken into account, although it seems that this extra amount can be taken care of at the extraction step.

The fines from shearing have to be separated by centrifuge or by filtration, but all these steps are similar to those of a usual reprocessing scheme, except for the dissolution time.

The dissolver offgases will contain somewhat more Kr-85, depending on the proportion of U-233, as the yield of Kr-85 by fission is about double that of U-235 (30). Iodine yield is about the same as with U-235.

The case of coated oxide or carbide HTR fuels is quite different and poses problems which have not been totally solved at industrial scale, although many engineering tests have been performed, mainly at Oak Ridge and General Dynamics in the USA and in Germany (Jülich), for their HTR programs. Limited tests have been performed in other laboratories (CNEN, Italy, especially).

Good summaries of these works are reported by C. MERZ (115) (116) and H. Vietzke (112), E.L. Nicholson (120).

The problem with coated oxide, especially carbide fuel, is that it is a very good physical component, but very difficult to process, due mainly to its PyC and impervious SiC coatings, and in part to the unavoidable carbon residue, as the ratio C/HM is around 20 in weight, more in volume, (with a small quantity of activated C-14 in the graphite).

Processing this carbide-coated fuel, to the author of these lines, is a headache, and apart from engineering laboratory scale, has not been applied industrially.

In order to "crack" the coated particles or kernels, two main processes have been proposed and tried :

a) crushing-grinding the kernels and dissolution, with filtration of the residual graphite,

b) crushing the kernels and burning the resulting powder in a fluidized bed.

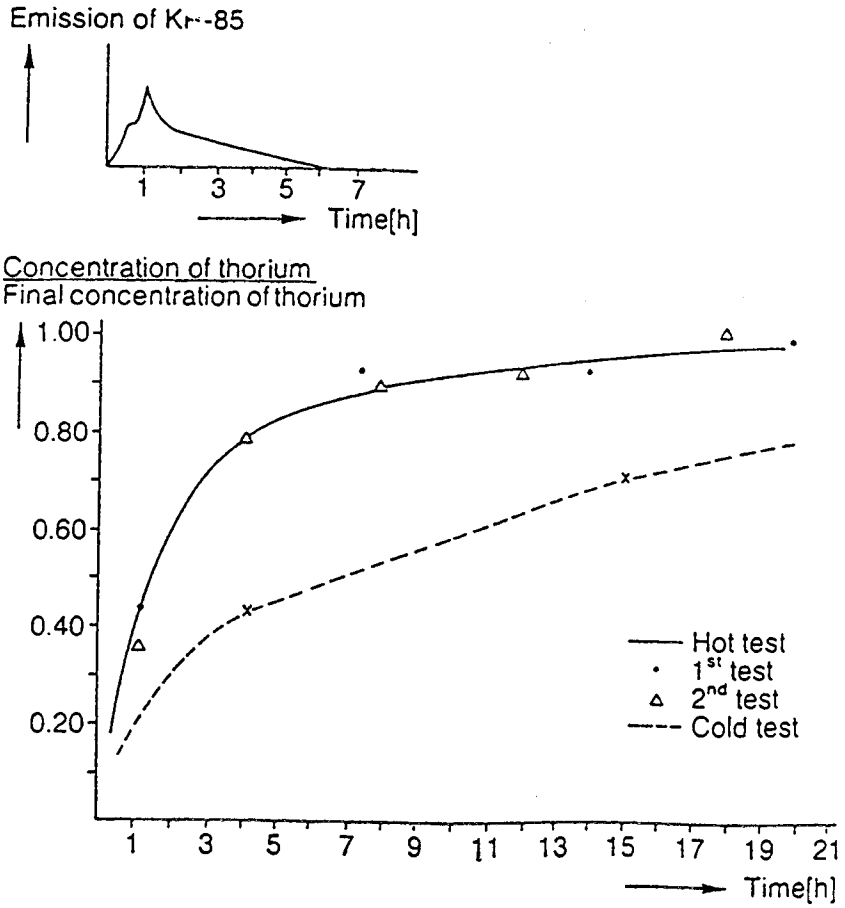


Figure 9: Dissolution of irradiated (Th, 5%U) O_2 in Thorex solution (12)

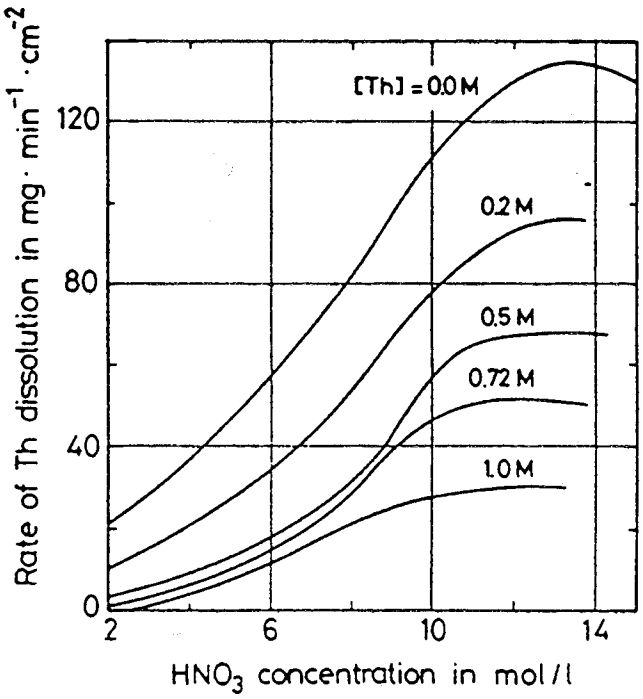


Figure 10: The dissolution as a function of HNO_3 concentration and that of Th already dissolved (116)

This second method seems preferable for the "armored" Triso-coated particles; besides graphite powder, SiC from the intermediate coating is also a solid impediment that has to be taken care of. (Problems in filtering silicate suspensions remain to be solved).

These different steps have been tested at the JUPITER Facility in Jülich, Germany, on the famous "pebbles" of the AVR and THTR reactors. Finally, most of the irradiated pebbles have been stored in casks and transported to the Ahaus site without further processing.

It is rather easy to imagine the problems created by crushing strongly radioactive, hard balls or kernels, with all the fine dust generated, the transport of the resulting dense powders, the possibilities of clogging, the necessary abatement of activated C-14 as CO2, generated in the burner if the powders are roasted, and the volatilization of some of the fission products (Fig. 11), or the filtration of graphite dust if the powders are chemically attacked.

All theses problems, and variants to these main processing lines, are analyzed by E. Merz (116).

Figure 12, right side, enumerates the possible head-end treatments, and the options chosen and experimented at KFA Jülich. The rest of the figure gives an idea of the type of equipment used in that process, prior to dissolution.

Figures 11 and 12 give the flowsheets tested at ORNL for grind/leach processing of Peach Bottom fuel elements, and the burn/leach processings (120).

Figure 13 gives details of a fluidized bed to be used for roasting the crushed kernels, and Figures 14 & 15 show how these recommended head-end processes mesh in with dissolution and the more "traditional" Thorex solvent extraction process. Figure 16 summarizes a most likely overall process.

Direct dissolution of the coated particles has been tested in some laboratories, especially at CNEN/ENEA (Italy) and in India (IGCAR), using electrolytic dissolution. The laboratory tests are promising, although the dissolution times are long and the equipment more complex than usual. It would be important to test such devices on large scale to assess if it works properly. It is probable that, with TRISO type kernels, a preliminary grinding may be necessary.

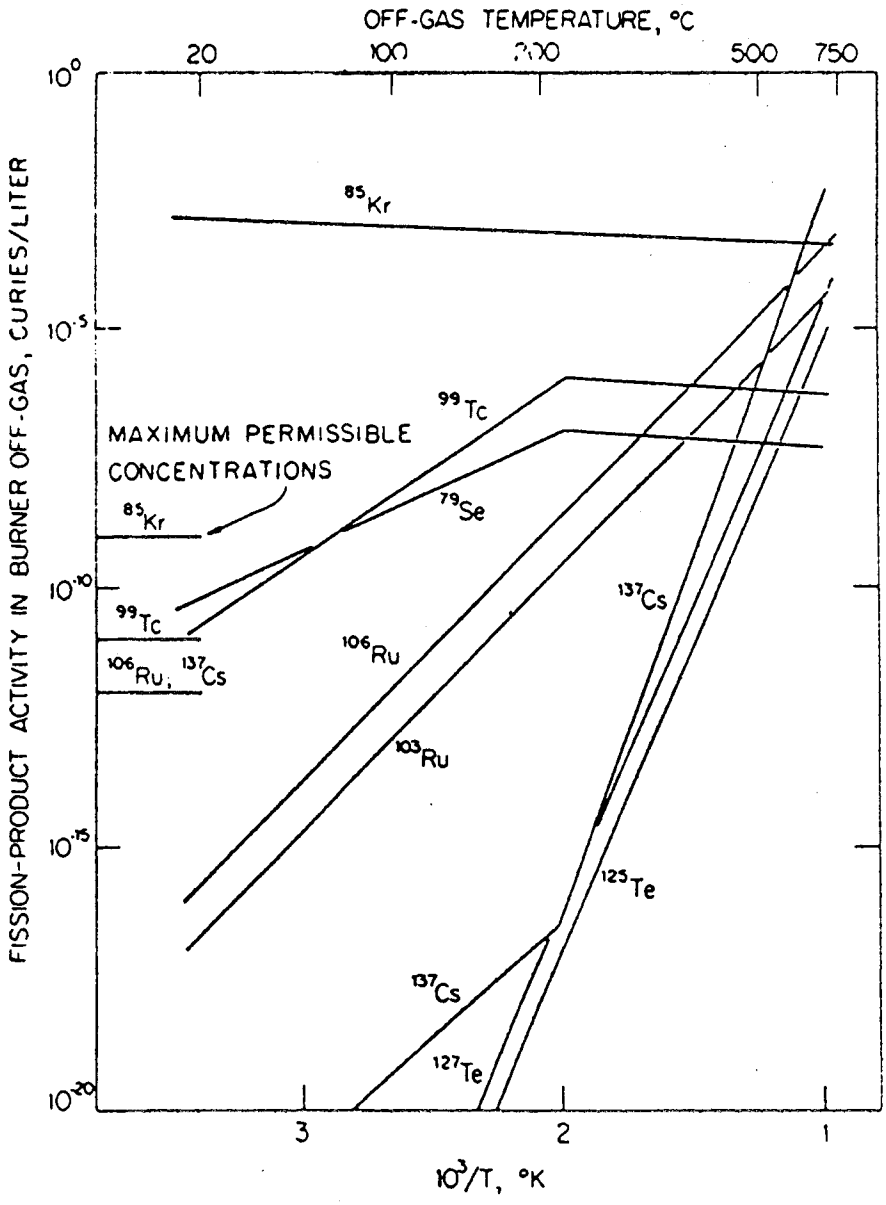


Figure 11: Calculated equilibrium fission product content of fluid-bed burner off-gas (oxygen pressure = 1 atm) (120)

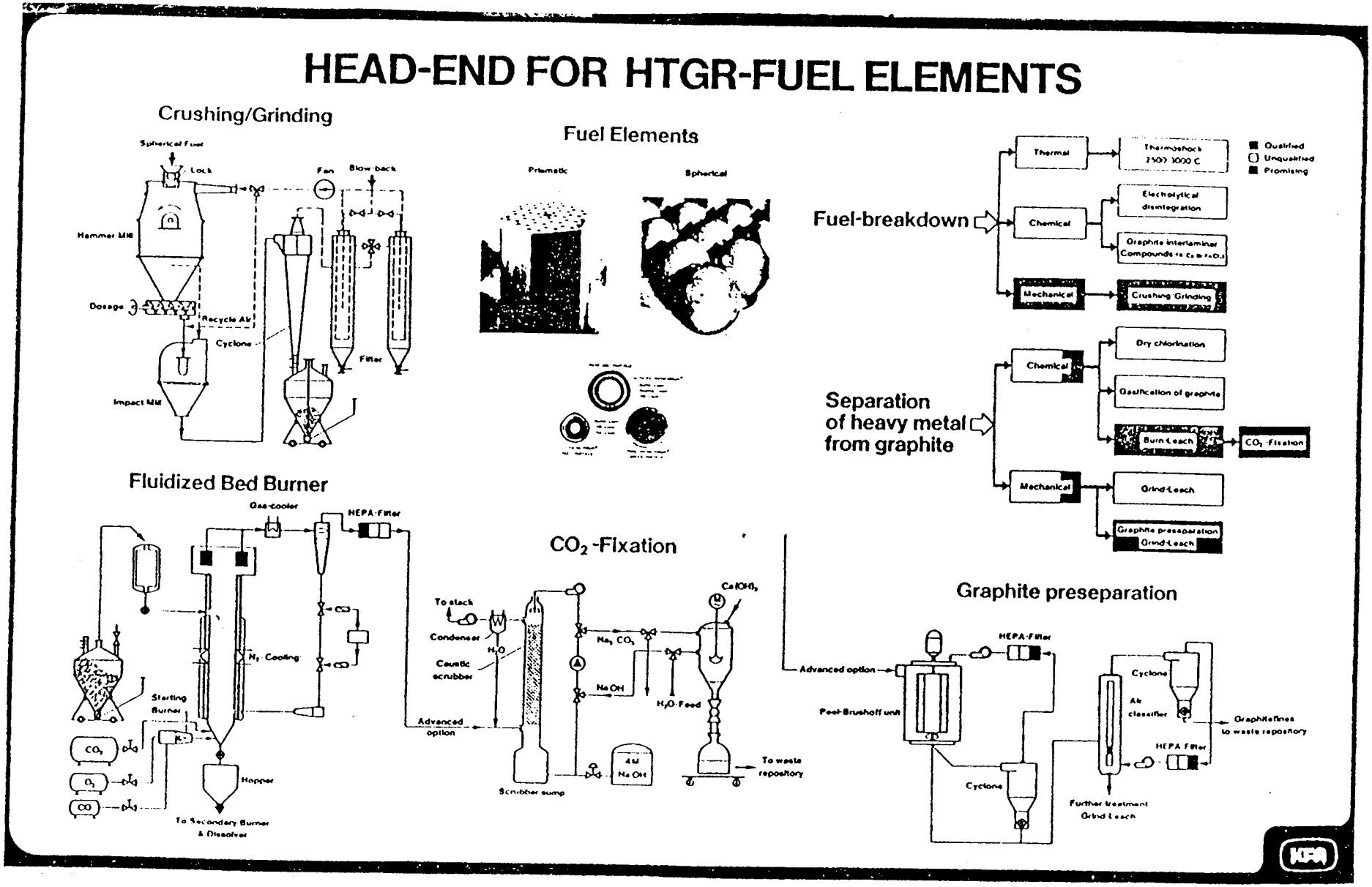


Figure 12: Head-end for HTGR fuel elements

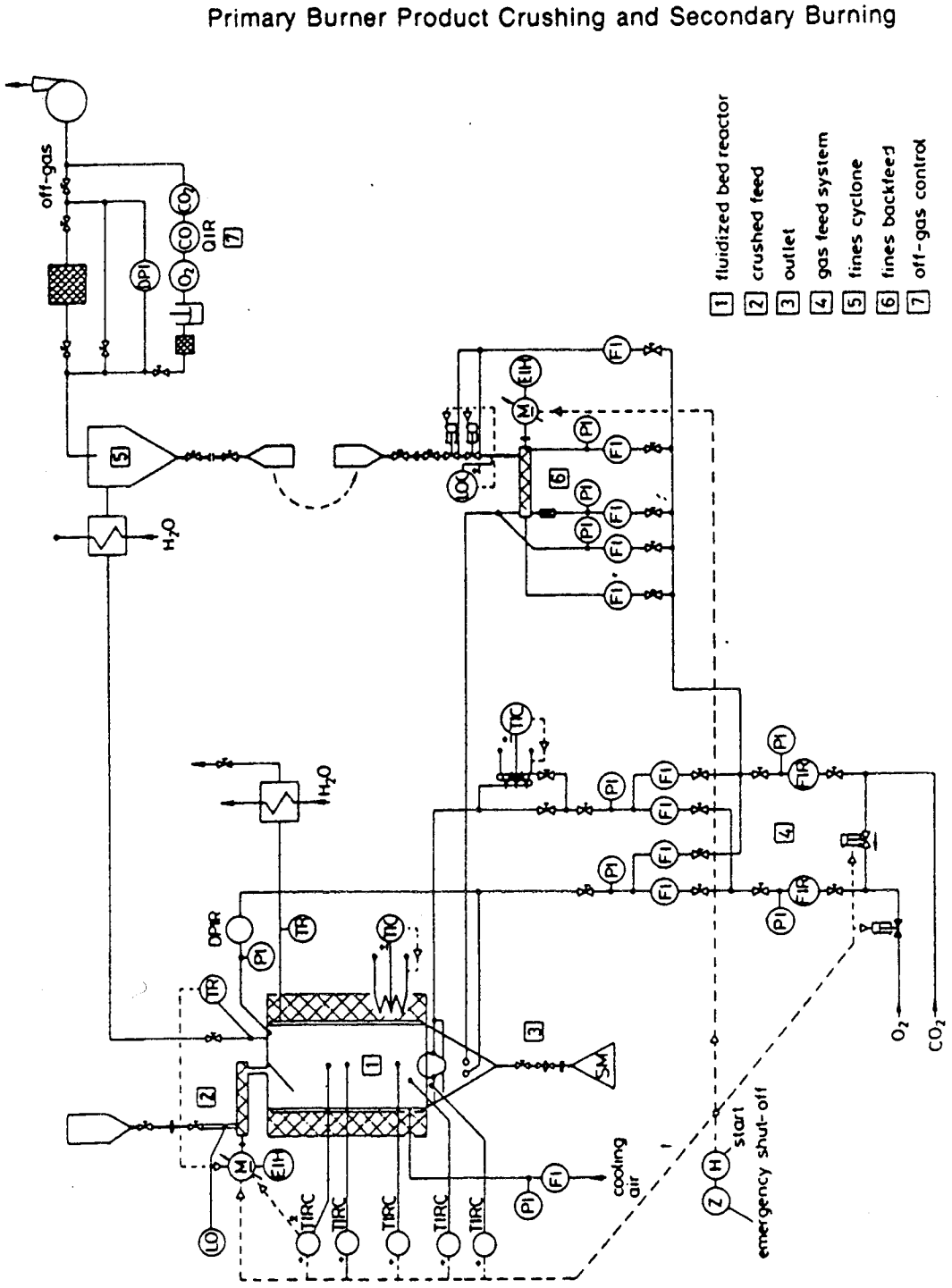


Fig. 13. Fluidized bed system of the "Kernforschungsanlage Jülich", Federal Republic of Germany (85)
(The abbreviations correspond to the DIN Standards.)

