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Nuclear Waste Disposal and Strategies for Predicting Long-Term Performance of Material

by

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Engineering with Ceramics- NUCLEAR WASTE DISPOSAL and STRATEGIES FOR PREDICTING LONG-TERM PERFORMANCE OF MATERIALS

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A. INTRODUCTION

Ceramics have been an important part of the nuclear community for many years. On December 2, 1942, an historic event occurred under the West Stands of Stagg Field, at the University of Chicago. Man initiated his first self-sustaining nuclear chain reaction and controlled it. The impact of this event on civilization is considered by many as monumental and compared by some to other significant events in history, such as the invention of the steam engine and the manufacturing of the first automobile. Making this event possible and the successful operation of this first man-made nuclear reactor, was the use of forty tons of UO_2. The use of natural or enriched UO_2 is still used today as a nuclear fuel in many nuclear power plants operating world-wide. Other ceramic materials, such as ^{238}Pu, are used for other important purposes, such as ceramic fuels for space exploration to provide electrical power to operate instruments on board spacecrafts. Radioisotopic Thermolecric Generators (RTGs) are used to supply electrical power and consist of a nuclear heat source and converter to transform heat energy from radioactive decay into electrical power, thus providing reliable and relatively uniform power over the very long lifetime of a mission. These sources have been used in the Galileo spacecraft orbiting Jupiter and for scientific investigations of Saturn with the Cassini spacecraft. Still another very important series of applications using the unique properties of ceramics in the nuclear field, are as immobilization matrices for management of some of the most hazardous wastes known to man. For example, in long-term management of radioactive and hazardous wastes, glass matrices are currently in production immobilizing high-level radioactive materials, and cementious forms have also been produced to incorporate low level wastes. Also, as part of nuclear disarmament activities, assemblages of crystalline phases are being developed for immobilizing weapons grade plutonium, to not only produce environmentally friendly products, but also forms that are proliferation resistant. All of these waste forms as well as others, are designed to take advantage of the unique properties of the ceramic systems (**1**).

In January, 2001, an important milestone was reached in the U.S. program to safely and effectively store and dispose of high level radioactive waste (HLW). A unique glass making operation in an equally unique vitrification facility, the Defense Waste Processing Facility (DWPF) in Aiken, South Carolina, poured its 4 millionth pound of environmentally safe nuclear waste glass, converting a significant portion of a very large inventory of HLW stored in a liquid form, to solid and inert products. These products will ultimately be disposed of in carefully selected geologic repositories, where they will be buried as part of a multi-barrier isolation system, designed to not only be safe for our

they will be buried as part of a multi-barrier isolation system, designed to not only be safe for our generation, but all generations yet to come. This paper will describe key features of this program and some of the important roles that ceramics play in this effort, including waste form development, processing and equipment design.

Also, it is necessary to quantify performance of nuclear waste glass systems in complex environments and for very long time periods. A strategy has been developed in the nuclear community to accomplish this important and challenging task. As a result of collaborations at last's years meeting in Cocoa Beach with colleagues working on biomaterials, we noted that important and challenging problems, such as being able to predict long term behavior of materials in aggressive environments, are common to many fields. We thought it might be worthwhile to see if the strategy, developed over many years for predicting long term performance of 40-component nuclear glass systems in a geologic environment for long time periods, could be useful in predicting long term performance of other ceramics and material systems in the body. Using an interdisciplinary approach, a master flowsheet is being developed. This will piggy back on the experience and strategy of the nuclear field and see if any of these elements can be applicable to those in the biomaterials area. This new initiative and collaboration will be introduced.

B. CASE STUDY- VITRIFICATION OF HLW

BACKGROUND

Every major country involved with long-term management of high-level radioactive [HLW] waste has either considered or selected glass as the matrix of choice for immobilizing and ultimately, disposing of potentially hazardous, high-level radioactive material. There are many reasons why glass is preferred. Among the most important considerations is the ability of glass to accommodate and immobilize the many different types of radionuclides present in waste, and make them into a highly durable, integrated, solid waste form. The waste glass product produced not only has outstanding technical properties but also possesses excellent processing features, which allow glasses to be produced with relative ease even under difficult remote-handling conditions, necessary for processing highly radioactive material. The single most important property of the waste glass is its outstanding chemical durability and its corrosion resistance to a wide range of environmental conditions. The following treatise will provide an introduction into the field of HLW management and emphasize the behavior and performance of nuclear waste glass systems studied under a wide variety of conditions.

- High Level Waste Inventory

Located in the United States today, are approximately 100 million gallons of high-level radioactive waste [HLW] containing over 1 billion curies of radioactivity **(2)**. Most of this waste has been generated from defense programs and results from the reprocessing of spent fuels used in the production of tritium and plutonium, both of which are used for military applications. Most of the defense waste is stored as a liquid

or semi-liquid in large underground tanks at the Hanford Site in Richland, Washington and at the Savannah River Site [SRS] in Aiken, South Carolina. While the largest volume of this waste is located at Hanford, the largest amount of radioactivity within this waste is contained at Savannah River. In addition to these inventories, a smaller quantity of defense waste has also been generated from the reprocessing of fuels used in naval reactors, and is stored in stainless steel bins as calcine at the Idaho Chemical Processing Plant in Idaho Falls **(2,3)**.

