PARTITIONING OF HIGH LEVEL LIQUID WASTE

Operational Experience and the Way Forward

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ABSTRACT

The Partitioning & Transmutation (P&T) strategy is perceived as a step in the direction of reducing the radio-toxicity associated with high level liquid waste for final disposal. In this regard, the partitioning step is well recognized as the first approach towards the goal of reduction in radio-toxicity. The first industrial experience in setting up and operation of a completely indigenous Actinide Partitioning Demonstration Facility (ASDF) at Tarapur in the year 2013 has paved the way for this strategy to be inducted into our back-end plants[1,2]. The second such facility was set up and commissioned in 2015 at Trombay, which has not only led to separation of radionuclides from the inactive content of HLLW but has also made it possible for the use of few of the radionuclides for societal applications. Important among them are ^{137}Cs, which is separated and converted to glass rods for its use as blood irradiators[3] and ^{90}Sr, which is purified further and milked for ^{90}Y, which is being used for therapeutic application as 90Y-acetate. Very recently, ^{106}Ru so separated from waste has also been used for preparation of eye plaques for treatment of eye-cancer with very positive feedback. The ^{241}Am separated from the ASDF demonstration trials, is presently being envisaged to be used for the transmutation experiments using fast neutron spectrum and alternatively, as a heat source/RTGs for the Indian space application. The smooth and uninterrupted operation of both these facilities have strengthened our knowledge base for extending such technologies to more of our plants including that at Kalpakkam in the near future. This article deals with the operational experiences and lessons learnt during setting up and operation of such facilities.

KEYWORDS: Partitioning, High Level Liquid Waste, Fission products.

HIGH Level Liquid Waste (HLLW), getting generated from reprocessing of spent nuclear fuel, is conventionally immobilized in glass matrix to isolate the radionuclide from human environment. The vitrified waste is stored in air-cooled vault for removal of decay heat for a few decades prior to their disposal in deep geological repository. HLLW mainly contains fission products, minor actinides, corrosion products as well as various inactive chemicals added during reprocessing. Out of which, minor actinides are major contributors towards high radiotoxicity associated with HLLW in the far time line of final disposal. Fission products like ^{137}Cs and ^{90}Sr are some of the main contributors to heat source for final disposal, which dictate the overall size and design of deep geological repository. Industrial adoption of partitioning technology has demonstrated technological feasibility of splitting HLLW into heat generating fission product stream and long lived alpha product stream impacting final repository design. Another advantage that has been exploited in the near time line, is that, by separation of the radionuclides from HLLW and then subjecting the separated stream to vitrification can lead to a substantial reduction in final waste volumes for final disposal. This is due to the presence of inactive salts which are contained in HLLW due to reprocessing steps involving additives. This approach has yielded substantial volume reduction in case of legacy high level waste stored at Trombay. Extending this scheme to some of the stored HLLW can also lead to a sizable volume reduction factors.

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HLLW Partitioning Facilities in India

The ASDF facility (Fig.1) set up at SSSF, Tarapur, is meant for demonstration of ‘Partitioning of Minor Actinide from HLLW’. Among the minor actinides, Americium is generally considered a prime candidate for partitioning and transmutation because it is present in relatively large amounts and is a major contributor to radiotoxicity. The three-cycle solvent extraction-based process was demonstrated with regard to bulk separation of actinides along with rare earths followed by separation of trivalent actinides from the lanthanides. Table 1 gives the details of solvents deployed for three different cycles of ASDF. The 241-Am rich solution so separated was subjected to appropriate polishing process, followed by a reconversion step leading to obtaining AmO_2 having acceptable level of β-Y contamination (of <3 mCi/g of product). Based on the successful demonstration of this facility, another such partitioning facility was designed incorporating the feedback of Tarapur experiences.

The Trombay flow sheet, three cycle based solvent extraction process, addressed separation of inactive

Table 1: Solvent systems deployed at ASDF, Tarapur.

Solvent Extraction	Objective	Solvent Deployed
Cycle 1	U separation	30% TBP in n-Dodecane
Cycle 2	Actinide + Rare Earthbulk separation	0.2 M TEHDGA in 30% IDA in n-Dodecane
Cycle 3	Actinide - Lanthanide separation	0.4 M D2EHPA in n-Dodecane

Fig.1: Actinide separation demonstration facility, Tarapur.

