Study the effect of gamma radiation on the some properties of glass and glass-ceramic immobilize nuclear waste

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Abstract

Vitrifications process one of the important methods to immobilize nuclear waste. In this research nuclear waste (Strontium Oxides) with molecular weight (5%) was immobilized by vitrification methods in two types of borosilicate glass (c-type) which are glass and glass-ceramics. To investigate the physical, chemical and mechanical properties of glass and glass-ceramic after immobilize nuclear waste these samples irradiated by gamma ray radiation. Co-60 was used as gamma a irradiation with dose rate 0.38 kGy/hr for different period of time. It's found that gamma radiation affected the glass and glass-ceramic properties. From phase analysis by the x-ray diffraction for glass-ceramic samples proved that at doses 343kGy change the crystalline glass to amorphous glass. A conclusion from these ionization studies is that the limited magnitude of Strontium ion leaching associated with ionization damage does not appear to pose any direct problems for the safe storage of nuclear waste glass.

Key words

glass-ceramic, immobilization nuclear, gamma radiation.

Article info

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تأثير اشعة كاما على بعض خواص الزجاج والزجاج – سيراميك المستعمل لتثبيت النفايات المشعة

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الخلاصة

ان عملية معالجة النفايات المشعة بواسطة تثبيتها في الزجاج واحدة من الطرق المهمة لمنع حركة النفايات النووية . في هذا البحث استعمل أوكسيد السنترنتيوم بنسبة وزنية 5% كنفايات مشعة، زجج في نوعين من الزجاج: البورسليكات (نوع C) الزجاج والزجاج-السيراميك .وتم دراسة الخواص الفيزيائية والكيميائية للزجاج والزجاج-السيراميك بعد تزجيج النفايات النووية المشعة بواسطة تشعيعها بأشعة كاما .تم استخدام الكوبلت 60-كنظير باعث لاشعة كاما بمعدل جرعة 0.38 كيلو كراي /ساعة لفترات مختلفة من الزمن .وجد أن أشعة كاما أثرت على خواص الزجاج والزجاج-السيراميك، وعند الجرعة الاشعاعية 343كيلو كراي تحول الزجاج-السيراميك من الطور البلوري الى زجاج عشوائي.ومن نتائج هذه الدراسة تبين أن نضوح أيون السترونتيوم بعد التعرض لاشعة كاما يكون قليل جدا بحيث لايشكل خطر، لذلك فان خزن النفايات المشعة في الزجاج تكون طريقة جيدة.

Introduction

Nuclear waste pollution is one of the biggest problems effecting in our environment and in our health. The nuclear technology used for different purposes has generated a wide range of different wastes. Each of them requires particular processes of conditioning, packaging and storage[1].

Radioactive waste are waste that contain radioactive materials and it is source

of hazard for human life because its generation from several sources around us (medical, artificial, commercial research activities). So the additional problem is how to disposal from nuclear waste and make it safety and isolation in our environment. Researchers focus about how to disposal from high-level waste because it emits high level radiation, and the isotopes diminishes requires hundreds or thousand years, so it represent the biggest hazard from another type waste so this will required to find suitable safety method to store it for this long-term of time, after invariance failed store it in the tanks because corrosion as a result of long-term preservation and this cause leakage of ions to the underground[2].

Vitrification or molten glass [3] is a muter technology and has been used for high-level waste immobilization for more than 40 years in France, Germany, Belgium, Russia, UK, Japan and USA vitrification methods employ heat up to 1200°C to melt and convert waste materials into glass or crystalline glass.

Radiation effects are of important consideration because of the potential influence on glass corrosion and chemical stability. At an irradiated Si, the first step corresponds to the trapping of a hole at the surface and the formation of intermediates (silicon surface atoms in oxidized state). These intermediates develop toward the final dissolution of Si species, through consecutive chemical species in which electrons are injected into the conduction band[4].

Experimental work

The borosilicate glass C-type (Celsian) prepared from the list oxides insert in the Table1. This table also contains weight percentage and the stage of oxides used for proper base glass. The methods of preparing glass based are list as follows: Mix and crushed the mixture at once, sort the mixture by using sieves, and make annealing process. Then the oxides put inside crucible from alumina inside the oven at temperature 500℃ for one hour and then height the temperature to (1250-1300)℃ for 3-4 hours, and shaking the molting inside the crucible to release the CO_2 bubbles for more homogeny. After ended the process of prepare the base glass, weight percent 5% from SrO which present waste (single ion) were added, the salt of hydroxide strontium was used instead of the ordinary oxide with calculated ratio. The glass-ceramics process consists of two stages of thermal treatment. The first stage of heat treatment is done by annealing the samples at a fixed temperature for appropriate time to have high degree of nucleation and the second stage to have maximum crystallization.

