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**IRON PHOSPHATE GLASSES: AN ALTERNATIVE FOR VITRIFYING CERTAIN
NUCLEAR WASTES**

Submitted By:

**Delbert E. Day (PI)
Chandra S. Ray
Cheol-Woon Kim**

Graduate Center for Materials Research
University of Missouri-Rolla, Rolla, MO 65409-1170
Ph: 573-341-4354
Fx: 573-341-2071
day@umr.edu

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Table of Contents

	Page
EXECUTIVE SUMMARY	2
INTRODUCTION AND RESARCH OBJECTIVES	5
NOTEWORTHY FEATURES OF IRON PHOSPHATE GLASSES	6
(1) Higher Waste Loading (Smaller Wasteform Volume)	6
(2) Excellent Chemical Durability	6
(3) Lower Melting Temperatures and Shorter Melting Times	7
(4) Minimum Corrosion of Refractory and Electrode Materials	8
a) Refractories	8
b) Electrodes	9
(5) Alternative Melting Methods	10
a) Cold Crucible Induction Melter (CCIM)	10
b) Hot Crucible Induction Melter (HCIM)	11
c) Microwave Melting	11
(6) Inexpensive Raw Materials	12
RELEVANCE AND IMPACT	12
PUBLICATIONS	13
PRESENTATIONS	16
COLLABORATIONS	17
REFERENCES	18

EXECUTIVE SUMMARY

The U.S. Department of Energy (DOE) Environmental Management Science Program (EMSP) has funded research on iron phosphate glasses at the University of Missouri-Rolla (UMR) for the period from September 15, 1996 to September 30, 2004 (DE-FG07-96ER45618). The final report for the initial period from September 15, 1996 to September 14, 2000 (project number 55110) has been submitted previously and can be found at www.osti.gov/em52/final_reports/55110.pdf. This final report mainly describes the research conducted during the period from September 15, 2000 to September 30, 2004 (project number 73976).

Vitrification of nuclear waste in a glass is currently the preferred process for waste disposal. DOE currently approves only borosilicate (BS) type glasses for such purposes. However, many nuclear wastes, presently awaiting disposal, have complex and diverse chemical compositions, and often contain components that are poorly soluble or chemically incompatible in BS glasses. Such problematic wastes can be pre-processed and/or diluted to compensate for their incompatibility with a BS glass matrix, but both of these solutions increases the wasteform volume and the overall cost for vitrification. Direct vitrification using alternative glasses that utilize the major components already present in the waste is preferable, since it avoids pre-treating or diluting the waste, and, thus, minimizes the wasteform volume and overall cost.

Iron phosphate glasses containing the following three types of nuclear waste, as recommended by the Tank Focus Area (TFA) group, have been investigated in collaboration with scientists from Pacific Northwest National Laboratory (PNNL), Idaho National Engineering and Environmental Laboratory (INEEL), Westinghouse Savannah River, V.G. Khlopin Radium Institute of Russia, Energy and Nuclear Research Institute of Brazil, and Ruder Boskovic Institute of Croatia.

- (1) a high sodium/sulfate Hanford Low-Activity Waste (LAW)
- (2) a High Level Waste (HLW) with a high chrome content at Hanford, and
- (3) a Sodium Bearing Waste (SBW) at INEEL

Over five hundred trial melts, ranging in size from 50 g to more than 10 kg, have been evaluated. The experimental work consisted of

- (1) evaluating the melting behavior and characteristics,
- (2) measuring of the viscosity and electrical conductivity of promising melts over their melting range,

- (3) determining the chemical durability by the PCT and VHT methods of both glassy and partially crystallized iron phosphate wasteforms,
- (4) determining the solubility limit for chrome oxide in selected iron phosphate melts,
- (5) examining the feasibility of melting iron phosphate glasses by Cold Crucible Induction Melting (CCIM), Hot Crucible Induction Melting (HCIM), and Microwave Melting,
- (6) and measuring the corrosion of Inconel 690 and 693, potential electrode materials, in an iron phosphate melt.

