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SYSTEMS APPROACH TO NUCLEAR WASTE GLASS DEVELOPMENT

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ABSTRACT

Development of a host solid for the immobilization of nuclear waste has focused on various vitreous wasteforms including borosilicate, high silica, and phosphate glasses. Consideration of the entire waste disposal system ensures that the final product will safeguard the public, and that the production process will be safe to operate. The systems approach requires that parameters affecting product performance and processing be considered simultaneously. Parameters affecting product performance, such as chemical durability, wasteform solubility, thermal stability, and mechanical stability must be optimized relative to processing concerns such as melt temperature, waste solubility, melt corrosiveness, and volatility of hazardous species.

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Application of the systems approach indicates that borosilicate glasses are, overall, the most suitable glasses for the immobilization of nuclear waste. Phosphate glasses are highly durable; but the glass melts are highly corrosive and the glasses have poor thermal stability and low solubility for many waste components. High-silica glasses have good chemical durability, thermal stability, and mechanical stability, but the associated high melting temperatures increase volatilization of hazardous species in the waste. Borosilicate glasses are chemically durable and are stable both thermally and mechanically. The borosilicate melts are generally less corrosive than commercial glasses, and the melt temperature minimizes excessive volatility of hazardous species. Optimization of borosilicate waste glass formulations has led to their acceptance as the reference nuclear wasteform in the United States, United Kingdom, Belgium, Germany, France, Sweden, Switzerland, and Japan.

INTRODUCTION

Development of a host solid for the immobilization of nuclear waste has focused on various vitreous wasteforms, including borosilicate, high silica, and phosphate glasses. Glass-ceramics and ceramic wasteforms have also been investigated. Both the product performance and the production process must be reliable in order to safeguard the public. Application of a systems approach simultaneously evaluates product performance and processing considerations. Paramaters affecting product performance, such as chemical durability, wasteform solubility, thermal stability, and mechanical stability are optimized relative to processing considerations, such as melt temperature, waste solubility, melt corrosiveness, and volatility of hazardous species (Table I).

TABLE 1

Systems Approach to Nuclear Waste Glass Development

Product Reliability

- Chemical Durability
- Thermal Stability
- Mechanical Stability

Process Reliability

- Melt Temperature
- Melt Corrosivity
- Waste Solubility
- Volatility*

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* of hazardous species

Development of glasses for the solidification of nuclear waste began at different times in the United States, Canada, Europe, and the USSR. Different glass formulations and processing strategies were developed. The borosilicate glass formulations were developed in the United States between 1956 and 1957, while the aluminosilicate (nepheline syenite) glass formulations were simultaneously being developed in Canada. Phosphate-based glasses were the last to be investigated and the first to be abandoned for solidification of nuclear waste. A systems evaluation of phosphate glasses demonstrated that the positive aspects of processing (e.g., low melting temperatures) were outweighed by other negative processing aspects (e.g., melt corrosiveness) and by poor product performance. The aluminosilicate glasses and the ceramic wasteforms are still being investigated for certain types of nuclear waste because the systems evaluation of these wasteforms is favorable under certain conditions. However, repeated systems evaluations of borosilicate waste glass formulations and the associated processing technology have found this family of glasses to be applicable to a wide range of waste compositions melted by various processing techniques. The favorable systems evaluations of borosilicate waste glasses have led to their acceptance as the reference nuclear wasteform in eight countries, and their consideration by a ninth, Canada (as of 1984).[1]

Figure 1 attempts to unify the conceptualization and systems development of borosilicate, aluminosilicate (nepheline syenite), and phosphate glasses from the late 1950's to the present. The historical development of the glass formulations and the importance of a whole systems evaluation will be summarized, with special reference to the family of phosphate glasses and the recently developed lead-iron-phosphate glass for nuclear waste disposal.[2,3]

HISTORICAL DEVELOPMENT OF GLASS FOR NUCLEAR WASTE DISPOSAL

Borosilicate Glasses

The first "glass" formulations for use in nuclear waste disposal were investigated by Goldman and others at the Massachusetts Institute of Technology as early as 1956.[4-6] They examined calcium aluminosilicate porcelain glazes to which B_2O_3 had been added to achieve a pourable glass by melting at 1300°C. The quality of these glasses did not suffer from the incorporation of such substances as ZrO_2 and Fe_2O_3 that were present in the nuclear waste solutions.[4-6] Eliassen and Goldman[5] felt that the most promising vitreous systems for future development were borosilicate based (e.g. $CaO-Al_2O_3-B_2O_3-SiO_2$ and $Na_2O-CaO-Al_2O_3-$ $-B_2O_3-SiO_2$).