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

Components	Alternative	Molecular Weight%
SiO_2		36.5
AlO_3		10
B_2O_3		5
CaO		10
Na_2O	Na_2CO_3	8.5483
Li_2O	Li_2CO_3	4.9453
TiO_2		6
ZrO_2		1
ZnO		5
MgO		1.5
BaO		18

The samples were irradiated by gamma rays from radioisotope Co-60 source which is emitting gamma radiation with two energies 1.17MeV and 1.35MeV and dose rate 0.38kGy/hr and the two types of samples (glass and glass-ceramics) was irradiated for different period of time as shown in Table 2. The radiation effects on density of glass and glass-ceramic was calculated by immersion two types of samples glass and glass-

ceramics that have nuclear waste (SrO wt 5%) in kerosene.

Table2: The gamma exposure time and the doses of irradiation gamma rays.

Dose	Time of exposure samples of gamma radiation
0.28kGy	45 min
11.71kGy	31hr
14.64kGy	39hr
46.848kGy	123 hr
343kGy	903 hr

Results and Discussion

1.Effect of gamma irradiation on the physical properties (density) of glass and glass-ceramic

The density calculated for samples before exposure to gamma ray radiation and after irradiation with different doses is shown in the Table3.

Table3: The variation of the glass and glass-ceramics density with different gamma doses

Dose of gamma radiation kGy	Glass density g/cm^3	Glass-ceramics density g/cm^3
0	3.023	3.025
0.28	3.005	3.143
11.71	3.005	3.143
14.64	3.005	3.143
46.8	3.005	3.143
343	3.005	3.143

Table 3 shows that the value of glass density is decreased after irradiation but the density of glass-ceramic increases after irradiation with gamma doses, after that the densities remain constant with increasing radiation dose.

The influence of radionuclide decay on vitrified nuclear waste may be manifested by density changes, (volume). The volume (or density) of glass and crystalline materials may change as a result of atomic displacements that occur after exposure to radiation sources.

Solid glass ionization damage results from the excitation and ejection of electrons from the valence orbitals of the target atom by incident radiation. Although ionizing radiation produces few direct displacements, electron excitations, if they are localized and persist long enough, may eventually lead to atomic displacements. This damage effect may manifest itself in several forms, including electron hole pairs, covalent bond rupture, valence changes, H_2O and OH- decomposition, Compton scattering, photoelectric effects, and decomposition of unstable molecular ions [5,6].

2. Effect of gamma ray on the chemical properties (Leaching rate) of glass and glass-ceramic

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 immersion in the water.

The release of glass components into solution, including radionuclides, may be changed by the presence of radiolytically produced nitric acid, carboxylic acid, and transient water dissociation products such as and O_2^- . Under batchtest conditions[6], glass corrosion has been shown to increase a maximum of three- to five-fold in gamma- and alpha-irradiated tests relative to non-irradiated tests, while in other studies; the presence of radiolytic products has actually decreased the release rates of some glass components. Bicarbonate ground waters will buffer against pH decreases and resultant corrosion rate increases arising from the formation of radiolytic acids. For all samples were calculated the leaching of strontium ions within ppm unit.

Table4: Leaching rate of strontium ions for samples before exposure to gamma radiation and after irradiated with 0.28kGy gamma ray.

Period of time kept samples in distilled water	Leaching rate of strontium for glass sample	Leaching rate of strontium ions for glass-ceramics sample	Leaching rate of strontium for glass sample after irradiation with 0.28kGy	Leaching rate of strontium ions for glass-ceramics sample after irradiation with 0.28kGy
After one week	<0.25	<0.25	<0.25	<0.25
After two weeks	<0.25	<0.25	<0.25	<0.25
After three weeks	<0.25	<0.25	<0.25	<0.25
After two months	<0.25	<0.25	<0.25	<0.25

Table 4: Leaching rate of strontium ions after exposure samples to 46.848kGy and 343kGy gamma radiation.

