Plutonium and the Indian atomic energy programme

Baldev Raj *

Indira Gandhi Centre for Atomic Research, Government of India, Department of Atomic Energy, Kalpakkam 603 102, Tamil Nadu, India

ABSTRACT

Plutonium is poised to play a key role in the Indian atomic energy programme, as fast breeder reactors with Pu fuels will be a major source of nuclear energy production in India in the coming decades. Starting from separation of Pu from irradiated PHWR fuel, to fabrication of Pu containing fuels and their utilization in fast reactors, India has mastered various aspects of Pu based fuel cycles. This paper describes the development of technologies for the fuel cycles of oxide, carbide and alloy fuels containing plutonium being pursued in India, and highlights the broad based programme of basic research on Pu science that has provided the confidence for effective utilization of plutonium.

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1. Introduction

Plutonium has a key role to play in the development of atomic energy in India which is based on a three stage programme tailored to suit the available resources of moderate uranium (84 600 t) and vast thorium (225 000 t). Pressurised heavy water reactors (PHWR) form the first phase of the program in which 17 reactors have been installed with a capacity of 4120 MWe and this program is already in the commercial phase. Fast breeder reactors (FBR) form the second stage. By enabling the production of ^{233}U, needed for the thorium based reactors of the third stage, FBRs serve as the vital link between the first and the third stages of Indian nuclear energy road map. Use of plutonium-based fuels in FBRs and breeding plutonium using a closed fuel cycle concept are inevitable for India because of the very limited sources of uranium. Fast breeder test reactor (FBTR) at Kalpakkam uses plutonium rich mixed carbide fuels and the 500 MWe prototype fast breeder reactor (PFBR) under construction will use uranium–plutonium mixed oxide fuels. Future FBRs will be based on alloy fuels containing plutonium. Hence the development of plutonium-based fuels and their fuel cycles is of paramount importance for the development of FBRs which will be discussed in this paper.

2. Historical perspective

In view of the importance of developing the plutonium technology, reprocessing of spent fuels to separate plutonium was established with the commissioning of plutonium plant in Trombay in 1965 which processed the fuels from the research reactors, CIRUS and DHRUVA. Subsequently, the spent fuels from PHWRs were

reprocessed at the plant in Tarapur. Technology for the fabrication of plutonium-based fuels was established in the Radiometallurgy Laboratory of Bhabha Atomic Research Centre (BARC) where the carbide fuels of FBTR are being fabricated. Uranium–plutonium mixed oxide fuels containing 1–4% plutonium oxide used in the boiling water reactors (BWR) at Tarapur are being fabricated in the BARC plant at Tarapur. It is proposed to use thorium–plutonium mixed oxides as fuels in the advanced heavy water reactor (AHWR) which is planned to be constructed.

3. Carbide fuel development

Uranium–plutonium mixed carbides containing 70% (Mark-I) and 55% (Mark-II) PuC along with 5–20% mixed sesqui-carbides are being used as the fuels in FBTR. Mark-I fuel pins have reached a burn-up of 155 GWd/t without any failure. Data on the thermophysical and thermomechanical properties available in the literature were for the mixed carbides containing up to 30% PuC. There were no in-pile or out-of-pile data for the plutonium rich carbides of Mark-I as well as Mark-II fuels. These properties were studied in detail [1–3] using experimental facilities set up at BARC and IGCAR, for the first time. The data thus generated were used as inputs for the design of the carbide fuel elements. Thermal conductivity and the melting temperature are the key parameters for determining the maximum linear heat rating to which the fuel can be subjected to. Restructuring behaviour as well as the transport processes that occur inside the irradiated fuel are highly dependent on the temperature gradients which in turn are determined by the thermal conductivity of the fuel. Incipient melting technique was employed to determine the melting temperature [4] and laser flash technique to determine the thermal diffusivity of the carbide fuel [1]. The thermal behaviour of the fuel was simulated out-of-pile in a set up by electrically heating the centre of

* Tel.: +91 44 27480240; fax: +91 44 27480060.
E-mail address: secdmg@igcar.gov.in

the fuel column whose periphery was cooled by flowing helium gas. The simulation was used to arrive at the maximum linear heat rating that can be achieved without melting [5].