All three of the wastes have been successfully vitrified in iron phosphate glasses at waste loadings ranging from a low of 32 wt% for the high sulfate LAW at Hanford, to 40 wt% for the SBW at INEEL, to a high of 75 wt% for the high chrome HLW at Hanford. In addition to these desirable high waste loadings, the iron phosphate glasses were easily melted, typically between 950 and 1200°C, in less than 4 hours in commercial refractory oxide containers. It is noteworthy that the chemical durability of both glassy and deliberately crystallized iron phosphate wasteforms not only met, but significantly exceeded, all current DOE chemical durability requirements as measured by the Product Consistency Test (PCT) and Vapor Hydration Test (VHT).

It has been estimated that the mass of glass needed to vitrify the high sulfate LAW at Hanford would be reduced by approximately 43%, compared to the baseline borosilicate glass, if the LAW was vitrified in an iron phosphate glass at a waste loading of 20 wt% Na₂O. This would mean a huge reduction in the time and cost to vitrify just this one waste. Similar savings in time and money could be realized for the Hanford HLW where the waste loading in an iron phosphate glass would be 2-3 times that projected for a borosilicate glass. As a final example, it was estimated that at a waste loading of 40%, the 1 million lbs of SBW at INEEL could be vitrified in only 520 days in a furnace with a melting chamber 2 ft in diameter and 2 ft in depth whose output was estimated at 200 lbs/hr.

These promising results combined with the easy melting of iron phosphate glasses in the CCIM can offer significantly reduced costs of vitrifying the Hanford LAW and high chrome HLW, and INEEL SBW.

The iron phosphate melt, which contained 30 wt% of the Hanford LAW, did not corrode the Inconel 690 to any greater extent than what has been reported for Inconel 690 electrodes in the borosilicate melt in the Joule Heated Melter (JHM) at the Defense Waste Processing Facility (DWPF). Furthermore, Inconel 693 appeared to be an even better candidate for use in iron phosphate melts since its corrosion rate (0.7 $\mu\text{m/day}$) was only about one half that (1.3 $\mu\text{m/day}$) of Inconel 690.

In review of the fact that iron phosphate glasses can contain high waste loadings of HLW, LAW, and SBW, they meet all of DOE's chemical durability (PCT and VHT) requirements and they can be melted in several types of furnaces, there are no reasons known why these glasses should not be considered for vitrifying those wastes that contain components which are poorly suited for borosilicate glasses.

This research has resulted in more than 30 refereed technical papers, 1 national laboratory report, and 10 presentations at national and international conferences. In addition to the principal investigator, two research professors, one post doctoral fellow, one graduate student, three undergraduate research aides, and two visiting scholars have been supported by this project.

INTRODUCTION AND RESEARCH OBJECTIVES

The U.S. has accumulated a large amount of nuclear wastes that is now stored temporarily at different DOE sites across the country. These wastes are spent fuel from nuclear reactors; liquid and solid wastes from spent fuel reprocessing for plutonium extraction; transuranic wastes which include clothing, tools, and other materials contaminated with plutonium, neptunium, and etc.; hazardous radioactive wastes from hospitals, research institutes, and remnants of decommissioned power plants; and uranium mill tailings. These wastes must be managed properly to avoid contaminating the environment and to minimize risks to the health of humans and other living species. The liquid wastes are of most concern because of their high mobility and radioactivity.

The overall idea behind the clean-up process for the mobile liquid waste is to immobilize the waste in a stable host matrix. Vitrification of liquid nuclear waste in a glass is considered the most effective process for waste disposal, and DOE currently approves only borosilicate (BS) type glasses for such use. However, many liquid wastes, presently awaiting disposal, have complex and diverse chemical compositions, and often contain components such as phosphates, sulfates, chrome oxide, and heavy metals that are poorly soluble in BS glasses [1,2]. Such problematic wastes can be pre-processed and/or diluted to compensate for the incompatibility with the BS glass matrix, but these two alternatives are expensive and involve risk to the operators. It is more desirable to avoid pre-treating or diluting the waste since these alternatives will greatly increase the wasteform volume and the overall time and cost for vitrification. Direct vitrification using an alternative glass that can incorporate the problematic components in the waste should minimize the wasteform volume and overall cost.