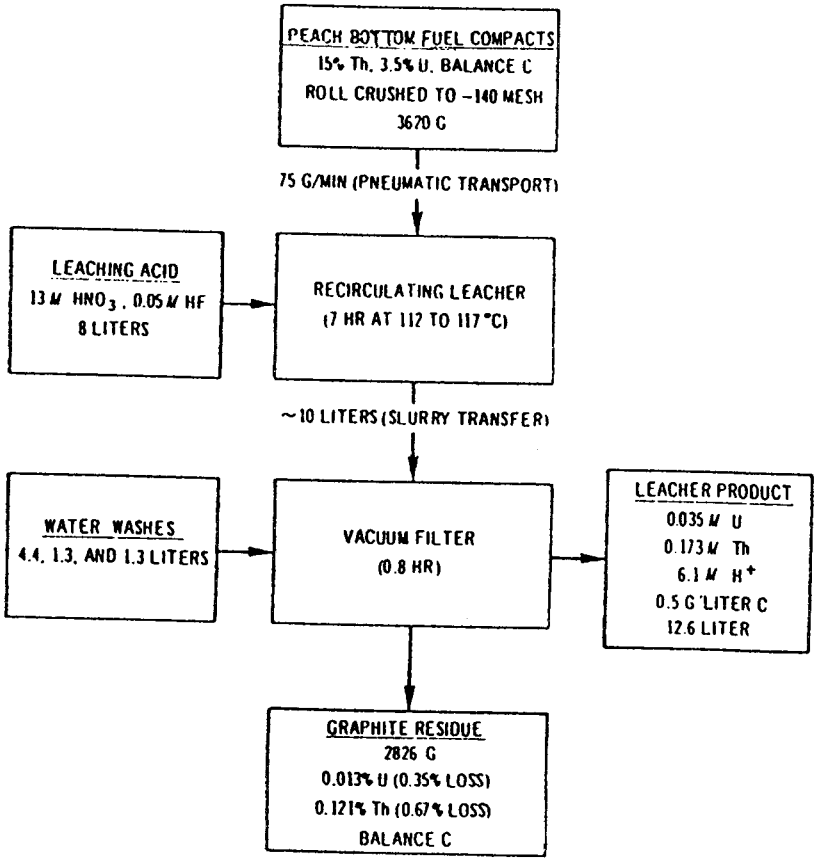


Fig. 14 -Engineering-scale grind-leach processing of Peach Bottom fuel. (120)

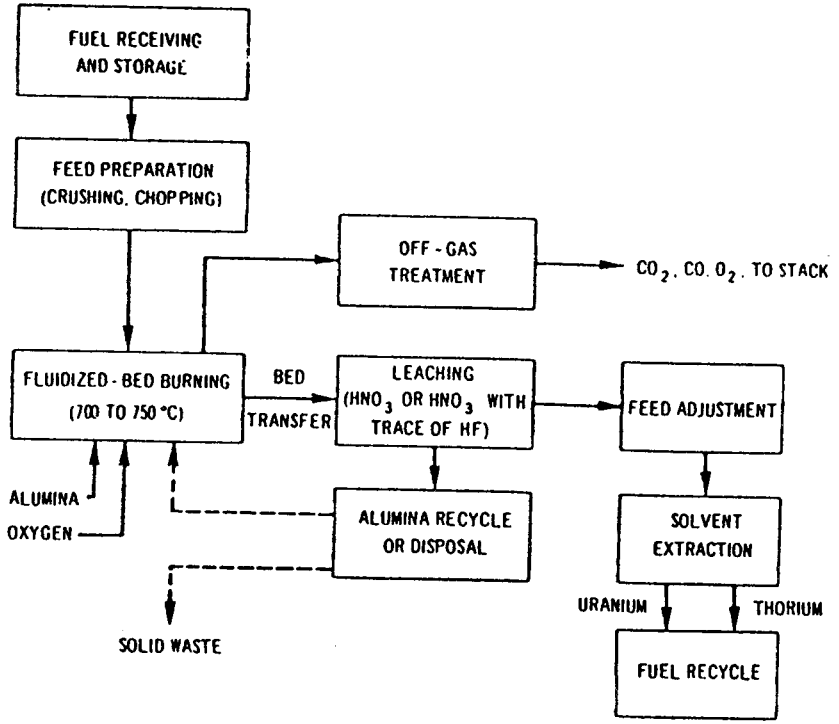


Fig. 15 -Burn-leach process for graphite-base reactor fuels. (120)

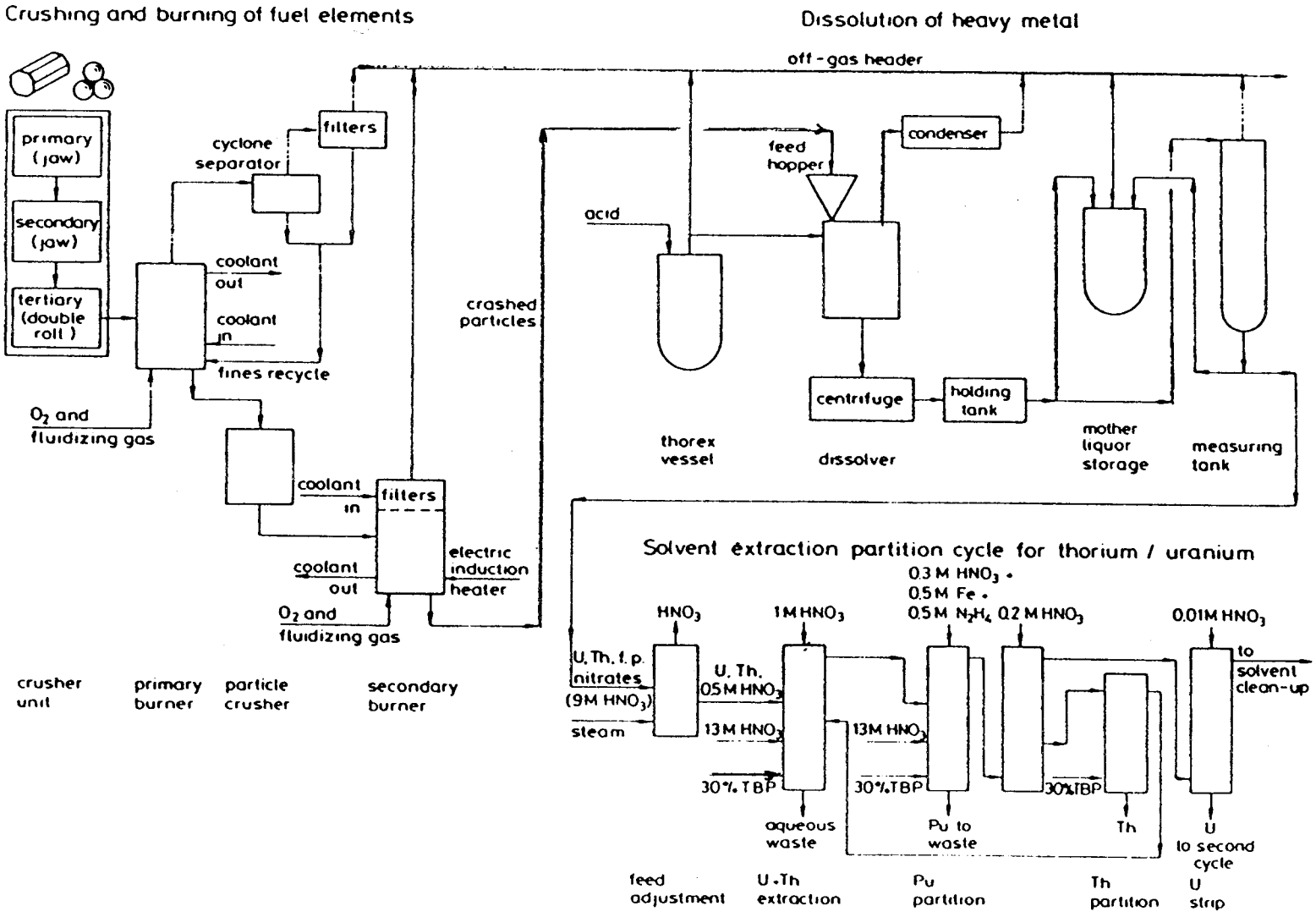


Fig. 16 Flowsheet of the reprocessing of HTR fuel elements (85)

Another possibility is attack of the kernels with hot steam to produce CO and CH_4, but it does not seem to have been tested to a great extent.

Note : the complexity of these pretreatments, and the technological problems associated with them, are one of the reasons why so far the High Temperature Reactor family has been postponed. (There are other political and commercial reasons, one being that the main proponent of commercial HTRs, General Dynamics, was acquired by the oil Company Gulf).

Broadly speaking, the kernel fuel is so good that it cannot be reprocessed simply by the conventional reprocessing methods. Maybe that a dry fluoride-volatilization process in fluidized beds could give simpler results in that case, but a preliminary grinding would seem unavoidable (116). It is possible that one has gone somewhat too far in one direction and it might be valuable to think the whole matter anew to find balanced solutions between "the best fuel" and "the best reactor" with a simpler fuel cycle, somewhat like what has been imagined for the famous PWR, which is a good compromise between neutronics, materials, feasible technology and economics.

The coated kernels, today, are, seemingly, the typical "once-through" fuel (at least for many years to come). They could be reserved to high temperature "burners" working on long (many years) once-through cycles, delivering high temperature gas. On the other hand, the "breeders" thermal or fast, should have fuel elements directly amenable to a simple reprocessing process, i.e. of the traditional oxide-in-clad type, water or liquid metal cooled.

In any case, the decontamination factors necessary, in view of the U-232 and Th-228 daughter products radioactivity, need not be very high anyway, the recycled fuel having to be remotely handled and refabricated; this poses new handling problems for transport and at reactor site, by the way, and in that context the AVR-type pebbles would be easier to handle than rod-type fuel elements.

7.3. Separation and Purification Operations

We assume that the different types of fuels have been, one way or the other, dissolved to obtain feed solutions consisting of clear (centrifuged or filtered) thorium and uranium nitrates in nitric acid solution. It is now necessary to separate thorium and uranium from the fission products and troublesome minor actinides.

For this, the traditional solvent extraction system using TBP in a diluent as an extractant, is the preferred way for which a sizeable experience has been obtained already, especially in the USA (cf. Table 1), using the so-called THOREX process.

Contrary to the PUREX process whereby plutonium is reduced to its III valency to be separated from uranyl solutions, thorium is constantly and naturally in this IV valency and this, to some extent, simplifies the flowsheet. Typical distribution coefficients for thorium, uranium and some fission products, between solvent and aqueous phase, are given on Figure 17. THOREX normally uses 30 % TBP in diluent as an extractant.

7.3.1. **The THOREX flowsheet** was originally developed at Knolls Atomic Power Laboratory (KAPL) and Oak Ridge National Laboratory (ORNL). It first used aluminium nitrate as a salting agent to enhance the distribution coefficients of uranyl and thorium nitrates, and, the so-called original "acid-deficient process" used an acid deficient feed of about - 0.15 mol/l HNO3 with 1.1 mol/l Th(NO3)4. As the solution from the dissolver was very acid (about 7-8 M), a deacidification step was needed during the feed adjustment step, usually by steam stripping to remain at low temperatures and avoid side effects (i.e. possibility of explosions, ruthenium peroxides, volatilization, corrosion, etc...).

Later it appeared, in the early 1960s, that not much was gained (mainly in zirconium concentration) by such acid-deficient feed, and that the salting out effect could be obtained readily with introduction of stronger nitric acid at the proper scrub stages. These two facts lead to the "acid-Thorex" process which is now recommended and is more convenient by avoiding too much salt in the waste raffinates to be later concentrated and solidified.

Important tonnages of low-irradiated, but short-cooled thorium metal rods or ThO2, aluminium-clad fuel elements, have been processed in the large defence canyon-type reprocessing plants of Savannah River (SRP, mixer-settlers, and

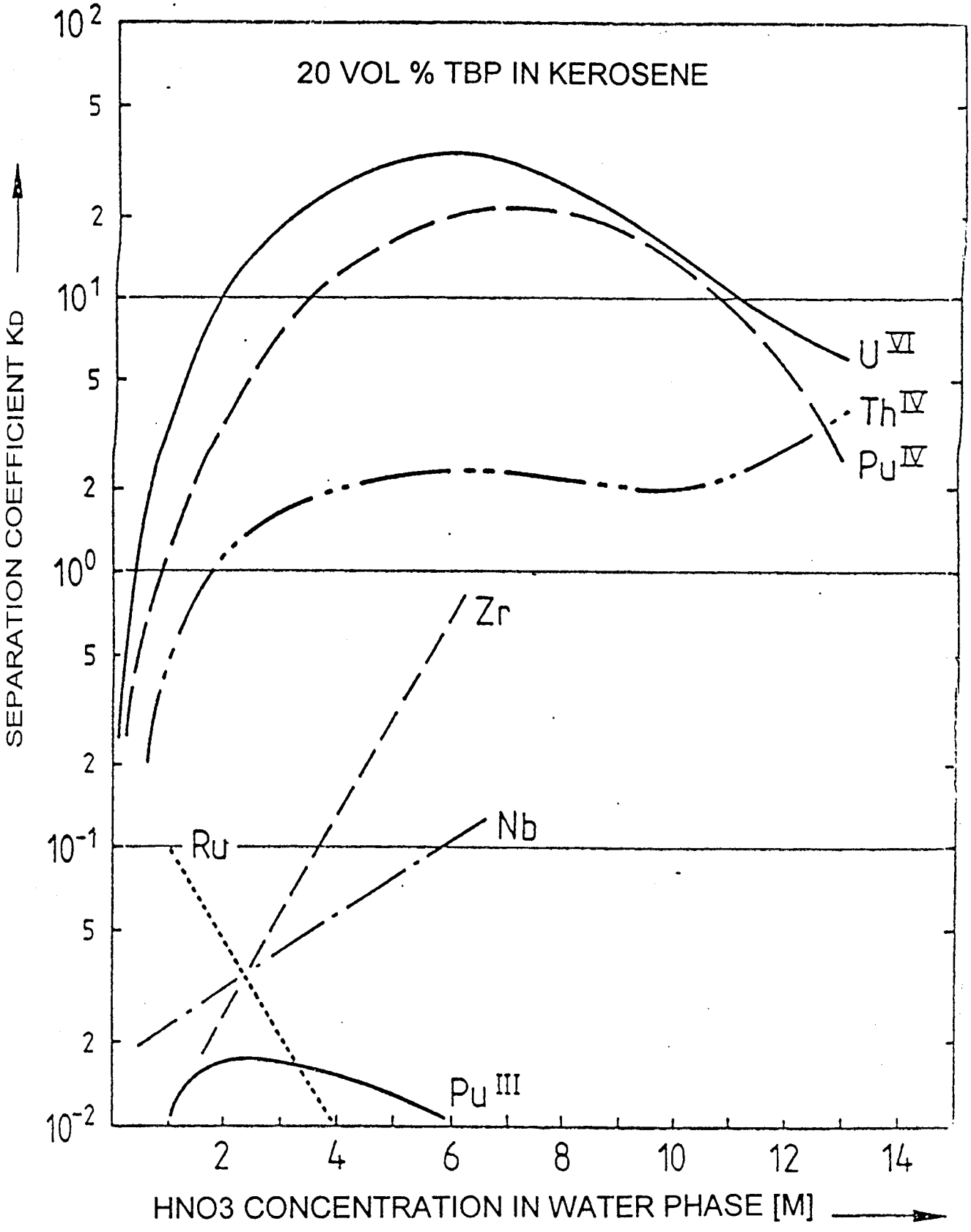


Fig. 17 : Separation coefficents of Thorium, Uranium and Plutonium, as well as of the fission products Zr, Ru and Nb with TBP 20 vol % in Kerosene, as a function of HNO3 concentration. (115)

Hanford, pulse columns), for recovery of U-233, and secondarily thorium, with capacities of the order of 1 ton thorium per day. The plants needed thorough flushing to avoid cross-contamination from U-238, U-235 and Pu for which they were designed. In spite of some adjustments, the results reported were satisfactory. (Fig .18). Detailed flowsheets can be found in Ref. 119. In the short-cooled fuel elements (called "targets" at Hanford and SRP), Pa-233 (27 days) present in mainly polymeric form was precipitated before extractions by MnO2 and centrifuged and allowed to decay into U-233. Other processes for Pa-233 call for sorption on silicagel or vycor glass beads, and elution by oxide acid.

