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Effect of Neutron Irradiation on Cerium Oxides Immobilized by Phosphate Glass

[1]Alyaa A. Abdaljabar, [2]Nagham Yassin and [1]Abdul Hussain K. Elttayef
[1]Ministry of Science and Technology, Baghdad, Iraq
[2]Department of Medical Instrumentation, Middle Technical University, Baghdad, Iraq

Abstract: 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 wastes need for storage until becoming safety. Therefore, in this study nuclear waste (cerium oxides) were stored by vitrification methods in two types of phosphate glass (glass and glass-ceramics) and the effect of neutron on the immobilized waste was done. Then the physical, chemical and mechanical properties phosphate glass contain cerium oxide before and after irradiated were investigated. It found that the physical properties and the leaching was not affected by irradiation while the values of surface strength and micro hardness of glass increased with increasing radiation doses and their values have remained constant in the glass-ceramics.

Key words: Neutron irradiation, phosphate glass, Al-Twitha site-Iraq, radiation, hardness, values

INTRODUCTION

The nuclear waste needs to store for a long time until loss the radiation effect (became safety). Therefore, must find a suitable method to immobilize nuclear waste to prevent leaching ions to surrounding and became hazards threatens environment and our health (Burns, 1988).

There has been considerably more research and technology development conducted on glass waste forms than any other waste from over the past 50 years. This is because their amorphous and relatively disordered structures can incorporate a wide range of chemical elements. Glass compositions that include 30-40 different elements are used routinely for High Level Waste (HLW). Such compositions produce highly durable glasses (Lee *et al*., 2008; Gray, 1982). Most elements play one of the three basic roles in glass structures: network formers, network modifiers and intermediates. In glass structures, the network is primarily formed by the chains of borate and phosphate, sodium and calcium are typical network modifiers that create non-bridging oxygen's or provide charge balance of some HLW elements (Simon, 1980; Turcotte *et al*., 1982). 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 60-85 weight% of the glass structure network modifiers make up 0-25% of the glass while 15-40 weight% are intermediates (Muller and Weber, 2001).

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 the glass of some important radionuclide's 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 (McCarthy, 1977).

In crystalline ceramic phases, radionuclide's 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 binding constraints on the radionuclide's that can be incorporated into the structure. This means that ideal waste formative phases usually have relatively complex structure types with a number of different coordination polyhedra of various sizes and shapes and with multiple substitution 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 binding 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

Corresponding Author: Alyaa A. Abdaljabar, Ministry of Science and Technology, Baghdad, Iraq

substitution schemes to allow for charge balance with radionuclide substitutions (Ewing *et al*., 1995). The aim of this study was to representation radiation damage result from decay of vitrification real waste, irradiation samples from glass and glass-ceramics with different time (1 day, 1-3 months) to study the effect on value of 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 100 million years or more in wet, thermal geologic environments. 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 on glazing waste in glass.

MATERIALS AND METHODS

In this research, phosphate glass and glass-ceramic materials were prepared as following ways.

Phosphate glass preparation: The glass prepared from the list oxides insert in Table 1. Also, this table contains the weight percentage oxides. The methods of preparing glass based are listed as follows.

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.

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

Make the annealing process (prepare the crucible) that made from alumina inside the oven at the temperature 600°C for 1 h and then leave to cool inside the oven gradually.

Put the above mixed oxides with Ce (5%) inside crucible from alumina inside the furnace at temperature 100-900 for 3 h, then to 1300°C for 2 h and shaking the molting inside the crucible to release the CO_2 bubbles for more homogenous. And left the mix inside the furnace cooling.

Table 1: Component of phosphate glass without waste (cerium ions)

Components	Alternative	Weight (%)
P_2O_3		60
CaO		10
Na_2O	Na_2CO_3	10
Li_2O	Li_2CO_3	10
TiO_2		5

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

The heat treatment to convert glass to glass-ceramics which consist of heat the samples from 25-600°C for 2 h which present the nucleation stage, followed by heating up to 100°C for 3 h to achieve maximum crystallization. To ensure the convert glass to glass-ceramics are result shows crystalline tops in diffraction pattern that indicates to generate crystalline phase through heat treatment.

The leaching rates for many glass and ceramic phases have been investigated as a function of time as follows way Leaching rate was measured in samples of glass and glass-ceramics prepared under the same conditions to study their chemical durability and its ability to store waste and to compare between the 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 at one side and the solution (leaching) for immersion sample is distilled water to (1 day, 1-3 months). The percentage between solution volumes that around the sample to its surface area must be not over 10 cm, so, this value was used, i.e., V/..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 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 indenture with distance of the indenture. 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 sec for all sample.

