Concurrent Trends in Indian Fast Reactor Fuel Reprocessing

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Abstract. The Indian fast reactor program, which began with the construction of the mixed carbide fuelled Fast Breeder Test Reactor (FBTR) at Kalpakkam, has reached a level of maturity with three decades of operating experience and is stepping into commercial operations with the construction of Prototype Fast Breeder Reactor (PFBR). The necessary technology for closing the fuel cycle, which is vital for the success of the fast reactor program, has been concurrently developed facing the unique challenges posed by the fast reactor fuel. CORAL (COmpact Reprocessing of Advanced fuels in Lead cells), a pilot facility, has been operating successfully since the year 2003, reprocessing the spent fuel discharged from FBTR, with burnup upto 155 GWd/t and very short cooling periods. This facility has served the purpose of validation of the process as well as the equipments that were developed for fast reactor fuel reprocessing. Operating the facility has given valuable feedback for the Demonstration fast reactor Fuel Reprocessing Plant (DFRP), which will be a regular reprocessing plant for FBTR and also serve as a demonstration for the reprocessing of mixed oxide fuel from PFBR. The CORAL experience was also vital in designing the Fuel Reprocessing Plant (FRP) of Fast Reactor Fuel Cycle Facility (FRFCF), which would be a regular reprocessing plant for spent fuel discharged from PFBR. Considerable experiences gained and feedback obtained in design and operation of the reprocessing facilities provided vital inputs for achieving the required robustness in the fast reactor fuel reprocessing program. With the construction of FRFCF, the Indian fast reactor fuel reprocessing program would step in to the realms of commercial reprocessing. R&D efforts are also concurrently under progress to develop efficient processes and equipments, aqueous processes for reprocessing of the U-Pu-Zr metallic fuel as a fall-back for the pyro-chemical process.

Key Words: Fast Reactor Fuel Reprocessing, R&D in fast reactor fuel reprocessing.

1. Introduction

The Indian Fast Reactor Reprocessing program is strategically conceived with four stages, to achieve the required robustness for commercial reprocessing. The first stage namely process and equipment development has been completed and a modified PUREX process has been developed to suit the fast reactor fuel, along with several unique equipments. As part of the second stage, a pilot plant CORAL (COmpact Reprocessing of Advanced fuels in Lead cells) has been constructed and commissioned in the year 2003 and has been successfully operating since then. This has served the purpose of validation of the process and equipments. With the rich operating experience of the pilot plant, a demonstration plant, DFRP (Demonstration fast reactor Fuel Reprocessing Plant) has been designed and constructed and is under commissioning, which will form the third stage. This will demonstrate the operation at plant scale with the required availability and capacity factors and also the scalability of the equipments. The construction of FRP FRFCF (Fuel Reprocessing Plant of Fast Reactor Fuel Cycle Facility) will mark the four stage, which will be the commercial scale reprocessing and the plant is meant for reprocessing fuel from PFBR (Prototype Fast Breeder Reactor) and future FBRs (Fast Breeder Reactors). During operation of the pilot plant CORAL, several processes were developed and the experience gained set the platform for development of several equipments. This paper brings out the concurrent trends in various field such as the process development, equipment development and R&D of the Indian fast reactor fuel reprocessing program.

2. Process development

The process for fast reactor fuel reprocessing has to take in to account the specific needs of the high burnup and high plutonium spent fuel, which is typical of fast reactors. A modified PUREX process has been developed to suit the specific needs of the fast reactor fuel and has been validated in CORAL. The plant has successfully processed fuel with burup upto 155 GWd/t and very short cooling with good decontamination factors. Further to this, the plant is acting as a hot test facility for various process development works being carried out simultaneously. This section describes the various significant developments in the process for fast reactor fuel reprocessing.

