Retention Mechanism of Cesium in Chabazite Embedded into Metakaolin-Based Alkali Activated Materials

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K-based geopolymer has recently attracted attention as a matrix material for spent synthetic chabazite encapsulation at the Fukushima Daiichi Nuclear Power Station. The aggregation of cesium with iron heterogeneously formed pollucite-like structures in chabazite, which was revealed by transmission electron microscopy images. Furthermore, Raman spectroscopy and thermodynamic calculation have proven the formation of pollucite structure.

Keywords: Geopolymer; Transmission electron microscopy (TEM); Raman spectroscopy; Thermodynamics calculation; Pollucite; Fukushima Daiichi Nuclear Power Station; Chabazite; Cesium retention

Disposal of the spent adsorbents generated from the decontamination of cooling water for the damaged reactor at the Fukushima Daiichi Nuclear Power Station (FDNPS) has become a crucial concern. Cesium-137 is one of the key radionuclides in the spent adsorbents. Hence, it is critically required to immobilize the radioactive cesium in the disposal system. The synthetic chabazite has been used for removing radioactive cesium for its high selectivity, even in the Simplified Active Water Retrieve and Recovery System (SARRY) and the Advanced Liquid Processing System (ALPS) at Fukushima Daiichi NPS site. Many previous studies have investigated a cement material as a matrix of the waste form, which matrix could immobilize radioactive nuclides. However, unfortunately, cesium retention cannot be expected for cement waste according to the results of many studies. On the other hand, geopolymer matrices recently attracts rising attention for radionuclides immobilization[1]. Geopolymer is an interlinked tetrahedral polymer containing amorphous alumina-silicate (Si-O-Al) with alkali cations (K^+, Na^+) as a charge balance. It has been reported that cesium could be effectively immobilized in the clinoptilolite-type of zeolite embedded with sodium aluminum silicate hydrate (N-A-S-H)-type geopolymer[2]. However, there are no further studies on the cesium retention mechanism in other zeolites such as chabazite embedded into potassium aluminum silicate hydrate (K-A-S-H).

In this study, the cesium retention mechanism of the synthetic chabazite embedded with K-A-S-H was investigated at low cesium concentration (10 ppm). The leaching experiment revealed that K-A-S-H could immobilize Cs^+ for 360 days with a leaching rate of 2.95%. The TEM images of leaching specimens after 90 days revealed that the cesium was concentrated on the aggregates with Fe, which were heterogeneously formed in the chabazite. From the electron diffraction patterns of the aggregates by TEM, the aggregates had a similar structure to Fe-pollucite in the cesium-contaminated soil nearby the FDNPS site[3]. Raman spectroscopy analysis and thermodynamic calculation have proven the structural change from chabazite to pollucite. It was clearly demonstrated that the pollucite-like aggregates were formed from the chabazite during the mixing with alkali-activated reagents. Pollucite has been well known as cesium-bearing natural zeolite, which can encapsulate cesium in the structure. Therefore, cesium retention was achieved in chabazite embedded into K-A-S-H as a spent adsorbent due to the formation of pollucite-like structures during the fabrication of chabazite embedded into K-A-S-H.

References

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