Our previous studies [3] have shown that iron phosphate glasses have the potential for vitrifying many nuclear wastes that are either completely unsuitable or poorly suitable in BS glasses in terms of reducing the wasteform volume and disposal cost. The present research is a continuation of our previous work and was focused on three specific wastes that were given a high priority by the Tank Focus Area (TFA) Report at Hanford [1]. The main objectives were to investigate the feasibility of vitrifying these wastes in iron phosphate glasses, with an aim to produce a wasteform having a higher waste loading than the borosilicate glass counterpart, combined with acceptable chemical durability, and to acquire scientific and engineering knowledge that is needed to utilize iron phosphate glasses for vitrifying selected nuclear wastes on a production scale.

NOTEWORTHY FEATURES OF IRON PHOSPHATE GLASSES

(1) Higher Waste Loading (Smaller Wasteform Volume). Unusually large amounts of simulated wastes (e.g., 32 wt% for LAW, 40 wt% for SBW, and 75 wt% for HLW), compared to the waste loading in borosilicate glasses, have been successfully vitrified in iron phosphate glasses (Figure 1). When the waste contains significant amounts of components such as sulfate, phosphate, and heavy metal oxides like Bi_2O_3 , Cr_2O_3 , UO_2 , and ZrO_2 , the waste loading in iron phosphate wasteforms can be up to five times higher than that for borosilicate glasses. For the Hanford LAW and INEEL SBW, it is unlikely that the waste loading in iron phosphate glasses will be limited by the SO_3 content of the wastes as is currently the case [4] in borosilicate glasses. If the sulfate content of the LAW did not limit its waste loading in the glass, then the amount of glass produced at Hanford could be reduced by as much as 43% [5].

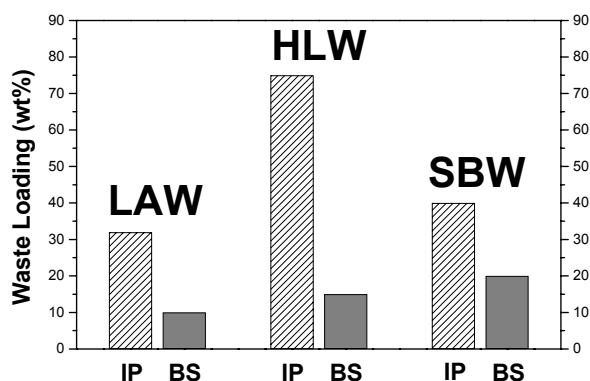


Figure 1. A comparison of the waste loading achieved in iron phosphate (IP) and that calculated for borosilicate (BS) glass wasteforms for three nuclear wastes. The waste loading in BS glass was calculated based on the low sulfate, < 1 wt% SO_3 , (for LAW and SBW) and the low chrome, < 1 wt% Cr_2O_3 , (for HLW) solubility in BS glasses.

To vitrify the Hanford high chrome HLW, it is important to note that only one additional component was added to the simulated waste, namely a source of P_2O_5 such as phosphoric acid. This is in contrast to the present plans to add 4 to 8 additional components to the LAW at Hanford in order to produce a borosilicate glass wasteform. Furthermore, because the solubility limit of Cr_2O_3 in iron phosphate glasses is higher (2.6 wt%) than that (< 1 wt%) [6] in common borosilicate glasses, the maximum waste loading is expected to be significantly higher in iron phosphate glasses.

(2) Excellent Chemical Durability. Vitrified iron phosphate wasteforms satisfy all known DOE requirements for aqueous chemical durability based on the Product Consistency Test (PCT) [7] and Vapor Hydration Test (VHT) [8], see Figure 2. Some iron phosphate wasteforms partially crystallize when slowly cooled or heat treated to simulate Canister

Centerline Cooling (CCC) [9], but the chemical durability of even these partially crystallized wasteforms is excellent and meets all DOE requirements.