Today, the normal procedure would be to let Pa-233 decay by allowing a 2-3 year cooling time for the spent fuel as is usually the case.

Some of the equipment used in the Hanford and SRP plants are shown in Figs. 19 and 20.

The THOREX process has been piloted in the THOREX plant at Oak Ridge, hot cells reconverted from a previous reprocessing plant, containing pulse columns. Extensive experience has been obtained with this plant, intended to reprocess higher burn-up fuels such as the Indian Point PWR fuel and Elk River BWR fuels, after a "chop and leach" head-end (120).

THOREX Process has been used and refined in Europe principally at the Rotondella, Italy pilot plant, in the frame of the DRAGON Project. Cf. Candelieri (114), Orseningo (153), and in Germany at Jülich for the JUPITER pilot plant (Merz, 116) with a flowsheet tested by Hoechst in the 1970s at laboratory scale (1 kg per day) on irradiated fuel elements (12) up to 54 000 MWd/ton.

7.3.2. **THOREX process applications**

A number of cases could arise depending on the reactor type and the stage in the nuclear programme development, being understood that to begin with, a U-235 or Pu "driver fuel" has to start any nuclear reaction involving thorium.:

1) Breeding U-233 in thorium rods in a LWR or FBR unit : in that case U-238/U-235.Pu oxide fuel is separated from Th/U-233 oxide fuel and by reprocessing, up to 1 % U-235 with some U-233 will be separated from thorium (some parallel with Pu separation from uranium base fuel can be made).

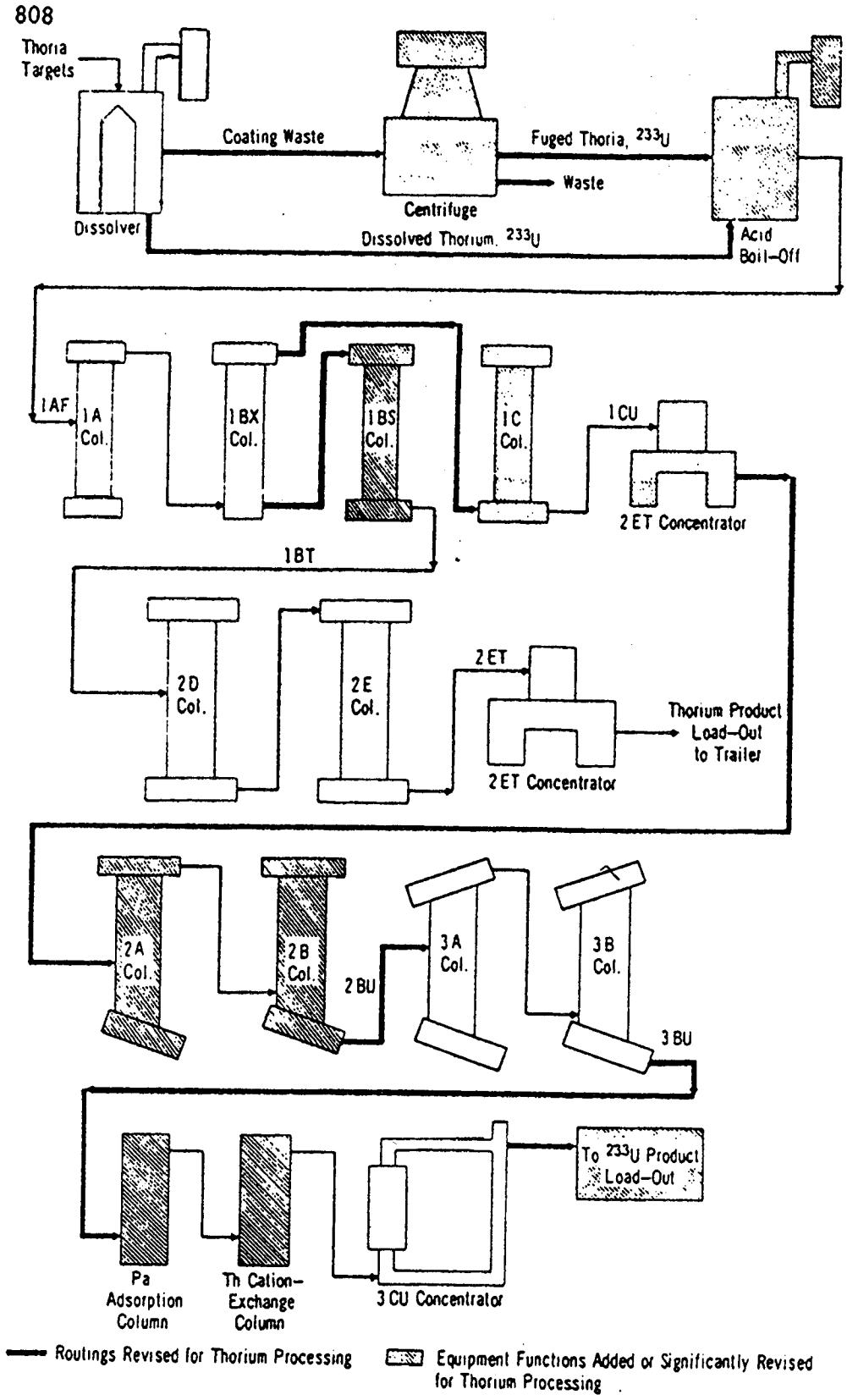


Figure 18: Hanford thorium process. Simplified flow diagram (119)

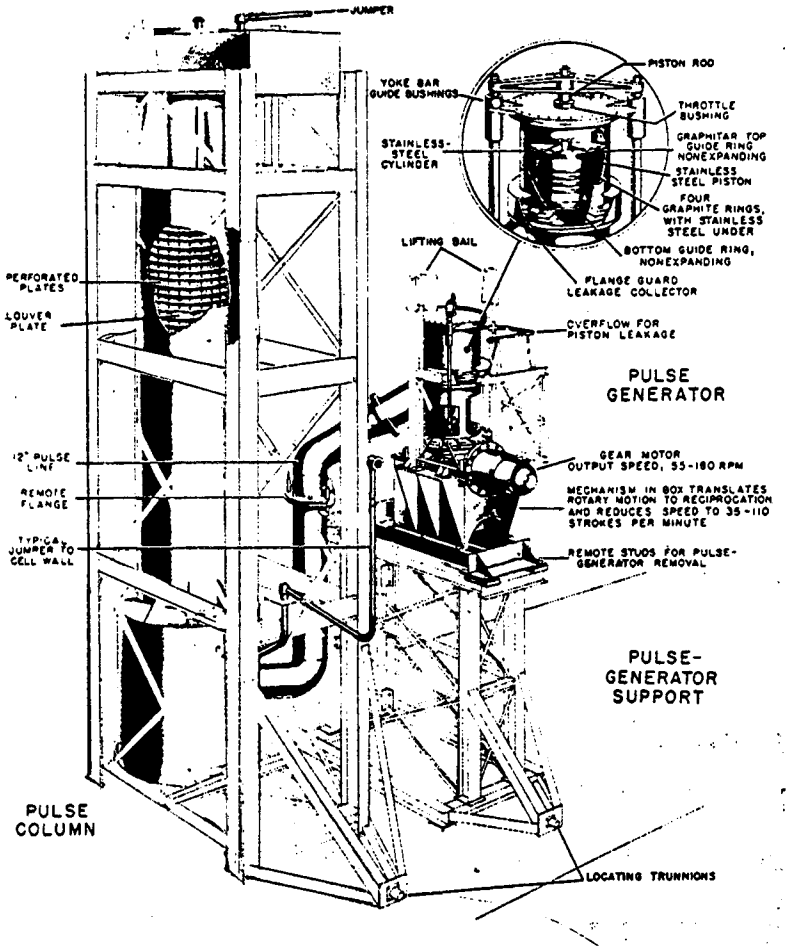


Figure 19: Hanford Purex pulse column (119)

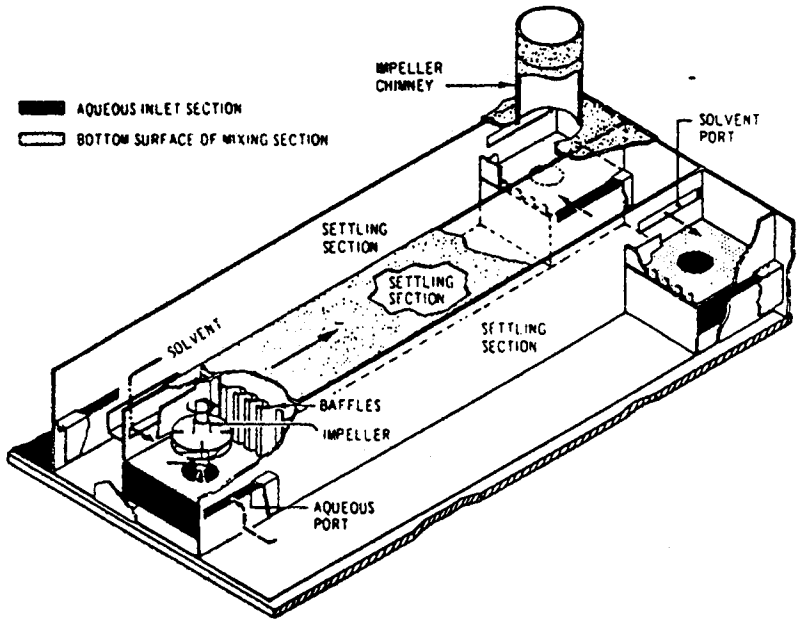


Figure 20: Savannah River plant mixer-settler (119)

2) Operating a Th/U-233 reactor (LWR or HTR or HWR) when sufficient U-233 has been bred : there, U-233 concentrations in the thorium could be from a few % to maybe 15 %.

3) Reactor operating on a mixed cycle, enriched U/Th or Pu/Th (this last one permitting to burn weapons' Pu) : the spent fuel will contain a complex mixture of maybe 50 % U, 50 % Th with Pu isotopes and U-233 in one case; in the other case, some percents of Pu isotopes, some U-233 and thorium.

It seems that the second case would be preferable from a reprocessing standpoint, as it permits to separate the chemical species Th, Pu and U-233 which could be recycled in a less complex manner possibly, as if a mixture of U-238, U-235, U-233 was obtained in the first case.

These examples show that a Thorex plant should be designed, if possible, for a variety of uranium contents, from 1 % to 50 %.

Separation of U, Th, Pu has been done at AECL Whiteshell on a small scale. It should be possible to extract Th IV from U VI-Pu IV, and Pu III from U VI later on, after Pu reduction. See also a different flowsheet on Fig. 16.

7.3.3. **Characteristics of Th/U separation by solvent extractions** (12) (115)

Today's knowledge permits to say that Th/U fuels can be reprocessed as well as U/Pu fuels (115). However, experience with Th/U fuels is much more limited, and operating conditions may have to be carefully adjusted to avoid a number of pitfalls. We should recall that such pitfalls have been encountered also at the beginning of the reprocessing of highly irradiated U/Pu fuels in the 1950s and 1960s, and have been avoided since by refining the reprocessing processes to achieve reliable industrial reprocessing. We have no doubt, for our part, that *the* same would be true for U/Th fuels, at least when we speak of "conventional" fuel elements with rod bundles. Reprocessing of HTR "compacts" in a graphite matrix certainly needs much development, or radical changes in the fuel design, to permit a sufficiently economical fuel cycle.

We will mention here a number of "pitfalls" which must be avoided when reprocessing Th/U fuels in the solvent extraction steps (12 (115):

- Thorium does not extract as readily as U VI or Pu IV in low or medium nitric acid concentrations. Thorium will be the limiting factor in a codecontamination U + Th cycle;

- A strong salting-out agent will be necessary for a satisfactory thorium extraction by solvent; this at some cost of lesser decontamination in a few fission products;

- It is not possible to play on the valencies to extract thorium. The only parameter is the concentration of nitrate ions present. The physical process is more difficult (more extraction stages) but the chemical process is simplified.

- A too high thorium concentration in the feed solution can result in a too high charge in the solvent phase, leading to a second solvent heavy phase with Th IV ("third phase"), so that practically the TBP should not be loaded to more than 30 % of its theoretical heavy metal capacity (ab. 35 g Th/l), and a maximum 1 M thorium in the feed is recommended, with about 1 M HNO3 ("Acid Thorex" process).

- A too low acid concentration in the extraction cycle may lead to Zr, Th precipitates. Higher acidities, on the other hand, especially with highly irradiated fuels, will cause TPB degradation into DBP and precipitation of DBP-heavy metal complexes, and a lower Zr decontamination.

It is recommended to use pulsed columns or fast extractors to avoid too much TBP degradation. The solvent must be thoroughly cleaned on line.

An organic-continuous, slightly heated extraction column, will be more efficient for the extractions and decontamination, at the cost of a slightly higher TBP degradation.

Conversely, the U/Th separation column will work better in aqueous continuous phase. There, one must be watchful for a uranium reflux causing high uranium nitrate concentrations with the risk of criticality. The thorium partitioning flow should not be less acid than 0.2 M HNO3 to prevent crud formation with thorium, and too high a uranium reflux.

Uranium stripping will take place in a third column with weak acid (0.01 M), preferably in a water continuous phase. Due to the weak acidity, any thorium traces left in uranium will create a DBP-Th precipitation. Hence U/Th separation has to be good in the second column. Typical distribution profiles for relevant constituents in the different extraction and partitioning steps are given in Figs. 21 and 22. If necessary, a second purification cycle for both thorium and uranium streams can be installed.

Experience shows that decontamination factors of up to 10^7 for uranium and 10^6 for thorium can be obtained. Uranium and thorium losses are less than 0.1%. The recommended flowsheet for high irradiated fuels is thus given in Table 2 and schematized on Fig. 23.

NOTE : A single codecontamination/partitioning/stripping cycle can result in about 10^3 decontamination factor for both thorium and uranium. Considering the residual activities of the purified streams due respectively to daughter products of Th-228 and U-233, this seems sufficient, as fuel refabrication will have to be carried out in hot cells anyway.