2.1.Aqueous partitioning

In thermal reactor fuel reprocessing, organic partitioning is followed, whereas the same may not be feasible for high Pu fuels, where the external Uranium load will be higher. To overcome this various partitioning techniques such as electrolytic partioning, aqueous partitioning etc are under development. Of these, the aqueous partitioning technique is particularly promising. In order to formulate flow sheet condition for aqueous partitioning of plutonium, a 20 stage ejector type mixer settlers was installed in an experimental Facility. Flow sheet conditions were designed based on results of batch studies and computer code simulation aiming to get maximum decontamination factor for uranium product in organic phase with respect to plutonium and vice versa.

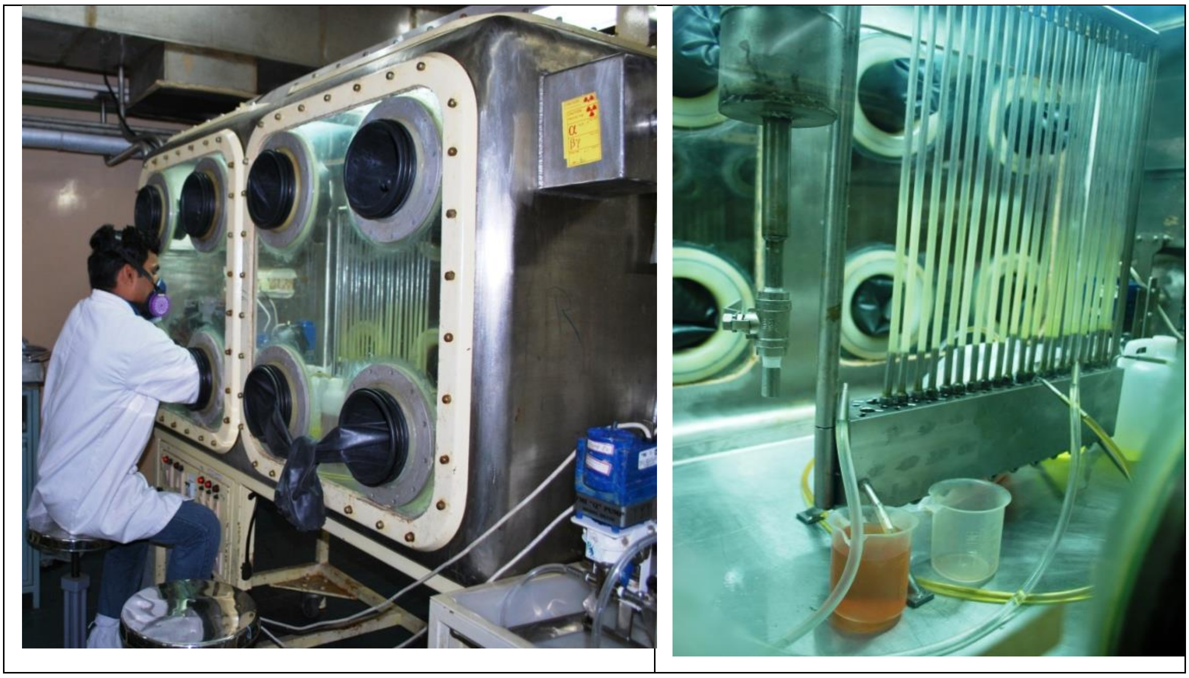

FIG.1. Laboratory scale demonstration of aqueous partitioning

Plutonium contamination in organic uranium product was found to be very low in the range of 30 to 40 ppm for different outlet samples taken at given time intervals. Results indicate that minimum 6-8 stages are sufficient for scrubbing of uranium from aqueous Pu product and for scrubbing of Pu from uranium product in organic phase respectively. This experimental run has given confidence that aqueous partitioning method for U/Pu is feasible on engineering scale and the same has been demonstrated in CORAL in centrifugal contactors.