Period of time kept samples in distilled water	Leaching rate of strontium ions for glass sample after irradiation with *46.8kGy*	Leaching rate of strontium ions for glass-ceramics after irradiation with *46.8kGy*	Leaching rate of strontium ions for glass sample after irradiation with *343kGy*	Leaching rate of strontium ions for glass-ceramics after irradiation with *343kGy*
After one week	<0.25	<0.25	<0.25	<0.25
After two weeks	<0.25	<0.25	<0.25	<0.25
After three weeks	<0.25	<0.25	<0.25	<0.25
After two months	<0.25	<0.25	<0.25	<0.25

Gamma irradiation of glass, on the other hand, is reported to lead mainly to surface damage, unstable charging, and migration of mobile (non-network) cations. The electron hole trapping sites in borate glass are believed to arise from the local non-stoicheometry, which is present in the glass as a result of either fabrication or radiation induced atomic displacement [7].

3. Phase analysis (x-ray diffraction and scanning electronic microscope)

1-The x-ray spectra shown in Figs.1,3 and,5 for glass samples proved that the glass was fully amorphous before and after exposure to gamma radiation, no peaks appearance in it.

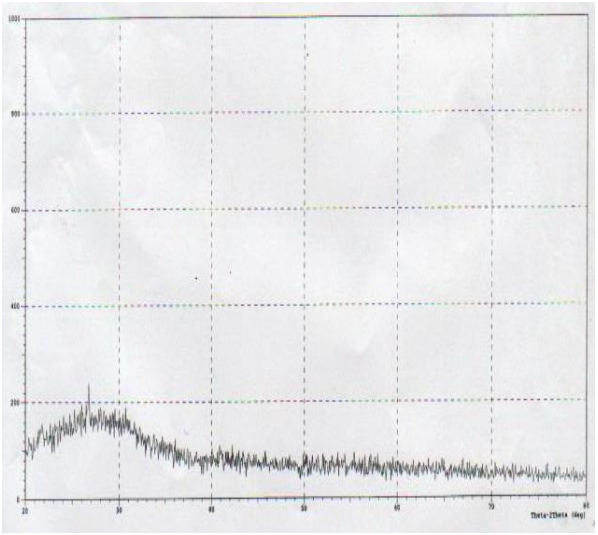

Fig.1: The x-ray diffraction for glass sample the glass before exposure to gamma radiation.

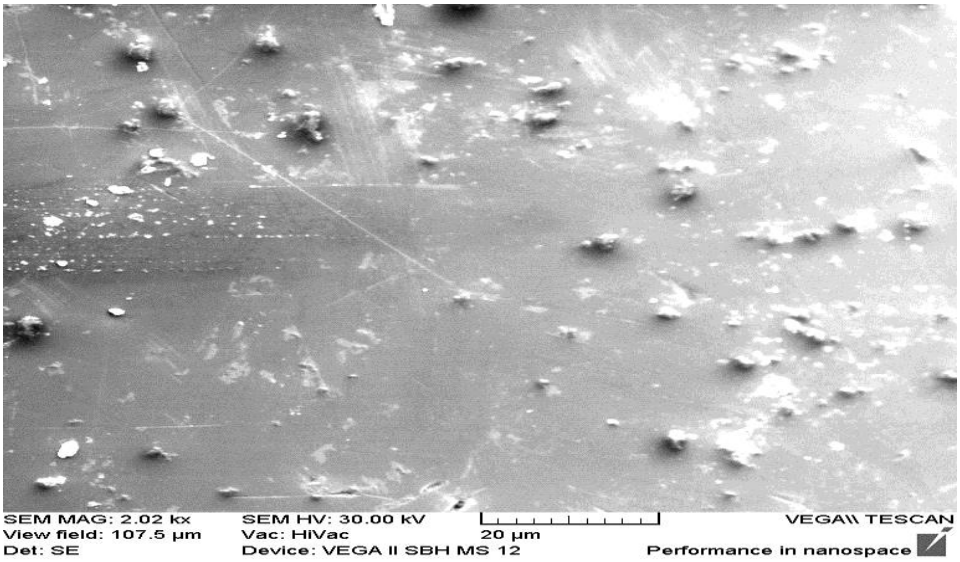

Fig. 2: The SEM micrograph for glass sample before exposure to gamma radiation.