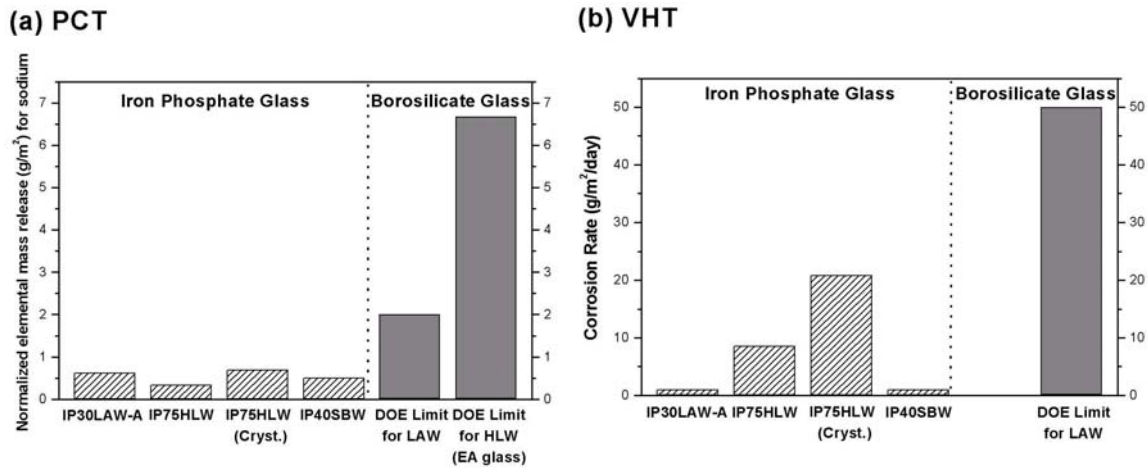


Figure 2. (a) Normalized elemental mass release (g/m²) after PCT for sodium from iron phosphate glassy and deliberately crystallized wasteforms containing 30, 75, and 40 wt% of simulated LAW, HLW, and SBW, respectively. (b) VHT corrosion rate (g/m²/day) for the same iron phosphate wasteforms. DOE specifications for PCT and VHT are given for comparison, but currently, there is no DOE specification for VHT for HLW.

It is important to recognize that, unlike borosilicate glasses, iron phosphate wasteforms inherently buffer any aqueous solution in which they come into contact. This buffering action slows the corrosion process and many iron phosphate glasses develop a corrosion resistant surface layer that inhibits further corrosion. On the contrary, the alkali ions released from borosilicate glass wasteforms during corrosion cause a rapid increase in the pH of the aqueous solution which accelerates the corrosion process since the silicate matrix of a borosilicate glass becomes increasingly soluble when the pH exceeds 9.

(3) Lower Melting Temperatures and Shorter Melting Times. Iron phosphate glasses can be melted as low as 950-1000°C compared to the 1150°C for the borosilicate glasses now being melted at DWPF. Because iron phosphate melts are fluid at their melting temperature (viscosity 200 to 900 centipoise), they rapidly become homogeneous and their melting times can be as short as a few hours (< 4 hours) compared to the > 48 hour residence time for borosilicate melts (viscosity 4,500 to 9,000 centipoise) in the DWPF joule heated melter. Also, because of their lower viscosity, there is less tendency for an insulating foam, which reduces the melting rate, to form on

the surface of an iron phosphate melt during melting. Lower melting temperatures and shorter melting times lead to less expensive and safer melting processes and mean that smaller furnaces can be used for any given output.

(4) Minimum Corrosion of Refractory and Electrode Materials. Experiments dealing with the corrosion of candidate refractory and electrode materials by iron phosphate melts (at the melting temperature) are encouraging since the corrosion observed has been less than expected based on experience with other phosphate melts. The reasons for this reduced corrosion are still unknown and need further investigation. This is another example of how the iron phosphate glasses are unlike other types of phosphate glasses and tend to be more like silica-based glasses than phosphate glasses.

a) Refractories: It was noted that crucibles made from high purity alumina or aluminosilicates were corroded very little by iron phosphate melts, even after being in contact with the melt for several days at temperatures well above the “normal” melting temperature, see Figure 3. For still unidentified reasons, these refractory oxides are poorly “wet” by iron phosphate melts.



