

# Studies on Corrosion of Simulated Nuclear Waste Glasses Under Varying Physical and Chemical Environments

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## ABSTRACT

Leaching investigations of nuclear waste glasses are significant since they will be buried for an extended length of time. In the Phosphate systems, glasses were melted. We were able to melt the glass at significantly lower temperatures (even at 750°C) with a soaking duration of 30 minutes to 1 hour, which had previously been reported to be melted at temperatures above 1000°C. Lead Iron Phosphate (LIP) glasses are a subset of this system of glasses; the influence of the specific source of lead as a modifier was also discernible. LIP glasses provide various benefits, including a decreased aqueous corrosion rate and a lower processing temperature. Lead-iron phosphate (LIP) glasses are a promising new waste form for the long-term immobilization of both high-level defence and high-level commercial radioactive waste. LIP glasses have a number of benefits, including a reduced aqueous corrosion rate, a lower processing temperature, and so on. The lower the glass processing because the glass will be used to combine nuclear waste with some of the extremely volatile fission products. The higher the temperature, the better the glasses melt. The pH measurement of the leachate solution at ambient temperature exhibits intriguing and regular fluctuations ranging from 6.43 to 8.18. The results are consistent with previous authors' observed changes in equilibrium pH with changing leaching period. The Soxhlet leaching investigation of such glasses reveals lead ( $Pb^{2+}$ ) to be a preferable contender as a modifier towards leach resistance. The findings are supported by the ionic size, ionic radius, and hence ionic potential of the modifier ions introduced into the glass structure.

**Keywords:** Radioactive Waste, Safe Immobilization, Glass Matrix, Leaching, Corrosion.

## I. INTRODUCTION

The term "radioactive waste" refers to materials that contain or are polluted with a radioactive nuclide at a concentration higher than the safe level and have no practical value. Radioactive waste is typically produced as a by-product of nuclear power generation and other nuclear fission or nuclear technology applications, such as research and medicine. Most forms of life and the environment are endangered by radioactive waste, which is regulated by government authorities to protect human health and the

environment. i.e., radioactive waste refers to all waste items that are radioactive in nature. Nuclear waste is dangerous because it is not biodegradable, which means it does not break down spontaneously in the presence of oxygen [1-5].

It poses a variety of health risks to everyone who comes into touch with the waste's radiation. Harmful radioactive emissions can cause skin cancer and modify the DNA of anyone who comes into contact with them, with the consequences being handed down to the victims' descendants for many generations. There is no known chemical or

mechanical mechanism that can remove radioactive elements (radionuclides). Their ultimate demise is caused by radioactive decay into stable isotopes or nuclear transformation caused by atomic particle bombardment. The management of nuclear waste is becoming an important study topic all around the world [6].

Radioactive/nuclear waste can be disposed of in a variety of ways. Glass or glass-ceramics have been the most extensively used material for the immobilization of both HIGH LEVEL WASTE (HLW) and LOW LEVEL WASTE (LLW) in underground repositories all over the world. Because of its versatility, the huge number of elements that can be integrated in the glass and the potential for a highly durable and small volume waste form, hazardous and radioactive waste verification is appealing. A diversity of nuclear waste glasses has been examined for immobilization due to their ability to tolerate ions with a wide range of radii and charges [1,7]. Another benefit of glassification is its strong resistance to leaching. The most of borosilicate-based glasses that have been explored and produced for the immobilization of HLW to date are based on borosilicate compositions. In comparison to borosilicate glasses, research on phosphate-based glasses for HLW and actinide immobilization began in the 1960s, owing to the perceived advantages of lower melting temperatures and higher potential loadings for some waste compounds (e.g. molybdates or chromates) and especially actinide oxides [3]. Phosphate-based glasses can lower the volume of vitrified waste by up to 50% when compared to borosilicate glass waste forms for certain wastes. Different issues with previous phosphate waste forms, on the other hand, quickly became apparent. Sodium phosphate melts, for example, are chemically corrosive and must be melted in platinum-lined furnaces. Furthermore, the chemical durability of sodium phosphate glasses was weak, and durability studies revealed unacceptably high radioactive leach rates. Devitrification occurs in sodium phosphate

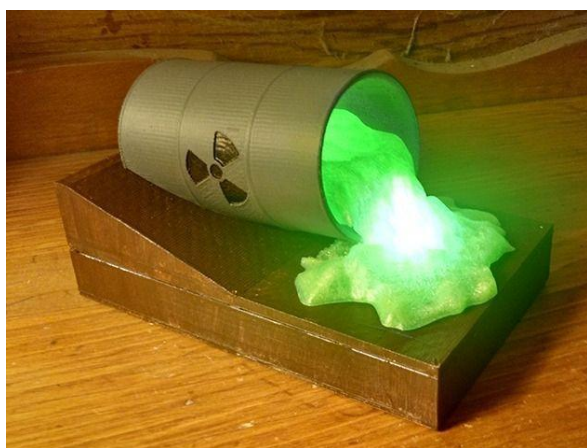
glasses as well, resulting in phases with even lower durability than the parent glass. Because of these issues with sodium phosphate glasses, all phosphate glasses were removed from the pool of viable host matrices in the early 1970s, and waste forms quickly emerged. Sodium phosphate melts, for example, are chemically corrosive and must be melted in platinum-lined furnaces. LIP glasses have several distinct advantages, including (i) 1000-fold reduced aqueous corrosion rate, (ii) a 100°C to 250°C lower processing temperature, and (iii) a very much lower melt viscosity in the range of temperature of 800°C to 1000°C. Most importantly, a method comparable to that established for borosilicate nuclear waste glasses can be used to process lead-iron phosphate waste [2,3]. When lead phosphate glasses were combined with various forms of simulated nuclear waste, it was found that a waste form might have a corrosion rate a thousand times lower than comparable borosilicate glass. The glasses in the systems of pure silicate, phosphate, borosilicate, and lead-iron phosphate (LIP), some of which were mixed with high uranyl acetate loaded with simulated nuclear waste and melted, were examined. In the case of silicate, phosphate, and borosilicate glasses, we were able to melt the glass at a much lower temperature (800 – 950°C) with a soaking period of 30 min to 1 hr. in comparison to earlier reports of 1000°C, whereas for one LIP glass in this work, it amounts to only 750°C for 30 min in which case the source of lead was PbO while in the situation of others it was Pb<sub>3</sub>O<sub>4</sub> against 1000°C for 1 hr. Leaching tests on these glasses were carried out over a 300-hour period using Soxhlet distillation and steam distilled water [1-3]. Weight losses and residual activities were tracked using the radiotracer technique in relation to the time period of leaching. The glass transition temperature for LIP glasses is in the range of 360 – 400°C, according to a thermal study (DTA). SEM micrographs of some of the selected glasses were reported. IR investigations show absorptions at 520, 1025, 1700, 2225, and 3450 cm<sup>-1</sup> [1,2]. The variation of

