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The thorium cycle for fast breeder reactors

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Abstract. The role that could be played by liquid metal-cooled fast breeder reactors (LMFBRs) in the utilization of India's considerable thorium resources is reviewed in this article. Distinct advantages of thorium-based fuels over plutonium-uranium fuels in LMFBRs pertain to a more favourable coolant voiding reactivity coefficient and better fuel element irradiation stability. The poorer breeding capability of thorium-fuelled fast reactors can in principle be overcome by improved core design and development of advanced fuel concepts. The technical feasibility of such advanced thorium fuels and core designs must be established by sustained research and development. It is also necessary to efficiently close the thorium fuel cycle of fast breeder reactors by appropriate development of the fuel reprocessing and refabrication stages. The Fast Breeder Test Reactor (FBTR) at Kalpakkam is expected to be an important tool for development of thorium fuel and fuel cycle technology. A quick look at the economics of the thorium cycle for fast reactors, vis-a-vis the more conventional uranium cycle indicates only a small and acceptable cost disadvantage on account of the need for remote fabrication of recycled thorium fuel.

Keywords. Thorium cycle; fast breeder reactors; nuclear physics features.

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1. Introduction

The fertile isotopes ^{238}U and ^{232}Th are several hundred times more abundant than the only naturally occurring fissile isotope ^{235}U. For complete utilization of the fertile isotopes it is necessary to operate breeder reactors which can transmute the fertile isotopes ^{238}U and ^{232}Th into the respective fissile isotopes ^{239}Pu and ^{233}U at a rate faster than the rate of consumption of fissile material for power production.

India's thorium resources are several times greater than the uranium resources and are sufficient to meet the energy needs in India for several centuries. Since thorium cannot be directly used to fuel a reactor, the Indian nuclear power programme has been started with heavy water-moderated, natural uranium-fuelled reactors (PHWRs) to generate power and also plutonium. The nuclear electric base using PHWRs is expected to reach about 10 GWe by the year 2000 and the readily available natural uranium in the country will be entirely committed to these reactors over their lifetime. The emphasis in the second stage will be on the utilization of plutonium and ^{238}U to rapidly increase the nuclear base to several hundred GWe by means of efficient breeder reactors. Of all the breeder reactor concepts the LMFBR is the most well developed and has been chosen for the second stage of the Indian nuclear power programme. The third

The authors felicitate Prof. D S Kothari on his eightieth birthday and dedicate this paper to him on this occasion.

stage will be to sustain and consolidate the nuclear electric base by means of thorium fuel.

Though the large scale utilization of thorium is expected only in the third stage of the Indian nuclear power programme there is considerable incentive for R&D in the technology of thorium reactors and the associated fuel cycle in order to have a balanced development strategy which will enable optimized retrofitting of the thorium cycle schemes into PHWR and LMFBR systems at the appropriate time. An advantage of early introduction of the thorium cycle and production of ^{233}U is that it leads to a diversification of energy resource bases and allows greater flexibility in the choice of breeder reactor concept and fuel cycle. It must be noted that any fuel cycle development takes a long time and it is necessary to generate essential data and establish semi-industrial experience well in advance of the actual large scale utilization. In the long term it may be necessary to have a symbiotic association of LMFBRs and PHWRs or to even introduce a new reactor type for efficient utilization of the thorium resources. In what follows the feasibility of the thorium cycle in LMFBRs is discussed.

2. Fast reactor fuel cycles

While the initial fissile material for LMFBRs in India will be the plutonium produced from PHWRs, alternate choices for the fertile material and core diluent are depleted uranium or thorium. Once ^{233}U has been produced from thorium a number of options become available for the fissile/fertile combinations to be used in LMFBRs.

Combining ^{233}U with Th or Pu with ^{238}U gives rise to simple cycles where the fertile material produces the same fissile material as present in the initial fuel. On the other hand the use of ^{233}U with ^{238}U as fertile material and Pu with Th as fertile material gives rise to hybrid cycles where the fissile production of one cycle is used to fuel the other. In mixed cycles both ^{238}U and Th are irradiated in the same reactor.

The most well-developed LMFBR fuel cycle at present is that involving mixed oxides of Pu and ^{238}U. Carbides and nitrides are more advanced fuel types being developed for use in future LMFBRs with better breeding performance. For thorium-based fuels in LMFBRs the development of metal alloy fuels appears to be important for good breeding.

