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ABSTRACT

The use of thorium as an alternative or supplementary nuclear fuel is examined and compared with uranium. A description of various reactor types and their suitability to thorium fuel, and a description of various aspects of the fuel cycle from mining to waste disposal, are included. Comments are made on the safety and economics of each aspect of the fuel cycle and the extension of the lifetime of nuclear fuel.

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THORIUM; THORIUM 232; THORIUM CYCLE; NUCLEAR FUELS; THORIUM REACTORS; BWR TYPE REACTORS; PWR TYPE REACTORS; LWBR TYPE REACTORS; CANDU TYPE REACTORS; MOLTEN SALT REACTORS; LMFBR TYPE REACTORS; GCR TYPE REACTORS; GCFR TYPE REACTORS; HTGR TYPE REACTORS; REACTOR SAFETY; PRODUCTION; ORE PROCESSING; FABRICATION; FUEL FABRICATION PLANTS; REPROCESSING; SOLVENT EXTRACTION; HEAD END PROCESSES; COST; HIGH-LEVEL RADIOACTIVE WASTES; LIQUID WASTES; SOLID WASTES; SOLIDIFICATION; RADIATION HAZARDS; MINING; PROLIFERATION; THORIUM ALLOYS; THORIUM OXIDES; THORIUM CARBIDES

PREFACE

This report is a critical review of the state of thorium use as an alternative fuel but, with a few exceptions, does not take detailed account of developments later than 1977. However, before publication a bibliography of later work was added. (See Section 16).

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(Continued)

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1. INTRODUCTION

The constantly increasing demand for energy (Figure 1) has resulted in the continually increasing production of coal, oil, natural gas and uranium. In some areas, e.g. oil in the USA, it has been shown that the change in production rate with time follows a bell-shaped curve. The area under that curve represents the total recoverable oil; the highest point is the peak production rate. This type of curve can be fitted to production rates for other types of fuel reserves wherever a large reserve is mined by several operators, as in the case of oil, coal and natural gas in the USA [Hubbert 1969].

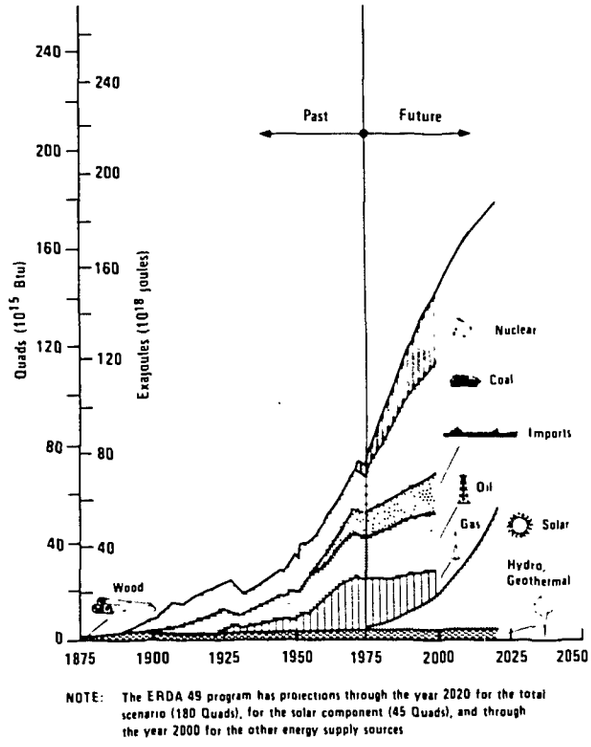

FIGURE 1 ENERGY PROJECTIONS OF THE ERDA-49 PROGRAM

When this type of calculation is applied to the world estimated initial recoverable reserves of coal (2400 x 10^9 t), the peak production occurs about the year 2150, and for crude oil (world reserves of 1250 x 10^9 barrels), the peak occurs about the year 2000 [Yannacone 1975]. Because of the very uneven distribution of these resources, there could be large variations in the time at which peak production occurs. For instance, 83 per cent of all coal resources is shared between the USA, USSR and China, whereas 3.2 per cent is shared by the continents of Africa, South America and Australia. Calculations of the Australian black coal reserves indicate that peak production would occur around 2120 using the rate of increase in production that has occurred between the years 1918 to 1972, or around 2050 using the production increases

over the years 1960 to 1972 [Stocks and Faulkner 1975]. Two interesting points are brought out in this type of calculation: a 50 per cent change in the reserves will make only 30 years' difference to the year of peak production, and shortfalls in production could occur around 2020.

Similar arguments apply to oil; the probable annual oil discovery rate in the 1980s has been predicted to be about 2×10^8 barrels. This is equivalent to only a few weeks' supply at current rates [Menard and Sharman 1975]. Such calculations do not allow satisfactory predictions; the fluctuations in energy requirements due to economic recessions and recoveries, and the increasing demands on fossil fuels as basic materials for the pharmaceutical and plastics industries, introduce large perturbations into the overall picture. All that the trends show is that the lifetime of fossil reserves is of the order of one or two hundred years, but shortages and consequent increase in costs, and the necessity to change energy sources will appear very much earlier.

To extend the energy resources, it is most likely that as many practical alternatives as possible will be used. One successful alternative is nuclear power which, although a satisfactory source of energy, will have similar resource limitations to fossil fuels if the current non-breeder operations continue.

At the most disadvantageous production costs envisaged, Lieberman [1976] predicted 1992 as the year of exhaustion of American uranium at ore concentrations greater than 600 μg/g. This prediction was made at the end of a period of eight or nine years, during which time no significant new uranium bearing region had been found in the USA. Since then, a mine has been started in California which originally was thought to contain an uneconomic 74 t of uranium, but which is now estimated to contain up to possibly 4600 t [Nucleonics Week 1978]. Apart from such reassessment of known reserves, new discoveries, such as the Key Lake ore body which may be the largest deposit in Canada, are still being made [Nuclear Energy International 1976]. Such finds, however, cannot continue indefinitely, eventual exhaustion being delayed as long as possible by the use of low grade ores, and the extraction of uranium from lignite, shale, seawater, etc., at greatly increased cost and increased scale of mining operations.

It is clear, therefore, that unless breeder reactors are brought into use, uranium reserves will have a short lifetime; only about 1 per cent of

the resources would be utilised, yielding about 824 EJ*. This represents some 2.5 per cent of the total world energy resources and will have the same effect as finding new coal or oil resources, i.e. it will only extend the time to peak energy production by a few years.

If breeder reactors are used, however, uranium would increase the total world energy resources by a factor of 1.5 to 2.5, and with thorium the same range of factors would apply [World Energy Conference 1974; Kleppe and Fisher 1976]. Thus the world's energy resource base can be more than doubled using the thorium cycle, in which ^{233}U is produced from ^{232}Th .

Thorium is about three times as abundant as uranium [Frondel 1956] and, if properly managed, could extend the lifetime of fission-produced power to around the year 2500 with fuel conversion, or even longer if fast breeders are used. The thorium cycle has been neglected in the past because the high costs involved in the development of nuclear power have made it impracticable to develop the two fuel cycles simultaneously. Increasing awareness of finite fuel resources appears to be changing attitudes towards the use of thorium. Over the years, many interested groups have examined the use of thorium either in existing uranium-fuelled reactors or in new systems designed specifically for use with thorium fuel, e.g. a number of public utilities in the US have formed a group to study the utilisation of high temperature, gas-cooled reactors (HTGCR) [Nucleonics Week 1977].

Examples of reactor types considered for adaptation to thorium fuel are the light water-cooled and moderated reactors (LWR: USA, Italy), pressurised heavy water-moderated reactors (PHWR: Canada, France) and graphite-moderated high temperature reactors (HTR: UK, Germany, USA). The USA, India and the USSR have considered the use of thorium in liquid metal-cooled, fast breeder reactors (LMFBR).

Systems designed specifically for the thorium fuel cycle are the molten salt reactors (MSR: USA) and the suspension slurry system (The Netherlands). A fuller description of the various types is given in Section 4. Only the HTR is nearing short-term commercial viability; its success in extending the resource base depends on the conversion ratio achieved.

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* EJ = exajoule. 1 EJ = 10^{18} J. 1 British thermal unit (Btu) = 1055 J. Another term in current use is the Q = Quad, short for quadrillion Btu's. 1 Q = 10^{15} Btu.

Kasten [1970] published an excellent review on the general utilisation of thorium and more specific reviews have been made of thermal breeder reactors [Perry and Weinberg 1972], HTGCRs [Landis 1973; American Nuclear Society 1974] and thorium fuelled LMFBRs [Seghal et al. 1975a,b] The physical and chemical properties of thorium were reported by Rand et al. [1975]. More recently, a number of papers on thorium fuel were presented at an International Conference on Nuclear Power and Its Fuel Cycle [IAEA 1977].

2. THORIUM USAGE AND RESOURCES

Thorium, discovered by Berzelius in 1828, is a widely distributed element making up some 12 to 15 micrograms per gram of the earth's crust [Rand et al. 1975; Frondel 1956]. Although the ultimate amount of thorium is about three times that of uranium, thorium occurs in fewer geologic environments and the bulk of the material may be more dispersed, residing in much lower grade accumulations [Searle and Platt 1975]. In 1970, the annual world consumption was about 270 t, of which about 50 per cent was used in the production of gas mantles and a gradually increasing amount (40 per cent) for metallurgical processes [Garg et al. 1977]. The remainder was used in refractories, catalysts and nuclear reactors. Estimates by the US Bureau of Mines put the demand for ThO_2 in the year 2000 as follows: non-nuclear uses as above, 450 to 620 t; nuclear reactor use 700 to 6500 t, depending on the degree of acceptance of thorium-fuelled systems.

A demand for up to 6500 t of ThO_2 per annum could be met as by-products of the extraction of rare earth elements from monazite sands and of the extraction of uranium. The increasing use of these materials will continue to keep thorium production well ahead of consumption. The available thorium supplies are estimated to be 0.5 Mt and inferred stocks are over 1 Mt, compared with the cumulative thorium usage of 40 000 to 80 000 t up to the year 2000.

Because the market for thorium is small, the resources and their recovery cost are not well known. The judgement of a resource as low cost sometimes depends on whether thorium is the principal metal mined or is a by-product from the mining of some other metal. Thus the Indian monazite sand reserves, which contain 8 to 10.5 per cent ThO_2 and are expected to yield 363 000 t of ThO_2 [Udas et al. 1977], were judged as low cost in 1965 but high cost in 1974. Information on this resource has been included in reports by the

European Nuclear Energy Agency (ENEA) Study Group [OECD 1968], the Organization for Economic Cooperation and Development [OECD 1973] and the World Energy Conference [1974] - see Appendix D, Tables D2-D5.

Kasten [1970] listed thorium resources in the USA as a function of recovery cost (Table D3) and predicted that this cost function would be applicable to the rest of the world. Thorium levels in seawater are much lower than uranium levels and cannot be considered as a potential source of future supply. Some production figures for monazite sands are shown in Table D4. The probability is that there is as much low cost thorium (< US10 per pound, 1975) as uranium. A summary of thorium resources and annual production figures is given in Table D5.

3. THE THORIUM FUEL CYCLE

3.1 Fissile Materials

Fissile materials are those isotopes which fission under thermal neutron bombardment. The only naturally occurring fissile isotope is uranium-235 (^{235}U); the other common fissile isotopes, plutonium-239 (^{239}Pu) and uranium-233 (^{233}U), are produced in reactors from the fertile elements uranium-238 (^{238}U) and thorium-232 (^{232}Th) respectively (see Appendix A).

3.2 Fertile Materials

The most common uranium isotope, ^{238}U (99.28 per cent abundance), is categorised as fertile and fissionable, i.e. it is fissionable only when bombarded with neutrons of energy higher than 1 MeV and fertile because it captures neutrons to form the fissile isotope ^{239}Pu.

Thorium consists of only one isotope, ^{232}Th, in the natural state, with properties similar to those of ^{238}U, i.e. it is not fissioned by thermal neutrons but, like ^{238}U, will fission with fast neutrons and capture neutrons to become, eventually, the fissile material ^{233}U (Figures 2a, 2b, 3 and 4) - see Appendix A.

Although the thermally fissile isotopes $^{233,235}U$, and ^{239}Pu can be assembled with a moderator and coolant to form a critical reactor, it is normal to include a non-(thermal neutron) fissile heavy element, usually ^{238}U

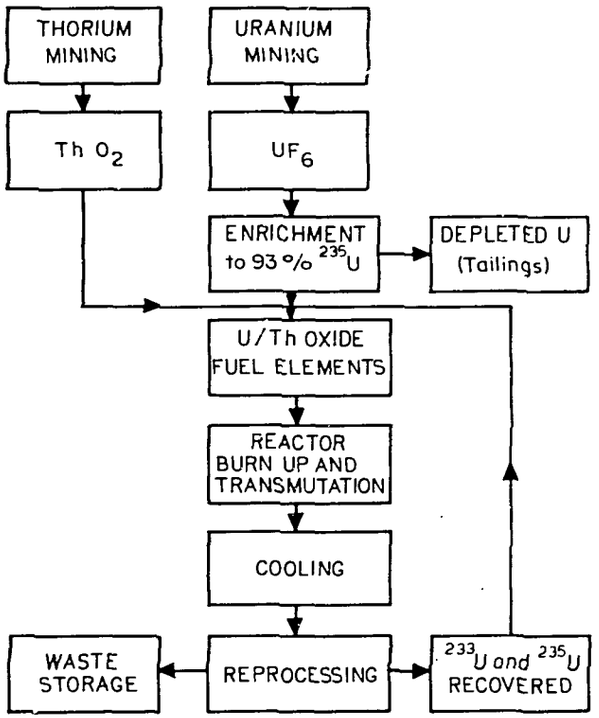

FIGURE 2a TYPICAL THORIUM CYCLE

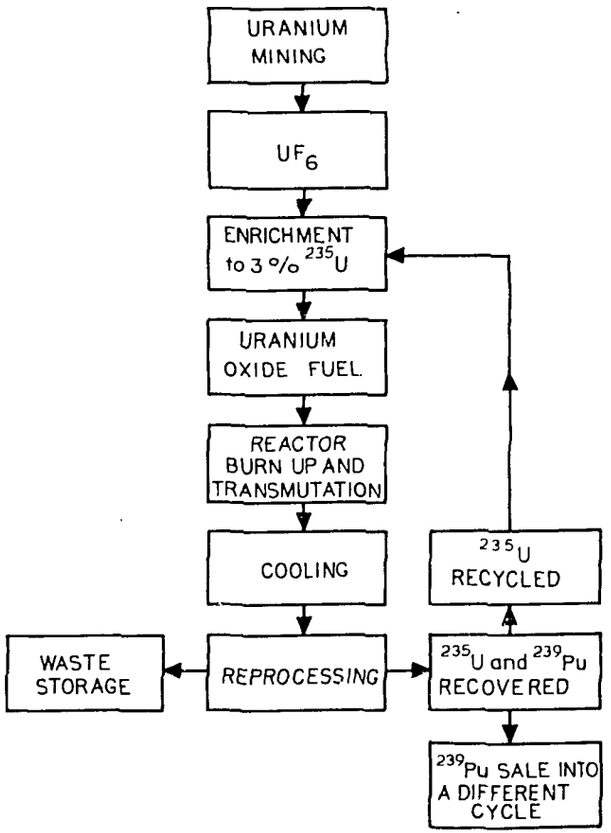

FIGURE 2b TYPICAL URANIUM CYCLE

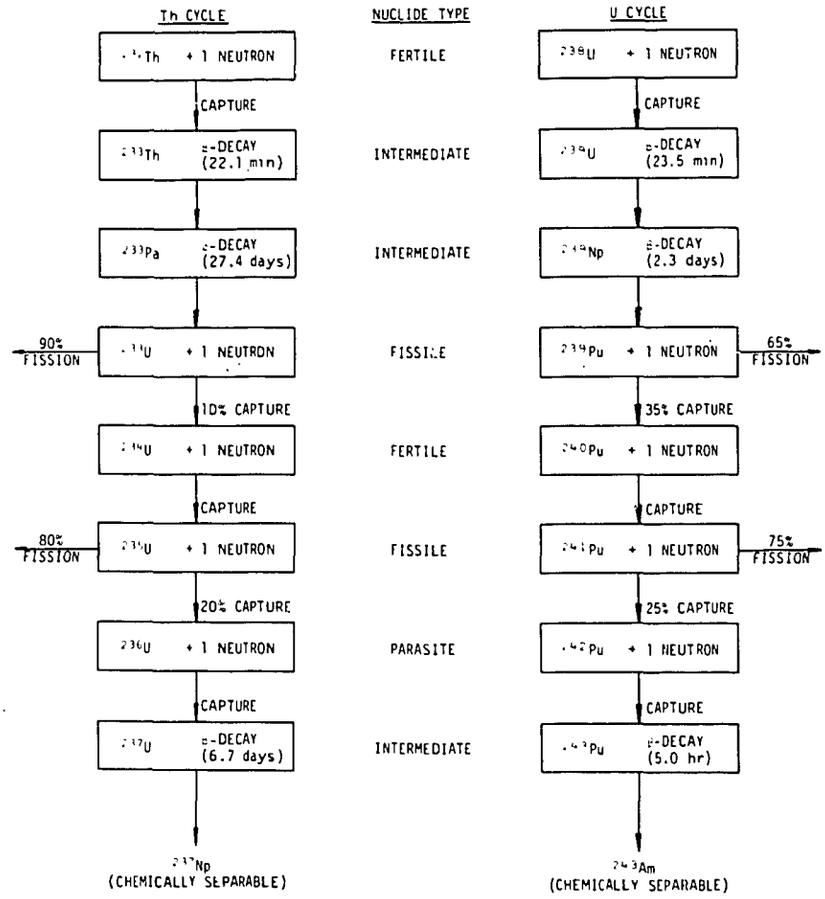

FIGURE 3 PRIMARY NUCLIDE CHAINS ASSOCIATED WITH THE THORIUM AND URANIUM FUEL CYCLES [after Kasten 1970]

or ^{232}Th, to give some reactivity control and reduce the radiation damage to the fuel which would arise from high power densities.

3.3 The Fuel Cycle

The fuel cycle covers all activities connected with the preparation and processing of nuclear fuel including mining, fuel preparation, reprocessing and waste disposal.

Unlike natural uranium, thorium cannot be used on its own as a nuclear fuel. Just as it is necessary to increase the ^{235}U content in natural uranium to fuel most reactor types, so it is necessary to add fissile material to thorium to make it suitable for use as a reactor fuel.

The fuel mixture, when exposed to neutrons within the reactor core, will undergo several transmutations (Figures 2 and 3) and a fraction of the fissile material will be burned up, eventually requiring replacement. At this stage, the fuel will be extremely radioactive and must be kept in a cooling pond until its radioactivity has decayed to a manageable level before it can be reprocessed.

During reprocessing, the contents will be chemically separated into waste material for storage or disposal, unchanged fertile material, unburnt fissile material and newly formed fissile material, i.e. ^{239}Pu from ^{238}U or ^{233}U from ^{232}Th. The unused fertile material may be disposed of or used again after purification; the newly formed fissile material may then re-enter its cycle, or be sold into another cycle. Figures 2a and 2b compare typical uranium and thorium cycles. The production of fissile material from fertile material is termed 'breeding' or 'conversion', depending on whether the original fissionable material in the reactor fuel is that produced from the fertile material, i.e. a reactor fuelled with $^{239}Pu/^{238}U$ breeds ^{239}Pu, whereas a reactor fuelled with $^{239}Pu/^{232}Th$ converts the thorium to ^{233}Th. It is common to use the term 'breeding' indiscriminately.

After the first recycle of the bred fuel, the composition of the fuel becomes much more complex with the uranium isotopes 233 to 236 being present in significant quantities owing to consecutive neutron capture. The ^{233}U bred from thorium has a relatively high η value* in thermal reactors in which it

* η is the number of neutrons produced for each neutron absorbed by the fissile material (see Appendix A: Figure A2 and Table A1).

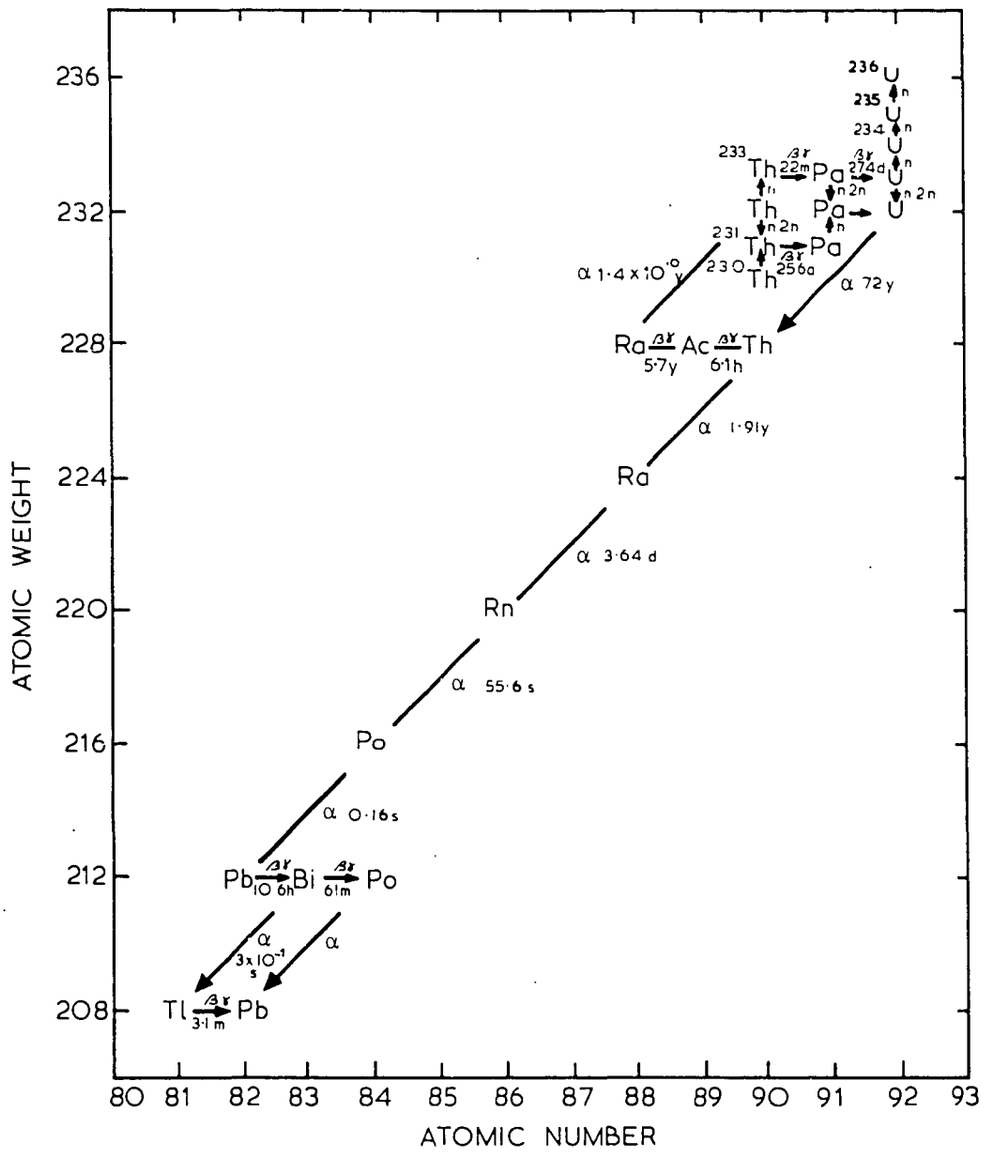

FIGURE 4 THE PRODUCTION OF FISSILE MATERIAL FROM ^{232}Th AND ITS DECAY SCHEME

TABLE 1

DIFFERENCES IN REACTOR PHYSICS PARAMETERS

BETWEEN THE VARIOUS FISSILE ISOTOPES

Parameter	Thermal	Fast
Potential breeding ratio	^{233}U superior	^{239}Pu + ^{241}Pu superior
Minimum critical mass	^{239}Pu and ^{241}Pu smaller	^{235}U, ^{239}Pu, ^{241}Pu smallest
Neutron capture cross section	Th significantly higher	Th slightly higher
	Resonance cross section generally lower for Th	
η the number of neutrons produced per neutron capture (Figure A1)	^{233}U superior, ^{241}Pu slightly lower	^{241}Pu superior, ^{233}U good at lower half of spectrum, Pu best at higher and best breeding region.
Doppler coefficient		Th produces more negative Doppler coefficient
Fast effect	U slightly bigger	U considerably bigger
Resonance integral	Favours U in heterogeneous systems, Th in homogeneous systems	Not important
Higher isotopes ^{234}U, ^{242}Pu, etc.	Favours Th	Favours U
^{239}Np, ^{233}Pa	Favours U but can be avoided in design	Little difference
Fission product poisoning	Favours Th	Little difference
Radioactivity of spent fuel	The production of ^{232}U daughter products significantly favours the U cycle	As for thermal
Delayed neutron fraction	Power more sensitive to reactivity changes with U/Pu but no significant economic disadvantage	Little difference
Energy release/fission	Very slightly favours U	As for thermal

TABLE 2

REACTORS USING THE Th FUEL CYCLE - ADVANTAGES AND DISADVANTAGES

Reactor Type	Advantages of Th Cycle	Disadvantages of Th Cycle
BWR & PWR	More economic if metallic fuel elements can be developed. Failure to develop other reactor types would favour Th cycle.	Power density would have to be increased by ∿15 per cent before the Th cycle would show a significant cost advantage.
LWBR (Seed blanket)	Good neutron economy; could be used if necessary in a reactor designed for U fuel. Metallic fuel elements would improve economics above that of U-fuelled PWRs. Remote fuel fabrication not required.	Th used in a U reactor reduces the power output by 20-40 per cent. In the Th designed system, unchanged Th from the blanket is diverted as waste, thus the resource is not fully utilised.
HWR (i) Pressure tube	Efficient use of resources. A self-sufficient equilibrium thorium cycle is possible allowing a large return of energy over a long period. CR ∿1.0.(a)	Little prospect for significant breeding without changing current operating conditions and increasing capital costs. This system applies to CANDU reactors but Canada has little experience in fuel cycle technology.
(ii) Pressure vessel	Homogeneous reactors could use Th cycle if necessary Aqueous suspension reactors (fuel as slurry). Good neutron economy and prompt negative temperature coefficient. Continuous fuel processing. CR in range 0.95-1.05.	Very little information available and what there is suggests that there is no incentive to use the Th cycle. Erosion problems at the pump impeller would have to be solved. Nothing approaching a commercial size has been designed. The on-line fuel processing has not been demonstrated.
Gas-cooled, graphite moderated reactor (excluding HTGCRs)	None.	High fissile inventory required. These would be about the last types to be considered for the Th cycle.