different properties was taken into account in terms of changes in the ionic charges and radii, and thus ionic potentials of the different modifier ions, the source of which were the respective modifier oxides, namely  $\text{PbO}$ ,  $\text{Pb}_3\text{O}_4$ ,  $\text{BaO}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{CaCO}_3$ ,  $\text{CeO}_2$ ,  $\text{SrO}$ , and  $\text{UO}_2(\text{CH}_3\text{COO})_2$  as for the various glass systems discussed above [8,9].

### Radioactive Waste:

Any substance containing or contaminated with a radioactive nuclide at a concentration greater than a permissible threshold is referred to be radioactive waste. In other words, radioactive waste is radioactive waste that contains radioactive elements that are no longer useful.

Nuclear power plants, medical isotope use, defense, the atomic industry, and laboratories are all sources of radioactive waste. The fuel cycle, which includes the mining and processing of uranium ores, generates the bulk of radioactive waste [10].



Figurer-1: Nuclear Waste

### Radioactive waste classification:

They are categorized into three groups:-

- 1) Low level radioactive waste (LLW)
- 2) Intermediate level radioactive waste (ILW)
- 3) High level radioactive waste (HLW)

Radioactive waste is produced in a variety of forms, including solid, liquid, and gaseous. The amount of

radioactivity produced varies depending on the source of production.

Exempt waste, low-level waste ( $37 - 3.7 \times 10^6 \text{ Bq/L}$ ), intermediate-level waste ( $3.7 \times 10^6 - 3.7 \times 10^{11} \text{ Bq/L}$ ), and high-level waste (above  $3.7 \times 10^{11} \text{ Bq/L}$ ) are the most prevalent classifications for radioactive liquid waste streams. Compressible or non-compressible solid radioactive wastes can also be classified as combustible or non-combustible, based on their physical properties. They are further subdivided by radioactive type and content [2,3].

### Low-level radioactive waste:

Low-level radioactive waste is defined as any radioactive waste that is not high-level radioactive waste, intermediate-level waste, or transuranic waste. Although it is larger in volume than high-level radioactive waste, intermediate-level radioactive waste, or transuranic waste, the radioactivity contained in low-level radioactive waste is significantly lower and made up of isotopes with much shorter half-lives than most isotopes in high-level radioactive waste, intermediate-level waste, or transuranic waste. Low-level radioactive waste is defined as large volumes of waste contaminated with tiny levels of radionuclids, such as contaminated equipment (glove boxes, air filters, shielding materials, and laboratory equipment), protective clothes, cleaning cloths, and so on. Even retired reactor components may fall into this group (after part decontamination procedures). Low-level waste has a low amount of radioactivity and has short half-lives of radioactive isotopes [11]. Most of the radioactive isotopes in low-level waste will decay after 10 to 50 years of storage, at which point the trash can be disposed of like regular garbage [11,12].

### High-level radioactive waste:

The waste comprising of spent fuel, liquid effluents from spent fuel reprocessing and solids into which the liquid waste is transformed is referred to as high-level

radioactive waste. Material from the core of a nuclear reactor or a nuclear bomb makes up the majority of it. Made up of fission fragments and transuranics, this waste contains uranium, plutonium, and other highly radioactive materials produced during fission. (Note that this definition does not define the amount of radioactivity required to be classified as high-level radioactive waste.) The decay rates of these two components differ. Transuranics, on the other hand, require roughly 500,000 years reaching such levels. The heat produced lasts for more than 200 years [13,14].

### What is glass?

A glass is a non-crystalline solid substance formed by a melt-quenching process, according to definition. Glass is also characterized as an amorphous/non-crystalline material that exhibits glass transition behavior and lacks long-range periodicity.

### Composition:

Amorphous (non-crystalline) solids include glass. Typical glasses are fragile and frequently optically clear. Soda-lime glass, which is made up of roughly 75% silica ( $\text{SiO}_2$ ) with  $\text{Na}_2\text{O}$ ,  $\text{CaO}$ , and a few other additions, has been used in windows and drinking containers for millennia. The term "glass" is frequently used in a limited meaning to refer to this specific use.

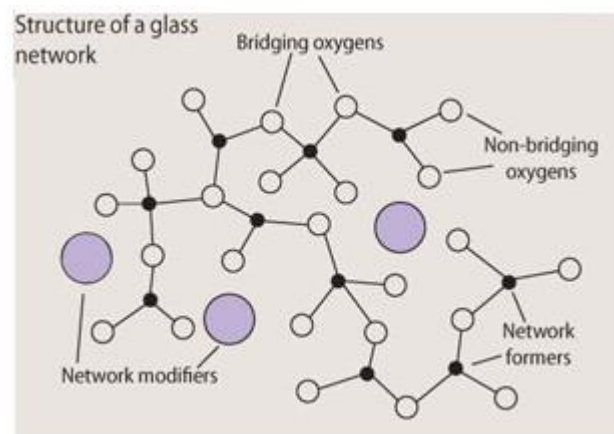
### Ingredients:

Quartz sand is the main raw materials in commercial glass production. Oxides such as  $\text{SiO}_2$ ,  $\text{B}_2\text{O}_3$ ,  $\text{GeO}_2$ ,  $\text{P}_2\text{O}_5$ ,  $\text{V}_2\text{O}_5$ , and  $\text{As}_2\text{O}_3$  are essential in the creation of glass because they constitute the foundation of random three-dimensional networks of glasses.

