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Fast Reactor Fuel Reprocessing Technology in India

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The limited indigenous uranium resource coupled with the need of energy independence necessitated the Department of Atomic Energy (DAE) in India to opt for a three-stage nuclear power program comprising of PHWRs in Stage 1, FBRs in Stage 2 and Th-U^{233} based reactors in Stage 3, respectively to meet the energy needs of the growing Indian economy. Presently the country has stepped into the 2nd Stage. The transition from the Stage 1 to 2 was fruitful thanks to the 2-decade long R&D experience gained in Fast Reactor Fuel Reprocessing (FRFR). Closing the fast reactor fuel cycle through FRFR was inevitable for the success of the Indian Nuclear Power Program (INPP). The latest achievement by the center was the successful reprocessing of 100 GWd/t burnup mixed carbide fuel with 70% Pu, discharged from the Fast Breeder Test Reactor (FBTR) which is also located in the same center. The designs of the various equipments and process flow sheet had stemmed from the above experiences thereby increasing the confidence level in the future plans of the Department. In this paper, an overview of the experiences in FRFR with glimpses of the various developmental activities towards the future plants is spelt out.

KEYWORDS: fast reactor, fast reactor fuel reprocessing, single pin chopper, centrifugal extractor, SIMPSEX

I. Introduction

The Department of Atomic Energy (DAE) in India has stemmed down to its present status through systematic and streamlined research and developmental activities in the field of nuclear science and technology. Its multidisciplinary activity in science and technology has paved the way for the successful completion of the first stage of the three-stage Indian Nuclear Power Program (INPP). Presently in the Stage 2 of the program, the closing of the nuclear fuel cycle has taken the center stage. Thus, the Fast Reactor Fuel Reprocessing (FRFR) figures at the top of the Department's agenda.

Though the Stage 2 has just begun, it has been envisaged long back with the operation of the Fast Breeder Test Reactor (FBTR) since 1985 at the Indira Gandhi Center for Atomic Research (IGCAR), Kalpakkam. FBTR is the first of its kind in the world to have the mixed carbide (MC) fuel with 70% Pu as its driver fuel. The facilities to reprocess the FBTR spent fuel were located at the Reprocessing Development Laboratories (RDL) in IGCAR. The laboratory backed by the confidence of the Department's experiences in Thermal Reactor Fuel Reprocessing (TRFR) achieved a major milestone in FRFR by commissioning a pilot plant, the CORAL (Compact Reprocessing facility for Advanced fuels in Lead shielded cells, **Fig. 1**), formerly known as the Lead Mini Cell (LMC), a facility to reprocess the MC fuel for the first time in the world. The CORAL targeted at demonstrating the flow sheet for FRFR with simultaneous validation of the various prototype equipments under hot realistic conditions and carrying forward the experiences in constructing the future FRFR plants. This paper gives an account of the Indian experiences with FRFR and the strategies to be adopted in the development of the technology of FRFR for the future.

II. Origin of FRFR in India

The technology of FRFR in India is planned to be executed in the four phases.[1] The design and development of equipments and systems followed by engineering scale experiments in non-radioactive conditions comprise the first phase; setting up of a Pilot Plant (CORAL) to run the active trials, the second phase; operating a plant of demonstration scale (DFRP), the third phase; and the construction and operation of a prototype plant (FRP) constitute the final phase. The realization of the CORAL was feasible owing to the systematic studies carried out in RDL on the various equipments like chopper, dissolver, centrifugal extractor, *etc.* The CORAL, commissioned in late 2003, has now successfully reprocessed the 2.5-years cooled 100 GWd/t burnup MC fuel with 70% Pu discharged from FBTR. The process has gained significance due to the very high Pu content and high specific activity[2] of the spent fuel. The Plutonium Uranium Extraction (PUREX) process[3] which has been well