In addition to the large amounts of defense waste currently on hand, another potential source of HLW is from reprocessing associated with nuclear power plants used to produce electricity. While reprocessing is in progress in other countries, commercial reprocessing was discontinued in the United States in the early 1970's. Hence, only 612,000 gallons of HLW were generated, all of which is stored at West Valley, New York. Existing inventories of defense and commercial HLW stored in the United States and their locations are summarized schematically in **Figure 1** **(2,3)**.

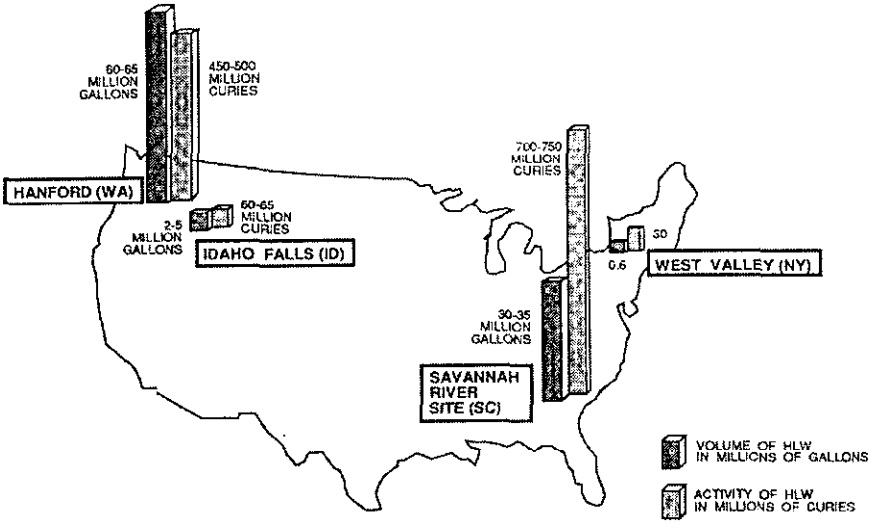


Figure 1
Volumes and Activities of High Level Radioactive Wastes in the U.S.

- High Level Waste Characteristics

High-level waste [HLW] often consists of three major components; *sludge*, *supernate* and *saltcake*. The sludge is the most important component of the waste in that it contains most of the radioactivity, including fission products and long lived actinides. Sludge comprises about 10% by volume of the entire waste inventory and settles to the bottom of the underground storage tanks as a thick, gelatinous precipitate. It consists mainly of aluminum, iron and manganese oxides and hydroxides and contains actinides, fission products and Sr-90. The supernate makes up the remaining 90% of the waste and consists mostly of sodium nitrate and sodium nitrite along with Cs-137.

In order to reduce the total amount of waste stored, the supernate is concentrated by evaporation and returned to the cooled tanks where saltcake crystallizes out of solution. Hence, in waste tanks today, these three phases can co-exisit. It is these forms of the HLW that must be considered in immobilization tasks.

C. ANALYSES

HIGH LEVEL WASTE DISPOSAL STRATEGY

Since the Manhattan Project of World War II, HLW in the U.S. has been stored mainly in a liquid slurry form in underground storage tanks (**Figure 2**). Many of these original tanks are now reaching the ends of their projected lifetimes and as a result, some leaks have developed. Even though no injuries from radiation have occurred as a result of these practices, it should be clear, that a new means to more effectively and more permanently isolate the waste is needed. The strategy for long-term management of HLW is now changing from a program developed in the 1940's, of temporarily storing the waste in a relatively mobile liquid form, to a policy of the late 1990's, of immobilizing the waste into a solid glass product and *ultimately*, *permanently* disposing of the waste glass forms in deep geologic burial, as part of a multi-barrier waste isolation system **(4)**.

Figure 2 HLW Storage Tanks at SRS

At the center of the proposed permanent disposal system is the durable, high integrity waste glass product. Adjacent to the waste glass, is the casting canister [304L stainless steel for the first HLW vitrified in the U.S.], followed by an engineered barrier tailored for the geologic conditions of the repository [may include an overpack and possible backfill], and finally, the geologic barrier itself. Geologic formations studied include salt, basalt, shale, clay, granite, and tuff, with the tuff site at Yucca Mountain in Nevada being selected as the reference repository setting. Each of the elements of the multi-barrier isolation system is designed to prevent the harmful release of radionuclides into the accessible environment and to allow the waste products to meet Federal release rate criteria. Among the most important regulations relating to waste form and waste package performance are 10 CFR 60 [Nuclear Regulatory Commission **(5)**] and 40 CFR 191 [Environmental Protection Agency **(6)**]. An attempt to define a time schedule for permanently disposing of HLW in the United States was provided by the Nuclear Waste Policy Act of 1982 **(7)** and subsequent legislation.

Unlike other types of hazardous wastes, high level radioactive waste can actually become less hazardous with time, due to decay of radionuclides. For example, about 99% of the many radionuclides and daughter products in SRS HLW decays with half-lives ranging from microseconds to less than 30 years. After a time period of about one thousand years, the activity of the waste decreases approximately 4 to 5 orders of magnitude and for much longer time periods, the activity can actually approach and eventually becomes less, than the natural uranium from which the waste was originally derived.

