Evolution of Back End Fuel Cycle in India

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Preamble

As India Celebrates her 75th Year of Independence, the present timeline seems most appropriate to revisit our important milestones in the evolution of back end technology in India and to identify a road map for the future. While doing so, one cannot but get inspired by the monumental decisions and ground breaking developmental efforts that have not only led to expertise in this sophisticated field but is also ensuring sustainability of our nuclear power programme. This present paper attempts to trace evolution of the back end of fuel cycle in the Indian context and highlights its present status.

Historical perspective

Closed fuel cycle involving reprocessing of spent nuclear fuel is considered a vital link to realise the Indian nuclear power program aiming at optimum utilisation of uranium and thorium resources. While spent nuclear fuel from nuclear reactor is considered as 'waste' in many countries, same is considered as 'material of resource' in the Indian context due to the presence of unused uranium and freshly formed plutonium in the 'spent fuel' that emanates from nuclear reactors. Recovery of such energy potentials, though highly desirable, is very complicated and involves a multidisciplinary approach which involves the complex technology of reprocessing. India today enjoys the special status of a country possessing this exclusive technology along with only a hand-full of countries in the world.

If Dr. Homi Jehangir Bhabha is credited with chalking out our well entrenched three stage nuclear power programme of our country, it was Dr. Homi Sethna who took the next epoch-making step towards the closed nuclear fuel cycle. As the adage goes…a Journey of a thousand miles starts with the first step and this first step taken by these great stalwarts paved the way for a self-reliant India and an Atma Nirbhar Bharat as it is today. The back end of the fuel cycle involves both complex technology with regard to reprocessing and the subsequent management of highly radioactive waste streams, both of which need massively shielded enclosures and completely closed environment so as to ensure negligible release of radioactivity into the environment. While embarking on this very challenging path, the highly radioactive waste was perceived as a hurdle, but the technological advancements in our country have changed this perception from that of a problem to a potential source of wealth. This is primarily due to adoption of a strategy that results in recovery of radionuclides like Cs-137, Strontium-90 and Ruthenium-106 for societal benefits

Recycling of Spent Nuclear Fuel

The Indian recycling programme was born with the setting up of India's first reprocessing plant at Trombay, Mumbai. It is indeed very interesting to note that the formal order to set up a plant to reprocess spent fuel from CIRUS reactor was issued dated December 31, 1958 at which timeline the reactor was still under construction. This project was aptly named “Project Phoenix”. Just as the mythological bird Phoenix obtains new life by arising from the ashes of its predecessor, the project phoenix was also aimed to recover new fuel from the spent fuel of reactor. In the absence of much design details, the 'Project Phoenix' (there after renamed as 'Plutonium Plant') was designed literally from first principles of chemistry and chemical engineering and the fabrication of the equipment was taken up in-house to ensure effective quality control. Ultimately, the plant went 'hot' in August 1964 within 5 ½ years from the date of order – a significant achievement and the first button of plutonium metal was produced in August 1965. As a tribute to this path breaking achievement, 18th August is celebrated as “Reprocessing Day” at BARC.

Hon'ble Prime Minister (then) Shri Lal Bahadur Sastri dedicating the Plutonium Plant to nation (Year 1965)

Visit of Hon'ble Prime Minister (then) Smt Indira Gandhi at Plutonium Plant (Year 1967)

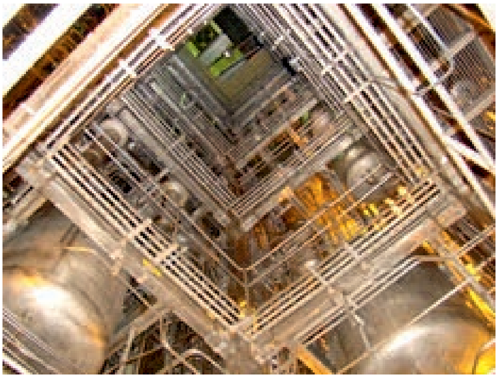
Initial challenges associated with reprocessing of spent nuclear fuel was addressed by Late Shri N Srinivasan in article of "IANCAS Bulletin, July 1998, Vol, 14, No. 2":

