Available online www.jocpr.com

Journal of Chemical and Pharmaceutical Research, 2016, 8(4):107-111

Research Article

ISSN : 0975-7384
CODEN(USA) : JCPRC5

Beta rays affect on verification the nuclear waste in glass and glass-ceramics

Aliaa A. Abdeljabbar and Asia H. Al-Mashhadani

Department of Physics, College of Science, University of Baghdad, Baghdad, Iraq

---

ABSTRACTS

Nuclear waste in Iraq has three major sources; spent nuclear fuel from the old Iraqi Atomic Energy Organization in Al-Twitha site, waste from radioactive isotopes used in medicine and waste generated by the nuclear weapons used in the war against Iraq in 2003. These waste need to storage until becomes safety. Therefore in this study nuclear wastes (strontium oxides) were stored by vitrification methods in two typesof borosilicate glass (glass and glass-ceramics)and the effect of beta ray on the immobilized waste was done. Then the physical, chemical and mechanical properties of borosilicate glass contain strontium oxide before and after irradiated by beta ray were investigated. It found that the physical properties and the leaching was not affected by beta-ray, while the values of surface strength and microhardness of glass increased with increasing radiation doses and their values have remained constant for the glass-ceramics.

Keywords: Beta rays, verification the nuclear waste, glass and glass-ceramics

---

INTRODUCTION

The nuclear waste needs to storage for long time until loss radiation effect (became safety). Therefore must found suitable method to immobilize nuclear waste to prevent leaching ions to surrounding and became hazards threatens environment and our health[1].

There has been considerably more research and technology development conducted on glass waste forms than any other waste form over the past 50 years. This is because their amorphous and relatively disordered structures can incorporate a wide range of chemical elements. Borosilicate compositions that include 30 to 40 different elements are used routinely for high level waist(HLW). Such compositions produce highly durable glasses[2-6]. Most elements play one of three basic roles in glass structures: network formers, network modifiers and intermediates. In borosilicate glass structures, the network is primarily formed of the chains of borate and silicate polyhedra, lithium, sodium and calcium are typical network modifiers that create non-bridging oxygens or provide charge balance for some HLW elements[7,8]. A commonality exists between the many different radioactive waste glass systems and the structural role components play in a glass. Compositionally, the glass forming elements in HLW glasses constitute 60 to 85 wt% of the glass structure; network modifiers make up 0 to 25% of the glass, while 15 to 40 wt% are intermediates[9].

Borosilicate glasses have some disadvantages, notably the off-gas systems needed to cope with HLW volatility at the glass melting temperatures and the low solubility in glass of some important radionuclides, such as Tc and actinides. With a strong premium on repository and interim storage space worldwide, the higher waste loadings achievable by crystalline ceramic waste forms are advantageous for specific waste streams. In addition, mineral analogues for many ceramic waste forms provide evidence of long-term durability[10].

In crystalline ceramic phases, radionuclides can be incorporated to occupy specific atomic positions in the periodic structures of constituent crystalline phases, which allows high loading of specific radionuclides. The coordination polyhedra in each phase impose specific size, charge, and bonding constraints on the radionuclides that can be incorporated into the structure. This means that ideal waste-form phases usually have relatively complex structure types with a number of different coordination polyhedra of various sizes and shapes and with multiple substitutional schemes to allow for charge balance with radionuclide substitutions.

Radionuclides may occupy specific atomic positions in the periodic structures of constituent crystalline phases, which are as a dilute solid solution. The coordination polyhedra in each phase impose specific size, charge and bonding constraints on the nuclides that can be incorporated into the structure. This means that ideal waste form phases usually have relatively complex structure types with a number of different coordination polyhedra of various sizes and shapes and with multiple substitutional schemes to allow for charge balance with radionuclide substitutions[11].