(Continued)

TABLE 2 (Cont'd.)

Reactor Type	Advantages of Th Cycle	Disadvantages of Th Cycle
Molten salt reactor	Potentially favourable economics, fuel utilisation and safety characteristics. High thermal efficiency. CR in range 0.85-1.07. Continuous reprocessing.	Considerable chemical and metallurgical progress needs to be made. Corrosion problems to be overcome.
FBR	With sodium cooling - a negative sodium void characteristic.	Poor economics compared with uranium cycle.
	Gas cooled. Net fuel cycle cost slightly favours Th cycle.	Slight increase in enrichment required.
	Metallic Th systems capable of higher breeding ratios than PuO_2/UO_2.	Lack of experience with metallic fuels. High burn-up (>7½ per cent) needs to be achieved.
	Symbiotic systems - see HTGCR.	
HTGCR	High temperatures allow the use of He gas turbine and process heat with very high fuel utilisation. Very high temperatures possible with little fuel development. CR in range 0.66-0.97 or even to 1.15.	At the higher temperature H_2O vapour is a problem to be solved. At BRs > 0.85(b) a considerable economic disadvantage occurs.
	Flexible binary cycles permitting very high fuel utilisation.	
	Symbiotic systems with FBRs permitting self-sustaining cycles.	

(a) CR = Conversion ratio
(b) BR = Breeding ratio

} See Appendix A.7

would normally be recycled. However, these are not hard and fast rules and it is possible to find either the roles reversed, or mixtures of all types of fissile material being used to suit particular design features.

From the above discussion, it is seen that if uranium and thorium cycles are both in use, fuel supplies become much more flexible and a short-term lack of one or other raw material or enrichment capacity need not be serious. Interchangeable fuel cycles are possible for some reactor systems (Section 4). In other reactors, a mixed cycle might be desirable for safety reasons, e.g. it is possible to produce a negative sodium void coefficient in a sodium-cooled fast reactor with ^{233}U or $^{239}Pu/^{232}Th$, but not without some diminution of the breeding ratio.

In general terms, the physics or neutronics principle of fission reactors is the same, regardless of the fuel used. It is therefore reasonable to expect that a particular reactor type could be made to operate with any fissile material provided that there was enough of it or it had the right enrichment. However, optimisation of the reactor involves the spacing of the fuel elements, their enrichment, and the type of moderator material; it is also governed by the physical parameters of the fuel (Table 1). Large changes in core geometry to accommodate a change in fuel would be out of the question on economic gounds, but some reactor types such as the HTGCR, are quite flexible in that the main fissile element in the fuel can be changed to suit the prevailing market conditions with very little effect on their economic performance.

At present, no country is known to have a plant manufacturing or reprocessing thorium-based reactor fuels on a commercial scale (see Sections 7, 8 and 9 and Zimmer et al. [1979]). However, the technology required for the thorium fuel cycle is very similar to that for the uranium fuels. The HTGCR systems will almost certainly use a $^{232}Th/^{233}U$ fuel cycle. France, Germany, USA, India and probably the USSR, have plans for commercial HTRs using thorium in the next century.

4. THORIUM-FUELLED REACTOR CONCEPTS

Table C1, lists reactors and critical assemblies that have used thorium and Table 2 shows the suitability of various reactor types for thorium use. A number of reactor types are suitable for use with this cycle and its

implementation would extend the fuel resources and help to make the cost of energy less sensitive to the cost of mining. Table 2 shows the effect of certain physical parameters on thermal and fast reactors; Table 3 compares the size and certain nuclear parameters of several of these reactors in their normal state, that is, using the fuel for which they were designed. It may be noted that overall, core size is much the same for thermal reactors; the variation in the table is due mostly to variation in power output. The liquid metal-cooled fast breeder reactors have smaller cores and the gas-cooled fast breeder will be even smaller. In general, the separation of the fuel rods or bundles of pins, depending on the type of moderator, is 1.5 to 2.0 cm for LWRs, 27 or 28 cm for heavy water reactors (HWRs) and 48 cm for advanced gas-cooled (graphite) reactors (AGRs). The microspheres of the HTGCR prismatic fuel element can be in fuel channels within the graphite elements or distributed throughout the graphite, whereas the 6 cm diameter spheres of fuel or fertile materials in the pebble-bed HTGCRs are arranged randomly within the pressure vessel.

Thorium could be used in each of these reactor types with greater or less advantage, but optimisation either for low cost or extension of resources will require modifications to lattice size, heavy metal content, power density, etc.

4.1 The Light Water Reactors, BWR and PWR

The present generation of LWRs use the uranium fuel cycle (Figures 5 and 6). The Indian Point 1 plant of Consolidated Edison (which went critical in 1962) at Buchanan, New York, USA, originally fuelled with thorium and uranium, is now fuelled with uranium only, for economic reasons. The relative performance of the thorium and uranium fuel cycles in LWRs is strongly dependent upon such factors as the cost of reprocessing and refabrication and the effective fuel inventory charge rate. These costs are not well established for the thorium cycle but, using a reasonable 15 per cent per year as the charge rate, $US25 for the cost of U_3O_8 and $US75/kg SWU*, Kasten and Tobias [1975] favour pressurised water reactors (PWRs) over boiling water reactors (BWRs) in the thorium cycle and have established that the uranium cycle would have about 1.5 mills+ per kWh(e) advantage over thorium because

* SWU = separative work unit. A measure of the effort required to enrich uranium, it is a function of the degree of enrichment, the ^{235}U content of the feed material and of the tailings.

\+ 1 mill = 0.001.

TABLE 3

COMPARISON OF REACTOR TYPES

Reactor Type and Name	Core Size (cm)		Lattice Type and Size (cm)		Pin/Rod o.d. (cm)
	Diameter	Height	Fuel Elements	Pins/Rod Separation	
BWR, Shimane No.1	344.0	366.0	Square 30.5	1.88	1.24
PWR, Millatone 2	347.2	347.0		Square 1.47	1.12
PHWR, Atucha	454.3	530.0	Triangle 27.1		1.07
HWR (CANDU), Pickering	780.0	634.0	Square 28.58	28 rod bundle inside 10.3 cm i.d. pressure tube	1.52
HTGCR, Fort St Vrain	594.0	475.0	Triangle 36.2	Fissile and fertile spheres are embedded in graphite moderator	Spheres: Fissile 0.1-0.3 Fertile 0.3-0.6
AGR, Hartlepool	930.0	820.0	Square 48.75	36 pin o.d. 19.0	1.45
LMFBR, Dounreay Prototype Fast Reactor	144.8 Surrounded by 1 row radial breeder 171.5 and 3 rows reflector to 243.8	91.5 Breeder and reflector to 146.3	Triangle 14.5	0.74	0.58
GCFBR, Dalle Donne Goetzmann [1976]	300.0	148.0		1.10	0.82

Z-2 = Zircaloy-2 Z-4 = Zircaloy-4

SECTION 1

TABLE 3

PARISON OF REACTOR TYPES

cm) Rod tion	Pin/Rod o.d. (cm)	Cladding Material and Thickness (cm)	Fuel and Enrichment	Burn-up (MWd/t) Average and Maximum	Conversion Ratio
38	1.24	Z-2 0.09	UO_2 2.09% initial 2.31% equilibrium	16 500 ave., 26 000 with initially 22 000 ave.equilib.	0.6-0.7
47	1.12	Z-4 0.10	UO_2 in 3 regions: 1.93%, 2.33%, 2.82%	22 000 ave. 50 000 max.	0.56
	1.07	Z-4 0.05	UO_2 Nat.	7 000 ave. 9 000 max.	0.814
e cm i.d. e	1.52	Z-4 0.038 ~0.047	UO_2 Nat.	8 000	0.81
res in erator	Spheres: Fissile 0.1-0.3 Fertile 0.3-0.6	Multiple layers of pyrocarbons and silicon carbide	Fuel $(U/Th)C_2$ with 93% enriched U, fertile ThC_2	100 000 ave. U+Th 200 000 max. U+Th	0.6
19.0	1.45	Stainless steel	Hollow UO_2. 1st change: 1.4% inner; 1.6% outer Fuel: 2.1% inner; 2.6% outer	18 000	
.74	0.58	Type 316 stainless steel 0.038	Inner zone 19% Pu 81% at 25% enrichment Outer zone 25% Pu 75% U at 30% enrichment	40 000 ave. 61 500 max.	1.12
.10	0.82	Stainless steel 0.05	Similar to LMFBR	100 000 max.	1.45

SECTION 2

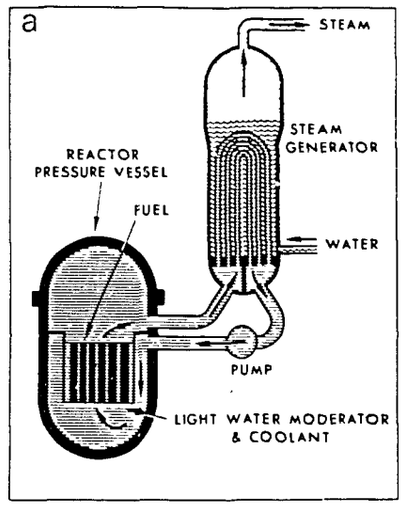

FIGURE 5 PRESSURISED WATER REACTOR (PWR) [after Robertson 1978]
Pressured light water is the moderator and coolant. The turbines are driven by steam produced in a heat exchanger, i.e. an indirect cycle. If all the light water is replaced with heavy water, this produces a larger size pressurised heavy water reactor (PHWR). With some modifications to the core, these systems could be used as light water breeder reactors (LWBR) - see Section 4.2 - and provision for mixing light and heavy water to a varying degree would result in a spectral shift control reactor (Section 4.2), another form of the LWBR. See Table 3 for core size, lattice, etc.

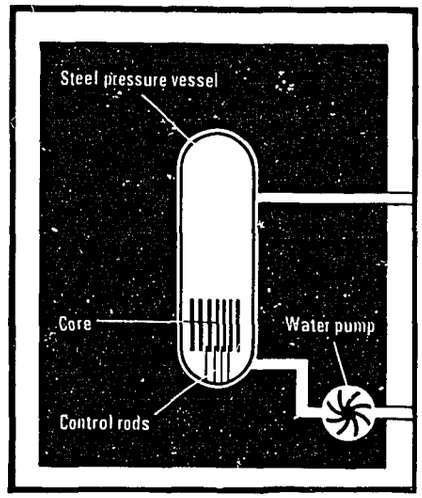

FIGURE 6 BOILING WATER REACTOR (BWR) [after UKAEA 1973/74]
Clusters of fuel pins inside a pressure vessel. Light water is moderator and coolant. A direct cycle is used, the steam formed in the pressure vessel drives the turbines.

the lower fuel inventory and reprocessing costs associated with uranium outweigh the higher conversion ratio and larger reactivity lifetime of the thorium. The increasing costs of separative work (US140/kg SWU from October 1981) may eliminate the uranium cost-advantage.

Using recycled Pu priced at 70 per cent of highly enriched ^{235}U,PuO_2 in ThO_2 in present PWR designs gives fuel cycle costs slightly less than those of the uranium cycle, assuming the establishment of a large fuel recycle industry. Failure to develop breeder reactors burning Pu would improve the prospects of Pu recycle in LWRs and this would favour the thorium cycle [Kasten 1970; Kasten and Homan 1977].

Zorzoli [1972, 1973a,b] calculated that by replacing the UO_2 fuel in a standard PWR with metallic Th and ^{233}U, and keeping the core geometry and thermal output unchanged, a significant reduction in fuel cycle costs could be achieved. Compared with a standard PWR with an average discharge burn-up of 33 000 MWd/t, the fuel cycle costs are reduced by 10 per cent at a burn-up of 10 000 MWd/t (U+Th) to 30 per cent with a burn-up of 25 000 MWd/t (U+Th). The calculations refer to a 1000 MW(e) PWR fuelled initially with Th/^{235}U but using ^{235}U as make-up for the recycled fissile material. If the core and plant of the reference PWR is redesigned to a lattice pitch of 1.36 cm instead of 1.43 cm, the fuel cycle costs will remain substantially unchanged, but the increased core power density should lead to a capital cost saving of about $5 per kW. This would reduce the cost per kWh by a further 7 to 9 per cent. The reduction in fuel cycle costs is achieved on two grounds; first the superior nuclear properties of ^{233}U will result in a higher conversion ratio, and second lower working capital, fabrication costs, and costs of UF_6 and separative work amount to more than the increased costs of reprocessing. (See Section 4.5 and Table A2 for metallurgical and radiation properties of thorium and uranium; see also Weissert and Schileo [1968].)

Zorzoli [1973a] further calculated that metallic Th-fuelled PWRs would achieve about 30 to 50 per cent saving on total uranium consumption by the year 2020. In the light of these costs and fuel savings, Zorzoli suggested that it would probably be more reasonable to improve PWR design than to proceed with light water breeder reactors.

4.2 Light Water Breeder Reactors

The seed blanket concept [Radkowsky 1962; Radkowsky et al. 1964] uses a central core of thin Th/^{233}U fuel pins surrounded by a fixed lattice of thicker Th pins (the blanket). The central core or cores can be moved up and down relative to the blanket to control reactivity. Thus there are no control rods to absorb the neutrons and consequently the neutron economy is substantially improved.

A very important feature of the light water breeder reactor (LWBR; see Figure 5) is that it can be installed in an existing PWR by rearranging the core and the top plate of the pressure vessel. The modifications are designed to increase the fuel-to-water ratio and the resulting increase in the neutron absorption in the thorium increases the conversion or breeding ratios. (See Appendix A7 for a definition of breeding and conversion ratios, etc.) However, the breeding gain is very small with conversion ratios close to unity and the specific power is low, reducing the available power to about 20 to 40 per cent less than that of the standard PWR [American Physical Society 1978].

Because of the low breeding gain, pre-breeders designed to operate with a high net production of ^{233}U are proposed [Kasten and Homan 1977] which imply an increased ore consumption during a long introductory period with savings only after very long operational periods. This system would appear to have a marginal advantage and therefore is not the best option to pursue.

The 'spectral shift control' reactor (SSCR) concept, in which the coolant is modified to improve the conversion ratio, may be a more realistic approach [Deuster and Levine 1960; American Physical Society 1978], but it would still be necessary to modify the core to get conversion ratios up to 1.0. The spectral shift control reactor works by having as coolant a variable mixture of light and heavy water. After each refuelling, the excess neutrons produced in a standard PWR would normally be absorbed in boron or some other non-fertile poison but, in the SSCR, the neutron spectrum is hardened by adding D_2O to the coolant (about 85% D_2O, 15% H_2O at the beginning of the cycle) and the excess neutrons are consumed by the absorption resonances of the fertile material. As burn-up proceeds, fewer neutrons are excess to requirements and it becomes necessary to reduce the neutron absorption in the fertile material. This is accomplished by increasing the H_2O content in the coolant (75% H_2O, 25% D_2O at the end of a one-year cycle) which becomes a more effective moderator and allows fewer neutrons to remain in the intermediate energy

region where the absorption resonances are so effective.

The good neutron economy and the higher conversion ratios attainable with this system offer a small advantage in terms of resource extension, but the added costs of the D_2O plant and the disadvantage of a higher tritium content in the coolant, making refuelling more difficult, are probably sufficient to make any uranium gains insignificant.

Lang [1975] suggested that LWBRs could ultimately be fuelled completely with thorium plus a fissile material. Initially, mixed loadings are used with thorium or thoria in an outer blanket, pins with added plutonium (preferably) as an inner blanket and a core region of enriched uranium. Ultimately, the mixed loadings will be abandoned as fast breeders are developed. In this scheme, it is envisaged that thorium with a low ^{230}Th content* is used in the blanket and the pins are discharged annually. When processed, the bred ^{233}U is added to depleted uranium and the plutonium added to new thorium in the inner blanket. The thorium and fission products left after removal of the ^{233}U would go to waste.

Figure 4 shows that if low ^{230}Th content material is used, the only way to arrive at ^{232}U is via (n,2n) reactions which have very small cross sections compared with neutron capture and occur only with very energetic neutrons. Since ^{232}U is the precursor of ^{228}Th and its daughter, which are sources of hard γ-rays, the need for remote handling of reprocessed fuel disappears. Uranium-232 at 5 mg/g can be achieved, but thorium ore not associated with uranium must be used otherwise the ^{230}Th content will be too high.

The advantages of the dual fuelling of LWBRs are as follows:

(i) The residual amount of ^{235}U in the reactor depleted fuel is eventually used since the replacement fuel will eventually be depleted uranium plus ^{233}U.

(ii) The thorium processing requirements increase as enrichment requirements decrease.

* By not using thoria produced as a by-product in uranium mining. See, for example, Boswell et al. [1966].

(iii) Abundant thorium supplies (Tables D2-D5).

(iv) Remote fuel fabrication will not be required since materials used are ^{233}U + Pu + new Th or ThO_2 + depleted U. Very little ^{232}U will be produced because it requires mainly (n,2n) reactions in the absence of ^{230}Th.

(v) Depleted uranium inventories will be reduced.

(vi) Transuranic isotopes and Pu inventories would be reduced (see Section 11, Tables 7 and 8).

(vii) Radiation damage to reactor hardware is reduced owing to the effective shielding of the blanket.

Ultimately, mixed loadings could become unnecessary if fast breeders are developed since abundant ^{233}U can then be produced, but with higher ^{232}U content [Mann and Scheuter 1978]. Since ^{233}U is likely to command a better price than Pu because ^{233}U produces more neutrons in an LWR than Pu, the revenues from such a breeder should be greater than from one using a uranium blanket.

On the basis of extensive irradiation experience with Zircaloy clad thoria and thoria/urania pellets obtained in the Westinghouse LWBR development program [e.g. Guessner 1969; Daniel 1971; Kreynes et al. 1976; Busby and White 1976; Crescimanno 1976; Eyles 1979], it has been shown that the irradiation performance of the fuel should be satisfactory since thorium oxide fuels show about half the swelling under irradiation and larger conductivity integrals than uranium oxide [Lafontaine et al. 1966]. The performance of PuO_2/ThO_2 fuels should be comparable to that of UO_2/ThO_2, but irradiation experience here is more limited. The higher thermal conductivity and higher melting point of ThO_2 are two factors in favour of the thorium fuel [Kasten and Homan 1977].

4.2.1 LWBRs with metallic thorium fuel

Zorzoli [1973b] concluded that it would be difficult to make the oxide fuelled LWBR attractive to electricity utilities; however, his figures show that metallic thorium is most attractive, with fuel costs between 0.75 and 0.5 that of a standard UO_2 fuelled PWR.

Experiments by Kittel et al. [1963] and Blumenthal [1963] on metallurgy and irradiation properties show that thorium metal and alloys of thorium with uranium and/or plutonium would be perfectly satisfactory fuels for PWRs, LWBRs and FBRs. In some respects, e.g. geometrical stability, these alloys are much superior to uranium.

4.3 Heavy Water Reactors

4.3.1 Pressure tube reactors

Pressure tube reactors (Figure 7) are two-fluid, large lattice systems in which the moderator is separated from the coolant. The Canadian CANDU reactor is a typical example. Since the moderator and coolant are physically separated, a variety of coolants can be considered, e.g. light water, organic fluids and gases [Atomic Energy of Canada Ltd 1974]. These fluids may also undergo phase changes (e.g. water to steam) as in the steam generating heavy water-moderated reactor (SGHWR).

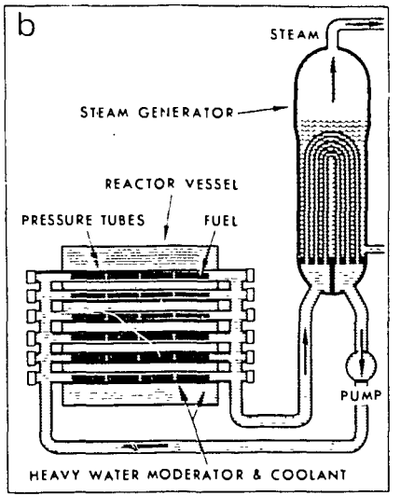

FIGURE 7 HEAVY WATER (PRESSURE TUBE) REACTOR [after Robertson 1978]
In CANDU reactors, the fuel elements consist of 28 or 37-rod clusters and these are contained within pressure tubes. The D_2O primary coolant flows through the pressure tubes and through a heat exchanger or steam generator. The H_2O steam produced drives the turbines. D_2O moderator is contained in the reactor vessel surrounding the system of pressure tubes. (See Table 1 for other details). Replacing the D_2O in the pressure tubes by an organic liquid produces the heavy water, organic-cooled reactor (HWOCR).

In 1963-64 in the United States, the Savannah River Laboratory provided plant and core designs for CANDU reactors using uranium and thorium fuel cycles. These designs were then evaluated at Oak Ridge National Laboratory (ORNL) where it was shown that the thorium cycle would have a distinct economic disadvantage.

Relative to ^{235}U, plutonium produces more neutrons per fission in fast reactors than in thermal reactors (Figure A2 and Appendix A). A better financial return should therefore be realised by refuelling a thermal reactor with enriched uranium and selling the plutonium produced to the fast reactor operators. This is called the ^{235}U-^{238}U/Pu (sale) fuel cycle. With this cycle in mind, Lewis [1968] envisaged the Valubreeder, a CANDU reactor using a mixture of ^{232}Th and ^{238}U as fertile material. Because the ^{233}U produced is worth more than the ^{239}Pu (USAEC purchase price in November 1967 was \$13/g ^{233}U; \$9.28/g ^{239}Pu), the cost of the power produced is marginally less for the Valubreeder. Although it lacked a worthwhile economic advantage, it was seen that engineering improvements allowing thinner pressure tubes would lead to a considerable increase in attainable uranium burn-up and therefore an economic and resource gain.

The coolant within the pressure tubes of the CANDU or similar reactors could be an organic liquid such as a terphenyl; these systems have been examined quite thoroughly. Extensive comparisons [USAEC 1968] of the uranium and thorium cycles in a heavy water-moderated, organic-cooled reactor (HWOCR) showed that the use of an organic coolant marginally improved the economics but improved the uranium cycle rather more. A later study [Whiteshell 1974] of a 500 MW(e) HWOCR using natural uranium as fuel showed a 10 per cent cost advantage over heavy water cooling. Lewis [1975] followed this with a comparison of the uranium and thorium fuels in a CANDU reactor of the type installed at Pickering (Canada). This system allows higher coolant temperatures which result in smaller turbines and increased efficiency. A higher power density can be used, resulting in a smaller core and consequently a smaller heavy water inventory; the latter is further reduced by the fact that the pressure tubes are filled with cheap, organic coolant. These improvements halved the calculated production cost of the electricity generated by the Pickering power station.

Critoph et al. [1975], Critoph [1977], Hatcher et al. [1975] and Slater [1976] directed their attention towards the efficient use of fissile material rather than the total unit energy cost. These authors showed that a 'self-

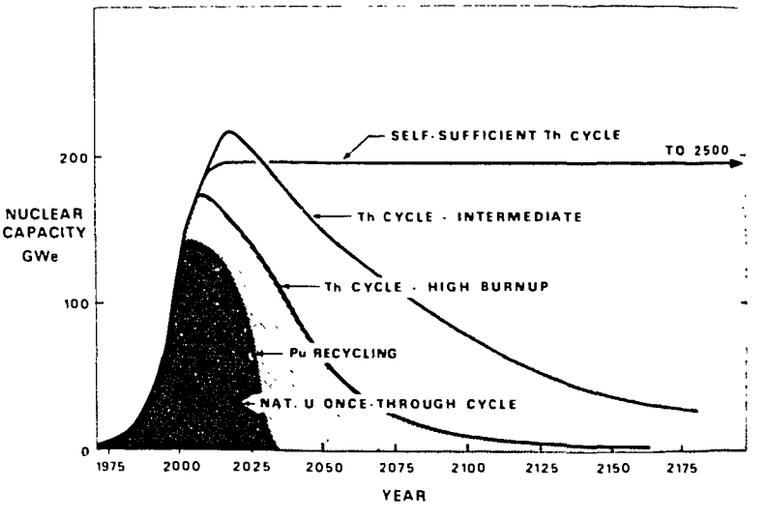

FIGURE 8 ENERGY FROM $3X10^5$ Mg URANIUM WITH VARIOUS CANDU CYCLES [after Robertson 1978]

sufficient equilibrium thorium' (SSET) cycle is a reasonable proposition. This cycle uses Pu from a natural uranium fuelled CANDU-PHWR to enrich a uranium/thorium mixture in another reactor of this type. After the fuel has been recycled many times, an equilibrium condition of constant isotopic content is established. For the SSET cycle, there is no need to 'top up' the feed fuel (max. recycled uranium) with fissile Pu hence, when this stage has been reached, natural uranium reactors are not needed to supply the plutonium, and only thorium needs to be added.

