

Chemical Durability of Uranium Oxide Containing Glasses

Enikő BITAY¹, Irén KACSÓ², Erzsébet VERESS³

¹ Sapiientia Hungarian University of Transylvania, Faculty of Technical and Human Sciences, Târgu-Mureș, Romania, ebitay@ms.sapiientia.ro

² National Institute for Research and Development of Isotopic and Molecular Technologies (INCDTIM), Cluj, Romania, iren.kacso@gmail.com

³ Transylvanian Museum Society, Cluj, Románia, veresserzsebet@gmail.com

Abstract

ZrO₂ doped Na-Ba-borosilicate glasses suitable as matrix materials for HLW immobilization were synthesized and corrosion behaviour was investigated in different aqueous media. Hydrolytic stability is increased with the doping level until 5 mol %; above this value the glass vitrification tendency is strongly intensified. Unexpectedly, ZrO₂ doping diminished the corrosion stability in 1M HCl solution, and low ZrO₂ content showed a low corrosion resistance in 1M Na₂CO₃ solution also. Doping effect was negligible in case of synthetic seawater. The glass structure is significantly stabilized by the integration of the 30% UO₃ added.

Keywords: borosilicate glasses, HLW immobilization, corrosion stability.

1. Introduction

Uranium glasses – uranium oxide (UO₂, UO₃, U₃O₈) doped oxide glasses – have been available on the market since the 1830s. The small quantity of uranium oxide added to the glass melt initially served only as a glass coloring agent, being used in the piece or small series production of diversely (typically yellow-green) colored, intense green UV fluorescence glass jewels (Figure 1.), decorative objects, dinnerware (glasses, bottles), enamels and fine ceramic glaze [1]

Uranium dental porcelain compositions of high esthetic aspect were developed starting from 1920-25, as uranium ions were found to repro-



Figure 1. Synthetic (glass) radioactive stones. Photo: R. Weldon. [9]

duce the broad fluorescence spectrum of natural teeth under daylight as well as under incandescent and UV light illumination, assuring a bright white effect of the artificial teeth (after the 1960s, the uranium content was limited to 0.05 %, and nowadays its use has been discontinued) [2–4].

In the 1930s, uranium glasses started their career as ion-sensitive (pH-sensitive) glass electrode membranes. Uranium oxide added in small quantities to a silicate glass melt significantly improved the hydrolytic stability and the electrical conductivity of the glass, opening up the possibility for development of glass electrodes with better functional parameters and longer working life. Furthermore, it gave rise to much more reliable glass electrodes for general and industrial use; and even biomedical microelectrodes suitable for *in vivo* monitoring. [5–8].

After 1945, the many and varied medical, industrial, agricultural, academic and scientific applications of the various radioisotopes were developed. The ever increasing use of nuclear power associated with civilian nuclear technologies has led to a rapid increase in radioactive waste as a by-product. The growing number of nuclear weapons and military applications, as well as the gradual destruction of nuclear weapon stockpiles

imposed by existing nuclear disarmament agreements, have also generated lots of active and depleted nuclear waste. Consequently, the environmentally secure long-term storage of radioactive waste is more and more critical. Presently, the preferred high activity (mostly uranium and/or plutonium containing) HLW (High Level Waste) encapsulation materials, besides Synroc-type glass-ceramics, are borosilicate or Fe-aluminophosphate matrix glasses. [10, 11].

Sufficient hydrolytic stability of uranium glasses has been an important requirement since their first commercial use. According to the published experimental data, in deionized water the mean U quantity leached from uranium drinking glasses is 0.052 (max. 0.63) $\mu\text{g L}^{-1}$, in diluted acetic acid it is 5.9 (max. 30.1) $\mu\text{g L}^{-1}$ and in the case of glazed ceramic tableware the U leached can attain 300000 $\mu\text{g L}^{-1}$ [12].