"While Canada India Reactor (CIR) was still under construction the formal order regarding the decision to set up a plant to reprocess irradiated fuel from it was issued. This was dated December 31, 1958. Apart from the historic nature of the decision it conveyed, the modalities set up for the implementation detailed in it were a veritable model for implementation of such path-breaking, scientifically challenging projects in a country which had been independent for only a decade. The freedom and flexibility in action allowed to the project management ultimately made it possible to complete the project within the sanctioned cost and the committed time schedule." [1]

A reprocessing flow sheet normally starts with a head end step commensurate with the fuel type, followed by complete dissolution of the spent fuel meat in nitric acid solution. The aqueous solution containing the nuclear material along with a plethora of radionuclides are subjected to solvent extraction cycles to separate and purify the uranium and plutonium from such undesirable elements. Such pure solutions are then subjected to a reconversion step to convert them into products meeting reactor specifications for their recycling into reactors. The PUREX based extraction process using 30% Tri-Butyl Phosphate (TBP) in n-Dodecane continues to be the heart of reprocessing plants, world over. While the TBP solvent preferentially picks up both uranium and plutonium, it is the partitioning agent which aids in their mutual separation. Replacement of ferrous sulphamate used during the earlier times with Uranous (U+4) is regarded as a special milestone, since the former would lead to choking issues in the plant leading to frequent down time of the plant. [2] Feedback generated from the plant was found very useful for construction and operation of successive plants. Although, formal regulatory body was yet to be established in the country, stringent safety review was carried out internally resulting in adherence of international safety stipulations with regard to radiation and industrial safety.

The experience of Plutonium plant, first reprocessing plant of India, resulted in to various developmental activities including process improvisation for better separation efficiency, solvent degradation and its management studies, analytical methods for analysing nuclear materials, corrosion studies and selection of better long lasting material of construction, which could be deployed in subsequent reprocessing facilities. Based on experience and technology development, second reprocessing plant of country, Power REactor Fuel REprocessing (PREFRE) plant with design capacity of 100 t/year, was built and commissioned at Tarapur site in year 1975 followed by Kalpakkam Atomic Reprocessing Plant (KARP), built and

PREFRE, Tarapur



Inside view of hot cell of reprocessing plant KARP

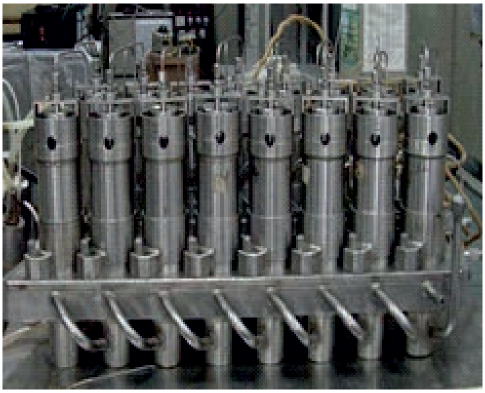
commissioned at Kalpakkam site for reprocessing of fuel from Madras Atomic Power Station. [3] This was followed by PREFRE II at Tarapur and more recently, another plant at Kalpakkam. The performance of these plants are rated at par with international standards both with regard to process performance and safety. The reprocessing process and flow sheet of PHWR fuel could be standardised for subsequent reprocessing facilities for enhancing the capacity.

With objective of bringing down the cost of fuel reprocessing and improving the economics of fuel cycle, high through put recycle facility, Integrated Nuclear Recycle Plant (INRP), is being constructed at Tarapur site. The plant is designed with 'Solid In Solid Out' concept with sharing of common utilities among various process block and thus minimising the overall cost. [4, 5] The valuable feed back of such operations of PHWR fuel reprocessing facilities were taken on board while addressing the closed fuel cycle for the upcoming recycle facilities for fast reactors. The challenges with regard to solvent degradation on account of high radiation dose and higher fissile material concentration are being addressed by deployment of centrifugal contactors. To meet the challenges of thorium based fuel cycle which is the nuclear fuel cycle of the future for India, R&D efforts are directed towards extractive metallurgy of thorium, fuel fabrication and its utilization in reactors, reprocessing of irradiated thorium for U-233 recovery and studies on U-233 based reactor systems. Demonstration facilities have been operated in all these domains and domain knowledge base is being built up for its implementation in the near future. [6]. In fact, India can claim to be one amongst the very few countries that have not only recovered U-233 from irradiated thorium/ thorium oxide, but also fabricated U-Al alloy fuel with the U-233 separated and commissioned a reactor (KAMINI at Kalpakkam) with this fuel as the driver fuel.