ADVANTAGES OF GLASS

Many different disposal options and potential waste forms, including both vitreous and crystalline ceramics, were evaluated to determine the best system and strategy for safely and effectively disposing of HLW **(8-13)**. Borosilicate glass was selected over other waste form alternatives which included calcines and supercalcines, cements, titanates, cermets, Synroc as well as other types of glass systems for immobilization of the first HLW to be treated in the United States, the waste at SRS. This decision was made based upon data and information supplied by more than three decades of national and international research and by reviews supplied by independent committees. Among the peer review committees used in this process, were groups sponsored by the National Academy of Sciences, the American Physical Society, and the U.S. Department of Energy, who all concluded that borosilicate glass would be a good material for immobilizing the SRS HLW. The selection of borosilicate glass was based on good characteristics in two categories; a) *processing considerations* and b) *technical performance* in five main areas. Following is a brief description of waste glass performance in each of these categories **(4,8,13)**:

PROCESSING CONSIDERATIONS

- Vitrification Facilities & Practical Operating Experience

Figure 3 Saltstone Facility at SRS

The only major HLW immobilization facilities in the world are glass-making operations. These include successful production facilities at Marcoule and LaHague in France, as well as a plant located at Mol in Belgium. There are also additional plants either in production, under construction or being planned in other countries such as Japan, Germany, India, the United Kingdom and the United States. Following is a brief description of the vitrification process and facility at SRS, which is currently in production, immobilizing the first HLW in the United States.

- SRS Vitrification Process

An important feature of the long-term management strategy of SRS HLW is to first separate the waste into radioactive and non-radioactive parts. Since most of the waste is non-radioactive, this allows the large bulk of the waste to be handled by less expensive and easier processing and disposal techniques. After a series of processing steps, the decontaminated portion of the waste is segregated and ultimately, mixed with a cement-based material to produce a waste product called saltstone, that is buried in engineered vaults on site, as shown in **Figure 3** **(9)**. This disposal strategy is designed to produce a durable waste form and subsequent system, that even allows groundwater at the disposal location to meet EPA drinking water standards.

Only the highly radioactive part of the waste is sent to a remote processing facility for conversion into borosilicate glass. After processing operations, which concentrate radionuclides and produce a waste stream amenable for vitrification **(10)**, glass frit is then added to the HLW stream. The slurry is then liquid-fed into a joule-heated ceramic melter in the vitrification facility. Here the waste mixture is melted at a temperature of 1150°C, using a unique off-gas system that is able to remove 99.999999% of the cesium in the original waste. The molten glass is then discharged from the melter and cast into Type 304L stainless steel canisters, 2-ft. in diameter and approximately 10-ft. in height. The units are then decontaminated by a frit blasting technique, which then recycles the used frit back to the front end of the process to help produce subsequent waste glass products. The canisters are welded shut using a resistance upset welding techique. This process and products produced are described in more detail elsewhere **(4,8,11-13)**.

- SRS DWPF

The Defense Waste Processing Facility [DWPF] represents the first major vitrification facility for immobilizing HLW constructed in the U.S. The facility, which contains 5 million cubic feet of volume and represents more than a billion dollar investment, is currently in operation at the Savannah River Site in Aiken, South Carolina. The DWPF, shown in **Figure 4**, is the largest plant of its type in the world today, and now the most prolific, recently pouring its 4 millionth pound of environmentally acceptable waste glass product. The design of the facility is based on the 35 years of successful operating experience of reprocessing plants within the Department of Energy (DOE) complex. Additional facilities are also planned

at other HLW sites in the U.S.

Figure 4 The Defense Waste Processing Facility (DWPF)

One of the most important pieces of equipment in the DWPF is a joule heated ceramic melter. Continuous melters using ceramic vessels have been used successful for many years in industry in production of a variety of different glasses. The joule heated ceramic melters operate by passing an electric current through the molten glass, causing heating. Continuous electric ceramic melters have resulted in higher production rates, more homogeneous products, and reduced volatilization for many applications. The DWPF melter is round in shape and consists of Monofrax K3 contact refractories and Inconel 690 electrodes. Another important feature of the melter is that it it is liquid fed, in which slurry of HLW and glass forming frit are injected directly onto the surface of molten glass at 1150ºC, where the components are then melted.

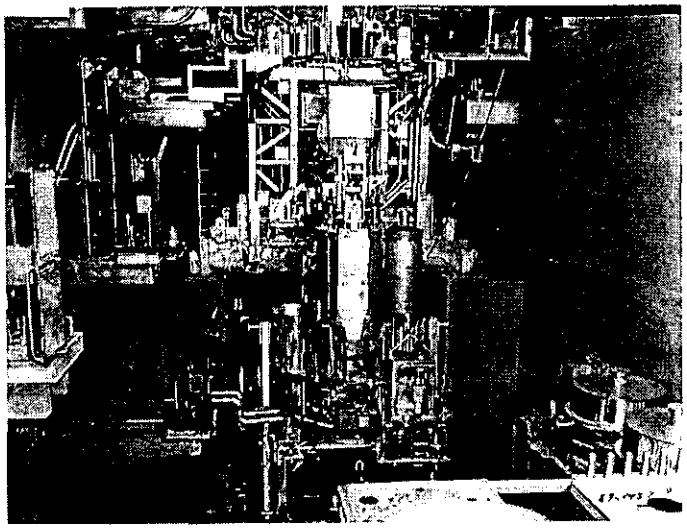


Figure 5 Joule Heated Ceramic Melter in DWPF Melt Cell

- SRS DWPF STATISTICAL PROCESS CONTROL (SPC) SYSTEM

The waste glass forms produced in the DWPF must be of high quality and able to be manufactured in both a highly reliable and effective manner. Therefore, properties of the waste glass must satisfy both processing constraints, to assure that production is optimized, and product quality considerations, to assure that a durable and stable wasteform is produced for storage and ultimate disposal. However, these important properties cannot be measured directly in situ and are instead, predicted from models relating the properties to composition, which can be routinely measured.

