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Investigating the effect of heat treatment on the diffusion behaviour of xenon implanted in glassy carbon

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ABSTRACT

The effect of sequential isochronal annealing on the diffusion behavior of implanted xenon in glassy carbon is reported. Glassy carbon substrates were implanted with 200 keV xenon ions to a fluence of $1 \times 10^{16} Xe^{+} cm^{-2}$. The sample was annealed in vacuum at temperatures ranging from 300 °C to 1000 °C for 5 h in steps of 100 °C. The RBS depth profiles obtained at temperatures above 800 °C showed that some diffusion occurred. The broadening of the peaks was not accompanied with a loss of the implanted Xe. Microstructural changes in the glassy carbon substrate due to Xe bombardment and annealing were monitored using Raman spectroscopy. The Raman spectrum obtained after xenon bombardment showed that the glassy carbon substrate became amorphized. However, a slight recovery of the glassy carbon structure was noticed after heat treatment. The SEM micrographs of the glassy carbon substrate showed an increase in the surface roughness of the glassy carbon substrate after implantation. The increase in the roughness of the glassy carbon substrate was attributed to the sputtering of the loosely bonded carbon atoms along the polishing marks after implantation and annealing.

1. Introduction

Recently, there has been an interest in employing glassy carbon to contain radioactive fission products. Glassy carbon is an apparently isotropic, continuous and non-porous material, showing conchoidal surface fracture. It is a non-graphitizing carbon which combines glassy and ceramic properties with those of graphite. Unlike graphite, glassy carbon has a fullerene-related microstructure [1]. This leads to a great variety of unique materials properties. Its high strength, high hardness and also high impermeability to gases, suggest that glassy carbon must have a unique structure different from those of regular carbons [2]. These properties allow glassy carbon to be used in a wide range of applications such as a nuclear waste storage containment material to lining of pressure vessels to acid-battery electrodes. Waste storage facilities should be designed and operated to minimize the probability and consequences of incidents and accidents. Factors that should be considered include the following: impermeability to the fission products, chemical stability against corrosion caused by processes within the waste and/or external conditions, protection against radiation damage and/or thermal damage, especially stability against the degradation of materials and resistance to impacts from operational loads or due to incidents and accidents [3].

Xenon (Xe) is a noble gas mainly produced in nuclear fission reactions. It consists of various radioactive isotopes includingXenon-135, which is of considerable significance in the operation of nuclear power reactors. Xenon-135 is a product of U-235 fission and has a very large neutron-capture cross section. This means that it acts as a neutron absorber or poison that can slow down or stop the chain reaction after a period of operation [4]. A major contribution to the sequence of events leading to the Chernobyl nuclear disaster was the failure to anticipate the effect of xenon poisoning on the rate of the nuclear fission reaction [5]. Decays radioactively with a half-life of 9.1 h. Little of the Xe-135 results directly from fission; most comes from the decay chain, Te-135 to I-135 to Xe- 135 [6].

The aim of this paper is to investigate the diffusion behavior of implanted Xe in glassy carbon. As was mentioned above, this

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information is necessary to determine the effectiveness of glassy carbon as a good construction material for the casks used for nuclear waste storage. The structural changes during annealing heat treatment at different temperatures were characterised by Raman spectroscopy. Surface topographical changes due to the annealing was investigated by SEM.

2. Experimental procedure

The glassy carbon (Sigradur®G) samples were polished with 1 μm and 0.5 μm diamond solutions respectively. The samples were implanted with 200 keV xenon ions to a fluence of 1×10^{16} ions/cm^2 at room temperature. The diffusion behavior of Xe in glassy carbon due to annealing was investigated using Rutherford backscattering spectrometry (RBS) with 1.6 MeV He^+ particles in a Van de Graaff accelerator. A total charge of 8 μC was used with an analysing current of 15 nA and a scattering angle of 165°. The resulting RBS depth profiles were fitted to the solution of the Fick diffusion differential equation for an initially Gaussian implanted profile [7]. The effect of Xe implantation and annealing on the microstructure of glassy carbon was monitored by using a Jobin Yvon, Horiba(C) TX64000 triple grating Raman spectrometer with a 514.5 nm (green) laser. The excitation laser used in this study was an Ar/Kr mixed gas laser. The 50 × objective was used to acquire the Raman spectra. One of the major setbacks of Raman spectroscopy is sample heating. In order to avoid this, the laser power was kept below 1 mW at the sample during analysis.

The surface morphology of glassy carbon due to Xe implantation and annealing was investigated by using the scanning electron microscope (SEM). SEM images were obtained by using the Zeiss Ultra Plus SEM. An analysing voltage of 2 kV was used. In-lens SEM images of the sample were taken before implantation, after implantation and after every heat treatment so as to investigate all surface changes due to these treatments.