At equilibrium, an SSET cycled system would produce 200 GW(e) from 3 x 10^5 t of uranium for an almost indefinite period; production is compared with other cycles in Figure 8. Under these conditions, the system is a converter with a conversion ratio of 1.0. Neutron balance calculations [Slater 1976] show that if all non-productive neutron absorption could be avoided, a conversion ratio of 1.3 could be achieved. However, while such absorption could be reduced considerably, it is likely that a reduction in power density would be necessary, implying higher capital costs, a large fuel inventory and the need for mid-cycle fuel changes, which could lead to increased operational costs. Irradiation and other studies are in progress to substantiate these estimates [Fanjoy and Bain 1977; Foster and Russell 1977] - see Section 16, especially reports from Ontario Hydro, for later developments.

Another possible advantage of this type of system is its ability to burn up excess plutonium. Figures 9 and 10 [Slater 1976] show that increased plutonium content causes a significant increase in burn-up, accompanied by a fall in the conversion ratio. It would obviously be possible to optimise the plutonium feed to extract the greatest amount of energy from excess plutonium stockpiles and total thorium availability (see, for example, Dormuth and Lidstone [1977]).

To summarise, it seems possible to achieve SSET cycles in CANDU-PWRs which would significantly extend the energy obtainable from all the fissile forms by allowing practically complete burn-up of uranium and excess plutonium with a ceiling on energy costs of no more than 25 per cent more than at present [Robertson 1976]. However, development of a significant breeding fuel cycle would decrease current operating capability and increase capital costs. Canada has had little experience in fuel reprocessing technology and is "consequently embarking on a 20-25 year program covering the major elements of fuel reprocessing waste management, recycle fuel fabrication and fuel testing, together with full investigations of the economic, safety, health and security

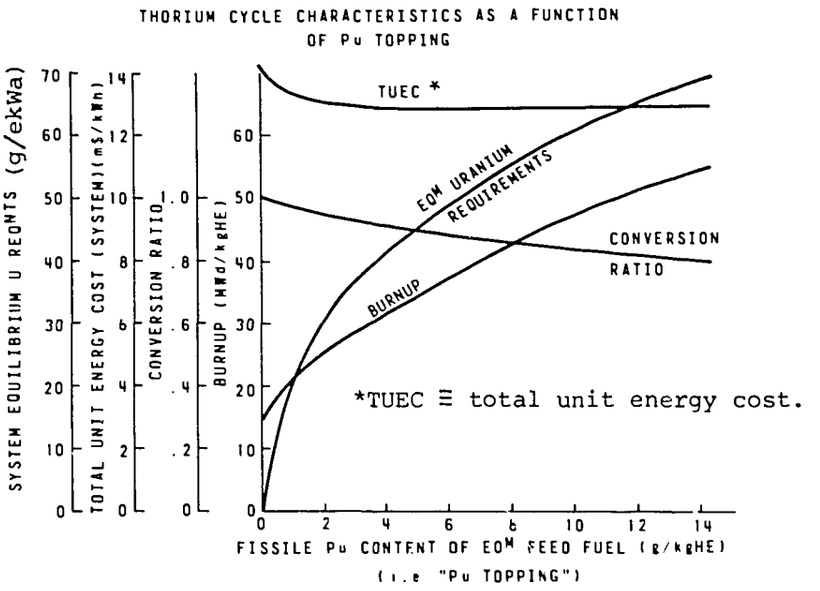

FIGURE 9

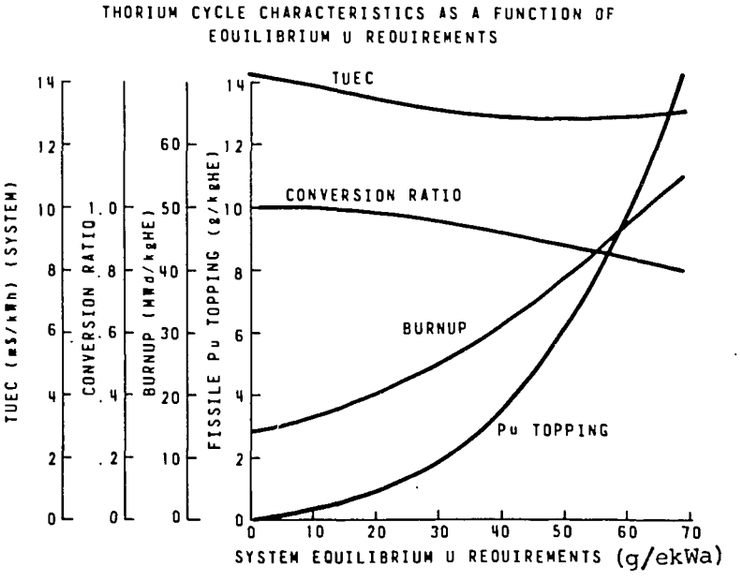

FIGURE 10

FIGURES 9-10 CONDITIONS AFTER ESTABLISHMENT OF ISOTOPE EQUILIBRIUM [Slater 1976]

aspects" [Went 1977] - see Section 16, especially reports from Ontario Hydro.

4.3.2 Pressurised heavy water reactors (Figure 5)

Pressure vessel reactors (PWRs or BWRs) use heavy water, thus requiring a physically larger reactor, but have reduced fuel enrichment requirements. The 12 MW demonstration reactor at Marviken, Sweden [Bergstrom and Nordinder 1969], which was shut down in 1974, and the first South American reactor at Atucha, Argentina [Herzog and Sauerwald 1969] are examples of this type of reactor. There is little published information on the comparative performance of the uranium and thorium cycles in these reactors; however Kasten [1970], making some reasonable but unproved assumptions, extrapolated results of calculations on PWRs and concluded that the thorium cycle could be slightly favoured by perhaps 0.05 mill/kWh(e). This difference is insufficient basis for development of the concept, but a switch to the thorium cycle may be economically sound should a large-scale thorium recycle industry be developed for other reactors.

Another type of pressure vessel is the aqueous suspension reactor in which dispersed fuel particles are suspended in a heavy water moderator [Went and Hermans 1971]. The suspension is pumped around the primary circuit which consists mainly of the reactor vessel, the heat exchangers and the pumps. Thorium and ^{235}U oxide spheres formed by a sol-gel process make up the fissile and fertile material. The size of the spheres (5 to 10 μm diameter) is such that most of the fission products leave the particle, thus simplifying continuous fuel reprocessing. The system has a large, prompt, negative temperature coefficient and no mechanical control devices are required. Conversion ratios in the range 0.95 to 1.05 are possible, the exact figure depending on the fuel concentration and reprocessing procedures. A 1 MW(th) test reactor of this type, the Kema suspension test reactor (KSTR), was in operation in The Netherlands from 1975 to 1977; it was shown that operation is possible up to twice the power densities required for full size systems and that the main expected problem, that of erosion, is only a problem at the pump impeller [Went 1977].

4.4 Molten Salt Breeder Reactors (Figure 11)

In molten salt reactors (MSBR) the fuel carrier is a mixture of molten LiF and BeF_2 at 500°C in which is dissolved thorium tetrafluoride and a fluoride of the fissile material which may be ^{233}U, ^{235}U or ^{239}Pu [Rosenthal

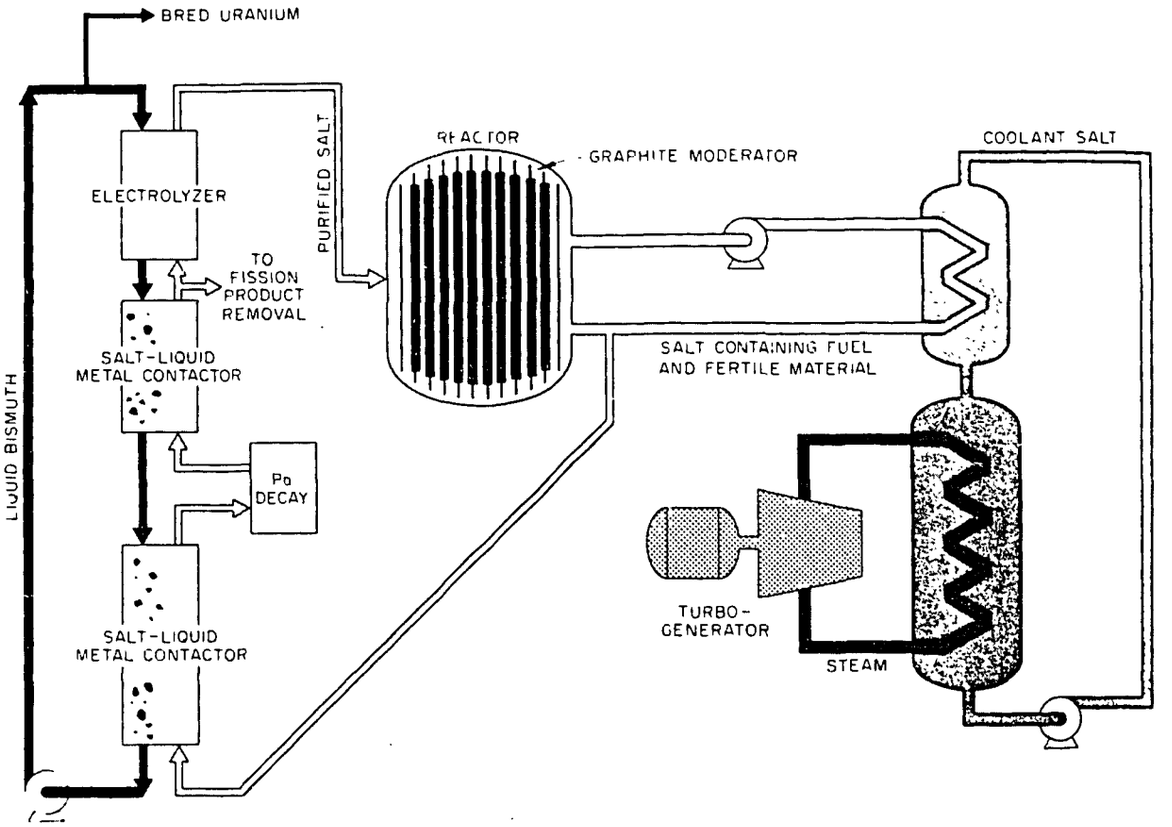

FIGURE 11 MOLTEN SALT BREEDER REACTOR [after Rosenthal et al. 1970]

et al. 1970]. This liquid is pumped through a core formed of unclad graphite which is not wetted by the salt and has low surface porosity to minimise fuel penetration. The graphite channels are spaced to provide a central, well-moderated core region surrounded by an under-moderated blanket region in which the power density is low and neutron capture in thorium predominates. Conversion or breeding ratios in the range 0.85 to 1.07 are possible, depending upon the reactor design and the reprocessing characteristics.

Experiments between 1954 and 1976, including the operation of an 8 MW(th) molten salt reactor experiment (MSRE) at Oak Ridge National Laboratory, have shown this to be a practical type of reactor. High thermal efficiency (44 per cent) and low primary system pressure give MSR converters and breeders potentially favourable economics, fuel utilisation and safety characteristics [Rosenthal et al. 1970].

In a review of molten salt reactors, Haubenreich [1973] commented:

> "Molten salt reactors promise breeding performance and power costs that are attractive, but some basic problems must be favourably resolved and a host of development tasks must be accomplished before these promises can be realised. The areas of greatest uncertainty are the measures that will be necessary for tritium containment and the solution to the cracking of the construction material, Hastelloy-N, by tellurium".

Major engineering development of steam generators and on-line processing plant was also seen to be necessary.

Because of the termination of the MSRE in 1976, not all of these problems have been entirely eliminated, but considerable progress has been made. Engel [1976] reported that the containment of tritium appears to be satisfactory; build-up to equilibrium in the coolant salt takes about four weeks with 1 to 4 per cent penetrating through the loop walls; the rest is removed by the loop off-gas system. This is equivalent to about 3.5 Ci/day (130 GBq/day) of tritium being released by a 1000 MW(e) reactor.

Progress has been made with materials and many Hastelloy alloys, modified by addition of titanium, cerium and niobium, have been tested. The addition of 0.5 to 2 per cent niobium to Hastelloy-N has proved to be the most successful, being highly resistant to intergranular penetration by Te under

irradiation up to 650°C. Its properties deteriorate rapidly with increased temperatures, however, becoming poor at approx. 760°C.

Feasibility of chemically reprocessing the MSBR fuel has been demonstrated, but technological development of a plant capable of coping with between 25 and 100 per cent of the requirements of a single 1000 MW(e) MSBR would take at least six years to accomplish if reasonable funding becomes available [Hightower 1975].

Finally, the resource utilisation of the currently envisaged system is reasonable. A 1000 MW(e) reactor with a breeding ratio of 1.07 (doubling time 19 years at 80 per cent plant factor) would have a specific inventory of 1.5 kg/MW(e) compared with an FBR with a doubling time of 10 years and a specific inventory of 4 kg/MW(e) [McNeese 1976]. Because of its low cost, thorium is discharged as waste from the processing plant, but modifications to the flow sheet are being developed to allow recycling and almost complete utilisation of the thorium if desired [Engel et al. 1975].

To summarise, the continuous reprocessing and adjustment of fuel loading afforded by the liquid fuel concept means that very little excess reactivity is required. This reduces reactivity control requirements and implies good neutron economy. The MSBR combines a positive breeding gain with high specific power, potential for high utilisation of fertile material and a high degree of inherent stability and safety in the Th/^{233}U fuel cycle. Against this must be weighed the very difficult chemical situation with high but not insuperable corrosion problems in the reprocessing areas, the operational difficulties inherent in reprocessing by remote control and the necessary use of new alloys which, while promising, have not been tested under irradiation for long periods. The current situation is that an MSBR could be constructed out of 1 to 2 per cent Nb-modified Hastelloy-N and operated satisfactorily at 650°C [McCoy 1978], but the chemical processing still faces formidable problems, some of which are well on the way to solution but some, perhaps as yet unrecognised, remain to be solved [Grimes 1978].

4.5 Fast Breeder Reactors (Figure 12)

The nuclear and economic performance of liquid metal-cooled, fast breeder reactors (LMFBRs) using oxide, or better still, carbide fuels and sodium coolant appears to be superior with the uranium fuel cycle. This is because the η-value and the smaller critical mass of plutonium (Appendix A1, A2)

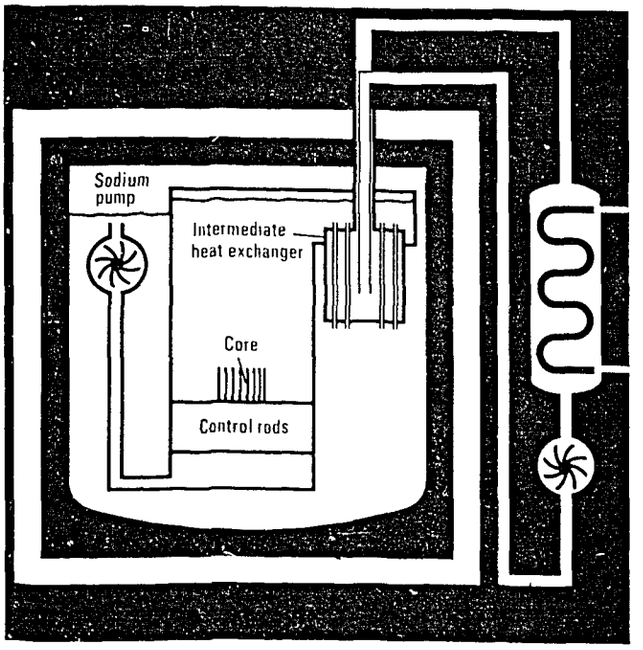

FIGURE 12 THE LIQUID METAL COOLED, FAST BREEDER REACTOR [after UKAEA 1973/74]

Fuel pins are assembled into clusters and cooled by liquid sodium. A blanket of pins containing the fertile material surrounds the core region. An intermediate heat exchanger containing liquid sodium ensures that no radioactive coolant appears outside the primary tank. Steam for the turbines is generated in a secondary heat exchanger outside the concrete shield.

favours the uranium cycle at the higher neutron energies of fast reactors (Table 2). All existing LMFBRs use PuO_2/UO_2 fuels.

Substituting ThO_2 for UO_2 causes a decrease in the breeding ratio from around 1.23 to 1.15 as a result of the lower fast fission cross section of ^{232}Th compared with ^{238}U, and also from the partial replacement of ^{239}Pu by ^{233}U as the latter builds up and fissions during the irradiation cycle. A core fuelled with $^{233}UO_2/ThO_2$ results in an even lower breeding ratio because of the decrease in η noted above.

The LMFBR designed for use with PuO_2/UO_2 fuel could probably be converted to use UO_2/ThO_2 fuel since the irradiation characteristics of these fuels are similar. However, a longer doubling time would result and the effect of this would depend upon the required rate of breeder introduction. Uranium/thorium fuelling would have the very attractive safety feature of a negative sodium void coefficient (Figure 13; see also Section 5).

Experience with the EBR-II experimental breeder [American Physical Society 1978] indicates that uranium metal alloyed with thorium may be an attractive fuel material. Thorium has a face-centred cubic (FCC) structure and is more stable than uranium to irradiation damage and swelling. It undergoes a solid phase transformation at 1365°C compared with the 660°C $\alpha \rightarrow \beta$ transformation temperature of uranium, and its melting point is 1725°C compared with 1132°C for uranium [Kittel et al. 1963; Blumenthal et al. 1969]; all of these parameters make thorium a potentially better fuel material. Because of the limited solubility of uranium and plutonium in thorium, the irradiation behaviour of alloys as fuel may not be as good as thorium metal and more research is required in this area. The higher thermal conductivity of the metallic alloys would produce higher specific powers than the oxide fuel; this, together with the higher atomic density of the metal, would result in an increased breeding ratio of about 1.21 with $^{233}U/Th$ or 1.31 with $^{239}Pu/^{233}U/Th$ [Seghal 1977]. The costs involved in thorium fuel manufacture are almost certain to be higher in the case of oxide, but for metal it is uncertain. One report [American Physical Society 1978] suggests that it may possibly be more expensive while others [Zorzoli 1973a,b] claim that the manufacture of metallic fuel elements by extrusion processes would reduce the overall cost to below that of the UO_2 fuel.

The gas-cooled fast breeder reactor (GCFBR; Figure 14) uses helium gas as coolant [Brooks et al. 1974]. Helium does not interact significantly with the

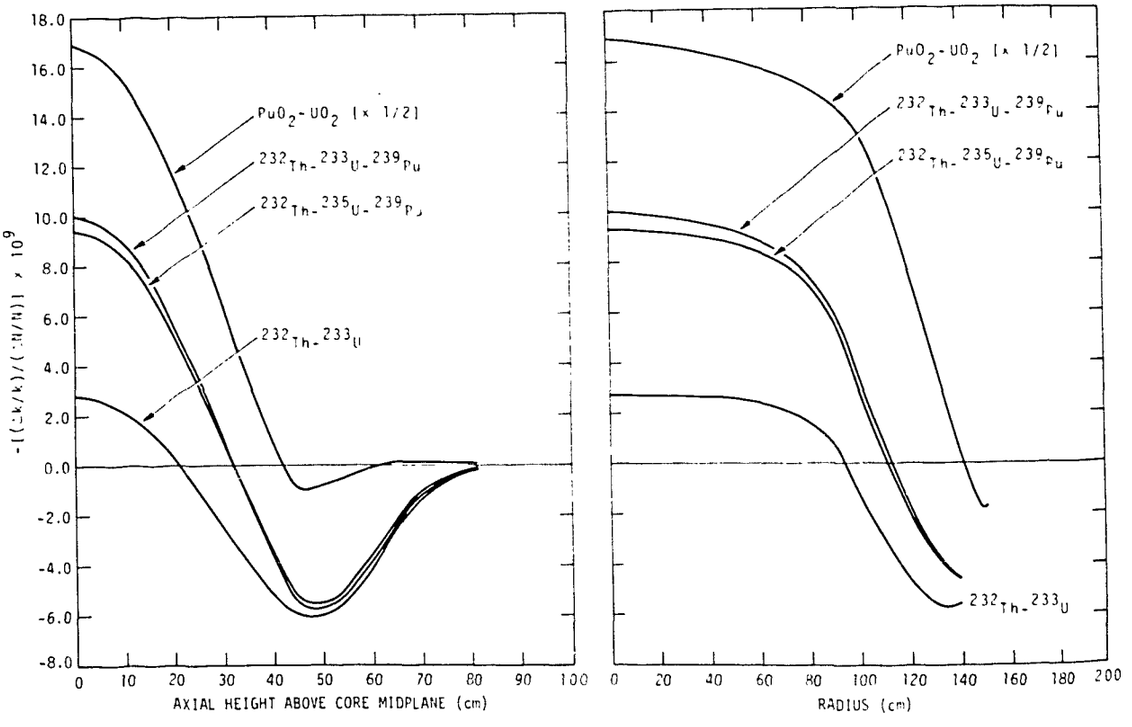

FIGURE 13 AXIAL AND RADIAL DISTRIBUTION OF SODIUM VOIDING WORTH [after Seghal et al. 1975]

fission neutrons and consequently a very hard neutron spectrum is possible. In the case of a reactor with a ThO_2 blanket this leads to a fuel doubling time of perhaps less than ten years and a breeding ratio of about 1.45. This value is slightly less than that obtained with a UO_2 blanket. A small increase in average core enrichment is required, but the net fuel cycle costs will be slightly less in the case of thorium. Such a design could readily revert to a UO_2 blanket if necessary. The overall breeding performance of an all thorium GCFBR is lower than that of a PuO_2/UO_2 core, having a breeding ratio of 1.27 [Cerbone 1974], but it certainly remains a reasonable option.

The higher fissile values of ^{233}U relative to ^{239}Pu create a real incentive to utilise thorium at least in the blanket of FBRs since the recycle of the ^{233}U bred in HTGCRs (Section 4.6) would significantly reduce the net fuel cycle costs. In addition, a symbiotic system (Section 4.8) of FBRs and advanced converters would greatly reduce the demand on fissile requirements.

4.6 High Temperature Gas-cooled Reactors (Figure 14)

Three HTGCRs (DRAGON, Peach Bottom and the Arbeitsgemeinschaft Versuchs-Reaktor (AVR)) using thorium as fertile material have operated for about eight to ten years [Bennett and Lane 1974; Cerbone and Tsoulfanidis 1975]. They are demonstration plants (20 to 116 MW(th)) which have performed well, and the ceramic fuel concept (based on the use of graphite coated fissile and fertile particles) and the ability to contain the reactor helium coolant have been proved (see Tables C3, C4 and C5).

The DRAGON reactor (Winfrith, UK) reached full power in 1966. Operation of this reactor provided much basic data and did much to prove the technical feasibility of the HTGCR. Latterly, DRAGON was used as an irradiation facility for different types of coated particle fuel and the levels of radioactivity appearing in the primary cooling circuit were extremely low. The DRAGON concept was developed for very high core temperature research, but *operations ceased in 1976 because of a lack of funding.*

Peach Bottom, Pennsylvania, USA, went into operation in 1967. The first core was unloaded after only half its normal life as a result of problems with the coated fuel particles. A second core using improved fuel particles operated satisfactorily. The availability of the plant was high and the improved fuel particles kept the primary system activity low. Peach Bottom was decommissioned during 1974 after achieving the full design life of 900

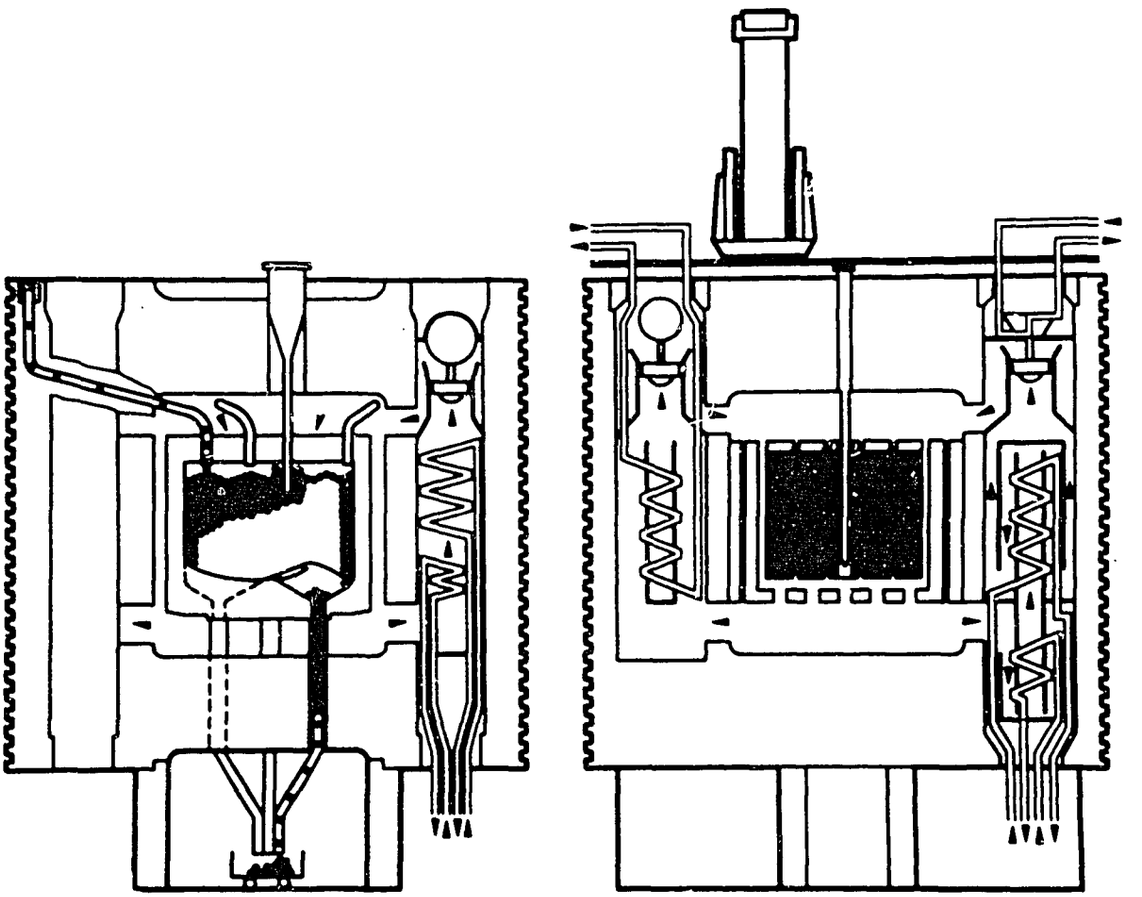

FIGURE 14 MAIN SYSTEMATIC DIFFERENCES OF TYPICAL HTGCR AND GCFBR DESIGNS [after Oehne 1974]

days (73 000 MWd/t)* for the second fuel loading.

