Migration behaviour of selenium implanted into polycrystalline-SiC

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Abstract. The migration behaviour of selenium (Se) implanted into polycrystalline SiC was investigated using Rutherford backscattering spectrometry (RBS). Se ions of 200 keV were implanted into polycrystalline SiC samples to a fluence of 1×10^{16} cm^{-2} at room temperature. Some of the implanted samples were annealed in vacuum at temperatures ranging from 1000 to 1500°C in steps of 100°C for 10 hours. No diffusion was observed at annealing temperatures up to 1300°C. Diffusion of Se was observed after annealing at 1400°C and increased with annealing temperature. This diffusion was accompanied by a peak shift towards the surface and loss of implanted Se. From fitting of the Se profiles, diffusion coefficients of 8.0×10^{-21} and 1.1×10^{-20} m^2s^{-1} were estimated at 1400 and 1500°C, respectively .

1. Introduction

Silicon carbide (SiC) is considered as one of the few lightweight covalently bonded ceramics with interesting properties, such as a low thermal expansion coefficient and high thermal conductivity, mechanical strength and hardness [1]. The outstanding properties of SiC, make it suitable for applications in the petrochemical and specifically, for the purpose of this work, the nuclear industries [2]. The safety of modern nuclear reactors depends on the retainment of all the radioactive fission products that may leak into the environment during its operation [3]. In the Pebble Bed Modular Reactor (PBMR) which is one of Very High Temperature Reactors (VHTR), the containment of fission products (FP) within Tristructural-isotropic (TRISO) fuel particles is critical to the successful and safe operation of the reactor. The SiC layer is a very important layer in these particles because it has a number of very crucial functions, such as structural support and acting as the main fission products barrier [4][5].

Selenium (Se) is a non-metallic element with atomic number 34. It has many radioactive isotopes such as ^{72}Se, ^{75}Se, ^{79}Se, ^{80}Se and ^{82}Se. ^{79}Se is a component of spent nuclear fuel, and is found in high-level radioactive wastes resulting from processing spent fuel associated with the operation of nuclear reactors and fuel reprocessing plants. The health hazards of ^{79}Se come from the beta particles emitted during its radioactive decay, and the main concern is associated with the increased likelihood of

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inducing cancer [6]. ^{80}Se is one of the stable isotopes, the most prevalent, comprising about half of natural selenium [6]. It is both naturally occurring and produced by fission [7].

The extremely low diffusivities for impurities in SiC is one of the reasons SiC is used as the fission product barrier in TRISO fuel [8]. The migration behaviour of fission products such as strontium, iodine, cesium and silver in SiC at temperatures above 1000°C have been studied extensively [9]. There is no reported information on the migration behaviour of selenium in SiC which is important in order to ensure the efficiency of SiC layer.

In this study, we investigate the migration behaviour of ^{80}Se implanted into polycrystalline 3C-SiC at room temperature to a fluence of 1×10^{16} cm^{-2} at temperatures above 900°C.

2. Experimental procedure

Polycrystalline 3C-SiC wafers from Valley Design Corporation were used in this investigation. Se ions with energy of 200 keV were implanted into the wafers to a fluence of 1×10^{16} cm^{-2} at room temperature. The implantation was performed at the Friedrich-Schiller-University Jena, Germany. Some of the implanted samples were isochronal annealed in vacuum using a computer controlled Webb 77 graphite furnace at temperatures ranging from 1000 to 1500°C in steps of 100°C for 10 hours. Se profiles of the as-implanted and annealed samples were monitored using Rutherford backscattering spectrometry (RBS) of the Van de Graaff accelerator at the University of Pretoria, which uses certain principles of operation [10]. RBS was performed at room temperature using He^{+}-particles with energy of 1.6 MeV. The beam current was approximately 15 nA. 8 μC was collected per measurement. The RBS spectra were converted to depth in nm using the energy loss data and density of pristine SiC (3.21 gcm^{-3}). The depth profiles were fitted to a Gaussian function to extract the projected ranges (R_p) and stragglings (ΔR_p) for each sample and also to the solution of the Fick diffusion equation for a Gaussian as-implanted profile to extract the diffusion coefficients [11].

3. Results and discussion

In Fig. 1, the Se depth profile of as-implanted sample is compared with that one simulated using TRIM 2012 software [12] assuming a displacement threshold energy (E_d) of 20 eV for C and 35 eV for Si [2]. The experimental projected range (R_p) of 87.7 nm was slightly lower than the theoretical value of 89.6 nm. The value obtained is within the experimental error of the RBS measurements about 2% and the uncertainties of the SRIM simulations. The experimental straggling (ΔR_p) value is about 11% larger than that obtained by theoretical simulation viz. 29.9 and 26.5 nm. This discrepancy in the ΔR_P might be implies to the fact that re-distribution of Se is already taking placed during implantation process. The implanted selenium profile is almost a Gaussian distribution with the kurtosis (β = 2.9) and skewness (γ = 0.28). For a true Gaussian distribution (β = 3) and (γ = 0). What is also evident in Fig. 1 is that the maximum damage of about 1.3 dpa is at about 70 nm below the surface as compared to the experimental R_p of 87.7 nm. If one assumes that 0.3 dpa amorphises SiC [13], it is quite clear that 125 nm layer of SiC from the surface is amorphized. From these results it is quite clear that the majority of implanted Se is embed in the amorphous SiC.

