Beta Ray and Neutrons Effect on Leaching Rate of Sr and Ce Doped in Glass and Glass-Ceramic

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Abstract

Nuclear waste in Iraq has three major sources; spent nuclear fuel from the old Iraqi Atomic Energy Organization in Al-Twatha 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 (SrO) and cerium (Ce)) were stored by vitrification methods in two types of borosilicate glass (glass and glass-ceramics) and the effect of beta ray on the immobilized waste was done. Then the leaching rate of strontium oxide and cerium in borosilicate glass before and after irradiated by beta ray and neutron flux were investigated. It found that the leaching was not affected by beta-ray and neutrons.

Keywords: Beta-ray; Neutrons; Glass; Glass-ceramics; Nuclear waste and Leaching

Introduction

Beta and neutrons decay from radionuclide waste affect materials through the interactions of the beta particles, neutrons, and recoil nuclei, with the materials that used to store the nuclear waste [1]. These interactions fall into two broad categories: the transfer of energy to electrons (ionization and electronic excitations) and the transfer of energy to atomic nuclei, primarily by ballistic processes involving elastic (billiard-ball-like) collisions. The partitioning of the energy transferred into electronic excitations and into elastic nuclear collisions is an important process controlling the effects of radiation [2]. For β -particles, the energy transfer is dominated by ionization processes. For recoil nuclei and neutrons, interactions involve ionization processes and elastic collisions with atomic nuclei.

Atomic Displacement (Damage) Atomic displacement can occur ballistically through kinetic energy transfer, or radiolytically by the conversion of radiation-induced excitation into atom motion (i.e., recoil). As a charged particle passes through matter, the particle energy dissipates by exciting orbital electrons and by elastic collisions with the material nuclei. An elastic collision can reject an atom from its normal lattice position. The ejected atom is known as a primary knock-on, which, in turn, may cause a cascade of atomic displacements before eventually coming to rest. The displaced atom becomes an interstitial, and

the position the atom formerly occupied becomes a vacancy. Together the interstitial and vacancy are referred to as a Frenkel pair. Some displaced atoms can lead to secondary displacements. For example, the displaced atom may collide with and replace another atom in the material. Displacement damage is the result of nuclear interactions, typically scattering, which cause lattice defects. Displacement damage is due to cumulative long-term non-ionizing damage from the ionizing radiations. The collision between an incoming particle and a lattice atom subsequently displaces the atom from its original lattice position. The particles producing displacement damage include protons of all energies, electrons with energies above 150 keV, and neutrons.

Ionization is the process of removing or adding an electron to a neutral atom, thereby creating an ion. The term is also often used in connection with the removal of an electron from a partially ionized atom. A closely related process is excitation, in which the energy level of an electron is raised; however, excitation occurs at an energy less than that required for ionization. The radiations which are charged (α , β , p) can directly ionize matter; however, those radiations which are neutral (n , γ) are said to indirectly cause ionization. Ionizing radiation tends to be increasingly damaging in the following order of molecular formation (largely due to the ability of ionization to disrupt

the bonds): 1. metallic bond (least damaged) 2. ionic bond 3. covalent bond (most damaged) [3].

The effects of radiation in nuclear waste were immobilized in glasses are complex, and the fundamental understanding of the radiation damage processes and results is limited. The high-radiation environment provided by the β -decay of radionuclide in nuclear waste and neutrons can affect the chemical, physical and mechanical properties which occur due to over relative long periods of storage, and at high temperatures dependents on waste loading, age of waste, depth of storage. Thus, a major challenge is to effectively simulate high-dose radiation effects that will occur at relatively low-dose rate over long periods of time [4,5].

Prediction of the long-term leach rates is important to aid in evaluating the safety because the radio nuclides are most likely to be released by leaching through groundwater, and because hazard of the radio nuclides lasts very long up to about 10^5 - 10^6 yr [6,7].

In this research the results of the leaching rate of glass and glass-ceramic with Sr and Ce before and after irradiation by beta rays and neutrons are presented.