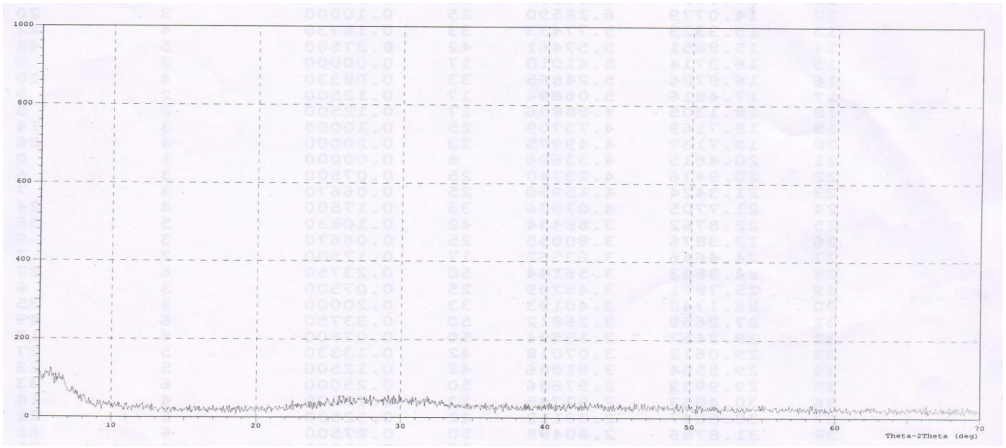

Fig. 3: The X-ray diffraction for glass after exposure to 0.28kGy gamma radiation.

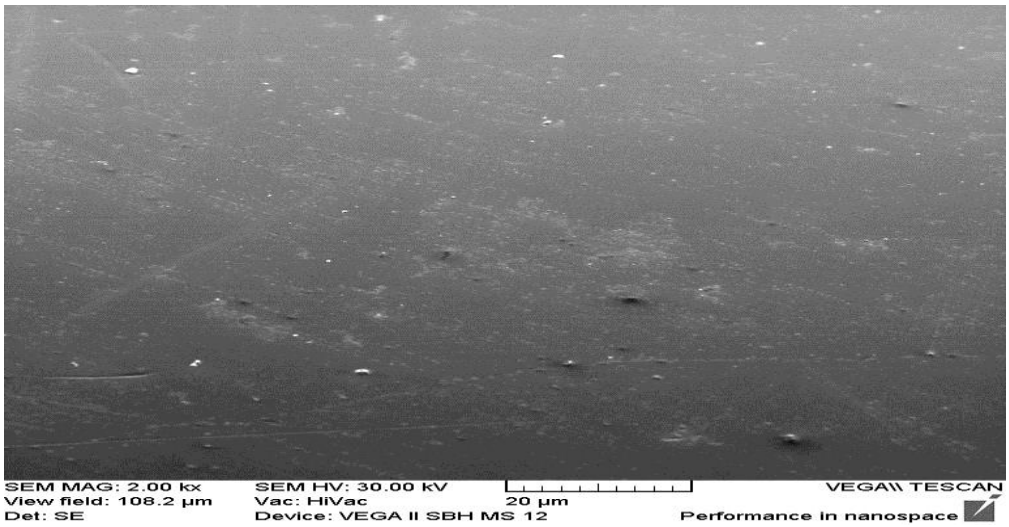

Fig. 4: The SEM micrograph for glass sample after exposure to 0.28kGy of gamma radiation.