The AVR, at Juelich in the Federal Republic of Germany, reached full power in 1967. The plant availability (74 per cent) indicates that only minor difficulties have been experienced. The primary circuit activity has decreased with time, even though the temperature has been gradually increased from 600°C to its design temperature of 850°C. Some fuel pebbles have reached a burn-up of 130 000 MWd/t, which is 30 per cent more than originally specified. To provide data for possible high temperature process heat applications and for gas turbine coupling, an increase in gas outlet temperature to 950°C is envisaged; eventually it is planned to increase the temperature to 1050°C.

A great variety of graphite fuel element designs has been proposed for HTGCRs [Price and Shepherd 1973] but, in general, they fall into two main groups: the pebble and the prismatic block.

The pebble elements lend themselves to standardisation of core design since the only geometric parameter is the radius of the spheres. Such elements have been extensively tested in the AVR and the thorium high temperature reactor (THTR). These spheres have a diameter of 6 cm; the graphite and fuel mixture is surrounded by a 1 cm thick wall, originally machined but now moulded. Design burn-up of these elements was nine per cent FIMA**, but some of the original elements achieved considerably more than twice this burn-up [Lotts et al. 1977; General Atomic Co. 1980].

Prismatic designs (e.g. DRAGON reactor) are based upon more or less complicated assemblies of rods or blocks of graphite with longitudinal channels containing the graphite-fuel matrix and open channels for coolant, gas flow, etc. The fuel for this type of element is fabricated by intrusion bonding of a blended bed of coated fissile and fertile particles contained in a metal mould. Experiments have shown that such designs retain adequate structural integrity after irradiation. Volumetric changes are small and predictable for Triso particles but the contraction of Biso particles is larger than predicted [Kovaks and Sedlak 1980; Kovaks et al. 1980]. See

* Burn-up is generally given in megawatt days per tonne of heavy elements.

** FIMA: When mixed fuel materials are in use, burn-up is sometimes defined as 'fission per initial metal atom'. When one fissile material is present as in, say, enriched uranium, burn-up is often defined as 'fission per initial fissile atom' (FIFA).

Section 7.2 for definitions of Triso and Biso.

Two second generation plants are well advanced. General Atomic's Fort St Vrain 330 MW(e) reactor, which uses hexagonal graphite fuel elements incorporating uranium thorium dicarbide fuel particles, went critical in January 1974. In May 1975, two per cent of full power was achieved, but because of a whole series of non-nuclear faults that have occurred since then, the reactor had only reached 30 per cent of full power by June 1981.

A 300 MW(e) pebble bed core THTR is under construction at Schmehausen, FRG, and expected to be in operation by 1985 [Nuclear Engineering International 1981]. The THTR has 675 000 spherical fuel elements, each of 60 mm diameter, including an outer fuel-free graphite shell of minimum thickness 5 mm. Embedded in a graphite matrix inside the shell are approximately 16 000 coated fuel particles. Initially, each fuel element contains about 1 g ^{235}U and 10 g thorium, both in the form of oxide. A significant difference from the AVR is that shutdown rods will be pushed directly into the pebble bed rather than run in graphite channels. This will require a force of several tonnes and is a potential mechanical disadvantage of the pebble bed system.

Interest in the thorium fuel cycle exists primarily because it can provide improved fuel utilisation over the uranium cycle. Interest in uranium continues, however, because it is possible to use low enriched uranium as the initial fuel (the LE cycle), whereas the thorium cycle requires more highly enriched uranium in the initial fuelling (the HE cycle). If irradiated fuel elments from HTGCRs could be disposed of inexpensively without any processing, the use of the LE cycle would be economically attractive for some time [Kasten et al. 1975]. Unfortunately, this is not possible and reprocessing is essential; recycle improves the fuel utilisation of both cycles but has a larger effect on the thorium cycle. However, comparative studies of the two cycles in HTGCRs give marginally less fuel costs for the thorium cycle [Kasten 1970; Oehne 1974].

Use of the thorium cycle in HTGCRs with ^{235}U as the initial fuel will reduce uranium requirements, but it will not significantly reduce the need for breeder reactors. The rapid increase in U_3O_8 costs in 1975-78 ($US55 to 88 per kg) prompted several studies aimed at increasing the conversion ratio and it was shown that significant improvements are possible [Brogli et al. 1975; Turner et al. 1976; Lane et al. 1976]. Fuel particle coating development should allow considerable reduction in coating thickness with the consequent

decrease in achievable C/Th atomic ratios. Using as reference the General Electric Co. standard ^{235}U/Th fuelled HTGCR design, it was shown that by increasing the thorium content to attain C/Th ratios of 120:140, the conversion ratio could be increased from 0.66 to 0.8 with little or no economic disadvantage, the cost of the extra ^{235}U being countered by the increased fuel and plant performance. At higher thorium contents, spectrum hardening demands excessive ^{235}U and fuel inventory costs rise very rapidly. In this initial core configuration, modest savings in the ^{235}U can be made, together with a 50 per cent increase in total energy output, by decreasing the initial thorium content and increasing the core cycle time to 1.5 years.

The above remarks refer only to the initial ^{235}U core. When ^{233}U is used, particularly in initial cores for high conversion or near-breeding reactors, the high fertile inventory leads to considerable hardening of the neutron spectrum. This has little effect on the η-value of ^{233}U which remains quite constant into the epithermal regions, whereas $\eta^{235}U$ and $\eta^{239}Pu$ fall rapidly in value (see Figure A2). Consequently, with ^{233}U fuel the HTGCR can yield conversion ratios in the range 0.94 to 0.97. If necessary, ratios up to 1.05 can be obtained, but at the cost of a very high initial fuel inventory. Such high conversion ratios depend entirely on the satisfactory performance of as yet untried thorium/carbon ratios in the advanced coated fuel particles. Table 4 compares the initial, annual and total U_3O_8 feed requirements for HTGCRs at various conversion ratios with those for a PWR.

TABLE 4
U_3O_8 FEED REQUIREMENTS (t/MW(e))
[after Brogli et al. 1975]

Reactor Type	HTGCR				PWR
Conversion Ratio	0.66	0.82	0.90	0.97	0.60
Feed:					
Initial	0.40	0.64	0.94	1.4	0.50
Annual	0.105	0.058	0.035	0.01	0.16
40 year total	4.53	2.90	2.30	1.90	6.66

The high coolant temperatures possible in the HTGCR system offer unique advantages: it can provide a direct source of process heat; produce electricity via steam with greater thermodynamic efficiency than any other reactor system; and has the potential to produce power even more efficiently if used with direct-cycle gas turbines.

Because the thermal qualities of ThO_2 are better than those of UO_2 (Table A2), thorium is of outstanding importance and provides the basis for the high temperature development potential of the HTGCR system. Using ThO_2, significantly higher temperature gradients across the fuel rods can be attained without excessive fuel migration [Kasten and Tobias 1975; Smith 1976]. The development status may be summarised as follows [Shepherd 1977]:

(a) Helium coolant temperatures of 750°C have been readily achieved or exceeded and no difficulties experienced in handling the helium at this temperature.

(b) Helium purity can be maintained and there are no significant corrosion problems.

(c) Helium leak tightness does not present a major difficulty.

(d) The graphite core structure has proved remarkably durable.

(e) Extremely low levels of fission product release to the coolant and radioactivity of the coolant. This leads to increased accessibility of primary circuit components.

Some chemical problems occur due to the reaction of H_2O vapour with graphite but, at steam producing temperatures, these are trivial and it is unnecessary to maintain very low water vapour leak rates. However, some difficulties may occur at higher temperatures. The mechanical strength and stability of the coated particle fuel has been demonstrated and irradiation experience [Shepherd 1977] has demonstrated the possibility of allowing further substantial increases in the fuel temperature (see also Section 7).

The high coolant temperature allows the use of smaller direct-cycle gas turbines which are less costly than steam turbines and, because of the higher temperatures, will run at higher thermal efficiencies. Such turbines are designed to run with gas entry temperatures of 800°C and have net efficiencies of 35 per cent, which is higher than the LWR systems. Shepherd [1977] suggested that gas outlet temperatures of 1000°C should be available in the future with very little necessity for further fuel development. Such temperatures would greatly increase the efficiency and, at the same time, gas will emerge from the turbine at temperatures in the range 180 to 200°C. This temperature increase is high enough for a number of industrial processes

which, if used, would further increase the overall use of the fuel, but not necessarily in the cheapest manner. It was shown by Bonnenberg and Schlenker [1974] that if such binary cycles producing electricity and district heating are adjusted for maximum use of resources, about 90 per cent of the total available energy could be used.

Many countries are highly dependent upon the importation of coal, oil and gas for their energy; considerable savings could be made in these import requirements by using nuclear power for process heat, i.e. the direct use of the heat in, say, a chemical process rather than via electricity production. Nuclear process heat systems also have significant environmental advantages:

(i) Possible large scale coal conversion systems with substantially reduced sulphur, NO_2 and particulate emission.

(ii) Reduced environmental impacts from mining.

(iii) Reduced needs for coal transportation.

(iv) Reduced CO_2 emission.

(v) Conservation of fossil fuels and their allocation to essential areas.

A number of diverse studies have been made of possible uses of process heat [Landis 1973; Hosegood and Greenhalgh 1975]. Fields of interest include process steam application [Main and Richards 1976], gasification and liquefaction of coal [Kugeler 1976], zinc and aluminium production [Huwyler et al. 1976], hydrogen production [Beglie 1976; Barnert 1976; Courvoisier et al. 1976] and energy pipeline systems [Daniels et al. 1980]. A number of geographical regions are experiencing problems which make them serious contenders for the use of nuclear process heat. Examples of the samples studied are the Texas Gulf coast, a heavily industrialised conurbation dependent on a natural gas supply which will be soon exhausted; increasing requirements of hydrogen for ammonia and methanol production in the Benelux countries; and newly developing iron and gas production in the Middle East and steelmaking in South America and Russia. In Japan, a large scale national project for nuclear steelmaking was initiated in the early 1970s and, in 1973 a similar project was commenced, on an international scale, by the European Steelmakers' Club (ENSEC) - see for example Spiewak et al. [1976] and Schulten

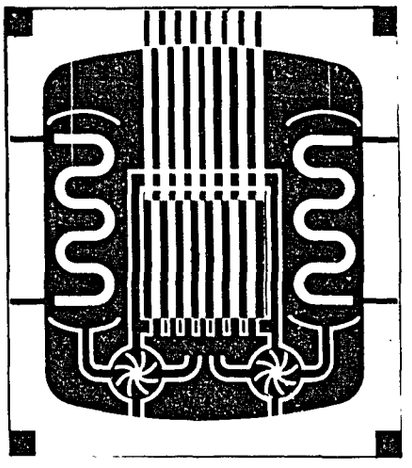

FIGURE 15 ADVANCED GAS COOLED REACTOR [after UKAEA 1973/74]

Clusters of fuel elements joined end to end in stringers are loaded into vertical holes in the graphite moderator. The coolant is carbon dioxide which produces steam in heat exchangers enclosed within the pressure vessel. The Magnox reactors are somewhat similar, except that the fuel is metal rather than oxide and the heat exchangers and carbon dioxide pumps are outside the pressure vessel.

et al. [1976].

Very high temperature reactors (VHTRs) have also been described and their capabilities assessed. Spiewak et al. [1976] considered an He/graphite system capable of operating at a coolant outlet temperature of 950°C using current technology or up to 1200°C with some development. The unique capabilities of such a reactor would be:

(a) the production of hydrogen by decomposition of water or via hybrid fossil-nuclear systems;

(b) the production of heat for direct industrial use, transported or stored for peak hour use; and

(c) high efficiency electrical production using direct cycle gas turbines, combined cycles or boiling potassium topping cycles*.

A 50 MW(th) VHTR reactor project is being developed in Japan [Nomura et al. 1977].

4.7 Gas-cooled Graphite-moderated Reactors (Figure 15)

In the natural uranium fuelled reactors (excluding high temperature reactors) there is no economic advantage in substituting thorium for ^{238}U because of limitations on fuel, specific power (MW/kg) and the relatively high fissile inventory cost associated with the use of thorium (kg/MW).

The AGR is also a large lattice system, analogous to the pressure tube HWR; the comparative performance of thorium and uranium fuel cycles in AGR lies between those for LWRs and pressure tube HWRs. Thorium does not appear to be a commercial proposition in these reactors.

4.8 Symbiotic Systems

The high breeding ratio GCFBR and the high conversion ratio HTGCR can be run as a team, complementing each other in combined fuel operations to form a self-sustaining system. Such a symbiotic system has attractive fuel economies

* Topping cycles are a means of improving thermal efficiency (see Appendix B).

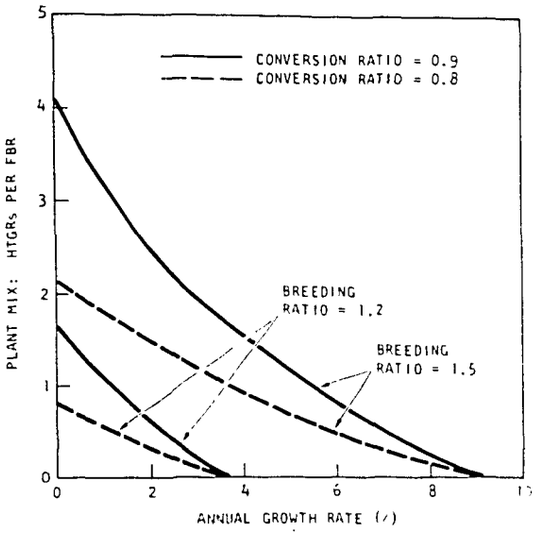

FIGURE 16 GROWTH POTENTIAL OF SELF-SUSTAINING POWER SYSTEMS [after Brogli and Schlueter 1975]

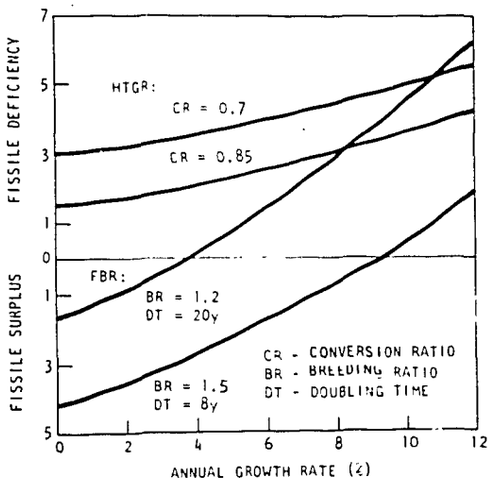

FIGURE 17 FISSILE MATERIAL BALANCE OF EXPANDING NUCLEAR REACTOR ECONOMIES [after Brogli and Schlueter 1975]

and exceptionally good resource conservation [Brogli and Schlueter 1974; Melese-d'Hospital and Simon 1977]. Current fuel technology could produce a GCFBR with a breeding ratio of 1.45 and its relatively simple plant technology means that it has a potentially low capital cost. Breeding ^{233}U in the radial blanket for use as HTGCR fuel is economically worthwhile because the ^{233}U is worth more than ^{239}Pu bred in a similar uranium blanket. The current value of 0.66 for the conversion ratio of uranium fuelled HTGCRs can be increased to 0.84 by changing to ^{233}U fuel, increasing the thorium load and changing to semi-annual refuelling.

Figure 16 shows the ratio of HTGCRs to GCFBRs necessary to be self-sustaining and to support an annual power growth rate, and Figure 17 shows the surplus fuel gained in the GCFBRs and the deficiencies occurring in HTGCRs as a function of the power annual growth rate.

During the initial introduction of such systems, the plutonium stockpile produced by the LWRs and other reactors could be burned up in the initial phase before processed ^{233}U becomes available (Figure 18). The total power costs of such a symbiotic system could be lower than for either of the individual plants and, in fact, lower than for any other reactor or combined reactor system yet envisaged [Brogli and Schlueter 1974].

A more comprehensive review of some of these reactor types and their ability to accommodate thorium fuel has been published by the American Physical Society [1978].

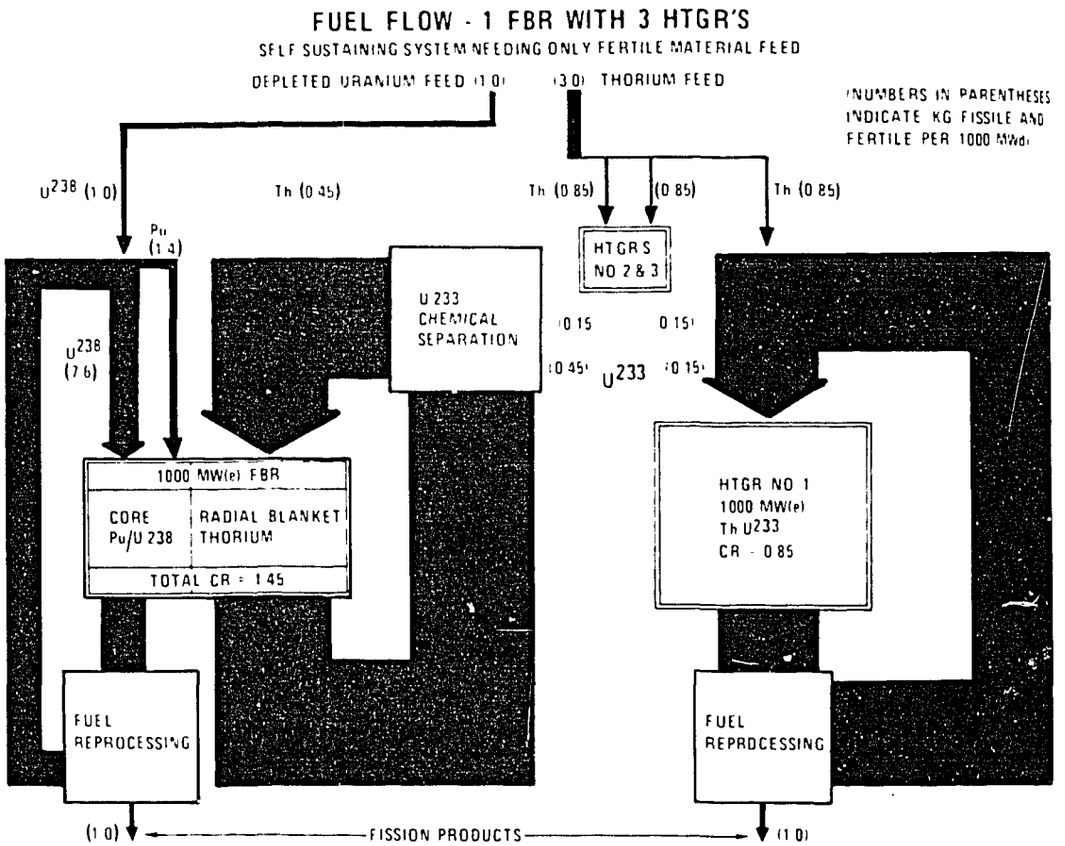

FIGURE 18 SYMBIOSIS OF FAST BREEDER REACTORS AND HTGRs [after Melese-d'Hospital and Simon 1977]

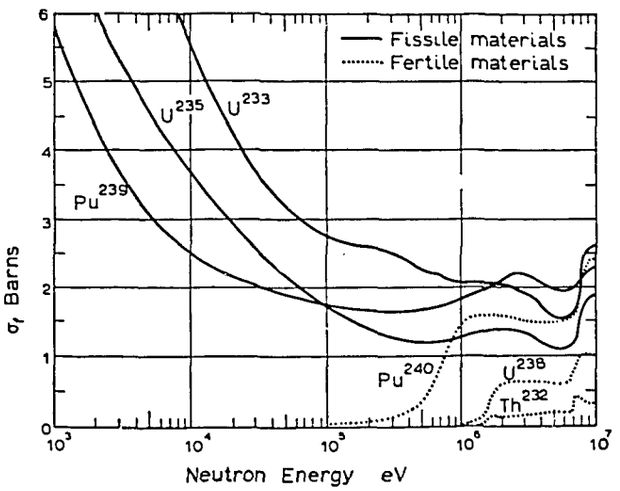

FIGURE 19 FISSION CROSS SECTIONS OF FISSILE AND FERTILE MATERIALS AT HIGH ENERGIES [after Jakeman 1966]

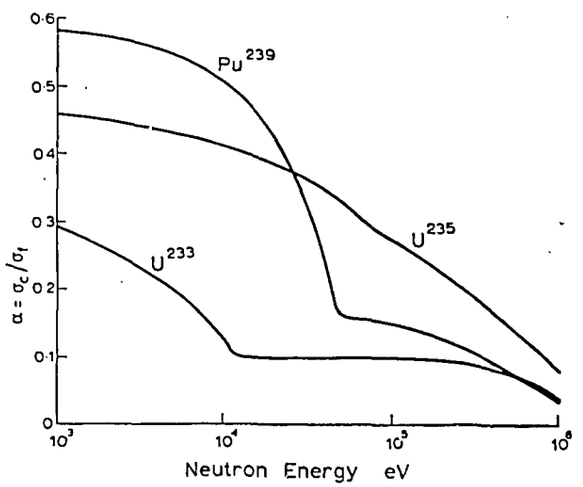

FIGURE 20 RATIO OF CAPTURE CROSS SECTION TO FISSION CROSS SECTION FOR FISSILE MATERIALS AT HIGH ENERGIES [after Jakeman 1966]

5. REACTOR OPERATIONAL HAZARDS

The safety of the public, the operating staff and the plant is a primary consideration in the design, construction and operation of a nuclear power station. Although all reactors should be designed to meet statutory safety criteria, different reactor types take necessarily different paths to achieve this, some of which are more complex and expensive than others. This is true whatever fuel is used. All fuels have their problems, but thorium has certain characteristics which may be developed towards improved safety; these are the fast fission of the fertile material (Figure 19), the α value* of the fissionable material (Figure 20) and the absorption resonances (Appendix A3). Some of the safety aspects of these characteristics are discussed below.

Liquid metal fast breeder reactors are cooled with liquid sodium; any change in the density of this coolant, whether by gas entrainment or temperature rise, has two major effects on the neutrons in the system. A decrease in density increases the leakage of neutrons and constitutes a negative and safe voidage coefficient; at the same time, a reduction in neutron scattering occurs, causing a hardening of the neutron spectrum which results in several changes. Figure 19 shows that an increase in fast fission of the fertile material will occur, but that ^{232}Th is the least affected. This is a positive coefficient. Figures 19 and 20 show that fission in the fissile material will decrease (negative coefficient), however, the α value changes quite dramatically, the loss of fission being more than compensated by the loss in captures with a consequent positive coefficient. With a large fraction of the neutron spectrum being above 10^4 eV, however, the effect on a ^{233}U system could be quite small.

In small LMFBRs, neutron leakage dominates the sodium voidage coefficient and it is thus negative and safe. In large LMFBRs with a somewhat softer spectrum, this is not the case. Only the outer regions of the core exhibit a negative coefficient; the inner regions are positive but, by introducing thorium as the fertile material, the dominance of the positive aspect can be considerably reduced and, with a $^{233}U/^{232}Th$ system, the negative coefficient can dominate (Figure 13).

The absorption resonances of thorium result in negative temperature coefficients due to Doppler broadening (Appendix A3). This effect is larger in ^{233}U than in the other fissile materials and the effect can be used to

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* α is the ratio capture cross section/fission cross section

ensure an overall negative temperature coefficient in fast reactors. The aqueous suspension reactor (Section 4.3.2) and the molten salt breeder reactor (Section 4.4) both have the negative temperature coefficients characteristic of thorium fuel, together with very small excess reactivity requirements and the resultant ease of control. These reactors, however, have some unsolved problems of corrosion or remote handling, both of which have hazardous connotations.

