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Polishing of spent fuel storage bay, dhruva water using synthetic zeolites

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ABSTRACT

Spent Fuel Storage Bay at Dhruva Research Reactor is meant to store spent fuel from the reactors for cooling purposes. Activity level and quality of the pool water is maintained within prescribed limit. Activity levels of the pool water were found to be much higher than the permissible level. Samples of the contaminated water were withdrawn from the SFSB and studies were carried out for characterization and selection of treatment method. Based upon the results obtained for chemical and radiochemical analysis of the pool water and from earlier experiences for treatment of such type of radioactive waste liquid, mixture of two different grades of synthetic zeolites i.e., 4A & 13X were selected for use in decontamination of the pool water. A 5 ml column bed was prepared by mixing synthetic zeolite 4A (90%) and 13X (10%). The feed solution was passed at a flow rate of 12Bvs/h. Samples were collected periodically and analyzed for gross beta as well as radionuclides. The column operation was terminated at 1% breakthrough with respect to radio cesium was achieved. The results revealed that gross beta activity can be brought down to <20 Bq/ml from 300–520 Bq/ml, no radiocesium or radiostrontium was detected in the effluent till 7000 bed volume. The effluent activity is mainly due to ^{125}Sb, the radionuclide which was not picked up the zeolites. Thus a mixed bed of synthetic zeolites 4A and 13X can be used efficiently to treat the pool water from SFSB, Dhruva.

Keywords: Spent Fuel Storage Bay; Synthetic zeolite 4A; Synthetic zeolite 13X; Radiocesium; Radiostrontium

1. Introduction

All the reactors and reprocessing plants are provided with spent fuel storage bay (SFSB) facilities for storing spent fuel in water for cooling purposes. The SFSB at Dhruva is having a total inventory of 1000 m^3 of demineralised water (DM) water. The pool water is continuously polished by circulating through a mixed ion exchange resin bed (Cation 350 l and Anion 300 l) to maintain the purity and clarity of pool water within the prescribed values i.e., Conductivity: <2 micro Siemens, pH: 5.5–7, Turbidity: <2 NTU. In addition, to this activity level is also kept below the prescribed limit i.e., <100 Bq/ml. It has been observed that sometimes the activity level in SFSB increases due to higher incidences of failed fuel as well as due to the nature of failure. In such cases the mixed bed alone is not able to bring down the activity level within the prescribed limit. Thus, an additional polishing unit is required to bring down the activity level. As the activity is mainly due to radio cesium and radio strontium, a bed of resin which selectively take these radionuclides can bring down the activity level below permissible level. A large number of inorganic and organic ion exchangers such as cobalt hexacyanoferrate, copper hexacyanoferrate ferrocyanate, resorcinol formaldehyde, ammonium molybdophosphate, zeolite etc.

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for cesium and poly antamonic acid, hydrous titania, imino diacetic acid, crown ether, zeolite etc. for strontium can be used for treatment depending upon the characteristics of waste. Synthetic zeolite has been chosen for the purpose due to following reasons [1].

- High selectivity for cesium and strontium.
- Easy availability in commercial quantity.
- High loading capacity.
- Compatibility with the waste i.e., no pretreatment is required.
- Good column properties.
- Compatibility of loaded zeolite with cement matrix.

The paper deals with the studies carried out in the Effluent Control Laboratory at Effluent Treatment Plant, Trombay for treatment of the contaminated water using inorganic ion exchange process.

2. Experimental

The contaminated pool water sample was withdrawn from the SFSB. The sample was characterized with respect to different chemical parameters such as pH, TDS using standard methods. The sample was also characterized for radio-chemical parameters such as gross beta measured by using GM counter, gamma radionuclides using HPGe detector coupled with multichannel analyzer, radio strontium by radiochemical analysis. Results obtained are given in Table 1. Based on our earlier experience and characteristics of pool water (Table 1), it was realized that synthetic zeolites are suitable sorbents for the treatment of such type of contaminated water. Detailed studies on synthetic zeolites indigenously available commercial grades with respect to their structure, selectivity for specific radionuclides, thermal and radiation stability, ion exchange capacity etc. and also their application in waste management have already been reported [2–7].