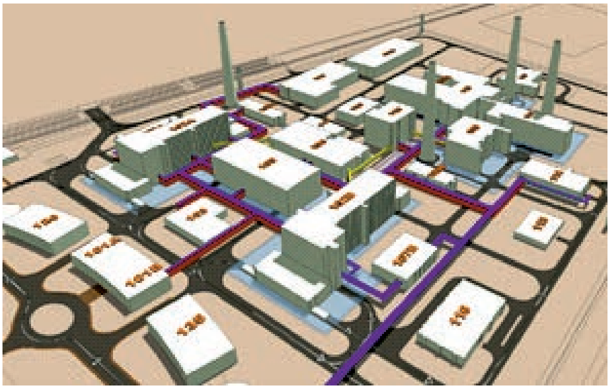
India has also built a Lead Minicell facility (CORAL) for reprocessing of uranium, plutonium mixed carbide fuel irradiated in Fast Breeder Test Reactor at Kalpakkam, and demonstrated recovery of plutonium from mixed carbide fuel irradiated to as high as 156 GWd/t. In fact, the Pu recovered was also used for fabricating fuel for FBTR subsequently, thus effectively demonstrating closure of fuel cycle. India is one amongst the very few countries in the world to have reprocessed mixed carbide fuel, and that too at high burn up and relatively short cooling.

Radioactive waste

Safe management of radioactive waste has been given prime importance since beginning of nuclear power program in India with objective to protect public, environment and future generation from harmful effect of radioactivity. Radioactive waste is generated at various stages



Centrifugal contactors used in CORAL plant



Conceptual image of integrated nuclear recycle plant of the nuclear fuel cycle, which includes the mining and milling of uranium ore, fuel fabrication, and reactor operation and spent fuel reprocessing. Besides these sources, radioactive waste is produced as a result of the ever-increasing use of radioisotopes in medicine, industry and agriculture. [7]

The essence of importance towards safe management of radioactive waste could be sensed from the address of Dr H N Sethna, Chairman, AEC (1972-1983) at IAEA General Conference in 1975:

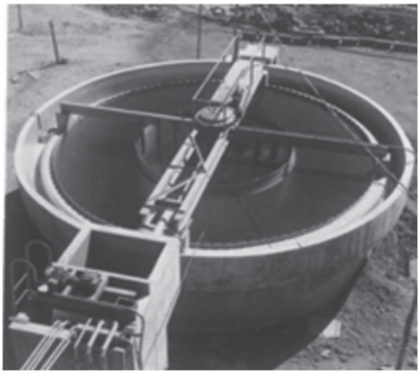
"The radioactive waste management in the Indian Nuclear Programme has continued to ensure that man and environment are not endangered to release of radioactivity....While we have worked on the basis of 'as low discharge as possible' as a practical reality, our current efforts are directed towards the concept of limiting discharge activity to the environment..."

Waste from Reactor Operations

Waste from Reactor operations normally fall into the category of low and intermediate level waste and all planning for treatment and management of such identified waste streams were carried out along with setting up of reactors in India. While considering the treatment of low active liquid effluent, the major challenges include very high waste volume and extremely low concentration of radionuclide which are aimed to be removed prior to discharge. In initial years, R&D was focused towards process technologies and plant operations for low level effluent treatment to meet the permissible environmental discharge limits. Chemical precipitation was developed as robust and effective processing techniques for low level effluent treatment for removal of radioactivity to meet the discharge limits. To date chemical precipitation is being used for low level liquid waste treatment in addition to various new membrane-based separation technologies have been developed and deployed for their plant level adoption.