In other types of reactor, the dominant safety concern is the coolant and the helium used as the primary coolant in the thorium fuelled high temperature gas-cooled reactor and the gas-cooled fast breeder reactor which is in a class of its own. Helium has the advantage of being a single-phase fluid, thus change in pressure cannot cause sudden effects in heat transfer or circulation; it is chemically inert and therefore does not interact with fuel or cladding or cause corrosion. It has good heat transfer characteristics and a practically zero neutron capture cross section, except for trace amounts of ^{3}He, which results in ^{3}H, a very soft β-emitter. The specific energy content of the He coolant is far lower than that of any liquid coolant pressurised to operate substantially above the atmospheric boiling point. This allows adequate containment at low cost. The Reynolds numbers and heat transfer coefficients are sufficient to permit efficient heat removal without introducing thermal stress and thermal shock problems in heat exchangers or primary circuit components [Buttemer and Torri 1977].

Helium has the disadvantage that it must be pressurised at 5 to 6 MPa in the HTGCR and 9 MPa in the GCFBR to provide the necessary heat removal. Natural convection cannot be relied upon for decay heat removal when the reactor is depressurised and certain contingencies have to be allowed for. The whole of the reactor coolant system is enclosed within a pre-stressed concrete reactor vessel (PCRV), the core within a central cavity, and the steam generators and He circulators within sidewall cavities. Internal ducting connects these components and there are no large external pipes. Potential openings in the coolant vessel are limited to small lines and to closures on penetrations used for the installation and maintenance of large items. The small penetrations are designed for He depressurisation times of more than one hour if they should fail; the larger closures have flow restrictors which increase the depressurisation time to tens of minutes in the case of a closure failure.

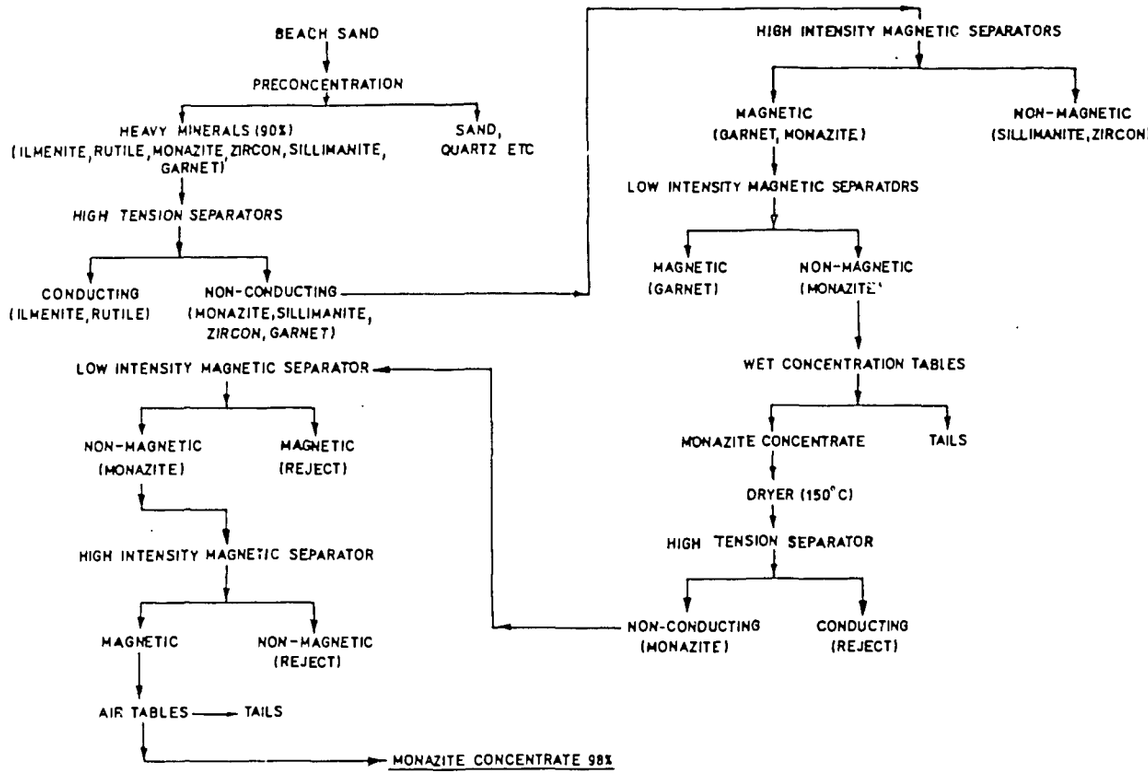

FIGURE 21 FLOWSHEET FOR CONCENTRATION OF MONAZITE FROM BEACH SAND [after Garg et al. 1977]

The extent of the depressurisation of the gas is limited to the ambient pressure within a secondary containment building (from 0.15 to 0.2 MPa). Residual heat removal (RHR) is achieved by two separate systems; first the operational RHR, in which a direct flow of steam from each steam generator through the He circulator turbines provides three independent heat transfer loops; and second, the core auxiliary cooling system (CACS), which is electrically driven by separate high frequency alternators and is normally used for long-term shut-down cooling [Melese-d'Hospital and Simon 1977; Larrimore and Moffette 1974].

The graphite structure of the HTGCR is advantageous in that the whole core and fuel system is built of a refractory substance which increases in strength as the temperature rises, reaching a maximum at about 2500°C; this is well above the operating range and usable strength is retained well above this temperature [Larrimore and Moffette 1974]. A high ratio of heat transfer surface to core volume, combined with good thermal conductivities in the core materials, minimises differentials between this maximum helium temperature and the peak fuel temperature [Shepherd 1977]. The intimate mixture of fuel and graphite within the fuel elements ensures a prompt moderator temperature coefficient and good control characteristics, leading to flexibility in operation with good ability to follow load fluctuations [Brown 1969]. The side and bottom graphite reflectors, normally at coolant inlet temperature, constitute a large, built-in heat sink in the event of temporary loss of main heat rejection system [Fortesque et al. 1960].

The fuel in the HTGCR and GCFBR has little similarity, except that the system for pressure equalisation or venting the GCFBR fuel is similar to that of some HTGCR designs. This venting procedure ensures that the fuel rods do not fail by creep collapse or fission gas pressure build-up. Charcoal traps within the element prevent the release of most fission products; only the noble gases are vented directly into an He purification system, where they are removed by low temperature delay beds. Experimental data show that volatile fission products are trapped in the blanket region of the fuel elements and are not a problem [Larrimore and Moffette 1974].

The coated particle fuel of the HTGCR has extremely good fission product retention and very low activities are found in the primary circuit [Shepherd 1977]. The ceramic fuel and core operate well below their temperature limits and, furthermore, the fuel coating does not fail rapidly, even when subjected to very excessive temperatures. There are substantial margins between

calculated fission product release and the US Energy Research and Development Administration (ERDA) guidelines for normal and accident conditions [de Nordwall and Bell 1974].

With regard to the substitution of thorium-based fuels into normally uranium fuelled reactors, Kasten and Homan [1977] report that thorium oxide plus uranium oxide pellets perform well up to 80 000 MWd/t in water-cooled reactors and satisfactory irradiation experience with zirconium clad ThO_2 and ThO_2/UO_2 pellets up to 10 000 MWd/t for LWBRs and 30 000 MWd/t for LWBR pre-breeders has been achieved. Limited experience indicates that PuO_2/ThO_2 should be just as good. The higher melting point and thermal conductivity of the thorium mixture should be beneficial in these cases.

These comments on the use of thorium as a fuel and on the reactors to which it is most suited clearly indicate a number of good inherent safety features which should make it less complicated and less expensive to achieve the required safety standards.

6. PRODUCTION OF THORIA AND THORIUM

Thorium occurs mainly as monazite, a variable mixture of oxides and phosphates (Table D1). Thorianite, an oxide mineral containing uranium, thorium and thorite, a silicate, are the other important sources. Most of the world's thoria is extracted from monazite; some production figures are given in Table D3.

Monazite is found on beaches along with other heavy minerals and various mining techniques are used for its extraction; rich seams are bulldozed but sands containing low percentages of heavy minerals are suction-dredged, a method favoured on the east coast of Australia. Concentration of sands containing low percentages of minerals is achieved by centrifugal or gravitational methods in a wet pond, resulting in 90 per cent heavy mineral content.

After drying, the various minerals are removed by electrostatic or electromagnetic methods from the non-conducting fraction left after the separation of rutile and ilmenite by high tension separators. The moderately magnetic monazite is separated by a series of high intensity magnetic separators and air and wet tables (Figure 21). The material then consists of

about 98 per cent monazite [Garg et al. 1977].

The chemical processing necessary to extract the thorium from the ore depends upon the starting material. The two methods most in use are the acid process, which is practised in the USA and USSR, and the alkali process used in India and Brazil. Since the latter countries produce the most thoria (Table D3), their method will be described.

Concentrated monazite is digested with caustic soda producing a hydroxide cake which is the starting point of the process shown in Figure 22 [Dar et al. 1971].

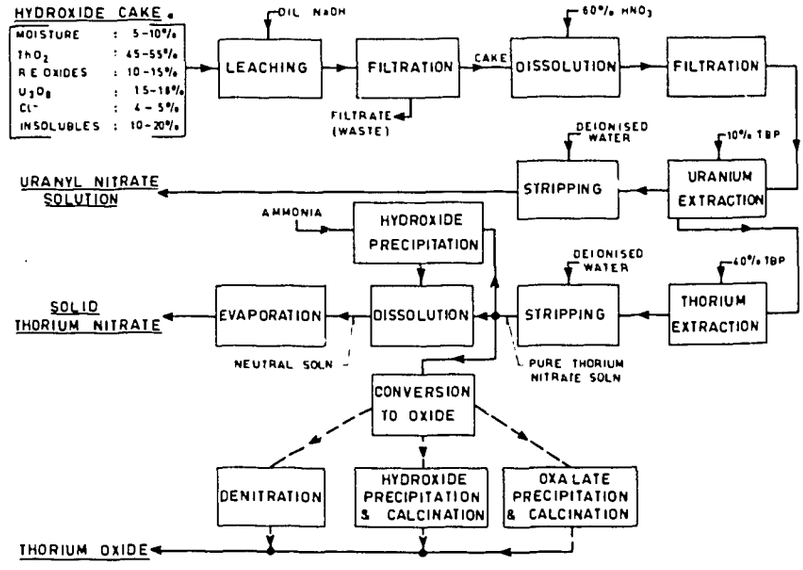

FIGURE 22 PRODUCTION OF NUCLEAR GRADE THORIUM NITRATE/THORIUM OXIDE [after Dar et al. 1971]

The high grade thoria required for nuclear purposes is achieved on an industrial scale by solvent extraction, the most widely used solvent being TBP (tributyl phosphate) diluted with commercial hydrocarbons such as kerosene. This extracts thorium nitrate from nitrate solutions and the pure nitrate obtained is converted to oxide.

Thorium metal is generally produced by one of the following methods [Garg et al. 1977]:

(a) Reduction of ThF_4 or ThO_2 by calcium.

(b) Fused salt electrolysis of $ThCl_4$ or ThF_4.

(c) Thermal decomposition of halides.

Methods (b) and (c) are only used on a laboratory scale at present.

7. FUEL FABRICATION

A detailed description of the methods of thorium fuel element fabrication and a description of the physical characteristics of thorium are given by Weissert and Schileo [1968]. Metallic thorium can be used pure or alloyed, sheathed or bare. As an oxide it may be compressed into pellets and canned in a conventional pin, or formed into microspheres coated with carbon and/or silicon carbide and contained in graphite spheres or prismatic graphite elements. Thorium carbide is only used in graphite cans.

7.1 Metallic Thorium or Thorium Alloys

Thorium metal is generally produced as a pyrophoric sponge or powder. It is consolidated by melting or by powder metallurgy. Th-U, Th-U-Pu and Th-U-Zr alloys can be made by consumable-arc melting of compound electrodes or by cold compressing and sintering alloy powder. The alloy slugs can be cast into shapes, extruded, co-extruded with cladding material, rolled and swaged to form fuel elements [Garg et al. 1977; Blumenthal et al. 1969; Weissert and Schileo 1968].

7.2 Oxide and Carbide Fuels

The ceramic and metallurgical processes used for uranium can be applied to thorium-based fuel. Pelletising, swaging and ceramic extrusion are well established processes and a method which promises to be more economical is the sol-gel process [Garg et al. 1977; Notz et al. 1978]. In this process, to produce oxide fuel, the appropriate mixture of uranium nitrate and thorium nitrate in solution is converted to a nitrate hydrosol in a paraffin, which is

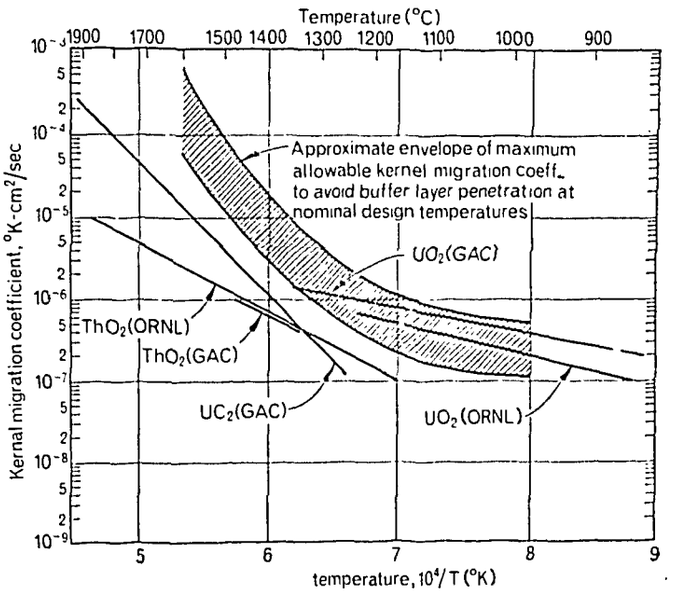

FIGURE 23 KERNEL MIGRATION COEFFICIENTS FOR UC_2, UO_2 AND ThO_2 [after Gulden et al. 1974]

converted to the oxide hydrosol. Spherical particles of gel are formed when water is extracted from hydrosol drops by a counter-current flow of alcohol. The gelled spheres, usually in the range 100 to 1000 μm diameter, are dried, calcined and formed into the desired bulk shape by vibratory compaction. Mixed ThO_2/UO_2 microspheres for HTGCR fuel are also prepared in this way. A similar technique is used to make (Th, U)C_2 microspheres.

The fissile/fertile microspheres are then coated with layers of pyrolytic carbon or carbon and silicon carbide. The first of these, called the 'Biso particles' has an inner layer of porous carbon which traps fission products and an outer layer of dense carbon strong enough to withstand the internal pressure generated by the fission products. Biso particles usually contain only fertile material. The second type of coating consists of an internal layer of porous carbon and an outer layer consisting of a sandwich of silicon carbide between two layers of dense carbon. Fuel coated in this way forms the Triso particles [Shepherd 1977]. The Triso coating is more resistant to the diffusion of some important fission products (Cs, Sr, Ag).

For use as fuel, the microspheres are bonded with graphite-loaded thermoplastic materials such as coal tar or petroleum pitch. These particles have shown an ability to withstand more rigorous temperature and radiation conditions than will be expected in a reactor [Gulden et al. 1974]. The use of thorium reduces the so-called 'amoeba effect'*, that is the migration of carbon in the presence of a temperature gradient. Figure 23 shows the distinct superiority of ThO_2 over UO_2 and mixtures of up to 10 per cent UO_2 in ThO_2 appear to be just as good. Some of the properties of these coated particles are given by Lauf and Braski [1981].

* The amoeba effect or kernel migration is the movement of a fuel kernel towards the hot side of a coated particle under the influence of a thermal gradient. Fuel coating failure can be caused by this phenomenon if a migrating kernel penetrates a structural layer to the point that the internal stresses cannot be supported [Smith 1976; Stansfield 1974; Stansfield et al. 1975].

8. FABRICATION AND REFABRICATION FACILITIES

Few countries appear to have acquired any expertise in this type of work. Canada is the only country embarking on a long-term (about 20 years) extensive thorium program.

8.1 Fabrication

The small-scale fabrication of thorium fuel has been carried out in India [Garg et al. 1977], the Federal Republic of Germany - FRG [Holzer and Knodler 1977], the USA [Burch and Lotts 1977], the UK [Graham et al. 1974] and France [Moreau et al. 1977]. Pilot plant facilities have been in operation in the USA and the FRG [Lotts et al. 1977] and both countries are actively developing advanced pilot or full-scale plants. A demonstration plant capable of processing 2800 fuel elements per annum has been designed in the USA. So far, feasibility of the basic processes has been established and the design of full-scale equipment initiated [Lotts et al. 1977]. In FRG, the consortium Nuken/Hobeg, which made the fuel for the AVR, has developed a system which can readily be expanded to a commercial scale [Balthesen et al. 1974]. France also is developing technology for thorium fuel fabrication.

8.2 Refabrication

This step in the cycle begins with the receipt of the nitrate solutions containing reprocessed ^{233}U and culminates with the assembly of this material into fuel elements (Figure 24). The major difference between fabrication and refabrication is that the latter is done remotely. Uranium-233 can be difficult to handle because of the build-up with time of ^{232}U (Figure 4), some of whose daughters emit very high energy gamma-rays (see Section 6.2). The amount of shielding required depends upon the ^{232}U content of the fuel which, in turn, depends upon the origin of the thorium and whether thorium recycle is used. Concrete up to 1 m thick may be necessary [Arnold 1964].

A part of the thorium utilisation program sponsored by ERDA [Sease 1978] was a design study of an integrated, commercial scale recycle plant able to reprocess 50 000 blocks*/year and to refabricate 20 000 blocks/year. A similar study on refabrication work was conducted in West Germany.

* A term used to denote a handling unit; this could be a fuel element in the case of prismatic elements or a graphite can plus spherical fuel elements, etc.

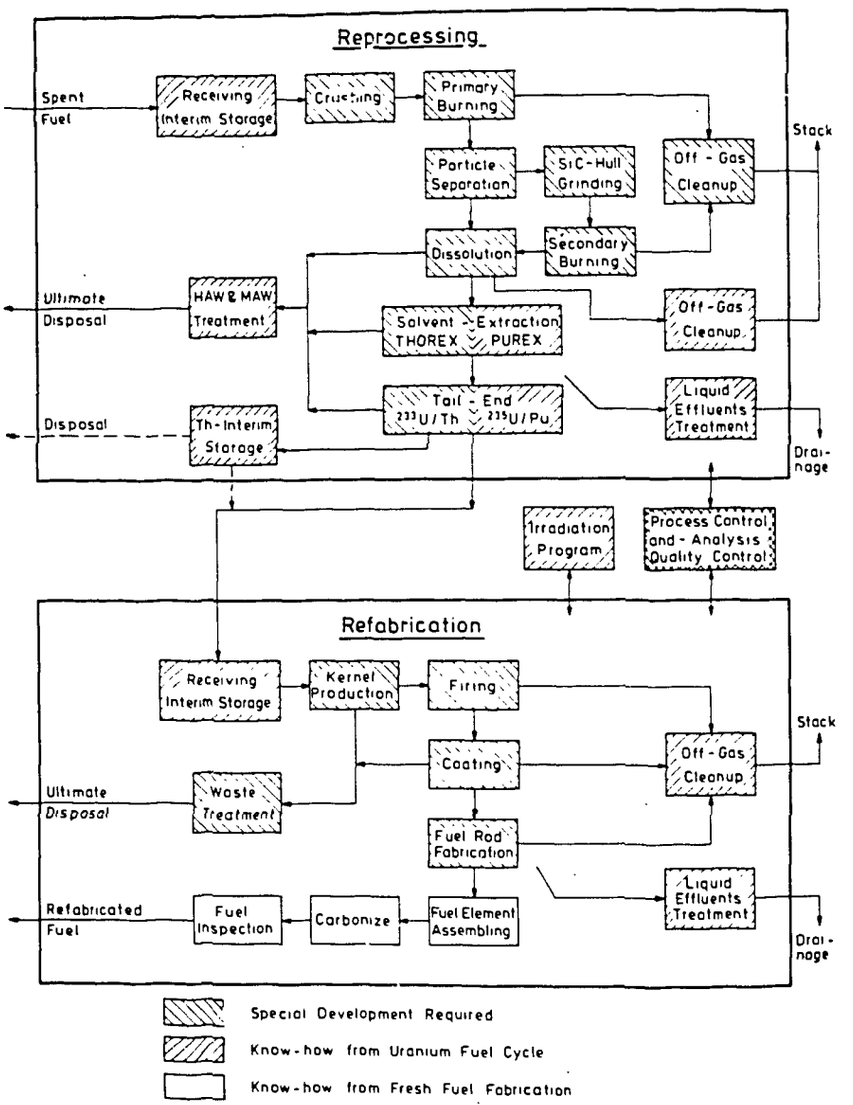

FIGURE 24 SIMPLIFIED FLOWSHEET FOR REPROCESSING, REFABRICATION AND WASTE TREATMENT OF FUEL ELEMENTS FROM HTR [after Merz 1977]

The principal refabrication work is being carried out at ORNL in the USA [Lotts et al. 1977] and KFK in the FRG [Burch and Lotts 1977]. Some pilot plant work has been carried out in Italy, but this is being phased out [Coa et al. 1977]. The US program includes the design and operation of laboratory scale equipment in all the major process steps and of some engineering scale equipment.

In the FRG program [Merz 1977], the major emphasis has been on process and equipment for kernel preparation and microsphere coating. A small coated particle refabrication plant, SATURN, was planned to be on-stream in 1981 and will have a throughput of about 1 kg of ^{233}U per day [Sease et al. 1975; Naefe 1979; Naefe and Zimmer 1979].

Merz [1977] discussed the timing and costing of the introduction of a thorium reprocessing and refabrication plant with the flowsheet shown in Figure 24. The time required is difficult to assess accurately because of the different stages and designs adopted by the various countries, but it is obvious that well over 20 years will be needed before a 5 GW(e) equivalent capacity plant can be commissioned. This is not a long time in relation to the building of reactors and the time required to establish an economic quantity of spent fuel as feed.

The costs involved are substantial; 1977 estimates were about $US250 million for research and development, $US600 million for the plant and up to $US30 million for experimental work on recycle elements. Merz considered that a cost benefit evaluation presented a very favourable picture in comparison with alternative future sources of energy production.

9. FUEL REPROCESSING

A description of thorium fuel reprocessing is given by Weissert and Schileo [1968]; it can be conveniently divided into several phases:

(a) The head end on which the fuel is prepared for later chemical separation processes.

(b) Solvent extraction separating fission products and other waste from the potentially useful material.

(c) The manufacture of new fuel elements.

9.1 The Head End Phase

In most cases, this phase will be similar to that in present-day uranium-fuelled systems in which the canning is removed from the fuel either mechanically or by preferential solution; the fissile, fertile or fission product mixture is then dissolved to produce an aqueous feed material for the separation phase.

Storage of kernels before reprocessing may require rigid control of atmospheric humidity to prevent fission product release but this will not pose difficult problems [Reitsamer and Falta 1980].

High temperature gas-cooled reactor fuels require a different head end procedure because in this case the canning is graphite blocks or spheres. The large quantities of graphite will be burned off [Merz 1977] releasing the volatile isotopes ^{3}H, ^{14}C, ^{35}Kr and ^{139}I [Pence 1976; Tischen et al. 1978]. The quantities involved are given in Table 5. Of these, ^{3}H presents no problem, the technique for removal being currently available. Iodine-139 absorbers, while effective, are expensive and need further development because of the large quantities involved. Krypton-85 removal is well in hand, the KALC* method being most promising. The ^{14}C problem, however, is not yet fully researched. Practically all of the ^{14}C is in the form of CO_2 (easily removed as carbonate from gas streams as is the case in the paint industry) and is of low activity since only 3×10^{-7} of the total carbon is the ^{14}C isotope [Notz et al. 1980]. The problem is the large bulk of carbonate for disposal; consequently there is considerable interest in isotope enrichment in this area. To reduce the bulk of the waste, the Japanese, using a similar reprocessing system, propose removing the fuel packs from the graphite sleeves before burning the fuel. The reduction factors necessary for the volatile isotopes appear to be achievable, but the continuous operation of the large rotary kilns necessary for voloxidation** is seen as a difficult operational

* KALC stands for krypton absorption by liquid carbon dioxide. In this process the solubility of Kr in CO_2 provides a basis for the separation of Kr from the light gases (O, N and CO) by fractionating, the Kr being the last to be given off [Burch and Lotts 1977].

** Voloxidation is a process for the isolation of tritium. The fuel is subjected to an oxidising atmosphere at 450°C to 650°C for 2 to 4 hours, releasing 90 to 99 per cent of the contained tritium as gaseous T_2O. These conditions also help to treat carbide fuels by converting them to CO_2.

TABLE 5

ESTIMATED MAJOR VOLATILE FISSION AND ACTIVATION PRODUCTS

IN FUEL ELEMENTS (180 DAYS AFTER REACTOR DISCHARGE)

[after Pence 1976]

Isotope	g/GW(e)y		GBq/GW(e)y	
	HTGCR	LWR	HTGCR	LWR
^{3}H	1.4	2.4	492 100	847 300
^{14}C	36	4	6 990	592
^{85}Kr	1 820	950	26 300 000	13 727 000
^{129}I	11 200	7 770	74	48

TABLE 6

SEMI-VOLATILE CHEMICAL ESTIMATED TO BE PRESENT IN HTGCR

REPROCESSING OFF-GAS (180 DAYS AFTER DISCHARGE)

[after Pence 1976]

Element	g/Fe	Est. Quantity Volatilised	Est. Quantity to be Collected (per GW(e)y)	
		(g/Fe)	g	Ci*
Ca	113	1.3	1350	92 000
Ra	48.4	0.26	280	13 900
Sb	0.20	0.001	1.1	520
Tc	28.8	0.15	160	2.7

* 1 Ci = 37 GBq

problem [Yabro et al. 1977; Kaiser et al. 1977].

