

STATUS OF PARTITIONING AND TRANSMUTATION IN INDIA: RESEARCH, DEVELOPMENT AND TECHNOLOGY

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Abstract

A programme is in place to recover the minor actinides from the high active waste solutions of the reprocessing operations and burn them in the fast reactors as well as Accelerator Driven Subcritical Reactor Systems(ADS). Recovery flowsheets based on indigenously synthesised Octyl Phenyl N,N'-diisobutyl carbamoyl methyl phosphine oxide (O Φ CMPO), malonamides and diglycol amides are being developed. A facility for production of test fuel pins incorporating minor actinides is being set up at IGCAR. A computational benchmark on effective incineration of minor actinides has been setup for analysis in IGCAR, as a part of work related to IAEA coordinated research programme. A 40 MWth fast breeder test reactor with unique (U,Pu)C fuel has been operating and the construction of a 500 MWe prototype breeder reactor has begun. Actions have been initiated for the development of ADS including high power LINACs and cyclotrons. This paper reports the salient features of the efforts being undertaken to achieve these targets.

Introduction

The success of the nuclear power programme of any country is influenced a great deal by the nuclear waste management policies of that country. At present almost half of the spent nuclear fuel generated in the world is disposed in once through mode and the remaining half is reprocessed. The disposal of both the spent fuel and the high level waste after reprocessing has been a matter of great environmental concern [1]. At present the countries which operate nuclear power stations to produce electricity are planning one of the two options of nuclear waste disposal, viz., 1) direct geological disposal of the spent fuel after sufficient cooling, 2) vitrification of the high-level waste generated during reprocessing operations followed by its interim storage in engineered facilities awaiting long term geological disposal. Partitioning the minor actinides and heat generating isotopes will reduce the load on vitrified borosilicate matrix destined for deep geological disposal. The approach of partitioning and transmutation aims at converting most of the long-lived problematic isotopes into either short lived or stable nuclides. A successful implementation of this approach will drastically reduce the stringent surveillance requirements of geological repository and meet many of the environmental concerns. Closing of fuel cycle is the central element of the Indian strategy for effective utilization of its uranium and thorium resources. Sustainability of the nuclear power programme and minimization of the alpha waste (which would demand deep geological disposal) are the important objectives of R&D in reprocessing and waste management in India. A programme has been taken up to recover the minor actinides from the high active waste solutions generated in reprocessing. Separation processes based on O Φ CMPO, DMDBDTMA and TODGA amides are under development in Bhabha Atomic Research Centre (BARC) as well as in Indira Gandhi Centre for Atomic Research (IGCAR). A sustained programme of laboratory scale testing of these extractants using simulated as well as actual waste solutions has been pursued at these Centres. Indigenous, commercial synthesis of O Φ CMPO and other extractants has been realized. It is envisaged that the forthcoming reprocessing plants for thermal as well as fast reactors would incorporate a scheme for minor actinide recovery.

Transmutation of minor actinides and utilization of the vast thorium resources available in India are planned with the aid of fast reactors and ADS. A facility for production of test fuel pins incorporating minor actinides is being set up at IGCAR. A computational benchmark on effective incineration of radioactive waste has been setup for analysis in IGCAR, as a part of work related to IAEA coordinated research programme. The benchmark is of a critical FBR of 500 MWe size. The core contains PuO₂-UO₂ fuel with ThO₂ as radial blanket. The MAs considered are ²³⁷Np, ²⁴¹Am and ²⁴³Am. The effects of MAs on core parameters especially those related to safety are studied.

A road map for the development of ADS has been drawn up and design and development activities related to high power LINACs, cyclotrons and other technologies are being initiated. A 30 mA, 10 MeV proton LINAC injector is being built at BARC, a high current low energy cyclotron injector is being designed at VECC, Kolkata and a spallation neutron source is being planned at Centre for Advanced Technology (CAT), Indore. An experimental development programme has been initiated for utilizing the attractive nuclear and physical properties of the heavy elements lead and bismuth in a spallation target. Conceptual reactor physics and system studies are being carried out for innovative possibilities with the ADS such as one-way coupled sub-critical reactor systems.

The paper presents salient features of our above activities, relating to P&T research and development.