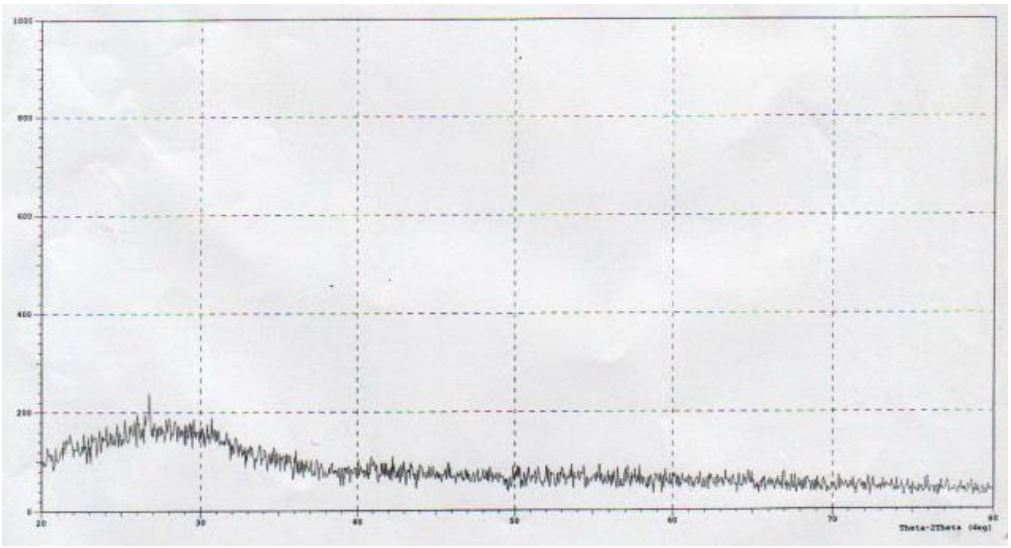

Fig. 5: The X-ray diffraction for glass sample after exposure to 343kGy gamma radiation.

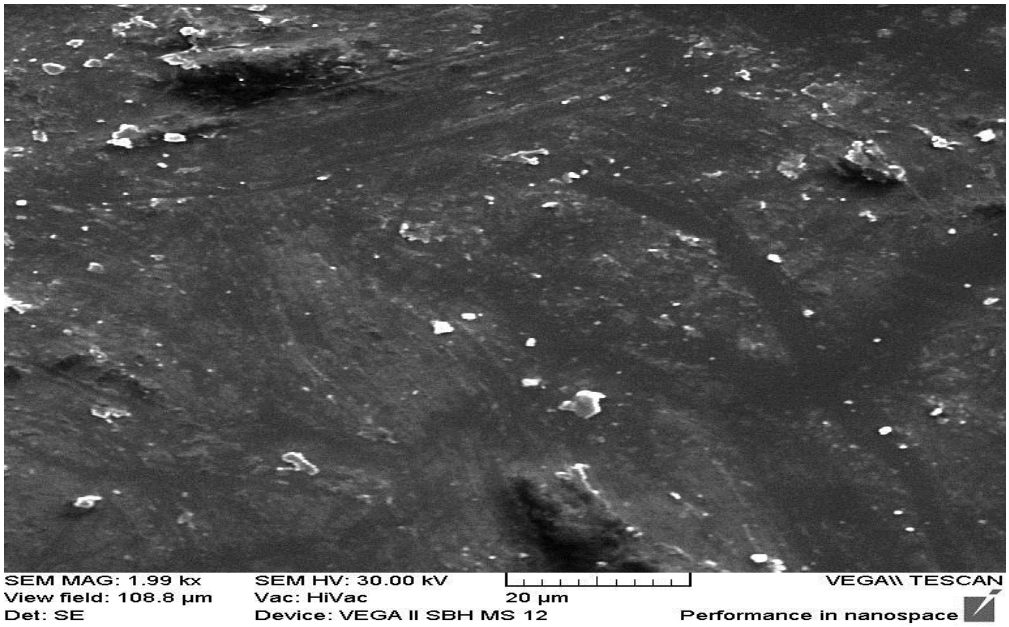

Fig. 6: The SEM micrograph for glass sample after exposure to 343kGy of gamma radiation.

2- The XRD spectra for glass-ceramics Figs. 7, 9, and 11 appear peaks in the spectra establish that crystalline phase was found (phase changing by heat treatment) and the appearance phase is Calcium Aluminum Silicate.

From the SEM Figs,2,4,6,8,10 and12 and X-ray diffraction spectra the threshold dose, where damage effects first appear, and a saturation dose for glass-ceramic materials can be determined. In present study the threshold doses for change the crystalline glass to amorphous glass is 343kGy.