The network former, the essential building block of the glass, is one of the most fundamental compositional alterations that can be made. The former may be compared to the framework of the glass, and altering it will have a significant impact on how the finished product behaves. The glass is

referred to as borate glass when boron oxide is used as the former.

$\text{Al}_2\text{O}_3$ ,  $\text{Sb}_2\text{O}_3$ ,  $\text{ZrO}_2$ ,  $\text{TiO}_2$ ,  $\text{PbO}$ ,  $\text{BeO}$ , and  $\text{ZnO}$  are examples of intermediates. To maintain structural continuity, these oxides are introduced in considerable amounts to join up with the fundamental glass network.



$\text{MgO}$ ,  $\text{Li}_2\text{O}$ ,  $\text{BaO}$ ,  $\text{CaO}$ ,  $\text{SrO}$ ,  $\text{Na}_2\text{O}$ , and  $\text{K}_2\text{O}$  are examples of modifiers. These oxides are applied to glass to modify its characteristics.

### Structural and properties of glasses:

Such glasses have been roughly categorized in two ways. The first is based on the actual components contained in them, and the second on their intended application. Glasses based on silicate, phosphate, borate, borosilicate, and other networks fall into the first group, whereas optical glasses, sealing glasses, and laser glasses fall into the second. Metallic glasses, halide glasses, organic glasses, mixed anion glasses, and ionic glasses, are examples of other types of glasses. The following section provides a summary of the many structural characteristics and qualities of some of the oxide glasses [11,15].

### Why glass used?

Inside solid glass, dangerous radioactive waste has been confined. Glass has been the most extensively used material for the immobilization of both HLW and LLW in subterranean repositories all over the world. Glasses are adaptable with waste loadings and

can the capacity to incorporate most of the waste constituents, in addition to chemical durability, mechanical integrity, and thermal stability. Vitrification is the technique of integrating HLW into glasses by chemically dissolving them. Glass is now being employed on an industrial scale to immobilize high-level wastes. Several nuclear waste glasses have been examined for immobilization due to their ability to tolerate ions with a wide range of radii and charges. Because of its relative insolubility, glass is a good choice for long-term storage as well as being a compact and nuclear waste material. This format enables better storage and handling, resulting in space savings and cost savings.

### Which kind of glass is preferred?

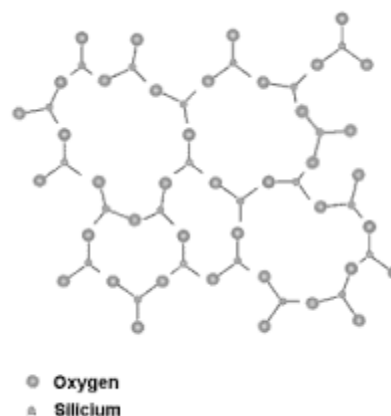
- 1) One of the most attractive qualities of the glass is its high leach resistance.
- 2) Because a significant percentage of Ru and Cs activity escapes the glass at higher temperatures (over 1100°C), a low melting glass is required.
- 3) Various waste compositions, including anions, can be accommodated.
- 4) High heat conductivity and radiation resistance.
- 5) Disperse the heat generated by radiation absorption.

## II. Some radioactive waste glasses

### 1) Silicate glasses:

"Silicate glasses," based on the chemical substance silica (silicon dioxide, or quartz), the principal ingredient of sand, are the most recognizable and historically the earliest varieties of made glass.

The coordination requirement of the  $\text{Si}^{4+}$  in the structure leads to the formation of silicate glasses, which may be thought of as an aggregation or polymerization process. The coordination of  $\text{O}^{2-}$  ions around  $\text{Si}^{4+}$  ions is four in vitreous silica, where each  $\text{O}^{2-}$  ion is exposed to two  $\text{Si}^{4+}$  ions.



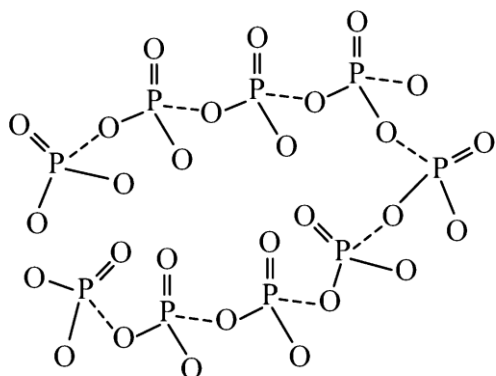
### Properties:

- 1) Excellent Transparency: Silica glasses have good transmission properties over a wide range of wavelengths, including visible, ultraviolet, and infrared.
- 2) Exceptional purity Silica glasses are made entirely of  $\text{SiO}_2$  and have very few metallic impurities.
- 3) Chemical Inertness of the highest kind: Chemically, Silica glass is very stable and chemically resistant. However, it should be used with caution if it will be exposed to hydrofluoric acid, phosphoric acid, Or alkaline solutions for a lengthy period of time, since these chemicals may etch it.

### 2) Phosphate glasses:

These glasses have been studied for over 150 years; however, while being one of the four classic glasses producing oxides (together with  $\text{SiO}_2$ ,  $\text{GeO}_2$ , and  $\text{B}_2\text{O}_3$ ), their nature, uses, and research and development have been limited due to their hygroscopic Nature. It wasn't until the addition of at least 30 mol% metal oxides was discovered to considerably increase the glass's durability that potential uses were aggressively researched. They are inorganic polymers based on the tetrahedral phosphate anion, which is formed by the phosphorus outer electrons forming  $\text{sp}^3$  hybrid orbital's ( $3s^2 3p^3$ ). The fifth electron is promoted to the 3d orbital, where strong-bonding molecular orbital's with oxygen 2p electrons are created, with charges balanced by polymerization or the presence of metallic ions.