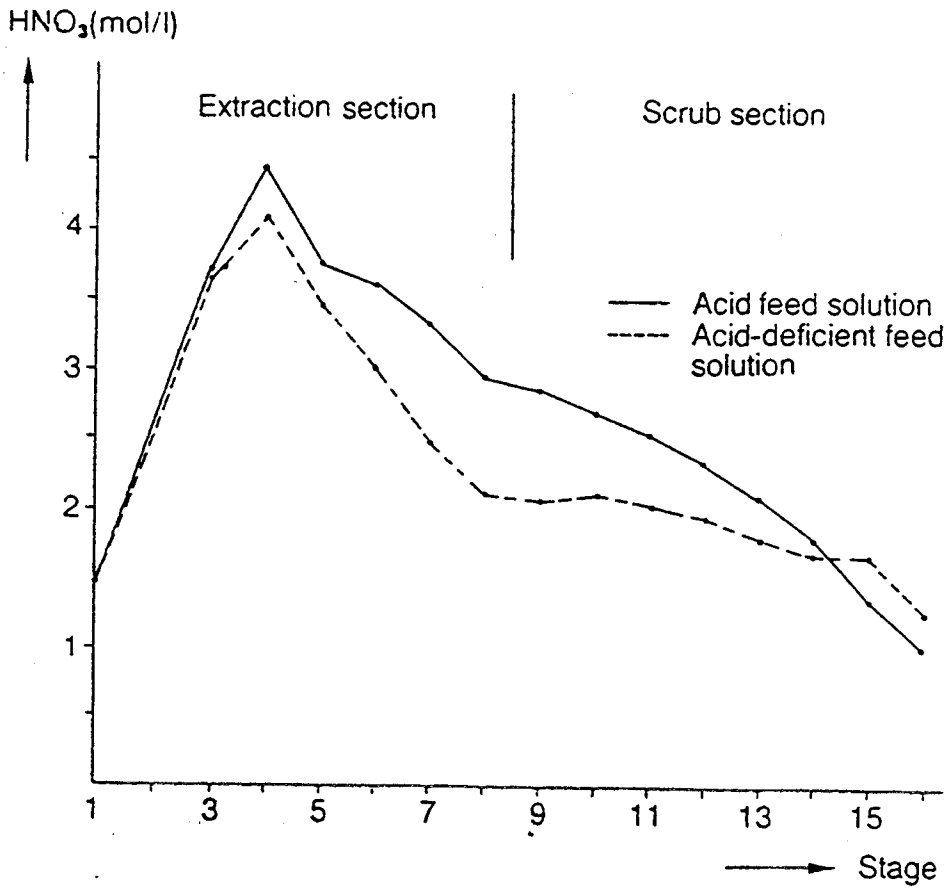


Figure 21A: HNO3 profile in Thorex process extraction (mixer-settler) (12)

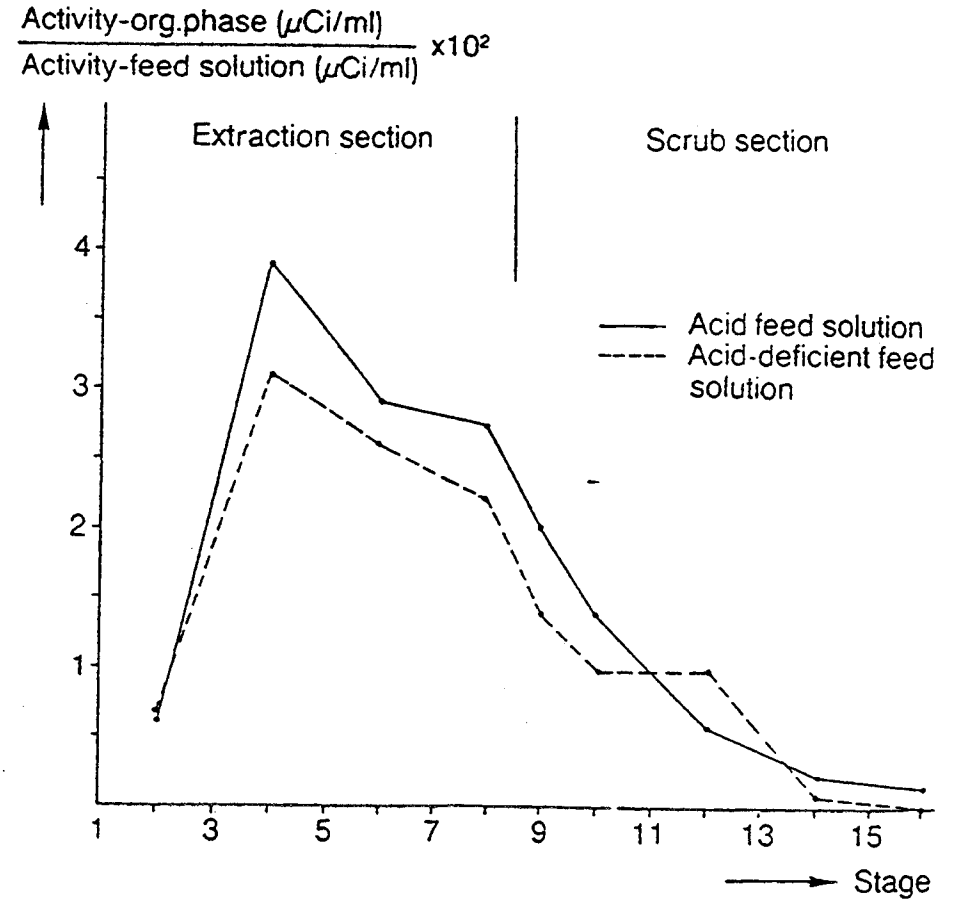


Figure 21B: Zr-95 profile in Thorex process extraction (mixer-settler) (12)

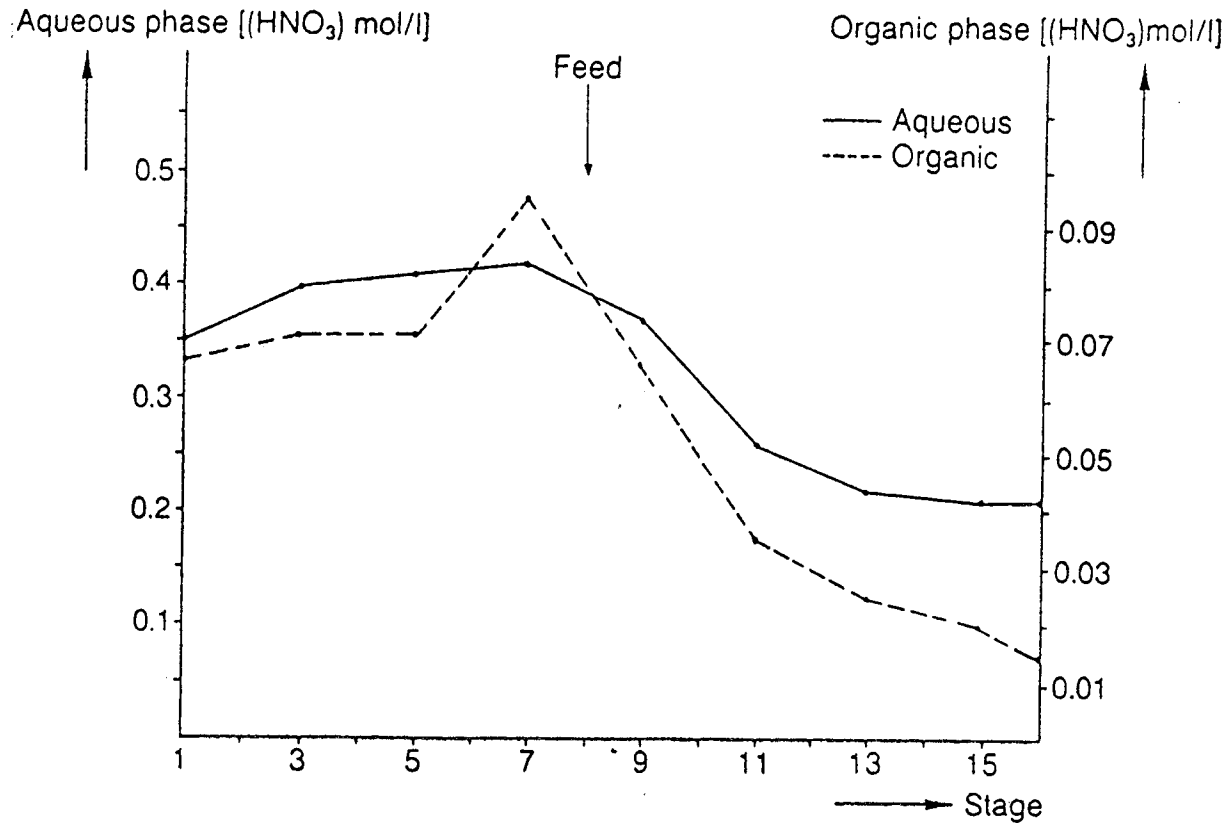


Figure 22A: HNO3 profile in Thorex process: partitioning (mixer-settler) (12)

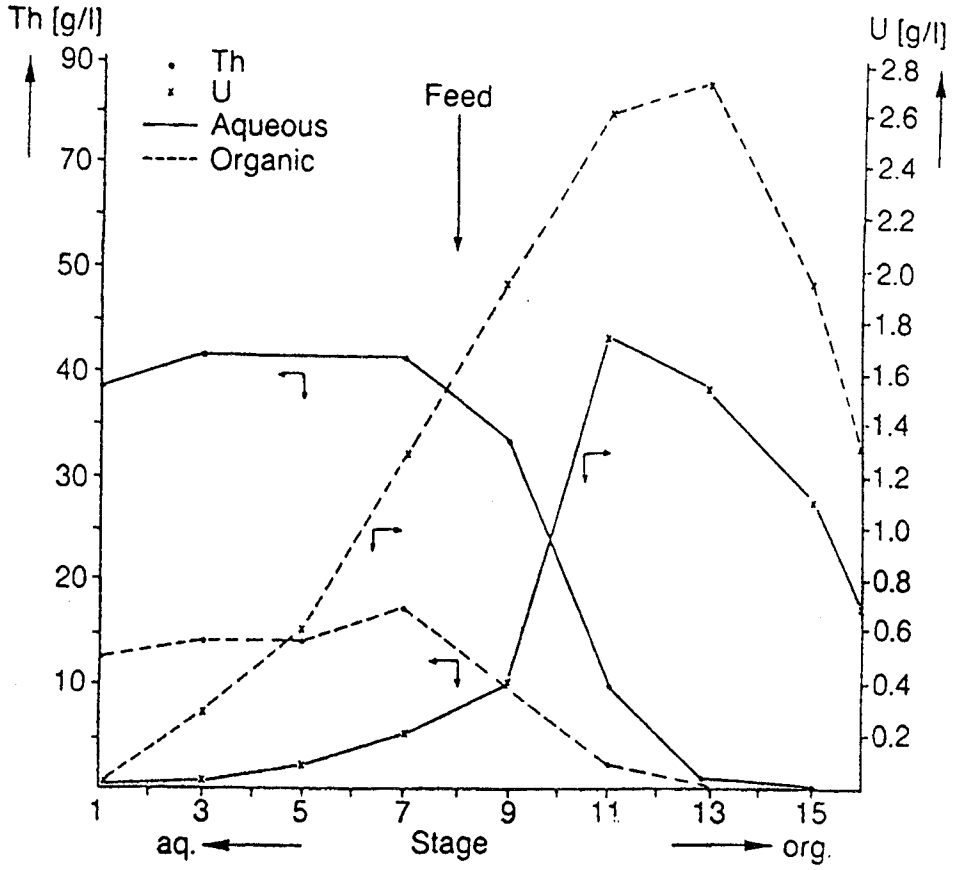


Figure 22B: Th and U profiles in Thorex process: partitioning (mixer-settler) (12)

Table 2 - RECOMMENDED ACID THOREX FLOWSHEET (12) (115)

CYCLE	FLUX	COMPOSITION	RELATIVE FLOW
1. U + Th Codecontamination (1 A)			
	Feed	1.0 M $Th(NO_3)_4$; 1.0 M HNO_3	1
	Scrub	0.1 M HNO_3	1
	Salting out acid	13 M HNO_3	0.2
	Solvent	30 % TBP in diluent (dodecane)	9
2. U/Th Partitioning (1 B)			
	Feed	0.11 M Th; U; 0.15 M HNO_3	9
	Strip	0.3 - 0.5 M HNO_3	7.5 - 11
	Solvent Wash	30 % TBP/ dodecane	1.5 - 2.2
3. U Stripping (1 C)			
	Strip	0.01 M HNO_3	1 - 5 *

* Depending on the U concentration

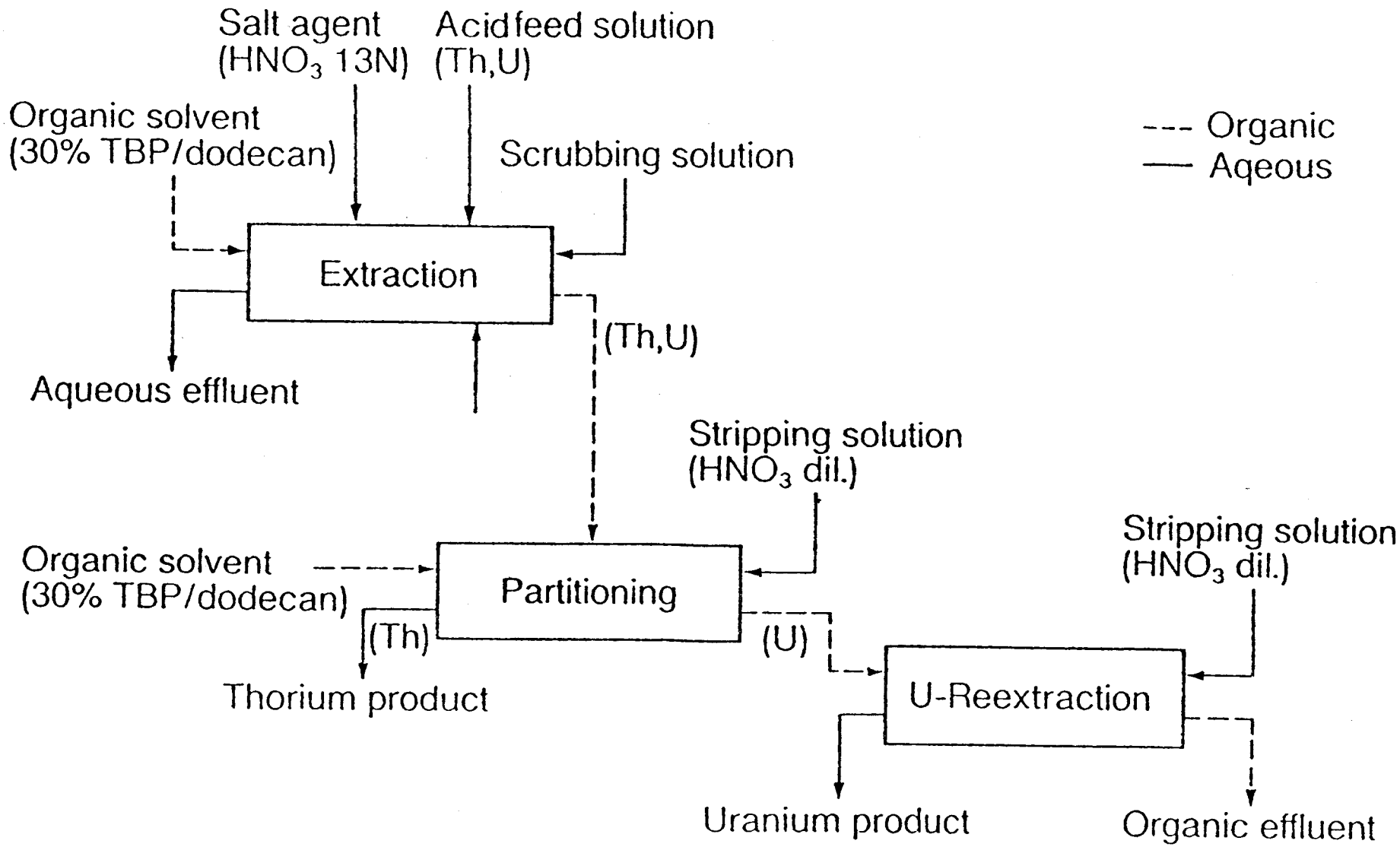


Figure 23: *One cycle Thorex Process* (12)

7.3.4. For **the final U-233 purification stages**, it is important to recall that U-233, to a lesser extent than plutonium, but higher than U-235, is a strong alpha emitter, and thus has to be manipulated in tight containment. Correspondingly, care must be exercised against criticality reactions, especially for U-233 containing solutions, hence criticality-safe and/or poisoned equipment must be used. The sizes will not be too different from those used with pure U-235 or Pu-239, and intermediate between those two.

Table 3 (42) recalls the alpha-heating rate and spontaneous neutron emissions of U-233 compared with related isotopes. Concentrated U-233 solutions are heat-emitters.

From the proliferation standpoint, the low spontaneous neutron emission of U-233, even with some U-232, would permit to fabricate simple "gun-type" weapons compared with more sophisticated "implosion-type" weapons in the case of plutonium (Fig. 24). (42)

However, the very presence of U-232 will act as an indicator due to the high gamma radiation of the decay chain, and may also detract from easy fabrication due to the rapidly increasing dose-rate with time (Fig. 25). Fresh reprocessed thorium nitrate concentrated, will also pose handling problems, due to the build-up during reactor irradiation of Th-234 (half-life 24 days) decay products, especially γ emitter Pa-234 (half-life 1,2 min), as can be seen in Figure 26 (119). Spent thorium also contains considerably more Th-228 (1.9 y) than its equilibrium value, due mainly to decay of U-232 (72 years). Hence fresh recycled thorium would have to be handled in hot cells, or else be stored for Th-228 decay for about 20 years. This fact is one of the negative aspects of the thorium fuel cycle.

7.3.5. **This presence of U-232 traces** (up to 700 ppm in U-233 for highly irradiated fuel), has been already discussed in Chapter 6.

