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Utilisation of thorium in reactors

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A B S T R A C T

India's nuclear programme envisages a large-scale utilisation of thorium, as it has limited deposits of uranium but vast deposits of thorium. The large-scale utilisation of thorium requires the adoption of closed fuel cycle. The stable nature of thoria and the radiological issues associated with thoria poses challenges in the adoption of a closed fuel cycle. A thorium fuel based Advanced Heavy Water Reactor (AHWR) is being planned to provide impetus to development of technologies for the closed thorium fuel cycle. Thoria fuel has been loaded in Indian reactors and test irradiations have been carried out with (Th–Pu) MOX fuel. Irradiated thorium assemblies have been reprocessed and the separated ^{233}U fuel has been used for test reactor KAMINI. The paper highlights the Indian experience with the use of thorium and brings out various issues associated with the thorium cycle.

1. Introduction

The worldwide nuclear energy programme is at present predominantly based on the uranium fuel cycle with very little of recycling. The use of uranium in the once through mode will enable us to use only 1% of the available uranium resource. Nuclear energy as a sustainable source of energy will not only require the adoption of a closed fuel cycle policy, but also the use of the other naturally occurring nuclear resource thorium. Thorium is 3–4 times more abundant worldwide than uranium. The use of thorium assumes greater significance in India's context, as our uranium resources are limited and our thorium resources are vast.

Unlike natural uranium which contains fissile isotope ^{235}U, thorium does not contain any fissile isotope. Its utilisation in the initial stage requires the aid of fissile material from the uranium cycle. India has devised a three-stage nuclear power programme to utilise effectively the limited uranium resource and the large thorium resource. The first stage involves utilisation of natural uranium in PHWRs (Pressurised Heavy Water Reactor). The second stage involves the utilisation of plutonium obtained from reprocessing the spent PHWR fuel in fast reactors. The third stage involves the Th–^{233}U cycle based reactor system. The large-scale utilisation of thorium requires the adoption of closed cycle and the (Th–^{233}U) fuel cycle is similar in most aspects to that of (U–Pu) fuel cycle. The development studies in India have focussed on both front-end and back-end activities of thorium in reactors [1].

2. Irradiation performance and reactor operation

Thoria has higher thermal conductivity, lower fission gas release characteristics, better dimensional stability at high burnups, stable stoichiometry and lower thermal expansion coefficient than urania, which shows that its operating performance will be superior to that of urania fuel.

The reactor physics aspects associated with thoria is different from that of urania. This is due to differences in the nuclear cross-sections of the fissile and fertile materials, the differences in fission product and actinide generation. Another important operational difference is due to the formation of ^{233}Pa, which has a half-life of 27 days in the intermediate stage of conversion of ^{232}Th to ^{233}U. During a long shutdown, there is a build-up of ^{233}U by decay of ^{233}Pa. This addition of reactivity has to be accounted for in the reactor operation after a long shutdown and may even require reactor operation below full power to keep fuel powers within limits. Multiple recycling in thoria based reactors will alter the uranium isotopic composition and the design must be able to accommodate the variation, and properly account in the equivalent fissile ^{233}U enrichment. As a typical case, the change in the uranium isotopes with multiple recycling of AHWR fuel is shown in Table 1.

To generate the required reactor physics and performance database for large-scale utilisation of thoria, various experimental campaigns have been carried out and a thoria fuel based AHWR is planned.

2.1. Irradiation of thoria bundles in PHWRs [2]

PHWRs use natural UO_2 as fuel and thoria bundles have been irradiated in these reactors for initial core flux flattening. The

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Table 1
Isotopic changes in Uranium during multiple recycling

	Isotopic composition of discharged Uranium (%)			
	^{233}U	^{234}U	^{235}U	^{236}U
Initial Core of (Th–Pu) MOX	92.8	6.5	0.65	0.04
1st Recycling	85.2	12.5	2.0	0.2
2nd Recycling	79.4	16.7	3.4	0.5

Table 2
Irradiation of thoria bundles in PHWRs

Reactor	Number of bundles
Madras- I	4
Kakrapar-I	35
Kakrapar-II	35
Rajasthan- II	18
Rajasthan -III	35
Kaiga-II	35
Rajasthan-IV	35
Kaiga-I	35

design of these thoria fuel bundles was almost identical to that of the urania fuel bundles to ensure compatibility with fuel handling and thermal hydraulic aspects. Table 2 gives the details of the thoria bundles loaded in PHWRs.

2.2. Irradiation of thoria assemblies in research reactors [2]

Thoria fuel assemblies known as 'J' rods have been irradiated in the reflector region of research reactor CIRUS. Three thoria fuel assemblies were loaded in research reactor Dhruva during its initial days of operation to take care of the excess reactivity of the initial core. These assemblies were similar in design to that of the natural uranium assemblies of the reactor and were successfully irradiated up to 100EFPDs (Effective Full Power Days).