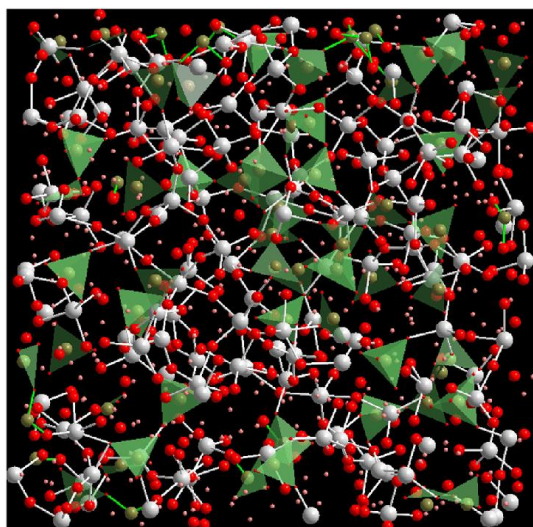


#### Properties:

- 1) Hydrofluoric acid is not a problem for phosphate glasses. They perform as excellent heat absorbers when iron oxide is added.
- 2) For the immobilization of radioactive waste, iron phosphate and lead iron phosphate glass are alternatives to borosilicate glass.
- 3) Phosphate glasses of various compositions have been studied with the purpose of vitrifying nuclear waste. Iron phosphate glass has recently emerged as a potential option in this application.

#### 3) Borate and Borosilicate glasses:

This Borosilicate glass is a kind of glass that is composed of silica and boron trioxide. Borate glasses with no moisture content are difficult to make under normal settings. Poch makes water-free Borate glasses by melting Ortho-boric acid over many hours at 1 mm Hg pressure.

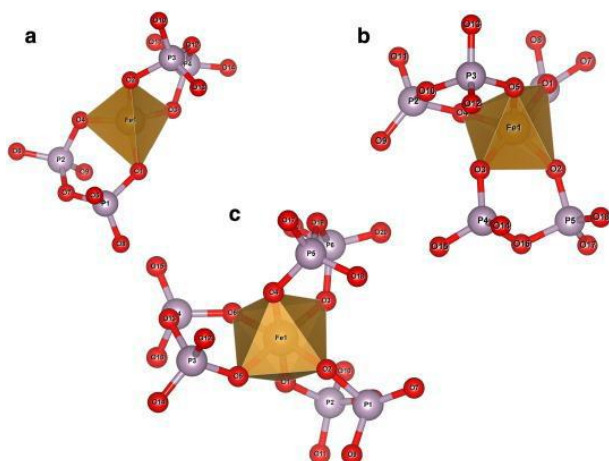


#### Properties:

Melting of high level nuclear waste should take place between 1100 and 1250°C; since higher temperatures cause excessive volatilization of both radioactive and non-radioactive waste components. Although the processing temperatures of alkali silicates are lower than those of vitreous silica, they are still too high for use in the vitrification of high-level nuclear waste. Glass formers other than silica are used to reduce the processing temperature even more. As the name implies, Borosilicate glasses are Laboratory glassware with low thermal expansion, high material strength, and chemical high stability.

#### 4) Lead-Iron-Phosphate (LIP) glasses:

In 1984 Sales and Boatner discovered and produced a unique phosphate glass frit as a stable HLW storage medium in 1984 [11]. Phosphorus pent-oxide, lead monoxide, and an appropriate proportion of ferric oxide are used to make lead-iron phosphate glass. The glass waste form of this new phosphate has several distinct advantages over borosilicate glass waste forms, including (1) lower aqueous corrosion rate, (2) lower processing temperature, (3) lower melt viscosity at processing temperature, and ease of making the solid waste form more than the ceramic phosphate waste form. Several various compositions of lead-iron phosphate glasses have been explored as possible radioactive waste hosts. Two basic procedures may be used to make the lead-iron phosphate glass frit that is mixed with nuclear waste and melted to generate radioactive waste monoliths. Most importantly, lead iron phosphate waste may be treated utilizing technology established for borosilicate nuclear waste glasses [15].



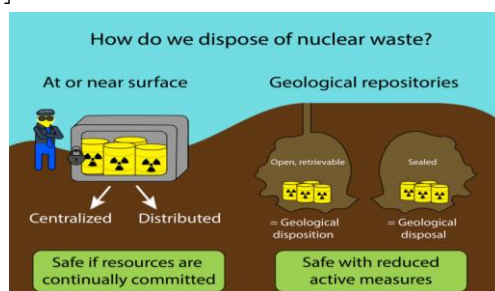
### Properties:

There are numerous significant advantages to using lead iron phosphate glasses:-

- 1) Aqueous corrosion rate that is approximately 1000 times lower.
- 2) A lower processing temperature of 100-250°C and a shorter melting time.
- 3) Melt viscosity is substantially lower in the temperature range of 800-1000°C.
- 4) Increased waste loading (smaller waste form volume.)
- 5) Excellent chemical resistance.

**Disposal technique:** Methods of disposal are entirely dependent on the type of the waste. LLW is not harmful to handle, but it must be disposed of with greater care than regular rubbish. It is usually buried in a shallow landfill. For disposal, ILW might be consolidated in concrete or bitumen. Short-lived garbage is often buried, but long-lived waste is

disposed of deep underground within a solid matrix [13-15].



## III. SOME EXPERIMENTAL STUDY

### Chemical Durability Testing:

Chemical reactions take place at the surface of a glass when it comes into contact with an environment, such as flowing or stagnant groundwater, corrosive gases and vapours, or aqueous solutions, and afterwards spread throughout the glass, based on the composition, pH of the solution, and temperature of the environment. In the last 30 years, scientists have gained a better knowledge of the chemical durability of glasses. The goal of these experiments was to better understand two characteristics of nuclear waste glasses' chemical durability. The capacity to anticipate glass durability and build glasses that fulfil certain leaching requirements based on short-term testing, and the ability to predict the long-term dissolving behaviour of glasses (on the order of 10,000 years or more) [16,17].

