Immobilization of Iodide and Iodate in Layered Double Hydroxide, Magnesium oxide and Silver embedded into Metakaolin-Based Alkali Activated Materials

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In March 2011, an accident had occurred in Fukushima Daiichi Nuclear Power Station (FDNPS), which caused massive damage to the reactor and released I-131 and I-129 products. The radioactivity of I-131 and I-129 in the environment has a high impact on human health under a long exposure as observed from the Chernobyl incident. The concentration of these two isotopes varies in the environment. A large quantity of I-131 was initially released, which was hard to detect owing to its short half-life (8 days). On the other hand, I-129 was initially released with a low quantity. However, it has a longer half-life (1.57 x 10^7 years); thus, this isotope persists in the environment [1]. Hence, it is crucial to solidify and immobilize I-129 isotope to achieve safe disposal system.

Geopolymer is considered to be one of the best candidates as an encapsulation matrix for radionuclide immobilization (i.e., cesium (Cs^+) and strontium (Sr^{2+})) [2]. Similar to zeolite, geopolymer is an alkaline material, whose structure contains alumina-silicate tetrahedra in polymers. It is known for cation immobilization due to association with the negatively charged alumina tetrahedra.

Previous studies have been investigating the possible natural and synthetic adsorbent materials for removing iodine. In an aqueous environment, iodine mainly exists as iodide (I^-) and iodate (IO_3^-). They may have a possibility to be immobilized by a positively-charged adsorbent within the geopolymer matrix. Layered double hydroxide (LDH), magnesium oxide (MgO), and silver (Ag) are commonly used for anion immobilization, which are proven to exert high iodine retention due to their ability to offer positively charged structures [3,4,5]. However, I^- and IO_3^- could be repelled by the geopolymer structure. There are few studies of these adsorbents' stability with iodine under alkaline environments and no openly published study of I^- and IO_3^- immobilization within the geopolymer matrix. Therefore, in this study, the adsorbed iodine in the LDH, MgO, and Ag embedded within geopolymer matrix was investigated by evaluating the leaching behaviour of iodide and iodate and their adsorbents' stability.

After 19 days of leaching experiments, the iodine leaching levels of the materials were observed as follows: geopolymer matrix had leached out 89% I^- and 96% IO_3^-, LDH had 100% leached I^- and IO_3^-, MgO had leached 98% IO_3^-, and Ag had leached out 58% I^- and 2.4% IO_3^-. The stability of I^- and IO_3^- in the adsorbents embedded within geopolymer matrix was clarified by nano-micro analysis (i.e., SEM, TEM), and their mineral phases were characterized by XRD and calculated by thermodynamic calculations. Firstly, LDH after encapsulation (0 day) and after 19 days of leaching did not change into any phase. However, LDH preferred $H_{4-n}SiO_4^n$ adsorption over I^- and IO_3^- due to the high silica activity in the geopolymer matrix. Secondly, MgO changed its phase into magnesium silicate hydroxide (M-S-H) in the geopolymer matrix after 19 days of leaching. However, M-S-H did not have the capability for iodine retention. Finally, Ag^+ was precipitated with I^- to form iodyrite or iodargyrite, which were stable after 19 days of leaching. Therefore, Ag is an excellent adsorbent for immobilization of I^- and IO_3^- in the geopolymer matrix.

Keywords: Geopolymer, Fukushima Daiichi Nuclear Power Station, Layered double hydroxide, Magnesium oxide , Silver, Iodine retention