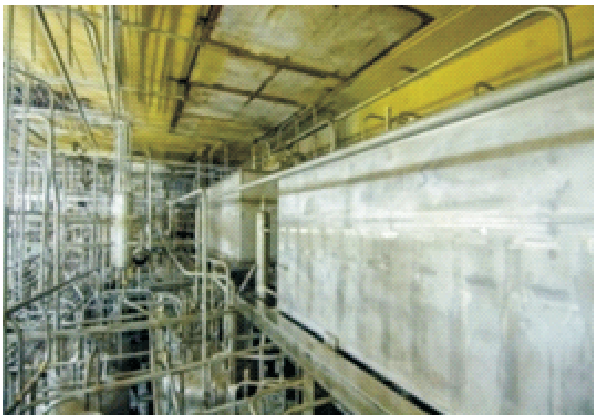

Fig.2: Solvent extraction system, Trombay.

components of waste from the radioactive elements, thereby precluding unwanted loading of vitrified glass with inactive components of waste, especially with regard to historic sulphate-bearing HLLW. In first cycle, residual Uranium and Plutonium are recovered from HLLW. While Cesium recovery is aimed in second cycle, Strontium-Actinide-Lanthanide combined separation is achieved in third cycle from HLLW. Table 2 gives the details of solvents deployed for three different cycles of solvent extraction system, Trombay. Fig.2 depicts the solvent extraction system installed at Trombay for pre-treatment of HLLW. The inherent composition of the waste stream resulted in only 250 L of waste being incorporated in 100 kg of glass matrix. By induction of the partitioning step, it is now possible to incorporate about 10,000 L of waste in 100 kg of vitrified product. As a spin-off of this program, it was also possible to recover a few fission products for direct use in societal application, namely ^{137}Cs, ^{90}Sr & ^{106}Ru. Fig.3 gives overall process schematic of solvent extraction process for treatment of sulphate-bearing Historic HLLW at Trombay.

Bulk Synthesis of Novel Extractant

The development of novel extractants, of desired quality, has been a key element for success of partitioning technologies in India. For example, selective recovery of cesium would not have been possible without the extractant named 'Calix Crown 6'. Similarly, extractants like TEHDGA and D2EHPA found necessary for separation of strontium, actinides & lanthanides from HLLW and partitioning of actinides from bulk of lanthanides respectively. Indigenous development of these extractants and its synthesis at large scale, to meet the requirement of engineering scale partitioning facilities, were the real challenge. Consistent R&D efforts in the field of development of extractants by nuclear chemists of BARC and participation of qualified vendors for its bulk synthesis with proper quality assurance program could realize the synthesis of these extractants at larger scale.

Air Lift Based Mixer Settler Unit

Air lift-based mixer settler units were selected as extraction equipment for the partitioning facilities as it imparts many benefits and better suits to desired applications. Air lift-based mixer settler unit gives flexibility in terms of number of stages for extraction, large range of organic to aqueous (O/A) ratios and flow rates, and also enable handling of varying densities of streams etc. Besides this, mixer settler unit does not require hot-cells having higher head room, as in case of air pulsed column, and helps in designing compact hot-cells. This further strengthens the selection of air lift-based mixer settler unit for said applications of extraction process. With the aim of eliminating all moving/maintainable parts of the mixer-settler, the agitator of conventional mixer-settler unit is replaced by an innovatively designed static mixing element CALMIX (**C**ombined **A**ir **L**ifting and **MIX**ing), which uses air to create high turbulence effecting the intimate contact of the two phases i.e., aqueous and organic. The CALMIX (Fig.4) has three identical square cross-section flow channel. One of them is the organic downcomer, which facilitates taking organic from top from previous settler irrespective of interface position. The other two flow channels are two identical annular flow path airlift created by locating airline at the center of flow channel. Both flow-channels get combined in the upper part of the mixing element which is similar to a vent pot with two inputs. This part is called mixing zone, where intense mixing takes place and the mixed phase along with air, are discharged through suitably sized holes provided in this region. As the energy for dispersion is supplied in the form of air, the liquid droplets do not experience shear and hence are easy to coalescence, resulting in a more compact settler design. The required contact time and the physical characteristics of the two phases form the design basis for the mixing element. The corresponding settlers are designed in a conventional manner. These units are provided with end-settlers to take care of any entrained phases in the terminal streams.

Table 2: Solvent Systems Deployed at Solvent Extraction System, Trombay.