To further complicate controlling the DWPF vitrification process, the repository requires that product quality constraints be satisfied to a very high degree of confidence, demanding a statistically-based control system for DWPF process and product control. The system developed for the DWPF is called the Product Composition Control System or PCCS (**14,15**). To provide the necessary confidence, all pertinent sources of variation are estimated and incorporated into PCCS, which simultaneously provides sufficient flexibility to process varying waste types and feed compositions from the SRS Tank Farm. Furthermore, the statistical process control algorithms at the foundation of PCCS are rigorous to assure that only acceptable waste glass will be produced. This is especially important since once waste glass products are made, the glass cannot be re-

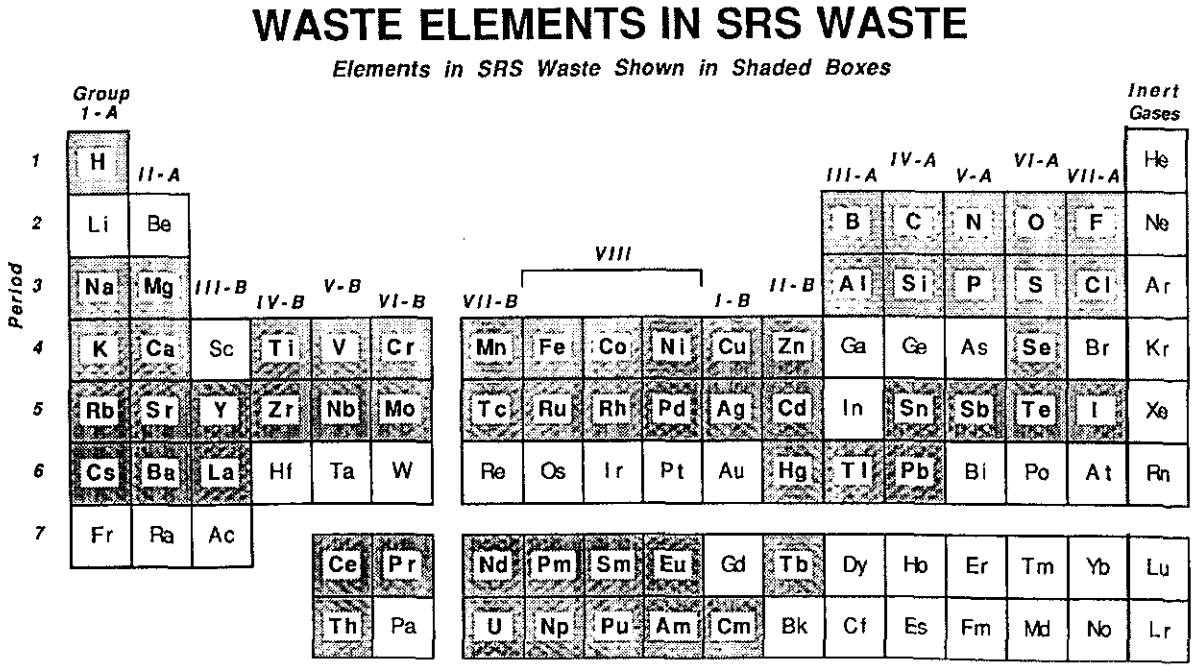
worked or re-melted. Finally, PCCS maximizes the efficiency of waste vitrified, so waste glass volumes and associated storage and disposal costs are minimized. No commercially-available alternative exists that can provide the rigor necessary for DWPF control while simultaneously furnishing the flexibility required to process all probable feeds. The PCCS is an important contribution to DWPF operations, because it represents a rigorous, statistically-defensible management strategy for a highly complex, multivariate system with multiple constraints imposed.

It is not possible to conduct routine measurements of key processing parameters, such as melt viscosity, electrical resistivity, and liquidus temperature, during radioactive operations. The key product parameter, glass quality, is able to be routinely measured using a seven-day crushed glass durability test on samples of the glass produced. Since the key process parameters are impossible to measure and glass quality requires more than a week to determine, models were developed from designed experiments that relate these key parameters to parameters that are easily measured in the process, i.e., composition. Consequently, these models help form the basis for the DWPF process and product control strategy.

TECHNICAL PEFORMANCE

As mentioned earlier, waste glass products that are fabricated must perform well in five main categories of technical performance. These include a) flexibility, b) thermal stability, c) mechanical integrity, d) radiation stability and e) chemical durability. These features will be discussed briefly below, with special reference to the designing of glass forms tailored for the vitrification process and for the ability of accommodating and immobilizing hazardous radionuclides (**4,13**).

Table 1
Waste Elements in SRS Waste Tanks



- Flexibility

Glass has demonstrated the ability to accommodate not only the 40 or more different elements that are found in individual waste tanks, but also the large variations in waste composition that occur **(13,16,17)**. The reason for this feature is a result of the relatively open random network structure that characterizes glass systems and its ability to accommodate elements or radionuclides of different sizes, charge, and characteristics, as well as differing amounts of constituents **(18,19)**. The diversity of the feed is illustrated in **Table 1**, which shows the many different elements found in SRS waste and frit formulations, which encompass many elements of the periodic table.