3. Nuclear physics features

The average number of neutrons, η, released per neutron absorbed in a fissile nuclide, is the most important parameter determining the breeding potential of that nuclide. For breeding to be possible η must be greater than 2. The values of η for different fissile nuclides are compared in table 1 for neutron spectra typical of a fast reactor and of a heavy water-moderated reactor. As noted from table 1, ^{233}U can be used for breeding both in thermal and in fast reactors, whereas Pu can be used for breeding only in fast reactors. Thus, availability of ^{233}U leads to much greater flexibility in the choice of breeder reactor concepts and fuel cycles. This flexibility is an important reason for the early development of the thorium cycle.

Cross-sections in a typical fast spectrum for the fertile nuclides are compared in table 2. Fast fission of the fertile nuclide contributes indirectly towards the breeding as extra

Table 1. Spectrum averaged η values of fissile nucides.

Nuclide	Fast reactor	Heavy water-moderated reactor
^{233}U	2·31	2·25
^{235}U	1·93	2·01
^{239}Pu	2·49	1·88

Table 2. Average cross-sections of fertile nuclides in fast spectrum.

Nuclide	Capture (barns)	Fission (barns)
^{232}Th	0·20	0·03
^{238}U	0·14	0·10

neutrons are produced without the loss of a fissile nucleus. ^{238}U has considerably higher "fast fission bonus" than Th on account of its lower fast fission threshold and higher fast fission cross-section.

Reliable nuclear cross-section data for ^{232}Th, ^{233}U and of some nuclides like ^{233}Pa are essential for the development of thorium fuelled reactors. While a large number of measurements have been made, there are still discrepancies and gaps in the data so that considerable scope exists for re-evaluation of available data and for making further measurements (Ganesan 1985).

4. In-core performance of thorium-based fuels

A commercial LMFBR is required to operate safely and reliably, produce power economically, and breed fissile material efficiently. Good economic performance with reasonable fuel cycle cost requires LMFBR fuel to attain burn-ups of the order of 100,000 MWD/Te. Good breeding performance means a short doubling time, which is defined as the operating time for the reactor to produce as much fissile material as is normally contained in the reactor (inpile inventory) and in the associated ex-reactor fuel cycle (out-of-pile inventory).

4.1 *Burn-up*

The fuel cycle cost is inversely proportional to the burn-up attainable by the fuel. The doubling time is also decreased for higher burn-ups on account of reduction in the out-of-pile inventory. First generation enriched uranium metal alloy fuelled LMFBRs were not capable of high burn-ups. They were replaced by mixed plutonium-uranium oxide-fuelled LMFBRs which were capable of burn-ups of the order of 50,000 to 100,000 MWD/Te. While both thoria and urania are capable of high burn-ups there is limited irradiation experience with thorium-based ceramic fuels for fast reactors. The bulk of the experience with the fabrication and irradiation performance of $(Th, U)O_2$ fuels has been gained from various thermal reactor programmes and this experience is not directly applicable to potential LMFBR fuels (Rodriguez and Sundaram 1981). Practically, no irradiation data are available for carbides or nitrides of Th, (Th, U) or (Th, Pu).

Thorium metal has a melting point of 1725°C compared to 1132°C of uranium metal. Further, unlike uranium metal, thorium metal has an isotropic crystal structure with no phase transformations from room temperature to $\sim$ 1300°C. Hence, thorium metal has much better irradiation behaviour and dimensional stability than uranium metal and remains a strong candidate for use in future fast reactors. However, much more development is required before the full potential of thorium metal alloy fuels can be realized. An important problem identified in thorium-based metallic fuel is the fuel clad metallurgical interaction leading to interpenetration of fuel and clad by inter-diffusion. The addition of zirconium in the metallic fuel (upto 10%) is required to reduce this interpenetration. In order to effect fission gas release and reduce fuel swelling and fuel clad mechanical interaction low metallic fuel smeared densities of 75% with a sodium bond between fuel and clad is found suitable. Another fuel concept of promise is the use of PuO_2 or $^{233}UO_2$ kernels in a thorium metal matrix. This concept attempts to exploit the better irradiation stability of thorium metal while avoiding the poor irradiation behaviour of plutonium or uranium metal.