During the burning operation, temperatures may reach 900°C; this will volatilise some fission products which, in other types of fuel, would not normally be found in the vapour phase during the reprocessing; most of these will condense before reaching the filters. Some of these semi-volatile isotopes will, however, find their way to the off-gas system; those of concern are given in Table 6.

Because a significant amount of these semi-volatile isotopes will probably condense onto micrometre or sub-micrometre size carbon particles, it may be necessary to use some type of pre-filtration, such as electrostatic precipitation, to prevent excessive loading of the high efficiency particulate air (HEPA) filters. The main problem appears to be the ability of Ru and Tc to form volatile carbonyl compounds which can penetrate HEPA filters. Engineering scale hot cell experience is required in this area.

Another previously unrecognised problem is that of sulphur which is present in the pitch used to bond the fuel particles. Most of this is volatilised during the curing of the 'green' mixture, but the remainder is present at about 85 μg/g and is sufficient to produce about 43 000 GBq of ^{33}P and 28 000 GBq of ^{35}S per GW(e)y, 180 days after fuel element discharge. No problems other than corrosion are foreseen for this source. A review of some of the problems in reprocessing HTR thorium fuel is given by Zimmer et al. [1979].

A great deal of research and development is required before a commercial thorium fuel reprocessing plant could be built, particularly in the head end areas of carbon burning, coated fuel crushing and off-gas treatment. Progress is being made in the hot cell handling of reprocessed ^{233}U and a plant is being constructed in Germany [Naefe and Mueller 1978; Zimmer et al. 1979].

9.2 The Solvent Extraction Phase

The Purex and Thorex processes used for the extraction of the fissile material from the burned up fuel elements have been described in detail elsewhere [e.g. Benedict and Pigford 1957; Roeh and Wheeler 1975; Srinivasan et al. 1973].

In the case of HTGCR fuels, primary burning, described above, removes fuel element graphite and the outer carbon coating of Triso coated particles (see Section 7.2), and the entire carbon coating from the Biso coated particles. The remaining fissile kernels (ex Triso), with their inner carbon coating and outer silicon carbide coating, have a density of about 3 g/cm^3, while the thoria kernels (ex Biso) have a density of about 10 g/cm^3 which allows gravity separation [Burch and Lotts 1977].

If it is necessary to process the uranium kernels, they must be further crushed to break the silicon carbide layer and then submitted to a secondary burning to remove the carbon [Kaiser et al. 1977].

At this stage, the raw material for the fuel is thoria and urania which may have come from the burning off processes in the case of HTGCR, or from other decanning processes; these are dissolved to produce a fertile derived liquor which goes through the Thorex process and a fissile derived liquor as feed for the Purex process.

The Thorex feed liquor is centrifuged to remove insolubles such as the crushed silicon carbide hulk and then undergoes liquid-liquid extraction by passing through a series of pulsed columns in which the thorium, ^{233}U, and fission products are separated from each other by differential solubilities in the second liquid (TBP), dissolved in various concentrations in a carrier (n-dodecane). No major problems are expected, but the equipment for solvent extraction must be designed for remote maintenance and this requires considerable development [Burch and Lotts 1977].

The Purex feed liquor containing uranium and fission products is similar in principle to Thorex, but differs considerably in detail, having different concentrations of TBP in a different carrier (say kerosine) and different decontamination or purification procedures, etc. The major problem is to determine whether the much higher burn-up (70 atoms per cent instead of, say, 3 atoms per cent in the case of LWRs) will cause difficulties. Hot engineering tests are necessary to determine this.

9.3 Manufacture of New Fissile Material

This is discussed in Sections 8 and 9. The main problem with the manufacture of fuel with reprocessed raw materials may be the necessity to use remote handling techniques.

10. FUEL CYCLE COSTS AND ADVANTAGES

From the discussion in Section 4 there is little doubt that a suitable reactor or reactor combinations using the thorium fuel cycle could result in improved economics and utilisation of material resources.

The HTGCR is an extraordinarily flexible system which allows an operator to change from one fuel to another almost at will, without sacrificing optimisation. This allows a choice of fuel to suit the existing fuel market conditions. The point is illustrated in a joint study by Euratom and the General Atomic Co. [Gutmann and Brogli 1974].

In this study, the basic reactor used was the then commercially available 1160 MW(e) General Atomic reactor with its fuel element block unchanged, but the fuel and fuel cycle varied. Comparisons were made between a reference four year annual reload cycle, semi-annual and on-line cycles using ^{235}U/Th and 2.5 year Pu/Th cycles. The overall variation in the total fuel costs was 16 per cent with the Pu/Th proving to be the cheapest.

Another calculation by the same authors involved fuelling the reactor with a well researched, low enriched uranium fuel (a DRAGON coated particle fuel used for a standard reference) running on a three-year fuel cycle for ten years, then switching to the reference thorium cycle used in the previous calculations. It was shown that the switch-over could be achieved during normal operation and there would be a cost benefit of about 16 per cent (Figure 25).

A French study [Audinet et al. 1974] concluded that if HTRs were built in Europe, the thorium cycle would show a considerable advantage over the low enriched uranium cycle. Exact costs were difficult to forecast because of the versatility of fuel cycle parameters in HTGCRs compared with LWRs, e.g. considerable variability being possible in the carbon to thorium ratio, power density, fuel residence time and refuelling frequency. Another factor favouring the introduction of the thorium cycle is the fact that certain of the French uranium fuel reprocessing plants could be easily adapted to thorium reprocessing, thus eliminating the need for initial excess fuel storage before the building of a reprocessing plant became economic.

With the rapidly increasing cost of U_3O_8 and coal, together with the depletion of oil and natural gas supplies, the economic benefit due to the use

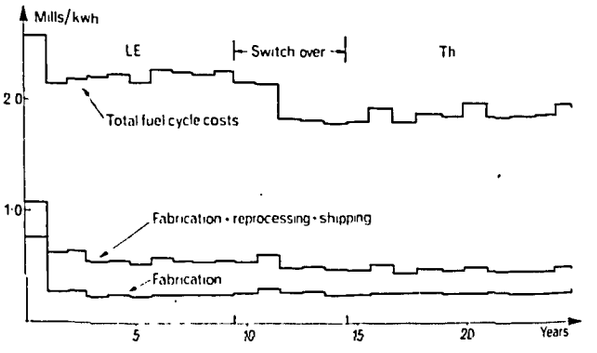

FIGURE 25 BREAKDOWN OF FUEL CYCLE COSTS IN THE LOW ENRICHED URANIUM CYCLE, DURING SWITCHOVER, AND IN THE THORIUM CYCLE [after Gutman and Brogli 1974]

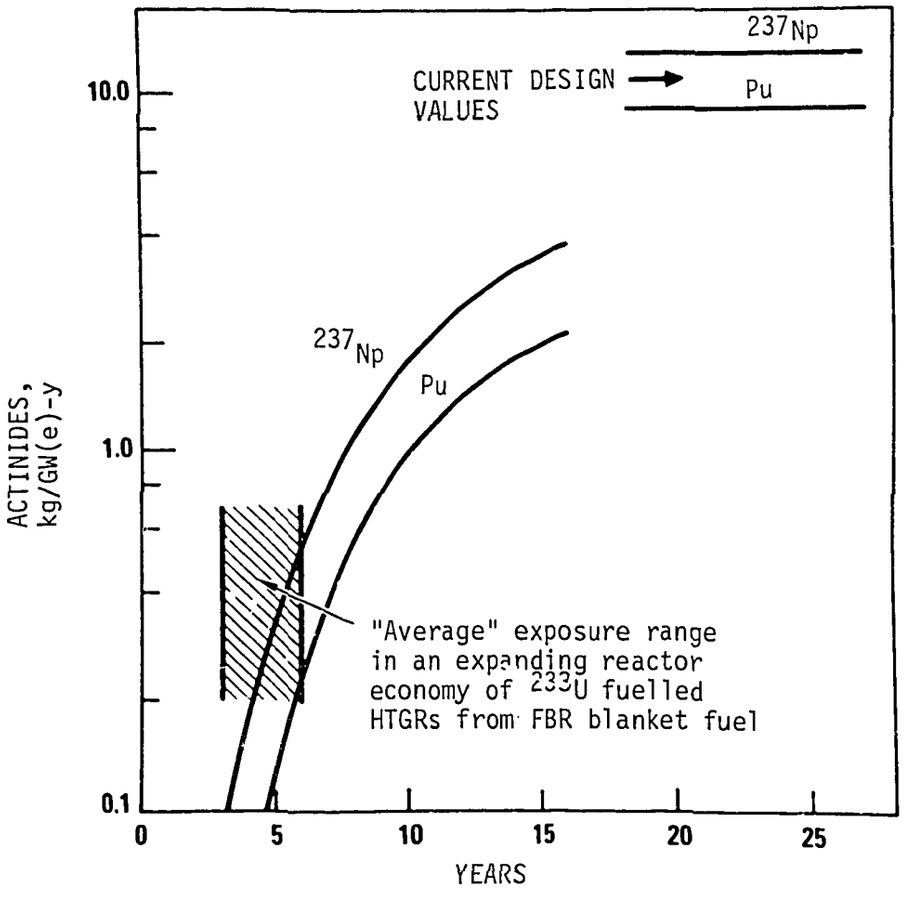

FIGURE 26 ACTINIDE BUILD-UP IN A ^{233}U FUELLED REACTOR (^{233}U FROM FBR BLANKET) [after Brogli et al. 1975]

of thorium as a fuel could improve rapidly since it allows more efficient use of the fissile materials. However, minimum energy costs may no longer be the prime target; rather it may be a balance between efficient use of material and reasonable cost.

Kasten and Homan [1977] examined the fuel cost of various combinations of LWR, HWR and HTGCR types with regard to the relative energy extraction from a given ore resource at specified power growth rates. The prices of uranium ore and separative work are included as extra parameters. Their conclusion was that the thorium fuel cycle provides better U_3O_8 utilisation, improved potential for long-term economics and additional flexibility with regard to fuel cycle alternatives. It also provides additional power generation capability in the case of delayed introduction of commercial FBRs. Similar studies of the self-sufficient equilibrium thorium cycle (Section 4.3) support these findings and the symbiotic system of GCFBR and HTGCR (Section 4.8) goes even further along the path toward low energy costs and resource conservation, together with a reasonable growth rate.

11. WASTE FROM THE THORIUM CYCLE

There are major differences between wastes from the thorium cycle and those from the uranium cycle. It is convenient to study these by looking at the different types of waste produced and comparing differences.

11.1 High Level Liquid Waste

The aqueous solution from the solvent extraction systems form the greatest part of this waste, whether it is from the thorium or uranium cycle. The total quantity of the fission products is practically the same, although individual elements may differ by up to about 30 per cent. In this respect, therefore, there is little to choose between the two cycles. Heavy metal (including transuranic) wastes however present a different picture (Figure 26).

In calculating Table 7, it was assumed that 0.75 per cent of the heavy metals Th and U in the spent HTGCR fuel and all of the remaining heavy metals, including Pu, would end up as high level liquid waste. For LWR fuel, 0.75 per cent of the U and Pu was assumed to go to the high level waste together with all remaining heavy metals. Because of the greater inventory of the heavy

TABLE 7

COMPOSITION OF HIGH-LEVEL LIQUID WASTE [a]
[after PENCE 1976]

	kg/GW(e)y [b]			
	HTGCR		LWR	
Fission Products				
Zr	141.8		122.0	
Cs	120.1		90.3	
Nd	116.8		131.0	
Mo	109.0		114.7	
Ce	85.7		95.0	
Ru	51.4		75.0	
Ba	44.8		46.7	
Pr	44.3		40.0	
Sr	43.8		29.8	
La	41.0		42.3	
Others	144.3		179.2	
		943		966
Heavy Metals				
Th	63.8		-	
Pa	0.2		-	
U	3.9		238.8	
Np	16.3		25.4	
Pu	10.5		2.3	
Am	0.2		5.3	
Cm	0.1		1.2	
		95		273
Chemicals				
Gd	178.0		300.0	
Al	123.1		-	
K	89.2		-	
PO_4	83.2		66.7	
Fe	50.0		-	
F	43.4		-	
Na	0.1		3.3	
		567		370
Corrosion Products and Burnable Poison				
Fe	80.0		66.6	
Cr	8.0		6.7	
Ni	3.2		2.7	
B	15.8		(3.0)[c]	
		107		79
TOTALS		1712		1688

[a] In about 20 000 L, 1 M HNO_3 (~90 g/L)

[b] Assumes 180 days from reactor discharge and 0.75% heavy metal loss to HLL waste (Th and U for HTGCR reprocessing and U and Pu for LWR).

[c] Gross estimates

TABLE 8

COMPARISON BETWEEN TRANSURANIUM PRODUCTION IN TWO 1000 MW(e) REACTORS WITH DIFFERENT FUELS AT THE END OF 1 CYCLE (3 YEAR POWER GENERATION, 3 YEAR COOLING)

[after Raman et al. 1975]

Uranium Fuel (kg)		Thorium Fuel (kg)
^{235}U - 2 630 ^{234}U - 20 ^{238}U - 77 350		^{233}U - 4 500 ^{232}Th - 75 500
Flux		Flux
2.9×10^{13} neutrons/cm^2/s		1.2×10^{13} neutrons/cm^2/s
Quantity (kg)	Isotope	Quantity (kg)
39	^{237}Np	0.04
14	^{238}Pu	0.003
442	^{239}Pu	0.0002
174	^{240}Pu	$<2 \times 10^{-5}$
72	^{241}Pu	$<2 \times 10^{-6}$
28	^{242}Pu	$<8 \times 10^{-8}$
13	^{241}Am	$<3 \times 10^{-7}$
0.07	^{242m}Am	$<4 \times 10^{-10}$
8	^{243}Am	$<4 \times 10^{-9}$
0.008	^{242}Cm	$<2 \times 10^{-11}$
0.006	^{243}Cm	$<3 \times 10^{-12}$
2.2	^{244}Cm	$<3 \times 10^{-10}$
0.16	^{245}Cm	$<5 \times 10^{-12}$

metals in the LWR fuel, its waste will contain about three times that in the HTGCR fuel waste. The amount of Pu in the LWR waste is smaller because it is ^{239}Pu and therefore can be extracted for future use, whereas the HTGCR waste contains mostly ^{238}Pu. From the same table, it may be noted that the HTGCR waste will contain some ten times less curium (Cm) than LWR wastes; this means considerably less neutron shielding requirements. However, it will not be ten times less, as the fluoride in the HTGCR waste will contribute neutrons from the (α,n) process.

Raman et al. [1975] compared two similar reactors fuelled, in one case, with about 3.3 per cent enriched uranium, and in the other, with thorium plus about 5.6 per cent ^{233}U (Table 8). They calculated that at the end of one fuel cycle of three years' operation, followed by three years' cooling, the amount of nuclides with mass numbers greater than ^{238}Pu would be less by $> 10^6$ in the Th/^{233}U fuel relative to the 3.3 per cent ^{235}U fuel. Although this calculation was made for an HTGCR, the results are applicable to the thorium cycle in general.

By recycling the wastes from a $^{235}U/^{238}U$ reactor repeatedly in a $^{233}U/Th$ reactor, it was shown that reductions of all the heavy isotopes except ^{246}Cm and ^{252}Cf can be achieved. Further extended calculations are expected to establish the reduction of these isotopes also. The rate of reduction depends upon the amount of ^{238}U originally in the waste. It is not yet clear if this is an overall advantage, since ^{252}Cf is produced in this system and is a starting point for the production of berkelium and californium isotopes. The activity of the latter may require extra shielding during processing or transport. More knowledge of the fission cross section of ^{245}Cm is necessary; to achieve this, a comprehensive program aimed at providing basic data for the recycle concept was initiated by Dabbs et al. [1975] and Cullen et al. [1977].

Radioactive waste material can be roughly divided into two main types: fission products and actinides*. The fission products decay to safe levels in about 1000 years; the actinides, however, live for a considerably longer period (Figure 27). Consequently, for the wastes from a $^{235}U/^{238}U$ cycle to be reduced to a 1000 year problem, it is necessary to remove 99.99 per cent Pu, 99.9 per cent U, Am, and Cm and 95 per cent Np during the processing, then

* Transuranic elements which occur in the actinide series of the chemical periodic table. They are produced in reactors by successive capture of neutrons by heavy metals.

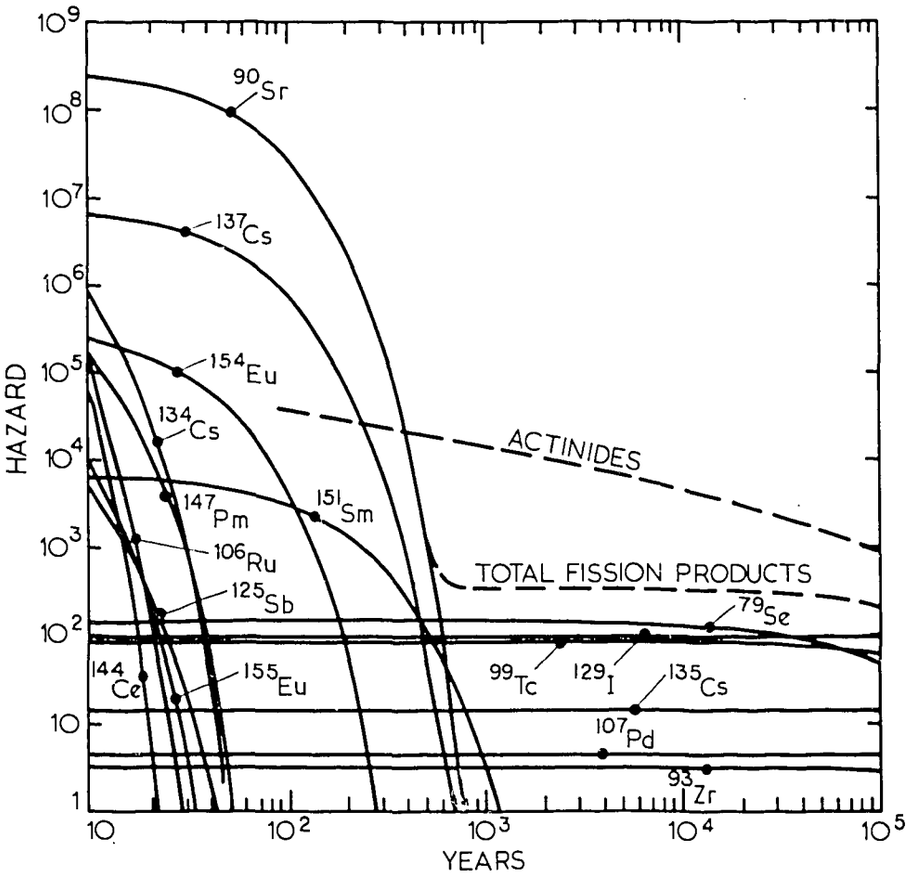

FIGURE 27 RELATIVE HAZARD OF THE FISSION PRODUCT ISOTOPES [after Harries 1974]

dispose of them, probably by burning in a reactor. It is reasonable to expect that actinides may be partitioned from waste [Campbell and Buxton 1976], but to achieve this considerable design and engineering effort is required. The fact that many more successive neutron captures are necessary in the Th/^{233}U to produce ^{237}Np, ^{239}Pu and the transplutonium elements means that the actinide content of the Th/^{233}U cycle waste is reduced sufficiently for storage to be considered a 1000 year problem without partitioning.

Using data of Raman et al. [1975], Brogli et al. [1975] calculated actinide build-up as a function of years of operation with ^{233}U feed for high conversion ratio HTGCRs (Figure 26); it is apparent that the actinide content of irradiated fuel could be up to an order of magnitude less than that for the uranium cycle, per unit of energy produced.

The other main difference at this stage is that the Thorex solution required to dissolve thoria contributes considerably more chemicals to the high level waste and consequently produces more corrosion products. Both these items are reflected in Table 7.

11.2 Solid Waste

There are considerable differences between the solid waste from HTGCRs and that from other reactors, particularly at the intermediate to high level. HTGCR silicon carbide hulls and (possibly) the untreated burned-up fissile particles represent less than 10 per cent of the bulk of discarded metal hulls. At low levels, the calcium carbonate from the graphite hulls of HTGCRs contains ^{14}C and represents an enormous volume even if it is a suitable substance for burial.

11.3 Waste Solidification

The techniques used for waste solidification will depend upon the type of solidification agreed upon: calcination, glass or ceramic. The principal difference between the U cycle and Th cycle wastes arises from the presence of large quantities of fluoride in the HTGCR waste which may require chemical stabilisation or an HF removal system in the vitrification off-gas treatment system.

12. HAZARDS

12.1 Relative to Other Energy Resources

The hazards of thorium must be compared not only with uranium, but also with other fuels. To do this, comparisons must be made first between fossil and nuclear power and then between uranium and thorium fuel cycles.

Apart from the incalculable damage to the world's ancient monuments by fossil fuel pollutants and the climatological changes possible with excessive carbon dioxide concentrations in the atmosphere (the 'greenhouse effect'), the health hazards in most cases are significantly greater than those of nuclear fuels. Hamilton and Manne [1977] have calculated mortality and morbidity rates for the USA and these are given in Table 9.

TABLE 9
MORTALITY AND MORBIDITY RATES FROM PRODUCTION OF 10^{10} kWh*
OF ELECTRIC POWER IN THE USA
[after Hamilton and Manne 1977]

Method	Deaths	Disabilities
Coal	20-200	300-500
Oil	3-150	150-300
Gas	0.2	20
Nuclear	1-3	8-30

*10^{10} kWh = 0.036 EJ

They also calculated that a hypothetical nuclear moratorium in the USA in 1977, in which all nuclear power is replaced by the most economic mixture of coal, gas and oil, would lead to between 1500 and 18 000 excess annual deaths by the year 2000. The upper limit is 1 per cent of the total projected annual death rate for this year. The morbidity values due to air pollution were estimated to be some five times greater than the mortality values.

12.2 Hazards of the Uranium and Thorium Cycles

These hazards are discussed and compared under several broad headings.

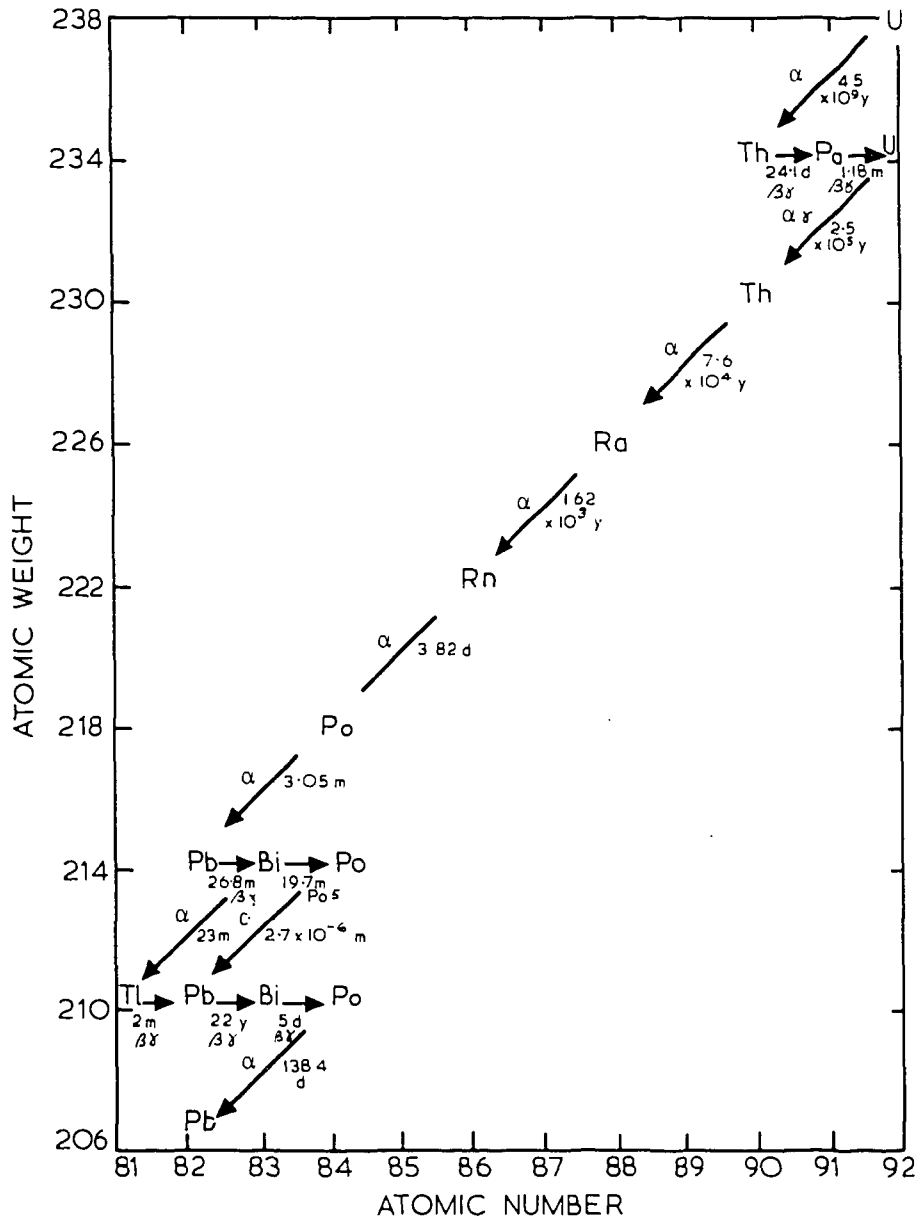

FIGURE 28 URANIUM RADIOACTIVE DECAY SCHEME

Mining

The principal hazards in uranium mining are the alpha-emitters ^{226}Ra and its first daughter, the inert gas ^{222}Rn (Figure 28). With thorium, the hazard is ^{224}Ra and ^{222}Rn, also both alpha-emitters (Figure 4). In both cases, the radon hazard can be minimised by adequate ventilation in underground mining.