Although the early borosilicate glass formulations were developed in the United States, vitrification process development and testing during the early 1960's was carried out primarily in the United Kingdom and Europe.[7-9] In the mid 1960's the Waste

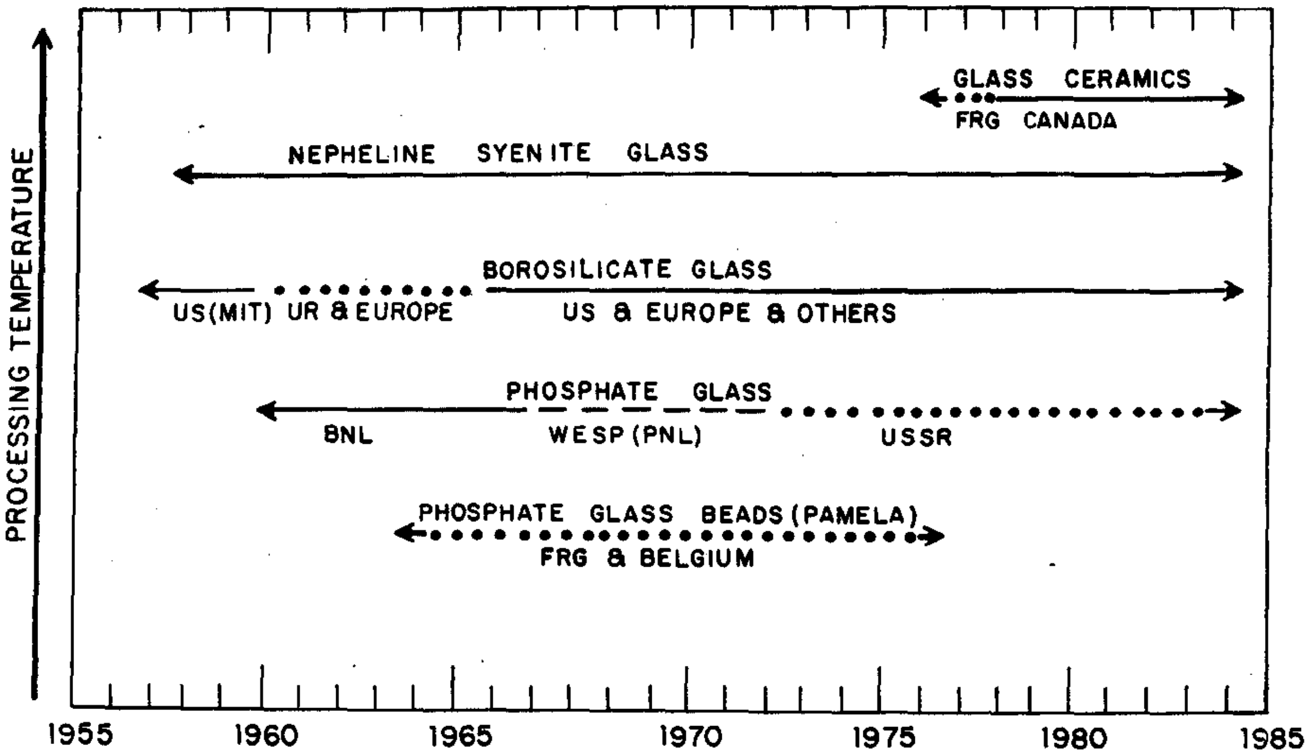

FIGURE 1. **Historical Development of Glass for Solidifying Nuclear Waste**

Solidification Engineering Prototypes (WSEP) program demonstrated the overall ease of processability and compositional flexibility of borosilicate-type glasses,[10] and Pacific Northwest Laboratories demonstrated the advantages of ceramic Joule-heated melters.[11] Other waste producers, such as the Savannah River Plant, built upon the European and the U.S. experiences and chose borosilicate glass as the reference solid wasteform.[12]