In order to develop glass matrices with the ability to immobilize this diversity in waste composition, various frit formulations have been developed world-wide. These are illustrated in **Table 2** **(4,20)**.

Table 2
Selected International Glass Frit Compositions for Immobilization of HLW

Component	Compositional Range	French AVH [R7T7]	German PAMELA [SM 513]	US SRS [165 Defense Waste]
SiO_2	45-77%	54.9%	58.6	68.0
B_2O_3	5-20%	18.9%	14.7	10.0
Na_2O	1-20%	11.9%	6.5	13.0
Li_2O	0-7%	2.4%	4.7	7.0
MgO	0-4%	-	2.3	1.0
ZrO_2	0-1%	-	-	1.0
Al_2O_3	0-3%	5.9%	3.0	-
CaO	0-7%	4.9%	5.1	-
ZnO	0-10%	3.0%	-	-
TiO_2	0-5%	-	5.1	-

As indicated in the table, borosilicate glasses have been the most widely studied systems for incorporation of nuclear wastes. Each component of the glass frit plays an important role by complementing the roles of waste constituents and by either assisting glass melting or by improving durability of the solidified product. Some of these key effects are shown in **Table 3** **(21)**.

- Thermal Stability, Mechanical Integrity and Radiation Stability

Waste glass products possess good *thermal stability*. Upon cooling from the melt or from self-heating due to radionuclide decay, waste glasses can phase separate or crystallize. Many different studies have been performed to assess the effects of these processes on performance of the glass **(22-25)**. In one series of experiments, SRS waste glass was purposely devitrified, even though extensive devitrification is not

expected for this system. The resulting time-temperature-transformation [TTT] curves identified phases formed and leaching tests showed that even in a 'worst-case' scenario, the effects of the crystalline phases on chemical durability of this system, were not significant.

Table 3
Effects of Frit Components on Processing and Product Performance

Frit Components	Processing	Product Performance
SiO_2	Incr. viscosity signif., reduces solubility	Incr. durability
B_2O_3	Reduces viscosity, Incr. waste solubility	Incr. durability in low amts, reduces in large amts.
Na_2O	Reduces viscosity & resistivity, incr. waste solubility	Reduces durability
Li_2O	Same as Na_2O but larger, incr. devitrif.	Reduces durability, but less than Na_2O
CaO	Incr. then reduces viscosity & waste solub.	Incr. then decr. durability
MgO	Same as CaO, reduces tendency for devitif.	Same as CaO
ZrO_2, La_2O_3	Reduces waste solubility	Incr. durability signif.

Waste glass products also possess good *mechanical integrity.* Cracking can occur due to stresses induced during fabrication or from accident scenarios during handling, transportation or storage operations. A variety of different mechanical tests have been performed on waste glass systems. The testing has ranged form laboratory-scale studies to drop tests of full-size canisters **(26-29)**. Based on these data, several important observations can be made. First, glass forms fracture into relatively large chunks inside canisters and fracture is generally localized to the area of impact. Second, the amount of increased surface area produced is low along with the amount of resulting fines or small particles. Waste glass products possesses more than adequate mechanical stability under anticipated as well as accident scenarios.

As radionuclides are incorporated into glass structures, a significant radiation field can be produced, so waste glasses must possess good *radiation stability.* Many studies of effects of radiation have been performed on waste glass systems and properties studied during these investigations include chemical and mechanical integrity, stored energy, helium accumulation, density changes and radiolysis **(30-35)**. Based on all existing data, waste glass forms perform very well under all of the radiation conditions expected during all stages of solidification and isolation of the HLW.

- Chemical Durability

The most important and most studied property of solidified waste glass forms is its *chemical durability*. This provides a measure of how well the waste glass will retain radionuclides under anticipated as well as accident scenarios. Nowhere is it more important than in the final resting place of the waste, the geologic repository. The two most important observations made after evaluating the chemical durability of a variety of waste glass systems under many different repository conditions is that 1) *leaching of glass is very low when subjected to realistic scenarios and conditions* and 2) *not only is chemical durability of waste glass good, but durability actually improves with time.*

Improved analytical capabilities have allowed investigators to obtain more insight into the corrosion processes of glass. In the late 1960's, an integrated study approach was first applied to studies of nuclear waste glasses, which combined solution analyses with a variety of bulk and surface studies of leached glasses, to obtain a more complete picture of the leaching process **(36)**. In subsequent years, new and more sophisticated surface analytical tools appeared along with more accurate solution analyses. The integrated study approach provides important information on the chemistry or structure of species of interest, on and within leached surface layers, as well as in solution. This, therefore, provides as complete a picture as possible of the leaching behavior and corrosion mechanisms of complex nuclear waste glass systems. Analytical tools used to study leached nuclear glasses are depicted schematically in **Figure 6**, along with
their sampling depths **(4,36)**.

