一般セッション | V. 核燃料サイクルと材料：505-1 放射性廃棄物処理

📅 2025年3月13日(木) 14:45 ~ 16:15 🏛 H会場(Zoomルーム8)

[2H08-12] セメント・ジオポリマー

座長:山田 一夫(国環研)

14:45 ~ 15:00

[2H08]

泊発電所3号機のセメント固化体に対するWAC適合性検討について（その２）

*小林 大和[1]、本山 光志[2] (1. 北海道電力、2. 日揮株式会社)

15:00 ~ 15:15

[2H09]

加熱すりもみ法による放射性コンクリート廃棄物の処理方法の検討

*東河 慶吾[1]、河口 宗道[1]、澤 和弘[1] (1. 北海道大学)

15:15 ~ 15:30

[2H10]

Enhancing Radionuclide Immobilization and Neutron Capture Potential in Metakaolin-Based Geopolymers through Boron Incorporation

XIAOBO NIU[1], *Yogarajah ELAKNESWARAN[1], Ang Li[1] (1. Hokkaido University)

15:30 ~ 15:45

[2H11]

Immobilization of Anions and Cations in Phosphoric Acid-Activated Geopolymers

*李 昂[1]、牛 暁博[1]、エラクネス ヨガラジャ[1] (1. 北海道大学)

15:45 ~ 16:00

[2H12]

Immobilization of Radionuclides in Fly-ash/Slag-based Geopolymer Matrices

*Mael VIAUD[1], Kanako Toda[1], Takumi Saito [1] (1. UTokyo)

16:00 ~ 16:15

座長持ち時間

泊発電所 3 号機のセメント固化体に対する WAC 適合性検討について（その２）

Countermeasures for conformity with WAC of cement-type waste packages arisen
from Tomari Nuclear Power Station Unit 3 (2)

*小林 大和 [1]，本山 光志 [2]
[1] 北海道電力株式会社，[2] 日揮株式会社

北海道電力㈱泊発電所 3 号機から発生する濃縮廃液を固型化したセメント固化体は、固化体製作方法及び放射能評価方法が、過去に類例のないものとなるため、WAC への適合性を検討している。

キーワード：低レベル放射性廃棄物、セメント固化体、WAC

1. 緒言

日本原燃㈱より提示されている廃棄物受入基準（WAC と呼ぶ、「日本原燃㈱　濃縮・埋設事業所　廃棄物埋設施設保安規定（認可日：2023 年 11 月 17 日）」にて提示）の適合性確認方法に関して、泊発電所 3 号機のセメント固化体について、①放射能濃度の確認方法、②セメント固化体の一軸圧縮強度、③セメント固化体の練り混ぜ、④健全性を損なうおそれのある物質、⑤セメント系材料の観点から、試験及び検討を実施し、この結果、いずれの課題についても、対応可能な見通しを得たため、本報告（その 1）[1] で報告した。

本報告では、①に関して分析データの拡充を進め、当初想定通りの結果を得たことについて報告する。

2. 放射能濃度の確認方法に関する検討結果

分析対象とした試料は、濃縮廃液のセメント固化体（10 点、各年度 1 点（1 年のみ 2 点）の 9 年分)、濃縮廃液（4 点）及び固体状廃棄物（液体フィルタ：3 点）の 17 点である。図 1 に平均放射能濃度法適用対象の一例として H-3 を、図 2 に SF 法適用対象の一例として Ni-63 の分析結果を Co-60 との散布図として示した。H-3 は発電所の廃棄物毎に平均放射能濃度が設定されているため、3 号セメント固化体独自に設定することが必要であるが、参考までに泊 1/2 号のアスファルト固化体と比較した結果、ほぼ一致した。PWR 共通の SF 及び平均放射能濃度が設定されている核種の分析結果は、Ni-63 と同様に、PWR 共通の設定値とほぼ一致していた。これらの結果から、従来と同じ考え方で SF 及び平均放射能濃度は設定できると判断される。