Experimental Work

Glass and glass-ceramic materials were prepared as following 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:

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

Components	Alternative	Weight Percentage %
SiO ₂		60
AlO ₃		2
B ₂ O ₃		2
CaO		10
Na ₂ O	Na ₂ CO ₃	10
Li ₂ O	Li ₂ CO ₃	10
TiO ₂		1
ZrO ₂		1
ZnO		1
MgO		1
BaO		1

I. 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.

II. 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.

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

IV. Put the above mixture oxides with SrO (5%) inside crucible from alumina inside the furnace at temperature 100 to 900 for 3 hours then to 1300°C for 2hours, and shaking the molting inside the crucible to release the CO₂ 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°C to 630°C for three hours, which present the nucleation stage, followed by heating up to 830°C 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 same procedure was used for immobilized Ce in glass and glass-ceramic.

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 (1-day,1-month,2-months-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.

$$V/(S.A)=10\text{cm.}$$

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

Calculating the leaching rate for glass and glass-ceramics

The most important chemical properties for glass immobilize the radioactive waste is leaching rate. Radiation can affect the release rate of radionuclides from waste glasses by increasing the surface area for radionuclide release and by changing the dissolution rate. All glasses dissolve to some extent in aqueous solutions. Waste leaching is regarded as the most important process by which long-lived radioactive elements incorporated in glass matrices. The considered for the disposal of highly active nuclear and might be carried in to ground water, and subsequently by returned to the environment. The actual dissolution rate of nuclear waste glasses may be affected by the radiation-induced changes in chemistry properties along radiation-damage tracks. The changes in leach rates due to radiation induced structural changes range from insignificant to significant, and the effects of radiation on radionuclide release are highly controversial.

I. A measurement of leaching rate that has therefore become one of the standard tests for such glasses. To measure leaching rate for the waste that immobilize inside glass and glass-ceramics, there are some properties for samples and their container must be note:

II. The properties for container

III. The container must be not interaction with samples and the solution.

IV. The container does not absorb the ions that outer from samples.

V 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.

Two types of samples first were prepared with waste (5% SrO), and second sample was prepared with (5% CeO).

Stage of exchange distilled water: change water after one week, after two weeks and after three weeks and 1 month, 2 months, 3months and then analysis percentage strontium ion and cerium ion for every part of million (ppm) by use Atomic Absorption device Varian type F-S 240 (Fast-Sequential 240) by putting the solution (distilled water) that wanted to be checked in the device which gave the result directly to know the percentage of leaching strontium ion for the two types of samples glass and glass-ceramics, to compare between their values in same condition and to know which is better leaching resistance (better chemical durability) for long term store.

Results

Table 2 presents the leaching rate values of strontium ions doped after exposure samples to different dose rates of beta ray the results showed its constant in glass and glass-ceramic (0.16) before and after exposure.

Table 2: Leaching rate of strontium ions after exposure samples to beta ray.

Dose (Gy)	Leaching Rate for Glass	Leaching Rate for Glass-Ceramic
After 1-day	0.16	0.16
After 1-month	0.16	0.16
After 2-months	0.16	0.16
After 3-months	0.16	0.16

The leaching rate for all samples were smaller or equal than 0.16 (where 0.16 is a sensitive of atomic absorption device) 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.

Table 3: Leaching rate of cerium ions after exposure samples to neutrons source.

Time	Leaching Rate for Glass	Leaching Rate for Glass-Ceramic
Slow Neutrons		
After 1-week	0.22	0.22
After 1-month	0.22	0.22
After 3-month	0.22	0.22
Fast Neutrons		
After 1-week	0.26	0.26
After 1-month	0.26	0.26
After 3-month	0.26	0.26

Table 3 shows the leaching rate values of cerium ions doped in glass and glass-ceramic samples we can see that its constant (0.22) after exposure to neutron source.

The leaching rate for all samples were irradiated by slow and fast neutrons are smaller or equal than 0.22 and 0.26 respectively, it means that there is no leaching of Ce from glass host due to radiation. In addition, the leaching 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.

Table 3 shows the leaching rate values of cerium ions doped in glass and glass-ceramic samples we can see that its constant (0.22) after exposure to neutron source.

Conclusion

A conclusion from these ionization studies is that the limited magnitude of Sr and Ce leaching associated with ionization damage does not appear to pose any direct problems for the safe storage of nuclear waste glass.

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