The aim of this work was to study the radiation damages on the glass and glass-ceramic due to decay of vitrification real waste, this is done by irradiation samples of glass and glass-ceramics contain Srwhich has the same chemical properties of the radioactive Sr by beta ray with different doses(0, 480Gy, 14400 Gy,28800Gy and 43200 Gy).Then study the effect of beta rays dose on the physical properties (density), chemical properties (leaching rate) and mechanical properties (microhardness and surface strength).

Chemical durability is an important technical performance property of waste forms in groundwater environments. Leaching provides a physical measure of how well waste forms can retain radionuclides if exposed to water in a repository setting. While thermodynamics can give the equilibrium states, kinetic information is needed to understand rates of leaching, especially in open systems. An advantage of waste forms based on mineral analogue phases is that the mineral phases can be shown to have survived for several hundred million years or more in wet, thermal geologic environments.

Self-radiation from radionuclide decay can affect microstructural evolution, phase stability, and thermodynamic properties in waste forms. The principal sources of radiation in HLW are β-decay of the fission products (e.g., ^{137}Cs and ^{90}Sr). Beta-decay produces energetic β-particles (~0.5 MeV), low energy recoil nuclei, and γ-rays; whereas, α-decay produces energetic α-particles (4.5 to 5.5 MeV), energetic recoil nuclei (70 to 100 keV), and some γ-rays. These particles and γ-rays interact with solids primarily through energy transfers to electrons via ionization processes, producing electron-hole pairs, or to atomic nuclei via elastic collisions, displacing atoms to produce defects[2]. In general, β-decay of the short-lived fission products is the primary source of radiation (and heat generation) from HLW during the first 600 years of storage. Due to the danger of using a real radioactive nuclides in research laboratories and not to expose students and teaching staff to a different dose of radiation, in this research stable nuclides act as radionuclides were used and studying the effect of beta particles on glazing waste in glass.

EXPERIMENTAL SECTION

In this work glass and glass-ceramic materials were prepared as fallowing ways:

Glass preparation

The borosilicate glass c-type (Celsian) prepared from the list oxides insert in Table 1. Also this Table contains the weight percentage. The methods of preparing glass based are list as follows:

1-Intialize the constituent oxide glass that insert in Table 1 or replace the oxide with its carbonate or nitrate by using the molecular weights with calculated ratios to ensure the required percentage of oxide.

2-Mix and crushed the mixture at once by using electric mixer contains Teflon balls for 24 hours to get smooth and soften for crushed oxides.

3- Make annealing process (prepare the crucible) that made from alumina inside the oven at the temperature 600℃ for one hour and then leave to cooling inside the oven gradually.

Table 1: Component of borosilicate glass C-type without waste

Components	Alternative	Weight percentage %
SiO_2		60
AlO_3		2
B_2O_3		2
CaO		10
Na_2O	Na_2CO_3	10
Li_2O	Li_2CO_3	10
TiO_2		1
ZrO_2		1
ZnO		1
MgO		1
BaO		1

4-Put the above mixture oxides with SrO (5%) inside crucible from alumina inside the furnace at temperature 100 to 900 for 3 hour then to 1300℃ for 2hours, and shaking the molting inside the crucible to release the CO_2 babbles for more homogeny. and left the mix inside the furnace cooling.

Glass-ceramics preparation
The glass-ceramics preparation process consists of two stages of thermal treatment. The first stage from heat treatment is done to have high degree of nucleation and the second stage to have maximum crystallization.

The heat treatment to convert glass c-type to glass-ceramics which consist of heat the samples from 25℃ to 630℃ for three hours, which present the nucleation stage, followed by heating up to 830℃ for five hours to achieve maximum crystallization. To ensure the convert glass to glass-ceramics is x-ray used to check diffraction the result shows crystalline tops in diffraction pattern that indicates to generating crystalline phase through heat treatment.