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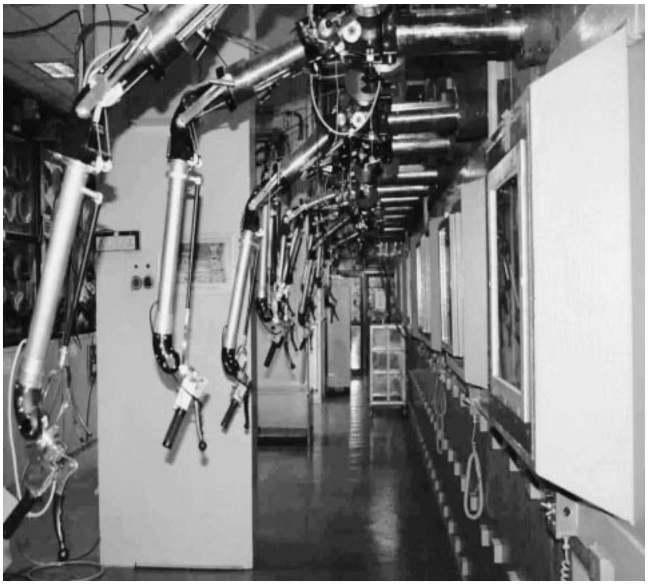

Fig. 1 CORAL–Hot cell operating area

understood now is likely to remain the workhorse of FRFR for the next couple of decades at the minimum.

III. Foundation for FRFR in India

The Fast Reactors are important in the Indian context due to the high breeding and low doubling time if the fuel cycle is closed as early as possible with the shortest possible cooling time. But all these could be made feasible only if the fuel cycle is closed. This realization of the FRFR had been the foundation stone for the technology in India. Only through its sustained research and developmental work, DAE was able to dwell over this foundation and bring the technology of FRFR in India to its present status. The R&D work in the field of FRFR aimed at process and equipments development, material development, and development of remote handling system, all of which could be tested at the CORAL during the FRFR campaigns.

1. Process and Equipments Development

The R&D work for the development of process and equipments for FRFR was carried out in the RDL at the IGCAR. The initial testing of all these equipments was done at the engineering laboratory of the RDL prior to their deployment in the realistic hot conditions. Some of the equipments developed were used in the reprocessing campaigns of thorium fuel rods to recover U^{233} used in the fabrication of FBR MOX fuel for the irradiation experiments in FBTR. The 30 KWt KAMINI reactor, the only reactor in the world operating with U^{233} as its driver fuel, uses this material. Some of the equipments developed are as follows.

(1) Single Pin Chopper

The dimension of the fuel pins of the thermal reactor is in favor of their chopping in bundles after their irradiation at the head-end step of their reprocessing. Whereas with the FR fuel pins being very slender with more Pu content (hence more safety with respect to criticality), harder cladding material and higher heat generating nature of fuels; the single pin chopper is the only option for a safe and smooth chopping operation. Hence it was essential to develop a Single Pin Chopper (SPC). The SPC developed at RDL is modular in nature with remotely maintainable parts using the Master Slave Manipulators (MSM). A prototype SPC installed in the CORAL, after a meticulous testing under inactive conditions, has performed satisfactorily so far.

(2) Dissolution and Electrolytic Dissolver

Nuclear fuels with Pu content exceeding 35% is difficult to dissolve. The MC fuel multiples the problem with formation of complex organic compounds during their dissolution.[4,5] The possibility of their interference in solvent extraction,[6] which is not ruled out, also adds on to the fear of MC fuel dissolution. These constraints were overcome with the development of an advanced dissolution process like silver catalyzed electrolysis.[7] A study with ozone for destruction of organic species[8] has been successful, which is planned to be tested under plant conditions. The electrolytic dissolver is made of Titanium Grade-2 material. The Titanium-Stainless steel interfacing was done by specially designed dissimilar joints.[9] Furthermore, as a part of our R&D work, gadgets for carbon estimation in the active dissolver solution have also been developed.

(3) Centrifuge for Feed Clarification

During chopping, numerous fine particles get generated from clad or from the fuel matrix and sometimes from both. The fuel being rich in Pu might yield fission product alloys at higher burnup which do not dissolve during dissolution, resulting in solvent degradation and their accumulation in the aqueous-organic interface during the SE process rendering unproductive extraction. A high speed air turbine operated centrifuge, operating at 15,000 rpm providing 125 times the acceleration due to gravity, has been developed for filtering the dissolver solution. Its design accomplices remote operation and maintenance by MSM.

(4) Solvent Extraction and Centrifugal Extractors

In comparison with TRFR, FRFR poses challenges in manifolds in accordance with the associated decay heat and Pu content. **Table 1** provides a comparison between TRFR and FRFR.

