

[Abstract for Global-2001, to be held in Paris, France, Sept. 8-13, 2001]

Transmutation of americium and curium using zirconia-based host materials*

P.E. Raison¹ and R.G. Haire²

¹ Commissariat à l'Energie Atomique, CEA-Cadarache DRN/DEC/SPUA/LACA
13108 Saint Paul lez Durance- France, Philippe.Raison@cea.fr

² Oak Ridge National Laboratory, P.O Box 2008,
Oak Ridge, TN 37831-6375 - USA, rgh@ornl.gov

"The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-00OR22725. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes."

A contractor of the U.S. Government under contract DE-AC05-00OR22725 has authored this manuscript. The U.S. Government retains a non-exclusive, royalty-free license to publish or reproduce the published form of this document, or allow others to do so, for U.S. Government purposes.

GLOBAL 2001
(2.2 Fuels and materials technologies)

**Transmutation of americium and curium
using zirconia-based host materials**

P.E. Raison¹ and R.G. Haire²

¹ Commissariat à l'Energie Atomique, CEA-Cadarache DRN/DEC/SPUA/LACA
13108 Saint Paul lez Durance- France, Philippe.Raison@cea.fr

² Oak Ridge National Laboratory, P.O Box 2008,
Oak Ridge, TN 37831-6375 - USA, rgh@ornl.gov

Abstract

We have investigated the incorporation of americium and curium in selected zirconia-based materials in conjunction with a research program at the "Commissariat à l'Energie Atomique" that addresses transmutation of long-lived radioactive elements. Both cubic zirconia and pyrochlore oxides $\text{An}_2\text{Zr}_2\text{O}_7$ ($\text{An} = \text{Am}, \text{Cm}$) are considered in the work reported here.

The strategy proposed is to treat americium and curium together in the same transmutation process. There are several incentives for this approach. One is the radiotoxicity benefits [1-2] while another is avoiding the difficult separation of Am and Cm. A third point is that curium must already be considered as it is generated in large amounts as a result of irradiating pure americium targets.

Outlined here are our efforts to examine the behavior of Am and Cm in selected uranium-free hosts, which avoids the generation of additional actinide products. The general concept consists of irradiating the host targets for extended periods, which would be then disposed in a suitable repository (the so-called "once through option").

The host matrix selected for Am and Cm must meet various criteria, such as a low neutron capture cross-section, a high melting point, phase stability, low oxygen potential, etc. Several potential candidates have been envisioned but the ultimate material may be a suitable "rock-like" product able to sustain harsh irradiation conditions as well as the long-term repository environment.

We initiated our studies for a host material with ZrO_2 -based compounds and concluded that the cubic forms, obtained by stabilizing zirconia with about 25mol% of Y_2O_3 , should be attractive for this technological application. We have demonstrated that $(\text{Zr}_{0.6}\text{Y}_{0.4})\text{O}_{1.8}$, or similar compositions, can incorporate significant amounts of americium dioxide by forming fluorite-type cubic solid solutions. It was also found that evolution of the cell parameters of these products is linear with the americium content.

Curium is generally considered as a trivalent element, although it can also exhibit a tetravalent oxidation state (e.g., CmO_2). Based on the consideration of both radius and oxidation state, we found that within certain limits Cm(III) can also stabilize the cubic form of ZrO_2 , as do other trivalent elements (e.g. Y^{3+} , R.E.^{3+} , etc.). To evaluate this capability of curium, we have prepared, on a milligram level, various $(^{248}\text{Cm}_x\text{Zr}_{1-x})\text{O}_{2-2/x}$ materials (with $x = 0$ to 0.5) and studied them by X-ray diffraction.

Another zirconia-based host for Am(III) and Cm(III) was also examined in this work; specifically, pyrochlores of the formula, $\text{An}_2\text{Zr}_2\text{O}_7$ ($\text{An} = \text{Am}, \text{Cm}$) which also form a cubic structure. These zirconia pyrochlore oxides are refractory ceramics with high melting points and are able to incorporate larger quantities of actinides per volume unit than the cubic solid solutions of (Zr-Y) -oxides. These pyrochlore oxides may also offer other advantages over cubic solid solutions of zirconia (e.g., higher thermal conductivity, lower oxygen potential, phase stability, etc.).

In our presentation we shall report on our recent experimental results concerning the incorporation and the physicochemistry of both ^{243}Am and ^{248}Cm in cubic zirconia and Zr-based pyrochlore oxides. In addition, a comparison of the advantages and disadvantages of these two host materials for transmutation of Am, Cm and/or Am-Cm mixtures will be discussed.

- [1] M. Delpech, J. Tomasi, A. Zaetta, H. Money, M. Salvatores, G. Vambenepe, Proc. 5th Actinide Partitioning and Transmutation, Mol, Belgium, Nov. 25-27, (1998) 283.
- [2] "Actinide and fission product partitioning and transmutation"
Status and assessment report, Nuclear Energy agency, OECD, 1999

* Research sponsored by Commissariat à l'Energie Atomique, CEA-Cadarache DRN/DEC/SPUA/LACA 13108 Saint Paul lez Durance- France, and by the Division of Chemical Sciences, Gepsciences and Biosciences, Office of Basic Energy Sciences, U.S. Department of Energy, under contract DE-ACO5-00OR22725 with Oak Ridge National Laboratory, managed and operated by UT-Battelle, LLC.