These data explain why after the 1970s, due to the potential damage to health – particularly from the swallowing U – commercial use of U compounds has been gradually restricted. However, investigations carried out on the stability of uranium glasses today are equally important because the quantity of HLW resulting from the nuclear industry, and the destruction of nuclear warheads (which necessitates environmentally safe long-term disposal options) is a constantly growing problem.

1.1. Uranium oxide containing HLW encapsulating glasses

The most important specific characteristics of the first generation HLW host glasses are their glass forming domain, crystallization properties, corrosion behavior, thermal, mechanical and radiation resistance. At the same time, of course, the economic aspects also have to be considered.

1.1.1. Borosilicate glasses [13–15]

Presently borosilicate glasses are the first choice as nuclear waste material matrices. The matrix glass composition is generally a Na-borosilicate, easily modified by the addition of further glass forming and glass modifying oxides (Al_2O_3 , Li_2O , CaO , BaO , ZnO , ZrO_2 , etc.) in order to optimize certain specific properties. They can integrate a significant amount of actinides (U, Pu) in their structure and are generally able to rapidly dissolve a broad spectrum of radioactive waste of different activities and provenance.

Borosilicate glasses are characterized by large glass-forming interval, and excellent chemical

(hydrolytic), thermal and (external and internal) radiation stability.

1.1.2. Phosphate glasses [16]

Phosphate glasses, the second most important HLW hosting glass family, differ in many of their properties compared with the borosilicate glasses.

Thermal stability is poorer, and excepting the iron oxide containing phosphate glasses, the chemical stability is much lower, especially in aqueous media. The melting temperatures and the molten glass viscosity are generally much lower, but this is offset by the high corrosivity of the molten material, which significantly shortens the working life of the melting furnace.

The chemical stability of the newly developed Na-Al-phosphate glasses is in some ways improved: leaching rates in the case of aluminophosphate-HLW and borosilicate-HLW are comparable. In corrosion experiments carried out in aqueous solutions and realistic long-term deposition conditions, the main normalized leached radionuclide quantity of the borosilicate-HLW tested was $0.3 \cdot 10^{-6}$ g/cm² Cs and $0.2 \cdot 10^{-6}$ g/cm² Sr daily, comparable with $1.1 \cdot 10^{-6}$ g/cm² Cs and $0.4 \cdot 10^{-6}$ g/cm² Sr obtained in the case of the aluminophosphate-HLW. [11]. However, the thermal stability is rather low; glass crystallization and temperatures above 100 °C significantly affect the hydrolytic stability (in the interim storage facilities temperatures can easily exceed 100 °C); the glass melt is still very corrosive.

Even if glass-forming properties of the newest Pb-Fe-phosphate glasses are below of those of borosilicates, they are sensibly improved: thermal stability is acceptable; chemical stability is good; the molten material is less corrosive. Matrix glass mixtures melt at (commonly by 100-250 °C) lower temperatures than the borosilicates (melting temperature ranges at 800-1000 °C), however for the complete dissolution of the HLW a temperature of at least 1000 °C is necessary.

By increasing the Fe_2O_3 content, decreasing the PbO content and adding CaO in the matrix glass mixture, the glasses chemical stability can be improved, especially in aqueous salt solutions.

1.1.3. Rare earth oxide glasses [17, 18]

The composition of the so-called Löffler glass, developed in the 1930s as an optical glass is 10-70% lanthanide oxide, 9-20% Al_2O_3 , the remaining part being SiO_2 . Lanthanide-borosilicate glasses are derived from the Löffler glass. Due

to the relatively high solubility of U, Pu and Am Löffler-type lanthanide-borosilicate glasses are convenient hosts for the disposal of these elements, nevertheless at present these glasses are without practical importance.

1.1.4. Aluminosilicate glasses [19, 20]

Aluminosilicate glasses could be good HLW host materials as they are capable of integrating 20 V/V% UO_2 . The disadvantages are their relatively high melting temperature and relatively high crystallization tendency.