At the RDL, we also developed a simulation code, SIMPSEX[10] for optimizing the process parameters for the

Table 1 Typical comparison of TRFR and FRFR

S. No.	Property	TRFR	FRFR
1	Pu/(U+Pu)	0.004	0.15 to 0.7
2	Specific β-γ activity	~200 Ci/l	~1,000 Ci/l
3	Third phase problem	—	Possible at some conditions

extraction units. Appropriate models[11)] have also been developed for tackling the third phase during extraction so that the conditions leading to their formation could be avoided.

The higher decay heat of the fast reactor spent fuel offers great restriction in the time of contact between the fuel and solvent, breaching which the TBP get heavily degraded giving rise to its degraded products DBP and MDP. The development of several variants of centrifugal extractors[12–14)] took care of this problem thereby improving the solvent extraction performance.

2. Material Development

In an effort to improve the performance of the AISI type 304L austenitic SS as the main construction material for the FBR reprocessing and waste management plants, IGCAR is collaborating with other industries to optimize the specification of tramp elements and carbon. All such materials developed and also those available in the market are being tested in the nitric acid loop at IGCAR for obtaining suitable guidelines for specifying the SS grades and welding procedures for the future plants.

Corrosion resistant alloys like Ti-5%Ta-1.8%Nb are being developed for dissolver and evaporator. Dissimilar metal joining for Ti-SS has been already developed and deployed[15)] and an improvement of these techniques is in progress. A small scale dissolver with Zircaloy has also been fabricated and is undergoing testing.

3. Development of Remote Handling System

The decay heat associated with FRFR plants demands for a contained operation of the equipments within a cell with heavy shielding. These equipments have to be operated remotely. The articulated arm type MSM developed for this purpose was deployed in the cells. These MSMs are used for a payload of 5 kg and 7 degrees of motion for remote replacement or repair operations. Heavy duty manipulators of 25 kg capacity with an extended arm type and a three-piece type are under advanced stage of development for deployment in the concrete cells. They have the special features of α-tightness so that α contamination during maintenance could be reduced.

IV. Fuel Reprocessing Experiences at CORAL

The CORAL is a compact lead shielded leak tight SS containment box designed on a no-direct maintenance concept to handle β,γ radioactivity of the order of 10^5 Curies and to protect the working personnel from α contamination. It is an $11\,\text{m} \times 2\,\text{m}$ box, housing all the equipments like single pin chopper, dissolver, centrifuge, CE (Centrifugal Extractors), sampling units, *etc.* Gadgets like radiation-shielding windows, in-cell crane, and articulated arm type MSM for remote operation and maintenance of equipments and systems in the containment box make the CORAL a self contained facility. The process vessels, located below the containment box, are specially designed to either an annular or a slab type for avoiding criticality. All the equipments, process vessels, and other service lines constituting the entire plant flow sheet are connected by intricate SS piping with about 3,000 bends and stretching to a total length of about 2 km. The facility offered great challenges en route to its commissioning. The CORAL took up progressive reprocessing exercise of low burnup fuel pins, 25 GWd/t fuel pins, 50 GWd/t fuel pins, and 100 GWd/t fuel pins in that order with stage-wise clearance from the Safety Review Committee for Operating Plants (SARCOP) of the Atomic Energy Regulatory Board (AERB) in India. Due to the lack of experience in handling MC fuel and too with burn-up of this magnitude, safety clearance in each stage was given in accordance with the operating experience of the earlier stage.

1. Fuel Receipt

The fuel subassemblies after their irradiation at the FBTR are discharged and disassembled at the post irradiation examination cells into separate pins. Ten such pins are then loaded into an SS fuel magazine and once such a magazine is transported at a time to the CORAL in a special α tight container housed inside a lead shielded cask.

2. Chopping

The indigenously developed single pin chopper chops the irradiated fuel pins at the CORAL prior to their processing. The operation is simultaneously purged with dry Ar gas to avoid flashes that occur due to the pyrophoric nature of the carbide fuels. During the various reprocessing trials, the Ar flow rate was optimized.