R&D on Minor actinide recovery

The high level aqueous waste from reprocessing typically contains 3-6M nitric acid which prohibits the use of the conventional tri butyl phosphate and calls for special extractants which can extract the trivalent lanthanides and actinides. Countries such as U. S. A., France, Japan and China are among the nations pursuing diverse R&D programmes employing different extractants viz. dihexyl-N,N-diethylcarbamoylmethyl phosphonates (DHDECMP), O Φ CMPO, diphenyl CMPO, N,N',N,N'-dimethyldibutyltetradecyl malonamide (DMDBTDMA), trialkyl phosphine oxide (TRPO), and diisodecyl phosphoric acid (DIDPA) and N,N,N',N'-tetraoctyl diglycolamide (TODGA).

In the TRUEX process developed at ANL (USA), uses O Φ CMPO in the form of a mixture of 0.2M CMPO + 1.2M TBP. The addition of TBP hinders third phase formation, contributes to better acid dependencies of $D_{Am(III)}$ and also reduces hydrolytic and radiolytic degradation of O Φ CMPO[1]. The TRUEX process is applicable to HLW solutions in the acidity range (0.7 –5M HNO₃). Several successful laboratory scale campaigns have been carried out to demonstrate its applicability for HLW solution of different origins. At BARC, extensive studies have been conducted to get basic distribution data of MAs, fission products and corrosion products[2]. Also several batch and counter-current mixer-settler runs have been carried out to evolve a suitable flowsheet for the recovery of actinides from a real Sulphate Bearing High Level Waste solution (SBHLW) and two synthetic solutions with compositions corresponding to high level waste from PHWR and FBR[3-9]. In all the cases, the HLW raffinate leaving the extraction section showed the alpha activity near background level. Quantitative extraction (>99%) of lanthanides was also observed. Oxidation state specific stripping of actinides from the loaded solvent has been proposed in three steps: 0.04M HNO₃ (for trivalent actinides), dilute HNO₃-HF mixture or oxalic acid (for tetravalent ions) and 0.25M Na₂CO₃ for uranium recovery. Major drawbacks of the TRUEX process are (i) use of large concentration of TBP as solvent modifier; (ii) inefficient stripping of the metal ions, and (iii) necessity of solvent cleanup using Na₂CO₃.

DIAMEX process developed by CEA, France [10] uses completely incinerable extractants comprising essentially atmospheric elements such as C, H, N and O. The use of amide has the advantages of low secondary waste generation, ease of synthesis and elimination of the uranium removal prior to MA recovery. In BARC a number of pentaalkyl malonamides, have been synthesised and evaluated for the recovery of Am(III) from nitric acid medium [11]. Of these extractants, DMDBTDMA was found to be the best candidate with respect to Am(III) extraction and third phase formation.

TODGA has also been identified as one of the most powerful extractants for the partitioning of trivalent actinides from HLW solutions by scientists at JAERI. Distribution behaviour of various metal ions viz. Am(III), U(VI), Pu(IV), Fe(III), Eu(III), Sr(II) and Cs(I) as a function of acidity in the absence as well as in the presence of Simulated High Level Waste (SHLW) using 0.1 M TODGA + 0.5 M DHOA solution in *n*-dodecane as the extractant has been evaluated in BARC. Interestingly, relatively higher extraction of Sr(II) is observed compared to other extractants proposed for actinide-partitioning.

DIDPA process was also developed at JAERI, using a mixture of 0.5M DIDPA and 0.1M TBP [12]. However, this process has the drawbacks such as, (i) poor extraction kinetics, and (ii) the necessity to adjust acidity $\leq 0.5M$ HNO₃. It cannot be applied to HLW without denitration or neutralisation.

TRPO process developed in China employs a mixture of trialkyl phosphine oxides containing alkyl groups with carbon number ranging from 6-8, in an aliphatic hydrocarbon as the solvent. Metal ion extraction is high at low acidity and vice versa, which makes it essential the neutralisation or dilution of HLW. TRPO process has been extensively tested for partitioning of minor actinides from low acidic HLW in BARC[13].