Carbon potential of the fuel is an important thermochemical property, since it is the driving force for any fuel–clad chemical interaction (FCCI) which could severely affect the integrity of the clad. The carbon potentials of Mark-I as well as Mark-II fuel compositions were measured using two techniques, namely, methane–hydrogen equilibration technique [6] and the isopiestic technique [7]. The measured carbon potentials of Mark-I and Mark-II fuel compositions and that of the clad indicated that carburization of the clad will not occur. These plutonium rich fuels are expected to be more prone to gas phase carburization of the clad by carbon monoxide due to the higher solubility oxygen compared to uranium rich carbide fuels. However, the measured carbon monoxide pressures indicated that they were too low to cause gas phase carburization. Besides these measurements, modeling activities were carried out to provide a thorough understanding of the chemistry of the carbide fuel and its thermochemical behaviour [8,9]. Loss of plutonium by volatilization during the fabrication of the carbide fuel by carbothermic reduction route and the final oxygen content of the fuel are highly dependent on the equilibrium pressures of CO, U(g) and Pu(g) over the Mark-I and Mark-II fuel compositions. Thermodynamic modeling of the U–Pu–C–N–O system was used to calculate the equilibrium pressures as a function of composition and to arrive at the optimum oxygen content to ensure minimum Pu loss by volatilisation. Capsule experiments established the compatibility of the fuel with sodium as well as stainless steel clad [10].

3.1. Carbide fuel fabrication

Carbides being highly pyrophoric and more prone to hydrolysis compared to oxide fuels are more challenging to fabricate. Stringent control of carbon stoichiometry is also needed as the carbon to metal ratio tends to decrease from centre to periphery as well as with the burn-up. It results in the formation of more noble metals at the fuel–clad interface and bonding of the fuel and cladding causing fuel–clad mechanical interaction. The specifications for the sesquicarbide, oxygen and nitrogen contents as well as the levels of impurities were arrived at taking these into consideration. After very systematic studies, the flow sheet for the carbide fuel fabrication was developed at the Radiometallurgy Laboratory of BARC which is shown in Fig. 1. Stringent quality control procedures and recycling the rejects produced during fabrication were ensured.

3.2. Post-irradiation examination of the carbide fuel

For enhancing the burn-up and linear heat rating beyond the conservative limits, performance of the FBTR fuel was evaluated through post-irradiation examinations (PIE). A series of hot cells have been established at IGCAR to carry out PIE of mixed carbide fuel of FBTR. Hot cells are provided with state of art fuel transfer systems, high purity inert atmosphere ventilation system for handling pyrophoric carbide fuel, remote handling devices and in-cell equipment for non-destructive and destructive examinations. The techniques and facilities have been augmented in tune with the increasing demands on the characterization of materials used for the fast reactor programme. PIE of FBTR fuel was carried out starting with experimental fuel pins for beginning of life performance evaluation and thereafter on fuel subassemblies at different burn-ups of 25, 50, 100 and 155 GWd/t.

Axial and radial fuel swelling was measured after different burn-ups using non-destructive and destructive techniques. Percentage increase in fuel stack length, pellet-to-pellet gap and pel-

let-to-clad gap were estimated using X-radiography and neutron radiography. Evaluation of the radiographs of the fuel pins after 25 GWd/t and 50 GWd/t revealed the presence of pellet-to-pellet gaps and pellet-to-clad gaps at the end of fuel columns. In the case of fuel pins of 100 GWd/t burn-up, the pellet-to-pellet gap and pellet-to-clad gaps are not observed at the centre of the fuel column. However, pellet-to-pellet gap was still seen in the end of the fuel column. The maximum increase in stack length was observed to be 2.61%. Radiography of 155 GWd/t fuel pins also revealed pellet-to-pellet gap at the end of the column. The increase in stack length varied from 2.7% to 3.7%. Maximum fission gas release estimated for 155 GWd/t burn-up fuel pins was 16% and the corresponding internal pressure in the fuel pin was measured to be 2.1 MPa. Xe/Kr ratio was estimated to be around 13. The trend in the fission gas release [11] in the fuel pins as a function of burn-up follows the trend in swelling behaviour.