TABLE-1: Preparation of various Borosilicate and Silicate Glass Compositions (wt percent %)

Glass ID	SiO <sub>2</sub>	Na <sub>2</sub> B <sub>4</sub> O <sub>7</sub>	PbO	BaO	Fe <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>	CaCO <sub>3</sub>	CeO <sub>2</sub>	SrO <sub>2</sub>	Melting Point (+2°C)
BS1	29.6	20	38.4	12	-	-	-	-	-	-	950
BS2	40	24	-	-	13	3	20	-	-	-	950
BS3	33	25	-	-	13	-	-	29	-	-	950

BS4	39.6	20	33.4	7	-	-	-	-	-	-	800
BS5	40	24	-	-	13	3	-	-	20	-	950
BS6	29.6	20	38.4	-	-	-	-	-	7	5	800
BS7	40	24	-	-	13	3	-	-	-	20	900
S1	33.7	22.7 (Na <sub>2</sub> CO <sub>3</sub> )	43.6	-	-	-	-	-	-	-	1000

TABLE-2: Preparation of various LIP Glass Compositions (wt. %)

Glass ID	Pb <sub>3</sub> O <sub>4</sub>	PbO	Fe <sub>2</sub> O <sub>3</sub>	P <sub>2</sub> O <sub>5</sub>	CeO <sub>2</sub>	SrO <sub>2</sub>	BaO	Uranyl acetate
LIP3	49	-	8	33	10	-	-	-
LIP4	-	52.4	7.9	33.2	6.5	-	-	-
LIP5	49	-	8	33	-	10	-	-
LIP6	49	-	8	33	5	5	-	-
LIP7	49	-	8	33	-	-	10	-
LIP8	-	52.4	7.9	33.2	-	-	-	6.5
LIP9	-	51.4	7.9	33.2	-	-	-	7.5
LIP10	-	45.8	7.0	30.2	10	-	-	7.0
LIP11	-	44.05	6.76	28.45	-	-	-	20.47

**LEACHING STUDIES:**

Because radionuclides are more likely to be discharged through leaching through groundwater, and because the radionuclide danger lasts a very long time (up to roughly 10<sup>5</sup>-10<sup>6</sup> years), predicting long-term leach rates is vital to help in evaluating the safety. Immobilization of waste for long-term storage and disposal should be an irreversible procedure that prevents pollutants from escaping the matrix during storage and disposal. Leaching resistance is an important feature of any waste form because it dictates how effectively radionuclides of concern are maintained inside the waste form in a moist environment. A crucial aspect in evaluating an immobilization process is estimating the rate of leaching from a matrix during disposal. Radionuclide leakage is less likely when the matrix solubility is low. Radionuclide leakage is less likely when the matrix solubility is low. Because long-term leach rates

cannot be readily monitored, we must forecast them using a leaching model [18].

The Soxhlet technique is a leaching procedure for compounds that are poorly soluble. It relies on the leachate (solvent) evaporating and condensing as a result of heating. The water then drops down to the leachant (solid waste form). As a result, leachate is constantly wetting the leachant. Once the level has risen to the arm, the leachate returns to the bottom flask. The leachate can be supplied while the initial leachate is removed for examination, allowing for the construction of a time series of dissolution while avoiding saturation [1,2].

**Calculating the Leach Rate**

To estimate the leach rates, the BET surface area of the U-containing LIP glasses was obtained at liquid nitrogen temperature, and then leach rates were



computed using the equation 1 from the total weight loss for the total cumulative leaching duration.

$$LR = (W_i - W_f) / (SA \times t) \text{ ----- 1}$$

The weights of the sample before and after leaching are  $W_i$  and  $W_f$ , respectively. The leachant's surface area (glass powder) is  $SA$ , and the leaching time is  $t$ .

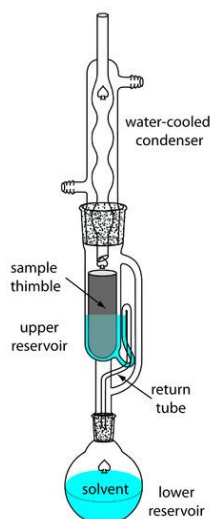


Figure- 2: The Soxhlate equipment used to leach test the glass samples is depicted schematically in this diagram.

### The Corrosion of LIP Glass in Aqueous Media as a Function of pH:

The resilience of iron lead phosphate glasses to water assault varies significantly with pH. According to Figure 3, the strongest chemical resistance is demonstrated for a neutral pH of around 7. However, the leaching of anions and cations is not always consistent. The unusual behavior depicted in Figure 3 can be explained by evaluating the size of the migrating Species at the water/glass interface. In the simplest scenario, the modifying cations will exchange with  $H_3O^+$  and the phosphate groups with  $OH^-$ . Cross links in the  $(PO_4^{3-})$  chains will be broken in the most acidic situations when both modifying cations are brought into solution, resulting in phosphorus loss and the concomitant leaching of cations and anions as seen (Figure 3). At high pH, however, the shortage of  $H_3O^+$  reduces cation

exchange, result in an increase in anion exchange. Modifying metals will not be casualties if phosphorous is lost from chain ends, as is anticipated, and the incongruent leaching behavior will not occur. Both nuclear waste glasses have the same compositions as those mentioned in the legend to figure 3; nonetheless, the corrosion rates of the other elements, including the waste components, follow the same general pattern when the solution pH is adjusted for each glass type [1,2,11].

When lead phosphate glasses were combined with various forms of simulated nuclear waste, it was discovered that a waste form might have a corrosion rate a thousand times lower than a similar borosilicate glass. Only when the lead phosphate glass was mixed with adequate simulated wastes was this impact noticed. As the solution pH increased, the 30-day corrosion rate of lead phosphate glasses in aqueous solution at 9000C reduced. For  $pH > 4$ , corrosion rates of  $10^{-3} \text{ gm}^{-2} \text{ d}^{-1}$  are possible. The corrosion resistance of the lead phosphate nuclear waste glass was mostly due to the presence of iron (a component of the simulated nuclear waste). Iron was shown to improve the chemical durability of lead phosphate glass and reduce the tendency for crystallization when it was cooled or reheated. After a 28-day corrosion test at 900°C, corrosion rates in distilled water for sintered lead iron phosphate (LIP) glasses are  $10^{-2} \text{ gm}^{-2} \text{ d}^{-1}$  [5,6]. Because the quantities of all components in the lead-iron phosphate leachate were below the analytical delectability limits of common analytical chemical procedures, the limiting values reported for lead-iron phosphate represent the analytical delectability limits for each element mentioned. Nuclear waste glasses made of lead-iron phosphate and borosilicate had the same waste-per-volume loading [19-22].