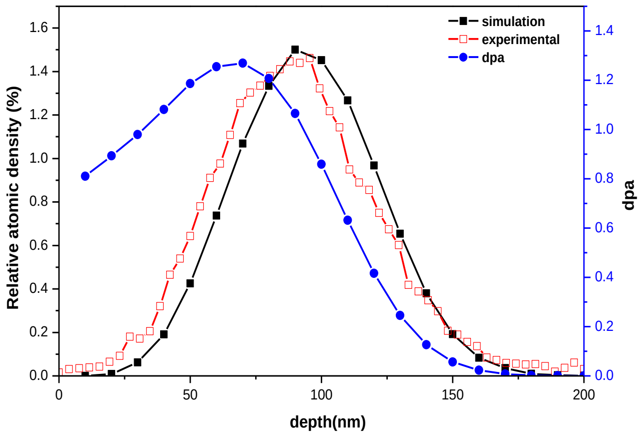

Figure 1. The RBS depth profile of Se implanted into SiC, TRIM2012 simulation and damage profile.

To investigate the migration behaviour of Se in polycrystalline SiC, the implanted samples were subjected to sequential isochronal annealing at temperatures ranging from 1000 to 1500°C in steps of 100°C for 10 hours. The Se depth profiles obtained from RBS before and after annealing are shown in Fig. 2. Neither a change in implanted Se profile nor broadening was observed after annealing at temperatures from 1000 up to 1200°C. These indicated the lack of detectable diffusion after annealing at these temperatures. The RBS profiles for the 1300°C samples indicated a small broadening of the profile and shift of the peak position of the Se profile. However, both were within the experimental error of the depth scale of our RBS measurements. For the 1400°C and 1500°C annealed samples there were measurable (only just for the 1400°C sample) broadening of the profiles and shift of the peak positions towards the surface (see Fig. 3(a) for the latter). Broadening of the profile is an indication of Fickian diffusion of the Se [11]. What was also noticeable was a general decrease in the heights of the profiles. To quantify this, the total integrated counts of the RBS Se signal (counts) were taken. The results are shown in Fig. 3(b). There was also a very slight asymmetry near the surface (i.e. $x = 0$) in the Se profiles at these two temperatures. This is due to evaporation into the vacuum of the Se atoms which diffused to the surface. The boiling point of Se is 685°C is significantly less than the annealing temperatures.

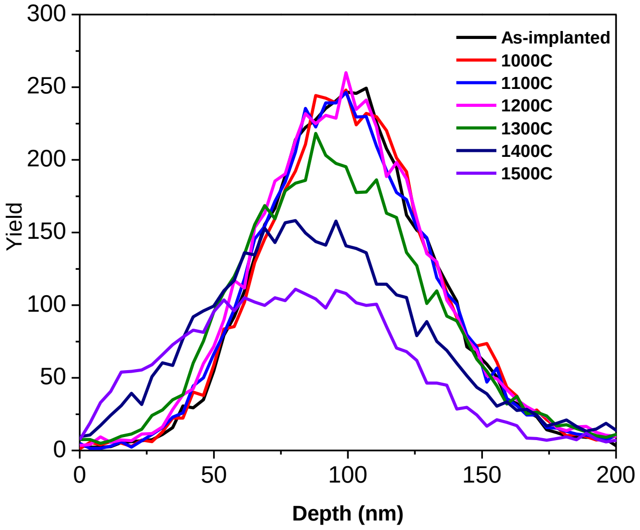

Figure 2. Depth profiles of selenium implanted in 3C–SiC at room temperature and after sequential isochronal annealing from 1000 to 1500 °C for 10 hours.

