PROGRESS IN RADIOACTIVE EFFLUENT MANAGEMENT

Hybrid Process Towards "ALARA"

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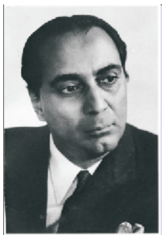
ABSTRACT

Management of radioactive effluents is vital to the success and acceptability of nuclear energy in public domain. Extensive research & development and five decades of operational experience at Trombay have resulted in standardization of the treatment methodologies. In order to follow ALARA philosophy, newer technologies like ion exchange and membrane processes have been evaluated in laboratory and pilot scale. A plant-scale ion exchange facility has been established. The use of membrane processes has also provided a firm foundation for demonstrating recycling of water recovered from LLW thus, proving the "Water Recycle" concept.

KEYWORDS: Radioactive effluent, Low level waste, Treatment, Ion exchange, Membrane.

THE safe management of nuclear waste has been a priority for this Research Centre from the early days of its inception. The importance placed on this area is evident from the extract of an order issued by our Founder-Director, Dr H. J. Bhabha.



"Radioactive materials and sources of radiation should be handled in the Atomic Energy Establishment in a manner which not only ensures that no harm can come to workers in the Establishment or anyone else, but also in an exemplary manner, so as to set a standard which other organisations in the country may be asked to emulate".

Feb.27,1960 ***-Homi Jehangir Bhabha***

Research and Development activities in radioactive waste management were initiated in parallel with ongoing research activities on Reactors and Nuclear Fuel Cycle. These activities eventually resulted in the commissioning of India's first plant for treatment of liquid radioactive waste *viz.*, Effluent Treatment Plant (ETP, then known as Waste Treatment Plant) at Trombay on July 26, 1966. The clarity of thought process is indicated from the fact that ETP was set out as facility to implement a "single discharge point" philosophy to control discharges, a model that is emulated even today. From the days of its commissioning, continuous endeavors for reduction of discharges to the environment were made. A quote from Dr. H. N. Sethna, second Director of this Research Centre, amply demonstrates this.

"...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 a discharge as possible' as a practical reality, our current efforts are directed towards the concept of limiting discharge activity to the environment..."

*-**H. N. Sethna**, Chairman, AEC (1972-1983)*
Address at IAEA General Conference, 1975.

This division has not only continued to follow the strategy of "limiting discharge" but, has over past five decades, progressed to the philosophy of "as low as reasonably achievable". It has now expanded it for the demonstration of "Water Recycle" concept.

Early Years

Even before CIRUS Reactor went operational, detailed studies were undertaken and discharge limits finalized. The early years of R&D and plant operations were focused on meeting the prescribed discharge limits and mastering the technologies for the entire cycle of low level radioactive waste (LLW) treatment[1]. Initial R&D activities had resulted in selection of a robust treatment process *viz.*, chemical co-precipitation for achieving initial aim of meeting discharge limits[2]. Subsequently efforts were directed towards optimizing the upstream and downstream processes of chemical co-precipitation. An example of this is that the plant, in response to operational challenges, changed its chemical co-precipitation sludge concentration techniques from rotary vacuum drum filtration to solar evaporation and finally to use of dewatering centrifuge. An overview of the LLW processing scheme is illustrated in Fig.1.

Towards "ALARA"

With the commissioning of new nuclear facilities in Trombay over the years, the challenges involved in LLW management has increased. Even as processes at waste