Third phase formation has been a major problem with all the extractants employed for MA recovery. With the expertise already available in this field, in IGCAR, efforts have been made to reduce this problem by replacing the TBP with tri n-amyyl phosphate(TAP) in the TRUEX solvent to permit higher loadings without forming the third phase[14]. Another way to overcome the problem is by using extraction chromatography mode by impregnating the CMPO on polymeric adsorbent substrates. Experiments have also been carried out at IGCAR to identify the best substrate with respect to kinetics of uptake. A hot-cell facility is being commissioned for developing and testing a flowsheet for the recovery of MA from HLW generated from the Fast Breeder Test Reactor (FBTR) spent fuel reprocessing operations. This facility will deploy a 16 stage novel ejector mixer-settler and will have on-line assay features.

Efforts are also underway in both BARC and IGCAR for separating the MAs from the bulk of trivalent lanthanides. The processes proposed envisage employing conventional TALSPEAK process or nitrogen/sulphur ligand based extractants.

Efforts on transmutation

In Fast Breeder Test Reactor (FBTR) and Prototype Fast Breeder Reactor (PFBR)

India operates a 40 MWth, liquid sodium cooled, (U,Pu)C fuelled FBTR at the Indira Gandhi Centre for Atomic Research (IGCAR), Kalpakkam. The reactor achieved first criticality in October 1985 and the unique carbide fuel of the composition $(U_{0.3}Pu_{0.7})C$ used for the first time in world in this reactor has seen a burn up of 120,000 MWd/T without any failure far exceeding the design value of 25,000MWd/T. This reactor is being used as a self-driven fuel irradiation test facility. Irradiation of the first U-Pu mixed oxide (MOX) test subassembly for the 500MWe PFBR has been started for performance assessment. Further $(U_{0.55}Pu_{0.45})O_2$ MOX subassemblies will be gradually introduced in FBTR to get irradiation data on fast reactor fuels and also to enhance the experimental capabilities for minor actinide burning in the fast spectrum. The operating experience of FBTR is also being extensively utilised for the design of PFBR.

A 500 MWe sodium cooled pool type prototype fast breeder reactor(PFBR), with $(U,Pu)O_2$ fuel is also under construction and will start operation by 2010. This reactor has two secondary sodium loops. The purpose of the reactor is to demonstrate the techno-economic viability of fast breeders on a commercial scale. In PFBR a homogeneous core concept with two enrichment zones of 21% and 28% PuO_2 is adopted for power flattening. The clad material is D9 alloy. The maximum linear power is 450 W/cm and the initial peak burnup of 100 GWd/T envisaged will be enhanced to 200 GWd/T in the longer run with improved wrapper material. It is envisaged to utilise both these reactors for burning the minor actinides.

A remote fuel fabrication facility with sol-gel based microsphere production and vibropacking facilities is under an advanced state of commissioning in IGCAR and will be utilised to prepare test fuel pins containing AmO_2 .

