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Development of Composite Materials with Zeolite and Magnetite for Radioactive Cs Decontamination in Soil

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1. Intorduction

The decontamination of radioactive Cs from the accident at the Fukushima No.1 nuclear power plant is an urgent problem. Zeolites are the most promising material for the Cs decontamination in water such as ponds and rice fields. The movement of the Cs^+ ions in the soil to the zeolite should be possible when the powdered zeolite mixes with the soil during the wet process using a K^+ or $NH_4{}^+$ ion-containing solution for ion exchange with the Cs^+ ions in the soil. However, the collection of the zeolite after the decontamination of the radio Cs^+ ions is impossible when the powder material mixes with the soil. Magnetic collection is one of the methods using a composite material composed of the zeolite and a magnetic material after the Cs^+ ion adsorption.

An Na-P1 type artificial zeolite ($Na_6Al_6Si_{10}O_{32}?12H_2O$) having a high cation exchange capacity (CEC) is able to be synthesized at a low cost using alkali from the waste coal fly ash of thermal power stations. On the other hand, the synthesis method using alkali for the nano-sized magnetite (Fe_3O_4) is very similar to that for the Na-P1 type artificial zeolite. We considered that a new composite material using alkali from a suspension of both starting materials would be a promising material for the Cs decontamination.

In this study, we synthesized the composite material (magnetic zeolite) of the Na-P1 type zeolite and nano-sized magnetite by alkali processing from a mixed solution of the fly-ash and iron chlorides for the magnetic collection of the zeolite after Cs^+ ion adsorption.

2. Experimentals

Fly ash (JIS II type) from thermal power stations (Shikoku Electric Power Co.) was used for the preparation of the Na-P1 type zeolite. For the preparation of the Na-P1 type zeolite, the fly ash and 2M NaOH were mixed and refluxed at 100 ^{o}C for 24 h. The powder was collected and washed several times by centrifugal separation, and then dried at 80 ^{o}C. For the preparation of the nano-sized magnetite, $FeCl_2?4H_2O$ and $FeCl_3?6H_2O$ (mole ratio=1:2) were dissolved in pure water. The mixed solution was placed in a water bath at 100 ^{o}C, and then a 2M NaOH solution was added with stirring and held at the same temperature for 30 min. For the preparation of the composite material (magnetic zeolite) of the Na-P1 type zeolite and magnetite, the synthesized magnetite in water and then fly ash was added to the mixed solution. A 2M NaOH solution was added to the mixed solution and refluxed at 100 ^{o}C for 24 h.

3. Results and discussion

For the material without magnetite , the main peaks for the XRD were the Na-P1 type zeolite with mullite ($Al_6Si_2O_{13}$) as the second phase. The peak intensity of the magnetite increased with an increase in the magnetite content. The peaks of the magnetite were very broad due to its small crystalline size. The particle size for the magnetic zeolite was 5˜30 μm for a SEM observation. These particles of the magnetic zeolite were easily attracted by the neodymium magnet. For the TEM observation, the Na-P1 zeolite, the magnetite, and amorphous phases were confirmed using the electronic diffraction of the center of the particle. The nano-sized and aggregated magnetite particles were observed in the bright-field image. Due to the slow formation of the zeolite crystals after formation of the nano-sized magnetite, the magnetite particles existed at the grain boundary between the polycrystalline zeolites.

We tested that the magnetic zeolite (200 g) and the soil (2 kg) obtained from the rice fields in Fukushima were mixed using a shaking apparatus with $NH_4{}^+$ ion containing solution. The magnetic zeolite with radioactive Cs collected using neodymium magnet (8000 gauss). We succeeded to decontaminate ca. 80 % radio active Cs from the soil using the magnetic zeolite.

Keywords: Racioactive Cs Decontamination, Na-P1 type zeolite, Magnetite, Composite Material