4.2 *Doubling time*

The important parameters affecting the doubling time are the breeding ratio and the total fissile inventory. While the former depends on the neutron spectrum and the nuclear properties of the fissile and fertile nuclides the latter depends on the fuel thermal rating and the time lags associated with the ex-reactor fuel cycle processes. Oxide-fuelled cores have the softest neutron spectrum leading to low breeding ratios. The breeding ratio improves considerably for carbide or nitride fuels. Metal-fuelled cores have the hardest neutron spectrum and the highest breeding ratios.

Table 3 summarizes calculated breeding performance for alternate fuel cycles in advanced fast breeders which are expected to be feasible from the year 2000 onwards (INFCE 1980). It is observed that the use of thorium in the blankets of $Pu/^{238}U$ fuelled reactors has little effect on the breeding ratio. However, when thorium is introduced into the core there is substantial decrease in breeding ratio. Though the $^{233}U/Th$ reactor has a long doubling time, the reactors on hybrid cycles Pu/Th or $^{233}U/^{238}U$ have fairly short doubling times, especially those using metallic fuels. Development of such fuel technology appears to be important for the proper utilization of thorium in fast breeders.

4.3 *Reactivity coefficients*

A factor of some concern in the design of LMFBRs is the reactivity change associated with sodium density changes or coolant voiding. This reactivity coefficient is positive in large $Pu/^{238}U$ fuelled LMFBRs. As shown in table 3 this coefficient becomes less positive for thorium-based fuels and is even negative in the case of $^{233}U/Th$ fuelled reactors. This enhanced inherent safety of thorium-fuelled LMFBRs comes about because of the slow variation of the ^{233}U capture to fission ratio with increasing energy as compared to that of ^{239}Pu and because of the fewer fast fissions in ^{232}Th as compared to ^{238}U.

Further, the capture cross-section resonances of Th go up to higher energies than ^{238}U so that fast reactors with thorium-based fuels tend to have a slightly greater negative Doppler coefficient of feedback reactivity than with ^{238}U based fuels.

5. Thorium fuel cycle technology

The following are the recognized stages for a closed fast reactor fuel cycle:

— fabrication of fuel from separated fissile and fertile material and fabrication of the fuel subassemblies;

Table 3. Summary of performance characteristics for alternate fast breeder fuel cycles: 1 GWe generation: 0.75 capacity factor: external cycle time: 2 years: fuel cycle losses: 1 % for uranium and 2 % for thorium (INFCE 1980)

Fuel type	Core material	Blanket material	Cycle fissile inventory (kg)	Net fissile gain (kg/year)			Breeding ratio	System doubling time (years)	Sodium voiding reactivity gain ($)
				^{233}U	Plutonium	Total			
Oxide	$Pu/^{238}U$	^{238}U	6315	0	245	245	1·325	17·8	5·7
	$Pu/^{238}U$	Th	6396	292	−93	199	1·305	22·3	—
	Pu/Th	Th	7317	740	−668	72	1·184	70·8	4·1
	$^{233}U/Th$	Th	6608	43	0	43	1·099	108	−0·05
	$^{233}U/^{238}U$	Th	5799	−301	457	156	1·240	25·9	—
Carbide	$Pu/^{238}U$	^{238}U	5229	0	354	354	1·479	10·2	5·3
	$Pu/^{238}U$	Th	5311	318	−32	286	1·426	12·9	—
	Pu/Th	Th	6150	734	−610	124	1·223	34·5	3·0
	$^{233}U/Th$	Th	5807	58	0	58	1·114	70·0	−1·1
	$^{233}U/^{238}U$	Th	5021	−237	454	217	1·330	16·0	—
Metal	$Pu/^{238}U$	^{238}U	4977	0	412	412	1·582	8·5	8·2
	$Pu/^{238}U$	Th	5080	361	−62	299	1·459	11·8	—
	Pu/Th	Th	6189	774	−594	180	1·301	23·8	4·7
	$^{233}U/Th$	Th	6080	56	0	56	1·115	75·1	−1·4
	$^{233}U/^{238}U$	Th	5134	−210	509	299	1·468	11·9	—

- —transportation of the fuel subassemblies to the reactor;
- —irradiation in the reactor followed by cooling in storage facilities;
- —transportation of cooled, irradiated fuel to the reprocessing plant.
- —reprocessing of the irradiated fuel for separation of the bred fissile material from fertile material and removal of fission products.
- —transportation of the separated fissile and fertile materials to the fuel fabrication plant for refabrication.

