学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　博士（工学）　氏名　NIU Xiaobo

学 位 論 文 題 名

Evaluation of metakaolin-based geopolymer for the treatment of simulant radionuclides: Analysis of surface chemistry and immobilisation behaviour
(模擬放射性核種処理におけるメタカオリン系ジオポリマーの評価: 表面化学と取り込み挙動の分析)

The development of nuclear power has significantly contributed to the progress of productivity in modern society. However, it has also brought forth formidable challenges. Issues such as nuclear fuel back-end disposal, radioactive waste management, and nuclear accident response pose inevitable hurdles, resulting in the generation of numerous radionuclides. Effectively containing these radionuclides is crucial to prevent environmental pollution and ensure a safe ecosystem for living organisms. Certain radionuclides, both cationic (e.g., ^{137}Cs, ^{90}Sr, and ^{60}Co) and anionic (e.g., ^{131}I and ^{79}Se), present significant environmental risks when it comes to the storage or disposal of nuclear waste. This is primarily due to their relatively long half-life and high mobility in soil and aqueous systems.

Geopolymer materials, emerging as a potential alternative to conventional Portland cement, have garnered widespread attention as a potential waste form for immobilizing radionuclides. However, research in the field of radionuclides immobilization within geopolymers is still in its early stages, necessitating further systematic and in-depth studies. This paper focuses on investigating the adsorption behaviour of alkali-activated metakaolin geopolymers on both cationic and anionic radionuclides. Predictive models based on thermodynamics are developed to better understand and forecast this behaviour. Moreover, efforts are made to address the lack of anionic radionuclide immobilization capacity in alkali-activated metakaolin geopolymers. By tailoring the materials, their capability to immobilize anionic radionuclides is enhanced. Additionally, the study explores the immobilization of anions using phosphoric acid-activated metakaolin geopolymers, providing valuable insights into the surface chemistry and electrostatic properties of acid-activated geopolymer materials.

In Chapter 1, background information on the study was presented, including the stages of nuclear power development, opportunities, and challenges. In addition, the application scenarios of geopolymer materials in the disposal of radionuclides, methods are presented. Finally, based on the presented background, the research objectives and structural organization of this thesis are discussed.

In Chapter 2, a literature review of geopolymer materials was presented, including basic properties, different activation pathways, and their hydration processes. In addition, basic information on radionuclides is presented, including chemical properties, toxicity, isotopic properties of radionuclides, and their general disposal methods.

In Chapter 3, two types of metakaolin-based geopolymers (Metastar501 and Sobueclay) were synthesized. They were evaluated for their binding capacity and interaction with Cs^{+}, Sr^{2+}, Co^{2+}, I^{-}, $IO_3{}^{-}$, $SeO_3{}^{2-}$, and $SeO_4{}^{2-}$. Results showed that both geopolymers effectively immobilized cationic ra-

dionuclides, while neither of the geopolymers could effectively incorporate anionic radionuclides. Cs^+ uptake involved a one-to-one exchange with K^+, while Sr^{2+} and Co^{2+} exhibited one-to-two and one-to-one exchanges with K^+. Co^{2+} binding was influenced by the formation of cobalt blue ($CoAl_2O_4$). Thermodynamic modelling predicted low-concentration binding of Cs^+ and Sr^{2+} based on the ion exchange mechanism.

In Chapter 4, a modification to metakaolin-based geopolymers to enhance their uptake capabilities for selenium oxyanions was proposed. Through co-precipitation experiments, binding analysis, and structural analysis, it was discovered that the in-situ formation of ettringite enables effective uptake of SeO_3^{2-} and SeO_4^{2-}. The modified geopolymer, with its in-situ ettringite, not only retains its capacity to uptake cationic radionuclides but also acquires the ability to uptake SeO_3^{2-}. In addition, the developed thermodynamic modelling accurately predicts the uptake of SeO_3^{2-} at low concentrations.

In Chapter 5, the study explored the electrostatic properties and anion immobilisation potential of PGPs (phosphoric acid activated metakaolin-based geopolymers). Results indicate that acid activation triggers geopolymerisation, forming Al_x-PO units that control surface charge. PGPs possess a positive zeta potential in the pH range of about 2-5 and exhibit a positive peak zeta potential at a pH of about 4, which is the pH at which PGPs reaches equilibrium in water. In acidic conditions, leaching of Al_{VI}-PO units decreases zeta potential, while loss of a proton from Al_{VI}/Si-OH group decreases zeta potential in alkaline conditions. PGPs effectively immobilize SeO_3^{2-}, SeO_4^{2-}, I^- and IO_3^- anions through stabilization/solidification, with the electrostatic attraction being the primary association between the anions and the positively charged PGPs surface.

Finally, chapter 6 presented the key findings and conclusions of this thesis. It offered valuable insights into the immobilization of with Cs^+, Sr^{2+}, Co^{2+}, I^-, IO_3^-, SeO_3^{2-}, and SeO_4^{2-} in both alkaline-activated and acid-activated metakaolin geopolymers. Additionally, a summary of the surface chemistry and electrostatic properties of the newly proposed acid-activated geopolymers was provided. Furthermore, suggestions are made for future in-depth studies on geopolymers for radionuclide immobilisation.