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# High temperature annealing studies of strontium ion implanted glassy carbon

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ABSTRACT

Glassy carbon samples were implanted with 200 keV strontium ions to a fluence of $2 \times 10^{16}$ ions/cm$^2$ at room temperature. Analysis with Raman spectroscopy showed that ion bombardment amorphises the glassy carbon structure. Partial recovery of the glassy carbon structure was achieved after the implanted sample was vacuum annealed at 900 °C for 1 h. Annealing the strontium ion bombarded sample at 2000 °C for 5 h resulted in recovery of the glassy carbon substrate with the intensity of the D peak becoming lower than that of the pristine glassy carbon. Rutherford backscattering spectroscopy (RBS) showed that the implanted strontium diffused towards the surface of the glassy carbon after annealing the sample at 900 °C. This diffusion was also accompanied by loss of the implanted strontium. Comparison between the as-implanted and 900 °C depth profiles showed that less than 30% of the strontium was retained in the glassy carbon after heat treatment at 900 °C. The RBS profile after annealing at 2000 °C indicated that no strontium ions were retained after heat treatment at this temperature.



## 1. Introduction

The burning of fossil fuels to generate electricity has led to numerous environmental problems such as global warming, greenhouse effect and urban smog. Most countries including developing countries are now looking into having a cleaner alternative for power generation [1]. Nuclear energy is a cleaner alternative way of generating electricity because of its environmental benefits. Despite all the advantages of using nuclear energy in lieu of fossil fuels, it is still the most controversial source of power generation because of the toxicity of the wastes generated, the difficulties in the storage of the wastes produced and the dangers associated with nuclear reactors. Due to the environmental and health hazards associated with nuclear waste i.e. fission products, there is a need to store them in containers made of suitable materials that can contain them for a long period of time [2].

Glassy carbon is a graphitic form of synthetic carbon with remarkable properties. Some of its relevant properties for this study include high strength, high temperature resistance, low permeation of gases and an extremely high resistance to chemical attacks. Based on these properties, glassy carbon has some industrial applications such as electrodes in electrochemistry [3] and high temperature crucibles used in laboratories [4].

The aim of this study is to investigate if glassy carbon will be a good material for nuclear waste storage. In order for glassy carbon to be a good nuclear waste storage material, it must be a good diffusion barrier for nuclear fission products. Glassy carbon must also be able to retain its properties even under radiation conditions. To study the effectiveness of glassy carbon as a good storage material, a nuclear fission product, namely strontium, was implanted in glassy carbon to a fluence of $2 \times 10^{16}$ ions/cm$^2$ at room temperature. Ion implantation has the additional advantage for this study in that it introduces radiation damage in the substrate. Recent studies [5,6] have shown that the structure of glassy carbon changes to that of amorphous carbon after high dosage ion implantation. This change in structure is usually accompanied with a change in the atomic density of the bombarded glassy carbon.

The diffusion behaviour of the implanted strontium in the glassy carbon substrate was then monitored using Rutherford backscattering spectroscopy (RBS) while the corresponding structural changes (i.e. radiation damage) were monitored using Raman spectroscopy. The effect of high temperature annealing on the diffusion behaviour of strontium in glassy carbon and the corresponding structural changes are reported in this paper.

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## 2. Experimental procedure

Sigradur® G glassy carbon samples were mechanically polished to a mirror finish with a 1 μm and 0.25 μm diamond solutions finish respectively. After polishing, the samples were cleaned with soap solution, de-ionised water and methanol in an ultrasonic bath. The samples were then implanted with 200 keV strontium ions to a fluence of $2 \times 10^{16}$ ions/cm$^2$ at room temperature. Because of our interest in investigating the radiation damage due to ion bombardment, the flux during implantation was kept low at about $10^{12}$ cm$^{-2}$ s$^{-1}$ to minimise the temperature increase during implantation. During implantation the temperature rose to about 55 °C.

Two samples were then annealed in vacuum at 900 °C for 1 h and 2000 °C for 5 h, respectively.

Rutherford backscattering spectroscopy (RBS) was used to study the diffusion behaviour of the implanted strontium in glassy carbon after heat treatment. Helium particles of energies 1.4 MeV and 1.6 MeV were employed, in a Van de Graaff accelerator. A total charge of 8 μC was used throughout this study with an analysing current of 15 nA and a scattering angle of 165°. In order to ensure accuracy and noise reductions, repeated readings were taken.

The structural changes as a result of strontium ion bombardment and heat treatment were monitored by using a Jobin Yvon T64000 Raman instrument. The instrument consists of a Raman spectrometer with Ar/Kr mixed gas excitation laser of 514.5 nm wavelength. One of the major problems encountered in Raman studies is sample heating. In order to avoid this, the laser power at the sample was kept lower than 1 mW.