Solvent Extraction	Objective	Solvent Deployed
Cycle 1	Uranium separation	30% TBP in n-Dodecane
Cycle 2	Cesium separation	0.03 M Calyx Crown(CC)6 in 50% IDA in n-Dodecane
Cycle 3	Strontium, Actinide & Lanthanide separation	0.4 M TEHDGA in 15% IDA in n-Dodecane

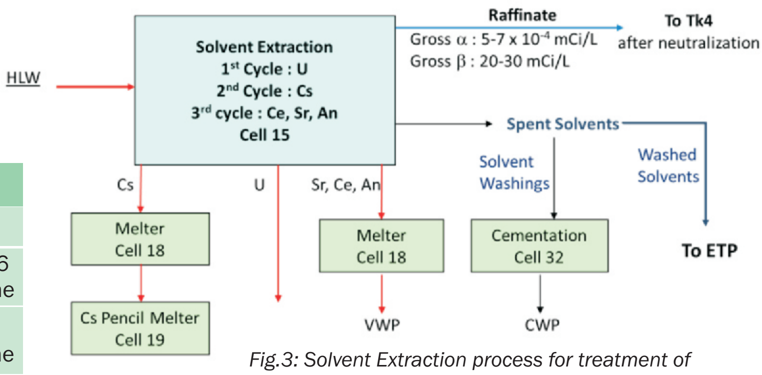

Fig.3: Solvent Extraction process for treatment of Sulphate bearing HHLW at Trombay.

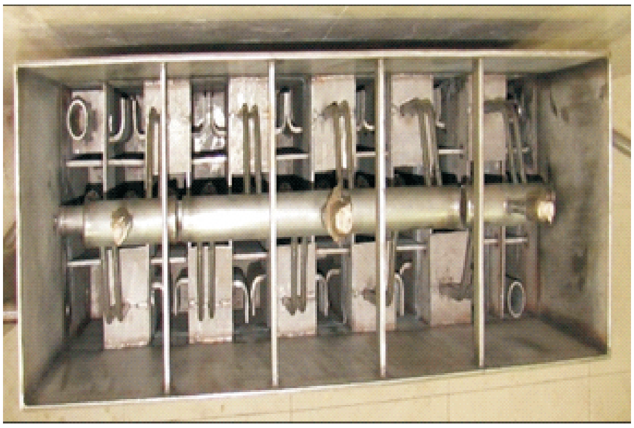

Fig.4: Combined Airlift based Mixer Settler Unit.

Design Considerations of Partitioning Facilities

Design & setting up of such operational facilities called for identification of robust engineering system to suit in-cell application and integration of such facilities to the vitrification plants. Setting up and operation of such facilities, being the first of its kind called for appropriate safety analysis and thorough documentation preceding a rigorous review process that culminated in obtaining regulatory clearances for these facilities.

In view of first time demonstration of partitioning technology, various features have been provided right from design stage to overcome unexpected challenges. Design provisions were made to address the uncertainties with regard to separation process. Recycle provisions were inbuilt in the system to ensure product purity with high decontamination factors. An elaborate solvent management provisions was worked out and incorporated leading to handling of multiple organic systems in the same cell. Focused attention was also given to secondary stream management including spent solvent streams.

Retrofitting of System in Existing Hot-cells

Both partitioning systems, of Trombay and Tarapur, have been retrofitted in existing hot-cells of facilities for demonstrating the technology of partitioning in the smallest possible time. Retrofitting of system with comprehensive design provisions in existing hot-cell is one of the major challenges. For example, three cycle solvent extraction based partitioning system is retrofitted in a hot-cell of WIP, Trombay having capacity to accommodate only one set of mixer settler units (one for extraction and one for stripper) and associated storage tanks. This necessitates use of single set of mixer units for three different solvent extraction cycles utilizing three different solvent system for recovery of varying radio-nuclide as mentioned above. The system needs to decontaminate effectively, to prevent cross contamination of radionuclide as well as solvents also, prior to change over to next cycle. Comprehensive design features and operating procedures are established to realize the desired decontamination of system. As a result, more than 100 m^3 of HLLW has been successfully treated for separation of radionuclides with desired purity fulfilling the objective of partitioning technology.

The Way Forward

Both the partitioning facilities have been operated successfully with actual HLLW demonstrating the partitioning technology at engineering scale. ASDF, Tarapur demonstrated the partitioning of minor actinides from HLLW by treating few tens of m^3 of HLLW for recovery of minor actinides of desire purity. Solvent extraction-based pre-treatment system at WIP, Trombay enabled effective treatment of more than 100 m^3 of HLLW with recovery of ~0.5 million Ci of Cs-137 and multi-fold reduction of vitrified waste volume to save precious repository space. Success of partitioning technology paves way for their deployment as pre-treatment step for facilities managing HLLW generated from reprocessing of power reactor spent nuclear fuel. Further focused efforts are also being directed towards secondary waste stream management including solvent management. Alternate and improved solvent systems are also being researched, especially for addressing the challenging task of An/Ln separation.

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