S.T. Reis et al. studied the 43.3PbO -13.7Fe<sub>2</sub>O<sub>3</sub>-43P<sub>2</sub>O<sub>5</sub> (mol percent) LIP glass, which has a

dissolving rate in water of  $1.26 \times 10^{-9} \text{ g/cm}^2 \text{ min}^{-1}$  and an O/P ratio of 3.5 [11].

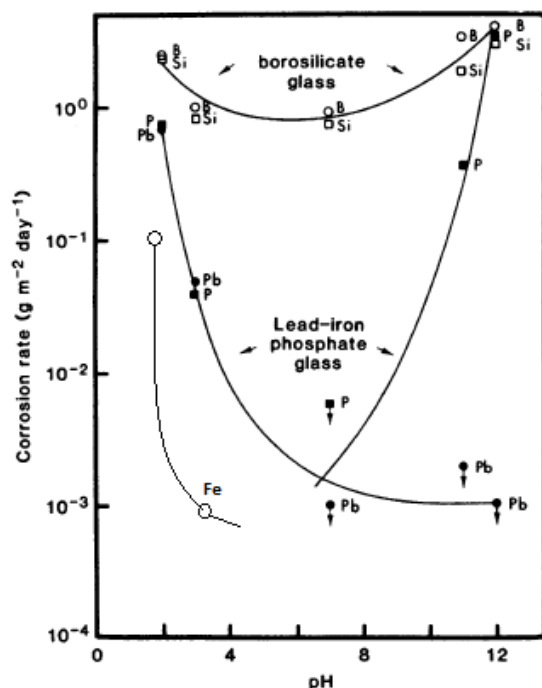


Figure 3: The correlation between the solution pH and the 30-day corrosion rates of lead-iron phosphate and borosilicate nuclear waste glasses at 90°C

#### IV. RESULT AND DISCUSSION:

The corrosion of nuclear waste glasses in various physical and chemical settings is critical since they have to be chosen for the long-term fixing of HLW or ILW. Many variables influence the corrosion or leaching of nuclear waste glasses (NWG), which may be divided into three categories: (i) system factors, (ii) leaching factors, and (iii) waste form factors. Time, temperature, radiation, and the ratio of waste form area to leachant volume are the system variables. Leachant variables, on the other hand, include (a) pH effects on leaching, (b) flow or replacement frequency, and (c) leachant composition. (a) Waste form composition, (b) Surface condition, (c) Porosity, and (d) Surface area to volume are the waste form parameters [23-26].

#### Modifier ions have an effect on the melting point of glasses:

The change in melting points is caused by the combined impact of several modifier ions, and the precise trend is difficult to predict. The crucial point to note here is that in the current study, we were able to melt the borosilicate glass composition at substantially lower temperatures (even at 800°C) with a soaking duration of 30 min. to 1 hr., which had previously been reported to be melted at temperatures above 1000°C. Given that the glass will be used to combine nuclear waste oxides with volatile fission particles such as Cs and Ru oxides, the lower the glass processing temperature, the greater the usefulness of the glass melt [27-32].

The melting temperature of PbO-containing borosilicate glasses (e.g., BS4 and BS6) is found to be lower (800°C) than that of other borosilicate glasses that does not include PbO. This suggests that PbO decreased the melting point of borosilicate glasses [4,5,].

#### PH analysis of borosilicate glass leachate solution:

The pH of the glass powder leachate solution was determined using a pH meter. pH was measured at 1 hour, 2 hours, 3 hours, 4 hours, and 5 hours intervals. The magnetic stirrer was used to stir the mixture every 15 minutes. Table-3 displays the results of such a pH investigation on chosen glassware, as well as graphs of pH change with regard to different time intervals.

Table-3: Soxhlet Leaching Investigation of Borosilicate Glasses

Glass	pH of the leachate solution Glass				
	1hr.	2hr.	3hr.	4hr.	5hr.
BS3	8.18	7.98	7.76	7.85	7.71
BS4	7.18	7.33	7.53	7.06	6.96
BS5	7.46	7.34	7.41	7.35	7.10
BS6	7.95	7.48	7.44	7.27	7.28
BS7	7.79	7.44	7.51	7.37	7.39

**pH of LIP Glasses' Leachate Solution:**

1g of definite size glass powder (0.30 0.42mm B.S.) was accurately placed in a beaker with a specified amount of distilled water of 40c.c. A magnetic stirrer was used to agitate the mixture for 2 minutes. A pH meter was used to determine the liquid's pH. pH levels were measured at 1 hour, 2 hours, 3 hours, 4 hours, and 5 hours intervals. The magnetic stirrer was used to stir the mixture every 15 minutes. Table-4 depicts the results of such a pH analysis on chosen glasses.

Table 4: Leachate pH after various time durations (LIP glasses)

Glass	pH of the leachate solution Glass				
	1hr.	2hr.	3hr.	4hr.	5hr.
LIP1	6.85	6.89	7.03	6.95	6.43
LIP3	7.05	7.25	7.35	7.25	7.07
LIP4	7.36	7.25	7.26	7.24	7.24
LIP5	7.02	7.06	7.09	7.13	7.12
LIP6	6.15	6.39	6.54	6.64	6.76
LIP7	6.78	6.84	6.76	6.80	6.63
LIP8	6.75	6.77	6.76	6.76	6.76

Because of the dissolved ambient CO<sub>2</sub>, the pH of pure distilled water is always somewhat acidic in nature. The pH of distilled water is also affected by the quantity of dissolved CO<sub>2</sub> inside it. In most situations, the pH of the leachate solution of the LIP glasses increases somewhat during the early stage. In certain circumstances (for LIP1 and LIP3), the pH of the leachate gradually increases, reaches a maximum, and then drops. The pH change of the leachate solutions over time for various LIP glasses was displayed in Fig 4.