Metallographic examination of the fuel was carried out at various burn-ups up to 155 GWd/t. Fig. 2 shows the photomosaic of fuel cross sections at the centre and end of the fuel column after 155 GWd/t burn-up. The observed circumferential cracks are due to the friction between the fuel and clad and the differential thermal expansion/contraction during the cooling down of the fuel under shutdown condition. A distinct zone devoid of fuel porosities was observed near the outer diameter of the fuel. The porosity exhaustion is attributed to the creep of the fuel due to FCMI. Micrograph of the fuel cross section at the end of the fuel column also reveals circumferential cracking and complete closure of the gap indicating complete closure of the fuel clad gap along the entire fuel column. However the dense, pore free zone similar to those observed at the centre is not noticed.

4. Fast reactor fuel reprocessing

The carbide fuel pins discharged from FBTR that have been irradiated up to 25, 50 and 100 GWd/t have been successfully reprocessed in the pilot plant CORAL (compact reprocessing facility for advanced fuels in lead cells) for the first time in the world. The flow sheet for carbide fuel reprocessing has been developed based on the ongoing, comprehensive Research and Development programme.

PUREX process, using tri-*n*-butyl phosphate (TBP) as the extractant is being used for reprocessing the carbide fuels. TBP has been used successfully as the extractant for the processing of spent thermal reactor fuels worldwide because of its excellent extraction behaviour for U(VI). However, it has limitations for fast reactor fuel reprocessing due to third phase formation in the extraction of tetravalent metal ions [3], aqueous solubility, chemical and radiation degradation, etc. Systematic studies for the development of alternate extractants which do not have the limitations of TBP but at the same time retain its advantages have been carried out at IGCAR. Several trialkyl phosphates such as tri-isobutyl phosphate (TiBP), tri-sec butyl phosphate (TsBP), tri-*n*-amyl phosphate (TAP), tri-isoamyl phosphate (TiAP), etc. were synthesized at IGCAR and their efficacy for actinide separations has been evaluated [12,13]. Batch extraction studies revealed that the enhancement of the extraction of U(VI) and Pu(IV) by trialkyl phosphates with increase in the alkyl chain length is very small whereas introduction of branching on the first carbon atom of the alkyl groups increased the extraction of U(VI).