Borosilicate glasses are alkali aluminosilicate-type glasses which are fluxed with boron (Table II). The lower alumina content and the presence of boron lower the melt viscosity and hence the processing temperature (about 1150°C) relative to that of the aluminosilicate glasses. The boron increases the solubility of many waste constituents in the silica-based glass[13] while maintaining their thermal and mechanical stability.[14] Boron decreases the chemical durability only slightly [14,15] relative to the highly durable, but difficult to fabricate, aluminosilicate glasses.

The typical borosilicate waste glasses currently in use for solidification of nuclear waste are quite different from Pyrex® (Corning) borosilicate glass (Table II). Since the borosilicate waste glasses are 15-30 wt% waste, much of the chemistry is still dominated by the chemistry of the alkali oxide-silica-boron oxide system. In Figure 2, the regions of phase separation for Pyrex® and Vycor® borosilicate glasses in the Na_2O-B_2O_3-SiO_2 system are shown.[16]

TABLE 2

Typical Range of Waste Glass Compositions

Oxide Wt%	Pyrex®*	Boro-Silicate*	Alumino-Silicate*	Glass-Ceramics*	WSEP P_2O_5*	$PbO-Fe_2O_3-P_2O_5$**
SiO_2	81	25-50	33-40	32-60	0-6	0-0.25
B_2O_3	13	9-22	--	1-11	--	--
P_2O_5	--	0-2	--	--	30-55	25-42
Alkali Oxides	4	8-19	18-22	0-13	5-25	1-2
Alkaline Earth Oxides	--	0-6	13-16	2-33	--	--
Fe_2O_3, NiO, Cr_2O_3, MnO	--	1-20	--	--	0-30	6-13
Al_2O_3	2	0-10	26-30	10-24	0-35	1-2
TiO_2	--	0-3	0-3	--	--	--
ZnO	--	0-22	--	0-13	--	--
PbO	--	0-50	--		0-30	36-53
Fission Products	--	30	5	20	30	0-0.25
Actinides	10	1	7	10	1-2.5	--

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* Mendel et al [13]

