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学 位 論 文 要 旨
Dissertation Abstract

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学位論文題目：
Title of Dissertation
Development of zeolite-embedded sheets for facile decontamination: radioactive cesium in soil and disease-causing bacteria in water
(簡便な除染のためのゼオライト包埋シートの開発：土壌中の放射性セシウムおよび水中の病原性微生物)

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This study aimed to investigate the potential of the mordenite-embedded sheet in decontaminating polluted environments and is consists of two parts. First, the decontamination of Cs-contaminated soils using a mordenite-embedded sheet, and second, the disinfection of water containing an *E. coli* species using a protonated mordenite-embedded sheet.

Part 1

The accidents in Fukushima Daiichi Nuclear Power Plant in 2011 had contaminated soils with radioactive Cs. In the Cs-decontamination, washing methods prevail due to cost and ease of use, but conventional washing methods remove little Cs^+ because the Cs^+-exchange reaction between the soil and solution reaches equilibrium after releasing only a few Cs^+. Here, adding a Cs^+-adsorbent to the washing suspension removes Cs^+ in the solution, causing more Cs^+-release from the soil. This adsorbent-coexistence method requires separating the adsorbent powder from the suspension after the washing, and this study developed a technology that uses a sheet embedded with an adsorbent powder for easy separation of the adsorbent from the suspension.

The mixtures of $^{133}Cs^+$-retaining wet/dry montmorillonite and water/0.1 M KNO_3 were shaken with/without the natural mordenite-embedded sheet (MES). Adding the MES increased Cs^+ release in all cases, with the highest increase for wet montmorillonite (more than nine times), and 0.1 M KNO_3 and water gave a similar Cs-decontamination level. The Cs concentration in the final washing solution was lower with water than with 0.1 M KNO_3, as was indicated by the isotherm experiments. Therefore, surface water or tap water is preferable as the washing solution because they are cost-effective, and the concentrations of both Cs and other solutes in the resultant waste solutions are low.

The proposed one-step washing method is a potential manageable decontamination method for Cs-contaminated solids. When this method is applied to Cs-contaminated soils, operating in the early stages of contamination is desirable because the desorption of Cs^+ becomes difficult with time.

Part 2

Safe water is essential for humans but is not readily available in rural areas of developing countries. The contaminants in the water are mainly heavy metals, arsenic, and pathogenic bacteria. The World Health Organization reported that at least two billion people drink fecal-contaminated water containing pathogens, associated with 485000 diarrheal deaths each year, mostly infants. The best way to solve this problem is to develop infrastructures that provide clean water, but in the short term, the priority should be supplying simple and effective methods to purify drinking water for individual use.

Besides boiling, chlorination and heavy metals (Ag and Cu) kill pathogenic bacteria in the water. This study used high concentrations of protons to kill bacteria because many bacterial species do not survive at low pH, below around 4, except for acidophiles. Adding acid solutions also lowers the pH, but using reagents at home may not be recommended for safety reasons. Hence, we used a mordenite-type zeolite holding protons to lower the pH, where the zeolite releases protons into the water through cation exchange with cations in the water.

A protonated natural mordenite-embedded sheet (H-MES) was prepared as a new tool for disinfecting drinking water. Proton retention was 1.2 mmol per gram of mordenite, 75% of its cation-exchange capacity. The H-MES released protons through cation exchange with cations in aqueous solutions, lowering the pH below 4. The low pH led to disinfecting 100 mL of 100-fold diluted TSB solutions containing an *Escherichia coli* species (DH5α). For example, an initial viable count of around 5000 CFU mL^{-1} decreased to 14 CFU mL^{-1} after 24 h shaking at 25 °C with added H-MES containing 0.2 g protonated mordenite; 3.8×10^7 CFU mL^{-1} without the H-MES.

Adding a nitric acid solution showed a similar effect, but using reagents at home might lead to unexpected accidents. Adding and removing the H-MES to and from household waterpots in developing countries will be done by hand.

Conclusion

This study showed the capacity of natural mordenite for both the Cs-decontamination and the disinfection of water. Its low price and availability make the natural mordenite a potential material for the above purifications. Embedding the natural mordenite powder to give sheet-form is crucial because it enables more release of Cs^+ from coexisting Cs-contaminated soils and easy separation of the mordenite powder from soil suspensions and the treated drinking water.