TREATMENT OF RADIOACTIVE AQUEOUS SOLUTIONS BY ADSORPTION ON FLY ASH AND SYNTHETIC ZEOLITES

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Liquid radioactive waste is one of the most difficult forms of waste to manage due to its mobility, ability to spread in the environment and complex chemical composition. Immobilization - the process of converting liquid waste into stable solid forms - is a key step in the management strategy of this waste.

Sorption-based processes offer useful and effective approaches for converting solutions into solid forms and immobilizing waste streams. The selection of appropriate sorbents depends on waste characteristics, regulatory requirements, economic considerations, and long-term stability needs.

Zeolites effectively immobilize liquid radioactive waste through their porous crystalline structure that enables selective ion exchange, particularly for Cs-137 and Sr-90. Their high sorption capacity, radiation stability, and compatibility with solidification matrices make them ideal for nuclear waste treatment. Both natural and synthetic zeolites are used in power plants, accident remediation, and waste decontamination by passing contaminated liquids through zeolite columns before final solidification.

Synthetic zeolites are primarily manufactured via hydrothermal synthesis, combining silica and alumina sources with alkali hydroxides in autoclaves (60-200°C). Production requires precise control of temperature, pH, and composition, followed by washing, ion exchange, calcination (500-600°C), and milling.

Major commercial varieties include types A, X, Y, ZSM-5, and Beta, each optimized for specific applications.

Synthesis of the zeolites

The Na-A, Na-P1, and Na-X zeolites were synthesized from the milled fly ash and the NaOH solution. To decrease the molar ratio of Si/Al, the Na-A zeolite was produced using an additional portion of the aluminium foil. Based on the previous work performed in the group of Prof. Franus [1], the process required:

- ✓ **Na-A zeolite:** 3 M NaOH solution;
- ✓ **Na-P1 zeolite:** 2 M NaOH solution;
- ✓ **Na-X zeolite:** 3 M NaOH solution.

The resulting suspensions were autoclaved for 24 hours at the crystallization temperatures established earlier for each material (from the range of 40-140 °C). Then, an excess of the alkaline solution was removed, and the final material was washed extensively with the distilled water followed by drying at 105 °C.

[1]. W. Franus, *Characterization of X-Type Zeolite Prepared from Coal Fly Ash*. J. Environ. Stud. **2012**, 21, 337–343.

Experimental

Sorption tests consisted of spiking the working solution with the radionuclides forming the carrier-free concentration and adjusting it to the desired pH. Except for the sorbent mass dependence experiments, 50 mg of the zeolite placed in the polyethylene test tubes was mixed at ambient temperature with 5 mL of the radioactive solutions by shaking for 3 hrs. In the following, the suspensions were separated by centrifuging, and the aqueous phases were withdrawn for measurement of the specific concentration of the radionuclides.

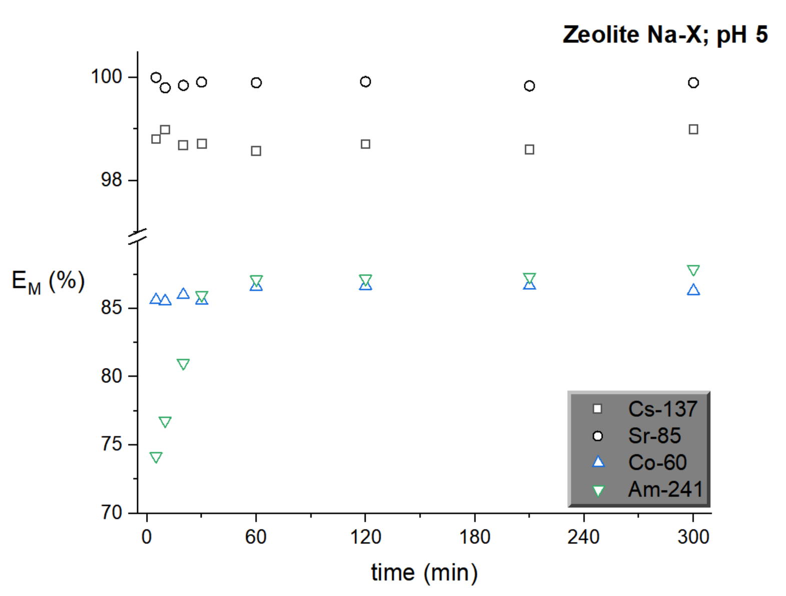
Mono-, di- and trivalent radionuclides (namely: Cs(I)-137, Sr(II)-85 and Am(III)-241) have been studied.