Detailed studies have been carried out on the various parameters that influence third phase formation. Some of the important results are: (a) 1.1 M TAP/*n*-dodecane has high capacity to load Pu(IV) without third phase formation under normal extraction conditions as compared to 1.1 M TBP/*n*-dodecane, (b) tri-cycloalkyl phosphates are highly prone to third phase formation making them unsuitable for solvent extraction applications, (c) for a given
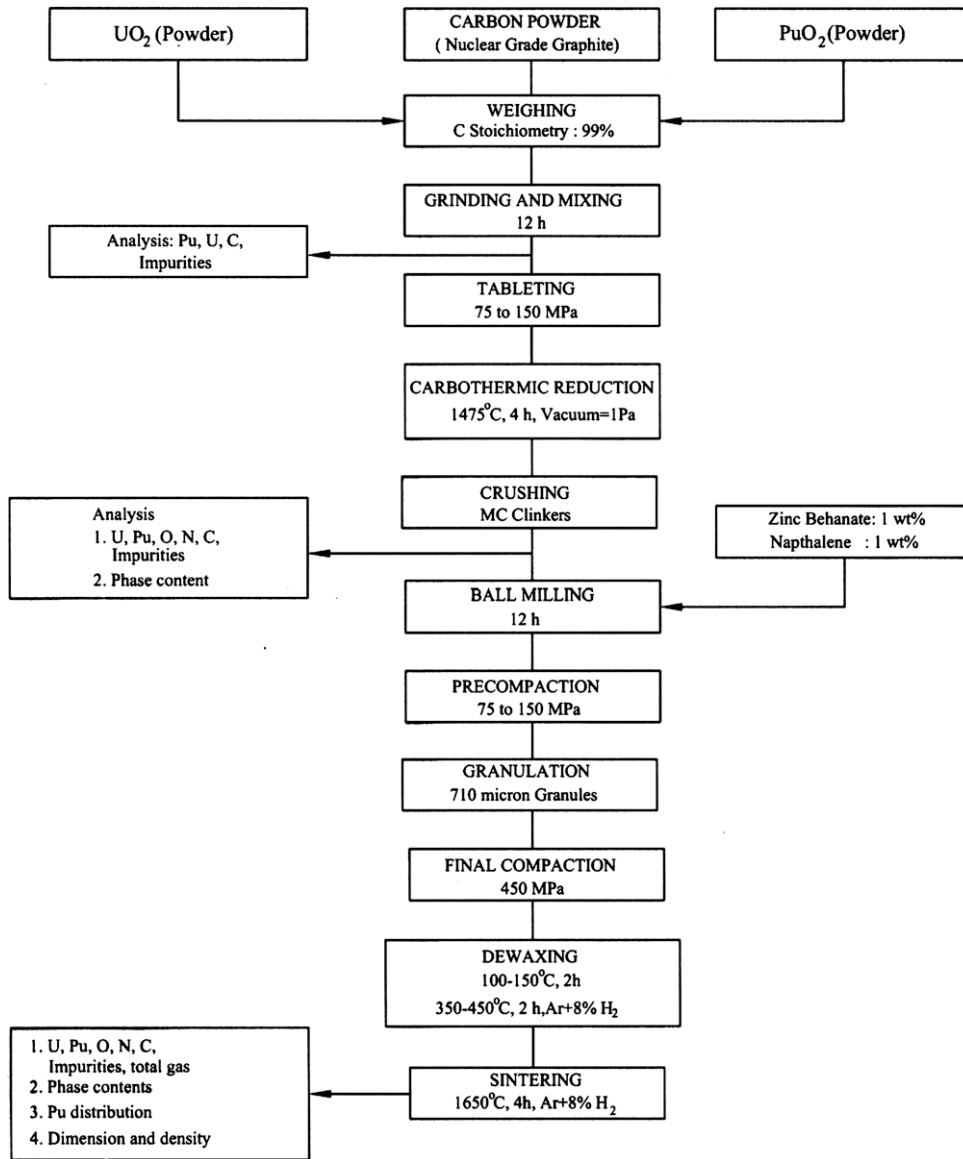
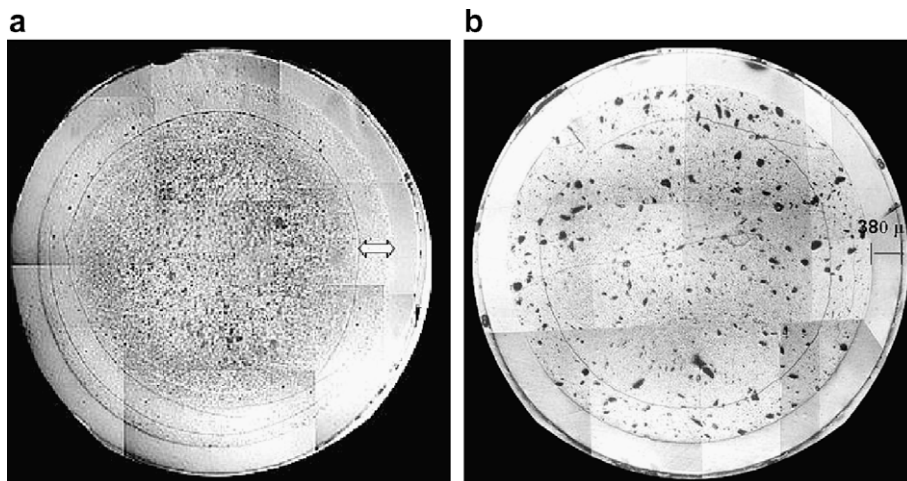**Fig. 1.** Flow sheet used for the fabrication of mixed carbide fuels of FBTR.

Fig. 2. Photomosaics of fuel pin cross sections at (a) the centre and (b) end of the of fuel column after 155 GWd/t burn-up.

phosphate, the limiting organic concentration (LOC) decreases with increase in the carbon chain length of the diluent and (d) LOC decreases with increase in the equilibrium aqueous phase acidity in the extraction of Th(IV), whereas initially it decreases and beyond 2 M HNO$_3$, LOC increases with increase in the equilibrium aqueous phase acidity in the extraction of Pu(IV) by TBP. Long chain monamides are another set of alternate extractants which have also been studied. The amides especially have the advantages of simple synthesis, complete incinerability, innocuous degradation products and low aqueous solubility.

