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Waste Forms for Plutonium Disposition

by

S.G. Johnson, T.P. O'Holleran, S.M. Frank, M. Meyer, M. Noy,
B.A. Staples, D. Knecht, and P.C. Kong.

Argonne National Laboratory-West
P.O. Box 2528
Idaho Falls, ID 83403-2528

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DISCLAIMER

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S. G. Johnson*, T. P. O'Holleran, S. M. Frank, M. K. Meyer, M. Hanson
Argonne National Laboratory West
P.O. Box 2528
Idaho Falls, ID 83403

B. A. Staples, D. A. Knecht, P. C. Kong
Lockheed Martin Idaho Technologies Co.
Idaho Falls, ID 83415

Introduction

The field of plutonium disposition is varied and of much importance, since the Dept. of Energy has decided on the hybrid option for disposing of the weapons material. This consists of either placing the Pu into mixed oxide fuel for reactors or placing the material into a stable waste form such as glass [1]. The waste form used for Pu disposition should exhibit certain qualities: 1) provide for a suitable deterrent to guard against proliferation, 2) be of minimal volume, i.e., maximize the loading and 3) be reasonably durable under repository-like conditions. This paper will discuss several Pu waste forms that display promising characteristics.

Results and Discussion

Our experiments are focused on two separate areas of interest in the plutonium disposition arena. The first involves taking metallic plutonium and placing it directly into a waste form with no prior processing. Most current waste form schemes call for a step involving oxidizing the metal to PuO_2. This is a process that requires additional facilities and subsequently adds further contamination sources to the overall process.

We have attempted to place Pu metal directly into a stable glass-ceramic form using only an ordinary furnace. This can be accomplished using conventional glass processing temperatures (1100 °C) with reasonable casting times (72 hr) and with attractive plutonium loadings of 15 wt%. Our results indicate that stable Pu-containing phases are formed, such as cubic (fluorite structure), that incorporate high Pu loadings. This experiment was analyzed using x-ray diffraction (XRD), scanning electron microscopy (SEM) and performance testing methods. The low magnification SEM micrograph of a core sample from a crucible-sized experiment is featured in Fig. 1. A small portion of the original Pu piece, now oxidized, is visible in the micrograph as are the various phases formed as the Pu dissolves into the glass. The XRD pattern confirms that no metallic plutonium is present.

The ability for the zirconia phase mentioned above to contain relatively high Pu-loadings. > 50 wt% makes it an attractive host for plutonium. An attempt to synthesize a phase pure c-zirconia that incorporates both Pu and Sm was undertaken. As a starting point the process parameters were first derived from surrogate experiments using CeO_2. Once this had been accomplished an experiment using PuO_2 was performed. This experiment involved two separate samples, one with PuO_2 and the second with PuO_2 and Sm_2O_3. These were analyzed using XRD, SEM and performance tests. The XRD pattern is featured in Fig. 2 below. This clearly shows a single c-zirconia type pattern that is different from PuO_2 and Sm_2O_3 in lattice parameter terms.

References

1) T. P. O'Holleran, S. G. Johnson, S. M. Frank, M. K. Meyer, M. Noy, E. L. Wood, D. A. Knecht, K. Vinjamuri, B. A. Staples, Proceedings of the Fall 1996 Materials Research Society Meeting, Boston (1997).

Figure 1. The SEM back scattered image from a waste glass used to dissolve and stabilize a piece of plutonium metal. The various regions of interest are discussed in the text. The Pu, if totally dissolved, would represent 15 wt% of the product. The magnification is 15X.

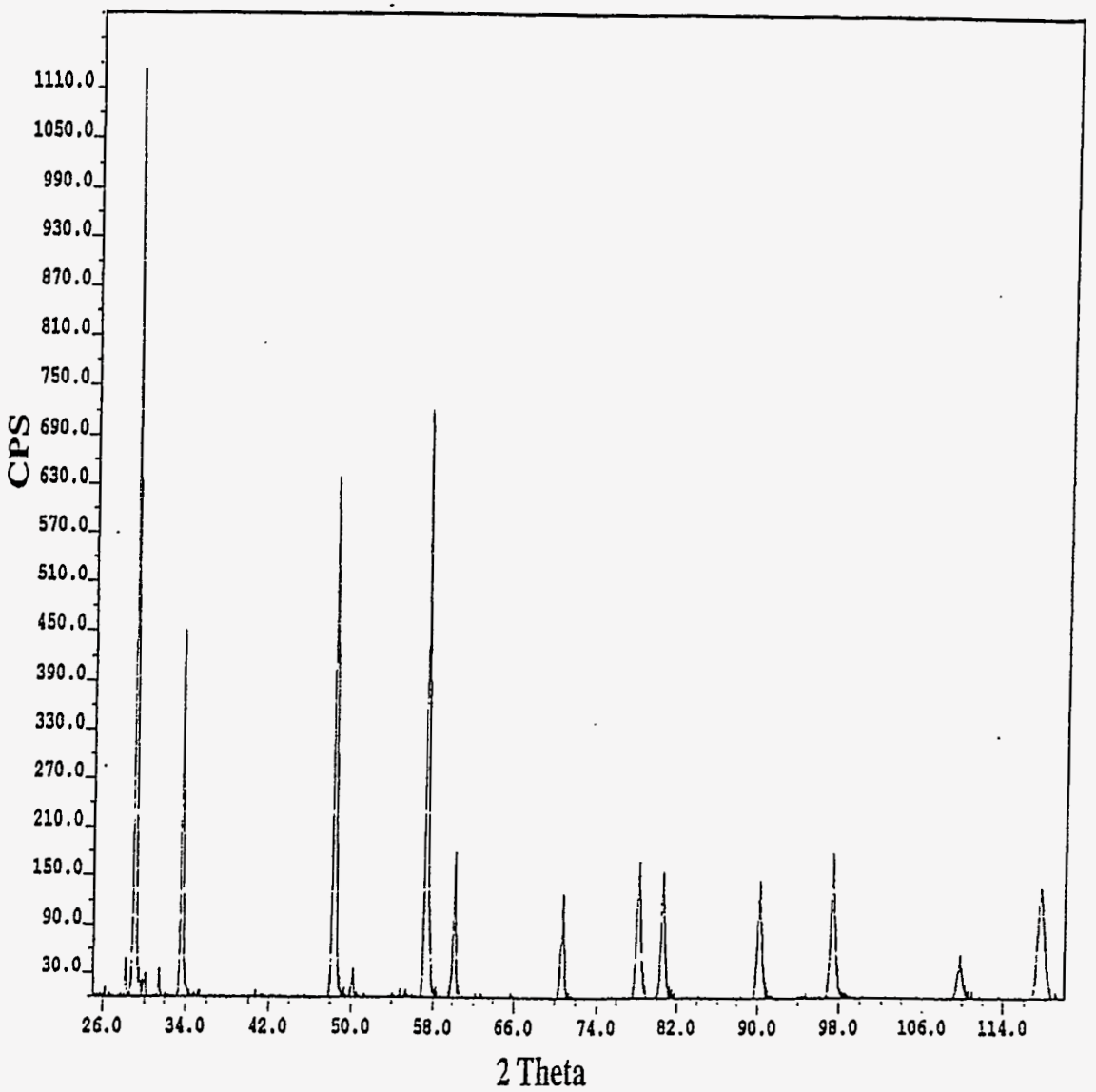

Figure 2. The XRD pattern of the pure phase zirconia with PuO_2, ZrO_2 and Sm_2O_3 constituents.