U-233 can be "repurified" from U-232 daughter products, by simple solvent extraction or ion exchange, (A. Ramanujam, 141), and a "window" of a few days (1 week) will theoretically permit to manipulate U-233 directly in unshielded glove boxes. However, in an industrial facility this cannot be counted on and remote handling behind shielding will be mandatory.

A radical way of eliminating the problem would be to separate by a laser process the U-232 isotope from U-233. An economic costs/benefits study would show if such a scheme would be realistic for an established Th/U-233 industry.

7.3.6. This brings us back to the concept of **"close-coupled processing/fuel fabrication plants"** which are one answer for the future, not only for the thorium fuel cycle, but probably also for the U fuel cycle, when the fuels are loaded with higher Pu isotopes which emit neutron and gamma radiation.

This concept was in the air in the case of thorium, as early as 1966 (120), however the time was not ripe for such bold concepts. It is much more so today with the advancement of cheaper robotics and streamlined processes.

The underlying idea is to take for granted that the fuels, even cleaned from their neutronic poisons, will be radioactive and will have to be handled remotely from the spent fuel stage to the reprocessing, refabrication, transport stages and at the reactor. This entails a revolutionary viewpoint of the fuel recycling, and special design around the reactor.

Hence, the chemical purification process could be simplified to usually one solvent extraction step and one partitioning step U/Th. This simplification would, to some extent, counterbalance the extra costs of remote fabrication and fuel handling costs. One advantage will be the proliferation resistance of such a schema.

The remote fabrication of MOX fuels today paves the way to such concepts.

Table 3. Alpha heating and neutron emission rates for uranium and plutonium isotopes. (42)

Isotope	Half-life(y)	alpha heating rate (W/Kg)	Half-lfe(y) spont. fission	neutrons (/Kg/s)
U-232	69.8	6.93e2	8.0e13	1.78e3
U-233	1.59e5	2.75e-1	2.7e17	5.25e-1
U-234	2.46e5	1.76e-1	1.4e16	9.95
U-235	7.04e8	5.60e-5	9.8e18	1.42e-2
U-236	2.34e7	1.71e-3	2.4e16	3.64
U-238	4.47e9	8.34e-6	8.2e15	1.69e1
Pu-238	87.7	5.55e2	4.7e10	2.95e6
Pu-239	2.41e4	1.87	5.5e15	2.51e1
Pu-240	6.56e3	6.93	1.2e11	1.19e6
Pu-241	14.4	7.34e-2	-------	-------
Pu-242	3.74e5	1.14e-1	6.8e10	2.01e6
Pu-244	8.00e7	4.96e-4	6.7e10	2.02e6

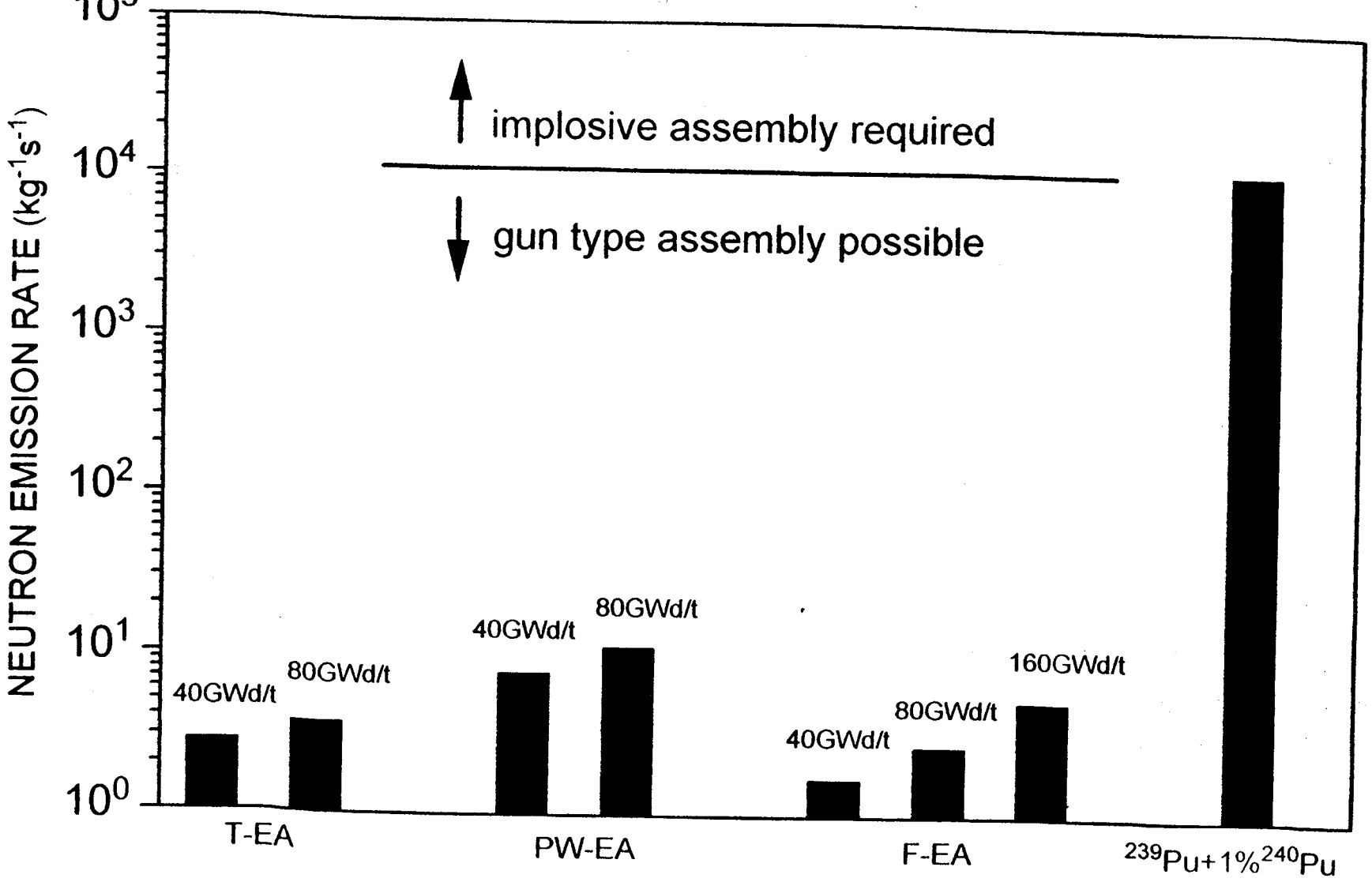


Figure 24. Neutron emission rates for spent thorium fuel from the T-EA, PW-EA and F-EA at different burnups. The reference value for an implosive assembly is given by a 1% mixture of ^{240}Pu in ^{239}Pu. (42)

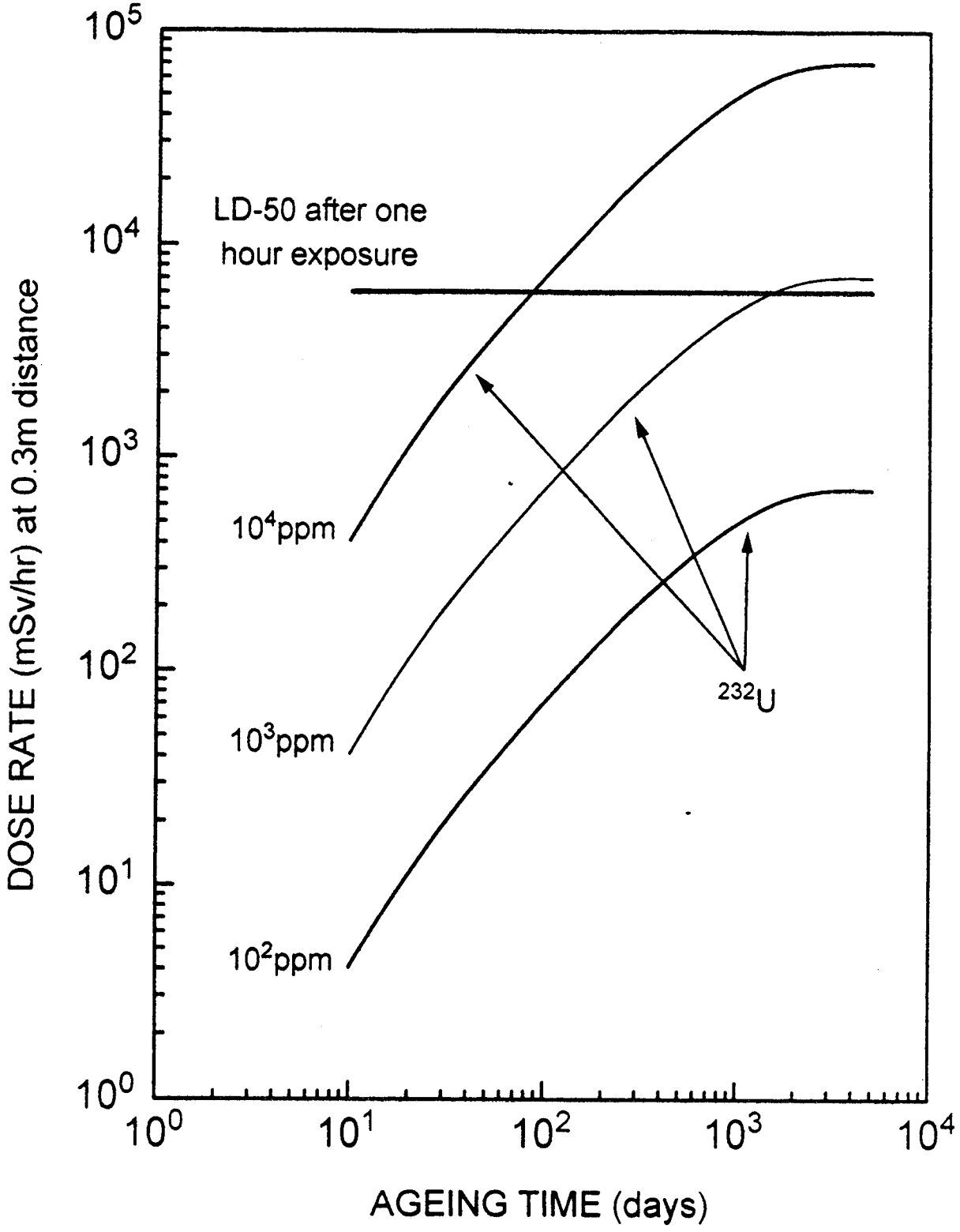


Figure 25: Dose rate as function of ageing time from 5kg of recycled U-233 contaminated with U-232 (42)

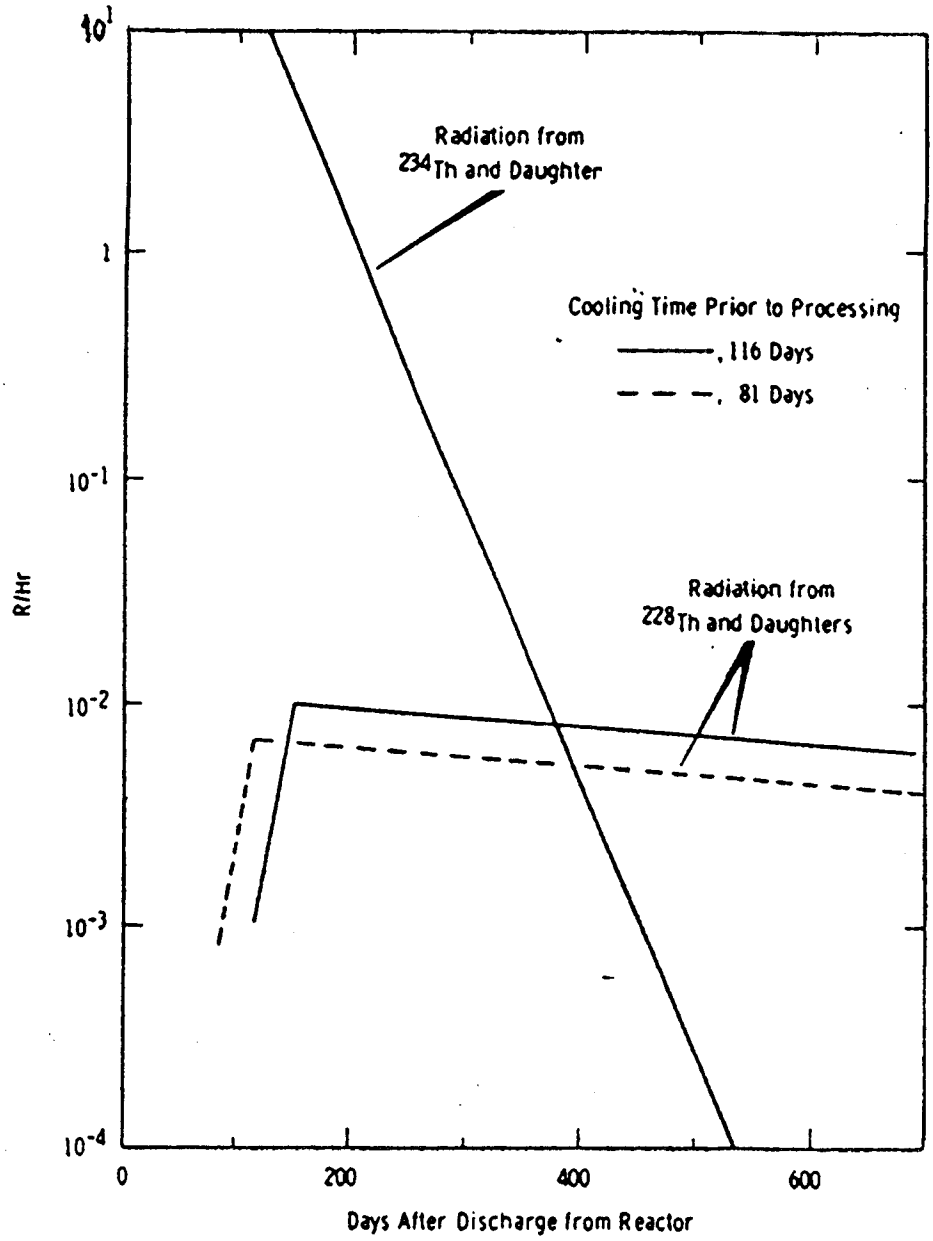


Figure 26: Savannah River plant thorium nitrate product. Radiation level at 3ft from side of railroad tank car (119)

7.4. Waste Treatment

The waste treatment from a Thorex reprocessing plant will not differ much from that of a modern Purex reprocessing plant and we do not intend to elaborate on it then.

Two remarks seem appropriate at this point :

1) The fact - as explained in more detail in Chapter 1 and Chapter 4 - that the thorium fuel cycle waste will contain less "minor actinides" (if a little more Np-237) and hence will be less radiotoxic for the first 10 000 years or so. Whether this is something really important, is in our view, debatable, owing to the excellent containment characteristics of natural media, supplemented by man's techniques.

2) Dissolution of thoria-based fuels requires a small proportion of HF, buffered by aluminium nitrate. The fluoride ion may possibly have an influence on the glass-making process of the waste, and this must be ascertained. In our view, this effect should be minimal and can be mastered.

7.5. The "Dry" Processes

Either for the processing of the carbide kernels, or for the in-line processing of the molten salts of the molten salt breeder reactors, the idea comes to resort to "dry" processes.

For the kernels, a heat treatment to up to 2800 - 3000°C followed by chloridation has been envisaged (116). For the molten salt fluorides, the fluoride volatilization process has been rather extensively experimented in a few countries, generally with success (USA, Argonne Laboratory, France, UK, etc...). The interest of the process is its relative compactness and absence of aqueous effluents. The drawbacks are the problems of working with fluorine and the corrosion of the equipment.

Figure 27 shows a possible process for a 2-zone molten-salt breeder (124) and Figure 28 represents a fluorinator permitting to extract the volatile fission products continuously from a reactor core. Such a fluorinator has been tested at ORNL. Needless to say, we are not ready yet for an industrial application !