2.3. Irradiation of thoria based MOX fuel [3]

Thoria based MOX fuels and thoria fuel have been irradiated in the Pressurised Water Loop (PWL) of CIRUS reactor. These tests were carried with short-length fuel pins of about 0.5 m under simulated power reactor operating conditions. Table 3 gives details of the irradiation experiments carried in PWL, CIRUS.

2.4. Thorium fuel cycle based AHWR [4,5]

A thorium fuel cycle based AHWR is being planned to provide impetus for large-scale utilisation of thorium. The reactor is a vertical pressure tube type, heavy water moderated, boiling light water cooled reactor generating 300 MWe. The reactor uses mixed oxide of (Th–Pu) and (Th–^{233}U) as fuel. The ^{233}U requirements for the reactor are met by recycling after reprocessing of its spent fuel. The adoption of closed fuel cycle helps in generating a large fraction of energy from thorium and the quantity of plutonium provided as an external fissile feed is kept low. As the reactor depends only on self-generated ^{233}U, it has two types of fuel cluster: the All-Pu MOX fuel cluster and the Composite fuel cluster. The All-Pu MOX fuel cluster will be used in the initial core and for reloads during the transition to equilibrium core. The composite cluster will be used in the equilibrium core and during the gradual transition to equilibrium core. The All-Pu MOX cluster has all the pins with (Th–Pu) MOX as fuel and the Composite cluster has pins of both the fuel types – (Th–^{233}U) MOX and (Th–Pu) MOX.

Unlike uranium-based fuels, very little database exists on reactor physics aspects for the thoria based fuel. A Critical facility is being constructed in BARC for carrying out reactor physics experiments as part of AHWR design and development programme. This facility is a low power research reactor where lattice physics experiments will be carried out for validation of reactor physics design parameters of AHWR. The experiments will be carried out with representative Initial Core configuration and Representative Equilibrium core configuration of AHWR. Fuel irradiation tests are also planned with AHWR type fuel pins in the experimental loops of research reactors CIRUS and Dhruva. The experiments will be carried with fuel pins of both short-length and full-length and over a range of linear heat ratings and burnup. The peak linear heat rating and the maximum discharge burnup planned for these experimental irradiations are 40 kW/m and 40 GWd/t, respectively. The experimental irradiations planned along with the Post Irradiation Examination (PIE) of these fuels will provide an insight into the performance behaviour aspects of thoria based MOX fuel.

Table 3
Details of fuel irradiation tests

Fuel design	Fuel type	Clad type	No. of pins	Peak linear heat rating (kW/m)	Burnup (GWd/t)
BWR	(Th–4%Pu) MOX	Free standing	6	40	18.5
PHWR	(Th–6.75%Pu) MOX	Collapsible	2	42	10.2

3. Fabrication

3.1. Fabrication experience of thoria fuel [6]

High-density thoria pellets have been fabricated for irradiation of fuel bundles/assemblies in PHWRs and research reactors. The fabrication was done by the conventional powder metallurgy technique of cold compaction and high temperature sintering in reducing atmosphere. The production campaign of high-density pellets of PHWR specifications provided an insight into the major hurdles in the large-tonnage scale production of thoria fuel. Moisture absorption on powder due to high surface area, caking of powder during milling, die-wall lubrication during powder compaction, defects in green compacts, attainment of high density of greater than 96% TD, reject recycling, control of aerosol generation were some of the major challenges faced during production campaign.

3.2. Fabrication of thoria based MOX fuel [5,7]

The use of thoria requires addition of fissile materials and these can be in the form of enriched uranium (^{235}U), plutonium or uranium (^{233}U) obtained from reprocessing of thorium fuel. The fabrication aspects of these three MOX fuels differ from one another. The (Th–^{235}U) MOX fuel can be fabricated like the conventional uranium fuel and the (Th–Pu) MOX fuel can be fabricated inside glove box like that of well established (U–Pu) MOX fuels. The fabrication of (Th–^{233}U) MOX fuel however requires a considerable technological development. The fabrication activities right from handling and storage of material to the manufacturing processes used must be able to accommodate the higher level of radiological activity due to the presence of ^{232}U in the recycled urania. The major radiation source is its hard gamma emitting daughter products ^{208}Tl and ^{212}Bi. The fabrication activities have therefore to be structured to ensure low material hold-up and quick recycling of the reprocessed materials to keep the radioactivity dose levels as low as possible and also calls for a high level remotisation and automation. The feasibility of using advanced fabrication techniques like Pellet Impregnation, Advanced Agglomeration, Sol–Gel Microsphere Pelletisation or Vibropac that are more amenable to remotisation are also being explored.