Leaching rate
The leaching rates for many glass and ceramic phases have been investigated as a function of time as fallowing way. Leaching rate was measured for samples of glass and glass-ceramics prepared under same conditions to study their chemical durability and its ability to store waste and to compare between amount of ions (strontium ions) leaching from both, when they immersed in the water. And to study the effect of gamma radiation on the amount of leaching strontium ions (nuclear waste) and effect of keep time of samples in the distilled water

The material of container was used for this test from stainless steel as a cylindrical shape open in one side and the solution (leaching) for immersion sample is distilled water to (1day,1month,2months, 3months).The percentage between solution volumes that around sample to its surface area must be not over10 cm so this value was used i.e. $\frac{V}{S.A} = 10\ cm.$

Then analysis percentage strontium ion for every part of million (ppm) by use Atomic Absorption device Varian type F-S 240

Microhardness Hardness is the property of a material that enables it to resist plastic deformation, usually by penetration After ending glass and glass-ceramic preparation process, microhardness measured by Vickers method used Digital Micro Vickers Hardness Tester TH714 between the tip The mold putted which has the samples on the rotate board under indenter with distance of the indenter. The suitable weight (load) used to make indentation, in this test was used load 50 pound for glass sample and 20 pound for glass-ceramics samples and load time was 5 second for all sample

The surface strength
The strength of material is the ability of that material to with stand an applied stress.After ending preparation process the mold which has two types of samples(glass and glass-ceramics) is tested by digital surface strength tester, where the mold is putting under probe made of stainless steel and pass on the surface of the samples this gave the value of surface strength for samples directly in digital way by method Center Line Average (CLA).

The physical properties (density measurement) for glass and glass-ceramics
The densities of glass and glass-ceramics immobilize radionuclides waste is measured by Archimedes Method

$$\rho = m/\mathrm{v}$$

RESULTS AND DISCUSSION

Table 2 represents the surface strength for glass and glass-ceramic sample before and after exposure to different doses beta ray.

Table2:The surface strength for glass and glass-ceramic samples before and after exposure to beta ray

Dose (Gy)	The surface strength for glass	The surface for glass-ceramic
0	0.3	1.8
480	0.3	1.8
14400	0.33	1.8
28800	0.35	1.8
43200	0.4	1.8

The changing on the surface strength of glass is little and no changing on the glass- ceramics after exposure to beta ray.

Table 3 represents the microhardness of glass and glass –ceramic sample before and after exposure to different doses of beta ray.

Table 3:The microhardness of glass and glass –ceramic sample before and after exposure to beta ray

Dose(Gy)	Microhardness for glass	Microhardnessfor glass-ceramic
0	56	88
480	60	88
14400	64	88
28800	70	88
43200	74	88

The hardness (resistance to scratching or cutting) is increased in glass after irradiation and remains constant after irradiation.

Table 4represents the leaching rate of strontium ions after exposure samples to different doses of beta ray

Table 4:Leaching rate of strontium ions after exposure samples to beta ray

Dose(Gy)	Leaching rate % for glass	Leaching rate % for glass-ceramic
480	0.16	0.16
14400	0.16	0.16
28800	0.16	0.16
43200	0.16	0.16

Table 5 represents the effect of different doses of beta rays on the density of the glass and glass- ceramic.

Table 5:The density of the glass and glass- ceramic after irradiation with beta ray

Dose(Gy)	Density(g/cm^3)for glass	Density (g/cm^3)for glass-ceramic
480	3.203	3.22
14400	3.203	3.22
28800	3.203	3.22
43200	3.203	3.22

It was found that the there is no effect of beta radiation on physical properties (density) for glass and glass-ceramic; i.e its values remain constant after irradiation with different beta doses.

The leaching rate for all samples were smaller or equal than 0.16 (where 0.16 is a sensitive of atomic absorption devise) it means that there is no leaching of Sr from glass host due to radiation. In addition, the leach rate itself can depend on SA/V, particularly at high SA/V where the concentration of leached elements can build up in solution. The self diffusion rates of radioisotopes in the waste form can also affect elemental leach rates by changing the local surface concentration exposed to water.