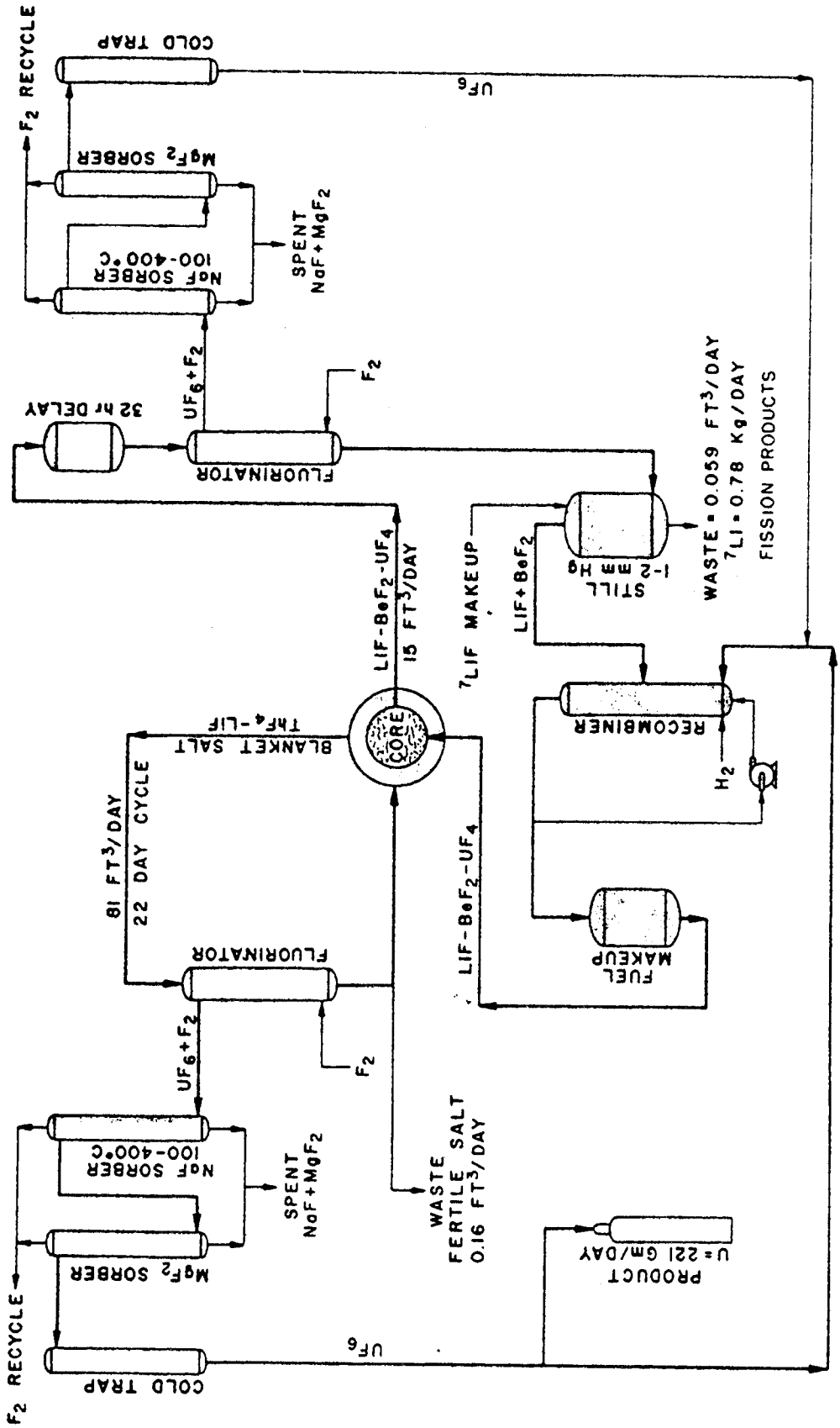


Figure 27: MSBR fuel and fertile stream processing (124)

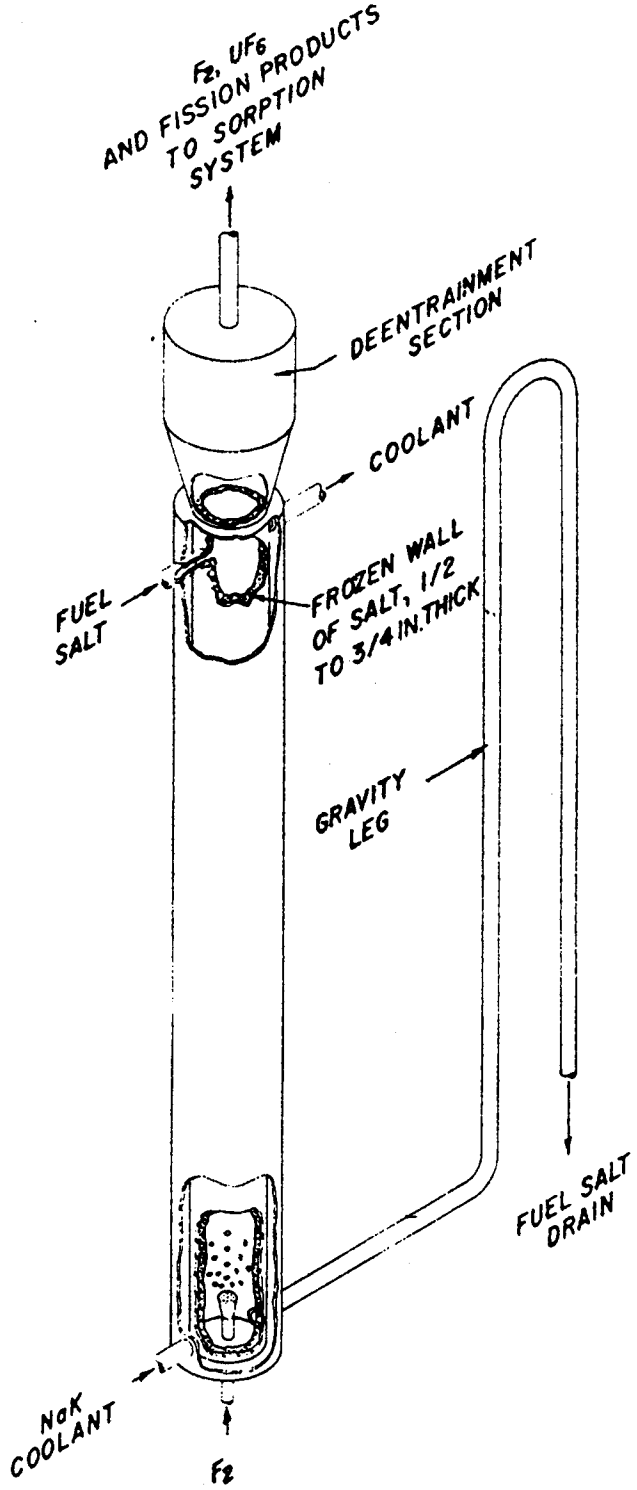


Figure 28: Continuous fluorination with frozen-salt wall for corrosion protection (124)

7.6. Concluding Remarks to this Chapter

The reader of this Chapter may think : how complicated the whole thing is ! He may be right for the advanced techniques put forward to reprocess the *oxide* carbide kernels or possible molten salts.

However, plain UO2-ThO2 fuel, or PuO2-ThO2 fuel as could be found in a light water reactor, will not present, in our opinion, more complications to reprocess than U-Pu fuels today. Once the right operating process criteria have been selected to avoid such difficulties as poor dissolution and settling, HF corrosion, third phase formation, uranium concentration peaking by reflux, solids precipitates... We must keep in mind all such problems were raised in the 1950-1960s with U/Pu separation, and have been solved with elegance. Thorium is a much simpler element than plutonium is. Conversely, one must face the fact that reprocessed thorium and uranium will have to be handled remotely in an established industry. Hence, the reprocessing steps could be simplified in a "close-coupled" reprocessing-refabrication plant in a longer-term view.

The reprocessing problem, however, exists for the HTR fuel. Much development is still needed to reprocess it readily. It is our opinion that a closer look at the real necessity of these wonderful coated "kernels" should be taken, or at the necessity to reprocess them, should their advantages be so overwhelming, at least in a medium-term perspective.

8. OVERVIEW OF NATIONAL AND INTERNATIONAL PROGRAMMES FOR THE THORIUM FUEL CYCLE

8.1. Historical Reminder (beginnings until 1975-1980)

8.1.1. General remarks

To understand the present, it is useful to refer to a more glorious past where a great number of avenues were investigated with dynamism, in the view of a rapid development of cheap nuclear energy where uranium would have to be spared and thorium could be an interesting, maybe necessary, supplement.

Quite naturally, following the Manhattan Project, the USA investigated thorium and its daughter U-233 as a possible weapon (Argonne, Oak Ridge, Los Alamos). Experimental weapons were made. The interesting neutronic properties of U-233 and the excellent breeding and high temperature characteristics of thorium quickly interested the scientists for peaceful applications. Soon thereafter most nations with a nuclear programme started to investigate the thorium fuel cycle.

USA : three extensive thorium-fuel reactor programs have been undertaken in the USA. These are the Molten Salt Breeder Reactor (MSBR), the Light Water Breeder Reactor (LWBR) and the High Temperature Gas Cooled Reactor (HTGR).

HTGR has used thorium since 1976 in the 300 MWe Fort St Vrain power plant. The behaviour of the graphite based coated particle fuel with (Th,U)C2 was good. Because of problems with some components the Fort St Vrain reactor did not reach a satisfactory performance. The later fuel charges used uranium only.

It should be reminded that the small commercial HGTR of Peach Bottom (1967-1974) (General Atomics) has given good results.

This study and development of **Molten Salt Reactors** was begun in the USA at Oak Ridge National Laboratory in 1947. The potential of MSBR for civilian power production was recognized and a development programme was established in 1956. It was fuelled with a U-235/U-238 mixture during the initial two years of operation and with U-233 during the remaining 1.5 years of operation. The successful operation of the thorium molten salt reactor and a favourable projected system characteristics attracted significant US industry and utility interest. The development of molten salt reactors was interrupted in 1973. The programme was resumed briefly in 1974 but finally terminated in mid 1976.

The **Light Water Breeder Reactor** concept, called afterwards seed-blanket, was originally introduced in 1951 as a means of minimising the separative work required for the fuel of the light water reactor. The seed-blanket concept was employed in the last core design of the first commercial PWR plant at Shippingport.

It was long thought to be impractical to breed with light water, however, since the eta value of U-233 is only slightly lower in the epithermal region, while that of U-235 and Pu-239 are greatly reduced, the thorium cycle appeared to be most attractive for a thermal breeder. After preliminary work in the early 1960s indicated the feasibility of breeding in a light water seed-blanket core on the thorium cycle, the US-AEC authorised a demonstration in the Shippingport plant. The demonstration in full-power operation cores started in December 1977.

The reactor operated on thorium and U-233 cycles until 1982 at which time it was shut down. The fuel has been reprocessed and it has been shown that a sustainable LWR reactor on thorium cycle could be operated in some conditions.

We must also recall the small commercial BWR reactor of Elk River, fuelled with U-Th classical fuel with a graphite reflector (1963-1968), as well as the Indian Point PWR (1962-1980).

To accompany these achievements, reprocessing of irradiated thorium was performed at Oak Ridge, Hanford, Savannah River, West Valley, for about 700 tons, producing about 1.6 tons U-233. More of it seems to be available from defence programmes.

Fuel fabrication of thorium, U-Th fuel elements has been performed by ORNL and industry (Babcock-Wilcox), for all sorts of fuel and all sorts of processes and matrices, and a great experience has been accumulated.

It is clear that the ban on reprocessing after 1975 has stopped much of the research on thorium, which was the most advanced in the USA at the time.

8.1.2. **Canada**

The possible use of thorium has been recognized very early in Canada, which has great reserves of uranium but also of thorium.

Candus operated on a thorium cycle are feasible and can be self-sustainable, i.e. producing enough U-233 to sustain the next charge. As such reactors need a U-235/U-238 driver, or even, if one goes slowly enough, natural uranium with a few thorium fuel elements to start with, the problem was to work on the chemical

separation of U, Th and Pu. This work has been conducted at Whiteshell, Manitoba, in a series of hot cells with mini-mixersettlers. Feasibility of operations was demonstrated.

However, in the end of the seventies, the work was discontinued; the use of the thorium cycle in Candu reactors was abandoned. The once-through fuel cycle not involving reprocessing was considered the more convenient, after a number of studies of fuelling strategies, including the use of slightly enriched uranium (SEU), which was finally chosen.

However, as will be seen, a long-term research program has been recently reestablished.

8.1.3. **Ex-USSR/Russia**

The use of thorium was regarded in USSR as a contribution to assure the fuel supply to its extensive nuclear power programme. In fact the combination of uranium-plutonium and thorium cycles ensures long-term fuel supply, makes the nuclear energy production more flexible.

A realistic view at the fast reactor breeders permits to conclude that there could be room also for fast breeders with a thorium blanket, or other types of self-sustaining reactors based on thorium, in the future.

Research on the thorium fuel were started at the beginning of the nuclear era in the former USSR and are still going on at a relatively low level. These studies were concentrated on :

- Physics of WWER and BN type reactors;
- Development of the basic fuel for WWER and BN reactors;
- Development of reprocessing technology.

These studies have shown that the many problems with the U based nuclear fuel cycle (radiotoxicity, plutonium accumulation) could be solved by changing to the thorium fuel cycle.

BN type fast reactors cores could be fuelled with Pu/Th fuel to burn plutonium and produce U-233 for WWER reactors. A specialized reactor of BN-800 type with core based on Pu/Th fuel will be capable of burning up to 600 kg of Pu per year and produce up to 700 kg of U-233 per year.

Possible improvement of safety of WWER reactors by use of thorium has been analyzed.

New thorium-based fuel compositions of the cermet-type are verified, mastered and adaptable to commercial production. Use of U-233 based fuel utilization, for instance in space or in remote nuclear power units, is possible, according to a IAEA seminar (1).

8.1.4. **Japan**

The research on the thorium fuel cycle was also active in Japan in the 60s, but has declined in both government institutions and private companies. On the other hand, a large number of university professors were interested in the thorium fuel cycle, and a research group on thorium fuel was organized by T. Shibata as a part of the Special Project Research on Energy, under a Grant-in-Aid for Scientific Research by the Ministry of Education, Science and Culture from 1980 to 1986. They carried out many works; nuclear characteristics, fuel, down-stream chemistry and biological effects related to the thorium cycle. A Japan-US Seminar on Thorium Fuel Reactor was held by this group (Shibata, et al., 1985).

One year after the completion of the Special Project, a new cooperative research program on Thorium as an Energy Source in the 21st Century was organized. It was supported by Grant-in-Aid for Scientific Research by the Ministry. Thereafter, another new program on the Thorium Fuel Cycle as a Promising Energy Source in and after the 21st Century was supported again from 1990 to 1992. In 1990, the Indo-Japan Seminar on Thorium Utilization was organized (142).The leading figure in Japan has been Professor I. Kimura, of Kyoto University, followed now by Professor K. Furukawa.

Research Works and Results (5)

More than 60 papers and reports were published and large number of papers were presented at various meetings. The list of the publications is shown in a brief report of the Cooperative Research Programme of the IAEA. (144)

Main works in the assigned subjects are as follows :

1. Design studies of thorium fuelled reactors, including

- Nuclear data evaluation,
- Neutron cross section measurements for thorium and related nuclides,
- Core designs of Th/U in PWRs, HCLWRs (with high conversion factors), MSRs (molten salt),
- Actinides production analysis in Th-U loaded reactors,
- Critical experiments (i.e. in the KUCA facility at the Kyoto University,
- Reactivity measurements for thorium and other samples for MSRs,
- Neutron transport experiments.

2. Study of thorium fuels and related materials, including :

- Thermochemical study of thorium and Th-U monocarbides,
- Chemical diffusion of constituent elements in mixed Th/U oxides,
- Preparation of Th/UO2 powder by a freezing process,
- Fission product release from coated particles of Th/UO2 fuel at very high temperature (2 000°C),
- Solubility of oxides in molten LiF-BeF2 and NaBF4-NaF,
- Mass transfer in molten salts,
- Microscopic study of crudding on nuclear fuel rods,
- Basic study on thorium fuel reprocessing,
- Molten salt technology.

3. Radiation safety of the thorium fuel cycle.

4. Preliminary work for fusion-fission hybrids.

Besides university work, R&D has been performed by the Japan Atomic Energy Institute (JAERI) : JAERI started in 1975 basic R&D works on thorium and thorium-uranium mixed oxides to develop laboratory-scale fabrication methods, examine irradiation behaviour and to measure physico-chemical properties of these fuels. The main accomplishments have been :

- Development of a new sol-gel process to prepare crack-free microsphere fuel with better sphericity aiming at HTGR applications and to prepare the starting material for making high-density pellets with varying Th/U ratios.

- Measurement of fission-gas release/irradiation-induced damage and data analyses to predict irradiation stability and densification mechanism.

- Measurement of new data on equilibrium oxygen potential/stoichiometry and its effect on chemical behaviour of (Th-U)O2 at simulated burn-ups.

In Japan, the use of thorium as a fertile material in nuclear reactors is considered as a very long-term issue.

8.1.5. **India** (141, 142)

India is the only country where a self-sustained programme has continuously been implemented without any interruption, contrary to other countries. Whereas in other countries there are no more reactors operating on a thorium fuel cycle, there are such in India. This is justified in principle by the huge monazite sands deposits of India compared to relatively smaller uranium resources.

When nuclear power was started in India, a three stage program was chosen, based on :

(1) Heavy water reactors,
(2) Fast breeders,
(3) Thorium based reactors (as a long-term objective set by late Dr. Bhabha).

Serious study for the third part was initiated in 1977 with a small reactor.

The situation as of 1994 was as follows (1, 139), see Fig. 1 :

1. 500 kg of thorium fuel has been used for initial power flattening in the KAPS-1 reactor which was commissioned in 1993, after preliminary tests in MAPS-1.