3. Results and discussion

Fig. 1 shows the RBS depth profile of 200 keV xenon implanted in glassy carbon at room temperature. The profile was compared with the spectra obtained from a SRIM simulation. Fig. 1 also shows the vacancy distribution obtained from SRIM. The experimental projected range, R_p, was estimated by fitting the Xe depth profile with a Gaussian equation and the value obtained was 120 nm. This R_p value obtained for the room temperature implanted Xe depth profile is comparable to the 119 nm value obtained from SRIM. The experimental straggling, ΔR_p value obtained is about 34 nm which is higher than the 21 nm obtained from SRIM.

Some of the implanted samples were annealed in vacuum at 300 °C -1000 °C in steps of 100 °C for 5 h – see Fig. 2 (a) and (b). Two diffusion mechanisms can be clearly observed from the spectra shown. The first diffusion regime is between 300 °C - 800 °C. The RBS profiles obtained after annealing the samples at these temperatures showed no noticeable diffusion of the implanted Xe. The lack of diffusion at these temperatures might be due to the presence of defects in the implanted region of the glassy carbon substrate acting as traps for the implanted xenon. The diameter of Xe atom of 216 p.m. is much larger that the C atom of 77 pm. In Fig. 1 the SRIM vacancy distribution is compared to the implanted distribution. The $R_{p(vac)}$ is about 79 nm which is significantly lower than the R_p value of the as-implanted Xe depth profile (120 nm). This discrepancy implies that the defects introduced into the glassy carbon substrate are concentrated towards the surface. The fact that the Xe atoms deeper inside the glassy carbon also did not diffuse indicates that the concentration of the vacancies was enough to trap the (also fewer) Xe atoms.

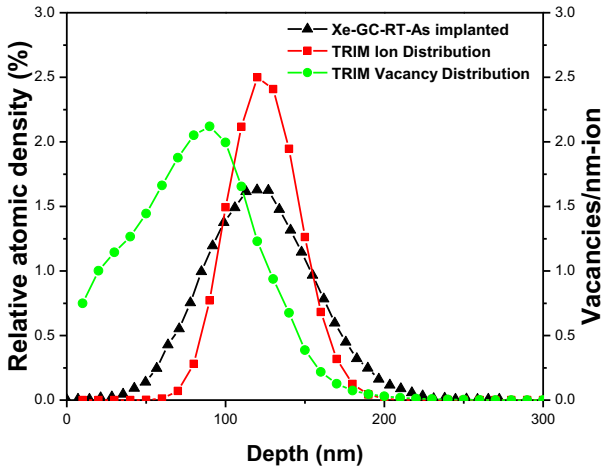

Fig. 1. RBS depth profile of 200 keV Xe^+ ions implanted in glassy carbon at room temperature and ion distribution profile together with vacancy distribution obtained from SRIM [8].

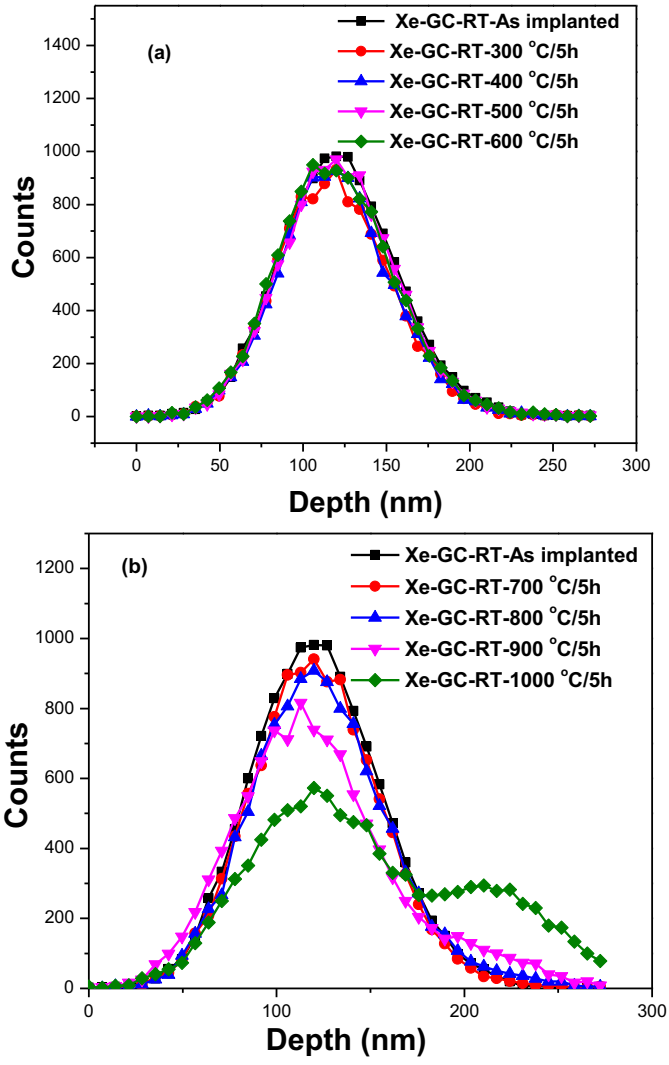

Fig. 2. RBS Depth profiles of Xe implanted at room temperature after isochronal annealing at (a) 300 °C - 600 °C for 5 h and (b) 700 °C - 1000 °C for 5 h.