The effect on Leaching Rate of Y 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-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 types of 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 leaching was not affected by beta-ray and neutrons.

1. Introduction
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.[1][2]
In this research the results of the leaching rate of glass and glass-ceramic with Ya andCe before and after irradiation by beta rays and neutrons are presented.

2. Experimental work
In this work glass and glass-ceramic materials were prepared as following ways:

2.1. 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-Initialize 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 °C for one hour and then leave to cooling inside the oven gradually.

4-Put the above mixture oxides with Yttrium oxides (5%) inside crucible from alumina inside the furnace at temperature 100 to 900 for 3 hour then to 1300 °C for 2 hours, and shaking the molting inside the crucible to release the CO2 babbles for more homogeny. and left the mix inside the furnace cooling [3].

2.2. 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 leaching rates for many glass and ceramic phases have been investigated as a function of time as following 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 (yttrium 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 [3][4].

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-month-3-month). The percentage between solution volumes that around sample to its surface area must be not over 10 cm so this value was used i.e. \( \frac{V}{S_A} = 10 \text{cm} \) (IAEA) then analysis percentage strontium ion for every part of million (ppm) by use Atomic Absorption device Varian type F-S 240.

3. Result and discussions
3.1. Calculate (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 [4].

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:

- The properties for container
- the container must be not interaction with samples and the solution.
- The container does not absorb the ions that outer from samples.

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% Yo), and second sample were 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 month, 3 month 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 [5].

3.2. Leaching Rate of Ya and Ce Doped in Glass and Glass-Ceramic after Exposure to Beta Ray and Neutrons

Tables (1,2) 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.

| Dose (Gy)       | Leaching rate for glass | Leaching rate for glass-ceramic |
|-----------------|-------------------------|---------------------------------|
| After 1-day      | 0.13                    | 0.13                            |
| After1-month     | 0.13                    | 0.13                            |
| After 2-month    | 0.13                    | 0.13                            |
| After 3-month    | 0.13                    | 0.13                            |
| After 6-month    | 0.13                    | 0.13                            |

The leaching rate for all samples were smaller or equal than 0.13 (where 0.13 is a sensitive of atomic absorption devise) it means that there is no leaching of Y from glass host due to radiation [6]. 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) 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.

Table(3): Leaching rate of Cerium ions after exposure samples to neutrons source

| Type of Exposure | 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                            |

The leaching rate for all samples were smaller or equal than 0.22 it means that there is no leaching of Ce from glass host due to radiation and 0.26. 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 selfdiffusion rates of radioisotopes in the waste form can also affect elemental leach rates by changing the local surface concentration exposed to water[7-9].

4. Conclusion

A Conclusion from these ionization studies is that the limited magnitude of Y 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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