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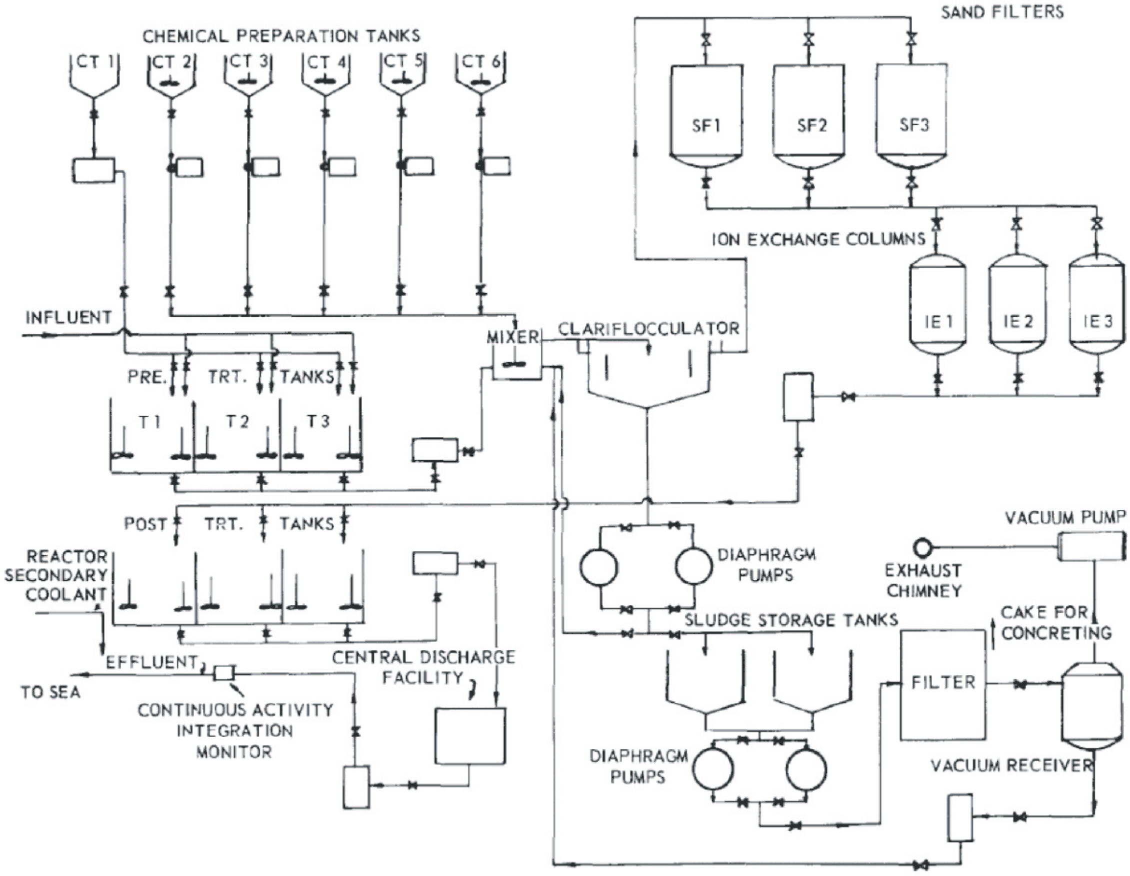

Fig.1: Process Flowsheet for Treatment of LLW.

generators improved, the overall quantum of LLW has increased along with the nature of wastes. The complexity of the LLW stream is highlighted by the fact that LLW is received from multiple facilities and even within a facility, widely differing streams ranging from process condensates, decontamination effluents, potentially active effluents, chemical wastes etc are generated. The broad characteristics of the LLW are given in Table 1.

For limiting the discharges, polishing of the supernate from chemical co-precipitation process was considered essential right from the design stage[3]. This was achieved by metering the supernate through ion exchange columns (Fig.2) filled with Cs-selective sorbent. The operating parameters of the column systems are also presented (left of Fig.2).

Due to the low loading capacity of the columns (500-800 bed volumes), the use of this sorbent was discontinued in the 1990s. Numerous laboratory scale studies were carried out to investigate possible high capacity sorbents as alternatives. Two classes of sorbents were explored and tested in laboratory and pilot plants. These were Synthetic Zeolites and Copper Ferrocyanide (CFC) impregnated on suitable substrates. Though CFC is a known high capacity Cs-selective material, due to its powdery form, it is not easily amenable to column operations. Impregnation on suitable substrates like anion exchange resins, polyurethane foam (PUF) and Zeolite 13X were explored over the years[4, 5, 6]. A 500 L scale ion exchange (IX) pilot plant using CFC-impregnated on anion exchanger was operated at WIP Trombay during the late 1990s for treatment of reprocessing effluents. Approximately 2400 bed volumes of LLW (1-5 x 10^{-4} mCi/lit) were passed through the column prior to its complete loading. Decontamination factor of 7-10 was obtained in the trial runs. The organic nature of substrate, non-regenerability and disposal considerations precluded use of this sorbent for regular operations. Pilot plants were also operated on 50 L scale for testing of CFC-PUF sorbents. Trial runs at Trombay indicated that 4000 bed volumes of LLW at 10 bed volume/hr were treatable with 10^{-4} mCi/lit as effluent quality. These sorbents, though promising, due to their low bulk density, would have resulted in large column sizes on plant scale considering the large processing rate requirement. CFC-Zeolite 13X and CFC-Zeolite 4A configuration were also tested on 50 L scale. Promising results were obtained with actual LLW. Loading of approximately 10,000 bed volumes at 10 bed volume/hr flow was obtained for few trials[7]. However due to non-reproducibility of results, further research is underway. Use of Zeolite 4A for selective separation of Sr was proven in

