

令和 2 年 7 月 7 日現在

機関番号：11601

研究種目：基盤研究(C) (一般)

研究期間：2017～2019

課題番号：17K00622

研究課題名(和文) A new approach for the selective removal of radionuclides of different half-lives from liquid waste

研究課題名(英文) A new approach for the selective removal of radionuclides of different half-lives from liquid waste

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交付決定額(研究期間全体)：(直接経費) 3,600,000円

研究成果の概要(和文)：この研究では、選択的分離とそれに続く廃棄物からの濃縮により、溶液からさまざまな半減期の放射性核種を選択的に除去するための新しい手法を紹介します。イオン選択的固相抽出システムを使用して水溶液からCsとSrを分離する2つの手法が開発されました。このプロジェクトの研究期間中の研究結果から、20件の査読付き学術論文が発表されました。

研究成果の学術的意義や社会的意義

The research was aimed to address the radiological issues for Fukushima's revitalized future after the FDNPP accident. The findings can be used to treat radioactive waste from nuclear power plants or the health sectors. Government personnel dealing with regulatory issues can also use it.

研究成果の概要(英文)：The research introduces new approaches for the selective removal of radionuclides of different half-lives from the liquid matrices via selective isolation followed by concentration from bulk waste. We developed two techniques for the selective separation of Cs and Sr from the aqueous matrix using ion-selective solid-phase extraction assemblies. During the project tenure (FY2017 to FY2019), the research results that so far obtained either from the current project or related research theme in progress in my laboratory have been reported in peer-reviewed journals resulting in 20 articles altogether. Several presentations in international conferences also used to share the preliminary results from the research project.

研究分野：Environmental remediation

キーワード：Solid-phase extraction Selective separation Radionuclides

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様式 C-19、F-19-1、Z-19 (共通)

## 1. 研究開始当初の背景

The decommissioning process of tsunami plus earthquake-crippled Fukushima Daiichi Nuclear Power Plant (FDNPP) continues to be weighed down by the accumulation of tons of radioactive waste water.

The radionuclides, which are the standard components of liquid radioactive waste, can be categorized to short, medium, long and extraordinary long-lived depending on the corresponding decay period. The radioactivity of spent fuel often modeled as the exponential decay of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , which are known as long-lived radionuclides ( $t_{1/2} \sim 30$  years).<sup>(1)</sup> The  $^{36}\text{Cl}$  ( $t_{1/2} = 0.3$  million years),  $^{135}\text{Cs}$  ( $t_{1/2} = 2.3$  million years) and  $^{129}\text{I}$  ( $t_{1/2} = 15.7$  million years) are some extraordinary long-lived radioisotopes, which represents the extreme biohazards.<sup>(2, 3)</sup> There is a notable time-lag in the half-lives of long and extraordinary long-lived radioisotopes, but it should be isolated in the longest decay period defensible repository. The existence of other natural compounds along with the radioisotopes also evokes concerns in using the nuclear transmutation as a disposal method. Therefore, selective removal of radionuclides of varying lifespans and the other elements will be useful in a cost-effective radioactive waste management.

Our previous research focused on the selective removal of potentially toxic elements and rare metals from waste matrices using molecular recognition approach.<sup>(4-9)</sup> The mechanism of molecular recognition by a host molecule considers the particular electronic and spatial features of another chemical species, called the guest, and capture selectively (Fig. I).<sup>(10, 11)</sup> The supramolecular technology offers the option of selective isolation of multiple ionic species of different charge states within the same structural arrangement (Fig. II).<sup>(12, 13)</sup>

There have been researches, which focused on the elimination or separation of radionuclides from waste matrices. The applications of ion exchange, membrane processes, and adsorptive technologies have been explored for removal of radionuclides from liquid wastes.<sup>(1, 14)</sup> Other efforts include co-precipitation,<sup>(15)</sup> electrocoagulation,<sup>(16)</sup> adsorption using activated carbon and/or zeolite-based media,<sup>(17, 18)</sup> sorption with different biomaterials,<sup>(19-26)</sup> and so forth. The removal techniques, however, lacks in selectivity, which is the major limiting issue of those methods.

The learnings from previous research on selective removal of hazardous species from the liquid matrices have been used in the current project, and it is a unique option to be explored till the date.

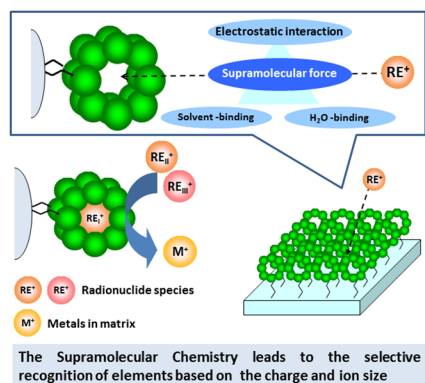


Fig. I: Mechanism of molecular recognition

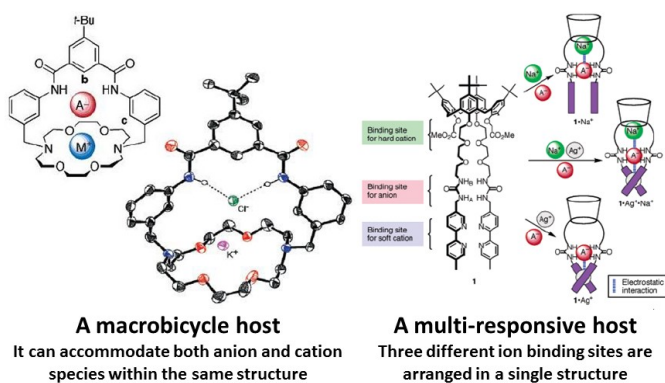


Fig. II: A supramolecular structure (host molecule) can accommodate multiple guest molecule

## 2. 研究の目的

The aim of current research is to develop a protocol for the removal of radionuclides from liquid waste based on the corresponding lifetimes. The solid-phase materials capable of molecular recognition, and designed using supramolecular technology will be employed. It will be a unique approach for treating the liquid wastes containing radionuclides. It is a zero-emission technique, expected to reduce containment period, monitoring time and cost.