Underground mining is used to produce most of the uranium in Canada, South Africa and France. About half of the American and all of the Australian production is by open-cut mining; Jabiluka in the Northern Territory of Australia is, however, likely to be underground. A good deal of the current thorium production is a by-product of uranium mining but, should the cycle develop further, production is more likely to come from monazite sand (Tables D1 to D6) which would be an open-cut or beach mine and therefore would present only a very small radon hazard.

Most of the radiation from the early members of the chain (Figures 4 and 28) is absorbed within the ore body and is relatively harmless, but ^{222}Rn from uranium and ^{220}Rn from thorium are gases and can diffuse from the ore into the atmosphere. The radon, being an inert gas, does not attach itself chemically to any substance and, when breathed into the lungs, is rapidly eliminated, only a minute quantity being dissolved in the body fluid and circulated. The daughter products of Rn, however, attach themselves to dust particles or form the nucleus of water droplets [Fry 1975], but some remain free, as unattached radioactive atoms.

Inhalation of air containing these daughter products results in the free atoms being deposited in the upper respiratory tract and the attached atoms being deposited more deeply in the lungs. The half-lives of these particles (Table 10) are short compared with the effective biological half-life of about eight hours [Khan et al. 1974]; as a consequence, practically all of the β- and α-particles emitted from the chain descending from the inhaled particles are absorbed in the lung tissue. This absorbed radiation may kill a few cells but, more importantly, it may damage others and cause them to become malignant.

The damage occurring within the lung is a function of the energy deposited; and Table 10 shows that for the same equilibrium dissociation rate, the energy available for deposition by the ^{220}Rn daughters is about 13.7 times more than that from ^{222}Rn. However, because the biological half-life is

TABLE 10

COMPARISON OF THE α-ENERGY POTENTIAL OF ^{222}Rn AND ^{220}Rn DAUGHTERS

^{222}Rn from Uranium Series						^{220}Rn from Thorium Series					
Isotope	α-energy (MeV)	Half-life	No. of Atoms in 100 pCi	Ultimate α-energy Available per Atom	Total Ultimate Energy (MeV)	Isotope	α-energy (MeV)	Half-life	No. of Atoms in 100 pCi	Ultimate α-energy Available per Atom	Total Ultimate Energy (MeV)
^{218}Po	6.00	3.05 min	977	6.00 + 7.69*	0.134×10^5	^{216}Po	6.78	0.15 s	0.8	6.78 + 0.36 x 6.06 + 0.64 x 8.78	11.6
^{214}Pb	0	26.8 min	8580	7.69	0.660×10^5	^{212}Pb	0	10.64 h	204420	0.36 x 6.06 + 0.64 x 8.76	1.595×10^6
^{214}Bi	0	19.7 min	6310	7.69	0.485×10^5	^{212}Bi	6.06	60.60 min	19405	0.36 x 6.06 + 0.66 x 8.78	0.151×10^6
^{214}Po	7.69	2.7×10^{-6} min	0.0009	7.69	negligible	^{212}Po	8.78	3×10^{-7} s	1.6×10^{-6}	8.78	negligible
				Total	1.279×10^5					Total	1.746×10^6

*Referring to Figure 28 each ^{218}Po atom will emit an α-particle of 6.00 MeV, producing ^{214}Po after two β-decays and this will emit an α-particle of 7.69 MeV. The α-decay of ^{214}Bi is from a very few short-lived daughters and may be neglected; the α-particle from ^{210}Po has a half-life very much longer than the half-life of biological evacuation and may be neglected also. Thus each ^{218}Po atom present will be the cause of two α-particles being absorbed in the lungs. Similar arguments apply to the ^{216}Po chain, but in this case account has to be taken of ^{212}Bi decay, 36% of which is by α (6.06 MeV) to ^{208}Tl and 64% by β to ^{212}Po, which then decays by α (8.78 MeV) to ^{208}Pb (see Figure 4).

shorter than the half-life of ^{212}Pb (10.6 h), the effective half-life for the ^{220}Rn daughters is 4.5 h and the actual dose received by the lungs is about 5.8 times that from ^{222}Rn. Because of the shorter half-life of uranium compared with thorium, then gram for gram present in the ore, the dose rate in the thorium case would be about twice that of uranium. This means that an underground thorium mine is likely to require a more effective ventilation system than uranium. There are fewer underground thorium mines than uranium and at least one, mining 3 to 6 per cent thorium, has no radon daughter problem from a ventilated system [du Toit 1963]. Most thorium mines are open-cut or monazite placers; in these, normal atmospheric dilution ensures that ^{220}Rn and its daughters are not a serious problem [Raghavayya 1974]. This argument applies to open-cut uranium mines also.

The other radiation hazards of mining, that is the gamma-radiation from the ore body and the inhalation of dust containing the long-lived daughter products, are in general not a great problem, the dose rates from both being generally below the recommended values for radiation workers. In a very few areas there are higher dose rate regions and miners in these regions are carefully watched for evidence of over-exposure [Fry 1975; Raghavayya 1974].

Milling

Milling is a relatively straightforward hydrometallurgical operation which gives rise to only minor radiological hazards for the mill workers. Essentially, the decay chain is split into two fractions, with the most radioactive part going into the tailings and the least, into the concentrated uranium or thorium oxides.

Not all of the daughter products are removed in the process; a small fraction of the radon is released during the grinding and leaching stages, most of it being retained in the tailings. In the case of the concentrated uranium, the ^{234}Th and ^{234}Pa will decay quite rapidly (Figure 28); a small trace of ^{226}Ra emitting a few soft gamma-rays will remain as an impurity in the uranium concentrate (yellowcake) produced but most of the gamma activity is now due to the ^{235}U fraction. This leads to radiation levels of the order of 2 to 3 mrem/h near drums of stored material [Fry 1975]. The thorium case is rather different; its pre-radon daughters are longer-lived and the ^{228}Ac (Figure 4) emits more and harder gamma-rays than the ^{226}Ra of the uranium chain. This leads to radiation levels of around 20 to 30 mrem in the region of the stored thorium concentrate [Murthy and Nambiar 1964].

The greatest occupational hazard in the milling area is due to the inhalation of dust. This does not appear to have been a significant hazard even in the days when occupational hygiene was less strictly controlled than now [Fry 1975].

Tailings

The tailings from both uranium and thorium mining have radioactive constituents, but there is a significant difference. The presence of ^{230}Th in the uranium tailings, with its half-life of 7.6×10^4 years (Figure 28) ensures a continuing supply of ^{226}Ra and subsequent daughters. In the thorium tailings, however, once the thorium has been removed, the longest-lived daughter is ^{228}Ra with a half-life of 5.75 years; consequently, the radiation will eventually decay to background levels. In India, it is usual to dump the tailings on the beach and allow them to be dispersed by the tide [Murphy and Nambiar 1964].

Fuel reprocessing and fabrication of recycle fuel

The problems of fuel reprocessing are described more fully in Section 9. Fabrication of recycle fuel presents a problem because the thorium cycle involves the production of ^{232}U from ^{233}U directly by a (n,2n) reaction* or indirectly from various thorium isotopes by various neutron reactions. Uranium-232 (half-life 72 years) produces a series of daughter elements (particularly ^{212}Bi) and their decay products (Figure 4), which produce such high levels of radioactivity within about two weeks of chemical purification, that fabrication of recycle fuel may require shielded facilities and remote active-handling equipment.

13. THE THORIUM CYCLE AND NUCLEAR PROLIFERATION

Although ^{233}U is a potential weapons material, most versions of the thorium-uranium cycle use ^{233}U diluted with natural uranium, which is considered to be 'denatured'. In this state, the fresh fuel would contain about 2 per cent ^{233}U, 14 per cent ^{238}U and 84 per cent Th. Fresh fuel, which has no fission product content and is therefore easily handled, would have no plutonium content and the ^{233}U can only be separated with difficulty.

* If a neutron with energy greater than about 8 MeV is absorbed by a heavy element, two neutrons may be emitted without fission.

However, if isotope separation technology continues to advance as rapidly as it has done over the last two decades, denaturing may become ineffective [see, for example, American Physical Society 1978]. The fact that there is a five-mass unit gap between the ^{233}U and the ^{238}U makes isotopic separation relatively easy.

The highly radioactive irradiated fuel, although requiring costly and sophisticated handling equipment which is economic only in very large plants, has the disadvantage from a safeguards point of view that immediately on discharge from the reactor, ^{233}Pa, the precursor to ^{233}U, can be separated using a chemically simple co-precipitation process. The ^{233}Pa decays with a half-life of 27.4 days to pure ^{233}U (Figure 4).

Plutonium-239 is formed from the ^{238}U in the denatured fuel and its separation is not difficult, but irradiated denatured fuel such as that described above would contain 0.1 or less of the Pu in irradiated low enriched uranium in current use [ERDA 1977; Science 1977].

Dahlberg [1978], discussing the proliferation cycle, concluded that if the risk of weapons proliferation is the only criterion, then once-through fuel cycles are the preferred form of recycle. However, when criteria relating to power cost, resource utilisation and preservation of future non-fossil energy options are applied, the recycle options appear favourable. Those cycles using medium-enriched uranium and thorium rank higher than those using low-enriched uranium, both overall and with respect to safeguards criteria. Thus it seems that because of the cost and complexity of the necessary separation plant, the thorium fuel cycle would make diversion by terrorist groups more difficult; this would not be the case nationally.

In 1980, the US Department of Energy (DOE) published a comprehensive report on the interrelation of fuel cycles and non-proliferation [DOE 1980].

14. CONCLUSIONS

Fission reactors will continue to play a large and continually increasing part in the energy field; to ensure the most efficient exploitation and conservation of natural resources, the thorium fuel cycle must eventually be developed to its full potential.

If necessary, all the modern reactor types can be adapted to the thorium cycle, thus prolonging the life of fission reactor energy production. At present, however, most types suffer from some disadvantage in using the Th cycle: this includes economic inferiority, engineering research, low breeding ratios, etc. Table 1 summarises these problems.

One or two thorium systems offer considerable advantages over uranium fuelled reactors. The MSBR has very favourable fuel utilisation, safety characteristics and high thermal efficiency, but requires a very large research effort to solve the on-line reprocessing problems. The CANDU system, especially if using the organic liquid-cooled option, is an obvious potential thorium cycle user and the stated objectives of Atomic Energy of Canada Ltd (AECL) on the study of this cycle should ensure its use in a few years' time. Regardless of fuel utilisation considerations, the HTGCR should be developed in its own right as a process heat and high efficiency electricity producer. Above all, the whole thorium cycle-energy production theme should be optimised along the CANDU-PWR self-sufficient equilibrium thorium cycle and the symbiotic GCFBR-HTGCR systems, which could solve the energy supply problems provided that the whole world, developed and under-developed, can limit the average annual increase in energy demands.

The thorium cycle has a number of advantages over uranium but these are offset to a great degree by the large development costs involved, particularly in the fuel reprocessing area. The widespread use of thorium for power generation is not likely to occur until low cost uranium reserves are substantially depleted, possibly in the first half of the 21st Century. This time scale is not so long that the development of the necessary techniques can be temporarily shelved. The successful development and introduction of plutonium fuelled breeder reactors should accelerate the introduction of thorium as a fuel.

15. REFERENCES

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16. FURTHER READING

Delays in publishing this report, due mainly to staff movements within the Commission, has meant that more recent publications have not been examined in detail. For the benefit of the reader a list of recent literature not specifically referred to in the text is given below. At the time of writing all are available in the AAEC library at Lucas Heights.

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17. ABBREVIATIONS

AAEC	Australian Atomic Energy Commission
AECL	Atomic Energy of Canada Ltd
AGR	Advanced gas-cooled (graphite) reactor
AHCF	Aqueous homogeneous critical facility
AI	Atomics International
ANL	Argonne National Laboratory (USA)
ASR	Aqueous suspension reactor
AVR	Arbeitsgemeinschaft Versuchsreaktor, Juelich (FRG)
BB/K	Brown-Boveri-Krupp Reaktorbau GmbH
BCC	body-centred cubic
BNL	Brookhaven National Laboratory (USA)
BNWL	Battelle North West Laboratory (USA)
BR	breeding ratio
BWR	Boiling water reactor
CA	Critical assembly
CACS	Core auxiliary cooling system
CNEN	Comitato Nazionale per L'Energia Nucleare (Italy)
CR	Conversion ratio

DOE	Department of Energy (USA)
EBR	Experimental breeder reactor
(E)NEA	(European) Nuclear Energy Agency (from 1972, OECD dropped 'European' from the name of this agency)
ENSEC	European Steelmakers' Club
ERDA	Energy Research and Development Administration (USA)
ERR	Elk River Reactor
FBR	Fast breeder reactor
FCC	Face-centred cubic
FIFA	Fission per initial fissile atom
FIMA	Fission per initial metal atom
FRG	Federal Republic of Germany
GCFBR, GCFR	Gas-cooled fast (breeder) reactor
HE	Highly enriched (uranium cycle)
HEPA	High efficiency particulate air (filter)
HTGCR, HTGR	High temperature, gas-cooled reactor
HTR	High temperature reactor
HWR	Heavy water reactor

HWOCR	Heavy water-moderated, organic-cooled reactor
IAEA	International Atomic Energy Agency
INFCE	International Nuclear Fuel Cycle Evaluation
JAERI	Japanese Atomic Energy Research Institute
KALC	Krypton absorption by liquid carbon dioxide
KFK	Kernforshungzentrum Karlsruhe GmbH (FRG)
KSH	Kernkraftwerk Schleswig-Holstein (FRG)
KSTR	Kema suspension test reactor (The Netherlands)
LE	Low enriched (uranium cycle)
LMFBR, LMBR	Liquid metal-cooled (fast) breeder reactor
LPR	Low power reactor
LWR	Light water-cooled and moderated reactor
LWBR	Light water breeder reactor
MSBR	Molten salt breeder reactor
MSR	Molten salt reactor
MSRE	Molten salt reactor experiment
NRTS	National Research Testing Station, Idaho (USA)

OCR	Organic-cooled reactor
OECD	Organization for Economic Cooperation and Development
ORNL	Oak Ridge National Laboratory (USA)
PCRV	Pre-stressed concrete reactor vessel
PDP	Process development pile
PHWR	Pressurised heavy water-moderated reactor
PWR	Pressurised water reactor
RE	Rare earths
RHR	Residual heat movement
ROSPO	Reaktore organico sperimentale potenza zero, CNEN (Italy)
SBR-1	Soviet Breeder Reactor 1
SGHWR	Steam generating, heavy water-moderated reactor
SLE	Small lattice experiment
SRE	Sodium reactor experiment
SSCR	Spectral shift control reactor
SSET	Self-sufficient equilibrium thorium (cycle)
SWU	Separative work unit
TBP	tributyl phosphate

THTR	Thorium high temperature reactor
THUD	Thorium, uranium, deuterium
UKAEA	United Kingdom Atomic Energy Authority
USAEC	United States Atomic Energy Commission
VHTR	Very high temperature reactor

APPENDIX A
A COMPARISON OF THE PHYSICS PARAMETERS OF THORIUM, URANIUM AND PLUTONIUM ISOTOPES

A.1 THE NUMBER OF NEUTRONS AVAILABLE FOR BREEDING

For each neutron absorbed by the fuel a number, η, are emitted of which one is necessary to continue the chain reaction, leaving a maximum of η-1 available for breeding. This value is energy dependent; Figure A2 shows that in a thermal reactor ^{233}U has the highest potential for breeding, while at fast reactor energies ^{239}Pu and ^{241}Pu have the higher potentials. These features influence the present choice of uranium as the preferred fuel cycle in fast reactors.

A.2 MINIMUM CRITICAL MASS REQUIREMENT

Figure A1 shows how the minimum critical mass of each of the more common fissile materials varies with neutron energy. A minimum fuel requirement is important from the point of view of cost and availability.

Figure A1 also shows that for thermal systems, plutonium has the lowest critical mass. Furthermore, for a $^{235}U/^{238}U$ fuel, if the burn-up of ^{238}U is equalled by the breeding of ^{239}Pu, a gain in reactivity results because the critical mass of the $^{235}U/^{239}Pu$ mixture decreases. This desirable quality (to compensate for fission product absorption) is slightly enhanced in a similar way by the build-up of ^{241}Pu. For a $^{235}U/^{232}Th$ initial mixture, such an increase in reactivity does not occur. Since there is no significant difference between the initial mass of ^{235}U and ^{233}U, there is no significant change in the critical mass of the resulting mixture of these isotopes.

The critical mass factor considered above does not account for the difference between the conversion ratios of the two cycles. The thorium cycle breeds more fuel than the uranium and as a result the uranium advantage largely disappears, especially when comparing the equilibrium conditions of $^{235}U/^{232}Th/^{233}U$ (recycle) versus $^{235}U/^{238}U/^{239}Pu$ (recycle). In general, the larger the fuel exposure in a thermal reactor, whether first cycle or recycle, the less is the advantage obtained with the uranium. Consequently, if low power costs are the result of the use of first fuel cycle and low fuel exposure, the uranium cycle is preferred, and if low power costs are

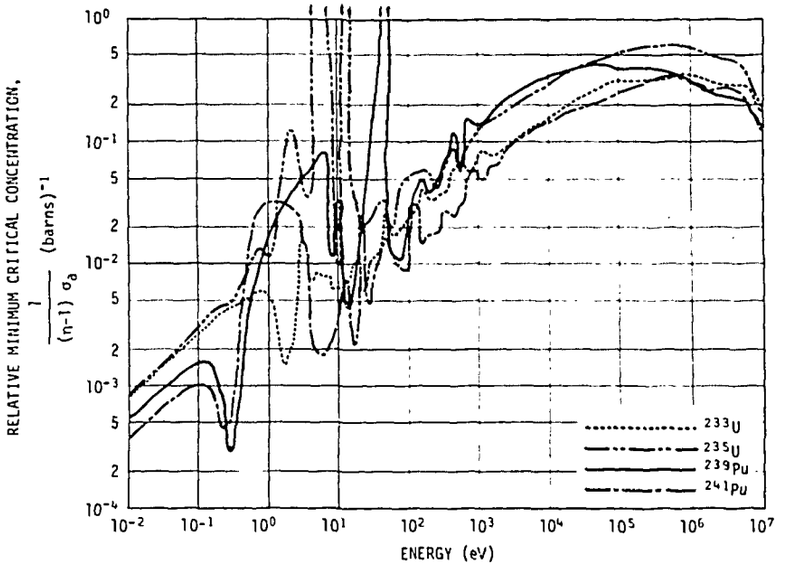

FIGURE A1 RELATIVE MINIMUM CRITICAL CONCENTRATION FOR THE FISSILE FUELS AS A FUNCTION OF NEUTRON ENERGY [after Kasten 1970]

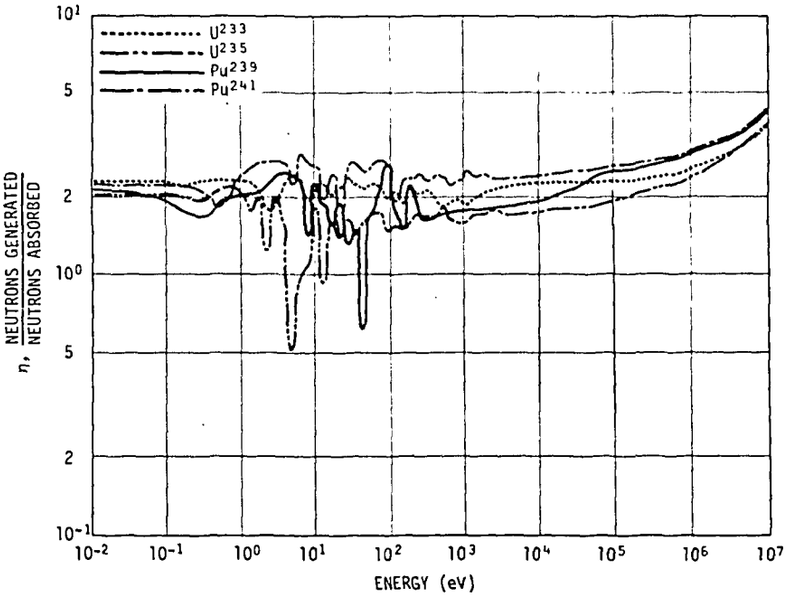

FIGURE A2 VALUES OF η FOR FISSILE FUELS AS A FUNCTION OF NEUTRON ENERGY [after Kasten 1970]

associated with fuel recycle and long fuel exposures, the thorium cycle becomes the more attractive.

In the high energy spectra of fast reactors (i.e. greater than, say, 100 keV), ^{235}U has a higher critical mass over the large part of the energy range than any of the other fissile isotopes. This, together with its low η-value (Figure A2), means that ^{235}U is not a good fissile isotope above about 100 eV. Although ^{233}U would have a lower critical mass, lower energy region of fast reactor spectra, the tendency is to try for harder spectra with better potential breeding characteristics, which means that in general the uranium cycle, with recycle of ^{239}Pu, is preferred in fast reactors.

A.3 THE ABSORPTION OF NEUTRONS IN ^{238}U AND ^{232}Th

From Figure A3 it will be seen that the capture cross section of thorium is considerably higher than that of ^{238}U at low or thermal neutron energies. This means that a thermal reactor using thorium as a fertile material needs relatively more enrichment (i.e. added fissionable material) than if using ^{238}U as the fertile material. The resonance behaviour tends to favour the thorium but, unless the fuel mixture is homogeneous, i.e. not in 'lumps' such as fuel pins or spheres, the critical mass with the thorium is the larger and therefore the more costly.

The resonance peaks affect the temperature coefficient of the fuel mixture. If the temperature increases, the peaks become wider (this is called the Doppler effect) and the mean neutron absorption cross section increases. The net effect is a tendency to increase neutron absorption relative to production and thus cause a negative temperature coefficient of reactivity. It is not obvious in Figure A3, but thorium has resonance peaks at higher energies than uranium and this tends to give fast reactors an overall negative temperature coefficient when using thorium as the fertile material; this favours the thorium cycle.

If the neutrons being absorbed have energy greater than about 0.82 MeV in the case of ^{238}U, or 1.4 MeV in the case of ^{232}Th, fast fission occurs, enhancing the system. This favours the uranium cycle.

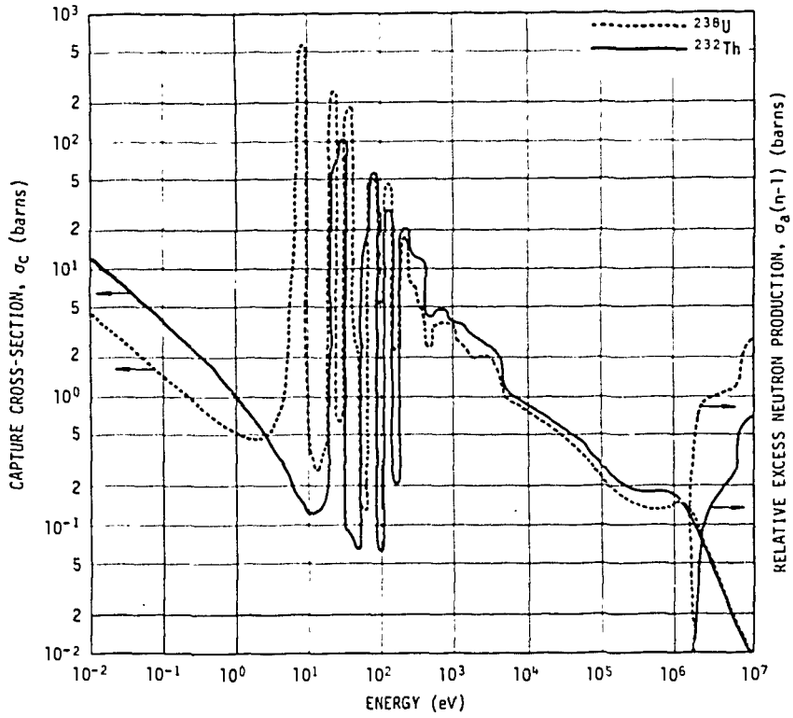

FIGURE A3 CAPTURE CROSS SECTIONS AND RELATIVE EXCESS NEUTRON PRODUCTIONS FOR ^{232}Th AND ^{238}U AS A FUNCTION OF NEUTRON ENERGY [after Kasten 1970]

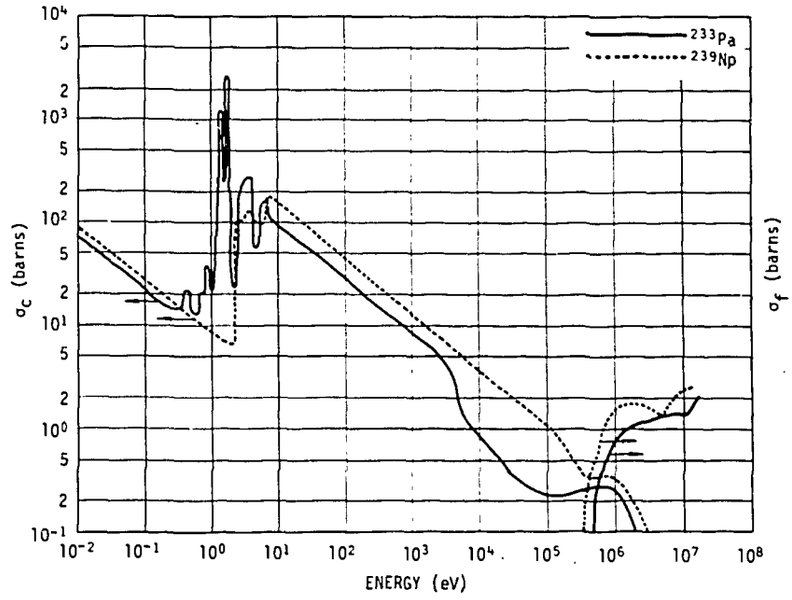

FIGURE A4 CAPTURE AND FISSION CROSS SECTIONS FOR ^{233}Pa AND ^{239}Np AS A FUNCTION OF NEUTRON ENERGY [after Kasten 1976]

A.4 NUCLEAR CHARACTERISTICS OF HIGHER ISOTOPES

Uranium-233 and plutonium-239 can capture a fraction of neutrons without fissioning; this leads to the production of higher isotopes such as ^{234}Pu (fertile), ^{235}U (fissile), ^{236}U (parasitic), ^{240}Pu (fertile), ^{241}Pu (fissile) and ^{242}Pu (parasitic).