2.2.Solvent extraction with dual Scrubbing

A unique challenge in the reprocessing of fast reactor fuel when compared to thermal fuel, because of its short cooled nature, is the decontamination to be achieved w.r.t the troublesome fission products namely Ruthenium and zirconium. Detailed modelling of the extraction behaviour of the various fission products have been carried out and a dual scrub extraction process was developed. This was validated in CORAL with hot fuel and was highly successful and a DF of more than 10^6 was achieved. This process will be adopted for DFRP as well as for the FRP-FRFCF.

2.3.Dissolution with soluble poison

In large capacity plants such as the FRP-FRFCF, contradictory requirement of large throughput meeting criticality safety has to be met. A continuous dissolution process would be more ideal, but in view of the technological challenges associated, it is proposed to use a batch dissolution process, using a soluble poison for criticality control. Further requirements such as ensuring the presence of the soluble poison before dissolution in a fool proof manner is also being implemented. Subsequently the impact of the presence of the soluble poison on the performance of the solvent extraction is also studied and found to be satisfactory. Since Gd is a neutron poison, its extraction should be minimum so as to meet the product specifications. An experiment was conducted to estimate the distribution co-efficient of Gd in the TBP Nitric acid system and parametric studies have been carried out for the effect of nitric acid concentration and Gd concentration on the distribution co-efficient of Gd. It is seen that the distribution ratio of Gd is between 10^{-3} and 10^{-2} in and hence the required product specification can be met.

2.4.Solvent treatment and recycle

Conventional solvent treatment technique using sodium carbonate has been found not suitable for the FBR fuel reprocessing and various solvent wash agents have been tested in CORAL. Recently solvent wash employing oxalic acid has been tested with hot solution and the treated solvent has been successfully reused for solvent extraction.

2.5.Aqueous reprocessing of metallic fuel

Though pyro-processing would be the ideal route for metallic fuels, as a fall back arrangement, aqueous reprocessing route is also being concurrently developed. Solvent extraction studies have been carried out with a simulated solution of typical U-Pu-Zr fuel. During the initial runs with a scrub acidity of 2M, crud formation was observed and subsequently the scrub acidity was raised to 3M where no crud was seen. The run had good recovery and the Zr pickup was found to be very low of the order of <0.01 mg/g of U+Pu, in the product.

3. Equipment Development

As part of the first stage of the reprocessing program, systematic efforts were put in for equipment development and several unique equipments were developed for the fast reactor fuel reprocessing such as the single pin chopper, Titanium dissolver with electrolyser, high speed centrifuge for feed clarification, compact centrifugal extractors with very low residence time etc. These equipments have been operating in CORAL successfully and based on the feed back their design have been refined, to make them robust. Improved designs are being evolved for applications in large capacity commercial reprocessing plants, where their robustness is an important requirement for the plant availability. A few significant developments are listed in this section.

3.1.Sub assembly dismantling machine

A Laser dismantling machine has been developed, deployed in the hot cells of Post Irradiation Examination and dismantling of many FBTR spent Fuel Sub Assemblies (FSA) have been successfully carried out. Based on this experience a fuel dismantling machine for dismantling PFBR FSAs have been designed and is being manufactured. It has the advantages like high capacity, less powder/chip generation, less solid waste generation, and modular in design amenable for remote maintenance. The machine is equipped with a mechanical slitting saw as a standby to the laser and is fully automated so that the entire dismantling operation will be completed in a sequential and automated manner.

