論文 / 著書情報
Article / Book Information

題目(和文)	
Title(English)	Development of Novel Micro Total Analysis System for Radioactive Waste Management and Decommissioning
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出典(和文)	学位:博士(学術), 学位授与機関:東京工業大学, 報告番号:甲第12047号, 授与年月日:2021年6月30日, 学位の種別:課程博士, 審査員:塚原 剛彦,竹下 健二,加藤 之貴,鷹尾 康一朗,吉田 克己
Citation(English)	Degree:Doctor (Academic), Conferring organization: Tokyo Institute of Technology, Report number:甲第12047号, Conferred date:2021/6/30, Degree Type:Course doctor, Examiner:,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

（博士課程）
Doctoral Program

論 文 要 旨

THESIS SUMMARY

系・コース： Department of, Graduate major in	応用化学系 系 原子核工学 コース	申請学位（専攻分野）： Academic Degree Requested	博士 （学術 ） Doctor of
学生氏名： Student's Name	BRANDT Aileen	指導教員（主）： Academic Supervisor(main)	塚原 剛彦
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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

Since the Fukushima Nuclear power plant accident, increasing decommissioning efficiency is imperative to revive the disaster-struck area, making waste management a top priority. The decommissioning strategies outlines the necessity of improving R&D in the radioactive waste characterization area, relating to the development of portable screening and detection devices in and around the decommissioning site. Therefore, this research will focus on the development of screening devices utilizing microfluidic extraction and detection technology. The microfluidic screening systems were developed based on future target elements of ^{79}Se, ^{154}Eu, ^{241}Am, ^{244}Cm and lanthanide group elements.

Chapter 1 - Introduction & literature review - Will describe the need to measure specific nuclides related to the decommissioning strategy, emphasizing the advantageous properties of using microfluidic devices as separation and detection technologies. A literature review was conducted on Fukushima decommissioning strategies and the challenges therein. Highlighting current analytical practices and their need to improve these methods for sampling fuel debris, contaminated wastewater and environmental samples. Microfluidic technology was examined in the nuclear field context and including the relevant theory needed to understand microfluidic operations.

Chapter 2 - Se(IV) microchip extraction with fluorescence microscope detection - The future target nuclide is ^{79}Se, thus developing Se(IV) extraction and detection system. This system used 2,3- Diaminonaphthalene(DAN) ligand which selectively reacts with Se(IV) in 0.1 M HCl conditions, and forms a fluorescent selenium complex 4,5-Benzopiazselenol (BPS). This property could be used to set-up a microfluidic platform coupled to a fluorescence microscope for in situ detection for Se(IV) analysis. In addition, the in-situ detection results were validated with ICP-MS and a fluorescence spectrometer. Extraction kinetics for Se(IV) was determined using the microfluidic set-up with fluorescence microscopy. The overall extraction efficiency of microfluidic device was 75 % for a 20-cm length channel in 6 s, evaluation of the extraction kinetics determined that a residence time of 20 s would be needed for the microchip to reach an equilibrium batch extraction of 93 %. Moreover, the fluorescence microscope detection limit was found to be 0.7 µM for Se(IV) ions. The pH region Se(IV) extraction conditions make it suitable for this system to be used in contaminated wastewater screening.

Chapter 3 - Microchip lanthanide group extraction - The future target detection nuclides for this system were ^{241}Am and ^{244}Cm, where Nd and Sm were used to simulate their extraction in multi lanthanide solution using conventional ICP-MS detection. The same parallel flow microchip in chapter 3 was used to investigate extraction performance into dodecane with ligands N,N,N′,N′-Tetraoctyl diglycolamide (TODGA), octyl (phenyl)-N, N-diisobutyl carbamoyl methyl phosphine oxide (CMPO), and 2 -ethylhexyl diamide amine (ADAAM). It was found that the ADAAM system achieved 35 x higher Nd/Sm separation in the microchip compared to the batch, with a separation factor of 706 and 40 % Nd extraction. In addition, low concentrations of TODGA at 3 M HNO_3 could also improve separation factor of light/heavy lanthanides in the microchip. Furthermore, high TODGA concentrations at 3 M HNO_3 allowed for overall lanthanide extraction of 94-96 % in 7 s. This system could expedite group lanthanide extraction, improving mutual separation between light and heavy lanthanides and improving separation between ^{241}Am and ^{244}Cm. It proved to be a suitable compliment to existing detection systems for samples in highly acidic

conditions such as those derived from spent nuclear fuel.

Chapter 4 - Microtube Eu(III)extraction and thermal lens spectroscopy (TLS) system - The future target analyte was ^{154}Eu, which was simulated with Eu(III) in microtube extraction system with plug flow coupled to a TLS system. Eu(III) was extracted in two different phases, the ionic liquid phase 1-butyl-3-methylimidazolium bis[(trifluoromethyl)sulfonyl] imide ($[C_4min][NTf_2]$) and the organic dodecane(DD) phase, using the same TODGA extractant. The two extraction systems were evaluated based on extraction performance by varying HNO_3 concentration, residence time and flow velocity. The DD microtube system achieved 98 % extraction corresponding to 100 % extraction efficiency at 3 M HNO_3 within 5 s, where the IL system achieved 96% extraction with 97 % efficiency from 0.001 M HNO_3 in 15 s. In addition, IL extraction results were also confirmed using TLS obtaining a detection limit of 0.1 μM for Eu(III) ions. The IL and DD systems showed high extraction efficiencies in low and high acidic media making it possible to use the same system configuration for a range of analytical samples just by changing the extraction solvent. Furthermore, successful detection using the TLS system for sub-micromolar concentration provides an alternative for analytical device for efficient in field elemental screening a result of its small footprint.

Chapter 5 - Microtube lanthanide group extraction - Future lanthanide separation and detection was the focus of this section. Based on the TODGA extraction performance for lanthanide in the microscope and high extraction efficiency provided by the microtube system, investigation was extended to extraction of 14 lanthanides in the microtube system with the TODGA extractant. Change in HNO_3 concentration, residence time and velocity was evaluated and found improved extraction of all lanthanides over the microchip system with 96-98 % in 25 s. Furthermore, it was found that by adjusting the extraction velocity 100 % separation of La,Ce and Pr could be done while maintaining > 90 % elemental extraction of Dy-Lu at 0.5 M HNO_3 in 15 s. The results again proved the efficient use of microfluidic systems for group lanthanide extraction and coupling to existing detection systems.

Chapter 6 - Conclusions and future work - In conclusion, this research provided ample evidence for deploying microfluidic extraction and detection for use in elemental screening at Fukushima, and proved its separation efficiencies all in less than 30 s. Furthermore, the coupling of novel and existing devices was also successful making device integration on-site possible. Future work for this research includes hydrodynamic modeling for optimal chip designs to further improve performance and also do back pressure studies for enabling tube to chip integration for improving in-situ detection.