The recently synthesized and studied aluminosilicate glasses are characterized by lower melting temperatures, lower viscosity and significantly higher durability. These being considered, based on the results obtained in the light of new developments, in perspective they could be the first representatives of the second generation HLW hosting glass family.

1.1.5. Sintered glasses [21]

By reason of the difficulties raised by the obtainability of high SiO_2 content quartz glasses, researchers are trying to prepare them by sintering of the sol-gel synthesized starting material. Use of the sintering procedure could reduce the temperature required to obtain and process a good HLW hosting quartz glass by several hundred grades, making it possible at the same time to avoid not only the major energy expenses, but also the loss of the volatile radionuclides (Cs, Ru, Mo, Tc).

The HLW to be disposed is practically permanently integrated in the structure of the host quartz glass. As indicated by the microstructural investigations performed, despite the diffusion of the Cs and Ba nuclides and the chemical reaction of the matrix with the Ce, Nd and Zr nuclides, structural integration of the most part of the waste material in the sintered host takes place by clustering processes, not dissolution.

This paper presents experimental results obtained by investigations of the chemical stability of some HLW hosting sodium-borosilicate glasses.

2. Glasses preparation and characterization

2.1. Synthesis

Matrix glasses (Table 1) were prepared by the melt-quench method, without specific thermal treatments.

The thoroughly homogenized mixture of the finely pulverized dry p.a. grade starting compo-

nents (oxides, carbonates, boric acid) was melted in atmospheric conditions in a platinum crucible, stirring the melt periodically with a platinum rod. The progress of the synthesis was followed by visual examination of sample drops removed at the end of every stirring. At the end of the synthesis the homogeneous glass melt was quenched by pouring the melt onto a 5 mm thick stainless steel plate.

Under the same conditions the 70% matrix glass + 30% UO_3 (9 mol% UO_3) compositions was also melted.

2.2. Investigations performed, results

2.2.1. Hydrolysis of the borosilicate glasses

Borosilicate glasses are solid electrolytes structured in a locally ordered atomic network consisting of Si, B and O atoms forming mostly $[\text{SiO}_{4/2}]$ and $[\text{BO}_{3/2}]$ electrically uncharged units and $\equiv\text{SiO}^-$ and/or $\equiv\text{SiOB}\equiv$ anions. The negative charges of the

Table 1. The matrix glass compositions

Glass type	Oxidic composition (mol %)				
	SiO_2	B_2O_3	Na_2O	BaO	ZrO_2
B05Ba	65.00	5.00	25.00	5.00	
B05BaZr1	64.00	5.00	25.00	5.00	1.00
B05BaZr2	63.00	5.00	25.00	5.00	2.00
B05BaZr3	62.00	5.00	25.00	5.00	3.00
B05BaZr4	61.00	5.00	25.00	5.00	4.00
B05BaZr5	60.00	5.00	25.00	5.00	5.00
B10Ba	60.00	10.00	25.00	5.00	
B10BaZr5	57.14	9.52	23.80	4.76	4.76
B15Ba	55.00	15.00	25.00	5.00	

anionic units are neutralized by ionically linked neighboring alkaline and alkaline earth cations of differing mobilities. Penetration in the glass bulk of the H^+ ions from the aqueous solutions contacting the compact glass surface is strongly hindered (Na^+ mobility in the quartz glass e.g. is 104 greater than H^+ (H_3O^+) mobility). [22].

Referring to silicate glasses Baucke [23] conceived several hypotheses which are also valid to borosilicate glasses:

- Structural cations in the glasses in contact with aqueous electrolytes are influenced by the potential gradient generated by the concentration gradient
- $[\text{SiO}]^-$ and $[\text{SiOB}]^-$ type anionic groups of the glass surface belong equally to the glass phase

and the aqueous electrolyte solution in contact with the glass surface and are in equilibrium with the ions existing in electrolyte solution

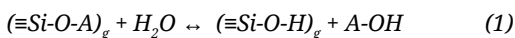
c) Due to the equilibrium processes after the hydration of the glass surface the hydrolysis of the silicate (borosilicate) glass network occurs (glass corrosion).