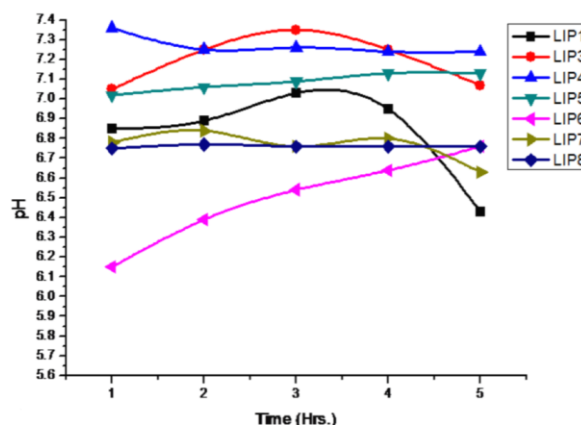
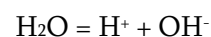


Figure 4: Leachate of pH vs leaching time (Hrs.) [for LIP glasses]

The little rise in pH that corresponds to the results might be explained by the H<sup>+</sup> exchange corresponding to hydration of an outer layer of phosphate chains. The hydration of the phosphate chain's outer layer released extra OH<sup>-</sup> into solution, causing an initial pH rise. The ion exchange and hence the glass composition will definitely influence pH variation. Phosphate ions can combine with H<sup>+</sup> ions to generate H<sub>3</sub>PO<sub>4</sub> (phosphoric acid), whereas cations would combine with OH<sup>-</sup> ions (producing hydroxide). It is also worth noting that the pH of the leachate solution (for LIP1-LIP8) remained almost neutral and hence had no further effect on dissolving rate. In the instance of LIP6, where the modifier oxides CeO<sub>2</sub> and SrO are evenly dispersed (5 wt%), there is a significant rise in pH during the time span. This might be due to the mixed oxide effect [1-5]. The slightly rising trend in pH values might possibly be attributed to the mixed oxide phosphate system. The measured pH values accord well with the equilibrium pH values of a glass system with a composition similar to that of the current work. The following equilibrium in the aqueous system can explain the result:



The different modifier ions produced during the leaching of the glass sample impact the amount of which (i.e., the forward or backward direction). In

our situation, the ionic radii ( $\text{\AA}$ ) of the various modifier ions are  $\text{Pb}^{2+}$  (1.33),  $\text{Ba}^{2+}$  (1.35),  $\text{Fe}^{3+}$  (0.69),  $\text{Ce}^{4+}$  (1.18), and  $\text{Sr}^{2+}$  (1.13). The equivalent ionic potential rises as  $\text{Ba}^{2+}$  (1.48),  $\text{Pb}^{2+}$  (1.5),  $\text{Sr}^{2+}$  (1.54),  $\text{Ce}^{4+}$  (3.39), and  $\text{Fe}^{3+}$  increase (4.69). The higher the ionic potential of  $\text{Mn}^{n+}$ , the more  $\text{OH}^-$  ions it will attract, forming the equivalent hydroxides  $\text{M}(\text{OH})_n$ . This creation of hydroxide causes the dissociation of  $\text{H}_2\text{O}$  to go forward, releasing additional ions into the medium. As the pH of the resultant solution decreases, it becomes progressively acidic. In the instance of LIP 8, the same figure reveals a plateau over the Results and Discussion time (hr.) of pH study. The sole difference between this glass and others studied is that it includes uranyl acetate (7.0 wt percent). Now that U can operate as a good glass maker, the glass does not dissociate, resulting in a flat pH value that is just below (6.75). As a result of the glass composition, more alkaline entities would be liberated, resulting in a rise in pH. It's also worth noting that the pH of the solution remained neutral, which had no effect on the dissolving rate [1-5].

#### LIP Glasses Leaching in Distilled Water under Soxhlet Conditions

Leaching The LIP glasses that did not include U (LIP3 to LIP7) were studied under soxhlet conditions for 24 hours, and the cumulative weight loss with respect to time (hrs.) is presented in Table-5, and the plot of these data is displayed in Fig.6 In the absence of U, the observed total weight loss (or percent weight reduction) with regard to cumulative time is as follows:

$$\text{LIP 6} > \text{LIP 4} > \text{LIP5} > \text{LIP3} \approx \text{LIP7}$$

Whereas LIP3, LIP4, and LIP6 include  $\text{CeO}_2$  as a modifier, LIP7 has  $\text{BaO}$ , and LIP5 contains  $\text{SrO}$ . The  $\text{CeO}_2$  concentration of the three glasses (LIP3, LIP4, and LIP6) changes as follows:

$$\text{LIP3 (10\%)} > \text{LIP4 (6.5\%)} > \text{LIP6 (5\%)}$$

and the observed weight loss with regard to cumulative time varies as; varies  $\text{LIP 6} > \text{LIP 4} > \text{LIP3}$ , Indicating that as  $\text{CeO}_2$  concentration increases, the percent weight loss decreases. As a result,  $\text{CeO}_2$  ( $\text{Ce}^{4+}$ ) can serve as an effective binder modifier [1,2].

Table5: Leaching Study of the LIP glasses under Soxhlet condition

Cumulative Leaching Time (Hrs.)	Cumulative Wt. loss (g)				
	LIP3	LIP4	LIP5	LIP6	LIP7
4	0.0008	0.0004	0.0033	0.0010	0.0000
8	0.0011	0.0022	0.0040	0.0030	0.0003
12	0.0017	0.0056	0.0040	0.0054	0.0003
16	0.0021	0.0062	0.0040	0.0082	0.0012
20	0.0021	0.0068	0.0050	0.0104	0.0022
24	0.0021	0.0074	0.0052	0.0114	0.0022