Figure 3. Cross sectional view of a dense high silica crucible (DFC 83% silica 17% alumina) after 14 days at 1250°C with an iron phosphate melt containing 27 wt% Hanford LAW simulant. No significant corrosion was evident on the crucible (Photo courtesy of MO-SCI Corp., Rolla, MO).

Our early measurements [10] showed that the dynamic corrosion rate (rotating refractory rods) for several commercial refractories (AD998 alumina, two AZS refractories, zircon, silica and chrome refractories) partially immersed in different iron phosphate melts (containing up to 16.4 wt% Na_2O) was typically less than the 0.19

mm/day rate used in the design of the DWPF melter. In many cases, the corrosion rate of these refractories by iron phosphate melts was less than that of a model borosilicate glass.

A further indication that commercial refractories, such as AZS, should provide an acceptable service life in a JHM melting iron phosphate glasses is the experience reported [11] from Russia where nuclear waste has been vitrified in sodium-alumino phosphate glasses. Sodium-alumino phosphate melts are by nature chemically corrosive so they are expected to chemically attack commercial refractories significantly more than iron phosphate melts. Even so, sodium-alumino phosphate glasses have been successfully melted in Russia for up to six years in a JHM lined with commercial AZS refractories. While the experience in Russia for melting sodium-alumino phosphate glasses in a JHM is only an indirect indication of the behavior expected for the less chemically corrosive iron phosphate melts, that experience combined with our own measurements of the dynamic corrosion resistance of several commercial refractories by iron phosphate melts indicate that commercial refractories should be satisfactory as the glass contact refractories and will have an acceptable service life in a JHM.

b) Electrodes: The corrosion of Inconel 690 and 693 has been investigated using samples partially and fully submerged in an iron phosphate melt that contained 30 wt% of the Hanford LAW at 1025°C for 155 days. The weight loss for the partially submerged Inconel 693 was only 2.5% (corresponding to $< 0.7 \mu\text{m/day}$), see Figures 4 and 5, which is comparable to the corrosion that has been observed [12] for the Inconel 690 electrodes in the JHM used to melt borosilicate glass at DWPF.

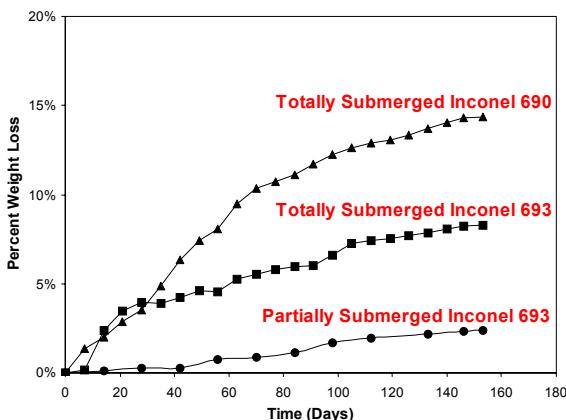


Figure 4. Weight loss for Inconel 690 and 693 samples submerged in an iron phosphate melt containing 30 wt% LAW simulant at 1025°C.

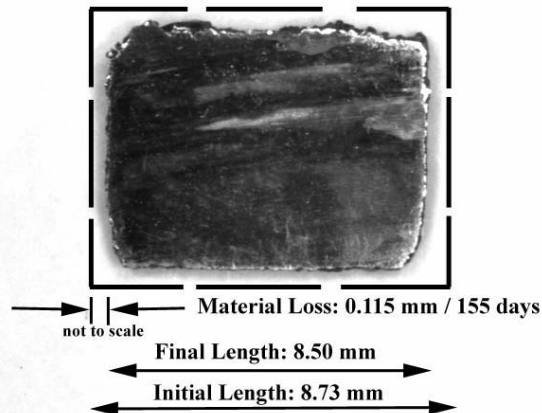


Figure 5. Cross sectional view of the Inconel 693 sample after the corrosion test at 1025°C for 155 days ($0.115 \text{ mm}/155 \text{ days} = 0.7 \mu\text{m/day}$). Note box around sample not to scale.