Typical time lags associated with the different steps are (INFCE 1980), 120 days for storage and fabrication at the fabrication plant, 210 days for storage at reactor, 130 days for storage and treatment at reprocessing plant and 90 days for various transportations giving a total of 550 days. One of the aims of R&D in fuel cycle technology is to reduce the total out-of-pile time lag to 365 days or less.

Fabrication processes for fresh thorium and its compounds as nuclear fuel are similar to that of uranium and its compounds. However, the build-up of ^{232}U on account of $(n, 2n)$ reactions in thorium and ^{233}U causes complications in the processes for fabrication of recycle fuel. Since the daughter products of ^{232}U are highly gamma-active, remote handling with fabrication processes in shielded enclosures is necessary. This has a significant impact on the process control sample analysis system, the scrap recovery and the off-gas system with consequent economic penalties. The bulk of the fabrication experience with thorium-based fuels in different countries, including India, is for thermal reactors. However, the experience is limited to fuel of low ^{232}U content (< 10 ppm). There is little experience with fabrication of thorium-based fuels for LMFBRs.

The thorex flow sheet is used for reprocessing thorium. India too has developed and utilized the thorex process for separating ^{233}U from irradiated thorium. One of the problem areas is the fact that to dissolve ThO_2, hydrofluoric acid must be added. The HF is very corrosive and special processes to prevent corrosion are necessary. Further, in the solvent extraction step, partitioning the thorium from the uranium is more difficult than with plutonium-uranium. There are also several problems in three way separation process for U-Pu-Th fuel which require considerable research and development.

R&D is directed towards reduction in the costs associated with fuel cycle processes and reduction of the fissile losses to 1%.

6. Utilization of FBTR for thorium fuel development

Fuel elements of large commercial fast power reactors not only have to withstand peak burn-ups of upto 100,000 MWD/Te but also have to attain fast neutron fluence exposures of upto 2×10^{23} n/cm^2 in order to have reasonable fuel cycle costs. Fast neutron irradiation of core structural materials leads to phenomena which are not encountered in thermomechanical treatments, like irradiation enhanced creep, void swelling, production of new phases, segregation/precipitation effects, and helium embrittlement. Research in these areas requires energetic charged particle beams from accelerators as well as high fast neutron fluxes provided by materials testing fast reactors like the fast breeder test reactor at Kalpakkam.

FBTR is a 40 MWT/13 MWe mixed plutonium-uranium carbide fuelled LMFBR which will have a maximum neutron flux of $3{\cdot}6 \times 10^{15}$ n.cm^{-2} sec^{-1}. The neutron spectrum and core temperature conditions make FBTR an ideal tool for irradiation testing and development of fast reactor fuels and core structural materials. Important data to be generated by irradiation testing in FBTR pertain to swelling rates, creep rates and evaluation of physical, chemical and thermo-mechanical properties of candidate fuel and structural materials under fast neutron irradiation at high temperatures in a liquid sodium environment.

The availability of FBTR makes possible the irradiation testing and development of candidate thorium-based fast reactor fuels in a meaningful manner. FBTR will also play an important role in the development of thorium fuel cycle technology. Thorium blanket elements have been fabricated for utilization in FBTR. Flow sheets and processes are being developed for reprocessing of these blankets and recovery of the bred ^{233}U. The recycle of ^{233}U into FBTR core, possibly in the form of test fuel pins, will be an important aspect of thorium fuel cycle technology development in India.

7. LMFBR economics

LMFBR economics not only depends on the costs of the power plant, but also on the costs associated with the whole fuel cycle consisting of reprocessing, refabrication and waste disposal plants. The costs of these plants are very sensitive to the scale of operations and attainment of actual costs under commercial-scale conditions implies the existence of a substantial LMFBR capacity (say $\sim$ 10 GWe). In India, the LMFBR is expected to be commercialized in the beginning of the next century and firm cost data especially on fuel fabrication and fuel reprocessing for plants operating on commercial scale under Indian conditions are not available. However, certain projections can be made based on the experience gained from the construction of FBTR and from the construction and operation of PHWRs in India.

