Australian Journal of Basic and Applied Sciences, 11(7) May 2017, Pages: 23-28

AUSTRALIAN JOURNAL OF BASIC AND APPLIED SCIENCES

ISSN:1991-8178 EISSN: 2309-8414
Journal home page: www.ajbasweb.com

Neutron radiation effects on the nuclear waste (Ce) vitrification in glass and glass-ceramics

[1]Muna A. Saeed,[2]Jinan F. Mahdi, [3]Asia H.Al-Mashhadani,[4]Aliaa A. Abdeljabbar

[1]Lecturer in Physics Department, Department of Physics, College of Science, University of Baghdad, Baghdad, Iraq.
[2]Associate Professor in Medical Instruments Department, Department of Medical Instruments, College of Engineering, Electrical and Electronic Engineering Techniques, Middle Technical University, Baghdad, Iraq.
[3] Professor in Physics Department, Department of Physics, College of Science, University of Baghdad, Baghdad, Iraq.
[4]Ph.D. in Physics Department, Department of Physics, College of Science, University of Baghdad, Baghdad, Iraq.

Address For Correspondence:
Asia H.Al-Mashhadani, University of Baghdad, Department of Physics, College of Science.

ARTICLE INFO

Article history:
Received 18 February 2017
Accepted 5 May 2017
Available online 10 May 2017

Keywords:
Vitrification, Nuclear waste, Neutron, Glass

ABSTRACT

Background: During the coming decades, one of the largest and most expensive challenges facing the world is the stability and immobilization of the high-level nuclear waste (HLW) in solid form. Include nuclear waste in Iraq volumes, high total activity very diverse and complex compositions. This is due to the emergence of a variety of sources increasingly from nuclear waste, such as spent nuclear fuel from the ancient Iraqi Atomic Energy Organization at the site of Al-Twitha, waste of radioactive isotopes used in medicine and waste from nuclear weapons used in the war against Iraq in 2003. This waste need to be stored until it is safety. **Objective:** Therefore in this study it was stored nuclear waste (oxides of cerium) through the vitrification method in the two types of borosilicate glass (glass, glass-ceramics) and the effect of neutron radiation on waste immobilized been done. Glass has emerged as favorite to immobilize a wide range of radioactive materials, potentially hazardous radioactive and nonradioactive materials. Then the mechanical properties, physical and chemical of the glass and borosilicate containing cerium oxide before and after irradiated by neutron irradiation were studied. **Conclusion:** It found that the physical properties and the leaching rate were not affected, while the values of surface strength and micro hardness of glass increased with increasing radiation doses and their values have remained constant for the glass-ceramics

INTRODUCTION

The existence of technology to manage all types of radioactive waste, and to demonstrate the safety of facilities and activities predisposal and disposal has progressed significantly. A comprehensive set of the International Atomic Energy Agency safety standards to support the safe development programs radioactive waste management have been published, and developed a set of technical reports to provide information on best practices applied by Member States to comply with the requirements and guidelines contained in the safety standards. In parallel, the review process under the Joint Convention on the Safety of Spent Fuel Management and the Safety of Radioactive Waste Management, which has gained in maturity, thus contributing to enhance safety in this area (James and Audeen, 2002; Ojovan *et al*, 2005). Nuclear waste need to store for a long time until the (safety became) the impact of the loss of radiation. Therefore must found suitable method to immobilize nuclear waste to prevent leaching of ions surrounding the way and become a risk to the environment and our health (IAEA, 2016; Burns. 1988; Gray, 1982).

There was so much more research and technology development in the field of glass waste forms than any other waste form over the past 50 years. This is because the structure of amorphous and disordered relatively