The genesis of radioactive waste management in India was with the setting up and commissioning of an Effluent Treatment Plant (ETP) at Trombay in 1967 for treatment of low active effluent generated from CIRUS reactor with chemical precipitation as major treatment step for removal of radioactivity prior to discharge to meet discharge criteria. This plant continues to be operational and has adapted itself to low level effluents from reprocessing & other laboratories at Trombay as well. Operational experience of ETP, Trombay has gone a long way in designing and setting up of effluent treatment facilities for power reactor wastes and low-level effluents from reprocessing plants. [8]

Construction of Low level tanks at ETP (Year 1962)



Clariflocculator at ETP (Year 1966)

While India's first PHWR (RAPP I) was constructed and commissioned in active collaboration with Canadians at Rajasthan, Liquid Effluent Management Facility (LEMP) for this reactor was set up in parallel and commissioned solely as an indigenous effort. Though this facility continues to serve the intended purpose, it had to be augmented by a Facility based on Solar Evaporation, which is a preferred mode of evaporation for larger volumes of waste with low activity at sites, which have favourable climatic conditions such as high ambient temperature, low humidity and high wind velocity. In mid-seventies, augmentation of waste management facilities for TAPP I & II was carried out as the inbuilt radioactive waste system provided was found inadequate with respect to decay of Iodine and regenerant waste that required treatment. Accordingly, Tarapur Radwaste Augmentation plant (TRAP) was set up incorporating two numbers of 1000 cu.m. tank serving as delay tanks for I–131 followed by chemical treatment & ion-exchange. This augmentation plant has resulted in substantial reduction of activity discharge to the environment. Presently, all the Pressurised Heavy Water Reactors (PHWR) at various sites like Rajasthan, Madras, Narora, Kakrapar and Kaiga have dedicated waste management systems consisting of Liquid Effluent Segregation system (LESS), Treatment & Conditioning system (TDS), and waste disposal system (WDS) for discharge/ disposal of waste.

Similarly, practices for management of radioactive solid waste was also conceptualised well in early 1960's. Focus was mainly on safe disposal of low and intermediate level solid in early years. Concept of 'multi barrier disposal system' based Near Surface Disposal Facility (NSDF) was adopted right from beginning & has given greater assurance of isolation to retard the migration of radionuclide to environment. The multiple barrier could be combination of engineered as well as natural barrier. The preliminary work on low-level solid waste disposal started in 1962 at Trombay, where the first laboratories of radiochemistry and isotope production had started functioning. This was followed by setting up of near surface disposal facility at Trombay. In subsequent years, developmental efforts were focussed towards further minimisation of discharge following As Low As Reasonably Achievable (ALARA) principle. For limiting the discharges, polishing of the supernatant from chemical co-precipitation process was considered essential right from the design stage. This was achieved by metering the supernatant through ion exchange columns with Cs specific sorbent, e.g. natural vermiculite. However, same was discarded due to low loading capacity of sorbent in early 90's. Efforts were continued for development of high capacity sorbents with better decontamination effect. Two class of sorbents, i.e. synthetic zeolites and Copper Ferro-Cyanide (CFC) impregnated on suitable substrate, were considered for further developments considering the large volume and characteristics of low level effluent. Parallel developments include, membrane based techniques including reverse osmosis based process that has been developed and demonstrated on pilot scale for treatment of low level effluents in early 2000's. Same also could not be deployed in regular operations due to operational problems such as fouling of membrane, lower volume reduction factor and higher cost associated with replacement of membranes.

In the recent decade, efforts were further channelized to attain concept of 'Near Zero Discharge' of radioactivity with advancements in membrane technology. Zeolite 4A based ion exchange resin has been demonstrated for effective removal of radioactivity, by more than 5 folds, from low level effluent. In view of the simplicity of ion exchange process, a plant based on ion exchange process was installed at ETP. The process flowsheet is provided with sufficient flexibility to use improved sorbents in future.

Recently, developmental works in low level effluent treatment have been directed towards realise the 'Hybrid Process' consisting of various advanced membrane technologies in combination to reduce the radioactivity at such a low level that recovered water can be reused as 'process water'.