2. 500 kg of thorium fuel has already been loaded as part of the initial charge of KAPS-2. This was to be critical in 1995. (7)

3. Thorium rods ("J rods") have been irradiated in the reflector of a research reactor core (CIRUS). They were reprocessed to obtain U-233.

4. A critical assembly with U-233 nitrate solution was built (PURNIMA 2).

5. A critical facility (PURNIMA 3) using U-233/Al fuel was put in operation at BARC in 1990, leading to a small 30 KW material test reactor on the same principle at IGCAR near Madras, almost complete (KAMINI) in 1995-1996.

6. Reprocessing technology for extracting U-233 from thorium has been developed and a commercial scale facility is now ready for operation.

7. (Pu-Th)O2 pins were fabricated and a cluster of 6 pins was irradiated in an in-pile loop to 19 000 Mwd/T.

8. Advanced fabrication technology for thorium U-233 fuel is being developed.

9. India has made a proposal for introducing thorium into one of the power reactors along with MOX. This is under consideration.

10. The advanced heavy pressurised water reactor (AHWR) specially designed for thorium, is being developed. Some funding has been provided for a study project. The feasibility report was expected to be submitted in 1995.

A large staff is mobilized in the two largest Indian Research Centres of BARC under Dr. A. Kakodkar, and of IGCAR under Dr. P. Rodriguez.

8.1.6. **Germany**

Germany occupies a special place in the thorium fuel cycle development, due to its use of the very particular concept of the High Temperature "pebble bed" Reactor.

The development of thorium fuelled reactors has been closely connected in the FRG with the activities related to the High Temperature Reactor. In the HTRs the use of thorium as fertile material decreases the consumption of U ores and then it was adopted as a basis for the AVR as well as the THTR The high-temperature physical and neutronic properties of Th and U-233 were also considered.

8.1.6.1. HTR Program

In the sixties, comprehensive R&D work on the development of Th-Oxide fuel - ThO2 or (ThU)O2 - including irradiation tests have been performed in Germany at KFA and Nukem/Hobeg. The ThO2 fuel was successfully used in the AVR (for about 15 years) and in the THTR (for about 3 years).

The 15 MWe AVR reactor has been successfully operated since 1967 with graphite based coated-particle oxide thorium fuel. Burn-ups of 15-10 % fima (*) or about 140 000 MWd/t, have been reached for a significant portion of the fuel showing good structure and hardly any defects. Up to 1988 about 2 000 kg of thorium heavy metal pebbles have been fabricated and inserted in the reactor. The 300 MWe demonstration THTR plant in Schmehausen also using thorium-based coated particle (Th,U)O2 fuel has been in operation since 1986 showing good fuel behaviour under irradiation. It was shut-down in 1989 due to technological problems within the reactor vessel, but not due to the fuel cycle itself.

Operation of those reactors required the use of HEU, which suffered objections in view of the weapon proliferation. HEU was then substituted by LEU, and the once-through fuel cycle was tested (in AVR) and finally chosen.

In theory, the thorium cycle is also possible with a medium enriched uranium (MEU) with a certain advantage for U ore consumption. (16)

(*) 1 % FIMA is a bout 9500 MWd/t.

In the last years in Germany, a modular version of HTR was proposed (the MODUL) whose goal is essentially in the use of passive safety systems. The use of thorium cycle in MODUL does not correspond to the first priority for these reactors.

In relation with these HTRs the respective reprocessing technology has been demonstrated in a pilot plant at the Jülich Centre.

Related to this reactor program, activities directed to spent fuel treatment have been concentrated more recently on the intermediate and direct storage of the (Th/U)O2 fuel.

8.1.6.2. Liquid metal-free breeders or converters

Another effort in FRG has been concentrated on the development of the heavy water moderated thorium breeder reactor. This activity incorporated, beside nuclear core design and fuel development, also an irradiation program; in the course of which Vipac (Th/U)O2 fuel was irradiated up to 5 % fima (i.e. about 50 000 MWd/t), showing good fuel behaviour.

In parallel, theoretical and experimental work at laboratory scale has been done by KfK/Siemens on LWR-converters using thorium as the breeder material. That also included irradiation tests.

The recycling of Pu and Th oxide fuel was investigated in a few fuel assemblies in the Lingen BWR in the early seventies. The task was terminated in 1973 after changes in the boundary conditions.

All those activities were terminated due to the lack of financial support at the beginning of the 80s.

However, a German-Brazilian cooperation in Scientific Research and Technological Development, which included a program of R&D on the thorium utilization in PWRs (1979-1988), concluded to the feasibility of using a thorium cycle in conventional PWRs without change in the reactor essentials. (12)

8.1.7. **Italy**

This is another example of a country where not only nuclear energy has been energically promoted, but also the thorium fuel cycle.

At the beginning of the sixties a very high interest appeared in Italy for the uranium-thorium cycles, under the pressure of economical arguments as well as

of energy independence. Italy, as the majority of the European countries, was concerned about the availability of uranium (remember for instance that the Chapter 6 of EURATOM Treaty was inspired by this concern, establishing a "Supply Agency" whose aim was to insure that all the states of the European Community would be equally treated for the uranium supply). Consequently, research was addressed to the use of thorium, very abundant in nature, which permitted savings in uranium ore consumption for the same energy production. Comparisons of U-Th cycle with all sorts of U-235/238 cycles with U and/or U+Pu recycling, showed a clear advantage for the uranium-thorium cycle, the main reason being related to the higher conversion ratio, due to the high eta value of U-233. In fact, the thorium fuel cycle appeared to be the most economic of all recycled fuel systems for thermal reactors, provided the costs of construction and operation of the recycle facility would be of the same order of the recycling costs of uranium-plutonium spent fuel reprocessing. Recycle systems in general being considered essential for the sound development of nuclear energy, CNEN and Allis-Chalmers Manufacturing Company undertook a Uranium-Thorium Fuel Cycle Program (PCUT), consisting of a Reactor Evaluation Study and a project for the design, construction and operation of a facility to process and refabricate Thoria-uranium oxide fuel assemblies.

An advanced prototype plant with remote maintenance, rack-mounted reprocessing plant, with a remote fuel refabrication plant, were built with Allis-Chalmers engineering at the South Italy Rotondella ITREC Center. The plants were due to operate originally with spent BWR U-Th Elk River elements, later on DRAGON-type fuels.

The PCUT plants were planned in the sixties, built and cold-tested until 1974, but then activities were stopped. The fuel fabrication plant was definitively stopped but the reprocessing plant has been used subsequently as a pilot plant for hot and cold tests of advanced components (centrifugal contactors, continuous dissolver, etc...)

A first campaign on 7 Elk River (ERR) spent fuel elements was conducted between 1975 and 1985; many small incidents occurred and much operational and chemical experience was accumulated. A second campaign was planned on 16 ERR spent fuel elements in 1986-1987.

The plants are now under dismantling.

8.1.8. **France**

Like in other countries, France started early with its interest for the thorium fuel cycle.

Rich Monazite (uranothorianite) deposits in the Madagascar sand permitted to operate a thorium separation pilot plant at Le Bouchet. About 2 000 tons of thorium have been produced until 1955 and most of it sold to the USA.

The CEA conducted preliminary studies based on thorium fuel cycle.

For some years EDF has carried out studies concerning the thorium cycle. These studies started in 1969 in relation with the HTR and have been performed jointly with the CEA Saclay as well as Cadarache for integral experiments needed for having better accuracy on nuclear data.

With regard to the PWRs, the studies were firstly concerned with the use of Pu with Th to start the cycle. A satisfactory solution was obtained by recycling the Pu with Th as from the first core loading. The solution proposed consisted of loading the whole reactor with Th/Pu assemblies from the first core and no longer using the checker-board loading scheme where the fuel assemblies are loaded in rings. Two positive aspects appear : burnable poisons are no longer necessary and the fuel assemblies are no longer divided into several zones (as with Pu recycle together with U in reloadings). There are however, two rather negative characteristics : the second generation Pu may be used only in fast breeder reactors (FBRs) and the conversion factor is rather small. Nevertheless the proposed solution gives acceptable pin power peak and temperature coefficients.

Satisfactory solutions have also been obtained for core loaded with Th/U-233 assemblies. The main problem is, in this case, the moderator temperature coefficient.

However, the theoretical advantages of the U ore consumption *savings* which could be considered with the use of a thorium cycle can be obtained with the development of advanced reactors using different means (i.e. spectral shift, use of MOX fuel, etc...).

Hence, apart from a technological watch and nuclear data acquisition, a real programme has been interrupted.

8.1.9. **The UK**

Like in other countries, interest for thorium was at a peak in the 1950s-1960s. Of course, being the host country for the DRAGON HTR prototype, has given the UK a special interest in HTRs and thorium for some time.

UKAEA had a thorium research programme and built a prototype thorium reprocessing plant at Dounreay called D 1203. This plant has been used on and off until rather recently.

At present, there is no definite programme going on the thorium fuel cycle.

8.1.10. **Netherlands**

At Kema-Arnhem, a team under Dr. J.J. Went designed and built a small UO2/ThO2 suspension reactor KSTR or SUSPOP which was operated between 1974 and 1977. The underlying ideas were that the suspension of UO2/ThO2 particles in water is as near as possible to a homogeneous reactor with a good neutronic efficiency, a high degree of safety : temperature control, boiling if necessary, breeding capacity, and on-line purification of most of the fission products which are ejected from the fuel microspheres (diameter 5 microns), especially Xe-135 gas which constitutes one of the high-yield and neutrons absorbing fission products; these could be removed on line, and U-233 would eventually replace U-235.

This unusual prototype has been operated rather successfully, but no extrapolation has been envisaged.

The whole reactor has now been dismantled.

8.1.11 **Other countries**

Australia : the thorium resources are those contained in monazite sands (RAR 13 000 tonnes) exploited for other minerals. Monazite concentrates production in 1983 was of 15 000 tonnes containing about 900 kg Th.

Theoretical studies and experiments have been performed by the AAEC, especially nuclear data acquisition experiments. The AAEC has carried out four experiments in the area of nuclea data on thorium isotopes : fission fragment angular distributions for Th-232; the average number of prompt neutrons emitted in the fission of Th-232; fission fragment angular distributions for Th-230, and the fission cross-section of Th-230. The first three experiments (concluded by 1980) aimed to examine the existence of a triple-humped fission barrier in the fission of

thorium. The data from a fourth experiment in 1985 measured the sub-threshold fission resonance in Th-230 near 715 keV to confirm the energy resolution and energy accuracy of the first three experiments.

Sweden, Austria are known to have conducted theoretical studies on thorium use in the 1960s.

8.1.12. **International cooperation**

The OECD-NEA HTR DRAGON prototype reactor was built in England in participation of EC-EURATOM and operated successfully between 1966 and 1973. It was originally fuelled with rather sophisticated (Th/U)C fuel elements in a graphite reflector, with helium cooling in a once-through cycle. Later the fuel elements were of the General Atomics hexagonal, simplified type (Peach Bottom type) but with less and less thorium, finally only with uranium, although towards the end the French and German were in favour of HTR thorium cycles.

The project has been interrupted in 1974, but has given a lot of experimental data, showing that the thorium breeding elements could be left for long periods in the reactor (240 days or more), with irradiations higher than 100 000 MWD/t. Temperatures as high as 1 500°C were reached, however at the cost of graphite sleeve shrinking. The programme was stopped in 1974, mostly for political and economical reasons.

Paper projects of a 500 MWe HTR were derived from the DRAGON experience.

Much ceramic/pyrocarbon fuel research was conducted at the UKAEA for the DRAGON project, and reprocessing tests were conducted at Dounreay and at Oak Ridge.

At the **Kjeller Research Centre**, Norway, thorium was also investigated, and a full course on thorium fuel cycle could be attended there for some years.

The ORGEL EC Concept of natural uranium, organic-cooled, heavy water moderated 500 MWe reactor has been pursued in the 1960s at the EC Ispra Centre. A similar research reactor has been built and operated at Whiteshell in Canada. Adaptations of ORGEL to slightly enriched (U-235 or U-233) thorium have been calculated (Lafontaine and al). They would be more attractive for large-type reactors (1 000 MWe) but the mandatory reprocessing and fuel refabrication step with thorium would create an economic disadvantage compared with uranium.

With the progress of the simpler light water reactors, the ORGEL programme was finally abandoned.

Thus, the 1960s saw the great days of the thorium cycle. Almost all avenues were investigated, and a huge corpus of knowledge and practical experience has been accumulated in these days. Among others, these golden days are reflected in the Thorium Fuel Cycle Symposia at Gatlinburg, Tenn. in 1962 and 1966.

8.2. Difficulties, the Oil Shock, Focusing on U-LWRs

We shall not dwell too long on the reasons which have led to a practical progressive abandonment of the thorium fuel cycle between 1973 and 1985 in most countries.

Even some of the originally thorium-fuelled high temperature reactors, such as Dragon, AVR, THTR, were later fuelled progressively with uranium. In that case, the main official reason was objections against the use of highly enriched uranium.

8.2.1. **The first reason for abandonment is an economical one.**

The extensive uranium mining development in prevision of great strides in uranium consumption, have led to an excess of uranium, of enriched uranium, at affordable prices. The fuel cycle was well demonstrated and fuel fabrication could be done simply with hands-on processes.

Conversely, the thorium fuel cycle is less straightforward; one more neutron is necessary to the chain reaction compared to uranium. Reprocessing is somewhat more complicated, especially when Pu is present at the beginning. Fuel refabrication is uneasy and expensive.

8.2.2. **Political reasons**

The far-reaching interdiction of reprocessing in the USA enacted by presidents Ford and Carter following a sort of proliferation hystery has ruined the FBR and thorium fuel cycles in USA within years. The movement has been followed in many other countries, with a great loss of human experience. The situation in the Western countries as of today still suffer from this unconsidered move, in my opinion.

The designer-builder of HTR in USA, General Atomics, became part of Gulf, which stopped these activities.

8.2.3. The PWR and the oil shock

The outstanding performances of the US PWR submarine reactors developed by Admiral Rickover, their simple design, came at a time when many nations revised their energy programmes following the oil shocks. Industry had to move quickly, and the light water reactors became the answer. Money also was less available and thorium became progressively less attractive and much less urgent. Other priorities were developed, like space and electronics.

8.3. The End of the Thorium Programmes, Keeping Technological Awareness (1980-1990)

If the industry, for the reasons indicated, had chosen its course, which will continue until well into the next century, the national research centres and institutions, and the universities of course, continued in their interest for the thorium, at least for some time :

Large experiments were underway : the Molten Salt Prototype reactor in Oak Ridge, the Fort St Vrain HTR industrial prototype, continued some time in the USA, until respectively 1976 and 1989. AVR and THTR continued also in Germany until 1988 and 1989. The scientists in charge, convinced of the merits of their solutions, battled to keep their experiments, their staff and their ideas, but the fundings were stopped progressively.

To keep the memory, many seminars were held, especially at Jülich in Germany (1978, 1980, 1984) and at the IAEA Topical Meetings. A rather good summary of the achievements has been published in 1980 in the course of the INFCE exercises under the auspices of IAEA.

Meanwhile, however, the Indian Programme was continued, undeterred, at its own pace.

In most countries thorium is at rest, except in India; Japan pursues the project of a High Temperature Engineering Test Reactor (HTTR) under the leadership of JAERI of the Science and Technology Agency.

If, for reasons of simplicity, this prototype 30 MWth reactor, now under construction, will be fuelled with low enriched (3 % - 10 %) UO2 in pyrolytic carbon spheres arranged in fuel element "compacts", the possibility exists to test also the thorium fuel cycle. The reactor, at O-Arai, should start operation in 1998.

8.4. Renewed Theoretical Interest (1990 -)

After long years of latence, the flame maintained by some of the old hands and by some professors, has brought renewed interest in the thorium fuel cycle, but for reasons quite different from those which prevailed in the 60s.

Sometimes these reasons look like a re-discovery of known properties of the thorium fuel cycle - so the world goes.

Here are the advantages put forward by some proponents of the thorium cycle, either by Russian, German, Indian specialists. We quote the Russian specialist (Mr. V. Kagramanian) who claims in a quite straightforward way (IAEA, 1994) (1) :

"Advantages of the thorium fuel cycle.

"(1) The development of nuclear power without plutonium and with minimum level of the long-lived radiotoxic nuclide in the wastes.

"The change from uranium to thorium permits the development in Russia of nuclear energy in the long-term on using WWER and BN type reactors, but without plutonium and minor actinides production. This closed fuel cycle is characterized by a much lower level or radiotoxicity of long-lived nuclides in the waste compared with traditional uranium-based systems. From the point of view of radiotoxicity, the thorium option could be considered as an alternative to the transmutation option.

"(2) Increase of proliferation resistance.