Computational benchmarking

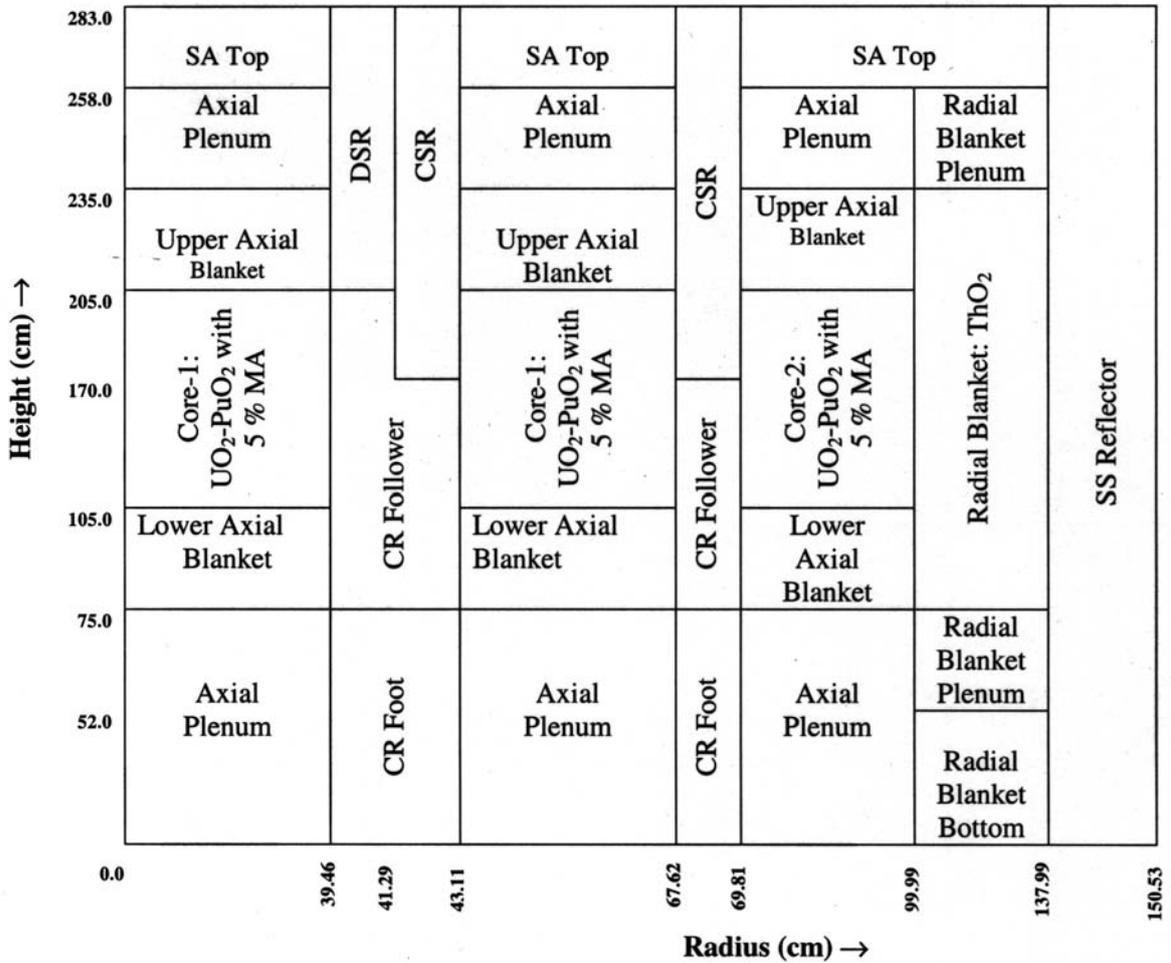
IAEA has initiated a Coordinated Research Programme (CRP) on “Studies of Advanced Reactor Technology Options for Effective Incineration of Radioactive Waste” [15] with an overall objective to perform R&D tasks contributing towards the proof of practicality for the transmutation of MAs and long-lived radioactive isotopes using fast reactors and Accelerator driven systems (ADS). IGCAR is participating in this CRP and has chosen the option of fast reactor core similar to that of PFBR for burning MAs separated from typical Pressurized Heavy Water Reactor (PHWR) fuel reprocessing.

The FBR benchmark used for this study has a power of 1150 MWth, with two cores of (U,Pu)O₂ MOX type fuel with PuO₂ enrichments of 19.5 and 27.1% in core1 and core2 respectively. It also has three rows of ThO₂ as the radial blanket while the axial blanket comprises depleted UO₂. To facilitate this study, 5% MA (by weight) has been put uniformly in core 1 and core 2. The R-Z model of the FBR benchmark used for MA incineration is given in Figure 1. The benchmark has a multiplication factor of 0.99844 and a total breeding ratio of 1.127. The MAs considered are ²³⁷Np(88.599 %), ²⁴¹Am(5.401 %), ^{242m}Am(0.045 %), ²⁴³Am(5.131 %), ²⁴²Cm(0.674 %), ²⁴³Cm(0.004 %) and ²⁴⁴Cm(0.146 %). This composition is typical of the discharged fuel from PHWRs in India. The reactivity coefficients for this benchmark have been computed and compared against those of PFBR and presented in Table 1.