Vizes elektrolitoldatban tehát a határfelületi folyamatok következtében a boroszilikátüveg felületi rétege megduzzad (hidratálódik), majd oldódni kezd (korrodálódik, hidrolizál).

Thus, in aqueous electrolyte solutions the surface layer of the borosilicate glass becomes swelled (hydrated), the hydration process being followed by its dissolution (corrosion, hydrolysis).

The corrosion rate and extent is defined by two basic processes [24, 25]:

- ion-exchange controlled diffusion



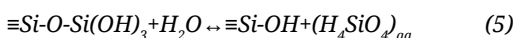
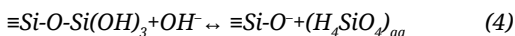
(A – any alkaline cation)

- hydrolysis of the silicate network:



The rate of the ion-exchange reaction is defined by the pH-dependent effective diffusion coefficient D_A of the interdiffusion process, and the E_A^d activation energy characterized by an Arrhenius-type temperature dependence (the activation energy of leaching processes is typically $\approx 30\text{--}145$ kJ/mol)

The alkaline components leaching due to selective dissolution and exchange to hydrated protons (H_3O^+) during the ion-exchange reactions, generates an alkaline ions shortened hydrated glass surface layer. In the following hydrolysis process hydroxyl ions contacting the hydrated glass surface react with the silicate network-resulting silicic acid (H_4SiO_4) which leads to the complete dissolution of the glass network:



2.2.2. Hydrolytic stability investigations

Corrosion stability of the synthesized matrix glasses and the corresponsive 30% UO_3 containing glasses was determined in different aqueous media: distilled water, acidic solutions, alkaline solutions, and saline solutions corresponding to seawater salinity. In the case of distilled water the method applied was that specified in the ISO 719 standard, in the other cases the properly modified version of the before mentioned method was used.

2.2.2.1. Experimental

Before carrying out determinations, the ground glass samples of controlled particle size were conditioned according to the ISO 719 standard: thorough washing with bi-distilled water, rinsing with p.a. ethanol, drying at 60 °C. The corrosion media used were bi-distilled water and the following solutions, prepared in bi-distilled water solvent and analytical grade reagents: 1M HCl, 1M Na_2CO_3 , and synthetic seawater (Table 2.).

Static corrosion experiments were performed on the following ground glass fractions obtained using Retsch laboratory sieving equipment:

- in bi-distilled water: 0,3–0,5 mm
- in 0,1 N HCl solution: 0,5–1,0 mm
- in 0,1 N Na_2CO_3 solution: 1,0–1,5 mm
- in synthetic seawater: 1,0–1,5 mm

Samples of 0.5 ± 0.05 g of the proper glass fraction were analytically (± 0.0001 g) transferred in 25 cm³ volumetric flasks filled to the sign with the corresponding corrosive solution. The prepared open volumetric flasks were kept at 98 °C for 2 hours, then the room-temperature flasks were refilled with the corrosion solution and were homogenized. The samples used for the measurements were taken from these stock solutions.

The mass loss measuring procedures of the usual standards given for glasses in case of acid corrosion (ISO 1776), and corrosion in alkaline and salt solution media (ISO 695) couldn't be applied because of the limited quantity of the glasses, therefore, the leached elements in these cases were determined by ICP-AES spectrometry. ICP-AES method compliance was verified by comparing the experimental results with the ISO 719 method results obtained on the same samples in the case of hydrolytic stability in bi-distilled wa-

Table 2. Synthetic seawater composition [26]

Component	Content (g/L)
NaCl	23.9985
Na_2SO_4	4.0111
KCl	0.6986
$NaHCO_3$	0.1722
KBr	0.1000
H_3BO_3	0.0254
NaF	0.0029
$MgCl_2$	5.0290
$CaCl_2$	1.1409
$SrCl_2$	0.0143

ter. Due to the parallelism of the two data series, concordance between the two different methods was considered satisfactory.