"The uranium-thorium fuel cycle could be developed as a more proliferation resistant fuel cycle than the plutonium-uranium one, because of high energy gamma rays from a descendant nuclide of U-232 that usually accompanies U-233. There is also a possibility to denature U-233 with natural U-238.

"(3) The burning of plutonium.

"About 30 t of separated civil plutonium has been accumulated in Russia. Nearly 100 t of ex-weapon plutonium is expected to be released due to nuclear disarmament. Use of Pu/Th fuel in BN and WWER reactors could be the most faster and simplest way to burn this plutonium. The production of new fissile U-233 in spent fuel due to plutonium burning could be the fuel basis for the U/Th fuel cycle in future."

Comment : we do believe indeed, for our part, that burning of some of the weapons plutonium could be one interesting way to enter the thorium fuel cycle.

To these arguments, we would like to add the following ones :

(4) India adds the fact that thorium cycles offer more flexibility with respect to policy decisions. Thus if it is decided today to destroy all stockpiled fissile material, and 10 years from now the objective changes to conserve those resources, the thorium cycle offers the best system to meet these changing goals.

(5) It goes almost without saying that thorium remains an important fertile material to supplement U-238 for the future energy needs of mankind.

The argument that the radiotoxicity of the thorium fuel cycle is lesser than that of the uranium fuel cycles due to much less long-lived minor actinides present, such as plutonium, americium, curium isotopes, is now taken quite seriously in the West, and at the recent Global '95 Conference in Versailles, many papers were devoted to the subject. Whether this is a real issue or not, considering the perfection of the planned high level waste underground repositories, remains to be seen.

(6) Another issue is the **Accelerator-driven Reactor System**, recommended by C. Rubbia, or the ABC system (Accelerator Based Converter), recommended by C.D. Bowman at Los Alamos. In this concept, the large quantity of neutrons created by spallation by impingment of heavy ions accelerated by a high-intensity 1 - 1.5 GeV accelerator on a heavy metal target - liquid metal for example - would permit to drive a subcritical reactor. The disadvantage of the thorium-fuelled reactor which needs a large excess of neutrons to reach criticality would be partly overcome, and the thorium fuel cycle would present the interesting features exposed above. However, one would have to pay for the extra cost of an accelerator plus that of a reactor, even if the reactor could - theoretically at least - be simplified, as claimed by its proponents .

(7) Finally, the example of **India - and to some extent - of Japan and Russia**, pursuing their long-term objectives, are observed with interest.

Recently, **China**, which also has large thorium reserves, has decided to launch a HTR programme, in which thorium cannot be totally absent.

None of the above points are likely to be seen by industry as overriding incentives, but they are all the same, very real advantages of the thorium cycle. Therefore, many countries are turning themselves towards IAEA whose role should be to collate and coordinate the thorium work being done by different countries, and encourage those countries where small groups are working on the subject.

Besides India which has a thorium programme alive and going, where do we stand ?

In **Japan**, in the Universities...

After the two cooperative research programmes described above, application was made to start a new extensive programme by Grant-in-Aid for Specially Promoted Research, but unfortunately it was not adopted by the Ministry. However, the recognition of the importance of the thorium fuel cycle is growing, therefore many staff of the cooperative programmes are still continuing research on the thorium fuel cycle, for example critical experiments with thorium at the KUCA accelerator, design study of Th/U-233 fuelled reactors, new thorium fuel development and thorium fuel irradiation, and molten salt technology. Recently new groups have started to investigate the thorium fuel cycle.

At Jaeri, R&D continues within the HTTR programme.

In **Canada**

A small but rather broad programme of research was reestablished in the last couple of years because of interest in the marketplace in the potential to burn thoria fuels in Candu reactors. The work involves fuel fabrication, irradiation, post-irradiation examination, reactor physics and waste management for thoria-based fuels. This programme is under the direction of Dr. Peter Boczar at Chalk River Laboratories. A number of rather recent reports have been published on these subjects.

In **Russia**

Recent efforts have been developed to produce isotopically pure U-233 for experimental goals. Thorium fuel elements were fabricated and irradiated in the BN-350 fast reactor. Fuel composition for different reactor types is important from the standpoint of reprocessing. While metallic thorium can easily be used in fast reactor blankets, special studies are required for VVER and BN core fuels. Also studies must be conducted on thorium-based fuel reprocessing. This is

because acid is required at levels which may be dangerous from the standpoint of corrosion of special chromium-nickel alloys.

Current compositions of UO2-ThO2 type are attractive in this respect, especially in the case of Pu/Th fuel of PuO2-ThO2 type.

A study is underway to develop a concept of changing from the present U/Pu fuel cycle to future nuclear power systems with thorium without plutonium and minor actinides accumulation.

The research works on thorium in Russia are authorized and subsidized (at a very low level) by Ministry for Nuclear Power.

Among the proponents of the thorium fuel cycle, are members of the Institute of Physics and Power Engineering at Obninsk (Prof. V.M. Murogov) and the Kurchatov Institute (Prof. N. Ponomarev-Stepnoi).

In **Germany**

At present no thorium programme is running. Some small studies on actinide generation and Pu-burning in thorium cycles (or burning of pure Pu in HTRs) have been and are still being performed (latest study by Siemens under contract from EC, Brussels, is still underway).

The long term R&D work for Th/U cycles should be done as "basic research" in universities and research centres (and not in the industry).

On the other hand, the **EU Institute for Transuranium Elements** (ITU) has undergone basic studies to assess some renewed ideas on thorium use, especially in the context of accelerator-driven reactor projects and of minor actinides by-products. Research is also progressing on remote fuel fabrication techniques, a must with the thorium fuel cycle.

In **France**

The SPIN Programme of the CEA evaluates ways to separate the minor actinides and incinerate them in reactors, preferably fast flux reactors, or in accelerator-driven reactors, due to the high neutron flux available, or dedicated accelerators.

These studies have led inevitably to the thorium fuel cycle which, for the time being, receives a motion of esteem among the physicists, but not in the industry, something which can be understood in these rather depressed times for the nuclear industry.

Dr. A. Lecocq of Euriwa, in cooperation with Japan's Dr. K. Furukawa of Tokai University, and the Radiology Institute of the Academy of Sciences of Minsk, continues to pursue the idea of the thorium molten-salt reactor which can be activated by an accelerator, the molten salt reactor having many physical advantages as far as breeding, ease of operation, in-line purification from the fission products and continuous incineration of the minor actinides.

However, corrosion, erosion and contamination risks from the core fluids are tremendous problems, even if the MSRE could show its operability.

In **Israel**

At Tel Aviv University and Brookhaven National Laboratory, in liaison with the Kurchatov Institute, Prof. Radkovsky, proposes a novel PWR thorium converter based on the seed-blanket concept of the Shippingport reactor, an intelligent concept fostered by Raytheon Corporation.

In **China**

Basic R&D is being conducted on nuclear data and ThO2 fuel, principally at the Shanghai Institute of Nuclear Research.

8.5. Conclusion

It is reasonable to assert that in a condition of high uranium ores demand for a large expansion of nuclear energy, as it was expected, the thorium cycle would have been developed in the seventies. However, the strong reduction of the nuclear programmes worldwide resulted in a practical stop of the development of the thorium cycle, except in India.

A novel expansion of nuclear energy is needed to launch the thorium fuel cycle industrially, at least in Western Europe.

Meanwhile, basic data, small scale experimental results are being collected. The Indian Programme continues. Progress is done on remote fuel fabrication techniques (like the sol-gel process at the EC Institute for Transurane in Karlsruhe) and practical experience on that subject is also obtained in the new MOX fuel fabrication plants. It is to be hoped that a small prototype accelerator-driven reactor (homogeneous reactor for example) be built to test the concept.

Whereas the added difficulties of thorium fuel reprocessing can be overcome, in our opinion, those of reprocessing coated oxide and carbide fuels from high temperature reactors are greater.

Thus the techniques which permit to take full advantage of simple thorium fuel cycles exist, like in heavy water reactors or light water reactors, but do not bring considerable economic advantages today; those for advanced concepts where thorium could bring a decisive advantage, as in HTRs, in the West at least, are not yet fully operational. Some thought should be given there.

In view of its potential advantages, the thorium fuel cycle has to be considered again as a promising energy source in - and after - the next century.

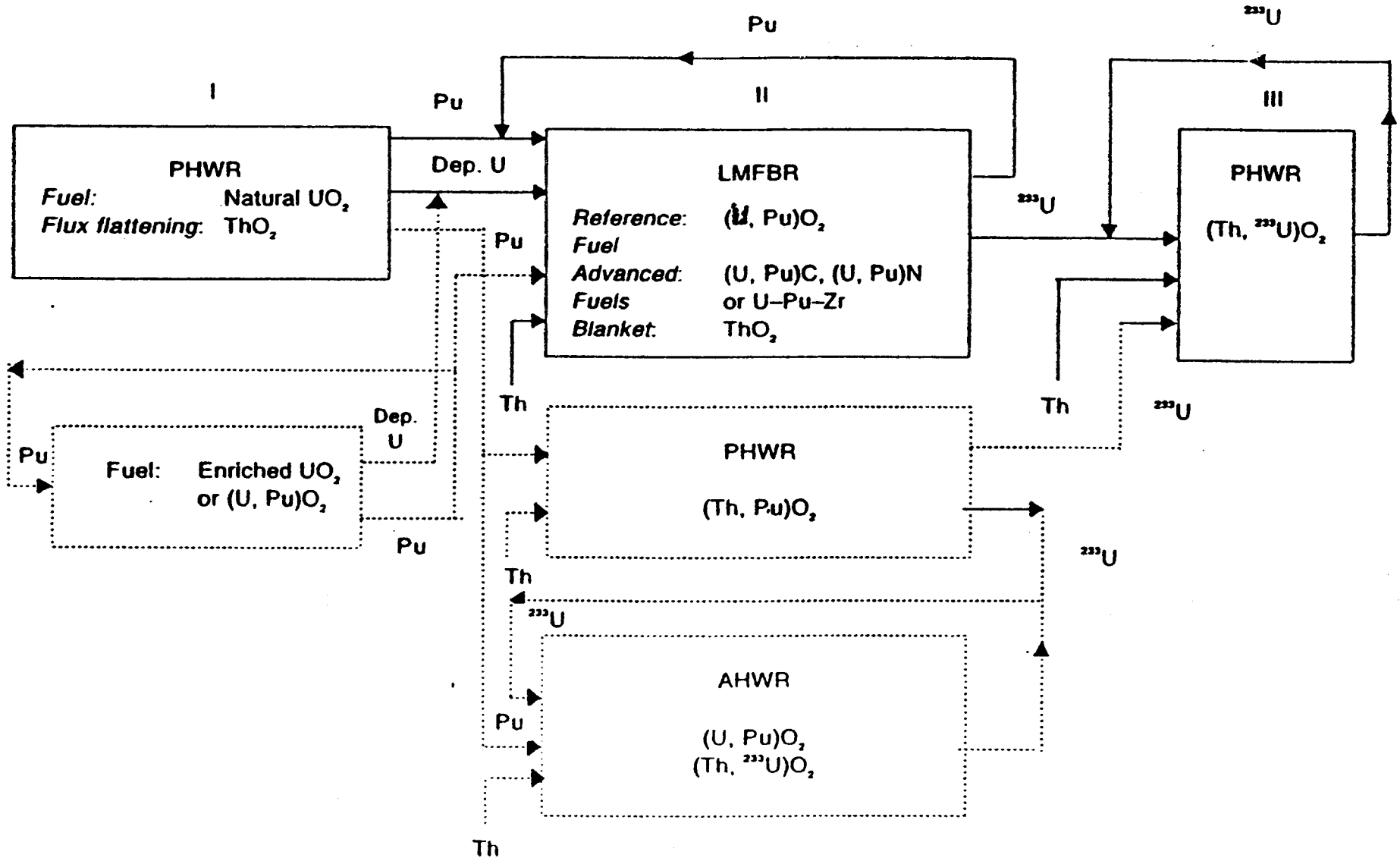


Figure 1. Nuclear fuel cycles and power programme in India.

9. CONCLUDING REMARKS

At the end of this survey, we are conscious of the difficulty to summarize 20 years of past - and rich - experience, and we apologize in advance for involuntary omissions or slight errors. We hope, however, that this work may help to find in a more condensed form a huge amount of data for which the memory of the actors of the time is progressively lost.

9.1. Observations

1) The large existing quantities of thorium to be found in earth's crust permit to at least double the potential energy contained in uranium, and save uranium for future generations. Thorium is an important energetic resource, whatever its disadvantages and advantages;

2) Thorium is not a fuel in itself, but it is a good breeding material for uranium-233, which seems to present many advantages as a fissile material;

3) Experience so far accumulated in the past 30 years shows that it could be possible to build an energy programme based on thorium, starting with fissile material U-235 or plutonium together with thorium. All the different steps of this industry have been tested at pre-industrial stage;

4) A Th/U-233 fuel cycle will need solid nuclear infrastructures with thorium irradiation reactors, and integrated reprocessing/fuel fabrication plants;

5) Once U-233 has been made available, its remarkable neutronic properties would be a bonus :
   - good neutron economy : smaller hold-up, thermal conversion,
   - good neutronic properties combined with thorium oxide, for High Temperature Reactors,
   - lower content of minor long-lived actinides in the spent fuel and reprocessing waste, compared to uranium;

6) U-233 breeding from thorium can be achieved with U-235, or Pu, (weapon's Pu for example) or U-233 : this permits a great deal of versatility,

7) The costs of a Th fuel cycle today, with the right infrastructures, may be estimated to about 20 % above those of a U fuel cycle, but these two will eventually become similar due to the experience gained with remote fabrication of MOX fuel elements, and the impact on the cost of kWh will be small anyway.

8) The main technical problem lies in the remote fuel fabrication with U-233, but today a good deal of these difficulties are already met with MOX fuel fabrication. Another complication is in the reprocessing which obliges to use particularly strict technical procedures, especially if zirconium cladding is present.

   The problem of reprocessing oxycarbide kernel fuels (with or without thorium) must be addressed separately from the thorium fuel cycle itself.

9) The Accelerator-driven System claims a number of theoretical advantages, probably more interesting from the point of view of small reactor hold-ups, flexibility of operation, degradation of the long-lived actinides, than in reactor safety and power generation as such. The cost of such a system may be an impediment. This is true with thorium or uranium as well.

9.2. General considerations

1) For those who have no thorium available, or simple infrastructures, it does not seem to be worth to embark on a thorium fuel cycle.

2) For those who have thorium and a strong nuclear infrastructure, thorium should be considered for the longer-term, hence a minimum R&D on thorium should be kept alive.

3) For those countries which have a huge energy demand, thorium is a good solution to pursue, after that of uranium, especially if the U reserves are limited : this is the case of India, for example, but it could be the case of others (China, Turkey ?).

4) In the farther future, thorium seems to be a natural complement to uranium, for fission or fusion reactors, from the point of view of its natural resources.

5) The encouraging results obtained with the LWR "breeder" at Shippingport and with the High Temperature Reactors prototypes should pave the way to develop similar, if not identical, reactor types.

6) If the necessity arises, it is possible to resort to thorium. However, the important knowledge and experience accumulated in the West some 30 years ago should be kept, and this may become a problem if no efforts to that end are made.

9.3. Recommendations for further developments

1) In spite of a large number of accumulated data, there seems to be a need for better neutronic data for multigroup computations.

2) Closer cooperation with the "India Thorium Experiment" should be considered.

3) Construction of a "seed-blanket" prototype on the lines of Prof. Radkowsky's recommendations seems advisable, using also the Shippingport experience.

4) The encouraging preliminary results obtained so far with the Accelerator-driven Reactor concept have to be consolidated and a pilot plant seems advisable.

5) Close-coupled reprocessing/refabrication plants should be studied from the point of view of remotization and costs.

6) The HTR concept should be revisited, in our view, to take advantage of the better thermodynamic yields offered by these reactors, with limited reactor waste and remarkable fuel stability for once-through cycles (or ulterior reprocessing, e.g. after 50 years). In parallel, pure oxide fuelled HTR fuel elements compatible with a simple reprocessing, should be studied, to arrive possibly to industrially more viable and economic solutions from the point of view of the fuel cycle. (Practically, HTRs can start with a well-known uranium fuel cycle, and could later use thorium).

7) Finally, it is important to keep the memory of a large sum of past experience. This document is a modest contribution.

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M. Lung

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Recently, a renewed interest in thorium has been expressed by distinguished scientists. Thorium as a nuclear breeder fuel, and the thorium/U-233 fuel cycle, could present some advantages as a back-up to the conventional uranium fuel cycle, and in matters of safety, non-proliferation, etc.

This work is a survey and compilation of the physical properties of thorium as a nuclear fuel, R&D on practical applications in the past and at present, (especially in India) in matters of resources, mining and metallurgy, fuel fabrication, prototype reactors, fuel reprocessing and waste disposal. Comments are given on past and present projects and on possible applications in the future.

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