ToCite ThisArticle: Muna A. Saeed, Jinan F. Mahdi, Asia H.Al-Mashhadani, Aliaa A. Abdeljabbar., Neutron radiation effects on the nuclear waste (Ce) vitrification in glass and glass-ceramics. ***Aust. J. Basic & Appl. Sci.,*** *11(7): 23-28, 2017*

can be incorporated into a wide variety of chemical elements. These structures produces highly durable glasses (Roy *et al*, 1982; Loida *et al*, 1991; Strachan *et al*, 2005; Stefanovsky *et al*, 2008).Most elements play one of three basic roles in glass structures: network formers, network modifiers and intermediates. In borosilicate glass structures, mainly the formation of a network of chains and borate polyhedra silicate, lithium, sodium, calcium and rates of typical network that creates an oxygen bridge or provide balance charge for some elements HLW (Malow *et al*, 1980; Turcotte *et al*, 1982). A commonality exists between the many different radioactive waste glass systemsand 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(Mueller and Weber, 2001).Borosilicate glasses have some flaws, especially away from the gas necessary to deal with the volatility in the HLW glass melting temperatures and low melting temperature systems in a glass of some of the important radionuclides, such as Tc and actinides. With a strong premium on warehouse space and caching all over the world, and higher shipments of waste are subject to investigation by the forms of crystalline ceramic waste are useful for specific streams of waste. In addition, mineral analogues of many forms of ceramic waste provide evidence of long term durability (McCarthy, 1977).

In crystalline ceramic phases, can be incorporated radionuclides to fill specific atomic positions in the periodic crystalline structures of the constituent phases, which allows for high load of specific radionuclides. The coordination polyhedra at every stage imposes certain size, charge, and restrictions on radionuclides that can be incorporated in the structure Association. This means that the ideals of the waste in the form of higher stages and types usually have a relatively complex structure with a number of different polyhedra of different sizes and shapes and in coordination with various alternative plans to allow the administrator to achieve a balance with the alternatives of radionuclides.

Radionuclide's may occupy specific atomic positions in the periodic structures of constituent crystalline phases, which are as a dilute solid solution. The coordination polyhedral 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 polyhedral of various sizes and shapes and with multiple substitutional schemes to allow for charge balance with radionuclide substitutions (Ewing *et al*,1982).

The aim of this study was to investigation the radiation damage result from decay of vitrification real waste, by irradiation samples from glass and glass-ceramics with different doses of neutron (4.4 mrad/h),then studying the effect of neutron dose on the physical properties (density), chemical properties (leaching rate) and mechanical properties (micro hardness and surface strength).

Chemical durability is an important performance characteristic artistic forms of waste in groundwater environments. Leaching provides a physical measure how lost as well as the forms can be retained in the radionuclides if exposed to water in the vicinity of the repository. While thermodynamics can give a balance, there is a need to kinetic information to understand the leaching rates, particularly in open systems. Advantage of the waste on the basis of mineral phases analog is that the mineral phases can be shown to have survived for several hundred million years or more in the humid geological and thermal environments.

Since neutrons without charge, interact almost exclusively with atomic nuclei mainly produces no direct ionization. Often cause the products of these reactions ionization, and thus lead to chemical reactions traditional radiation. Principle ions produced are protons and heavier positive ions, and chemical reactions similar to those resulting from exposure to radiation and heavy positive particles. Because of the large penetration, and the results of neutron radiation is not limited to the surface layer of the radioactive substrate. The principle include interactions of neutrons with the issue of fragmentation of elastic, inelastic scattering, nuclear reaction, and was arrested. And it can be used for neutron radiation in three ways (Majdi and Fadhel, 2017). Fast neutron dissipate their energy by elastic collisions resulting in numerous atomic displacements and the sample become moderately radioactive .neutron irradiation damage all of the phase in the waste form ,and therefore ,will not simulate the phase selective α-decay damage experienced in actual waste forms.

Neutron irradiation have been used to simulate damage in synroce phase (Reeve and Woolfrey,1980**)**. boron contain glass (borosilicate glass) or boron doped waste forms can be irradiated in thermal neutron flux and because of high cross section and E=1.7Mev is generated with this technique High rate of He formation are possible, but this does not simulate the α-recoil damage of the α-decay event.

Finally thermal neutron capture by selected nuclide (U^{235}) can lead to nuclear fission which resulted in the formation of extensive zone of atomic displacement, fission tracks (1979).this is not good simulation of α-decay damage and the number of fission event in nuclear waste forms is so low as to make the process unimportant damage mechanism (Majdi and Fadhel, 2017) .