3. Dissolutions

The importance of the CORAL to the FRFR technology in India was evident from the design input of the spent fuel dissolution process. The parameters for the electrolytic dissolution in the plant were based on the laboratory experiments on a single pellet. The necessity to enhance the dissolution rate and to destroy the carbon compounds formed during the dissolution of the MC fuel prompted for adopting the electrolytic dissolution process. The optimization of the process parameters was based on the results of the various reprocessing trials at the CORAL. The difficulties in conditioning the feed prior to the solvent extraction process were overcome successfully. The air operated centrifuge designed for feed clarification performed successfully so far.

4. Solvent Extraction

After two batches of dissolution, initial batch runs for extraction and stripping were carried out in the laboratory using samples of dissolver solution. This enabled in establishing the extraction parameters in the plant. On the basis of the outcome of this study, the conditioning step for the destruction of dissolved carbon was ruled out in the plant. The Pu lose in the raffinate stream during the various CORAL campaigns was generally low. Based on the experiences of the initial campaigns, the Pu loss was further minimized in the later campaigns by the multiple point introduction of uranous nitrate stream in the strip section. Hence, Pu retention in the lean organic stream has been overcome.

All the three models of CE used in the CORAL performed well and were provided with electrical devices. After hot commissioning of the plant, modification in the mixer section in one type of CE was carried out using a specially designed internal. In the CE of a variable interface type, the maintenance problem was reduced by modifying the sealing. As and when the requirement of motor change arrived, it was done remotely using the MSM and in-cell crane. The electrolytic mixer settler also performed well, though it required a change of electrodes once. As an alternate partitioning method, a selective precipitation route using oxalic acid was adopted and the results were very satisfactory.

5. Reconversion

The reconversion operation carried out during the various CORAL campaigns has benefited us with enormous confidence in handling appreciable quantity of Pu at a higher concentration. During the later campaigns, some minor modifications in the precipitator and furnace were carried out for a hassle free operation.

6. Solvent Treatment

The Pu retention in the used solvent was pretty high. Hence a method based on ammonium carbonate[16] was developed and adopted in the plant operation successfully. Using this method, Pu was recovered and the solvent was either recycled or disposed safely depending on its properties.

7. Hot Cell Operation and Maintenance

All the equipments such as MSMs, in-cell crane, sampling systems, capping-decapping systems, and in-cell analytical systems were operated to the desired levels of requirements and found to be satisfactory. In-cell crane could be maintained remotely. An improvement in the pneumatic conveying system had been carried out. The sample bottle design and conveying method has been modified based on the operational feedbacks.

8. Instrumentation System

All the instrumentation systems for the measurement and data acquisition of process parameters, logic control systems in chopper, dissolver, centrifuge, centrifugal extractors, interlock systems for liquid transfer, sampling and exhaust & off-gas systems performed as per design specifications. The working of all the radiation monitoring instruments and fire alarm systems were satisfactory and are as per technical specifications. The spectrophotometric based system for direct measurement of Pu performed very satisfactorily without any secondary waste generation. This system enabled easy process control of valency conditioning of Pu which is very vital for controlling the losses of Pu in the solvent extraction.

V. Path from CORAL to DFRP and FRP

The decontamination and recovery factors achieved in the various campaigns of the CORAL with irradiated FBTR MC fuel up to 100 GWd/t burnup have given confidence to take up 150 GWd/t burnup fuel discharged from FBTR. The process flow sheet for the DFRP and FRP has been finalized based on the experience and feedback of the CORAL operation. The CORAL has paved the way for developing, fine tuning, and scaling up the process equipments for the future plants with higher throughput. Advanced equipments like the active neutron interrogation for hull monitoring, the multipin chopper, rotary dissolver, screw calciner, and a compact layout design with poison tube cylindrical vessels are some of the equipments which are being developed to meet the future requirements. The organic entrainment in a strip product observed during the CORAL campaigns is a safety-related issue which is being addressed. Its early detection during the plant operation will remain an important requirement. The DFRP operation will aid in estimating the various performance factors of the plant operation. The erection of this plant is under way.

The DFRP operation will enable further improvement in recovery and reduction in the number of cycles apart from the plant scale experiences on solvent management. This will aid in reduction of waste generation.