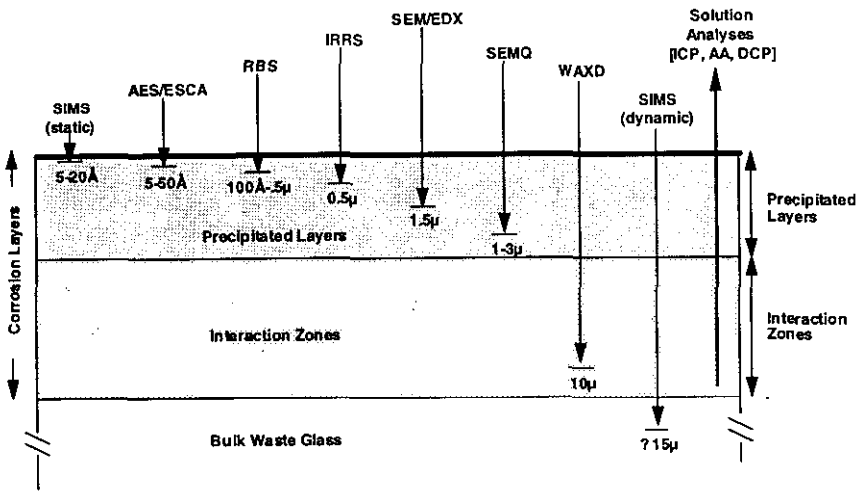


Figure 6
Integrated Study Approach

In order to assess, understand and ultimately, be able to predict the long term reliability of waste glass systems, chemical durability has been assessed as a function of important parameters that would be encountered during each stage of the solidification, transportation and interim and permanent disposal scenario. These important variables affecting the chemical durability of waste glass include time **(37-39)**, temperature **(40-42)**, solution pH **(43,44)**, Eh **(45,46)**, composition (waste, glass, leachate and

homogeneity) **(47,48)**, devitrification **(22-25)**, waste loading **(49,50)**, surface area of sample to leachant ratio [SA/V] **(51,52)**, flowrate **(53)**, pressure **(54)** surface finish, glass cracking and fines **(26-29)**, radiation effects **(30-34,)**, geology, hydrology, and package components [canister metal, possible overpack, potential backfills] **(21,55,56)**. Based on all data currently available, the chemical durability of waste glass forms should be extremely good when subjected to realistic values of these parameters.

WASTE GLASS STRUCTURE

As discussed earlier, it is the random network structure of glass that helps to make this structure so accommodating for waste constituents (**Figure 7**). Waste glass forms are composed of approximately 70% glass forming chemicals or frit added to about 30% waste constituents. Although there are many individual elements that comprise nuclear waste glass compositions, these components can play only one of three basic roles in the glass structure; *network formers*, *intermediates* or *modifiers* **(4,18,19)**.

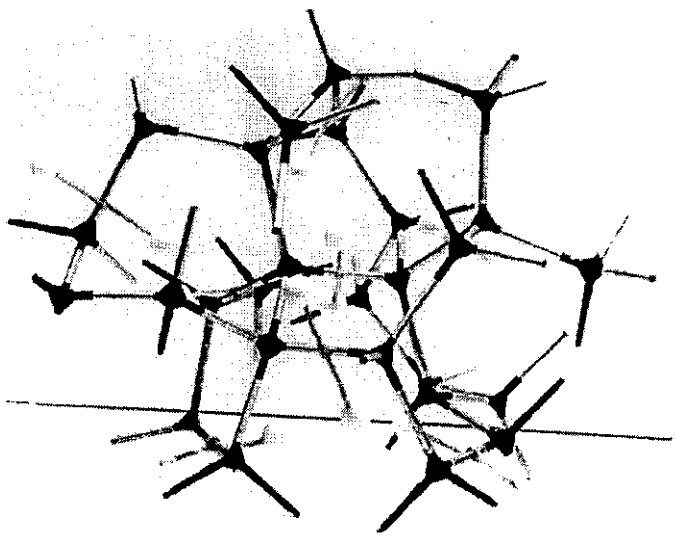


Figure 7 Random Network Structure of Glass

Constituents such as silica and boric oxide, are generally added to the waste as major components of the frit. The silicon and boron atoms are *network formers* and become located in the center of oxygen polyhedra in the configuration of tetrahedra or triangles. These polyhedra are then tied together by sharing corners, generally in accordance with Zachariasen's rules, which then makes up the 'framework or skeleton' of the random network structure of the solidified waste glass form. Another structural role that both glass frit and waste elements can play is that of *intermediates* , which is exemplified by major components found in the waste such as as alumina. These components can replace the network formers and still retain the framework structure of the glass. Other cations can move to the singly bonded oxygen ions that are created, for charge neutrality. The final role that components can play is the most prevalent, that of *modifiers.* In this case, important waste components such as cesium and strontium, along with alkali and alkali earth constituents, are located within the holes of the random network structure, and can also be associated with nearby singly bonded oxygen ions.

An important point to note is that both glass and waste components become an integral part of the random network structure of the glass. Components are incorporated by primary and/or secondary bonding which helps explain why the glass is able to retain radionuclides so well during leaching and why different elements can leach at different

rates. Based on this picture of waste immobilized in glass, a 3-stage corrosion process for nuclear waste glass was proposed, consisting of *Interdiffusion*, *Matrix Dissol*ution and *Surface Layer Formation* **(57)**. Both kinetic and thermodynamic modeling has been used successfully to describe and predict long-term behavior of these complex systems.

IN-SITU TESTING AND FIELD PERFORMANCE OF SRS WASTE GLASS

The ability of geologic formations to retain radionuclides is not without precedent. Two billion years ago, a chain reaction was started in a natural uranium ore deposit located at Oklo in Africa, and continued for hundreds of thousands of years before burning out. The waste that was generated by this natural event was trapped within this site. Hence, nature not only produced the first fission reator, but also the first geologic repository to contain the waste generated **(58)**.