Experimental work:

In this work the glass and glass-ceramics samples prepared as two fallowing means

Preparation the borosilicate glass c-type from the list oxides using weight percentage insert in Table 1. The methods of preparing glass based are list as follows:

1- Initialization constituent glass oxide inclusion in Table 1 or replace dioxide with carbonate, nitrate or using molecular weights with calculated ratios to ensure the required proportion of oxide.

2-Mix and crush the mixture at once using an electric mixer containing Teflon balls for 24 hours to get a smooth and soften the oxides crushed.

3. Make the hard process (preparation crucible) that made the alumina into the oven at a temperature of 600 ℃ for one hour and then left to cool inside the oven graduallyas shown in Fig.1.(Kadhim 1986).

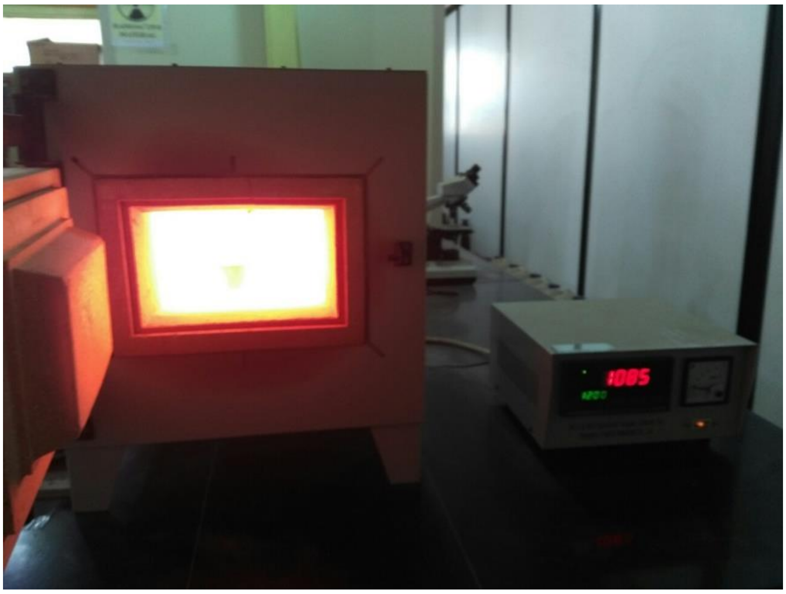

Fig.1: Oven used to prepare glass material

Table 1: Borosilicate glass components

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 Ce(0.5) 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, the prepared glass samples were shown in Fig.2.

Fig.2: Samples of prepared glass and glass-ceramic

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 650℃ for three hours, which present the nucleation stage, followed by heating up to 800℃ 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, the prepared glass-ceramic samples were shown in Fig.2.

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 (cerium ions) leaching from both, when they immersed in the water. And to study the effect of neutron radiation on the amount of leaching cerium 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 (1-day,1-month, 2-months and 3-months). The percentage between solution volumes that around sample to its surface area must be not over10 cm so this value was used i.e. $\frac{V}{S.A} = 10\ cm.$ (IAEA)

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 the material which enables it to resist plastic deformation, usually by penetration after the termination of the glass and the process of preparation, glass-ceramics, and micro hardness measured way Vickers used Digital Micro Vickers Hardness Tester TH714 between the tips. 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.

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).

Physical properties (Density measurement) for glass and glass-ceramics:

The densities of glass and glass-ceramics immobilize radionuclide's waste is measured by Archimedes Method

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

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 sample before and after exposure to fast neutron source

Neutron Dose (rad)	The surface strength of glass(kg/mm^2)	The surface strength of glass-ceramic(kg/mm^2)
0	0.3	2.1
0.7	0.32	2.1
1.4	0.32	2.1

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

Table 3 represents the micro hardness of glass and glass –ceramic sample before and after exposure to different doses of neutron source.

Table 3: The micro hardness of glass and glass –ceramic sample before and after exposure to different doses of fast neutron source.