High Level Waste Treatment:

In view of the importance of reprocessing in the Indian context, the challenges that could be faced for the management of high level liquid waste was well recognized. Accordingly, R&D activities with respect to management of high level waste were initiated from mid sixties. Efforts were directed to develop and characterise a number of alternative glass matrices suitable for immobilisation of HLW generated as first cycle raffinate (PUREX process) from the Indian reprocessing units on one hand and develop, evaluate and perfect conditioning processes and techniques on the other.

In line with the international practice, a three-stage programme for the management of high-level waste was evolved.

1. Conditioning of the highly radioactive liquid wastes wherein radio-nuclides present in the high-level waste are immobilised in suitable matrices that is inert, highly durable (resistant to chemical/aqueous attack), and in turn contained in high integrity storage units which are subsequently overpacked.
2. Interim storage under surveillance and cooling of overpacks containing conditioned wastes for periods ranging up to 30 years to allow reduction in decay heat to a level acceptable for geological disposal on the one hand and to ensure integrity of the waste form and its packaging on the other before a commitment is made for their irretrievable disposal.
3. Disposal in deep underground repository such that at no stage potentially hazardous radioactive materials are recycled back to human environment in concentrations that can subject living beings to a risk considered unacceptable.

Development of vitrification technology for high level waste – a challenging task

Even though immobilisation of high level waste in glass matrix could be demonstrated at laboratory scale with desire product characteristics like very low leach rate, the challenge was to develop engineered systems and scale up the process at industrial scale required handling of highly radioactive and corrosive nature of high level waste at elevated temperature of 1000^0 C. This called for development of special equipment – a melter, which can process high level waste up to 1000^0 C for immobilisation in glass matrix. The process of immobilisation of high level waste in glass matrix is called vitrification. The process of vitrification essentially involves sequential drying of the high level waste, calcinations and conversion to glass/vitreous product, usually with the aid of specially formulated glass forming additives, depending upon the chemical composition of the waste. In this process most of the radio nuclides present in the waste get immobilized in the vitreous matrix as network modifiers and a few get physically trapped in the interstitial spaces in the bulk of the vitreous mass. Induction Heated Metallic Melter (IHMM) was developed for vitrification of high level waste at industrial scale utilising 'pot vitrification' process. IHMM consists of a pot of high nickel-chromuim based metal alloy, Inconel, for glass making process utilizing induction based indirect method of heating, for various stages of vitrification and to reach 1000^{O}C for making glass. Based on laboratory scale experiments and pilot scale trials, a plan was drawn up to set up a facility for further development of the process on a plant scale. The aim was to carry out a thorough evaluation of the process in conjunction with the needed auxiliaries and fine tuning of the same before constructing the facility for processing of actual radioactive wastes from a near by reprocessing unit on a routine basis. Thus, the first industrial scale vitrification facility i.e. Waste Immobilization Plant (WIP) was born at Tarapur in 1980s. The plant was designed based on 'single cell concept' containing all the process systems in single concrete shielded hot cell. Active trials of high level waste vitrification resulted in generation of very valuable feed-back for next facility. Some of the important feed backs were to incorporate 'multi cell concept' as compared to 'single cell concept', design improvisation of IHMM, deployment of robust remote handling techniques for remote maintenance and dismantling in the future etc.

Incorporating the feed backs, the second vitrification plant was set up at Waste Immobilisation Plant (WIP), Trombay, for treatment of high level waste arising from first reprocessing plant, Plutonium Plant, of India. The plant was designed based on multi-cell concept, provision of maintenance cell above the melter cell, improvised design of IHMM etc. The plant was inaugurated in the year 2002 by then Prime Minister of India, Shri Atal Bihari Vajpayee. Even though the plant was built with improvised design, different sets of challenges were associated due to legacy nature of high level waste, as high-level waste of Trombay was stored since first batch of spent fuel reprocessing at first reprocessing plant of India. Over the period, reprocessing flow sheet was modified considerably resulting in generation of varied composition of high level waste collectively store in single tank. The Trombay waste is characterised with high concentration of inactive salts along many problematic components, such as sulphate, and hence has difficulties in processing. As a result, a special glass matrix was needed to be developed to accommodate the components of Trombay high level waste. Quite substantial numbers of operations could be carried out successfully at WIP, Trombay. The operation of WIP, Trombay has given good amount of experience during vitrification of high level waste with respect to process operations with handling of large amount of radioactivity, remote handling systems, dismantling and remote assembling of melter system etc. [9]

