学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Application of Alkali-Activated Materials to Solidification of the Wastes Generated from On-site and Off-site of Fukushima Dai-Ichi Nuclear Power Station
(福島第一原子力発電所のオンサイトとオフサイトで発生している廃棄物固化へのアルカリ刺激材の応用)

The accident at the Fukushima Dai-ichi Nuclear Power Station (FDNPS) led to a significant release of radioactive materials, impacting both the surrounding environment and human health with extensive consequences. On-site at the FDNPS, systems such as the Simplified Active Water Retrieve and Recovery System (SARRY), Advanced Liquid Processing System (ALPS), and KURION water treatment facilities played a pivotal role in treating water contaminated with radionuclides. This treatment resulted in secondary wastes such as spent chabazite and iron slurry. These systems are tasked with the intricate challenge of long-term management and disposal of radioactive wastes, which exhibit a range of characteristics. For instance, Cesium-137 is highly radioactive with a half-life of 30.17 years, whereas isotopes such as selenium-79 and iodine-129 have much longer half-lives, approximately 327,000 years and 15.7 million years, respectively. This variation significantly complicates their management and permanent disposal, considering their long-lasting nature and potential for environmental migration, making them key radionuclides of safety assessment. Additionally, the accident led to widespread soil contamination with Cs-137, affecting an area of 13,600 km^2, necessitating ongoing management and the development of long-term waste disposal strategies. In this context, the current doctoral thesis delves into the use of alkali-activated materials (AAMs), particularly potassium-based alkali-activated materials (K-AAM), for the immobilisation of these hazardous radioactive materials. Through the use of advanced analytical instruments and thermodynamic models, this research aims to gain a more comprehensive understanding of the mechanisms underlying the immobilisation process.
Chapter 1 of the doctoral thesis provides a comprehensive background on the aftermath of the FDNPS incident. This chapter delves into the complexities of managing fuel debris, contaminated debris, water, soil, and vegetation highlighting the limitations of current technologies. It introduces AAMs as a novel solution for these challenges. The chapter concludes with an outline of the study objectives and provides a framework for the subsequent structure of the doctoral thesis.
Chapter 2 presents a thorough review of the literature on AAMs, exploring their fundamental properties, activation mechanisms, and hydration processes. This chapter specifically concentrates on K-AAMs, providing a summary of the current knowledge and pinpointing the gaps that this research intends to address.
Chapter 3 targets the immobilisation of Cs, commonly found in spent chabazite adsorbents, using K-AAM as an encapsulating matrix. In-depth analysis suggests that the high Cs retention in K-AAM is primarily due to its amorphous structure and alkalinity, which lead to structural changes in the

aluminosilicate rings of chabazite, enhancing Cs ion retention.

Chapter 4 investigates the immobilisation of Cs in soils, specifically basaltic and granitic types from Fukushima, which replicate the actual Cs-contaminated soil. This chapter evaluates the use of soil as a binder with potassium-based alkali activators, exploring various recipes with different particle sizes to determine the optimal combination for encapsulation efficiency, mechanical strength, and waste volume reduction. The selected optimal formulations demonstrate strong cesium retention in K-AAM.

Chapter 5 examines the interactions between iron slurries, specifically 2-line ferrihydrite, and K-AAM. It discusses the low solubility of 2-line ferrihydrite when integrated into K-AAM, which inhibits the release of ferric ions. This phenomenon is investigated using nano-micro analysis.

Chapter 6 focuses on incorporating iodide into K-AAM. It assesses the effectiveness of various additives, including layered double hydroxide, magnesium oxide, and silver nitrate, in immobilising iodide ions. Despite certain limitations, silver nitrate is found to be the most effective, particularly when the silver concentration is precisely controlled. The chapter also examines the influence of competing ions and the structural integrity of K-AAM in immobilising iodide ions.

Chapter 7 investigates the behavior of selenite ions in K-AAM. The chapter reveals superior selenite ion retention in K-AAM, attributed to unique binding mechanisms within its matrix, as elucidated through detailed analysis.

Chapter 8 concludes the studies by recommending methods for the underground disposal of various waste types. It evaluates the suitability of trench and pit disposal methods for different radionuclide-contaminated wastes embedded into K-AAM, using leaching rate calculations and focusing on minimising potential radiation exposure pathways.

Finally, Chapter 9 provides a summary and draws general conclusions, highlighting the novel findings of the doctoral study, their implications, and suggesting directions for future research.