Based on these results, Inconel 693 is a likely candidate for the electrodes in a JHM melting iron phosphate glasses. Another candidate electrode material is molybdenum since molybdenum electrodes have been used successfully in joule heated melters in Russia to melt the more chemically corrosive sodium-alumino phosphate glasses (for up to six years).

(5) Alternative Melting Methods

a) Cold Crucible Induction Melter (CCIM): Melting iron phosphate glasses in a CCIM has been of interest, since this technique eliminates many materials and operating constraints, such as the chemical corrosion of the melter refractories and metal electrodes, which is unavoidable in a JHM such as that now being used to vitrify nuclear waste at DWPF.

An iron phosphate glass containing 40 wt% simulated SBW was successfully melted in a CCIM at only 50% of the power consumption needed for melting borosilicate glasses (Figure 6). This was the first experimental melting of an iron phosphate wasteform in a CCIM and was conducted in collaboration with the V.G. Khlopin Radium Institute (KRI) in St. Petersburg, Russia. The properties of the iron phosphate glasses melted in the CCIM were the same as those of the same glass composition melted conventionally in an electric furnace. These successful trials of melting iron phosphate glasses indicate that CCIM technology is a feasible and practical method of melting iron phosphate glasses, thereby avoiding potential problems of electrode or refractory corrosion.

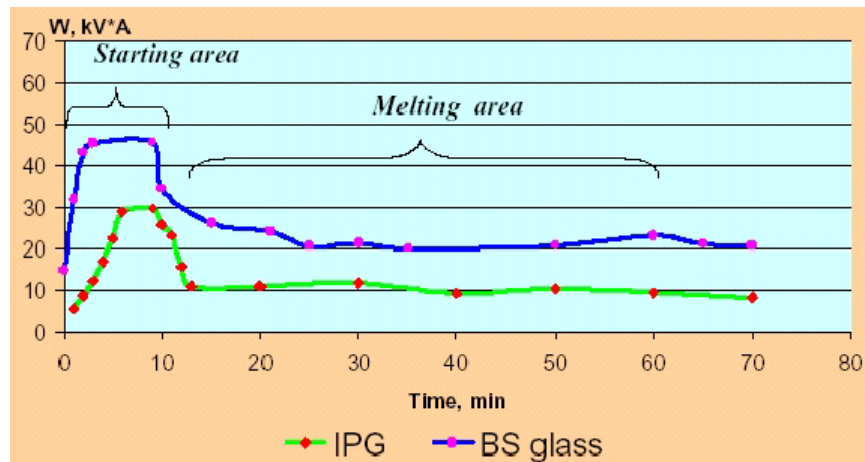


Figure 6. Power consumption at various stages of the melting process in the CCIM for iron phosphate (IPG) and borosilicate (BS) glasses.

b) Hot Crucible Induction Melter (HCIM): Small amounts (300 g) of an iron phosphate glass containing 35 wt% simulated Hanford LAW were successfully melted in a HCIM (Figure 7). The batch was contained in a dense high silica crucible inside a graphite susceptor (Figure 7(a)) and melted at 1150°C for 2 hours at a power level of 12 to 18 kW, depending on the stage of melting. The chemical durability and other properties of the glasses melted in the HCIM were the same as those of the same glass melted conventionally in an electric furnace. The successful melting of the iron phosphate glass in the HCIM is encouraging since this method eliminates the need for metal electrodes in the melt, as in joule-heated melting and the stirring of the fluid melt by the magnetic field rapidly homogenizes the melt, thereby, reducing the melting time.