** Sales and Boatner [3]

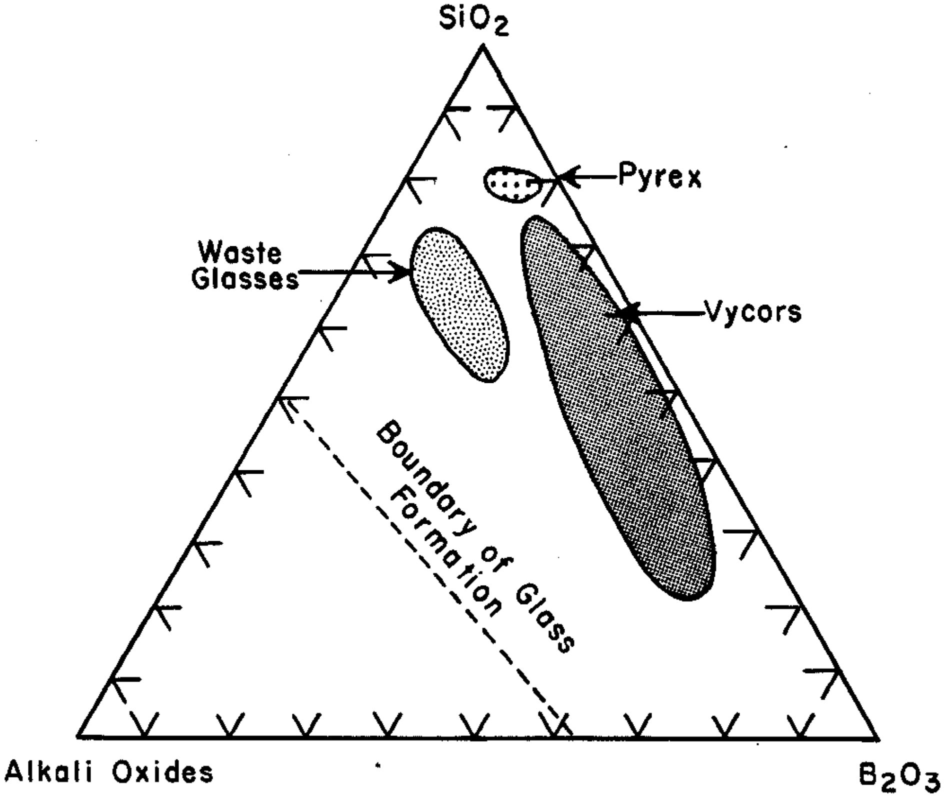

FIGURE 2. Pseudoternary phase diagram of the alkali oxide-silica-boron oxide system (after Reference 16). Compositional ranges for borosilicate glasses, including Pyrex®, Vycor®, and borosilicate nuclear waste glasses.

The range of compositions for commercial and defense borosilicate waste glasses expressed as a function of the alkali, boron, and silicon oxide content are superimposed on the ternary phase diagram (Figure 2). The compositional differences between the borosilicate waste glasses and Pyrex® show that Pyrex® has a higher silica content than the waste glasses and, hence, melts at a higher temperature. Although Pyrex® is phase separated on a fine scale, it is an extremely durable glass due to its high silica content.[16] The lack of phase separation in borosilicate waste glasses is also attributed to the compositional difference (Figure 2).

A systems evaluation of the borosilicate waste glasses indicates that these glasses exhibit good chemical durability, thermal stability, mechanical stability, and waste solubility while being processed at temperatures (1150°C) that limit the volatility of hazardous species (Table III). The borosilicate waste melts are generally less corrosive than commercial glass melts, such as Pyrex®, due to the lower temperature of fabrication. Therefore, the technology used for commercial waste glass fabrication can be easily applied. Borosilicate glasses exhibit favorable product performance as well as ease of processability.

Aluminosilicate Glasses

Aluminosilicate-based glass formulations were investigated by Atomic Energy of Canada, Ltd. (AECL) about 1957.[17] These glasses were fused from crushed nepheline syenite rock mixed with

TABLE III

Systems Evaluation of Aluminosilicate, Borosilicate and Phosphate Waste Glasses

	Type of Glass		
Product Reliability	**Borosilicate**	**Aluminosilicate**	**Phosphate**
Chemical Durability	Good	Excellent	Good
Thermal Stability	Good	Good	Poor
Mechanical Stability	Good	Good	Poor
Process Reliability			
Melt Temperature	1150°C	1350°C	850-1050°C
Waste Solubility	Good (28-35%)	Fair (10%)	Low (5-15%)
Volatility*	Good	Poor	Good

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* of hazardous species

waste at temperatures above 1350°C. The Canadian developers had considered use of crushed borosilicate Pyrex® (Table II) glass because of its superior durability, but the cost of the raw material was prohibitive when compared to the nepheline syenite rock.[17]

The aluminosilicate or nepheline syenite type glasses have undergone continual development in Canada since the late 1950's, as indicated in Figure 1. The major advantage of aluminosilicate waste glasses is the excellent chemical durability, and thermal and mechanical stability due to the high silica and alumina content. The disadvantages are the high melt temperature of 1350°C, which causes volatilization of hazardous species, and the limited waste loading.[13] Melt corrosiveness is comparable to Pyrex® glass but greater than that of borosilicate glasses due to the higher fabrication temperature of the aluminosilicate glasses. A systems evaluation of the aluminosilicate glasses, therefore, indicates that they have superior product characteristics but are difficult to process (Table III).

Glass Ceramics and Ceramics

Although glass ceramics and ceramics have also been examined as hosts for the solidification of nuclear waste, these forms were difficult to process and were not as flexible toward variations in waste composition as glass.[18-25] The objective of ceramics development was to provide chemical, thermal, and mechanical stability superior to borosilicate glass. However, intergranular glassy phases often formed in the ceramic materials, especially when alkali-containing wastes were processed. This intergranular glasss limited the product stability and durability.[26,27] The chemical durability of glass ceramics and ceramics was, therefore, found to be comparable to glass.[25,28] However, both the historical development and the systems evaluation of glass ceramics and ceramics as solid wasteforms parallel that of the vitreous wasteforms and will be discussed briefly.