Table 1. Comparison of Safety Parameters

Parameters	MA-Incineration Benchmark	PFBR [16]
Doppler constant: Fuel (pcm)	-443.7	-699.7
Sodium void worth (pcm)	+1340	+1256
β_{eff} (pcm)	469.5	340.0
Prompt neutron life time (μs)	0.34	0.39

Figure 1. R-Z Model of FBR Benchmark for MA Incineration (not to scale)



This benchmark has a cycle length of about 195 days. The reductions in the MA in core 1 for the burnups corresponding to 25 GWd/t (one cycle of operation) and 49 GWd/t (two cycles) are 12 and 22% respectively. Thus the core is an effective incinerator. No significant MA is produced in ThO₂ blanket. Overall reactor safety is comparable to PFBR.

PFBR has two cores of enrichments 20.7 and 27.7 % respectively in core1 and core 2, with depleted UO₂ radial blanket. It is seen that for this benchmark, there is an increase in β_{eff} (safer) and a fall in Doppler constant (less safe) with respect to PFBR. Changes in sodium void worth and prompt neutron life-time are not significant.

In ADS [17-19]

Reactor physics & Neutronics studies

A programme of theoretical and experimental studies in reactor physics of sub-critical system has been initiated. The theoretical programme is oriented towards development of codes for ADS core physics analysis and to study ADS designs for effective Th-utilization/actinides transmutation. Efforts are underway to develop Monte Carlo and transport theory codes with fuel burn up to facilitate accurate simulation of ADS fuel-cycle performance. These codes will be oriented toward analysis of ADS as thorium burner device; under the combinations of (i) lead bismuth eutectic (LBE) cooled fast reactor geometry, (ii) pressurized heavy water geometry with natural uranium as seeding material. Other configurations of ADS, such as the coupled fast-thermal ADS, are being studied to plan suitable sub-critical core with significant fission-power level. The fast neutron spectrum would be utilized for incinerating the actinides loaded fuel.

The experimental programme makes use of 14-MeV neutron generator and a light water-uranium rods sub-critical assembly. An existing 400-kV deuteron accelerator can generate maximum neutron yield of 3×10^9 n/sec, with deuteron beam current of about 0.3 mA and 260 kV energy on a 5-Curie tritium target (on titanium substrate). This is considered sufficient for reactor physics experiments with the sub-critical assembly having (estimated) k_{eff} of about 0.87. The deuteron ion source is currently being modified for (i) pulsing at 10 μs width and rep. rate up to 500 Hz, (ii) higher ion current up to 2 mA (iii) operating voltage with improved stability up to 350 kV etc.

The sub-critical assembly for these experiments is planned in an aluminium shell & tube type calandria vessel of about 1.4 m diameter using 300 metallic uranium fuel rods with aluminium cladding. The kind of experiments planned will help in validation of calculation tools and neutron cross-section data, neutron flux profiling and real time measurement of the k_{eff} .

Target technologies

Thermal hydraulics for window and windowless spallation target configurations are being investigated by theoretical modeling of the lead bismuth eutectic flow conditions. Rates of energy deposition by the high-energy proton beam have been calculated by using FLUKA code that was made available by the CERN ADS group. The results of volumetric heat deposition in the spallation reaction zone matched well with the published results of lead target design for energy amplifier.

An in-house developed computational fluid dynamics (CFD) code has been used for flow analyses in one dimension – single phase for buoyancy and two-phase axis symmetric for gas-driven, in the lead-bismuth spallation target region. The heat source terms from the FLUKA output are used in this simulation. More refinements in this code are being effected to represent actual target heating conditions.

The benchmarking of CFD code simulations is planned in an experimental LBE loop facility in which volume energy deposition is to be simulated by use of 10-kW electron beam and 50-kW plasma torch heating for windowless and beam window regimes respectively. A mercury circulation loop facility is under fabrication and assembly at a laboratory to conduct cold trials of diagnostics instrumentation for their subsequent use in hot LBE liquid metal system. Basic process system design of the LBE loop facility has been completed and its equipments are being ordered for procurement/fabrication. These include mechanical and electro-magnetic re-circulation pumps for hot liquid heavy metal at the maximum rate of 119 kg/sec (~36 m³/hr). This LBE experimental loop would also be utilized in the study of materials compatibility with regards to corrosion and erosion by the heavy liquid metals. A part of loop would consist of an active oxygen control facility for optimizing the operating parameters and studying effect of various passive coatings.

