

## 学位論文要旨 Dissertation Abstract

氏名 : Moses Wazingwa Munthali  
Name

学位論文題目 : Studies on Cation Adsorption Properties of Zeolites and the  
Title of Dissertation Development of Radioactive Decontamination Method Using  
Zeolites (ゼオライトの陽イオン吸着特性及びゼオライトを用いた放

射能除染法の開発に関する研究)

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Adsorption behavior of  $H^+$ ,  $Li^+$ ,  $Na^+$ ,  $K^+$ ,  $Rb^+$  and  $Cs^+$  ions on  $Li^+$ ,  $Na^+$ ,  $K^+$ ,  $Rb^+$  and  $Cs^+$ -saturated Linde-type A, Na-P1, mordenite, X type and Y type zeolites were evaluated at different pH-pM, where pH-pM is equal to  $\log \{(M^+)/\{H^+\}\}$  and  $M^+$  represents either  $Li^+$ ,  $Na^+$ ,  $K^+$ ,  $Rb^+$  or  $Cs^+$ . The cationic adsorption selectivity sequences on zeolites were also determined. The method for radioactive decontamination were developed for radioactive  $Cs^+$  and  $Sr^{2+}$  decontamination from water and soil using zeolites. In case of water  $Cs^+$  and  $Sr^{2+}$  were coexisted with different concentrations and types of competing cations. While for soil, the experiments were done by coexisting zeolites with contaminated soil in the presence of the different types of extracting salt solutions. Different variables such as solid/solution ratio, time, concentration, pH, zeolite species and method of coexistence were evaluated.

Generally, the results indicated that with decreasing pH-pM, the amounts of the alkali metals retention decreased due to the adsorption of  $H^+$  via cation exchange reaction. Adsorption selectivity of  $H^+$  to zeolites had a negative correlation with Si/Al ratio of zeolites. Although each zeolite has a constant amount of negative charge, the amount of  $M^+$  adsorption of each zeolite decreased drastically at low pH-pM values. By using the plot of the amount of  $M^+$  adsorption versus pH-pM, an index of the  $H^+$  selectivity, which is similar to pKa of acids, of each zeolite was estimated, and the index tended to increase with decreasing Si/Al ratio of zeolites. These results indicated that zeolites with lower Si/Al and higher negative charge density have higher  $H^+$  adsorption selectivity than with high Si/Al ratio and lower cation exchange capacity (CEC), and in fact, such zeolite species (4A and X) adsorbed considerable amount of  $H^+$  even at weakly alkaline pH region. Furthermore, in each zeolite species,

Cs<sup>+</sup>-saturated zeolite showed the lowest H<sup>+</sup> selectivity, and this suggested that Cs<sup>+</sup> had the strongest adsorption energy in the alkali metal cations. The adsorption of H<sup>+</sup> was strongly affected by diameter and hydration energy of the alkali metal cations, and also affected by framework type and Si/Al ratio of the zeolites. The adsorption of H<sup>+</sup> to zeolites decreases the amount of cation retention other than H<sup>+</sup>, and may cause the elution of Si and Al into aqueous solution.

In case of Cs<sup>+</sup> and Sr<sup>2+</sup> adsorption selectivity among the zeolites, mordenite had the highest Cs<sup>+</sup> adsorption selectivity, and the selectivity had no correlation to the CEC of zeolites. In contrast, Sr<sup>2+</sup> adsorption selectivity of the zeolites positively correlated with the CEC of the zeolites: Linde-type A with the highest CEC showed the highest adsorption selectivity, and its adsorption rate was more than 99.9% even in the presence of 0.5 M K<sup>+</sup>. Simulated soil decontamination experiments of Cs<sup>+</sup> and Sr<sup>2+</sup> from Cs- and Sr-retaining vermiculites by using mordenite and Linde-type A respectively showed decontamination rates of more than 90%. In case of radioactive decontamination experiments from soil, over 90% of Cs<sup>+</sup> and Sr<sup>2+</sup> were decontaminated from soil when large soil particles (>150 $\mu$ m) coexisted with small zeolite particles (<100 $\mu$ m) and 10-60% when small soil particles (<150 $\mu$ m) coexisted with large mordenite particles (>150 $\mu$ m) in presence of extracting salt solution depending on soil types. However, the method failed to remove 100% Cs<sup>+</sup> from the contaminated soils with fine clay particles due to: a) reduced effective contact distance between soil and zeolite particles in extracting solution; b) high concentration of already present competing soil K<sup>+</sup> and NH<sub>4</sub><sup>+</sup> due to applied fertilizers; c) encapsulation of mordenite by fine soil particles; d) Cs<sup>+</sup> fixation by clay minerals. In order to improve on the decontamination efficiency rate, the following recommendations are made: a) to extensively evaluate the effect of solid to solution ratios to find optimum ratio for effective contact; b) to change the concentration of extracting salts from 0.3M to 0.1 $\mu$ M-0.3M of extracting solutions; c) to characterize the clay mineralogy of different contaminated soils and their composition ratio including the amounts of exchangeable cations present; d) to screen for cation adsorption/desorption selectivity sequences by different soil clays i.e. vermiculite; and etc. and to compare with those that are highly selective by zeolites; e) to use cation saturated zeolites with same cation species of the extracting salt solutions.