

Accepted Manuscript

Influence of temperature on the dissolution kinetics of synthetic LaPO_4 -monazite in acidic media between 50 and 130 °C

Yulia Arinicheva, Clemence Gausse, Stefan Neumeier, Felix Brandt, Konstantin Rozov, Stéphanie Szenknect, Nicolas Dacheux, Dirk Bosbach, Guido Deissmann

PII: S0022-3115(18)30374-X

DOI: [10.1016/j.jnucmat.2018.07.009](https://doi.org/10.1016/j.jnucmat.2018.07.009)

Reference: NUMA 51068

To appear in: *Journal of Nuclear Materials*

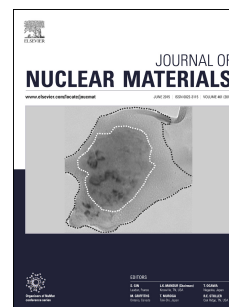
Received Date: 13 March 2018

Revised Date: 8 June 2018

Accepted Date: 5 July 2018

Please cite this article as: Y. Arinicheva, C. Gausse, S. Neumeier, F. Brandt, K. Rozov, Sté. Szenknect, N. Dacheux, D. Bosbach, G. Deissmann, Influence of temperature on the dissolution kinetics of synthetic LaPO_4 -monazite in acidic media between 50 and 130 °C *Journal of Nuclear Materials* (2018), doi: [10.1016/j.jnucmat.2018.07.009](https://doi.org/10.1016/j.jnucmat.2018.07.009).

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.



Influence of temperature on the dissolution kinetics of synthetic LaPO₄-monazite in acidic media between 50 and 130°C

Yulia Arinicheva^{1,2*}, Clemence Gausse³, Stefan Neumeier¹, Felix Brandt¹, Konstantin Rozov^{1,4}, Stéphanie Szenknect³, Nicolas Dacheux³, Dirk Bosbach¹, Guido Deissmann¹

¹Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research (IEK-6): Nuclear Waste Management and Reactor Safety, 52425 Jülich, Germany

²Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research (IEK-1): Materials Synthesis and Processing, 52425 Jülich, Germany

³ICSM, CNRS, CEA, ENSCM, Univ. Montpellier, Site de Marcoule, Bât 426, BP 17171, 30207 Bagnols/Cèze, France

⁴Institute of Environmental Geology, Russian Academy of Sciences, St. Petersburg State University, St. Petersburg 199004, Russia

*corresponding author.

E-mail address: y.arinicheva@fz-juelich.de (Y. Arinicheva).

Abstract

Single-phase monazite-type ceramics are discussed as waste forms for the safe disposal of surplus plutonium or separated minor actinides. To gain a deeper insight into the mechanism governing the dissolution of monazite-type compounds, the dissolution kinetics of synthetic LaPO₄-monazite was systematically studied by dynamic dissolution experiments carried out in the temperature range from 50 to 130°C in acidic media (0.01M HNO₃). The dissolution rates at far from equilibrium conditions increased from $3.2 \times 10^{-5} \text{ g m}^{-2} \text{ d}^{-1}$ at 50°C to $2.5 \times 10^{-4} \text{ g m}^{-2} \text{ d}^{-1}$ at 130°C. Two different temperature regions were observed, in which the normalised dissolution rates of LaPO₄ have a diverging temperature dependence, indicating two different mechanisms of dissolution: namely surface-controlled dissolution ($T = 50 - 90^\circ\text{C}$; $E_a = 44 \text{ kJ mol}^{-1}$) and transport-controlled dissolution ($T = 90 - 130^\circ\text{C}$; $E_a = 7.5 \text{ kJ mol}^{-1}$). Complementary thermodynamic modelling studies of the dissolution of LaPO₄ at the experimental conditions show that La-rhabdophane (LaPO₄·0.667H₂O) is the thermodynamically favoured phosphate phase in aqueous environments below about 100°C. Apparently, the hydration of monazite and the formation of a thin surface layer consisting of rhabdophane is an intermediate step controlling the dissolution kinetics of the LaPO₄ monazite ceramics in low temperature aqueous environments.

Download English Version:

<https://daneshyari.com/en/article/7963016>

Download Persian Version:

<https://daneshyari.com/article/7963016>

[Daneshyari.com](https://daneshyari.com)