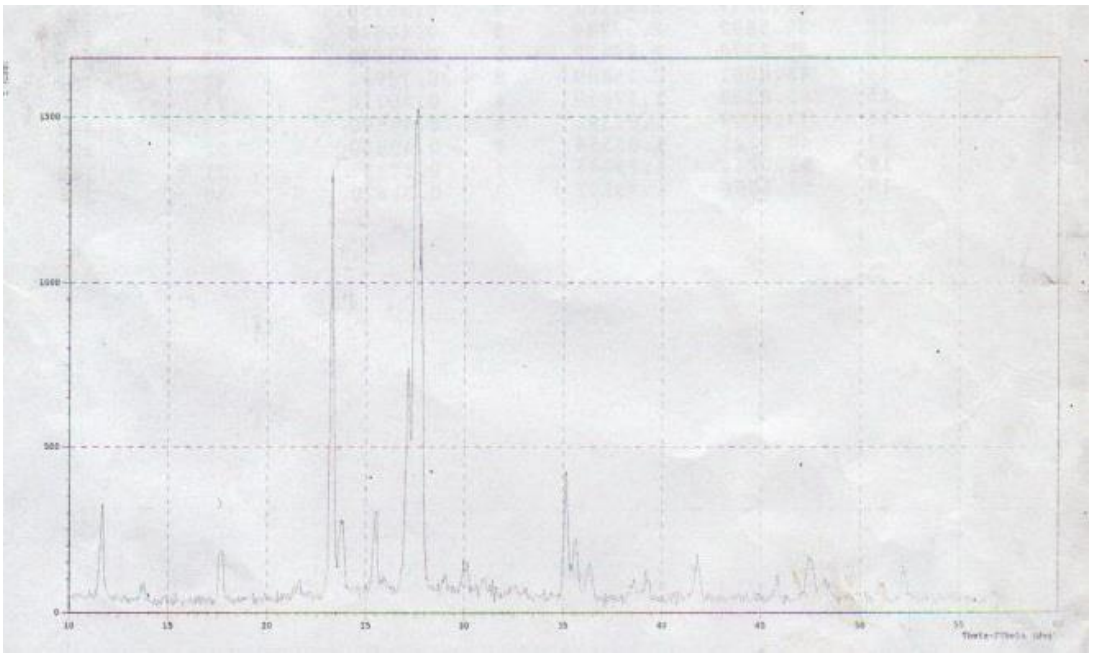

Fig. 7: The X-ray diffraction for glass-ceramics samples before exposure to gamma radiation.

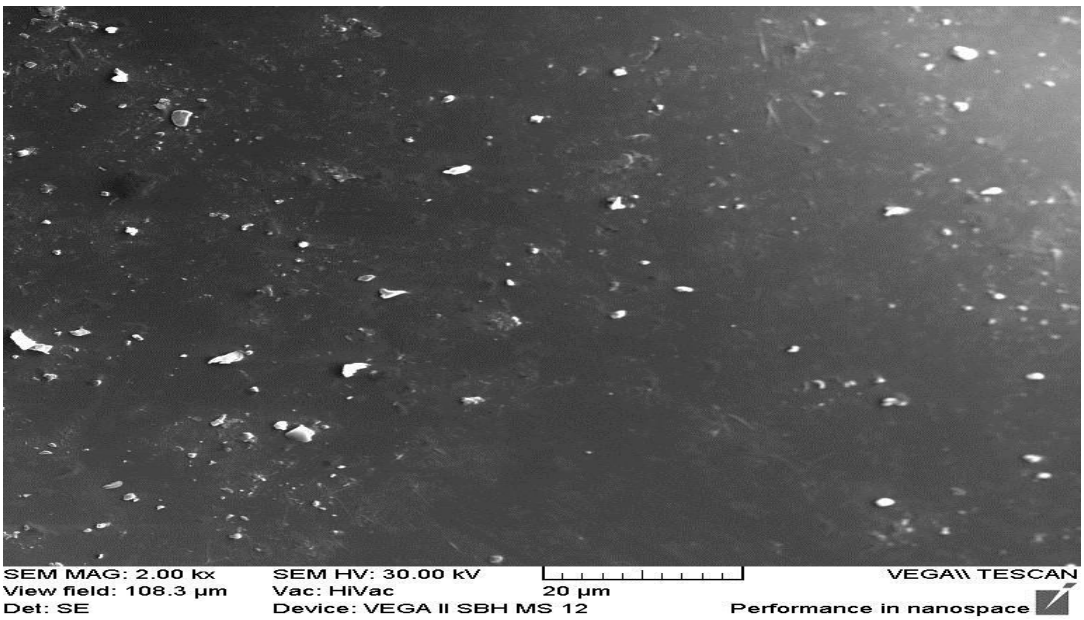

Fig. 8: The SEM micrograph for glass-ceramics samples before exposure to gamma radiation.