### 3. 研究の方法

The proposed process included following steps:

- a) Characterization of radionuclides in liquid waste;
- b) Evaluation and selection of solid-phase extraction (SPE) systems;
- c) Optimization of the selective removal process of radionuclides from liquid waste using different SPE systems in tandem or in series;

Selection of SPEs were based on trial-and-error approaches. The laboratory-based experiments was used ICP-MS or ICP-OES for stable element analysis, while HPGc detectors was used for radionuclides.

### 4. 研究成果

The research was progressed as planned. The laboratory is now equipped with necessary instruments, and future research on a similar theme can be continued if an additional fund is received. We developed two techniques for the separation of Cs and Sr from the aqueous matrix using ion-selective solid-phase extraction assemblies. The data-sets, after verification steps, now compiled for reporting in the peer-reviewed journals. During the project tenure (FY2017 to FY2019), the research results that so far obtained either from the current project or related research theme in progress in my laboratory has been reported in peer-reviewed journals as follows:

- 1) S. Barua, I.M.M. Rahman, I. Alam, M. Miyaguchi, H. Sawai, T. Maki, H. Hasegawa, Liquid electrode plasma-optical emission spectrometry combined with solid-phase preconcentration for on-site analysis of lead, *J. Chromatogr. B*, 1060 (2017) 190-199.
- 2) Z.A. Begum, I.M.M. Rahman, H. Hasegawa, Complexation behavior of Sr<sup>II</sup> and geochemically-related elements (Mg<sup>II</sup>, Ca<sup>II</sup>, Ba<sup>II</sup>, and Y<sup>III</sup>) with biodegradable aminopolycarboxylate chelators (GLDA and HIDS), *J. Mol. Liq.*, 242 (2017) 1123-1130.
- 3) H. Sawai, I.M.M. Rahman, C. Lu, Z.A. Begum, M. Saito, H. Hasegawa, Extractive decontamination of cesium-containing soil using a biodegradable aminopolycarboxylate chelator, *Microchem. J.*, 134 (2017) 230-236.
- 4) Z.A. Begum, I.M.M. Rahman, Y. Tate, T. Ichijo, H. Hasegawa, Binding of proton and iron to lignite humic acid size-fractions in aqueous matrix, *J. Mol. Liq.*, 254 (2018) 241-247.
- 5) H. Hasegawa, S. Barua, T. Wakabayashi, A. Mashio, T. Maki, Y. Furusho, I.M.M. Rahman, Selective recovery of gold, palladium, or platinum from acidic waste solution, *Microchem. J.*, 139 (2018) 174-180.
- 6) M.K.M.Z. Hyder, M.A. Saleh, F. Hossain, S.H. Mir, K. Iwakabe, I.M.M. Rahman, Viscometric studies of molecular interactions in binary liquid mixtures of isomeric xylenes with methanol, *J. Chem. Eng. Data*, 63 (2018) 1370-1381.
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- 10) I. Alam, S. Barua, K. Ishii, S. Mizutani, M.M. Hossain, I.M.M. Rahman, H. Hasegawa, Assessment of health risks associated with potentially toxic element contamination of soil by end-of-life ship dismantling in Bangladesh, *Environ. Sci. Pollut. R.*, 26 (2019) 24162-24175.
- 11) Z.A. Begum, I.M.M. Rahman, T. Takase, H. Hasegawa, Formation and stability of the mixed-chelator complexes of Sr<sup>2+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, Ba<sup>2+</sup>, and Y<sup>3+</sup> in solution with bio-relevant chelators, *J. Inorg. Biochem.*, 195 (2019) 141-148.
- 12) H. Hasegawa, M.A.A. Mamun, Y. Tsukagoshi, K. Ishii, H. Sawai, Z.A. Begum, M.S. Asami, T. Maki, I.M.M. Rahman, Chelator-assisted washing for the extraction of lead, copper, and zinc from contaminated soils: A remediation approach, *Appl.*

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2. 論文標題 Chelator-induced recovery of rare earths from end-of-life fluorescent lamps with the aid of mechano-chemical energy	5. 発行年 2018年
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2. 論文標題 Binding of proton and iron to lignite humic acid size-fractions in aqueous matrix	5. 発行年 2018年
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2. 論文標題 Complexation behavior of SrII and geochemically-related elements (MgII, CaII, BaII, and YIII) with biodegradable aminopolycarboxylate chelators (GLDA and HIDS)	5. 発行年 2017年
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2. 論文標題 Formation and stability of the mixed-chelator complexes of Sr <sup>2+</sup> , Mg <sup>2+</sup> , Ca <sup>2+</sup> , Ba <sup>2+</sup> , and Y <sup>3+</sup> in solution with bio-relevant chelators	5. 発行年 2019年
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2. 論文標題 Chelator-assisted washing for the extraction of lead, copper, and zinc from contaminated soils: A remediation approach	5. 発行年 2019年
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2. 出版社 IntechOpen: London	5. 総ページ数 76
3. 書名 Metals in Soil-Contamination and Remediation	

〔産業財産権〕

〔その他〕

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6. 研究組織

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