UCRL-JC-130180
PREPRINT

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This paper was prepared for submittal to the
DOE Spent Nuclear Fuel and Fissile Materials Management Topical Meeting
Charleston, SC
September 8-11, 1998

March 2, 1998

Lawrence Livermore National Laboratory

Investigations of Plutonium Immobilization into the Vitreous Compositions

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Disposal of radioactive waste is a central problem and among the most important concerns of the nuclear fuel cycle.The Russian concept of nuclear fuel-cycle management is aimed at reprocessing spent fuel with the maximum, economically justified extraction of useful components for their recycling. The technology currently used in Russia for reprocessing spent nuclear fuel gives rise to liquid high-level waste (HLW) with minor concentrations of valuable components such as uranium (U) and plutonium (Pu) [1]. The liquid radioactive wastes formed in the course of reprocessing are converted into the solid forms suitable for the transportation, storage, and burial.

Of special importance is management of high-level waste (HLW). Although various technological approaches underlying the processes for the solidification or immobilizatin of liquid HLW are used at the research institutes of the MINATOM RF [1–5], all these approaches have in common the idea of a strong bonding of radionuclides in the resulting solid matrices. Therefore, development of solidification technologies must include the mandatory stages of investigating the behavior of HLW components during the immobilization process and in the prepared solidified compositions and characterizing their properties under conditions for subsequent transportation, storage, and burial.

An important technological area of exploration is study of the behavior of long-lived alpha radionuclides during the course of the vitrification process and the ultimate long-range influence of these radionuclides on the properties of the immobilized forms. For the most part, immobilization of alpha radionuclides, particularly plutonium, in vitreous compositions involves investigations on the properties of final materials and the effect of alpha-decay radiation on the synthesized solid compositions.

Another direction of investigation is study on the behavior of plutonium and transplutonium elements upon vitrification of liquid HLW, as applied to the one-stage process for immobilizing HLW by using different types of melters. Such studies were carried out to forecast the behavior of the above radionuclides during long-term operation of the ceramic melter at the vitrification facility of PU “Mayak.”

The results of many investigations on the behavior of plutonium upon immobilization into phosphate and borosilicate vitreous compositions developed in Russia are generalized and summarized in the present work.

In the conducted investigations of plutonium immobilization into both phosphate and borosilicate vitreous compositions used for the solidification of high-level liquid wastes upon vitrification in ceramic melters, plutonium exhibited a limited solubility in the studied glass matrices. The solubility of plutonium, using plutonium dioxide powders, in phosphate and borosilicate glasses of specifically studied compositions was limited to 0.2 –0.4 wt %.

The degree of incorporation (i.e., solubility) of plutonium, using plutonium in the form of nitrate solutions, in borosilicate glasses was also equal to 0.2–0.4 wt %.

The degree of incorporation (i.e., solubility) of plutonium, using plutonium in the form of nitrate solutions, in phosphate glasses depended considerably on the chemical compositions of the solution to be solidified and on the specific glass matrix (i.e., on the composition of final solidified product) and was equal to 0.4–1.0 wt %.

Available experimental data also allow one to assume that the use of the cold-crucible-induction melter (CCIM) method for immobilizing plutonium-containing wastes [6–8] provides a means of synthesizing the high-quality final solid-glass products with a plutonium content.

Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

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