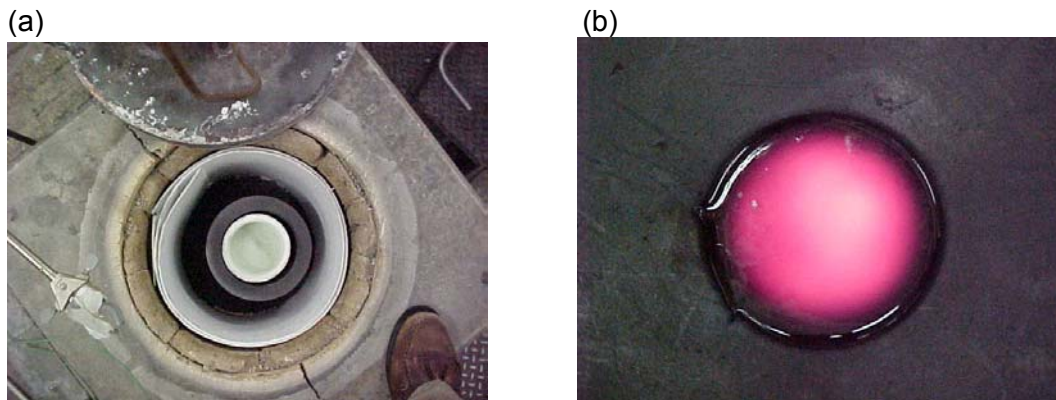


Figure 7. (a) Top view of HCIM and (b) iron phosphate glass wasteform (300 g) containing 35 wt% Hanford LAW that was melted at 1150°C for 2 hours.

c) Microwave Melting: Microwave melting is another alternative technology for vitrifying iron phosphate glass wasteforms. In collaboration with the Energy and Nuclear Research Institute, Brazil, small amounts (50 to 100 g) of an iron phosphate glass containing 35 wt% simulated Hanford LAW have been successfully melted, starting from a cold batch, in an ordinary microwave oven (1100 watts). Compositions containing significant amounts of alkalis, such as soda (~75 wt%) in the Hanford LAW, are an advantage since this improves the coupling of the microwave energy to the melt and promotes rapid melting. Microwave melting also eliminates the need for metal electrodes in the melt, but a refractory crucible is required (alumina and silica work well). The properties of the iron phosphate glasses prepared with microwave heating are the same as those of the same glass melted conventionally.

(6) Inexpensive Raw Materials. The number of additives that are added to a particular nuclear waste to form a glass with acceptable properties obviously depends upon the waste composition, but iron phosphate glasses typically require fewer additives than borosilicate glasses. In many cases, only a source of phosphate (such as phosphoric acid, rock phosphate, or industrial phosphate waste) and a source of iron oxide (as-mined iron ore) need to be added to the nuclear waste. These raw materials are all inexpensive.

Two industrial waste products have been found to provide an economical and technically acceptable source of phosphate. The first is the phosphate waste currently being generated by metal fabricators (automotive, construction equipment, appliance and office furniture manufacturers) which use a metal conversion process to improve the corrosion resistance of iron and steel. An estimated 12,000 tons/yr of iron and zinc phosphate waste is currently generated from this conversion process and buried in landfills [13]. Many of these wastes have been determined [5,13] to be an excellent source of iron oxide and phosphate for iron phosphate glass wasteforms.

The second waste providing phosphate is a relatively pure calcium phosphate powder that is currently being recovered, along with recycled glass, from burnt out fluorescent lights. An estimated 300 to 1000 tons/yr of this waste is being landfilled. This material has been used successfully as the source of phosphate in iron phosphate glasses.

The use of both of these wastes as a raw material in vitrifying nuclear waste would be a doubly beneficial use of these wastes and would eliminate the need to dispose of them in our Nation's landfills.

RELEVANCE AND IMPACT

The DOE high priority wastes including the Hanford LAW and high chrome HLW, and the INEEL SBW are not well suited for vitrifying in borosilicate glasses because their high sodium, high sulfate, high phosphate, and high chrome content can seriously reduce the maximum waste loading in borosilicate glasses. Special procedures designed to successfully vitrify such problematic wastes in borosilicate glasses can add billions of dollars to the DOE's cost of cleaning up the Hanford and INEEL sites. The iron phosphate glasses developed at the University of Missouri-Rolla appear to be an

excellent alternative host matrix for vitrifying these DOE high priority wastes that contain components poorly suited for borosilicate glasses.