Surface strength: The strength of material is the ability of that material to withstand 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 put under probe made of stainless steel and pass on the surface of the samples this gave the directly in value of surface strength for samples digital way by method center line.

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.

$$\rho = m/v$$

RESULTS AND DISCUSSION

Table 2 represents the surface strength for glass and glass-ceramic sample before and after exposure to different neutron doses. The changing on the surface strength of glass is little increased in the glass and constant on glass-ceramic after exposure to neutron source. Table 3 represents the microhardness of glass and glass-ceramic sample before and after exposure to different doses. The hardness (resistance to scratching or cutting) is increased in glass after irradiation and constant after irradiation. Table 4 represents the effect of different doses on the density of the glass and glass-ceramic. It was found that the there is no effect on physical properties (density) for glass and glass-ceramic, i.e., its values remain constant to glass and increased in glass ceramic after irradiation with different doses. Table 5 represents the leaching rate of cerium ions after exposure samples to different doses of neutron source.

The leaching rate for all samples were smaller or equal than 0.22 (where 0.22 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 increased several orders of magnitude above levels that are pertinent to the actual waste to accelerate glass damage processes into reasonably short time periods.

Table 2: The surface strength for glass and glass-ceramic sample before and after exposure to neutron irradiation

Dose (Gy)	Glass	Glass-ceramic
0	0.50	3.1
203	0.50	3.1
427	0.57	3.1

Table 3: The microhardness of glass and glass-ceramic sample before and after exposure to fast neutron

Dose (Gy)	Glass	Glass-ceramic
0	59	97
203	59	97
427	64	97

Table 4: The density of the glass and glass-ceramic after irradiation

Dose (mrad)	Density (g/cm^3) for glass	Density (g/cm^3) for glass-ceramic
1 day	4.003	4.21
1 month	4.003	4.21
2 months	4.003	4.21
3 months	4.003	4.21

Table 5: Leaching rate after exposure samples to neutron

Dose (mrad)	Leaching rate for glass	Leaching rate for glass-ceramic
After 1 day	0.14	0.22
After 1 month	0.14	0.22
After 2 months	0.14	0.22
After 3 months	0.14	0.22

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 adioisotopes into waste forms for interim storage over several 100 years this was affected on the surface strength and microhardness of glass.

CONCLUSION

It was found from the results that the and the leaching was not affected by neutron irradiation (that's means not ions seep to surround). The physical properties (density) was found that the there is no effect for glass and glass-ceramic. Its values remain constant to glass and increased in glass ceramic after irradiation with different doses. The values of surface strength and micro hardness of phosphate glass increasing with radiation doses and their values constant for the glass-ceramics. Therefore, waste radionuclide's can be stored in glass-ceramic (phosphate glass) with safe environment for a momentary time.

REFERENCES

Burns, M.E., 1988. Low-Level Radioactive Waste Regulations: Science, Politics and Fear. Lewis Publishers, Chelsea, Michigan,.

Ewing, R.C., W.J. Weber and F.W. Clinard Jr., 1995. Radiation effects in nuclear waste forms for high-level radioactive waste. Progress Nucl. Energy, 29: 63-127.

Gray, W.J., 1982. Fission product transmutation effects on high-level radioactive waste forms. Nature, 296: 547-549.

Lee, W.E., J.W. Roberts, N.C. Hyatt and R.W. Grimes, 2008. Scientific Basis for Nuclear Waste Management XXXI. Materials Research Society, Pittsburgh, Pennsylvania, Pages: 709.

McCarthy, G.J., 1977. High-level waste ceramics: Materials considerations, process simulation and product characterization. Nucl. Technol., 32: 92-105.

Muller, I. and W.J. Weber, 2001. Plutonium in crystalline ceramics and glasses. MRS. Bull., 26: 698-706.

Simon, R., 1980. Radioactive Waste Management and Disposal. Harwood Academic Publishers, Chur, Switzerland, ISBN:9783718600564, Pages: 693.

Turcotte, R.P., J.W. Wald, F.P. Roberts, J.M. Rusin and W. Lutze, 1982. Radiation damage in nuclear waste ceramics. J. Am. Ceram. Soc., 65: 589-593.