2.2.2.2. ISO 719 titrimetric method

Samples taken from the extraction solutions obtained were potentiometrically titrated using 0.01N HCl solution as titrant, pH glass measuring electrode and Ag/AgCl reference electrode. (Standard deviation: 0,1%). Corrosion (hydrolytic) stability is defined by the Na₂O quantity extracted from 1 g glass sample in µg units, and by the consumption of the titrant solution used in mL (Table 3.).

2.2.2.3. ICP-AES determinations

ICP-AES measurements were performed on a Jobin-Yvon 24 type sequential ICP-AES spectrometer provided with argon gas of 99.995% purity for the plasma. Determinations were repeated on three parallel samples for every element in the case of every glass, working on the spectral lines presented in Table 4.

In order to assure the correctness of the determinations, a five point matrix-identical multi-element calibration was performed in every corrosion medium at five properly chosen concentration levels. The concentration of the standard solutions used in case of the determinations on bi-distilled water extracts were 0.0; 0.5; 1.0; 5.0 and 10.0 ppm for every element (Figure 2). Standard deviation of the method was of 1-2%.

In case of the bi-distilled water and the acidic solution extracts the concentration level of the standard solutions and the matrix (the extract) was analogous and the sample solutions to be measured could be used "as is". As part of the metallic ions separated (precipitated) from the seawater

Table 3. Glass hydrolytic classes (ISO 719)

V 0,01M HCl mL	Na ₂ O µg	Hydrolytic class
≤ 0,1	≤ 31	1
0,1–0,2	31–62	2
0,2–0,85	62–264	3
0,85–2,0	264–620	4
2,0–3,5	620–1085	5
≥ 3,5	≥ 1085	> 5

Table 4. The spectral lines used for ICP- AES determinations

Element	Ba	B	Si	Na	Zr
λ (nm)	233.527	208.959	212.412	588.995	257.139

and the alkaline standards, in these cases the calibration standards and the measurable samples were previously diluted, applying a dilution ratio of 1:10.

2.2.3. Results

All the synthesized glasses, derived from the 65SiO₂·5B₂O₃·5BaO·25Na₂O [mol %] oxide system, can be arranged in the following two groups:

- B05Ba (alap); B05BaZr1.....B05Ba5Zr5;
- B05Ba (alap); B05Ba, B10Ba, B15Ba; in this latter the ZrO₂ doped B10BaZr is included, proven to be the optimal composition.

2.2.3.1. Titrimetric method (ISO 719)

The ISO 719 standard method of hydrolytic stability determination was only applied to the group (b) matrix glasses and their 30% UO₃ doped variants. Titrations were performed on three 5 ml extract samples for every glass sample.

As expected, in the case of similar B₂O₃ content the ZrO₂ addition increases the hydrolytic stability of the Ba-borosilicate glass system. At the same time it can be ascertained that the integration of the UO₃ in the glass network [27, 28] has the effect of stabilization of the glasses, raising their compactness and corrosion stability (Figure 3, Table 5.).

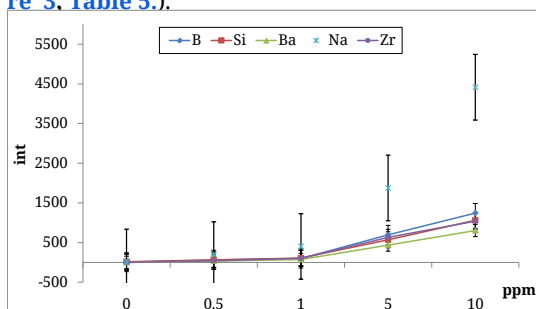


Figure 2. ICP-AES calibration in bi-distilled water

Table 5. Effect of UO₃ integration in the glass network on the hydrolytic stability