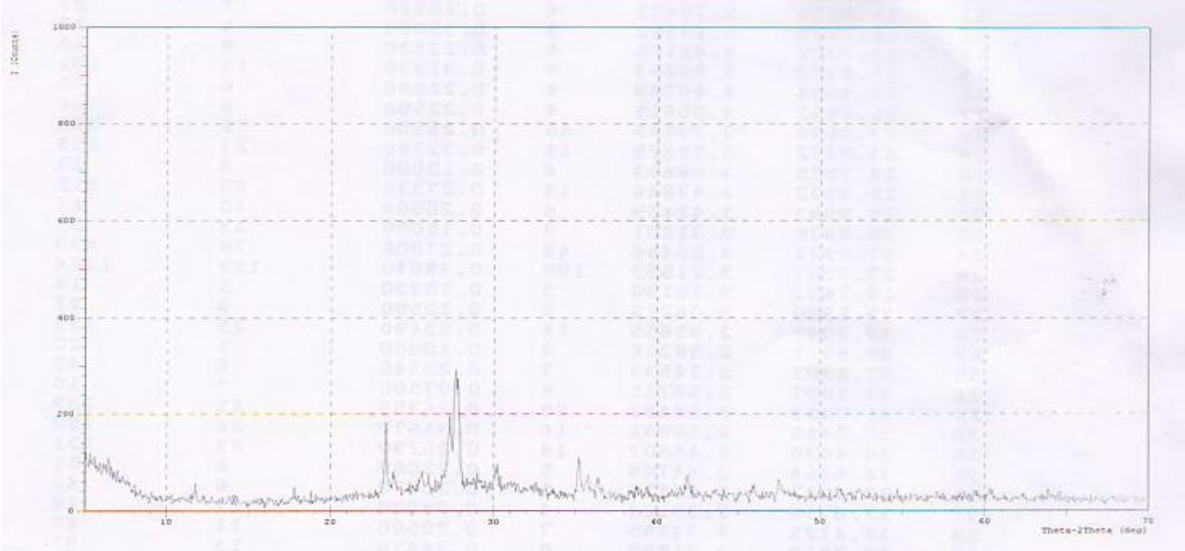

Fig. 9: The X-ray diffraction for glass-ceramics after exposure to 0.28kGy of gamma radiation

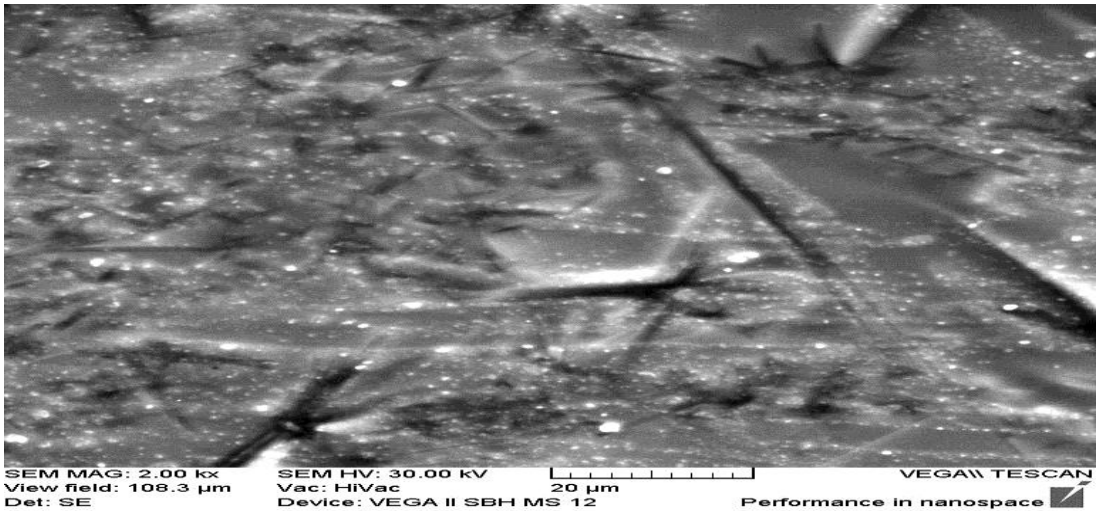

Fig. 10: The SEM micrograph for glass-ceramics sample after exposure to 0.28kGy gamma radiation.