Table 1: Typical LLW Characteristics as received at ETP, Trombay.

Parameter	Value
Generation rate	50 to 1500 m^3/day
pH	8 - 10
Gross ß	10^{-5} to 2 x 10^{-2} mCi/L
Total Solids	Upto 1000 mg/lit

Overall Size: 1.82 m dia x 2.42 m ht	
Sorbent: Natural Vermiculite	
Type: Non-regenerative	
Size: -20 to +30 ASTM Mesh Size	
No. of Columns: 4	
Quantity of Sorbent: 4000 kg/Column	

Fig.2: Operating characterisitics of Cs-sorbent column (left) and a 1970s file photograph of the same (right).

Fig.3: Self-shielded Ion Exchange columns.

Fig.4: Valving station of Ion Exchange system.

laboratory scale earlier. However, the sorbent also shows selectivity for Cs too. When operated at 50 L scale, loading of minimum 7000 bed volumes was obtained consistently at 10 bed volume/hr flow. Thus, after trials involving approximately 2500 m^3 of LLW treatment, this sorbent was chosen as a primary material. In view of the simplicity of ion exchange process and large body of scientific work in this area, a plant (Figs.3 and 4) based on ion exchange process was installed at ETP. The process flowsheet is provided with sufficient flexibility to use improved sorbents in future.

Towards "Water Recycle"

An alternate process involving use of membranes showed significant promise in its use for LLW treatment on laboratory scale in early 2000s. With the commercial availability of reverse osmosis membranes on the rise, during the period from 2009–2015, thin-film composite reverse osmosis membranes in spiral-wound and disc-tube configurations were operated in three campaigns at Trombay in consultation with Desalination Division. Actual LLW was used in these pilot plants[8]. A total of 33,00,000 L of LLW was processed. These runs provided valuable insights into usability of the membrane process on plant scale. Throughout its operation, effluent specific activity of <1 Bq/ml was consistently obtained. This is three times lower than the effluent quality with ion exchange. The trial runs proved that efficient pre-treatment technique is vital to operation of RO plants especially when mixed effluents, as generated at Trombay, are encountered. Conventional pre-treatment techniques like sand & activated carbon filtration and two-stage microfiltration was found to be ineffective for Trombay effluents as it choked the membranes and rendered in unusable. Ultrafiltration was found to be the most promising pre-treatment technique for RO systems with mixed wastes as feed material. Effluent streams, which led to long life of the membranes, were identified and qualified in pilot plant runs. Polishing of this very low active stream by mixed bed resin rendered the effluent specific activity to below detectable levels. These observations indicated that LLW could be decontaminated to a level where its use as "process water" could be conceptualized. A pilot plant for "Water Recycle" has been installed at Trombay for "proof of concept" demonstration. This can eventually pave way for recovery and reuse of water from LLW for large nuclear facilities planned in the country. This will further strengthen and extend the philosophy of "Recycle & Reuse" being followed in Nuclear Recycle Programmes.

Conclusions

During the period from 1966 till date, mastery over the technologies for low level liquid radioactive waste treatment has been achieved leading to standardization of process flowsheet. Without relying on laurels, continuous efforts towards research and development to further reduce discharges are underway. An Ion Exchange Plant at ETP Trombay will be a test bed for some of these promising

Fig.5: Ultrafiltration Pilot Plant.

Fig.6: Spiral RO Pilot Plant.

Fig.7: Disc Tube RO Pilot Plant.

sorbents in the future. The R&D in membrane processes carried out over the years has provided confidence for demonstration of concept of "Water recycle" which will be the need of the hour in near future. India thus, can be world leader in emulating the philosophy of "Wealth from Waste".

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