Dihexyloctanamide (DHOA), another completely incinerable amide is considered a promising candidate for the reprocessing of spent fuels in view of its (a) high decontamination factor (DF) values for U/Pu over fission products, (b) ease of stripping of U/Pu and (c) reasonably high U/Pu loading capacity. The radiolyic degradation products of DHOA have been identified, using gas chromatographic studies, as caprylic acid, dihexylamine and dihexylketone, which could be easily washed out. Mixer settler studies were carried out to evaluate the performance of DHOA vis-a-vis TBP using 260 g/l U at 3 M HNO$_3$ as feed solution. Batch process studies using TBP and DHOA were carried out at BARC to evaluate their potentials for co-processing of U and Pu. In view of the lower D_{Pu} values obtained for DHOA at lower acidity (0.5 M HNO$_3$) compared to that for TBP, DHOA promises to be a better candidate solvent for co-processing.

Modeling the PUREX process for the development of codes has been an important R&D activity in fuel reprocessing. A number of codes have been evolved to model the solvent extraction of uranium, plutonium, nitric acid, etc. as well as to predict third phase formation during extraction. SIMPSEX is a code developed in-house which has been validated by experiments. Flow sheets to be used in future commercial scale reprocessing plants would aim at co-processing of U and Pu rather than separation of U and Pu. SIMPSEX code has been used to propose flow sheets suitable for co-processing of uranium and plutonium.

Density of the solution is a very important parameter for solvent extraction operations and is used in the design of settlers (decanters) for separation of organic and aqueous phases, remote operation and nuclear material accountability besides prediction of H/fissile atom ratios in the criticality calculations involving solutions containing plutonium. Based on the critical review of literature data, a new form of density equation, which is simpler and more accurate than many empirical extensions has been developed [14,15] which has been used by other countries [16] as well, for calculation.

Aceto-hydroxamic acid (AHA) is a novel reductant having the potential to replace the conventional reductants such as ferrous sulphamate, uranous nitrate or hydroxylamine nitrate (HAN). Studies have been carried out at IGCAR on the effect of AHA on the distribution coefficient of Pu(IV) in 30% TBP and the results obtained by modeling have thus been validated. Studies have been carried out at BARC on the evaluation of organic soluble *tert*-butyl hydroquinone (TBH) as reductant for plutonium in uranium purification cycle.

These studies on PUREX process are not only applicable to carbide fuel reprocessing, but the results will serve as valuable inputs for implementation reprocessing plants of the mixed oxide fuels of PFBR .

5. Oxide fuel development

The 500 MWe PFBR under construction at Kalpakkam and to be commissioned in 2010 will use uranium–plutonium mixed oxide containing 21% PuO$_2$ as fuel in the inner core and that with 28% PuO$_2$ in the outer core. The thermophysical and thermochemical
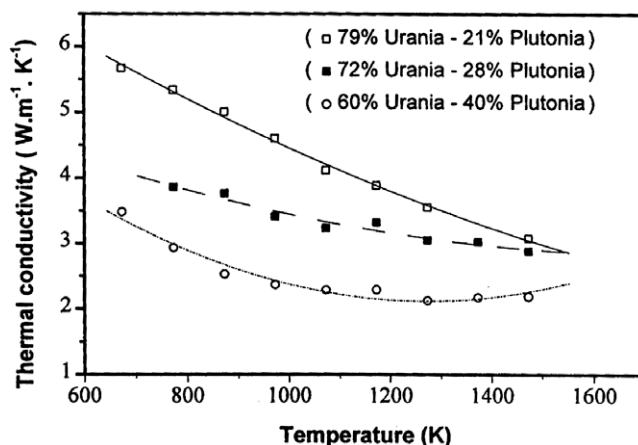

Fig. 3. Thermal conductivity of the uranium–plutonium mixed oxides.

properties of these fuel compositions such as heat capacity, thermal conductivity, oxygen potentials have been measured at BARC and IGCAR to establish a comprehensive data base. The thermal conductivity data of these fuels along with that of the mixed oxide containing 40% PuO$_2$ are shown in Fig. 3. Studies have also been carried out at BARC, using capsule experiments, on the compatibility of the mixed oxide fuel with sodium coolant in which no detectable reaction product was formed, though thermodynamically the reaction is feasible.

The mixed oxide fuel pellets of PFBR will be of annular shape. The fabrication flow sheet has been developed at advanced fuel fabrication facility (AFFF), Tarapur. A subassembly of 37 test fuel pins with mixed oxide fuel containing 27% PuO$_2$ and the rest UO$_2$ enriched with ^{233}U up to 53.5 at.% to ensure a linear power of 450 W/cm is undergoing irradiation in FBTR. It is performing well and has already achieved a burn-up of 80 GWd/t.

