Technical Aspects of the Plutonium Vitrification Option (U)

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A paper proposed for presentation at the
American Nuclear Society National Meeting
Panel Session on Plutonium Management
Philadelphia, PA
June 26 - 29, 1995

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Vitrification of Pu and placement in a repository was one of two disposal options recommended by the National Academy of Sciences in a report issued in 1994 [1, 2].

Studies done at SRS and a review of international literature confirm that vitrification of surplus plutonium as borosilicate glass is a technically viable option. The technical basis for this conclusion derives in part from many decades of commercial production of this type of glass and the 30+ years of international development and use of borosilicate glass for stabilization of high-level radioactive waste from reprocessing of spent fuels, which has gained wide international acceptance.

A variety of processes and types of equipment are in commercial use throughout the world, producing borosilicate glass for a variety of large and small commercial applications, both radioactive and non-radioactive. Most of these technologies could be adapted to produce plutonium borosilicate glass, with or without added fission products. Although there are many differences among these applications, including feed compositions, melter designs, scale (size), and heating methods, the compositions and properties of most of the final glass products are similar. Heating methods in common use include joule heating (heating by resistance to the flow of electricity through the molten pool), magnetic induction, microwave, plasma, and radiant energy from resistance heaters. All of these heating methods work, and the best choice depends on the particular application, the throughput that is needed, feed composition, and final form requirements. Vitrification of high-level and low-level radioactive wastes is now becoming commonplace world-wide.

Within the nuclear community, vitrification of high-level radioactive wastes has been studied for over three decades. The oldest and most thoroughly demonstrated process is the one that was used in France at Marcoule, which with minor alterations is being used in the United Kingdom, and France (LaHague). This process calcines acidic waste, feeding the dry calcine to the melter. In the U.S., most of the high-level radioactive wastes (Savannah River Site and Hanford) are alkaline, and contain significant quantities of sodium and aluminum. For vitrification of these wastes it was judged appropriate to use a liquid slurry-fed melter. Germany and Japan processes also plan to use liquid-fed melters. The melter in use at the Defense Waste Processing Facility (DWPF) at the Savannah River site (SRS) has joule and radiant heating. It also has a cooling

* The information in this article was developed during the course of work under Contract No. DE-AC09-89SR18035 with the U.S. Department of Energy.

water jacket that freezes the glass in the interior insulation, which prevents molten glass from reaching the melter's steel shell and maintains the exterior at a low temperature to prevent up-drafts of contaminated air. To satisfy the need to demonstrate compliance with waste form criteria, this melter has the capability for sampling the molten glass as it pours from the melter into the canister.

Vitrification plants for high-level radioactive wastes are in commercial operation in France, the United Kingdom, and Belgium (with Germany), and will soon be operating in the, the U.S. and Japan. Vitrification plants for low-level waste are becoming commonplace. During the development of these technologies some data was obtained on the behavior of plutonium in this type of glass [3]. More recently, plutonium glasses have been studied, yielding sufficient data to increase confidence that plutonium can be safely and securely incorporated into a high-level waste glass at concentrations up to at least 4 to 5 % [4, 5] by weight, and at higher concentrations in glass without high-level waste components [6].

Specific technologies exist to vitrify plutonium as borosilicate glass, with or without added fission products or radioactive spiking materials. Vitrification without added fission products or radioactive spiking materials could be done in glovebox facilities with only modest shielding to protect the workers from the 0.060 Mev gamma radiation from Am-241, the daughter of Pu-241. If the radioactivity from either high-level waste or separated Cs-137 were incorporated in the glass as a proliferation deterrent, the process would require heavy shielding, i.e., remote operations in large cell or canyon-like facilities.

Preliminary criticality studies indicate that a high quality borosilicate glass can be made without adding neutron poisons other than the boron and lithium that are inherent ingredients in the glass [1]. The boron and lithium in one of SRS's reference glasses would assure criticality safety up to about 14 weight % Pu. This amount of poisoning would be adequate for safe long-term vault storage, where no mechanism exists for separating the poisons from the Pu. However, if the plutonium borosilicate glass were to be placed in a repository, nuclear criticality avoidance would be a significant issue because of the potential over geologic time for leaching to separate the more soluble boron and lithium from the Pu. Adding a rare earth neutron poison such as gadolinium, with chemistry more similar to the actinides, would reduce the risk of criticality over geologic time. The potential for long-term repository criticality from plutonium borosilicate glass and from U and Pu in spent fuel is now being assessed by the technical community, but this issue is not yet resolved. Indeed, there is a session on that subject at this conference.

The chemistry of the plutonium glass-making process would require some additional laboratory development and demonstration for this application. For example, tests would be conducted to determine the optimum composition of the glass for this specific purpose. Properties of the glass product, such as durability and leach rates of plutonium and other components, must be determined. More detailed calculations and modeling of nuclear criticality safety must be made that include other glass compositions and poisons, a variety of neutron moderating conditions, and plutonium isotopic variations. The effects of other poisons on plutonium solubility and glass properties needs to be determined, as does the leach rate of other candidate poisons. The volatility and entrainment of plutonium into the melter offgas needs to be determined for candidate melter systems as a function of melter temperature, offgas flow rate, and other process and equipment variables. If any measurable carry-over of plutonium into the offgas is possible, the design must detect and prevent plutonium accumulations in the offgas system that could lead to nuclear criticality accidents. Design features must also assure that criticality cannot occur throughout the process, including in the melter feed tanks or in the melter itself.

Vitrification of plutonium without added radiation sources may prove to be desirable regardless of whether or not national policies elect to retain the plutonium for its future energy production value or permanently dispose of it in a geologic repository. This form can be produced much faster, in existing facilities, than the form with fission products, and is a critically safe, stable, monitorable form. It could not quickly be converted to a weapon, so it would be advantageous for with safeguards and security. Yet, the plutonium in this glass could be recovered and purified for later use in reactor fuels when its energy value is higher and competitive with other energy sources. This recovery could be achieved using existing technology and facilities in any nation with nuclear capability, but would be more difficult in non-nuclear nations or by terrorist groups. On the other hand, if the U.S. or any other nation chooses to permanently dispose of its surplus plutonium in a repository with a radiation source, the plutonium glass (without radiation source) makes a near-ideal feed for a vitrification process that would incorporate the high-level waste or Cs-137.

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