

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
著者(和文)	BRANDTAileen
Author(English)	Aileen BRANDT
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)	要約
Type(English)	Outline

# Development of Novel Micro Total Analysis System for Radioactive Waste Management and Decommissioning

*by*

Aileen Brandt

A Thesis submitted in partial fulfilment  
of the requirements for the Degree

*of*

DOCTOR OF PHILOSOPHY IN ENGINEERING  
(NUCLEAR ENGINEERING)

Department of Chemical Science and Engineering  
Graduate School of Materials and Chemical Technology

Supervisor

Assoc. Prof. Takehiko TSUKAHARA



TOKYO INSTITUTE OF TECHNOLOGY  
Tokyo, Japan

June 2021

## Table of Contents

Declaration .....	i
Abstract .....	ii
Acknowledgements .....	v
Table of Contents .....	vi
Abbreviations .....	xi
Glossary .....	xiii
Table of Figures .....	xiv
List of Symbols .....	xx
List of Tables .....	xxi
1    Introduction and Literature Review.....	1
1.1    Motivation for research.....	2
1.2    Aims and objectives.....	2
1.3    Scope and thesis layout.....	2
1.4    Fukushima decommissioning project .....	4
1.5    Fukushima decommissioning strategy .....	5
1.6    Nuclides selection waste management strategy for Fukushima decommissioning ...	6
1.6.1    Challenges associated with sample collection and analysis .....	9
1.6.2    Target nuclides for detection in this research .....	9
1.6.3    Nuclide sources at Fukushima Daiichi : fission products (FP).....	10
1.6.4    Radioactive waste categorization at TEPCO and internationally .....	10
1.6.5    Future of characterised waste.....	13
1.7    Difficult to measure (DTM) nuclides .....	14
1.8    Analytical methodologies and validation.....	14
1.9    Microfluidic background .....	15
1.9.1    What is microfluidics and microfluidic chips? .....	15

1.9.2	Applications, benefits and limitations of microfluidic devices .....	16
1.10	Nuclear field applications of microfluidic devices .....	17
1.11	Importance of flow types and extraction for detection .....	19
1.11.1	Flow theory of microfluidic devices .....	19
1.11.2	Design and fabrication of microfluidic chips.....	23
1.11.3	Extraction inside the channel.....	28
1.12	Detection techniques.....	31
1.12.1	Thermal lens spectroscopy (TLS).....	31
1.12.2	Solvent selection for TLS detection techniques.....	33
1.13	Ionic liquids .....	34
1.14	Research questions.....	35
1.15	References.....	36
2	Se(IV) Microchip Extraction with Fluorescence Detection .....	40
2.1	Overview of selenium .....	42
2.1.1	General information on selenium.....	42
2.1.2	Chemical properties, uses and effects of selenium .....	43
2.1.3	Current methods of selenium detection .....	44
2.2	Selenium relevance in nuclear decommissioning .....	46
2.3	Materials and methods .....	48
2.3.1	BPS formation.....	48
2.3.2	Liquid-liquid batch extraction.....	49
2.3.3	Microfluidic setup and operation .....	49
2.4	Results and discussion .....	52
2.4.1	Evaluation of BPS formation .....	52
2.4.2	Liquid-liquid extraction in batch .....	53

2.4.3	Micro-extraction: fluorescence calibration curve .....	55
2.5	Selenium system conclusions .....	64
2.6	References.....	65
3	Microchip Lanthanide Group Extraction.....	67
3.1	Introduction for lanthanide separation and detection in microchips.....	68
3.2	Lanthanide overview.....	68
3.3	Lanthanide detection in microfluidics .....	69
3.4	Materials and methods .....	70
3.4.1	Chemicals and solutions .....	70
3.4.2	Experimental methods .....	72
3.5	Results and discussion .....	72
3.5.1	Batch and microscale with change in ligands .....	72
3.5.2	Change in TODGA concentration and third phase formation (TPF).....	77
3.5.3	Change in nitric acid concentration with TODGA extraction system .....	83
3.5.4	Microscale overall mass transfer with change in TODGA concentration .....	85
3.6	Conclusions.....	85
3.7	References.....	87
4	Microtube Eu(III) Extraction and Thermal Lens Spectroscopy (TLS) .....	90
4.1	Introduction.....	90
4.2	Overview of europium in decommissioning.....	90
4.3	Materials and methods .....	92
4.3.1	Chemicals.....	92
4.3.2	Experimental setup for IL flow map.....	93
4.3.3	Experimental setup for DD/IL extraction .....	94
4.3.4	Batch experimental method .....	95

4.3.5 Microchannel extraction experimental method .....	96
4.3.6 TLS calibration and detection .....	96
4.4 Results and discussion .....	97
4.4.1 IL and DD Batch scale results .....	97
4.4.2 IL system flow map .....	99
4.4.3 Microchannel experiments.....	101
4.4.4 Eu(III) ions using TLS system.....	106
4.5 Conclusions.....	111
4.6 References.....	113
5 Microtube Lanthanide Group Extraction.....	115
5.1 Introduction.....	115
5.2 Materials and methods .....	116
5.2.1 Chemicals.....	116
5.2.2 Experimental setup.....	116
5.3 Results and discussion .....	117
5.3.1 Batch extraction .....	117
5.3.2 Microchannel extraction .....	117
5.3.3 Microchip and microtube comparison .....	127
5.4 Conclusions.....	130
5.5 References.....	132
6 Conclusions and Recommendations for Future Developments.....	134
6.1 Introduction.....	135
6.2 Chapter 2 - Se(IV) microchip extraction with fluorescence detection .....	135
6.3 Chapter 3 - Microchip lanthanide group extraction.....	137

6.4	Chapter 4 - Microtube Eu(III) extraction and detection with TODGA/IL and TODGA/dodecane systems .....	139
6.5	Chapter 5 - Microtube lanthanide group extraction.....	142
6.6	Future application - Cerium(IV) extraction with TLS detection system and portal total analysis system for future land remediation.....	145
6.7	References.....	147
7	Appendix .....	148
7.1	Appendix A – Selenium system.....	149
7.2	Appendix B – Lanthanide microchip extraction .....	159
7.3	Appendix C - Comparison of lanthanide extraction in parallel and plug flow .....	162
7.4	Appendix D - Derivation of mass transfer equations.....	163