Investigating the diffusion of Xe implanted into glassy carbon

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Abstract. Recently, there has been a renewed interest in employing glassy carbon to contain radioactive fission products. One of the fission products, Xe is significant by itself due to its high neutron absorption cross section and high production as a fission product. 200 keV Xenon (Xe) ions were implanted in the glassy carbon samples to a fluence of 1×10^{16} Xe^{+} cm^{-2} at room temperature. The diffusion of the implanted Xenon in the glassy carbon was measured using Rutherford backscattering (RBS) after vacuum annealing.

1. Introduction

Glassy carbon (GC) 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 [1]. Unlike graphite, glassy carbon has a fullerene-related microstructure 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 in nuclear waste storage containment material.

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].

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Xenon (Xe) is a noble gas mainly produced in nuclear fission reactions. It consist of various radioactive isotopes including Xenon-135, which is of considerable significance in the operation of nuclear power reactors. The reason being 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]. Xenon-135 is a product of U-235 fission and has a very large neutron-capture cross section. It also decays radioactively with a half-life of 9.1 hours. Little of the Xe-135 results directly from fission; most comes from the decay chain, Te-135 to I-135 to Xe- 135 [6].

In this study, the diffusion behavior of implanted Xe in glassy carbon was investigated. As has been mentioned above, this information is necessary to determine the effectiveness of glassy carbon as a good material for constructing the casks used in nuclear waste storage.

2. Experimental procedure

The glassy carbon (Sigradur®G) samples were polished with 1 μm and 0.5 μm respectively with diamond solutions. The samples were implanted with 200 keV xenon ions to a fluence of 1×10^{16} ions/cm^2 at room temperature. The implanted sample was annealed in vacuum at temperatures increasing in steps of 100 °C from 300 °C to 1000 °C for 5h at each temperature. The depth distribution of the implanted xenon before and after each annealing step was analyzed using Rutherford backscattering spectroscopy (RBS).The RBS spectra were acquired using 1.6 MeV He^+ ion beam at beam current of 15nA. The RBS spectra were converted into the respective depth profiles of the implanted xenon. The resulting RBS depth profiles were fitted with a MATLAB program that was written by Malherbe et al [7] to extract diffusion coefficients from an initial Gaussian profile.

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 the TRIM ion distribution simulation. Fig. 1 also contains the vacancy distribution obtained from TRIM [8]. The experimental projected range, R_p, was estimated by fitting the Xe depth profile with a Gaussian equation. The value obtained is 120 nm. This value obtained for the room temperature implanted Xe depth profile is comparable to the 119 nm obtained from TRIM. The experimental straggling, ΔR_p value obtained is about 34 nm which is higher than the 21 nm obtained from TRIM. The higher ΔR_p obtained from the as-implanted Xe profile suggests a broader ion distribution to that suggested by TRIM.

Some of the implanted samples were annealed in vacuum at 300 °C -1000 °C in steps of 100 °C for 5 hours – 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 can be attributed to the presence of defects in the near surface region of the GC substrate. This statement is supported by the TRIM vacancy distribution shown in Fig. 1. 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 GC substrate are concentrated towards the surface.

In the second diffusion regime (900 °C - 1000 °C), movement of Xe into the bulk of the GC was observed with the formation of a bimodal distribution. The migration of Xe into the GC was accelerated at 1000 °C with a new Xe peak formed within the bulk of the GC. The diffusion coefficients at temperatures 900 °C - 1000 °C were calculated to be $D = 4.3 \times 10^{-21}$ m^2/s and $D = 6.7 \times 10^{-21}$ m^2/s respectively. 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 pm is much larger that the C atom of 77 pm. Thus, it effectively represents the boundary between the radiation damaged glassy carbon and the pristine bulk. This penetration of xenon into pristine glassy carbon together with the segrational kind of diffusion of the

implanted xenon atoms towards the bulk at the higher annealing temperatures.