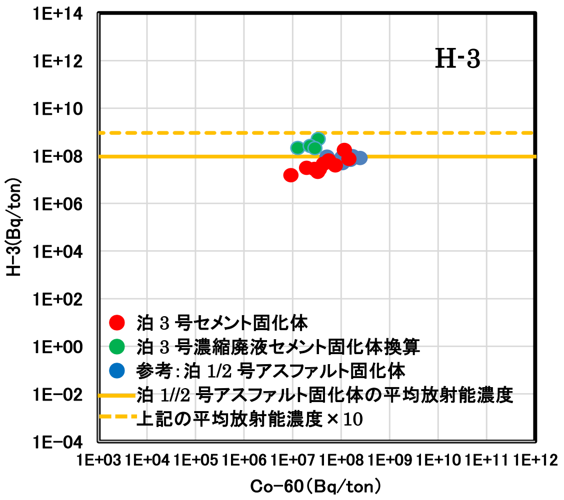

図-1 H-3 と Co-60 の散布図

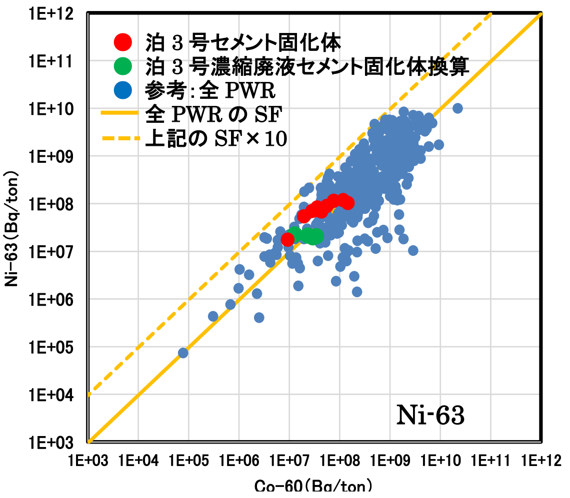

図-2 Ni-63 と Co-60 の散布図

参考文献

【1】小林ら：日本原子力学会 2024 春の年会[1A03] 泊発電所 3 号機のセメント固化体に対する WAC 適合性検討について.

謝辞　本検討に当たり、貴重なご助言を頂きました、日本原燃㈱ 熊谷守氏に、感謝申し上げます。

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加熱すりもみ法による放射性コンクリート廃棄物の処理方法の検討

Study on treatment method of radioactive concrete waste by heating and rubbing method

*東河 慶吾[1], 河口 宗道[1], 澤 和弘[1]

[1] 北海道大学

放射性コンクリートの再資源化・再利用に向けた処理方法の検討のために、模擬汚染コンクリートの加熱すりもみ法による骨材分離の基礎実験を行った。再生骨材に含まれる Cs 量の基礎データを採取した。

キーワード：模擬汚染コンクリート、加熱すりもみ法、骨材分離

1. 緒言

原子力発電所で用いられるコンクリートは膨大であるため、福島第一原子力発電所(1F)の事故により大量に発生する放射性コンクリート廃棄物の合理的な処理・処分が求められる[1]。本研究では実験室規模での骨材分離試験を行い、骨材分離により発生する再生骨材、粉末、気中に浮遊する粉塵の時間変化を測定した。骨材分離後の再生骨材、粉末、粉塵の Cs 濃度分析を行った。

2-1. 実験

本研究では、加熱すりもみ法の前処理として、約 30 mm に粗粉砕した模擬汚染コンクリート塊を 150 g を用意し、300℃で 3 時間加熱した。その後、小型ボールミル（容積 5L）に模擬汚染コンクリート塊と 750 g のボルト(M27×35 mm)を入れ、骨材分離試験を行った。骨材分離はミルを 50 rpm で 3 分間回転させ、その都度骨材と粉末の重量を測定した。また骨材分離中に気中に浮遊する粉塵をカスケードインパクター(LP-20)で分級／サンプリングした。この骨材分離の操作を 20 回（計 60 分間）行った。骨材分離試験後に再生骨材と粉末、サンプリングした粉塵に含まれる Cs 濃度を原子吸光分析により定量化した。

2-2. 結果

図 1 は再生骨材と粉末の質量変化を示す。3 分間の骨材分離により、再生骨材の質量は 5 g 減少し、粉末の質量は 1.5 g 増加した。60 分間の骨材分離試験で、再生骨材の質量は推定される粗骨材の質量に達した。図 2 は骨材分離前の模擬汚染コンクリート塊、図 3 は 60 分間の骨材分離試験後の再生骨材の外観である。一部の再生骨材にモルタルが付着しているものの、完全に分離された再生骨材も確認された。

