



## Monazite-type ceramics as nuclear waste form: crystal structure, microstructure and properties

Yulia Arinicheva

Energie & Umwelt / Energy & Environment

Band / Volume 459

ISBN 978-3-95806-397-6

Forschungszentrum Jülich GmbH  
Institute of Energy and Climate Research  
Nuclear Waste Management and Reactor Safety (IEK-6)

## **Monazite-type ceramics as nuclear waste form: crystal structure, microstructure and properties**

Yulia Arinicheva

Schriften des Forschungszentrums Jülich  
Reihe Energie & Umwelt / Energy & Environment

Band / Volume 459

---

ISSN 1866-1793

ISBN 978-3-95806-397-6

## Table of Contents

Abstract .....	1
List of publications .....	5
1 Introduction .....	7
2 Scientific and theoretical background .....	11
2.1 Monazite-type ceramics for conditioning of actinides .....	11
2.2 Rhabdophane compounds .....	14
2.3 Synthesis and fabrication of monazite ceramics.....	17
2.4 Structural investigations of solid solutions with monazite and rhabdophane structures ..	19
2.4.1 Surrogates for minor actinides and plutonium .....	19
2.4.2 Actinide incorporation into the monazite structure.....	21
2.4.3 Actinide incorporation into the rhabdophane structure .....	22
2.4.4 Analytical methods for probing the structural homogeneity of solid solutions ..	23
2.5 Properties of monazite-type ceramics.....	28
2.5.1 Thermodynamic properties.....	28
2.5.2 Thermal and mechanical properties .....	30
2.5.3 Dissolution kinetics .....	31
3 Aim of the work .....	35
4 Materials and methods.....	36
4.1 Synthesis and aftertreatment.....	36
4.1.1 Synthesis of rhabdophane and monazite samples for structural and thermodynamic studies of solid solutions formation.....	39
4.1.2 Synthesis and fabrication of $\text{La}_{1-x}\text{Eu}_x\text{PO}_4$ ( $x = 0 - 1$ ) samples for studies on sintering and/or physical and mechanical properties of monazite-type ceramics .....	41
4.2 Experimental and computational methods.....	45
4.2.1 Elemental analysis .....	45
4.2.2 X-ray diffraction and Rietveld refinement .....	46
4.2.3 Environmental scanning electron microscopy .....	46
4.2.4 Spectroscopic methods for short-range structural studies on solid solutions formation	
47	
4.2.5 Atomistic modelling .....	49
4.2.5.1. <i>Ab initio</i> and force-field simulations of $\text{La}_{1-x}\text{Gd}_x\text{PO}_4$ solid solutions .....	49
4.2.5.2. <i>Atomistic simulations of <math>\text{Cm}^{3+}</math> incorporation in monazite and rhabdophane.....</i>	50
4.2.6 Methods for studies on physical, thermodynamic and mechanical properties .....	52
4.2.7 Dissolution experiments .....	56
5 Results and discussions .....	59
5.1 The effect of the synthesis route on crystal structure and morphology of monazite precursor powders .....	59

5.1.1 Thermal behaviour of monazite precursor powders from different synthesis methods .....	59
5.1.2 Crystal structure of monazite precursor powders from different synthesis methods .....	60
5.1.3 Morphology of monazite precursor powders from different synthesis methods ...	65
5.1.4 Résumé of Chapter 5.1 .....	66
5.2 The effect of powder morphology on sintering kinetics and microstructure of sintered pellets.....	67
5.2.1 Characterization of monazite powders and green specimens.....	67
5.2.2 Sintering kinetics and microstructure evolution.....	68
5.2.3 Résumé of Chapter 5.2 .....	76
5.3 Structural characterization .....	77
5.3.1 $\text{La}_{1-x}L_n\text{xPO}_4$ -monazite solid solutions ( $Ln = \text{Gd, Eu}$ ).....	77
5.3.2 Rhabdophane-type solid solutions.....	92
5.3.3 Structural incorporation of $\text{Cm}^{3+}$ in lanthanide orthophosphates with rhabdophane and monazite structures .....	101
5.3.4 Structural incorporation of $\text{Pu}^{+3}$ in lanthanide orthophosphates with rhabdophane and monazite structures .....	121
5.3.5 Résumé of Chapter 5.3 .....	132
5.4 Properties of monazite-type ceramics.....	136
5.4.1 Thermodynamic properties of $\text{La}_{1-x}L_n\text{xPO}_4$ ( $Ln = \text{Gd, Eu}$ ) .....	136
5.4.2 Physical properties .....	148
5.4.3 Mechanical properties .....	152
5.4.4 Dissolution: Influence of temperature on dissolution kinetics of synthetic $\text{LaPO}_4$ -monazite in acidic media.....	154
5.4.5 Résumé of Chapter 5.4.....	161
6 Conclusions and outlook .....	164
6.1. Conclusions.....	164
6.2. Outlook .....	166
7 Appendix .....	168
8 References .....	175

Energie & Umwelt / Energy & Environment  
Band / Volume 459  
ISBN 978-3-95806-397-6

Mitglied der Helmholtz-Gemeinschaft