It is proposed to use subassemblies with uranium–plutonium mixed oxide fuels containing 44% PuO$_2$ in FBTR for enhancing the operating power by expanding the core. The choice of mixed oxide over mixed carbide fuel for the additional subassemblies stems from the need to gain experience with high Pu content oxide fuels for actinide burning, for validating the flow sheets for reprocessing of oxide fuels proposed through modeling and for fine tuning process flow sheets and nuclear material accounting protocols. Eight subassemblies of this mixed oxide fuel are undergoing irradiation in FBTR. It is known that higher plutonium content results in lower thermal conductivity and also increases concerns regarding fuel–coolant compatibility. Thermophysical properties of this fuel such as heat capacity and thermal conductivity have been measured. Though the thermal conductivity of the fuel is lower than those of PFBR fuels, the annular form of the pellet results in reduction of the centreline temperature and fission gas release. Out of pile compatibility studies carried out at BARC have shown that after 140 h of equilibration with sodium at 973 K, the uranium–plutonium mixed oxide pellet containing 44% PuO$_2$ has retained integrity with acceptable rates of reaction between the fuel and sodium.

6. Need for metallic fuels in future FBRs

Compared to mixed oxide fuel, carbide fuel gives higher fuel breeding ratio (BR). For a 500 MWe reactor, a more optimized design than that of PFBR could result in a BR of 1.09 and a simple fuel doubling time of 40 years. With carbide fuel, the BR can be improved to 1.19 and doubling time reduced to 20 years. Quantum increase in BR is achieved with metallic fuels. U–Pu–Zr fuels with varying Zr content have been studied in IGCAR [17]. It has been
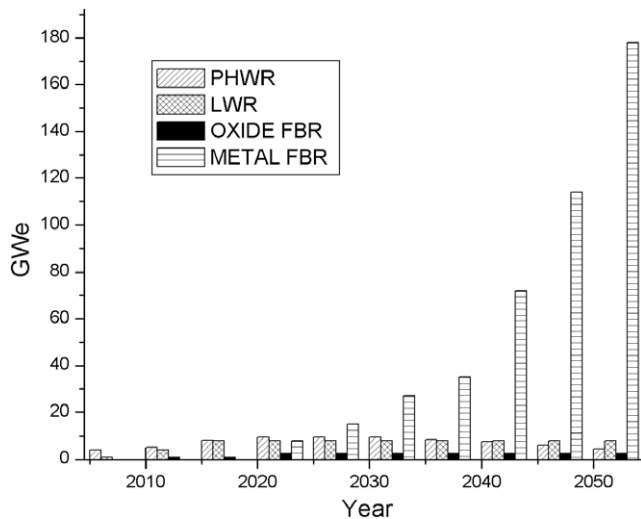
Fig. 4. Possible nuclear power growth scenario in India.

found that the BR increases with reduced Zr content. Thus a U–Pu fuel with 150 μ Zr liner on inner clad (to prevent fuel–clad interaction) is being studied for advanced FBR in India. The BR is calculated to be 1.45 with a fuel doubling time of 9 years. The internal BR of such a core is above unity, eliminating the excess reactivity requirement for core burn-up and the corresponding absorber rod worth requirement.

It has been estimated that with Pu reprocessed from PHWR used as input to the closed FBR cycle, rapid growth of power from FBR is possible beyond 2020 in India. Considering fissile and fertile material balances, the projected contribution from metallic fuel FBR is about 180 GWe beyond 2050 (Fig. 4). The remaining PHWR, imported LWR or MOX-FBR are expected to make only minor contributions.

7. Studies for metallic fuel development

A very rapid growth rate of 100-fold increase in the next 50 years has been targeted for nuclear energy in India for which metallic fuel will be introduced in FBRs after 2020. The fuel will either be sodium bonded ternary U–19Pu–6Zr alloy or mechanically bonded binary U–15Pu alloy with a Zr liner between the fuel and the clad. The decision on the choice of the fuel will be based on test fuel irradiations in FBTR, subsequent PIE results, modeling studies and closing the fuel cycle through pyrometallurgical route. Development activities for the injection casting technology for making the fuel alloy rod and sodium bonding technology for fabrication of fuel pins with ternary fuel as well as co-extrusion of alloy with Zr liner and clad tube for the pin with binary fuel are being pursued. Test fuel pins with metallic fuels will be introduced in FBTR by December 2009.