Neutron Dose (rad)	The micro hardness of glass (kg/mm^2)	The micro hardness of glass-ceramic (kg/mm^2)
0	60	95
0.7	65	94.12
1.4	65	94.0

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

Table 4 represents the leaching rate of strontium ions after exposure samples to different doses of neutron source

Table 4: Leaching rate of strontium ions after exposure samples to neutron source

Time	Leaching rate for glass	Leaching rate for glass-ceramic
After 1-week	0.22	0.22
After 1-month	0.22	0.22
After 3-month	0.22	0.22

Table 5 represents the effect of different doses of neutron source on the density of the glass and glass-ceramic.

Table 5: The density of the glass and glass- ceramic after irradiation byfast neutron source

Neutron Dose (rad)	Density(g/cm^3)for glass	Density(g/cm^3)for glass –ceramic
0	2.1	2.1
0.7	2.1	2.1
1.4	2.1	2.1

It was found that the there is no effect of neutron source 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 0.21 (where 0.21 is a sensitive of atomic absorption devise) it means that there is no leaching of Ce 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 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 Ce 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 micro hardness of glass.

At these high ionization doses and temperatures, many materials undergo decomposition, phase separation, and bubble formation under neutron particles) irradiation on laboratory time scales. Neutron irradiation have been used to simulate damage in synroc phase. Boron contains glass(borosilicate glass)or boron doped waste forms can be irradiated in thermal neutron flux, and because of high cross section and E=1.7Mev is generated with this technique. High rate of He formation are possible, but this dose not simulate the α-recoil damage of the α-decay event.

Conclusions:

The results show that the physical properties and the leaching rate was not affected by the source of the neutron, while the surface hardness strength and micro hardness of glass increased with increasing radiation doses and their values have remained constant for the glass-ceramics. Therefore waste radionuclide's can stored in glass-ceramic for long time but not glass phase.

REFERENCES

Burns, M.E.,1988. Low-level radioactive waste regulations: science, politics and fear. Chelsea, Mich.: Lewis Publishers.

Ewing,R.C., W.J. Webert and F.W. Clinard, 1982."Radiation effects in nuclear waste forms for high-level radioactive waste", Progress in Nuclear Energy.Gray, W.J. 1982, Nature296, 547.

IAEA, 2016. International Conference on the Safety of Radioactive Waste Management Vienna, Austria, 21-25.

James, H. Saling and Audeen W. Fentiman, 2002 “Radioactive waste Management”, Taylor and Francis, New York and London, 2nd ed

Kadhim A. Hubeatir, 1986, M.Sc. Thesis, Applied Physics, University of Technology.

Kays, S. Majdi and Heidar J. Fadhel, 2017.Journal of Radiation Research and Applied Sciences, 10(1): 53-56.

Malow,G., J.A.C. Marples and C. Sombret, 1980, in Radioactive Waste Management and Disposal, R. Simon and S. Orlowski, eds. (Harwood Academic Publishers, Chur, Switzerland), p: 341.

McCarthy, G.J., 1977.Nucl. Technol., 32, 92.

Michael, I Ojovan, William E Lee and William E. Lee, 2005. “An Introduction to Nuclear Waste Immobilization”, Elsevier.

Mueller, I. and W.J. Weber, 2001, MRS Bull., 26, 698.

Reeve, K.D. and J.L. Woolfrey, 1980.Aust. Ceram. Soc., 16:10-15.

Stefanovsky,S.V., A.N. Lukinykh, S.V. Tomilin, A.A. Lizin and S.V. Yudintsev, 2008. 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).

Strachan, D.M., R.D. Scheele, E.C. Buck, J.P. Icenhower. A.E. Kozelisky, R.L. Sell, R.J. Elovich and W.C. Buchmiller, 2005.J. Nucl. Mater., 345, 109.

Turcotte,R.P., J.W. Wald, F.P. Roberts, J.M. Rusin and W. Lutze, 1982 J. Am. Ceram., 65: 589.

Vance,E.R., R. Roy, J.G. Pepin and D.K. Agrawal, 1982.J. Mater. Sci., 17: 947.

Vernaz,E., A. Loida, G. Malow, J.A.C. Marples and Hj. 1991.Matzke, in Proc. 3rd EC Conf. On Radioactive Waste Management and Disposal, L. Cecille, ed. (Elsevier, London, pp: 302.