The FRP, an integral part of the commercial operation of the FBR fuel cycle, is being designed with a firm grip over its construction cost as well as time. An innovative design with inherent safety features, like modular construction with batch execution especially for the reconversion operation, is the order of the day as far as the construction of FRP is concerned as it handles a large amount of Pu. Various R&D activities on in-service inspection and online monitoring are concurrently being undertaken to improve the plant performance.

VI. Conclusions

The results of all the CORAL reprocessing campaigns were extremely encouraging. It has given valuable inputs to undertake the design of chopper, dissolver, and centrifugal extractors for the DFRP. A lot of experiences have been gained in the remote operation and maintenance of hot cell equipments using in-cell gadgets. The successful reprocessing of 100 GWd/t burnup short cooled fuel through the PUREX process has proved the credentials of the method even for the fast reactor fuels.

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References

1) Baldev Raj, R. Natarajan, "An integrated approach to reprocessing and waste management of fast reactor fuels," *IANCAS Bulletin*, **1**[4], 51–59 (2002).
2) R. Natarajan, "Challenges in fast reactor fuel reprocessing," *IANCAS Bulletin*, 27–32 (1998).
3) J. C. Boudry, P. Miquel, "Adaptation of the purex process to the reprocessing of fast reactor fuels," *Lyon, ISEC*, Paper No. 253 (1974).
4) L. M. Ferris, M. J. Bradley, "Reactions of uranium carbides

with nitric acid," *J. Am. Chem. Soc.*, **87**, 1710 (1968).
5) H. Bokelund, M. Cadeci, M. Ougier, "Dissolution of mixed carbide fuels in nitric acid," *Radiochim. Acta*, **30**, 49 (1982).
6) J. R. Flanary, J. H. Goode, M. J. Bradley *et al.*, *Hot Cell Studies of Aqueous Dissolution Process for Irradiated Carbide Reactor Fuels*, ORNL-3660, Oak Ridge (1964).
7) A. Palamalai, S. K. Rajan, A. Chinnusami *et al.*, "Development of an electro-oxidative dissolution technique for fast reactor carbide fuels," *Radiochimica Acta*, **55**, 29–35 (1991).
8) R. Geetha, M. K. Ahmed, P. Velavendan *et al.*, "Application of ozone in nuclear fuel reprocessing studies on destruction of residual carbon in solutions of uranium carbide and uranium plutonium mixed carbide," *Proc. NUCAR*, 169 (2003).
9) U. Kamachi mudali, B. M. Anandarao, K. Shanmugam *et al.*, "Corrosion and microstructural aspects of dissimilar joints of Ti and type 304L SS," *J. Nucl. Mater.*, **321**, 40–48 (2003).
10) Shekahar Kumar, S. B. Koganti, "Dynamic simulation of high plutonium flowsheets proposed for reprocessing FBR fuels," *Proc. Int. Solv. Ext. Conf.(ISEC- 96)*, Melbourne, Australia, March 19–23, 1996 (1996).
11) Shekahar Kumar, S. B. Koganti, "Empirical modeling of Pu(IV) third phase formation in 30% TBP/n-Dodecane systems," *J. Nucl. Sci. Technol.* **33**[12], 1003–1005 (1996).
12) S. B. Koganti, "Equipment for spent fuel aqueous reprocessing: The scope and the challenges. Special Issue on Nuclear Fuel Reprocessing," *IANCAS Bulletin*, **14**[2], July 1998 (1998).
13) S. B. Koganti, V. Sridharan, G. R. Balasubramanian, "Development of low capacity centrifugal extractors for small scale reprocessing facilities," *Proc. Int. Solv. Extr. Conf. (ISEC-86) Pre-print*, **1**, 413–420 (1986).
14) S. B. Koganti K. Periasamy, C. V. Rajagopalan *et al.*, "Experience with centrifugal extractors," *4th Int. Conf. Nuclear Fuel Reprocessing and Waste Management (RECOD '94)*, **1** (1994).
15) U. Kamachi Mudali, R. K. Dayal, J. B. Gnanamoorthy, "Corrosion studies on materials of construction for spent nuclear fuel reprocessing," *J. Nucl. Mater.*, **203**, 73–82 (1993).
16) P. Govindan, K. Dhamodharan, K. S. Vijayan *et al.*, "Development of a novel method for recovery of plutonium from lean organic," (under preparation).