Waste Immobilisation Plant, Tarapur

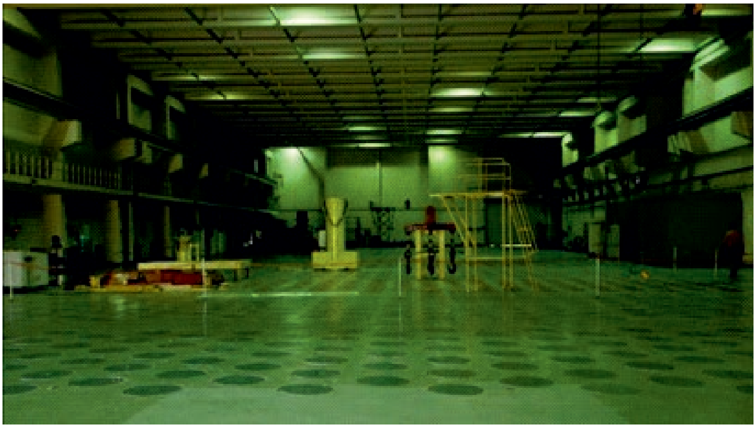
Hon'able Prime Minister Shri Atal Bihari Bajpayee inaugurating Waste Immobilisation Plant, Trombay (Year 2002)

In order to meet the higher throughput requirement, the second generation vitrification plants were set up based on Joule Heated Melter Technology (JHCM). This works on Joule principle of direct heating and offers higher through put than IHMM. This design has been deployed in subsequent facilities and has performed satisfactorily.

Joule Heated Ceramic Melter, Tarapur

High-level vitrified wastes are characterized by decay heat and need to be cooled to a level where transportation and disposal in geological repository become viable and economical. This period of cooling is also used to generate data on the product behaviour under constant surveillance and monitoring. These data are essential for prediction of long-term behaviour of the vitrified products. These requirements necessitate interim storage of vitrified waste containing overpacks spanning over 30 years and more.

The first Solid Storage and Surveillance Facility (SSSF) co-located with a vitrification plant was also conceived and constructed at Tarapur. This facility has been designed for storing vitrified canisters in overpacks for a period of about 30 years for ensuring decay heat removal. This is achieved by natural convective ventilation induced by a tall stack. This is an inherently self-regulating system and takes care of the changes in decay heat. The cooling system ensures that the temperature within the vitrified waste product, under no circumstances, exceeds softening point of the vitrified mass.



Solid Storage Surveillance Facility, Tarapur

Partitioning of High Level Waste – major breakthrough

Along with working on vitrification technology, back-end specialists in India also started looking at ways and means for reducing the radiotoxicity associated with high level waste. Partitioning & Transmutation strategy held the key to this. The Fast reactor programme that India had embarked on, could also be used for transmutation purposes along with the proposed ADSS. This propelled R&D in the field of partitioning of high level waste culminating in setting up and operation of India's first Actinide Separation Demonstration Facility at Tarapur in the year 2013. The feedback of the successful operations were utilised to set up another such facility at Trombay. While, ASDF demonstrated the very exacting task of actinide-lanthanide separation, the facility at Trombay helped in separating all the radionuclides from the inactive component of waste. This not only resulted multi fold decrease in volume for final disposal but also open an opportunity to harvest useful radionuclides for societal benefits. [10, 11]

Hon'ble President of India Shri. Pranab Mukherjee, dedicating Actinide Separation Demonstration Facility, Tarapur to the Nation (Year 2013)

