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Uptake selectivity for different types of zeolites in the presence of boric acid

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Introduction

The development of selective adsorbents is very important subject for the effective multi-nuclide decontamination related to the severe accident of Fukushima NPP-1. From the stand point of cost efficiency, stability for usage and safety handling, the decontamination method using zeolites is one of the most effective methods for the selective decontamination system. Thus in this study, several kinds of zeolites are tested through batch experiment in the presence of boric acid for a decontamination method in PWR-NPP.

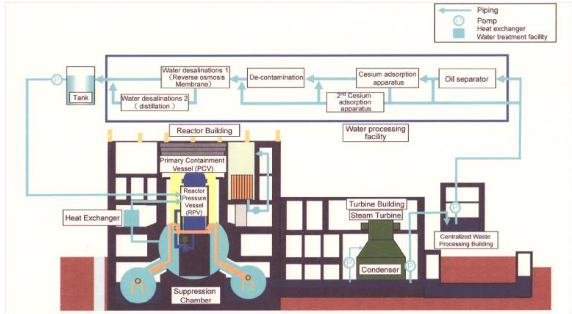


Fig. 1. Decontamination system for high-activity-level water.

Objectives

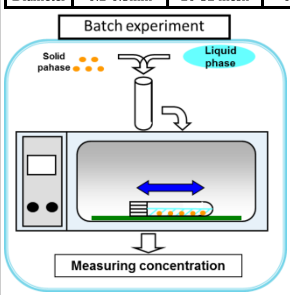
1. Check the uptake behavior of different kinds of zeolites
2. Develop and evaluate the adsorbents with adsorption performance for Iodine
3. Get the detail uptake information for several nuclides under Boric Acid

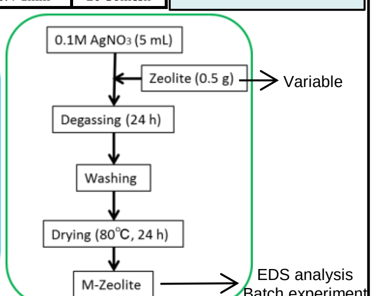
Materials and Preparation Procedure

Zeolite	ZeoliteA	ZeoliteX	ZeoliteY	ZeoliteL
DM	500 µm	500 µm	500 µm	500 µm
Diameter	0.5mm	20-32 mesh	20-32mesh	20-32mesh
Zeolite	Chabazite	SM	NM	CP
DM	500 µm	500 µm	500 µm	500 µm
Diameter	0.2-0.5mm	20-32 mesh	0.4-1mm	20-30mesh

Condition
1: DW (3,000 ppm H_3BO_3 + 10 ppb LiOH)
2: Sea water 30% (3,000 ppm H_3BO_3 + 30% sea water)
3: Sea water (3,000 ppm H_3BO_3 + sea water)
4: 0.1M NaCl

V/m= 100 / 300
Shaking time: 24 h
Temperature: 25°C
Career concentration: 1 ppm / 10 ppm / Free





Simple Nuclide Experiment ($^{60}Co^{2+}$, $^{85}Sr^{2+}$, $^{137}Cs^{+}$ and $^{152}Eu^{3+}$)

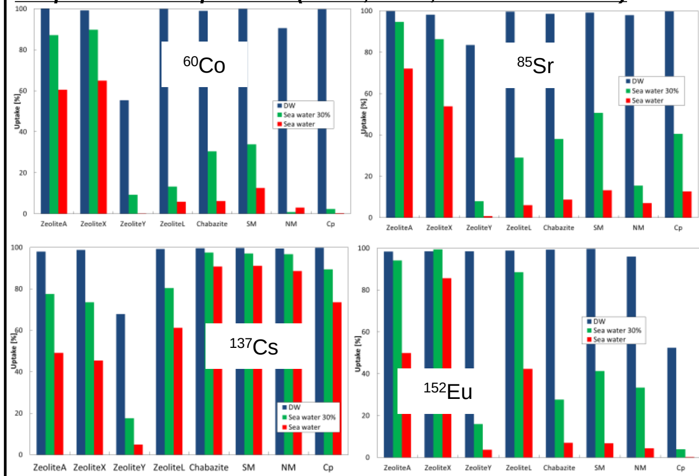


Fig. 2. Uptake performance of different kinds of zeolites.

1. Under the DW, most of the zeolites shows high adsorption performance for ^{85}Sr, ^{60}Co, ^{137}Cs, ^{152}Eu.
2. As the concentration of sea water is increase, uptake of some zeolites decrease due to the difference of selectivity.
3. To adsorb Sr and Co nuclides, zeolite A and X is effective under the condition with seawater.
4. To adsorb Cs nuclides, chabazite, mordenite and clinoptilolite is very effective even the concentration of sea water is very high.
5. Zeolite A, X and L show relatively high uptake (%) for Eu nuclides.