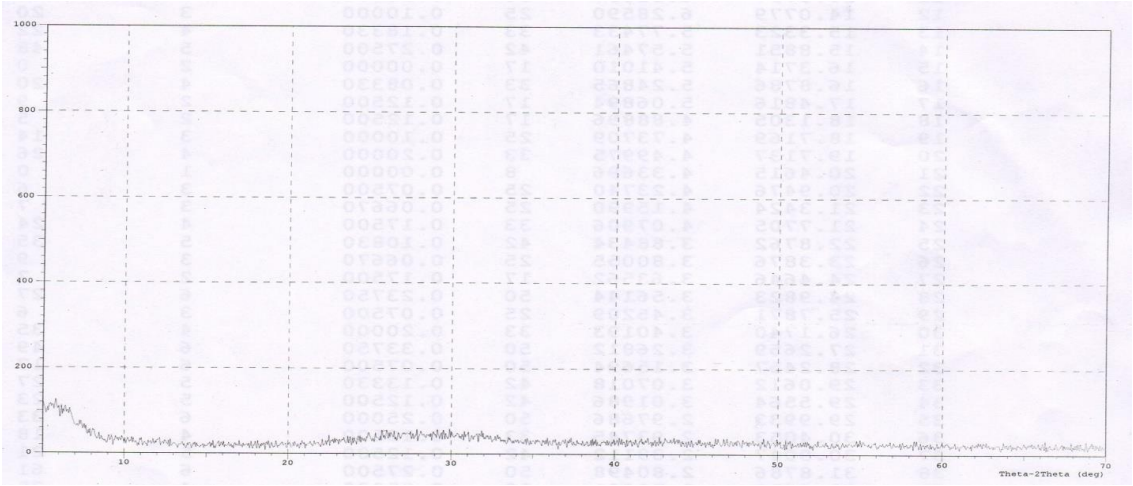

Fig. 11: The X-ray diffraction for glass-ceramics after exposure to 343kGy of gamma radiation.

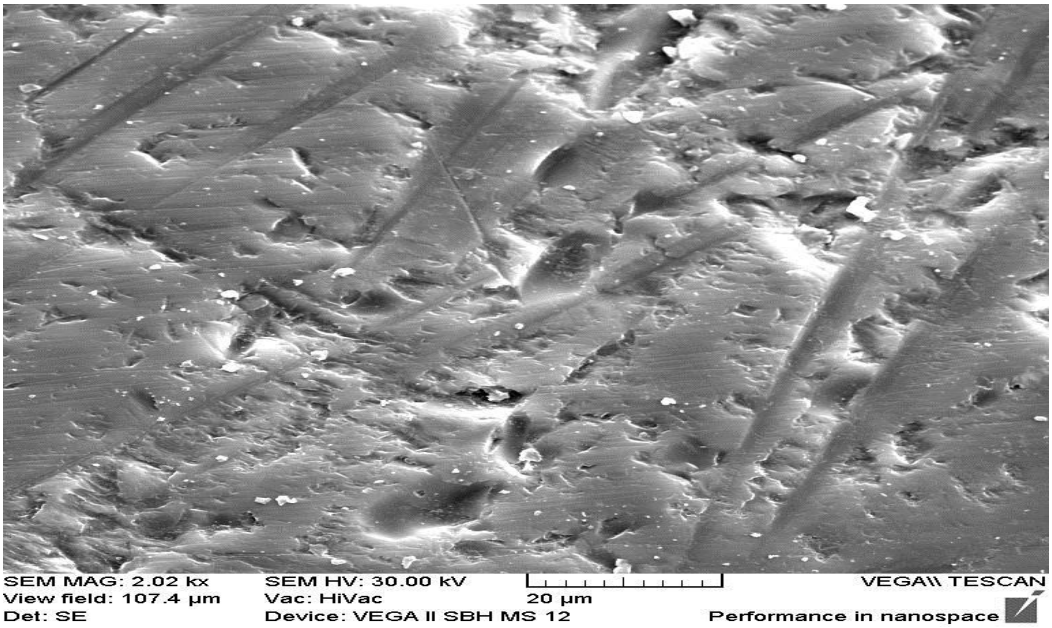

Fig.12: The SEM micrograph for glass-ceramics sample exposure to 343kGy of gamma radiation.

Conclusions

Glass melting is a complex process governed by several parameters including initial compositions, melting temperature, heating and cooling rates, and the type of equipment used. The main conclusions of the present study can be summarized as follows:

The effect of gamma ray irradiation on the density and leaching, for borosilicate glasses and glass-ceramic has been measured for doses between 0 and 343kGy, the radiation-induced change in density of these glasses as gamma ray doses increases. The density for glasses decreases after irradiation by gamma ray, and the value of density remain constant with more increasing irradiation doses.

A conclusion from these ionization studies is that the limited magnitude of Sr 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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