The first field tests assessing the ability of waste forms to retain radionuclides in geologic formations was begun at Chalk River in the early 1960's by Canada. Following this pioneering effort, subsequent in-situ programs were characterized by cooperative, international undertakings, such as the burial of glasses, waste forms and package components in granite in Sweden, in clay in Mol, Belgium, in a limestone formation in the United Kingdom, and in salt, at the Waste Isolation Pilot Plant [WIPP] in Carlsbad, New Mexico. This later effort, the WIPP/SRS in-situ testing program is the first field tests involving burial of simulated nuclear waste forms to be conducted in the United States. It also represents the single largest, most cooperative venture of this type yet undertaken in the international waste management community.

- Materials Interface Interactions Tests [MIIT]

The WIPP/SRS Materials Interface Interactions Tests [MIIT] represents a joint effort managed by Sandia National Laboratories in Albuquerque, New Mexico and the Savannah River Site in Aiken, South Carolina and sponsored by the U.S. Department of Energy. MIIT involves the field testing of simulated or non-radioactive waste forms and waste package components supplied by seven different countries. Included in MIIT are over 900 waste form samples comprising 15 different systems, almost 300 individual metal samples of 11 types of potential canister and overpack materials and over 500 geologic and backfill specimens. In total, there are 1,926 relevant interactions that characterize this effort **(59-63)**.

The MIIT program is part of a larger effort at Savannah River aimed at understanding and being able to predict long term performance of DWPF waste glass in realistic repository environments. All samples, including waste forms, canister and overpack metals and geologic specimens, were fabricated in the shape of pineapple slices. They were then stacked onto heater rods in various stacking sequences in order to produce interfaces and interactions of interest. There are fifty assemblies in MIIT in seven different stacking sequences, which comprise a 7-part MIIT program, each part with its own set of specific objectives. The assemblies were inserted into brine-filled boreholes about 655 meters below the surface at WIPP in bedded salt deposits of the Salado Formation and heated to a temperature of 90°C [**Figure 8**] Samples and aliquots of

brine were obtained and analyzed after 0.5, 1 and 2-years as part of this 5-year burial study **(53-63)**.

- Post-Test Analyses of SRS Burial Waste Glasses

Based on both solution analyses and surface studies, the performance of SRS waste glass was found to be very good during the 5 years of field testing. There were two distinct regions observed in surfaces of leached glasses; an outermost precipitated layer consisting of two sublayers, and an inner glass reaction zone containing 3 sublayers. **(64)**. Analyses of the leached layers correlated well with solution data in the field experiments and findings were consistent with a variety of earlier laboratory support studies. Hence, many different simulated waste glass systems were seen to perform well not only in controlled laboratory tests, but also in a variety of multi-year field experiments.

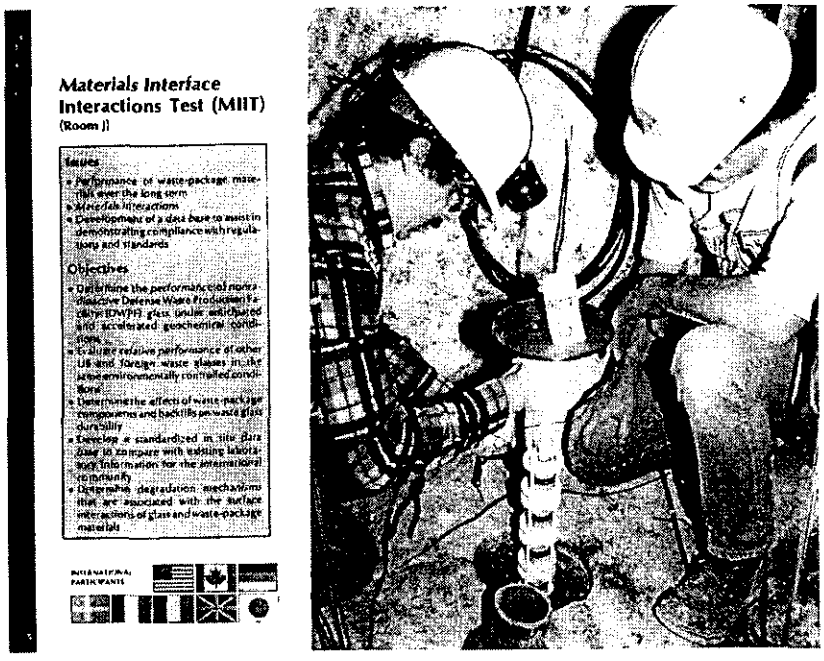


Figure 8 MIIT Field Tests at WIPP

D. PROPOSED NEW INTERDISCIPLINARY PROJECTS;

<u>LONG-TERM PERFORMANCE OF NUCLEAR WASTE GLASSES AND BIOMATERIALS</u>- A new interdisciplinary approach to improving and predicting performance

A common problem in many different fields and disciplines is to assess, understand and predict the long-term performance of materials of interest. This is necessary in order to determine the reliability of materials, often in very severe environments, and ultimately, to be able to extend their useful lifetimes.