## 3. Results and discussion

Fig. 1 shows the RBS depth profile of glassy carbon implanted with $2 \times 10^{16}$ Sr$^+$/cm$^2$ at room temperature. The experimental strontium concentration is about 1.7 relative atomic percentage. The as-implanted spectrum was fitted with a Gaussian distribution and the projected range $R_p$ was estimated to be about 165 nm and the projected range straggling; $\Delta R_p$ was calculated to be 61 nm. The experimental depth profile was compared with SRIM simulations. The SRIM simulation was carried out using the density of the Sigradur G glassy carbon (1.42 g/cm$^3$) and a predicted $R_p$ of 157 nm and straggling ($\Delta R_p$) of 34.5 nm respectively. The straggling value obtained experimentally is 43% larger than the value predicted by SRIM. This implies that re-distribution/diffusion of the strontium was already taking place during implantation. The larger value for the experimentally determined projected range, $R_p$ may be due to a change in the glassy carbon structure within the implanted region.

The RBS Sr concentration depth profile obtained after heat treatment of the sample at 900 °C and 2000 °C are shown in Fig. 2 below. The spectra were compared with the as-implanted depth profile. Annealing of the sample at 900 °C resulted in the migration of strontium towards the surface of the glassy carbon substrate. Since the melting and boiling points of strontium are 777 °C and 1377 °C, respectively, the strontium atoms on the surface of the glassy carbon sublimated/evaporated into the vacuum, leading to a loss of the implanted material. The retained strontium in the substrate after heat treatment at 900 °C was calculated to be 30%. The depth profile of the strontium implanted glassy carbon after heat treatment at 2000 °C showed that no implanted strontium was retained in the glassy carbon.

Because it is important for nuclear reactor applications that the glassy carbon retains its properties even in extreme conditions, the effect of ion implantation and subsequent diffusion of the implanted strontium on the structure of the glassy carbon was investigated using Raman spectroscopy. Raman spectroscopy has been proven to be an excellent probe for identifying different structural information especially in carbon based materials.

Fig. 3 shows the Raman spectrum of the virgin glassy carbon substrate. The spectrum consists of two prominent peaks namely; the D peak located at 1350 cm$^{-1}$ and the G peak located at 1585 cm$^{-1}$. These peaks exist in carbon based materials with sp$^2$ carbon bonds with disorder. The glassy carbon spectrum shows an additional peak at 1620 cm$^{-1}$, indicated by an arrow in Fig. 3. This peak is usually denoted with D′ and it is present in graphitic carbons with very small size of sp$^2$ domains [7,8]. In addition to these peaks, a small peak was also observed at 1100 cm$^{-1}$. This peak has been said to be due to the effect of intersection of acoustic and optic modes in the density of states of graphitic carbon materials [7,9]. The appearance of the peaks at 1100 cm$^{-1}$ and 1620 cm$^{-1}$ in the glassy carbon Raman spectrum indicates that glassy carbon is an sp$^2$ carbon material with small crystallite size. The virgin Raman spectrum was fitted with a combination of the Lorentzian equation (for the D peak) and a BWF (Breit–Wigner–Fano) equation (for the G peak). The heights of these peaks were taken as their intensities. The $I_D/I_G$ ratio was then calculated to be 1.4. The Tuinstra–Koenig relation [10] can be used to estimate

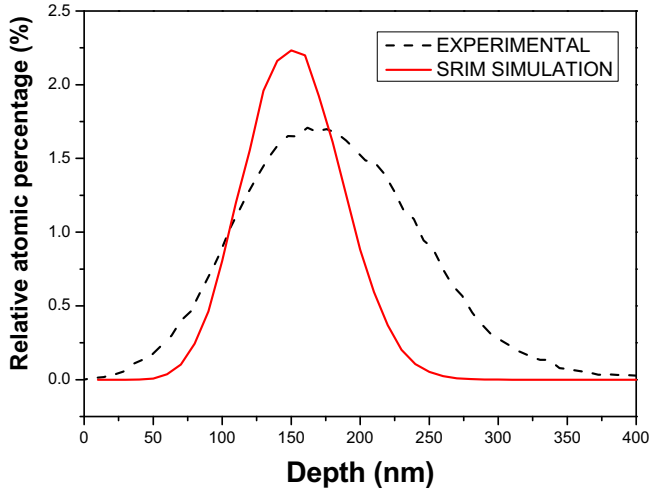


**Fig. 1.** Distribution of 200 keV Sr$^+$ in glassy carbon as a function of depth.

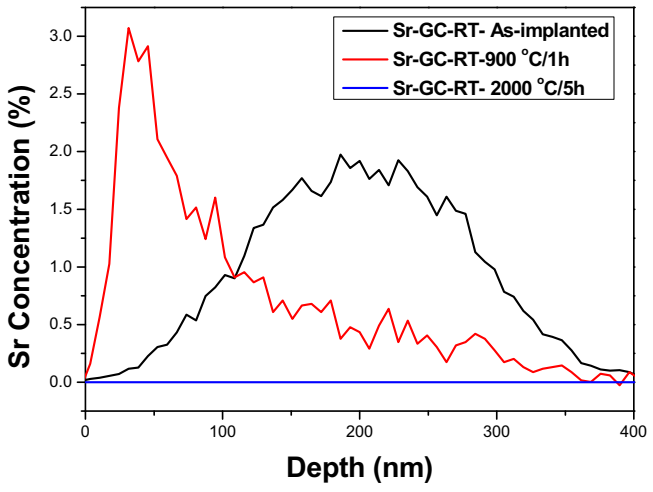


**Fig. 2.** Sr concentration depth profile showing the effect of high temperature vacuum annealing on the diffusion of strontium implanted in glassy carbon.