Recovery of valuable radionuclide from waste

The partitioning strategy, as adopted at Trombay, led to the opportunity of separating radionuclides for societal benefits. Recovered ^{137}Cs from High Level Waste is converted into non-dispersive sealed source of Cs glass pencil for its application in blood irradiation. India is the first country to deploy ^{137}Cs in non-dispersive glass form for irradiation application. ^{90}Sr is recovered and purified for milking of clinical grade ^{90}Y for radiopharmaceutical applications. ^{106}Ru based eye plaques, RuBy, of different configurations have been developed, utilising ^{106}Ru recovered from radioactive waste, for affordable eye cancer treatment. [12, 13]. These radioisotopes are normally handed over for commercial use through BRIT (Board of Research in Isotope

Handing over of first consignment of Caesium glass pencils for blood irradiator to BRIT (Year 2016)

Handing over of first consignment of 10 Ru plaque to BRIT (Year 2019)

Technology)

Management & Disposal of Radioactive Solid Waste

Well characterised solid/solidified wastes conforming to regulations with regard to radioactive content and physical properties are only permitted for disposal. In the Indian context, since the country is fairly large, we have Near surface Disposal Facilities co-located with reactors at various identified sites. These are well engineered disposal modules which cater to waste containing only permissible limits of long lived actinides.

During initial stage, the focus was mainly on safe disposal of these waste in 'multi-barrier disposal system' at NSDF, since then innumerable improvements and requirements have been incorporated in design and construction of these disposal modules. Earth trenches, Stone Lined Trenches, Reinforced Concrete Trenches, Tile Holes are examples of engineered barriers. Mathematical models and their experimental validations for radionuclide migration from the disposal of radioactive solid waste from NSDF have been studied in detail to endorse the design of disposal facility ensuring no adverse effect to environment. Provisions for monitoring and surveillance are incorporated in the design of the disposal facility. Many NSDFs are under operation at various nuclear sites of India. A graded approach has been adopted for disposal of radioactive waste involving segregation and disposal of waste in different disposal module based on categorisation of waste. For example, low active category-I waste were disposed in earthen trench or stone lined trench, while intermediate active category II & III waste in reinforced concrete trench.

Vitrified waste containing long lived radionuclides are required to be disposed in Deep Geological Repositories. However, in the Indian context adoption of the closed fuel cycle and induction of cross cutting technologies with regard to partitioning of waste are ensuring that the need for Deep Geological Repository (DGR) are shifted by around a 100 year or so. In this regard, R&D activities with regard to site evaluation, waste-host interaction and environmental migration aspects are all being studied to accumulate a sufficient knowledge base for use at a later date whenever required. India presently does not have any Deep Geological Repository operational in the country.

As part of waste volume minimisation, design and operating practices were improvised to reduce the generation of solid waste and segregate them at source for their ease of management. Efforts were dedicated towards development of techniques for reduction of solid waste prior to disposal. Heavy capacity mechanical compactors have been developed and deployed for compaction of low active compressible waste to reduce the volume by about five folds. Incinerator system was deployed for incineration of low active cellulosic waste attaining volume reduction factor of about 30. The waste volume reduction techniques are still being utilised for reducing waste volume prior to disposal at NSDFs.

Recently, plasma-based incineration system has been developed and demonstrated for polymeric waste, contributing major fraction of low active waste, for waste volume minimisation. Developmental activities are focused to design the disposal module with optimum utilisation of space with desired quality of engineered barrier. [14]

Management of radioactive solid waste including safe disposal in NSDFs is under practice since early 1960's without any adverse effect to environment. Experience of more than six decades in the field of radioactive solid waste management and disposal has endorsed the design of NSDFs fulfilling the objective of safe disposal and also assured the capability of safe management of radioactive waste.

Conclusions

Recycling of spent nuclear fuel, as a long-term strategy has both environmental and resource advantage. In countries, where the energy needs have plateaued out, direct disposal of spent fuel is projected as an attractive option. Whereas in India, the need for effective utilization of limited resources along with environmental concerns necessitates adoption of reprocessing & recycling. Indigenous development of back-end technology and its maturing into standardized plants are ensuring sustainability of our Indian Nuclear Programme.

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