The fuel fabrication for AHWR critical facility will be the first major fabrication exercise of the thoria based MOX fuels. The fuel fabrication for this experimental facility is going to start shortly and the entire fabrication will be completed in the next 2–3 years. This experience will provide the initial feedback on aspects related to fabrication and specification. A total of about 1000 fuel pins with 1.0 ton of (Th–Pu) MOX fuel pellets and 0.5 ton of (Th–^{233}U) MOX fuel pellets will be fabricated during this campaign. Fuel pins of both (Th–^{233}U) MOX and (Th–Pu) MOX fuel will also be fabricated for carrying out in-pile loop irradiation experiments. The experiments will also involve parametric study of various fuel fabrication specifications.

4. Reprocessing [8,9]

THOREX (Thorium–Uranium^{233} Extraction) process has not matured to the same extent as the PUREX (Plutonium–Uranium extraction) largely due to lack of its immediate application in the technologically developed countries. The versatile extractant TBP in hydrocarbon diluent still remains the best choice for the extraction. A pilot facility was installed in the late sixties at Bhabha Atomic Research Centre (BARC), Trombay to develop flow sheet parameters for the 5% TBP flow sheet. This is being used for carrying out various studies on the THOREX process. A facility has also been constructed at IGCAR, primarily for reprocessing fast reactor blanket fuel. In India, reprocessing of thoria fuel has been carried out on aluminium clad ThO_2 fuel irradiated in CIRUS reactor. The reprocessing of these rods which had been irradiated to a level up to 1.2 kg of ^{233}U/t of thorium, and cooled for more than two years was carried out in a pilot scale test facility at BARC and at Indira Gandhi Centre for Atomic Research (IGCAR). Zircaloy clad ThO_2 fuel bundles has been used for initial flux flattening in PHWRs and about a ton of this fuel has already been irradiated. The Zircaloy clad ThO_2 fuel bundles will be reprocessed in Facility for ^{233}U Separation (FUS) being constructed at BARC and the separated ^{233}U will be used for AHWR Critical Facility.

One of the main hurdles in thorium fuel reprocessing is the highly stable nature of thorium, which makes its dissolution more complicated than that of uranium. The problem of thoria dissolution during reprocessing is to a considerable extent overcome by having the fuel pellet density at a slightly lower level in comparison to that of the urania based fuel. Additives like MgO in fuel pellet during fabrication helps in improving the dissolution of thoria pellets and also in lowering of the sintering temperature. Experimental studies have been carried out for MgO additions ranging from 0.5% to 2.5% using HNO_3–HF and 1.5% has been found to be the optimum addition. The dissolution time has reduced almost by an order of magnitude compared to that without any MgO addition. Fig. 1 shows the dissolution characteristics of thoria for various concentrations of MgO in the pellet. When work on THOREX process started in BARC, the basic laboratory data for ^{233}U and thorium were scarce and most of the batch and counter-current extraction and stripping data were generated by in-house studies. As the recovery of U^{233} alone was contemplated during the initial phase of the DAE program, more stress was given to the process using 5% TBP-Shell Sol-T (SST) solvent. Different flowsheets with varying content of TBP in diluent have been developed for the recovery of ^{233}U alone, for both ^{233}U and thorium separately as well as co-processing of ^{233}U and thorium. The flowsheet for recovery of uranium, plutonium and thorium from reprocessing of (Th–Pu) MOX fuel is also being developed. These technological developments will be put to use in the large-scale reprocessing of AHWR fuel.

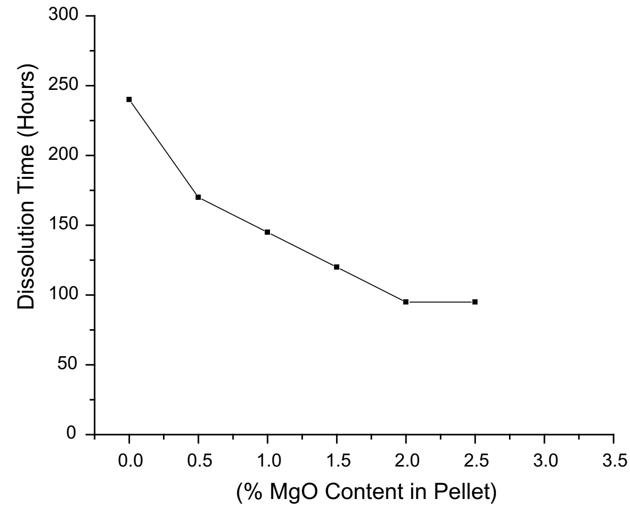

Fig. 1. Thoria dissolution time for various concentration of MgO.

5. Conclusions

The use of thorium in reactors is necessary from long-term objective of sustainability of energy resources. Its use requires reprocessing to separate the fissile ^{233}U for sustained operation using ^{232}Th–^{233}U. The thoria fuel fabrication and reprocessing is complicated by the presence of ^{232}U in ^{233}U and the inert nature of thoria. The thorium fuel cycle technologies that are being developed for AHWR will be useful for the large-scale thorium utilisation in the third stage of Indian nuclear power programme.

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