Development and Evaluation of the adsorbents for I⁻

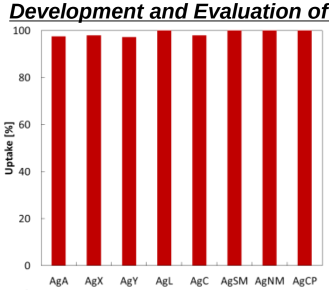


Fig. 3. Uptake performance for I⁻ by M-zeolites

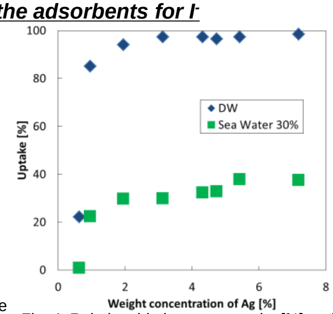


Fig. 4. Relationship between uptake [%] and weight concentration of Ag [%] on NM

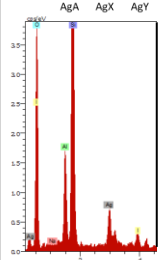


Fig. 5. EDS spectra of Ag-NM

1. Every Ag-zeolites showed very high uptake (%) for I⁻ under DW condition (more than 98 %).
2. Content of Ag increased up to 7 wt% with decreasing the added amounts of NM.
3. If the weight concentration of Ag is more than 2 wt%, the uptake (%) keeps more than 95%
4. If the weight concentration of Ag falls below 1 wt%, the uptake (%) was markedly dropped.
5. The uptake of I⁻ was lowered in 30% seawater due to the coexistent ion like Cl⁻.

Simple Nuclide Experiment ($^{239}NpO_2^{+}$)

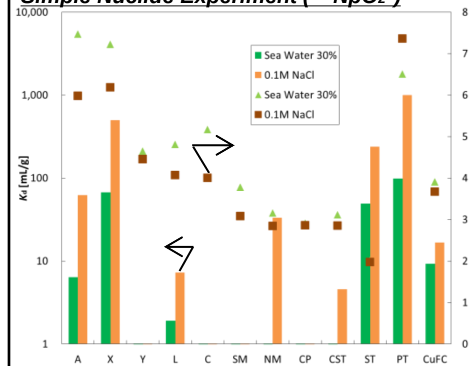


Fig. 6. K_d value for NpO_2^+ of different kinds of adsorbents.

CST: Crystalline silicotitanates
ST: Tin antimonate
PT: Potassium titanate
CuFC: Microcupsuls with insoluble ferrocyanides

1. In the zeolite group, zeolite X showed the highest K_d value.
2. However, the K_d value is very low (68 mL/g) under the presence of Seawater.
3. PT had eminent adsorption performance under 0.1 M NaCl condition.

Multi –Nuclides Experiment

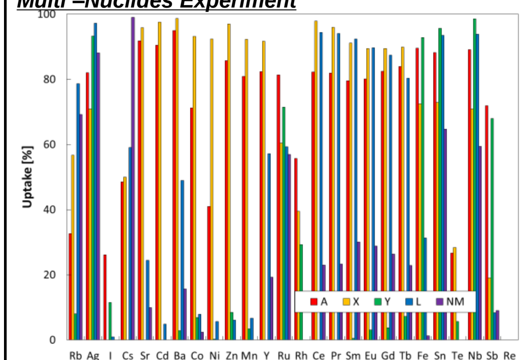


Fig. 7.Uptake selectivity of different types of zeolites .

These elements were referred to the nuclides in ALPS (Advanced Liquid Processing system) in Fukushima NPP-1.

1. Zeolite A and X shows high adsorption performance for divalent ions like Sr^{2+} and Co^{2+}.
2. Zeolite L shows high selectivity for trivalent ions like Ce^{3+} and Eu^{3+}.
3. Mordenite shows high selectivity for monovalent ions like Cs^{+}.
4. Zeolite Y seems to adsorb nuclides with high valence ions like Sn(IV) and Nb(V).

CONCLUSIONS

1. Mordenite has high adsorption performance for monovalent metal ions. Zeolite A has high adsorption performance for divalent metal ions. In contrast, zeolite L has high adsorption performance for trivalent metal ions. These uptake tendencies are similar to those under the condition without Boric Acid.
2. Ag-zeolites with high adsorption performance for I⁻ were developed.
3. Ag-NM has a high adsorption performance for I⁻ in DW, while the uptake of I⁻ in sea water was markedly lowered.
4. Zeolite X showed relatively high K_d value for NpO_2^+ under 0.1M NaCl condition (495 mL/g) even though the K_d value under 30 % sea water condition is low (68 mL/g).
5. Cs can be removed from waste water containing sea water very easily by using mordenite or chabazite even under sea water atmosphere.

Zeolites are effective for the selective decontamination system for PWR-NPP considering the difference in their adsorption properties.