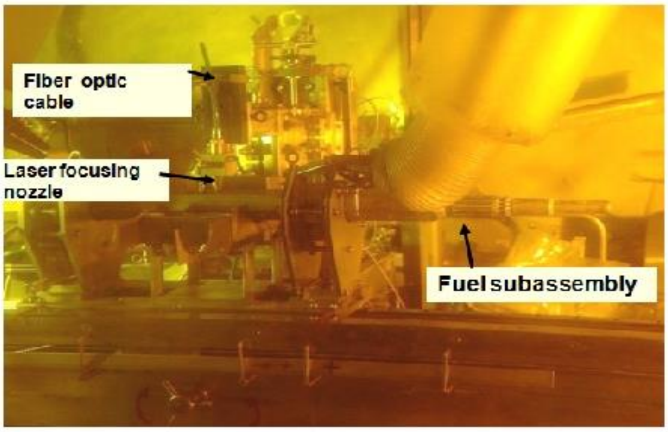

Fig.2 Typical laser dismantling machine

3.2.Fuel pin chopper

Because of the slender nature of the FRFR fuel pins, to avoid crimping, single pin chopper are being developed. A single pin chopping machine has been designed and developed for CORAL. The performance of the single pin chopper has given valuable input for the design of the chopper for DFRP. Based on CORAL experience, the pin feeding mechanism has been modified for increasing the reliability and throughputs. Various improvements have been carried out in the chopper design for DFRP like modifying the gripper, stationary and cutting tool for avoiding entanglement of the spacer wire avoided, change of material of bed plate to titanium to minimize wear and tear etc. DFRP chopper is designed for chopping three types of fuel pins of FBTR with different dimensions. The chopper for DFRP has been designed, manufactured, subjected to rigorous testing and has been erected inside the hot cell of DFRP.

To meet the capacity requirements of FRP-FRFCF, a special and unique shearing machine which is first of its kind has been designed, to shear fuel pins of Fuel Subassembly (FSA) of PFBR. It adopts a vertical shearing design. The machine shears ten pins at a time. Design of vertical machine is envisaged to meet the capacity requirement of FSA fuel pin in FRP and to avoid the use of multiple mechanisms such as shunter, Titanum Base plate, holder for stationary and gripper block, pin pusher etc. with respect to horizontal shearing machine.

Rigorous demonstration of fuel pins shearing without any remarkable notice on tool profile has proven the soundness of the cutting tool design. Measurement of actual cutting force at various cutting speed was found to be in close agreement with estimates. The tool life is also estimated to be 10^6 cycles based on fatigue analysis.

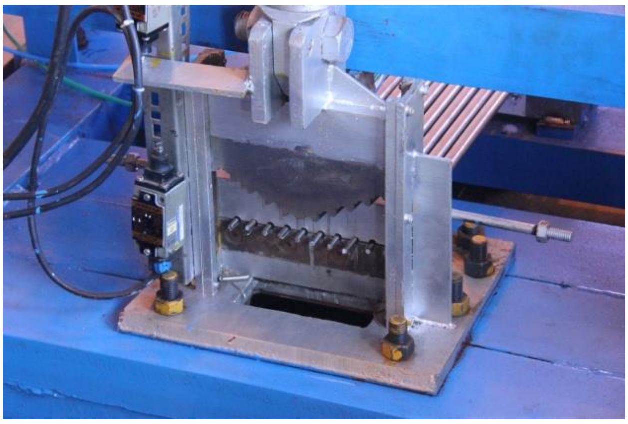

Fig.3 Multi pin chopper of higher throughput for future plants

3.3.Development of High Speed Centrifuges

Spent fuel with high Pu content upon dissolution may contain finely suspended particles which are mainly inter-metallic noble metal alloys. To avoid difficulties in downstream processing, the dissolver solution is required to be clarified. The centrifuge is intended for efficient separation of very fine solids from liquids. An air turbine driven centrifuge which can be operated and maintained remotely is deployed in CORAL whose performance is satisfactory. Since the centrifuge rotates at super critical speeds developing high centrifugal acceleration, detailed vibration analysis for evaluation of the critical speeds was carried out in order to address the various maintenance problems encountered at CORAL. Improvement in the design of bearings by changing from SS bearings to ceramic bearings have been carried out, to improve the availability.

The centrifuge system has been tested with a simulated feed of Ferric Oxide (Fe_2O_3) powder suspended in DM, upto a flow rate of 400 lph. From the studies it is seen that the system is capable of achieving separation efficiencies of above 98% for particle sizes upto 5 micro meter, for a feed flow rate of 300 lph. Based on the study, optimum number of units have been employed for FRP.