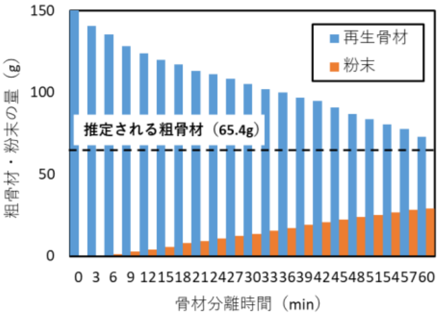

図 1. 再生骨材の質量

3. 結論

本研究は、加熱すりもみ法による骨材分離により発生する再生骨材、粉末、気中に浮遊する粉塵の時間変化を測定し、含まれる Cs 濃度の基礎データを採取した。

図 2. 骨材分離前

図 3. 骨材分離後

参考文献

[1] JAEA-Review 2022-038

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*Keigo Sokugawa[1], Munemichi Kawaguchi[1], Kazuhiro Sawa[1]

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Enhancing Radionuclide Immobilization and Neutron Capture Potential in Metakaolin-Based Geopolymers through Boron Incorporation

Xiaobo Niu[1], *Yogarajah Elakneswaran[1], Ang Li[1]
[1]Hokkaido Univ

Abstract: This study aimed to develop a modified geopolymer material with neutron-absorbing and masking properties by incorporating boron (B) into a metakaolin-based geopolymer. The mechanisms underlying the incorporation and stability of boron in geopolymers were investigated under various curing and activation conditions.

Keywords: Metakaolin-based geopolymer; Boron; Alkali-activation; Acid-activation; Borosilicate; Immobilisation

1. Introduction and Methodology

Metakaolin-based geopolymers have attracted increasing interest as a promising solution for radioactive waste treatment, with previous studies confirming their ability to immobilise radionuclides. However, their limited neutron shielding capability, due to the absence of a structure capable of effectively capturing neutrons, restricts their broader application in radioactive waste disposal and related safety concerns. Therefore, this study aims to develop a method for incorporating boron (B), which has strong neutron-capturing properties, into the structure of metakaolin-based geopolymers, thereby enhancing their neutron shielding capacity while preserving their radionuclide immobilisation effectiveness. Geopolymers synthesised via both alkali and acid activation methods were systematically compared under different curing conditions. Structural changes due to varying B content were analysed using mechanical strength tests, X-ray diffraction (XRD), scanning electron microscopy (SEM), solid state nuclear magnetic resonance spectroscopy (NMR), and Fourier transform infrared spectroscopy (FTIR). The stability of B within the geopolymer matrix, along with its effects on the overall structural integrity and its radionuclide immobilisation capacity, was assessed using leaching tests and assessment of the leachate with inductively coupled plasma optical emission spectroscopy (ICP-OES). This comprehensive analysis enhances the understanding of B incorporation into metakaolin-based geopolymers and supports the development of borosilicate structures, offering valuable technical insights and potential applications in nuclear waste disposal, particularly in neutron-rich circumstances.

2. Conclusion

B formed an unstable BO_4(xB, 4-xSi) structure with extra short-range Si tetrahedra in low-temperature conditions, making it prone to be leached out. High-temperature curing facilitated alkali-activated geopolymerisation, mitigating negative effects of boric acid. Additionally, in acid-activated geopolymers, B could be incorporated into the framework by bonding with numerous Al-unsaturated Si tetrahedra to form stable BO_4(0B, 4Si) structures. Overall, the B-containing acid-activated geopolymer effectively incorporated B into the main framework while maintaining radionuclide immobilisation capacity. This study provides valuable insights into the selection and incorporation mechanisms of the B-containing geopolymer matrix, contributing to strategies for radioactive waste disposal.