Development studies for the development of the molten salt electrorefining process for reprocessing spent metal fuels were initiated with the setting up of a laboratory scale facility comprising a train of glove boxes having high purity argon atmosphere. After the studies on all the unit operations of the pyroprocess flow sheet using uranium alloys, studies on plutonium-based alloys have been taken up. In view of the need to generate experience in bulk processing, an engineering scale facility for the demonstration of the process in kg scale has been set up which will be commissioned shortly. This facility will also enable demonstration of remotisation of the process. Thermochemical modeling of the molten salt electrotransport in the electrorefining cell has been carried out with the development of our own code PRAGAMAN [18].

8. Minor actinide (MA) incineration in Pu fuelled FBR

Pu fuelled FBR can be used as burner for MA produced in PHWR in India. The composition of MA discharged from PHWR is dominated by ^{237}Np and Am with small amount of Cm. A 540 MWe PHWR produces 112 kg of MA in its life time. This MA can be partitioned and incinerated in a FBR.

The effectiveness of a MOX fuelled PFBR core with a modified design for achieving significant MA incineration has been evaluated by calculation. In this design, the depleted UO_2 radial blankets are replaced with ThO_2 blankets, since there is no MA production in ThO_2. However, the axial blanket could either be depleted UO_2 or ThO_2. The MA inventory introduced in the initial core is considered to be 5% of fuel (∼400 kg) mixed uniformly. Studies carried out under an IAEA-CRP on advanced reactor technology options for effective MA incineration have shown that this is the amount of MA which can be introduced in existing FBRs keeping the transient behaviour well within allowed safety limits. But, it is possible to consider higher concentration of MA with adequate core design changes.

Our evaluation shows that the changes in core design parameters like fuel enrichment, delayed neutron fraction, Doppler and sodium void reactivity coefficients compared to that of PFBR are found to be acceptable and will not affect the safety of the burner reactor.

In the equilibrium cycle, the MA in core is found to reduce by 8.5% during the cycle. Thus, the amount of MA incinerated per cycle is 32 kg which is about one third of the life-time MA of PHWR (540 MWe). There is Pu gain in axial blanket and ^{233}U gain in radial blanket with negligible MA production in these. ^{233}U production can be augmented by using ThO_2 in the axial blanket also.

9. Sol–gel based fuel fabrication

Currently used fuel fabrication flow sheets are based on powder-pellet route involving large number of mechanical steps that are not conducive for remotisation. On the contrary, sol–gel-based fuel fabrication methods are more suitable for remotisation, since either the solutions or fluid like microspheres are handled here. Other advantages include absence of powder handling leading to less man-rem problems and the potential for integration with reprocessing plant for elimination of reconversion step. Sol–gel technology based on internal gelation has been developed at BARC for urania, thoria, and (U,Pu) mixed oxide with PuO_2 content up to 15%. Development of sol–gel process for mixed oxide fuel with higher Pu contents for FBRs is carried out at Kalpakkam jointly by BARC and IGCAR. A laboratory scale facility for test fuel pin fabrication based on sol–gel technology has been set up and the performance of sphere-pac test fuel pins produced in this facility will be evaluated by irradiation in FBTR.

10. Conclusion

Plutonium is a vital element for the Indian nuclear energy programme without which the vast resources of thorium cannot be exploited. The use of high Pu content carbide fuel in FBTR has posed a number of challenges, but at the same time, has enabled the development of facilities and expertise on a number of aspects of plutonium science and technology. This augurs well for the Indian nuclear power programme which in future would involve plutonium containing fuels of various types. The properties, behaviour and processing of fuels with high plutonium content also have unique features that have made scientific research in this area exciting and rewarding.

Technologies for the fuel cycles of various types of plutonium-based fuels, namely, oxide, carbide and metal, are being developed for deployment in India. A well conceived road map for the development of these technologies has been drawn and is being implemented using the versatile expertise, experimental facilities and large scale collaborations with research, academic and industrial organizations. Specials emphasis on human resources through extensive and quality training of technicians, young scientists and engineers in all aspects of reactors and fuel cycles enables us to have an effective knowledge management and mentoring future generations for mega scaling up of nuclear energy with closed fuel cycles.

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