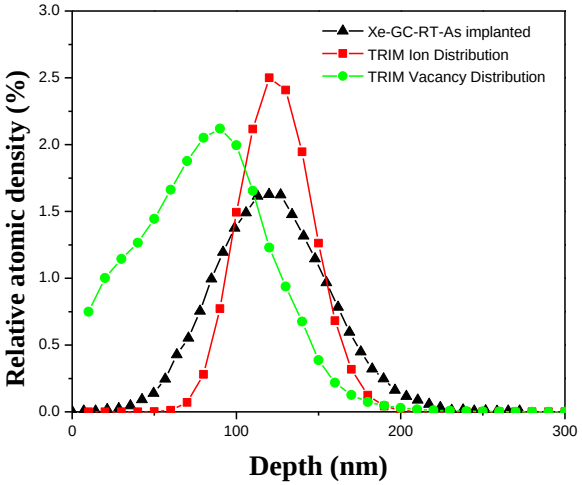

Figure 1. Depth profile of 200 keV Xe^{+} ions implanted in glassy carbon at room temperature and vacancy distribution from TRIM [8].

The movement of Xe deeper inside the GC substrate indicated that the implanted Xe will be diffused towards the bulk of GC after annealing at temperatures above 900 °C. This suggests that a Fickian type of diffusion is not responsible for the diffusion observed in this regime. Odutemowo et al [9] studied the diffusion of strontium in glassy carbon and observed a similar phenomenon when they annealed their Sr implanted GC samples at high temperatures. However, the segregation of Sr was observed towards the surface of GC instead of towards the bulk as observed in our results. They suggested that a prime factor to be considered in this segregational kind of diffusion is the Gibb's free energy of the system.

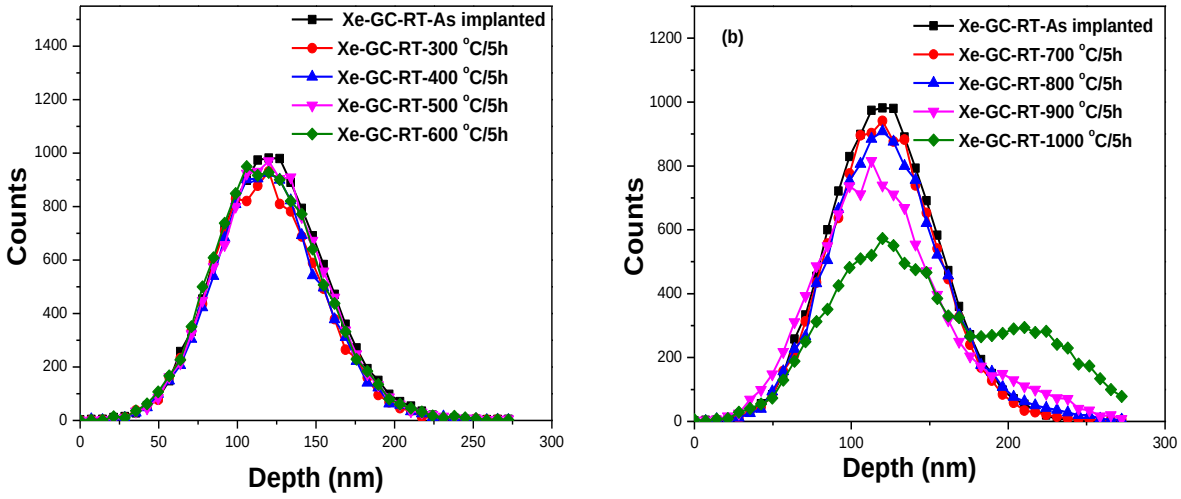

Figure 2. RBS Depth profiles of Xe implanted at room temperature after isochronal annealing at (a) 300 °C - 600 °C for 5 h and (b) 700 °C - 1000 °C for 5 h.

In order to estimate the diffusion coefficient of Xe in glassy carbon, the RBS depth profiles obtained at 300 – 1000 °C were fitted with an in-house MATLAB program. The MATLAB program fits experimental depth profiles to the solution of the Fick diffusion equation with originally a Gaussian profile and with a perfect sink at the surface. Examples of such fit are shown in Fig. 3. The fitting of the RBS spectra show that the diffusion coefficient of Xe in GC could not be estimated. This is in agreement with the results discussed earlier (no noticeable diffusion of Xe was observed).