In the thorium cycle, if the conversion ratio is less than 1.0, some ^{235}U make-up will have to be added at each new fuel change, leading to a gradually increasing fraction of fission in ^{235}U and a consequent slight lowering of the η-value.

In the uranium cycle, the build-up of ^{241}Pu is beneficial to the overall η-value; however, the absorption cross sections of ^{240}Pu and ^{241}Pu are larger than those of the corresponding nuclides in the thorium cycle. In general, the production of higher isotopes favours the thorium cycle for thermal reactors and the uranium cycle for fast reactors.

A.5 INTERMEDIATE ELEMENTS ^{233}Pa AND ^{239}Np

These isotopes are intermediate between the primary fertile material and the fissile material produced (see Figures 3 and 4). Neutron absorption by these materials causes a loss in the production of fissile material and also an increase in the parasitic neutron absorption. Both ^{233}Pa and ^{239}Np have small, fast fission cross sections, with ^{239}Np having slightly better characteristics, but the difference here is hardly significant.

Neutron loss in these materials is proportional to the flux, cross section (Figure A4) and time. The capture cross sections are comparable, but the half-life of ^{233}Pa is about 27.4 days and that of ^{239}Np is 2.3 days. This means that compared with the uranium cycle the neutron flux in the thorium cycle is limited by the necessity to keep absorption by ^{233}Pa acceptably low. However, certain designs of liquid fuelled reactors (e.g. MSBR) would not be restricted in this way, since ^{233}Pa would decay to ^{233}U outside the reactor.

A.6 DELAYED NEUTRON FRACTION AND ENERGY RELEASE PER FISSION

The smaller the delayed neutron fraction β (Table A1) the more rapidly a reactor will respond to changes in the multiplication constant. In the thermal systems, both ^{233}U and ^{239}Pu have relatively small β values compared

with ^{235}U but, after the first cycle, the $^{235}U/^{232}Th/^{233}U$ (recycle) case has a lower β value than the $^{235}U/^{238}U/^{239}Pu$ (sale) case. Although this would necessitate some changes in design criteria to eliminate possible safety consequences, it would not necessarily impose a significant economic penalty on a particular cycle.

TABLE A1

DELAYED NEUTRON FRACTIONS

Nuclide		β
^{233}U		0.0027
^{235}U		0.0065
^{239}Pu		0.0021
^{241}Pu		0.0049
^{238}U	fast fission only	0.0148
^{232}Th	fast fission only	0.0203

Fast reactors would run on a recycle system for both fuel cycles and there is a significant difference between the β values for ^{238}U and ^{232}Th. However, the lower fission cross section of ^{232}Th means that it contributes less to the effective β value than ^{238}U. Consequently, in fast reactors the β value is about the same for both cycles.

A.6.1 The Delayed Neutron Fraction β

If η is the number of neutrons emitted per neutron absorbed by a fissile nucleus and ν is the number of neutrons emitted per fission of such a nucleus, then

$$\eta = \nu\sigma_f/(\sigma_c + \sigma_f)$$

where σ_c and σ_f are the capture and fission cross sections for the conditions of the experiment.

Of the neutrons produced, most are emitted in a time less than 10^{-14} s; these are called prompt neutrons. A very small percentage (Table A1) are emitted over several hours afterwards, the numbers rapidly changing with time;

these are the delayed neutrons.

For each number of neutrons, ν, emitted per fission, some will be lost through non-fission capture in fuel, moderator and structural materials and others lost through leakage from the fuel regions, leaving a fraction $g\nu$, say, for the next generation of neutrons $(g\nu)^2$ for the next and so on. The factor $g\nu$ is called the multiplication factor k.

If it can be shown that, for an infinitely large matrix of core material (i.e. no neutron can be lost by leaking out of the system), the infinite multiplication factor k_∞ is greater than one, then it will be possible to construct a finite system with an effective multiplication factor k_{eff} equal to one. This is the criterion for a reactor being critical.

In practice, it is necessary to be able to make k_{eff} greater than one and to reduce it to one or less by inserting an absorber or control rod. Thus changes in the multiplication factor can be made which cause the reactor power level to rise or fall at a rate depending upon the value of the delayed neutron fraction β and the change in multiplication factor.

A.7 CONVERSION RATIO, BREEDING RATIO AND DOUBLING TIME

The conversion ratio is defined as the number of fissile nuclei formed (^{239}Pu) to the number destroyed (^{235}U). If the fuel contains a large proportion of fertile material, as in the case of natural or slightly enriched uranium, the ^{239}Pu produced in this way makes an important contribution to the burn-up. In fact, with a conversion ratio greater than about 0.8, an increase in reactivity occurs soon after start-up, peaks and then decreases steadily. For fuels containing more than about 2 per cent enriched uranium, the initial conversion ratio is too small for this to be noticed. At the end of its life time, the fuel can be reprocessed and the unburnt fraction, original and converted, extracted for further use. In the case of deliberate conversion, the fertile material may be kept partially separated from the fuel or core when used as a blanket.

The breeding ratio is defined in a similar way to the conversion ratio, i.e. the ratio of the number of fissile nuclei formed to the number destroyed, but in this case ^{233}Th is produced from ^{232}Th. A breeding gain may be defined as the excess of the breeding ratio over unity.

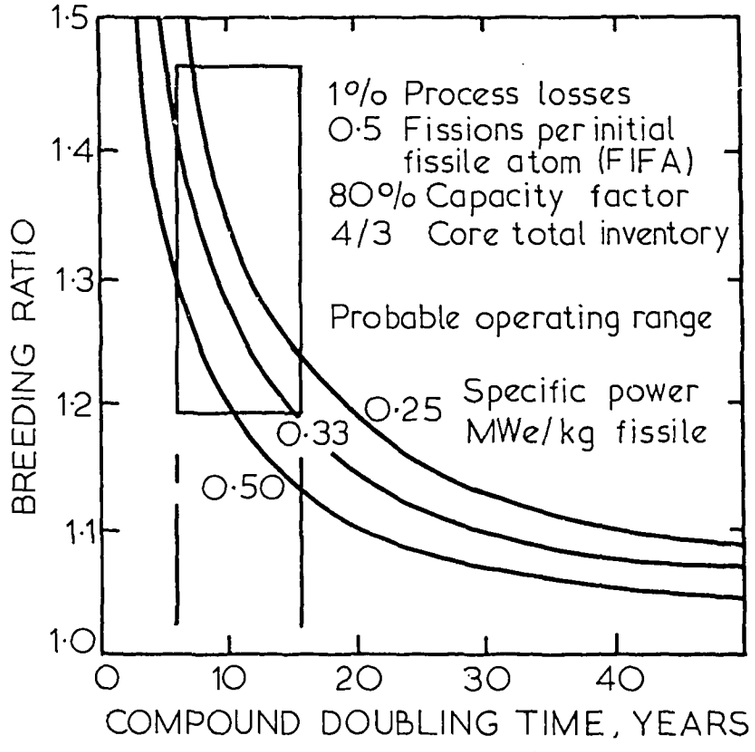

FIGURE A5 INTER-RELATIONSHIP OF LMFBR OPERATING PARAMETERS

Doubling time could be defined as the time required for a reactor to produce a surplus amount of fissile material equal to that required for the initial loading. This definition is not very useful because of the long time spent by the fuel in other parts of the fuel cycle such as reprocessing. It is therefore more usual to define the doubling time as the time taken to double the inventory of the fuel cycle.

If the initial amount is W grams and ω grams are consumed each day, then the doubling time in days is $T = W/\omega G$ where G is the breeding (or conversion) gain defined above. Of the ω grams consumed, some undergo fission, the rest capture and, if α is the ratio of capture/fission, the amount of material undergoing fission is $\omega/(1+\alpha)$. Since 1 g of fissile material consumed per day produces approximately 1 MW of power, then $T = W/GP(1+\alpha)$ where T is in days, and P is in megawatts.

Thus a desired short doubling time can be achieved by having a large gain which is limited eventually by the maximum number of neutrons produced per fission, or by a large value of P/W. The latter is obtained by having a high specific power and the attendant difficulties of getting the heat out of the core, or by having the smallest possible quantity of material in the fuel cycle outside the core; this introduces the difficulties of reprocessing fuel after short cooling (radioactive) periods. Figure A5 shows the relationship between breeding ratio, doubling time and power density.

A.8 OTHER PHYSICAL PROPERTIES

These are summarised in Table A2.

TABLE A2

SELECTED PHYSICAL PROPERTIES OF THORIUM AND URANIUM METALS, OXIDES & CARBIDES

[after Kasten 1970 : see also Rand et al. 1975]

	UO_2	ThO_2	U (metal)	Th (metal)	UC	UC_2	ThC	ThC_2
Melting point (°C)	2750	3290	1130	1700	2320	2480	2625	2655
Density at room temperature (g/cm^3)	10.5	9.7	19.0	11.6	13.0	11.0	10.6	9.6
Thermal conductivity* at 650°C (W/cm °C)	0.035	0.040	0.37	0.45	0.23		~0.2	~0.25
Temperature at which phase change occurs (°C)			665 (α to β) 775 (β to γ)	1375 (FCC TO BCC)				

*Ceramics generally suffer a decrease in conductivity with long reactor exposure at relatively low temperature, which is not considered in the above values. At high temperatures ($\gtrsim$ 1700°C) irradiation effects on k do not appear significant. FCC = face-centred cubic, BCC = body-centred cubic.

APPENDIX B
TOPPING CYCLE

The use of a topping cycle is to improve the efficiency of a power plant by matching the properties of the combustion temperature to the working fluid. In the steam cycle, for example, although the combustion temperature is around 1700°C, the fundamental properties of water limit the peak system temperatures to about 500°C and the plant efficiency to about 40 per cent.

The most obvious way to improve this efficiency is to convert part of the high temperature heat, i.e. that between 1700 and 500°C, into electricity before the balance is used in the steam phase. One favoured method is to use a metal vapour Rankine cycle (potassium and caesium are probably the best) in which a turbine is driven by the high temperature metal vapour and the non-condensing exhaust is used to heat the steam or vaporise the condensate exhaust from the main steam turbine before re-introducing it to the boiler or superheater. Such topping procedures can significantly improve the plant efficiencies to a degree depending upon the peak cycle temperature (Figure B1). Other topping cycles considered are gas turbine, magnetohydrodynamic (MHD) generators and thermionic converters.

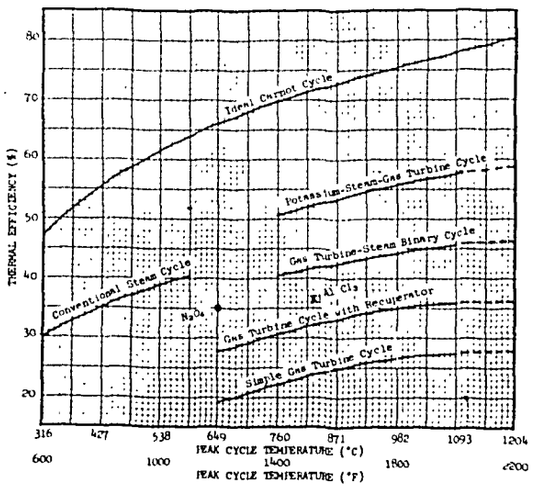

FIGURE B1 COMPARISON OF TYPICAL THERMAL EFFICIENCIES FOR REPRESENTATIVE THERMODYNAMIC CYCLES [after Frass 1971]

APPENDIX C
TABLES OF THORIUM-FUELLED REACTORS AND THEIR PARAMETERS

TABLE C1

THORIUM-FUELLED REACTORS AND CRITICAL ASSEMBLIES (CA)

[after Rostogi 1966]

Name	Location	Type	Power	Fuel	Year
SRE	Atomics International, USA	Graphite, Na	20 MW	*93% ^{235}U + Th metal	1951
PDP	Du Pont, USA	D_2O	1 kW	variable	1953
BORAX IV	National Research and Testing Station, USA	BWR	20 MW	*90% UO_2 + ThO_2	1958
ZENITH	Winfrith, UK	Graphite	100 W	*93% UO_2 + ThO_2	1959
Peach Bottom (CA)	La Jolla, USA	Graphite	-	*93% UC_2 + ThO_2	1960
AHCF	Japanese Atomic Energy Research Institute	D_2O	100 W	*20% UO_2SO_4 + D_2O core ThO_2/D_2O blanket	1961
ERR	Elk River, USA	BWR	22 MW(e)	*93% UO_2 + ThO_2	1962
ROSPO	Comitato Nazionale per l'Energia Nucleare, Italy	Organic		*90% UO_2 + Th blanket	1963
SLE	Lynchburg, USA	D_2O		*93% UO_2 + ThO_2	1963
THUD	Argonne, USA	D_2O		*93% UO_2 + ThO_2	1963
Indian Point	Indian Point, USA	PWR	265 MW(e)	ThO_2 + UO_2 (1st core only)	1962
SBR-1	Obninsk, USSR	FBR	100 W	Th blanket	1963
LPR (CA)	Bettis Laboratory, USA	Seed blanket H_2O	-	ThO_2 + 93% $^{235}UO_2$	1963
DRAGON	Winfrith, UK	Graphite, He HTGCR	20 MW	*93% UC_2 + ThC_2	1964
KSTR	Kema, Netherlands	Aqueous suspension	1 MW	*93% UO_2 ThO_2	1964
Kilorod Exp.	Brookhaven National Laboratory, USA	H_2O	-	$^{233}UO_2$ + ThO_2	1964
ZED-2	Chalk River, Canada	D_2O	200 W	*93% UO_2 + ThO_2	1965
Peach Bottom	Peach Bottom, USA	Graphite HTGCR	40 MW(e)	ThC_2-UC_2	1968
MSRE	Oak Ridge, USA	Molten salt	10 MW	*93% UF_4 + ThF_4	1965
AVR	Juelich, FRG	HTGCR pebble bed	15 MW(e)	ThC_2 + 93% UC_2	1967
KSH	Geesthacht, FRG	HTGCR Gas turbine	22 MW(e)	ThC_2 + UC_2	1973
Fort St. Vrain	Fort St Vrain, USA	HTGCR	330 MW(e)	ThC_2 + 93% UC_2	1973
THTR	Schmehausen, FRG	HTGCR	300 MW(e)	ThC_2 + UC_2	1977

*Percentages refer to uranium enrichment.

TABLE C2

SUMMARY OF HIGH TEMPERATURE GAS-COOLED REACTORS

Reactor	Country	Criticality	Full Power	Shutdown	Power MW(e)	Power Density	Aim	Achievements
DRAGON	UK	1964	1966	1976?	20 MW(th) No electricity generated	14	To prove helium coolant and coated particle concept. Technical feasibility of HTGCR.	Proved helium and fuel concept. Low primary circuit activity.
AVR	FRG	1966	1967		15	2.2	Prove pebble bed concept. Achieve burn-up of 100 000 MWd/t.	Pebble bed worked and fuel has lasted beyond design limits. Runs at 850°C.
Peach Bottom	USA	1966	1967	1974	40	8.3	Prove HTGCR as a power producer.	Some trouble with first core loading, but good availability of whole plant with second core.
Fort St Vrain	USA	1974			300	6.3	Demonstrate commercial worth of HTGCR.	Non-nuclear problems; 30% full power by end 1981.
THTR	FRG	1983			300	6.0	Demonstrate commercial worth of pebble bed fuelled HTGCR.	

TABLE C3

PARAMETERS OF HIGH TEMPERATURE REACTORS

[after Brown 1969; Dahlberg and Brooks 1974]

	Peach Bottom USA	Fort St Vrain USA	Fulton 1160 USA	DRAGON UK (OECD)	Pebble Bed FRG (AVR : BB/K)	THTR FRG (BB/K)
Power { MW(Th)	115	842	3000	20	49	750
Power { MW(e)	40	330	1160	-	15	300
Efficiency (%)	35	39	38.6		31	40
Gas Temp. { inlet (°C)	340	405	318	350	175	270
Gas Temp. { outlet (°C)	715	780	741	750	850	750
Core { height (m)	2.28	4.75	6.34	1.6	2.5	5.1
Core { diam. (m)	2.79	5.94	8.47	1.1	3.0	5.6
No. of elements	804	1482	3944	259	-	-
No. of columns		246	493		-	-
Elements per column		6	8		-	-
No. of balls	-	-	-	-	95 000	675 000
Ball diam. (cm)	-	-	-	-	6	6
Basic fuel component	Particles in graphite compact within cylindrical purge sleeve	Bonded rods of particles within a hexagonal graphite element	Bonded rods of particles within a hexagonal graphite element	Biso/Triso fuel bonded and in graphite compacts loaded into channel in graphite	Random packed bed of spherical fuel elements Biso/Triso fuel within graphite ball	Random packed bed of spherical fuel elements Biso/Triso fuel within graphite ball

BB/K = Brown-Boveri-Krupp Reactorbau GmbH

TABLE C4

COMPARATIVE FUEL CYCLES

[after Dahlberg and Brooks 1974]

	Peach Bottom	Fort St. Vrain	Fulton
Fuel lifetime at 80% capacity factor, y	3	6	4
Refuelling cycle, y	3	1	1
Fraction of core replaced each cycle	1	$^1/_6$	$^1/_4$
Number of refuelling regions	1	37	85
Average power density, W/cm^3	8.3	6.3	8.4
Carbon/thorium ratio (equilibrium cycle)	310	225	240
Fuel exposure, MWd/tonne heavy elements	73 000	100 000	98 000
Peak fast fluence, 10^{21} nvt	4.5	8	8

APPENDIX D
TABLES OF THORIUM RESOURCES

TABLE D1

COMPOSITION OF MONAZITE CONCENTRATES

[after Garg et al. 1977]

Constituents	India	Brazil	USA*	South Africa (Monazite rock)	Malagasy Republic
ThO_2	8.88	6.5	3.1	5.9	8.75
U_3O_8	0.35	0.17	0.47	0.12	0.41
$(RE)_2O_3$	59.37**	59.2**	40.7**	46.41**	46.2**
Ce_2O_3	28.46	26.8	-	24.9	23.2
P_2O_5	27.03	26.0	19.3	27.0	20.0
Fe_2O_3	0.32	0.51	4.47	4.5	-
TiO_2	0.36	1.75	-	0.42	2.2
SiO_2	1.00	2.2	8.3	3.3	6.7

* Florida sand containing about 70% monazite

** Includes Ce_2O_3

RE = Rare earths

TABLE D2

THORIUM RESOURCES OF THE WESTERN WORLD
(10^3 t of ThO_2)
[after Kasten 1970; Garg et al. 1977]

Country	Reasonably Assured Resources <$22*/kg ThO_2	Estimated Additional Resources <$22*/kg ThO_2
India	363	~550
Canada	90	90
USA	60	300
South Africa	20	80
Egypt	20	320
Brazil†	1	35
Australasia and S.E. Asia	10	
Europe (mainly Denmark)	10	20

*1973 US dollars

†Proved placer reserves are estimated at 15 000 t and total reserves at 182 000 t

TABLE D3

THORIUM RESOURCES IN USA AS A FUNCTION OF RECOVERY COST
[after Kasten 1970]

Recovery Cost (1965 $US/lb oxide)	ThO_2 (thousands of tonnes)*
5 - 10	100 - 800
10 - 30	200 - 1100
30 - 50	3 000 - 11 000
50 - 100	7 000 - 30 000
100 - 500	900 000 - 5 000 000

*Ranges given correspond to reasonably assured resources to estimated possible resources

TABLE D4

SUMMARY OF WORLD MONAZITE PRODUCTION

[after Mineral Yearbook 1968, US Bureau of Mines
Mineral Yearbook 1971, US Bureau of Mines
World Energy Conference 1974]

Name of Continent and Country	Year of Production[1]	Amount	
		Monazite (tonnes)	Thorium[2] (tonnes)
Africa			
Nigeria	1971	91	4.8
Mauritania	1969	105	5.5
Malagasy	1965	1087	57.4
Mozambique	1970	2	0.1
Zaire	1969	178	9.4
Total Africa		1463	77.2
Asia			
Korea, Rep. of	1965	25	1.3
India	1971	4000	211.2
Sri Lanka	1969	56	3.0
Indonesia	1964	140	7.4
Thailand	1971	112	5.9
Malaysia	1968	2143	113.2
Total Asia		6476	342.0
South America			
Brazil	1972	2363	359.0
Oceania			
Australia	1971	4521	228.8
World Total		14823	1007.0

[1]Maximum annual production during period 1964-1972

[2]Based on average ThO_2 content of 6% in monazite that might have been recovered.

TABLE D5

SUMMARY OF THORIUM RESOURCES AND RECENT ANNUAL PRODUCTION
(All values in terms of elemental thorium)

(Source - World Energy Conference 1974)

Name of Continent and Country	Ref. No.	Year of Ref.	Lower Cost Resources					Higher Cost Resources			Recent Annual Production	
			Maximum Cost ($/kg)	Reasonably Assured			Additional Resources (tonnes)	Maximum Cost ($/kg)	Reasonably Assured (tonnes)	Additional Resources (tonnes)		
				Amount (tonnes)	Energy Content, Breeders (millions of GWh)*	Energy Content, Breeders (millions of TJ)†					Year	Amount (tonnes)
Africa												
Malagasy	2	1960						U	1960			
Malawi	2	1959						U		8800		
Egypt	3							U	14700	279800		
South Africa	1	1972						U	20434			0
Other	4							U	~17200	30800		
Total Africa									54294	319400		
Asia												
India	1	1973						U	302370	U		U
Turkey	1	1972						U	3960	U		
Total Asia									305330			
Europe												
Norway	1	1973	22	132000	1890	6804		34		132000		0
USSR	4							U	~80000	~80000		
North America												
United States	1	1973	25	51573	744	2678	267443					U
Canada	1	1972	25	79832	1146	4128	79832				1968	56
Greenland	1	1969						U	26400	U		
Total N. America				131405	1890	6806	347275		26400			
South America												
Brazil	1	1972	25	58388	836	3010	1059080				1972	359
Uruguay	1							U	748	1496		
Total S.America				58388	836	3010			748	1496		
Oceania	1	1973						U	6160	10560		229
Australia												
WORLD TOTAL				321793	4616	16620	1406355		473932	543456		644

U = Unknown 0 = Nil *GWh = gigawatt hours = 1000 MWh = 10^9 watt h †TJ = terajoule = 10^{12} joules

References

1. Data from current survey; see also Tables D2 to D4
2. 'World Power Conference Survey of Energy Resources, 1968'. A. Parker, 1968
3. 'Mineral Facts and Problems, 1970', US Bureau of Mines Bulletin 650 (Resource data only).

Notes: Resources for Africa recoverable at costs up to $22/kg ThO_2 include 91000 metric tons in central and southern Africa (including Malagasy) divided equally between reasonably assured and additional resources.

TABLE D6

GENERAL REPROCESSING METHODS OF THORIUM-URANIUM FUELS

[after Srinivasan et al. 1972]

Type of Fuel	Head End Methods	Purification Process
1. Aluminium clad uranium oxide-*thorium oxide fuels*	Chemical decladding with NaOH and dissolution	
2. Pyrolytic carbon coated particles of thorium oxide-uranium oxide or thorium-uranium carbide	Burn leach process or grind leach process followed by acid leaching	
3. Silicon coated particles - oxides or carbides of thorium and uranium	Grind leach process followed by nitric acid leaching	
4. Zirconium clad carbides or oxides of thorium and uranium	Chop leach process or Zirflex chemical decladding followed by nitric acid dissolution	Acid thorex using 90% tributyl phosphate when thorium and uranium both have to be recovered. Using *di-sec-butyl-phenyl-phosphomate as* solvent or interim-33 process* using 5% tributyl phosphate, when uranium only is recovered.
5. Stainless steel clad oxides or carbides of thorium and *uranium*	Chop leach or sulfex chemical decladding followed by nitric acid dissolution	
6. *Molten fluoride salt of* beryllium, lithium contain-UF_4 and ThF_4	*Closed circuit processing by* reductive extraction with liquid bismuth containing lithium and salt metal transport process aided by fluoride volatility	

*A method to simplify the immediate recovery process and increase the plant capacity. The ^{233}U that builds up from the decay of ^{233}Pa must be recovered after an interim period, hence the name.