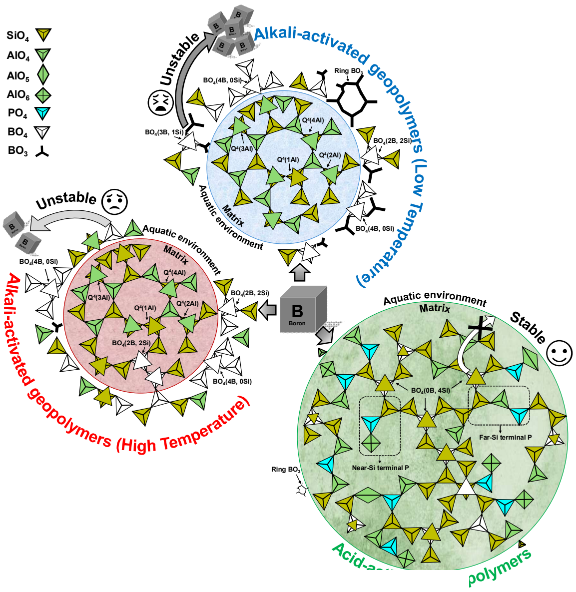

Fig1. Graphical Abstract

Immobilization of Anions and Cations in Phosphoric Acid-Activated Geopolymers

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Abstract: Compared to conventional alkali-activated geopolymers, acid-activated geopolymers excel in immobilizing anions and exhibit superior stability after being exposed to water. Changes in zeta potential under varying solution conditions reveal the immobilization mechanism, further supported by binding experiments. This study advances the understanding of ion immobilization, paving the way for efficient radioactive waste management.

Keywords: Acid-activated geopolymer, Immobilization, Zeta potential, Stability, Radioactive waste management

1. Introduction

The effective management of radionuclides is a critical global challenge due to their long-term persistence and potential risks to ecosystems and human health. The conventional alkali-activated geopolymers, with their permanent negative surface charge, demonstrate strong adsorption of cations but show limited effectiveness in immobilizing anions like SeO_3^{2-} and SeO_4^{2-}. Phosphoric acid-activated geopolymers, as an emerging material, have shown significant potential in immobilizing various anions. However, their ability to immobilize critical cations, such as Cs^+ and Sr^{2+}, has not been sufficiently studied. Therefore, addressing this gap is crucial for enhancing the applicability of phosphoric acid-activated geopolymers in managing radioactive waste. This study aims to investigate the immobilization efficiency of phosphoric acid-activated geopolymers for a range of anions and cations. Geopolymers synthesized using acid activation methods were systematically compared. The structural changes and ion immobilization efficiency were analyzed using leaching tests, binding capacity measurements, and structural characterizations, including X-ray diffraction (XRD) and zeta potential measurements. The stability of the immobilized ions and their binding capacity were further evaluated by analyzing the leachates with inductively coupled plasma optical emission spectroscopy (ICP-OES). The results will provide valuable insights into the ion immobilization mechanisms, supporting the development of phosphoric acid-activated geopolymers as an effective material for radioactive waste management.

2. Conclusions

Compared to traditional alkali-activated geopolymers, acid-activated geopolymers perform better in immobilizing anions. The stability of acid-activated geopolymers after exposure to water demonstrates superior performance (Figure 1). The change in zeta potential under different solution conditions explains the immobilization mechanism (Figure 2), and the binding experiments further corroborate this. This study provides valuable insights into the mechanisms of ion immobilization and highlights the potential of acid-activated geopolymers as an effective material for managing radioactive waste.

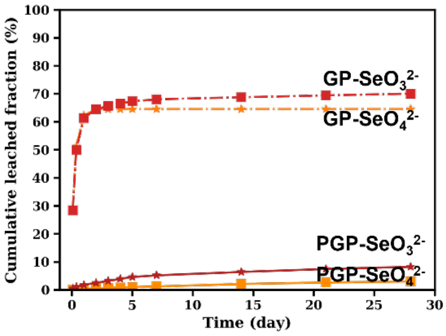

Figure 1 Cumulative fraction of anions leaching (GP: conventional geopolymer; PGP: acid activated geopolymer)

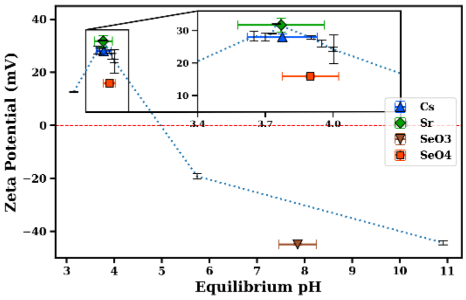

Figure 2 Zeta potential changes of acid activated geopolymer under different solution environments