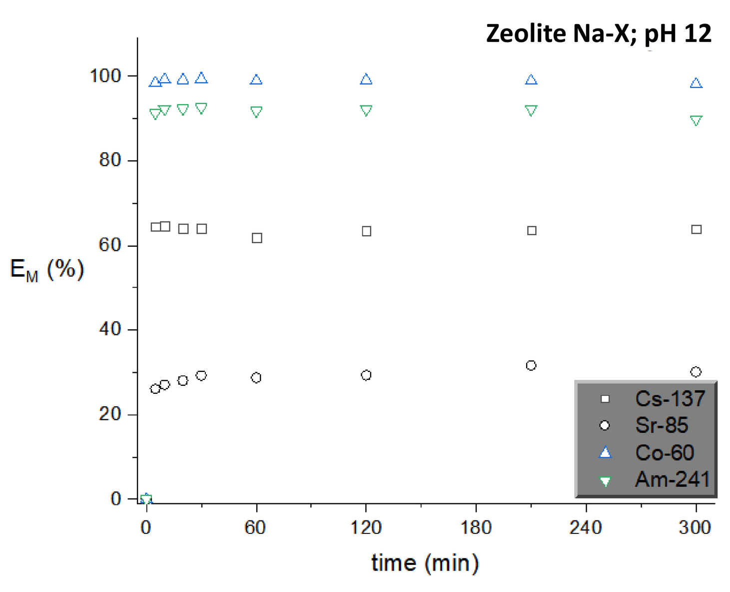
Co(II)-60 was also been examined as a representative of the corrosion products.

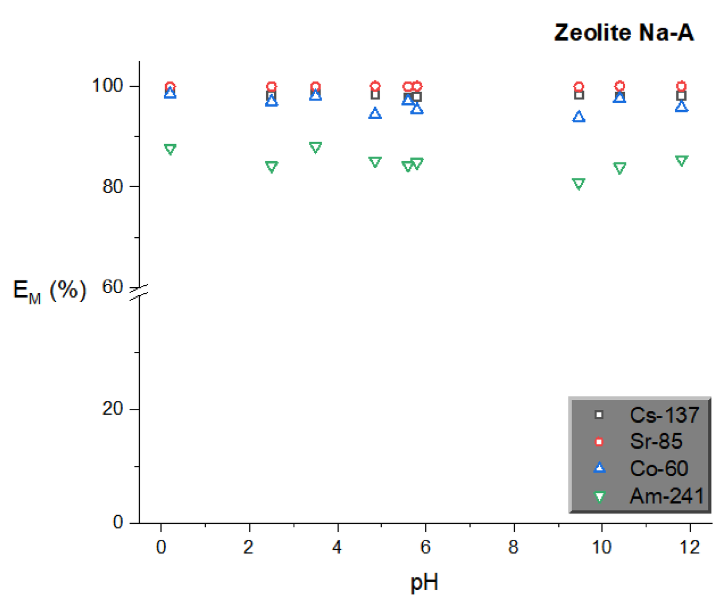
The possibility of removing Tc-99, which occurs in aqueous solutions in anionic form, was also studied.