Damage will occur only after damage zones overlap and provide interconnected fluid and/or solid diffusion channel ways between the interiors of the samples and the surface. If individual damage zones anneal in relatively short times, the damage zones will not overlap and significant increases in leach rates will not be realized. In laboratory experiments, dose rates may be increased several orders of magnitude above levels that are pertinent to actual waste to accelerate glass damage processes into reasonably short time periods.

In some advanced closed fuel cycle concepts, the separation of Sr into a separate waste stream provides an opportunity to immobilize these high heat-generating radioisotopes into waste forms for interim storage over several hundred years this was effect on the surface strength and microhardness of glass.

At these high ionization doses and temperatures, many materials undergo decomposition, phase separation, and bubble formation under electron beam (beta particles) irradiation on laboratory time scales. Electron beam interactions are similar to those of beta particles and the fast electrons produced by gamma interactions, but their dose rates are significantly higher than the 10^4 to 10^5Gy/h expected in Sr waste forms. Developing predictive models and validating such an approach will require a more fundamental understanding of ionization effects and the coupling of electronic excitations to atomic dynamics than currently exists.

CONCLUSION

It was found from results that the physical properties and the leaching was not affected by beta-ray, while the values of surface strength and microhardness of glass increased with increasing radiation doses and their values have remained constant for the glass-ceramics. Therefore waste radionuclides can stored in glass-ceramic for long time but not glass phase.

REFERENCES

[1] M.E. Burns. Low-level radioactive waste regulations: science, politics and fear. Chelsea, Mich.: Lewis Publishers, **1988**.
[2] W.J. Gray, *Nature*,**1982**, 296, 547.
[3] E.R. Vance, R. Roy, J.G. Pepin, D.K. Agrawal, *J. Mater. Sci.***1982**,17, 947.
[4] E. Vernaz, A. Loida, G. Malow, J.A.C. Marples, Hj. Matzke, in *Proc. 3rd EC Conf. On Radioactive Waste Management and Disposal*, L. Cecille, ed. (Elsevier, London, **1991**), p. 302.
[5] D.M. Strachan, R.D. Scheele, E.C. Buck, J.P. Icenhower. A.E. Kozelisky, R.L. Sell, R.J. Elovich, W.C. Buchmiller, *J. Nucl. Mater.***2005**,345, 109.
[6]S.V. Stefanovsky, A.N. Lukinykh, S.V. Tomilin, A.A. Lizin, S.V. Yudintsev, in *Scientific Basis for Nuclear Waste Management XXXI*, W.E. Lee, J.W. Roberts, N.C. Hyatt, R.W. Grimes, eds (MRS Symp. Proc. **1107**, Warrendale, PA, **2008**).
[7] G. Malow, J.A.C. Marples, C. Sombret, in *Radioactive Waste Management and Disposal*, R. Simon and S. Orlowski, eds. (Harwood Academic Publishers, Chur, Switzerland, **1980**), p. 341.
[8] R.P. Turcotte, J.W. Wald, F.P. Roberts, J.M. Rusin, W. Lutze, *J. Am. Ceram.***1982**, 65, 589.
[9] I. Mueller, W.J. Weber, *MRS Bull*,**2001**,26, 698.
[10] G.J. McCarthy, *Nucl. Technol.***1977**, 32, 92
[11] R. C. Ewing, W. J. Webert and F. W. Clinard, Jrf "Radiation effects in nuclear waste forms for high-level radioactive waste", Progress in Nuclear Energy, **1995**,29**(**2) 63-121.
[12] W.G. Burns, A.E. Hughes, J.A.C. Marples, R.S. Nelson and A.M. Stoneham, *Nuclear Materials*, **1982**,107, 245-270.
[13]D.E. Clark, Pantano, C.G., and Hench, L.L., Corrosion of Glass, Books forIndustry, New York, **1979**.