Accelerator technologies

At present, efforts are underway to develop key accelerator technologies for a high power proton accelerator. Its low-energy section has been planned as linear accelerator consisting of electron cyclotron resonance (ECR) ion source, radio-frequency quadrupole (RFQ) and drift-tube LINAC (DTL) tanks-all of normal conducting types. This accelerator would deliver 10-MeV, 30 mA (cw) proton beam initially and be upgradeable to 20-MeV. All infrastructures for this facility such as, plant building, RF power supplies and cooling water system etc., are under design. This accelerator would be located in Trombay. About 30 m long LINAC tunnel would have an adjacent klystron gallery. The floor above LINAC tunnel would be a hall and house all the modules of power supplies for, up to three klystrons of 1.3 MW cw rating each.

As a prototype for RFQ development, a short length RFQ has been planned to deliver 400 keV, ~2mA deuteron beam to serve as alternative high intensity neutron generator for sub-critical experiment mentioned above.

Until now, there has been no R&D programme in India on elliptical superconducting (SC) RF cavities. These are considered to be the mainstay of energy-efficient RF proton LINAC's high-energy section (when $\beta = v_p/c$ for proton energy $E_p > 100$ MeV is > 0.43), which would be best suited to drive the full-scale ADS reactors. An R&D facility has been planned under the present ADS technology development project to initiate electro-dynamics design analysis, construction and characterization of elliptical SC RF cavities. Two operating frequencies have been identified for the high power proton LINAC. These are-350 MHz (at low energy end of proton LINAC) and 700 MHz for the Superconducting section.

Activities at VECC & CAT

These two R&D units of the DAE specialize in the development and utilization of particle accelerators in India. The present accelerator R&D programmes at CAT focus on the electron synchrotrons and that at VECC on cyclotron accelerators. Considering the possibility of proton cyclotron as accelerator for driving an ADS, preliminary design of a suitable high current machine has been initiated in VECC. In the first phase, an ECR proton ion source and an injection beam line to the 10-MeV cyclotron is under development. The ion source would deliver 30mA cw, low emittance proton ion current that would be accelerated to 100-150 keV and passed through an RF buncher at 40-50 MHz. The scheme would include design and development of a spiral inflector to inflect this beam into the median plane of a cyclotron.

The accelerator R&D programme in CAT would be oriented toward developing a major facility of spallation neutron source based on proton synchrotron. Some studies have been initiated to work out details and utilization schemes of such a facility. Tentatively, it would consist of a linear RF proton accelerator (LINAC) as an injector to a rapid cycling, 1-GeV proton synchrotron. The proton LINAC would be high current, pulsed 100-MeV machine with duty factor as high as about 10%. The proton LINAC and the synchrotron would be planned to facilitate measurement of nuclear cross-sections as part of nuclear data development programme and also conduct sub-critical ADS reactor experiments in future. The project costs and construction schedule etc., of this scheme are under investigations at present.

Future Plans on ADS

As a first step, it is planned to build 5-MW fission power ADS. This would be a near-ambient operating temperature, sub-critical reactor, driven with a few mA average proton beam of 120-150 MeV. Such an accelerator could be scaled up from the technological developments of the 10 MeV proton LINAC or cyclotron. This accelerator can be a normal conducting linear accelerator (LINAC) or a cyclotron, both of which are well-proven technologies. This low power reactor would be used to verify the reactor-accelerator power coupling parameters, to adjust the reactor calculation code and data with new and innovative fuel such as thorium-uranium, and to study burn up reactivity kinetics without risks of “nuclear safety”.

In the long range, it is proposed to have a demonstration ADS version utilizing 10- 20 mA, 500-600 MeV proton beam to drive a sub-critical reactor of about 100 MW fission power. The main difference from the one mentioned above, would be that the operating temperature of the coolant, in this case, and would be around 350°C similar to that of a power generating plant. Also, the LINAC/cyclotron drivers should be based on partial/full superconducting technology. This ADS reactor could be applied to the study of the “nuclear safety” aspects with regard to the temperature reactivity feedback effects, as the case may be in a realistic ADS.

In both the above reactor plans, the main aspect of ADS reactor’s deterministic safety will be utilized to study various neutronics feedback effects which are of relevance to other innovative (including the critical) reactor systems based on new nuclear fuel combinations.

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