Immobilization of Radionuclides in Fly-ash/Slag-based Geopolymer Matrices

*Maël VIAUD[1], Kanako Toda[1] and Takumi Saito[1]

[1]UTokyo

This study investigates geopolymers as a potential solution for immobilizing radionuclides such as cesium and selenium. It focuses on fly ash-based geopolymers, emphasizing the importance of precursor selection. The feasibility of forming the matrix itself is studied with the capacity to immobilize the target radionuclides.

Keywords: Geopolymer, Radionuclide, Ion Immobilization, Fly-ash, Slag

1. Introduction

The 2011 Fukushima accident heavily contaminated soil and forests, demanding advanced strategies to immobilize radioactive cesium (Cs) due to its slow mobility in soils and long half-life. Worldwide occurrences of radionuclide-bearing ashes may also contain selenium compounds (e.g. SeO_4^{2-} and SeO_3^{2-}), which exist as anions, and present an immobilization challenge, too. Geopolymers (GP), formed via alkali activation of aluminosilicate precursors such as fly ash (FA) and slag, offer eco-friendly alternatives to Ordinary Portland cement (OPC) for the immobilization of such radionuclides [1]. Also, the direct use of contaminated FA as GP precursors could enable volume reduction by avoiding the use of cementitious precursors. However, the unpredictable content of silicon (Si), aluminum (Al), and calcium (Ca) in precursors complicates the optimization of GP matrices and the immobilization capacity of radionuclides.

The objective is to know if such GPs are suitable and effective to immobilize these radionuclides.

2. Method

GP were synthesized via NaOH/KOH activation. Fly ash (FA), FA and slag (FA/slag), and metakaolin (MK) based GP were synthesized using precursors with 100% FA, 50% FA and 50% slag, and 100% MK, respectively. The mixing ratio of precursors to alkaline activators was investigated to form specimens that could be mixed readily and undergo uniaxial compressive strength (UCS) development. The samples were cured for 1 month at 20 °C. UCS test, X-ray fluorescence (XRF), X-ray diffraction (XRD), and scanning electron microscopy (SEM) were performed to study the strength, the elemental compositions, the presence of crystalline phases, and the microstructure, respectively. 10 ppm of Cs^+, SeO_4^{2-}, and SeO_3^{2-} were added at mixing and cured for a week. The samples were ground and leached in deionized water for 24 hours, and the release of target ions was quantified with an inductively coupled plasma mass spectrometry (ICP-MS).

3. Results and discussion

The FA contained 7% Ca, 5% Si, and 1% Al. The 100% FA-GP was hardened with water glass (7% K_2O, 14% SiO_2), targeting Si/Al and K_2O/Al ratios of 15 and 25, respectively. All specimens achieved UCS > 0.98 MPa, meeting the required strength for cementitious waste bodies. Leaching tests confirmed good, poor, and moderate Cs retention in MK-GP, FA-GP, and slag/FA-GP, respectively (Fig.1). In contrast, retention of SeO_4^{2-} and SeO_3^{2-} was weak across all formulations. XRD analysis revealed that FA/slag-GP formed crystalline secondary phases but not in FA-GP. These findings underpin the potential of GP for cation immobilization while underlining the importance of precursor selection, and the possibility of using FA and slag to form waste forms instead of OPC or MK. Furthermore, for anion immobilization, this study emphasizes a need for further modifications—such as incorporating ettringite [2] or others.

References

[1] Provis et al., www.annualreviews.org, 2014 [2] Niu, *Thesis Hokkaido University*, 2023

Acknowledgment This research was performed by the Environment Research and Technology Development Fund (JPMEERF22S20910) of the Environmental Restoration and Conservation Agency provided by the Ministry of the Environment of Japan.

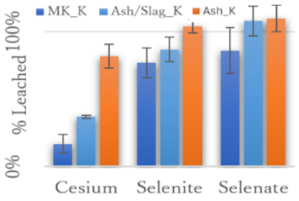

Fig.1 Leaching Behavior of Cesium and Selenium Ions in GP with FA and Slag Modifiers