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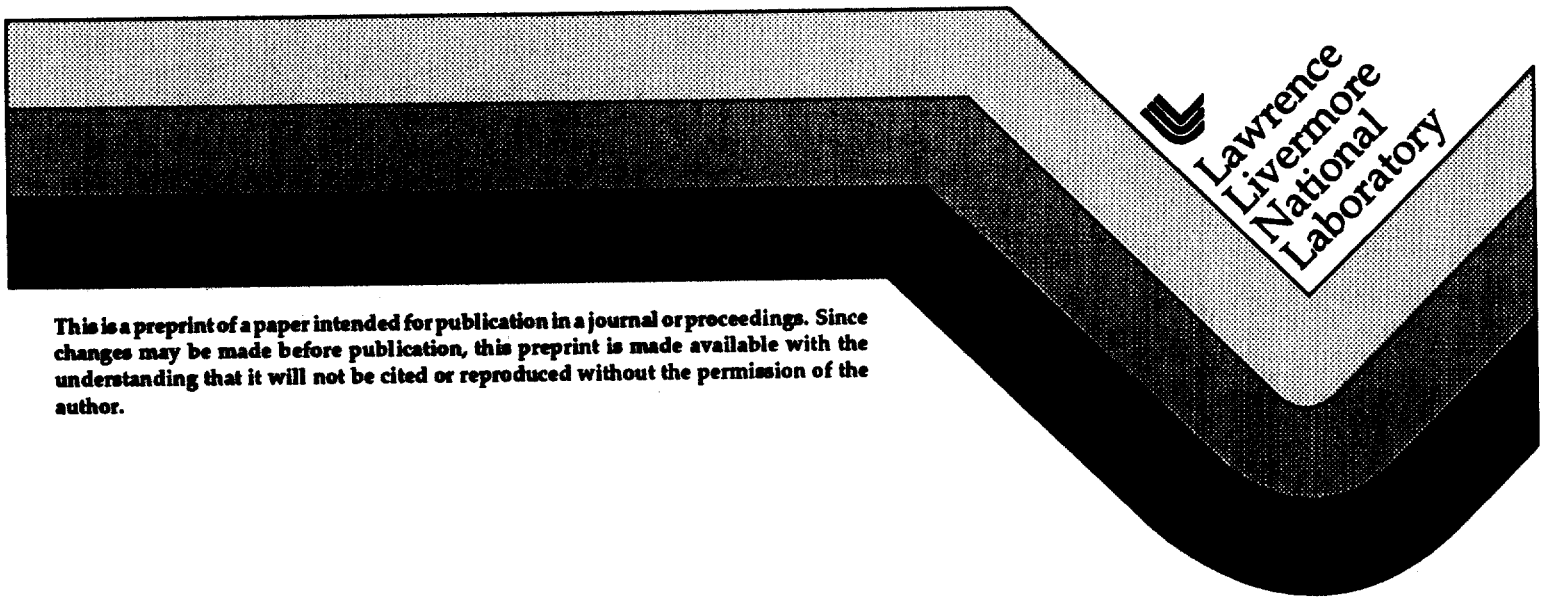

Dissolution Studies of Plutonium Oxide in LaBS Glass

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Introduction

As part of international agreement between the United States and Russia, a significant amount of plutonium requires disposition. One of the disposition paths is to immobilize it and dispose of it in a geological repository. The two favored immobilization forms are glass and ceramic. The plutonium, as an oxide, would be reacted with the glass or ceramic to form a homogenous material. The resulting solid product would then be encased in High-Level Waste (HLW) glass for the can-in-canister option. The HLW glass gives a radiation barrier to increase proliferation resistance. The glass canister would then be disposed of by geological emplacement.

1. Significant actinide solubility
2. Easily handled and processed in a glovebox environment
3. The product should be as durable as spent fuel at a minimum
4. The PuO_2 feed should be incorporated into the matrix without significant amount of unreacted material.

Both the glass and ceramic immobilized product need to meet all of these criteria. The ceramic option is described in other papers being presented at this conference. This paper discusses how glass meets criteria 1 and 4.

Description

The proposed glass formulation is a lanthanide boro-silicate (LaBS) glass. The formulation is shown in Table 1.

Table 1. Base LaBS Composition

Oxide	Weight %
SiO_2	25.80
B_2O_3	10.40
Al_2O_3	19.04
ZrO_2	1.15
Gd_2O_3	7.61
La_2O_3	11.01
Nd_2O_3	11.37
SrO	2.22
PuO_2	11.39

The initial poduction of the LaBS glass was done on the laboratory scale at Pacific Northwest National Laboratory (PNNL), Westinghouse Savannah River Company (WSRC) and Lawrence Livermore National Laboratory (LLNL). These mixes were static or with very little agitation (near static runs). They showed solubility of plutonium in the neighborhood of 10 wt% Pu.

Plutonium dissolution in the melt was also studied at LLNL using the Tilt-Pour furnace in the Plutonium Facility (Building 332). This furnace has the capability of making glass volumes of about 2 liters (about 10

kilograms of glass). The furnace has a platinum crucible and can stir the glass. After the completion of the dissolution, the product is poured into a stainless steel mold. The glass can be recovered from the molds. The Tilt-Pour Furnace was used to determine the effects of stirring, plutonium oxide particle size and residence time on the dissolution rate and solubility of plutonium in glass. These runs were a joint effort of LLNL, WSRC, and PNNL.

Results

In this paper the results of the static, near static and agitated runs will be discussed. The results will include the information about the dissolution rate as a function of particle size, residence time, Pu concentration and stirring. These results will greatly aid in the design and operation of the glass melter for whatever design is chosen. They will also help set the requirements for the feed pretreatment prior to the glass melter.

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