The concept of immobilizing the radioactive elements of nuclear waste in an assemblage of mineral phases was originally introduced by Hatch[29] at Brookhaven National Laboratory in 1953. The feasibility of making a ceramic of natural mineralogically stable phases was demonstrated by McCarthy[18,19] and Roy[30,31] at the Pennsylvania State University between 1973 and 1976. Since that time, a number of other mineralogic-ceramic assemblages have been developed. Among them are the Sandia titanate-based ceramic,[20] the Australian titanate-based ceramic "SYNROC",[21,32,33] the silicate-phosphate supercalcine-ceramics,[34] and the alumina-based tailored ceramics.[22]

Silicate glass ceramics were developed in the mid 1970's in Germany.[24] Silicate and phosphate glass ceramics were also developed in the USSR.[35] Silicate glass ceramics were developed in Japan,[36] and titanium aluminosilicate glass ceramics are currently being developed in Canada.[37] Implicit in the ceramics glass-ceramics formulations is the idea of using additives to alter the waste composition; tailoring the waste chemically so that the desired host radionuclide phases are produced after consolidation. The "SYNROC" concept is also under current examination in many countries for high heat producing wastes (Table IV).

Phosphate Glasses

The major research into phosphate-based waste glasses in the United States started at Brookhaven National Laboratory around 1967 by Tuthill and others[38] (Figure 1). The delay in the development of phosphate glasses for use in waste disposal has been attributed to a lack of industrial use for these types of glasses and, hence, the lack of commercial experience and technology that exists for the various silica-based glasses.[13] The attractive low melting temperature of the phosphate glasses is offset by the corrosiveness of the melt and the ease with which these glasses devitrify.[13] Phosphate glasses were also attractive because molybdenum and sulfate were more soluble than in borosilicate glass.[25] This was particularly applicable to wastes in Germany and, hence, the PAMELA process[39] was developed in the early 1960's. In this process, phosphate glass is formed as small beads that are then placed in a metal matrix within a steel canister (Figure 1). The small size of the beads plus the high thermal conductivity provided by the metal matrix ensure that the phosphate glass will not devitrify.

Development of the phosphate beads in Germany began in the early 1960's and subsequent solidification in a continuous metallic melter at Eurochemic in Mol, Belgium continued until the late 1970's. About 1976, Karlsruhe and Eurochemic changed to a

TABLE IV

Wasteforms Being Considered or Accepted by Various Countries (After Reference 1)

Country	Waste Matrix*	Comments
Belgium	BSG	Plans to use the PAMELA vitrification process
Canada	BSG, Alumino-silicate glass, ceramics, cast metals	Research in progress
FRG	BSG	Plans to use the PAMELA vitrification process
France	BSG	Industrial, commercial-scale technology (AVM) to serve domestic & foreign
Japan	BSG	Pilot plant operation planned for 1987. Studying SYNROC
Sweden	BSG	Accepts BSG from foreign reprocessing
Switzerland	BSG	Accepts BSG from foreign reprocessing
UK	BSG	Will use AVM technology. SYNROC studies underway
US	BSG	BSG was 1st choice of a study of 17 potential matrix materials

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* BSG = Borosilicate Glass

borosilicate waste glass produced by a Joule-heated ceramic melter. The experience in the glass industry and at Pacific Northwest Laboratory[11] demonstrated that this type of melter had a higher capacity per physical size, produced a more uniform glass, and had fewer problems with volatile losses than other melting techniques. The time interval for development of phosphate glass beads in a metal matrix was, therefore, relatively short (Figure 1).