3.4.Centrifugal extractors for solvent extraction

Centrifugal extractors of low capacities have been performing exceptionally well in the pilot plant CORAL and has delivered good decontamination factors even for fuel with very short cooling periods. Centrifugal extractors of larger diameters catering to flow rates as high as 300 lph are being developed and will be employed for commercial plants of the future, including FRP.

The centrifugal extractor motors are exposed to radiation and vapours of 4M nitric acid leading to corrosion. For the corrosion protection Polyetheretherketone, PEEK coating was chosen as it is a high performance engineering thermoplastic which has a radiation resistance of 10^9 rads. PEEK coating was developed and motors were fabricated employing the coating with a thickness of 80 to 90 micrometer. These motors have been extensively tested at CORAL and have considerably reduced failures due to corrosion. No swelling, thickness change and peeling were observed after testing.

Fig.4 Rotor and stator of centrifugal extractor with PEEK coating

3.5.In-service inspection

Inservice inspection (ISI) would be vital to ensure the healthiness of the equipment during its operating life. As the majority of the critical equipments in the reprocessing plant are not accessible to inspection during operation, special inspection techniques and devices are called for. In pursuit of this, IGCAR has developed several techniques for the inspection of equipment such as Dissolver, Evaporator, waste storage tanks and process vessels in general. Apart from visual inspection with the aid of a remotely operable camera, thickness measurement utilizing ultrasonic principle is also being adopted. These systems have been validated and have been successfully used in CORAL plant for inspection of dissolver and also the waste tank farm, which has given a good feedback. Special autonomous vehicle mounted systems are also being developed for DFRP and future plants.

Fig.5 Remote inspection of CORAL dissolver vessel

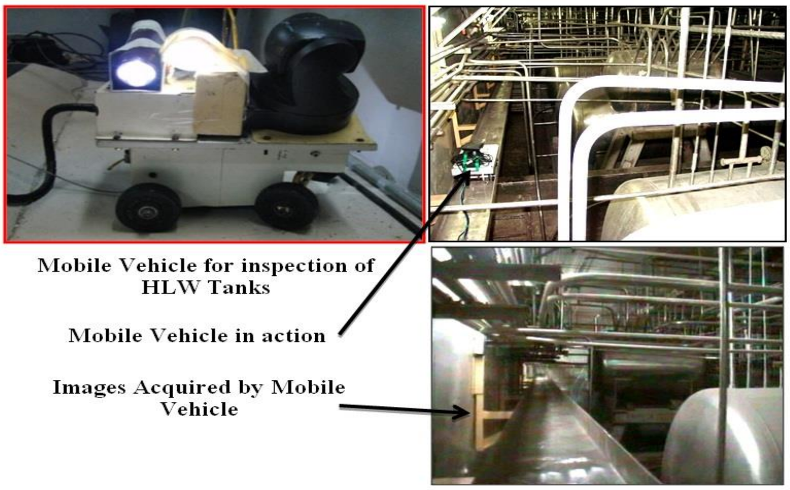

Fig.6 Inspection of Waste tank farm with mobile vehicle

4. Research and Development

4.1.Dissolution of PFBR inner core MOX pellet in nitric acid

MOX fuel will be the primary driver fuel for FBRs for the near future and it is necessary to have data on the dissolution kinetics of the fuel for the design of dissolution equipment. PFBR MOX fuel pellet dissolution studies in nitric acid were carried out to evaluate its dissolution kinetics. The fuel pellets used for the studies are the inner core pellets with the composition 21.7% PuO_2 and rest are UO_2. The time taken for complete dissolution is measured. Periodic samples were also taken to analyze U(VI), Pu(IV), Pu(III) and acidity to evaluate the dissolution kinetics. From the results it was concluded that all the MOX pellets dissolved rapidly and completely in comparison to UO_2 pellets. As the MOX pellets have annular gap, the dissolution was happening both from the outer and the inner surface and hence the pellets dissolved very rapidly.