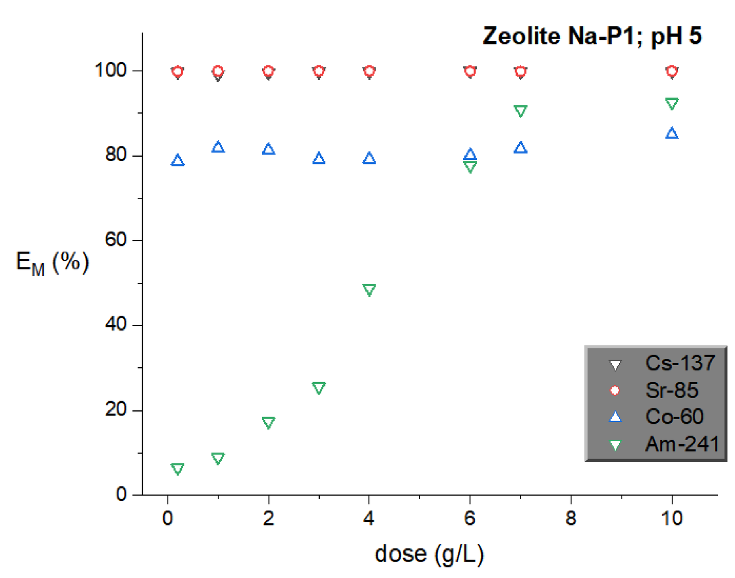
Results

In the static (batch) sorption experiments of removal of the radionuclides from aqueous solutions, for all the potential sorbents, we have determined the following parameters: dosage of the sorbent, time of contacting the phases, acidity of the solution and the salinity of the solution. Selected results are presented in the Figures forming the first line:

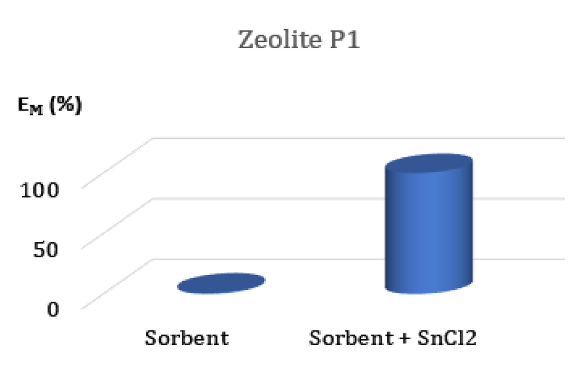


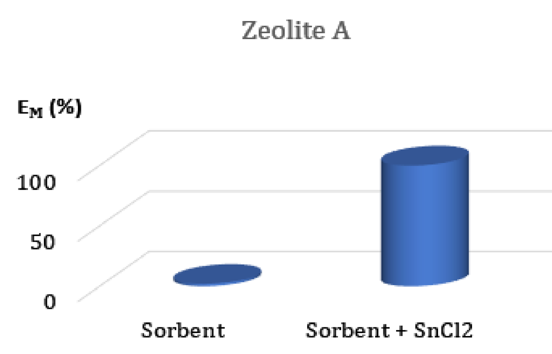


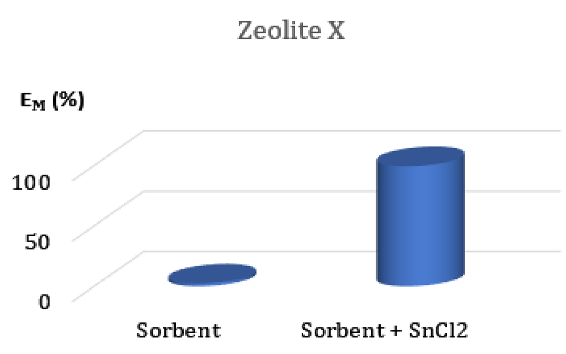




Removal from aqueous solutions of Tc-99m (an analog of dangerous Tc-99) is possible only after its reduction to the cationic form.







Conclusions

The obtained results show, that static (batch) adsorption of radionuclides using the fly-ash-based zeolites may be used to uptake cationic radionuclides, such as Cs^+, Sr^{2+}, Co^{2+} and Am^{3+}, from water. Radionuclides present in an anionic form must be reduced to the cationic form prior to sorption.

However, careful selection of the type of the zeolite must be done for any solution to be purified because the magnitude of sorption of each metal depends on the type (structure) of zeolite.