It is extremely important that the reader understand that iron phosphate glasses have unique properties which place them in a special category of phosphate glasses. The excellent chemical durability, tendency not to wet or chemically attack refractory oxides, high waste loading, easy melting in a cold or hot crucible induction melter, and other important properties of the iron phosphate glasses are far superior to those of all other known phosphate glasses, including the less chemically durable and more corrosive sodium-alumino phosphate glasses that have been used for waste vitrification in Russia for more than a decade. All of our existing knowledge of iron phosphate glasses and their wasteforms indicates that they can be a highly effective and low cost alternative to borosilicate glasses, especially for those wastes which contain components that are poorly soluble in or chemically incompatible with borosilicate glasses.

PUBLICATIONS

The following 34 papers on iron phosphate glasses have been published during the period (September 15, 2000 to September 30, 2004) covered by this EMSP final report. Over the entire period from September 15, 1996 to September 30, 2004 during which EMSP supported research on the iron phosphate glasses, a total of 47 papers and 3 theses were published.

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PRESENTATIONS

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with Cold Crucible”, MRS 2003, Scientific Basis for Radioactive Waste Management XXVII, Kalmar, Sweden, June 15-18, 2003.

7. C. W. Kim, D. E. Day, and A. S. Aloy, “Iron Phosphate Glasses for Nuclear Waste Vitrification”, Alternative Nuclear Waste Forms, Engineering Conferences International, Alaska, U.S.A., January 18-23, 2004 (invited).
8. C. W. Kim and D. E. Day, “Iron Phosphate Glasses for Vitrifying DOE High Priority Nuclear Wastes”, Environmental and Waste Management: Advances through the Environmental Management Science Program, 227th American Chemical Society Meeting, Anaheim, California, March 28-April 1, 2004.
9. C. W. Kim, D. Zhu, and D. E. Day, “Corrosion Resistance of Metal Electrodes in An Iron Phosphate Melt”, ACerS 2004 Annual Meeting, Indianapolis, Indiana, April 18-21, 2004.
10. C. W. Kim and D. E. Day, “Advantages of Using Iron Phosphate Glasses for Vitrifying DOE High Priority Nuclear Wastes and Korean LILW”, Expert Mission of International Atomic Energy Agency (IAEA) Technical Cooperation Program (ROK/4/031): Title- “Technology Development for Vitrification of Low- and Intermediate-level Radioactive Wastes from Nuclear Power Plants”, Nuclear Environment Technology Institute, Korea Hydro & Nuclear Power, Daejeon, South Korea, October 25-29, 2004.

COLLABORATIONS

Pacific Northwest National Laboratory (PNNL): Evaluating the feasibility of vitrifying Hanford Low-Activity Waste (LAW) in the iron phosphate glasses.

Idaho National Engineering and Environmental Laboratory (INEEL): Investigating the vitrification of Sodium Bearing Waste (SBW) in the iron phosphate glasses.

Westinghouse Savannah River Co.: Investigating the corrosion behavior of electrode materials in iron phosphate glass melts.

V.G. Khlopin Radium Institute, Russia: Provided glass compositions and evaluated the properties of iron phosphate glasses melted by Cold Crucible Induction Melting (CCIM) technique.

Energy and Nuclear Research Institute, Brazilian Nuclear Energy Commission, Brazil: Technical collaboration and experiments on microwave melting technique as an alternative melting process.

Ruder Boskovic Institute, Croatia: The structure of iron phosphate glasses using Raman spectroscopy and thermally stimulated current techniques.

Ishikawajima-harima Heavy Industry Co. Ltd. and Institute of Research and Innovation, Japan: Feasibility of vitrifying Japanese HLW using iron phosphate glasses.

Nuclear Environment Technology Institute, Korea Hydro & Nuclear Power, South Korea: Investigating the feasibility of vitrifying Korean low- and intermediate-level radioactive wastes from nuclear power plants in a cold crucible induction melter using iron phosphate glasses.

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