In 1966, the Waste Solidification Engineering Prototypes (WSEP) program was initiated at Pacific Northwest Laboratory. WSEP was a pilot plant designed for a radioactive demonstration of three solidification processes; two for borosilicate glass and one for phosphate glass.[10] Eleven canisters of radioactive phosphate waste glass were solidified. The results of the WSEP program showed that phosphate glass had several shortcomings when compared with borosilicate glasses. These shortcomings included the following:

- high ruthenium volatility during preparation (denitration) of the liquid waste slurry
- additions of ferric nitrate and sodium hydroxide required to adjust the melting point and melt viscosity
- extreme corrosiveness of the phosphate melt required the use of platinum melters
- extreme corrosiveness of the phosphate melt placed limitations on the temperature of the melt poured into the final metal canisters

- low solubility of certain waste components including alumina, alkaline earth oxides, sulfates, and fission products
- segregation of fission products in the glass
- rapid thermal devitrification at temperatures above 500°C
- a factor of 1000 increase in the leach rate of the glass after devitrification

At the conclusion of the WSEP program in 1972, phosphate glasses were abandoned for waste solidification in the United States, as indicated in Figure 1.

In 1984, lead-iron-phosphate (LIP) glasses were developed at Oak Ridge National Laboratories as a new, very stable, and easily prepared medium for the immobilization of all types of High-Level Liquid Waste (HLLW).[2,3,40,41] ORNL demonstrated that the corrosion rate of the lead-iron-phosphate waste glass is at least 10^2-10^3 times lower than the corrosion rate of a comparable borosilicate waste glass at 90°C.[2,3] In addition, they determined that (1) the lead phosphate glass could be prepared at temperatures as low as 800°C because the glasses had low melt viscosities (in the 800-1050°C range), (2) the glasses did not devitrify up to temperatures as high as 550°C, and (3) the glasses were not adversely affected by large doses of gamma radiation in water at 135°C.[2,3]

The developers stated that the improved chemical durability and thermal stability of phosphate glass is caused by the Fe_2O_3 content[3,40,41] and the structural role of iron in the glass, which strengthens the cross bonding between the polyphosphate chains.[41,42] The amount of Fe_2O_3 which must be added to the glass is dependent upon the iron content of the waste. A highly stable wasteform is realized when the iron concentration is adjusted to a content of $Fe_2O_3/PbO \cdot P_2O_5 \sim 9$ wt%. The PbO content of the lead metaphosphate starting material may be as low as 45 wt% but may not exceed 66 wt%, which has been experimentally determined to be the critical limit for glass formation.[2] The viscosity of the melt increases rapidly as the PbO content falls below 45 wt%. Although waste loadings of only 15-20 wt% were achievable in the 5.0 g/cm^3 LIP glasses, these were considered comparable to 30-40 wt% waste loaded 2.5 g/cm^3 borosilicate glasses on a waste-per-volume loading.[2] Comparing the recent LIP glass formulations to the WSEP phosphate glasses, the LIP glasses have a somewhat higher PbO content and a lower waste loading than the WSEP phosphate glasses (Table II). The iron and phosphate levels are, however, comparable. Likewise, melt corrosiveness[40,41,43,44] and incompatibility with certain canister materials[43,44] were observed for the LIP glass formulations. Low waste component solubility, even at elevated melt temperatures (>1150°C), produced nonhomogeneous glasses which gave leach rates comparable to those of borosilicate waste glass.[43,44] Thermal stability was poor, as evidenced by rapid thermal devitrification above 550°C.[45,46]

A systems evaluation of the phosphate glasses, including the WSEP, LIP, and European phosphate-based glasses, indicates that they have average to poor product characteristics (e.g. the durability is similar to that of borosilicate glass), but the thermal stability is poor and they are difficult to process (Table III). The only known work continuing on phosphate glasses for waste disposal is in the USSR where aluminophosphate formulations are being pursued to limit devitrification.[47]

CONCLUSIONS

The continued application of the systems approach for evaluating glass formulations and processing techniques between 1956 and the present has led to the acceptance of borosilicate waste glass for the immobilization of a wide range of nuclear waste compositions. A systems evaluation of aluminosilicate glasses indicates that they have superior product characteristics but are difficult to process. A systems evaluation of the phosphate glasses including the WSEP, LIP, and European phosphate-based glasses indicates that they have adequate to poor product characteristics (e.g. the durability is similar to that of borosilicate glass) but the thermal stability is poor and they are extremely difficult to process. Borosilicate glasses exhibit favorable product performance as well as ease of processing. As of 1984, nine countries had either chosen or were considering borosilicate glass as a solid wasteform (Table IV).

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