Table 6: Leaching study of LIP3 glass at different medium

Cumulative Leaching Time (Hrs)	Weight loss (g) in various media				
	Distilled water	0.001(M) HCl	0.01(M) HCl	0.001(M) NaOH	0.01(M) NaOH
4	0.0008	0.0014	0.0024	0.0009	0.0011
8	0.0011	0.0023	0.0038	0.0013	0.0017
12	0.0017	0.0032	0.0056	0.0018	0.0022
16	0.0021	0.0035	0.0062	0.0023	0.0027
20	0.0021	0.0041	0.0068	0.0025	0.0031
24	0.0021	0.0045	0.0074	0.0028	0.0035

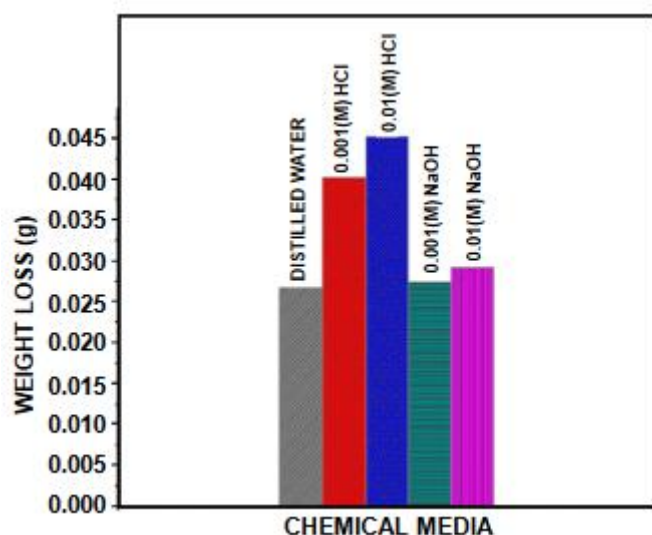


Figure 5: Leaching investigation of LIP3 glass in various media

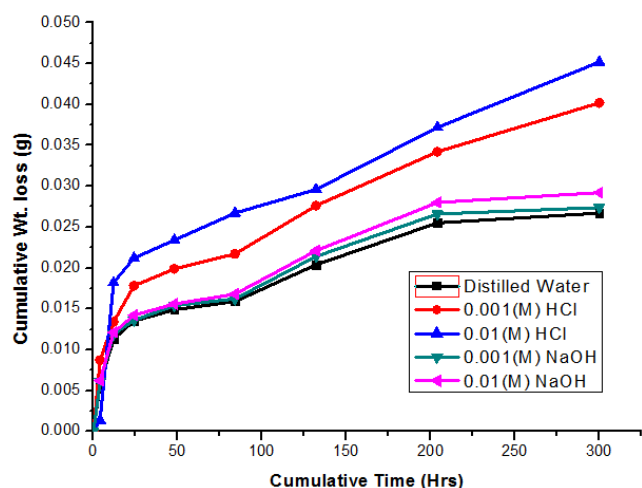


Figure 6: Weight loss in different mediums after 300 hours for LIP10 glass is depicted as a bar diagram. The LIP9 continuous leaching investigations.

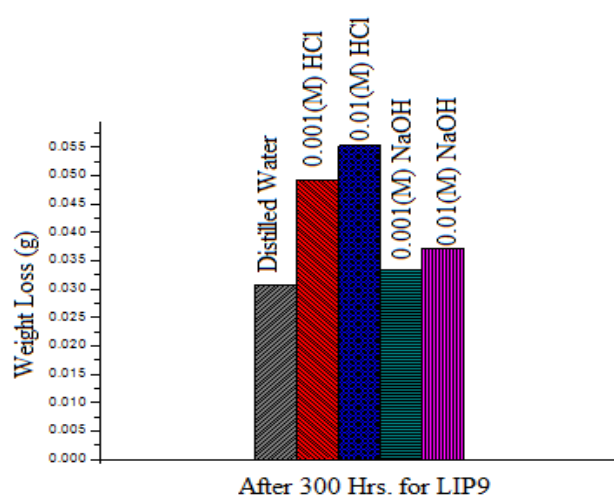


Figure 7: Weight loss in different mediums after 300 hours for LIP9 glass is depicted as a bar diagram.

## V. Conclusion of the Entire study with some Suggested Suppositions:

1. Glass melting temperatures can be decreased to some extent by using appropriate modifier oxide combinations. In the current study, we were able to melt the glass composition, LIP4, at a significantly lower temperature of 750°C after soaking it for 30-60 minutes. This is lower than the melting temperatures recorded previously (1000°C). The melting points of the PbO-containing BS4 and BS6 glasses are lower than those of the other borosilicate and silicate glasses. As a result of its glass forming role, PbO lowered the melting point of the glasses and the viscosity of the melted glasses. The melting values of all melted borosilicate glasses are lower than those of pure silicate glasses. The presence of modifier ions may be responsible for the temperature drop. The variation in melting point between the two systems might be attributed to differences in bond energies as well as the number of glass formers (two in borosilicate and one in phosphate and pure silicate).
2. The pH of leachate solution differs between the phosphate and borosilicate glass systems. In the case of borosilicate glasses, the pH of the leachate solution gradually falls as the extent of leaching increases (i.e., as the leaching period increases), whereas the pH of

the leachate solution gradually increases in the case of LIP glasses.

3. Lead was discovered to be an effective modifier in terms of leach resistance for both borosilicate/silicate and phosphate glasses.

4. Within the pH range of 5-10, the leaching of the LIP glasses does not alter. However, at extremely low pH (pH 4) the leaching rate increases to a greater extent, but at very high pH (pH > 10) the leaching rate grows slowly in comparison to the neutral situation.

#### Extrapolations from the Current Study:

The following suggestions for future research areas are based on an expansion of the research conducted in this study.

1. Real nuclear power plant liquid HLW or ILW must be examined and evaluated to determine the impacts of the contents in the LIP glasses.
2. To assess the actual leaching of the various compositions of the LIP glasses using the radio tracer approach, conductivity measurements and elemental analysis of the leachate solution are required.
3. To conduct a thorough leaching research on the LIP glasses over an extended length of time. Radiation damage, that is, the effect of gamma radiation on the leachability of LIP glasses.
4. LIP glasses Mössbauer and  $P^{31}$  NMR investigations.
5. Metal ion diffusion investigation to determine preferred dissolution.

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