During last year's ACerS Cocoa Beach meeting, a small group of interested individuals met to discuss a potential interdisciplinary strategy for extending lifetimes of materials

of interest. One objective included examining the methodology developed after more than 30 years of R&D development for assessing and predicting long term behavior of complex nuclear waste glasses in a complex repository environment, and to see if it is also applicable to other fields. Of special interest was to define ways of improving the performance of biomaterials and medical devices, within the challenging environment of the human body. For example, materials such as those used in hip replacements, work well for older, less active individuals, but for young adults and especially children with medical needs in this area, hip replacements may only last 8 to12 years. After that time period options for medical treatment are reduced and as these individuals approach middle age, their prognosis often becomes poor. If one can find a means to improve development of these and other important biomaterials and devices, and ultimately, improve their long-term performance and reliability, this could have a very important beneficial impact to the public.

After assessing the strategy for assessing and predicting long-term performance of nuclear waste glasses, it was found that this methodology may have relevance to other fields, including biomaterials and medical devices. A large flowsheet was develped examining potential similarities by a person who has worked for many years with nuclear waste glasses (George Wicks- Senior Advisory Scientist at DOE's Savannah River Site) in collaboration with someone who has much experience with biomaterials (Gary Fischman- Director of Biomaterials UIC, also previously with FDA). The team also received input from many other ACerS and NICE colleagues and may develop an interactive workshop at a future meeting on this subject, if there is enough interest.

The overall mission of this collaboration and interaction can be stated as follows:

MISSION

Examine a methodology to demonstrate long-term performance and reliability of important materials and systems in their final environment in an interdisciplinary manner, using experiences and expertise gained from the Nuclear Waste Management community.

A common, interdisciplinary approach for assessment and development of improved nuclear waste glasses and biomaterials/ medical devices was defined. Key common elements of this strategy and methodology include the following considerations:

INTERDISCIPLINARY TEAM

Nuclear Waste Glass Disposal vs. Human Implants, Prosthetics, Diagnostics, etc ;
In the nuclear community, a strategy and experimental program was developed and executed over a time period of more than 30 years, and involved many national and international collaborations. Stakeholders involved in key contributions came from national and federal laboratories, academia, government labs, agencies and organizations, the public, various peer review groups, as well as the industrial sector. Each of these groups provided important contributions and perspectives of the overall

program, and allowed the main objectives of the program to be met using all resources available, in a relatively cost effective, time efficient and productive manner.

INTERDISCIPLINARY OBJECTIVES

Nuclear Waste Glass Disposal; Predict the long-term performance of a 40-component waste glass system within a multi-barrier package in a complex geologic repository, in situ, out to time periods of 1,000 years (NRC regulations) and 10,000 years (EPA regulations).
Human Implants, Prosthetics, Diagnostics, etc.; Predict and improve the long-term performance and compatibility of materials and devices, in vitro, in a complex human system, out to time periods of 100 years.

INTERDISCIPLINARY R&D PROGRAM APPROACH

a) General 3-Stage Experimental Program
A 3-stage experimental program was used in the nuclear waste glass community that included thousands of individual and specific experimental studies. This overall program parallels that which is currently used to various degrees, in investigations of biomaterials and other related systems. This involves;

I. Laboratory Tests
II. Prototype Materials & Systems Tests
III. In-Situ Testing/ Clinical Trials

b) General Areas Performance
Important general areas of performance associated with R&D activities common to both fields include:

Nuclear Waste Glass Disposal	vs.	Human Implants, Prosthetics, Diagnostics, etc ;
• Compatibility	vs	• Bioactivity & Interactivity
• Chemical Durability	vs.	• Biocompatibility & Stability in Fluid Environments
• Mechanical Integrity	vs.	• Mechanical Integrity
• Flexibility	vs.	• Material & System Diversity
• Thermal Stability	vs.	• In-Vitro Stabilization
• Radiational Stability	vs.	• Photonic & Nuclear Effects
• Recycle/ Reuse	vs.	• Retrievability/ Reuse

c) General Experimental Features
There are general experimental features and considerations that were found to be especially important in nuclear waste glass studies. This considerations would also be expected to be relevant to other fields, including studies of biomaterials. These include:

- Standardized/ approved tests and procedures
- Accelerated tests
- Standards
- Natural systems
- M&I's
- Statistical significance of data
- QA
- Electronic databases
- Documentation and accessibility of data
- Interdisciplinary programs
- Many collaborative efforts
- Regulatory compliance issues
- Modeling for long-term predictions
- Peer review processes

SUMMARY

Based on all data currently available, the performance of nuclear waste glass systems is excellent, when tested under realistic conditions, as determined by many studies performed by many different investigators. In addition to possessing excellent chemical durability, the durability also improves with increasing time. This behavior has been observed not only in laboratory tests, but in actual field experiments as well. The observed time-dependent leaching behavior has been described by a variety of existing kinectic models, geochemical models and by the use of thermodynamic analyses, which relates the behavior of waste glass systems to natural glasses such as basalt, which have been stable for millions of years. While glass systems have demonstrated the ability to immobilize and retain radionuclides very well, it must also be emphasized that waste glass forms are only one part of a multi-barrier isolation system. This disposal scenario provides an extremely safe and effective means for disposing and isolating potentially harmful radioactive elements for not only our generation, but for generations yet to come.

Finally, the strategy and methodology used in more than 30 years of experimental studies for assessing, understanding and predicting nuclear waste glass behavior, was examined for possible applicability to other fields. It is believed that using the experience and expertise gained in the national and international waste management field, may be very helpful for predicting and optimizing the long-term performance of other materials in other areas, such as in development of biomaterials and medical devices, in the medical community.

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