Table 1
Chemical and radiochemical characteristics of contaminated water from sfsb, dhruva

S.No.	Parameter	Value
1.	pH	8.1
2.	TDS (ppm)	100
3.	Gross β (Bq/ml)*	300–520
Average %age of various radionuclides		
^{137}Cs, ^{134}Cs,		85%
^{95}Zr, ^{144}Ce, ^{95}Nb		3%
^{125}Sb		7%
^{90}Sr		5%

*Gross beta varies with the samples received at different intervals.

Two different grades of synthetic zeolites 4A and 13X were procured from Indian Petro Chemical Corporation Ltd. (CATAD Division), Baroda in the form of cylindrical pellets of length 1–5 mm and diameter of 1.5 mm. The samples were grounded and sieved to grain size of 0.3–0.6 mm, washed free of fines with distilled water and dried at ambient temperature (30°C). Although both the zeolites were procured in sodium form still to ensure 100% sodium form, they were equilibrated with excess of 1.0 M NaCl solution overnight, washed free of chloride with distilled water and once again dried at ambient temperature. They are finally stored in a desiccator containing saturated solution of ammonium chloride to maintain uniform moisture content and were used for experimental studies.

As indicated in the Table 1, 90% of the activity is due to radiocesium and radiostrontium, remaining 10% is due to other radionuclides, thus the column bed of mixed zeolites was used for the studies. The column bed was prepared by mixing zeolite 4A (selective for Cs & Sr) and 13X (for other radionuclides such as ^{95}Zr, ^{144}Ce, ^{95}Nb) in the ratio 90% and 10% respectively. Measured volume (5 ml) of mixed zeolites was packed in a glass column with height 15 cm and internal diameter 10 mm. The feed solution was passed through the fixed bed by using peristaltic pump at a constant flow rate of 12 bv/h (60 ml/h). Samples of effluent were taken periodically and analyzed by radiometric techniques. Gross beta activity and radio nuclides present in the effluent samples was determined. Feed water was periodically made available from SFSB, Dhruva. The column operation was terminated after achieving 1% breakthrough with respect to radiocesium. Total volume passed at the time of termination was about 8500 bed volumes.

3. Results and discussions

The results indicated in the Table 1, showed that 90% of the gross beta activity is due to radiocesium and radiostrontium and remaining 10% is due to other radionuclides such as ^{95}Zr, ^{144}Ce, ^{95}Nb , ^{125}Sb.

Results of column run shown in Table 2 indicated that, initially the effluent activity is <20 Bq/ml and is mainly due to ^{125}Sb. The column has removed radio cesium and radio strontium completely. Activity of the effluent sample increases after passing 7000 bed volumes and the analysis of the effluent sample showed the presence of radio-cesium (^{137}Cs + ^{134}Cs) also. The column operation was terminated at 1% breakthrough with respect to radiocesium (^{137}Cs + ^{134}Cs) which was obtained after passing 8500 bed volumes (42.5 l). The radiation dose on the column was measured periodically and at the time of termination it was 1mGy/h. As activity does not leach from the loaded zeolite [2]. Also being alumino

Table 2
Results of column operation. Bed volume: 5 ml (4.5 ml 4 A and 0.5 ml 13X zeolite). Flow rate: 12 Bvs/h. Grain Size of zeolite: 0.3–0.6 mm

S. No.	Bed volume passed	Radionuclide	Gross β (Bq/ml)
1.	1000	^{125}Sb	11
2.	2000	^{125}Sb	11
3	3000	^{125}Sb	14
4.	4000	^{125}Sb	14
5.	5000	^{125}Sb	17
6.	6000	^{125}Sb	17
7.	7000	^{125}Sb	17
8.	8000	^{125}Sb, ^{137}Cs, ^{134}Cs	22
9.	8500	^{125}Sb, ^{137}Cs, ^{134}Cs	25

silicate, zeolites are very much compatible with cement, thus loaded zeolite can be disposed off either as such or after immobilization into cement matrix [8].

4. Conclusions

Synthetic zeolite of Indian Origin can be successfully use for the treatment of contaminated water of SFSB with very high column throughput.

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