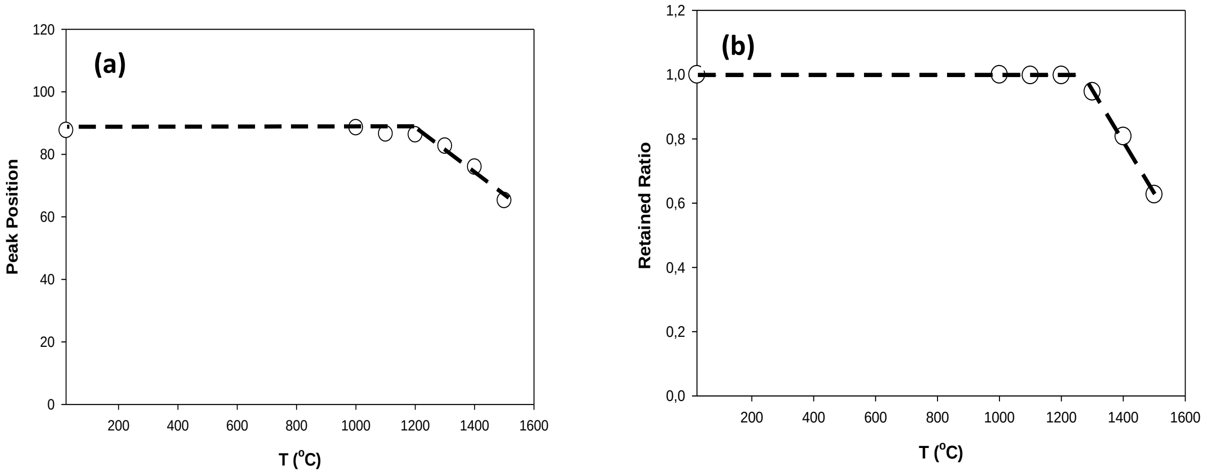

Figure 3. (a) The peak shift (b) retained ratio (calculated as the ratio of the total integrated counts of Se after annealing to that of as-implanted) of the Se profile as a function of annealing temperature.

To extract the diffusion coefficient of Se in polycrystalline SiC, the Se depth profiles obtained from RBS were fitted to the solution of the Fick diffusion equation for Gaussian as-implanted profile and with a perfect sink at the surface (see Fig. 4) [11]. The diffusion coefficients of $(8.0 \pm 0.24) \times 10^{-21}$ and $(1.1 \pm 0.33) \times 10^{-20}$ m^2s^{-1} were extracted at 1400 and 1500°C, respectively. No previous Se diffusion in SiC data were obtained in literature hence the obtained diffusion coefficients were not compared with any literature values.

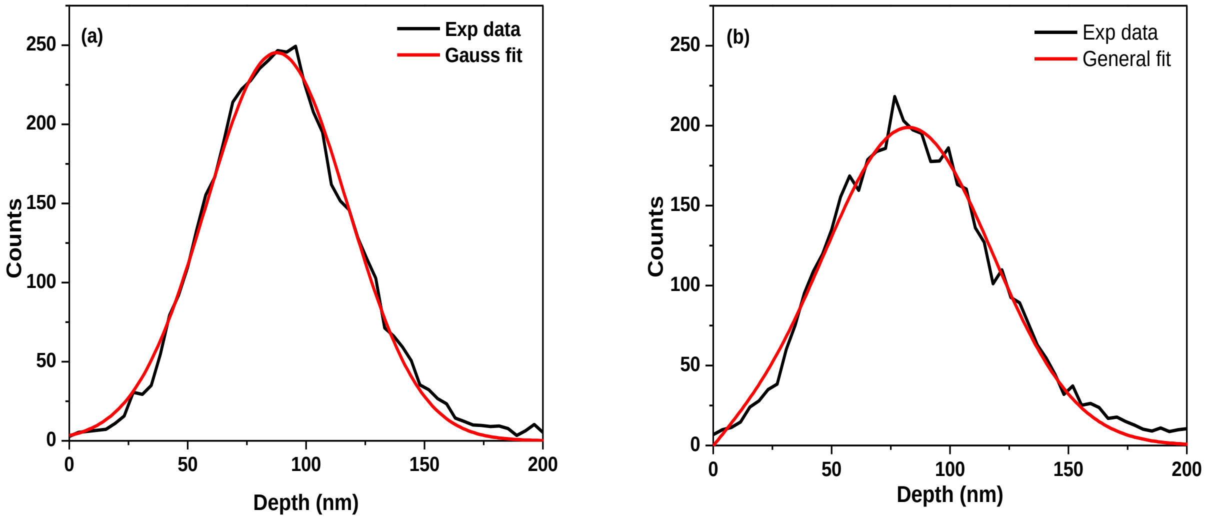

Figure 4. Example of the fitting of the diffusion equation solution to the depth profiles of the sample (a) as-implanted (Gaussian fit only) , (b) annealed at 1300°C

4. Conclusion

In this work, the migration behaviour of Se in polycrystalline SiC has been studied in terms of diffusion. Se^+ of 200 keV was implanted at RT to a fluence of 1×10^{16} cm^{-2}. The implanted sample was isochronally annealed at temperatures ranging from 1000 to 1500°C in steps of 100°C for 10 h. The effect of annealing on Se implanted on SiC and its migration behaviour was investigated using RBS.
No diffusion was observed after annealing at temperatures from 1000 up to 1300°C. The diffusion of Se began after annealing at 1400°C and increased with temperature. Also, the Se peak profile began shifting towards the surface after annealing at 1400°C and became more pronounced at 1500°C. This shift was accompanied by loss of Se from the surface. Significant loss, viz. about 40%, was observed at 1500°C. From fitting of the Se profile in the annealed samples, diffusion coefficients were extracted for the samples annealed at 1400°C and 1500°C.

Acknowledgement

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