4.2.Extraction of Zirconium by TBP

The ^{95}Zr - ^{95}Nb pair is responsible for the majority of gamma activity of dissolver solution of short cooled fuels and it contributes to around 1% of the total activity for a typical fast breeder reactor fuel with 6 months cooling. The fission yield of Zr is 7-7.5%. In the concentration range of interest in the study of FBR spent fuel reprocessing flow sheets, only Zr^{4+} is the dominant species. Hence, ^{95}Zr is to be considered while designing solvent extraction flow sheet for reprocessing fuels with a cooling period of less than two years. Hence, it is necessary to understand the extraction kinetics of zirconium with the solvent, tri-butyl phosphate (TBP) /n- dodecane (DD) system.

Extraction kinetics of zirconium in the mixture of tributyl phosphate (TBP) and n-dodecane was investigated experimentally. Based on the experimental results it was concluded that the reaction with the extractant takes place at the interface and the overall rate is controlled by intrinsic kinetics of the reaction of the species involved. The study indicated that improvement in the decontamination of uranium and plutonium from zirconium can be achieved when the extraction process is performed in short residence time contactors (i.e. centrifugal extractors) due to the difference in the rate of extraction of these elements.

4.3. Removal of Dissolved TBP from Aqueous Streams of PUREX Process by n-Dodecane Wash

Tri-n-butyl phosphate (TBP) is used as the extractant in the PUREX process for the recovery of uranium and plutonium from spent nuclear fuels, due to the mutual solubility of aqueous and organic phases. All the aqueous streams leaving the extraction cycle will contain some entrained and dissolved TBP and its degradation products. The presence of traces of organic compound in aqueous stream is undesirable as it would lead to violent reactions during the evaporation of waste solution. Diluent wash using n-dodecane is one of the established methods for the removal of dissolved TBP. The diluent used for the diluent wash process also contributes to this organic waste. Hence, kinetic studies were carried out for the removal of dissolved TBP from aqueous streams with different (a/o) ratio and mixing speed. Experiments were conducted with a solution containing around 200 ppm of TBP dissolved in 4 M nitric acid. Aqueous–organic solution with (a/o) ratio as 10:1 and 5:1 and vortex mixer's speed as 500 to 1700 rpm were used. Good separation of TBP from the aqueous streams were achieved.

a. Development of compact short path distillation unit for solvent recovery

Fig.7 Solvent purification system

Tri butyl Phosphate diluted with a suitable hydrocarbon is used as solvent in the extraction step of PUREX process for spent fuel reprocessing. The useful life of the solvent in reprocessing plant is limited by the extent of formation of heavy molecular weight secondary degradation products during the solvent extraction step. These degradation products are formed by the interaction of nitric and nitrous acids with various hydrocarbon compounds under the conditions of radiation and nitric acid. One of the easily deployable methods for the removal of these secondary degradation products is vacuum distillation. The instability of tri-*n*-butyl phosphate at elevated temperatures and the requirement that very low concentrations of variety of impurities that have to be removed, call for a careful design of process and equipment. A lab scale solvent purification system (Fig.7) based on short path distillation has been installed and commissioned at Reprocessing Research and Development Division, Reprocessing Group, IGCAR. Trail runs are being carried out with simulated degraded solvent to assess the purification capacity and solvent recovery of the compact short path distillation unit. Results were found satisfactory.

5. Conclusions

The Indian reprocessing program is at an important juncture where it is about to step into commercial operations after gaining vital operating experience. As detailed in this paper several improvements in process as well as equipments have been carried out based on the feedback, which will give the necessary robustness for commercial operations. Metallic fuel is being planned to be introduced in the fast reactor program in the near future, the reprocessing of which will be a challenging task. Aqueous route is being envisaged as a fall back arrangement to pyro reprocessing, at least for the initial period till the time pyro route is established.

Appendix 1: Reference

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