Glass type	Hidrolitikus stability (0.01N HCl, mL)		Density at 20°C (g/cm ³)	
	Mátrix	30% UO ₃	Mátrix	30% UO ₃
B05Ba	2.25		1.857	
B10Ba	4.50	0.44	2.014	2.183
B10BaZr	2.87	0.14	2.307	2.484
B15Ba	2.35	0.56	2.208	2.424

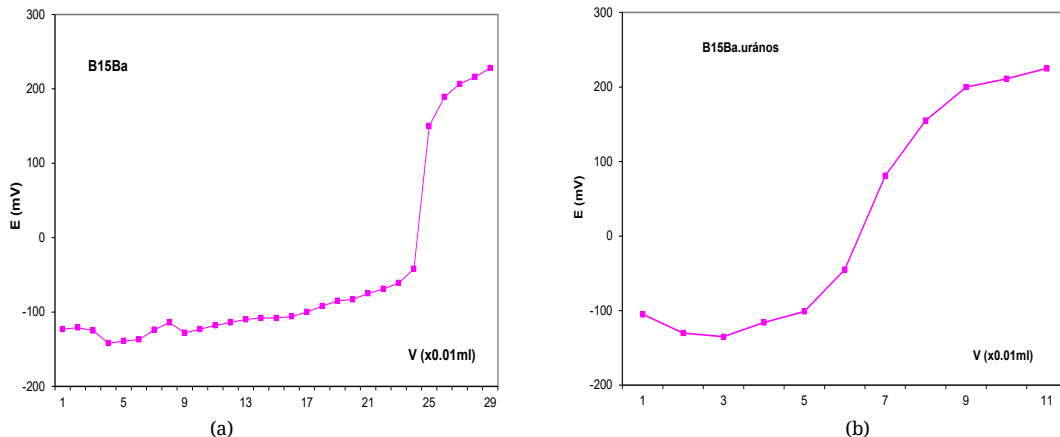


Figure 3. Titration curves obtained for B15Ba (a) and (70%) B15Ba-(30%)UO₃ (b)

Table 6. Influence of the ZrO₂ integration in the glass network on the corrosion stability vs. different aqueous media

Glass type	Normalized total extracted oxide quantity µg/g ground glass			
	H ₂ O	0.1N HCl	0.1N Na ₂ CO ₃	See water
B05Ba	8.88	1.99	3.37	0.69
B05BaZr1	30.15	29.67	9.92	0.42
B05BaZr2	27.66	24.67	2.33	0.31
B05BaZr3	19.98	18.63	3.55	0.47
B05BaZr4	14.93	14.93	5.83	0.36
B05BaZr5	10.91	13.39	2.93	0.42

2.2.3.2. ICP-AES method

Experimental results obtained by the ICP-AES method for the hydrolytic stability (vs. H₂O) and the chemical resistance (vs. acidic, alkaline and saline solutions) of the group (a) matrix glasses is presented in Table 6. (Normalized total metal ion quantity extracted in static conditions at 98 °C). Table 6 data indicate that at the B05Ba (basic) glass derived ZrO₂ doped glasses the influence of the ZrO₂ network integration [29, 30] on chemical stability of the glasses is observed starting from 2 mol % ZrO₂ added. In order to completely clarify the network stabilization effect tendency in this case, further study is required.

3. Conclusions

Hydrolytic stability of the studied Ba-borosilicate matrix glasses is increased by the B₂O₃ addition starting from 10 mol % B₂O₃ added.

At the same B₂O₃ content ZrO₂ doping increases the hydrolytic stability. The effect is maximal at 5 mol % ZrO₂. Further doping significantly increases the vitrification tendency.

In the case of vitrification of Zr-containing wastes, the fact that ZrO₂ addition decreases the corrosion resistance of borosilicate glasses in acidic media should be taken into account.

UO₃ integration in the glass network increases the chemical stability significantly.

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