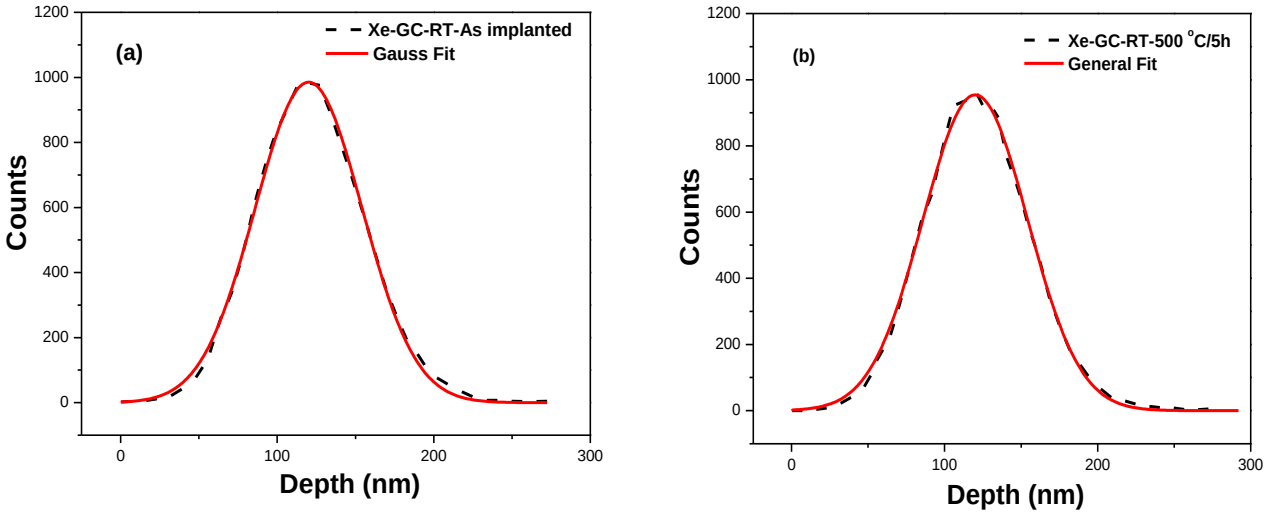

Figure 3. The MATLAB program fitted to the sample (a) implanted at RT, (b) annealed at 500 °C.

4. Conclusion

The diffusion behavior of Xe in GC was investigated using Rutherford backscattering spectrometry (RBS). RBS depth profiles obtained after annealing the sample at 300 °C – 800 °C showed that there was no diffusion of the implanted Xe into the bulk or towards the surface of GC. This non-diffusion was attributed to the presence of defect traps acting as a diffusion barrier towards the surface of GC. At the highest annealing temperatures (900 °C – 1000 °C), there was a movement of Xe into the bulk of the GC with a new peak (indicating segregation) formed within the GC bulk. The lack of diffusion of Xe in GC shows that glassy carbon is a good containment material for xenon.

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References

[1] Noda T, Inagaki M, and Yamada S 1968 A Comment on the Structure of Glassy Carbon *Bulletin of the Chemical Society of Japan*. vol. 41, no. 12. pp. 3023-3024

[2] SPI Supplies 2009 *Glassy Carbon Product Information* pp. 1–5. www.2SPI.com accessed Nov. 10, 2016

[3] Krause H 1972 *Storage of radioactive waste,* vol. 59, no. 9, pp. 412-417

[4] Wiley J & Sons Ltd 2010 *Radionuclides in the Environment* ISBN 978-0-470-71434-8

[5] Garland WJ 2003 *Xenon Effects on Nuclear Reactor Reactivity 4D03 course notes*, pp. 1-6

[6] Roggenkamp PL 2000 *The Influence of Xenon-135 on Reactor Operation* pp. 49-56

[7] Malherbe JB, Selyshchev PA, Odutemowo OS, Theron CC, Njoroge EG, Langa DF,

Hlatshwayo TT 2017 Diffusion of a mono-energetic implanted species with a Gaussian profile *Nucl. Instrum. Methods Rel. Phys*. **406** 708-713

[8] Ziegler J 2012 SRIM computer code-2012 www.srim.org. accessed May 24, 2017.

[9] Odutemowo O S, Malherbe J B, Theron CC, Njoroge EG, Wendler E 2016 *"In-situ RBS studies of tium implanted glassy carbon,"* Vacuum, vol. 126, no. March, pp. 101–105