Of the various cost contributions to LMFBR power generation the capital costs and fuel cycle costs are dominant. The capital costs of LMFBRs constructed to date in various countries vary considerably (by over a factor of ten per kWe installed) and cannot be used as a basis for cost projections in India. The capital cost of the 1200 MWe Super Phenix LMFBR in France as well as its electricity generation costs are about a factor two higher than the costs of a similar Pressurised Water Reactor power station (IAEA 1985a). LMFBR R&D in France is actively directed towards reduction in these costs and ultimately when commercial scale operations are established it is expected that LMFBR capital costs will be about 26 % higher and electricity generation costs will be about 13 % higher than for PWRs for constant uranium prices. As uranium prices increase the LMFBR is expected to become fully competitive. It must be noted that PWR power costs in France are already a factor 1·75 less than that from coal-fired stations, so that LMFBR power stations are already nearly competitive with coal fired power stations in France.

In India detailed costing of PHWR power and coal power is available as well as some preliminary estimates of LMFBR power costs. Table 4 gives a simplified overall comparison of projected capital costs and electricity generation costs in the 1990's from coal-fired plants, PHWRs and the proposed Pu/^{238}U ceramic fuelled 500 MWe prototype fast breeder reactor (PFBR). It is estimated (Paranjpe and Sundaram 1985) that the fuel cycle cost for PFBR consisting of Pu inventory charges (3 %), fuel refabrication (7 %), reprocessing (7 %) and waste management (3 %) constitute about

Table 4. Comparative cost ratios in 1990's.

	Capital cost ratio	Electricity generation cost ratio
500 MWe PHWR / 500 MWe coal fired station	1·25	0·8–1·0[(a)]
500 MWe PFBR including development costs / 500 MWe PHWR	1–1·5[(b)]	1·0–1·4[(b)]

(a) depending on the distance from coal pit head; (b) depending on the amount of development needed, and the apportioning of costs of reprocessing plants, waste management plants, heavy water plants etc.

20% of the total electricity generation costs for an average discharge burn-up of 70,000 MWD/Te.

The capital costs of thorium fuelled LMFBRs and the burn-ups achieved are expected to be similar to that of Pu/^{238}U fuelled ones. On account of the magnitude of the other contributions to the fuel cycle cost there is little reduction in fuel cycle cost due to credit for bred fissile material and the influence of the breeding ratio on the fuel cycle cost is small. Thus there is little economic disadvantage of the thorium cycle on account of the lower breeding ratios per se. The increase in fuel cycle cost of thorium cycles comes from the reprocessing and refabrication stages. Since thorium fuels are not being commercially fabricated and reprocessed, there is a large uncertainty in estimates of their reprocessing and refabrication costs. While reprocessing costs can be reasonably expected to be similar for the uranium and thorium cycles, the fuel refabrication costs for the latter cycle will be higher on account of the need for shielding and remotization of the fabrication processes. It has been recommended (IAEA 1985b) that a penalty factor of 1·75 be used for the cost of remote fuel fabrication as compared to non-remote fuel fabrication. Since the fuel fabrication costs constitute only about 7% of the electricity generation costs, the economic penalty for remotization of fabrication processes increases the electricity generation costs of thorium fuelled LMFBRs by only a small amount.

8. Conclusions

^{233}U can be used for breeding both in thermal and in fast reactors whereas Pu can be used for breeding only in fast reactors. Thus early utilization of thorium in LMFBRs and production of ^{233}U can lead to much greater future flexibility in the choice of breeder reactor concepts and fuel cycles for balanced and optimized exploitation of the nuclear resources. To improve the low breeding ratios with thorium-based fuels it is necessary to perform R&D and develop advanced fuels (like metal alloy fuels) and improved core designs. Quantitative irradiation data on thorium based LMFBR fuels are meagre and FBTR can play a role in the generation of this information. Finally, the economics of the thorium cycle in LMFBRs seems to show only a small disadvantage relative to the uranium cycle.

Dedication

We would like to join the others in this issue of *Pramana* to express our appreciation to the contribution to science by Prof. Daulat Singh Kothari who has reached his 80th birthday. We need hardly say that he has played a vital role in the development of Indian science especially after independence. As an acknowledgement of his vision this paper is written looking into India's power needs in the next century.

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