The effects of implantation temperature and annealing on glassy carbon implanted with Se ions

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Introduction

The management of high-level nuclear wastes (HLW) has always been a significant problem facing the nuclear energy industry. These challenges have negatively affected the acceptance of nuclear energy as a good source of a clean and stable form of energy. A common practice of HLW storage is direct disposal via wet storage systems, dry storage systems, and permanent geological repositories. Currently, there is a limited operational geological repository to dispose HLW permanently. Typically HLW can be stored for a longer period in dry storage systems than in wet storage systems. The demand for dry casks of longer lifespan (40 to 50 years) could be in high demand in the nearest future. Therefore, the improvement and performance of the materials used for the fabrication of dry storage devices are vital.

Glassy carbon (GC) is a non-graphitizing form of synthetic carbon that exhibits some ceramics and glassy properties with those of graphite. It has some remarkable physical, chemical, and mechanical properties, making it a perfect alternative to be considered a canister for HLW storage. The properties of glassy carbon include: high hardness and strength, low reactivity, high-temperature stability (does not graphitize at temperatures up to 3000 oC) [1], resistance to radiation and chemical attacks, and impermeability by gases and liquids [1].

Previous studies on the migration behaviour of fission products in glassy carbon have shown that glassy carbon can serve as an alternative material to be considered for the fabrication of the dry cask needed for nuclear wastes management [2–11]. Among the various isotopes of selenium, 79Se is the only radioisotope that falls into the seven most long-lived radioactive fission products. It can be found in trace amounts (low yield of about 0.0487 % [12]) in uranium ores, spent, and reprocessed nuclear fuel. The release of 79Se into different environmental media would pose a health hazard from the β-particles emitted during its radioactivity. The main concern is the increased likelihood of causing cancer, especially when selenium is swallowed or absorbed into the body at a dose > 400 µg per day.

This study aims to investigate the effect of implantation temperature and annealing on the migration of selenium (Se) in glassy carbon.

Experimental

- **SIGRADUR G** from Hochtemperatur-Werkstoffe GmbH, Germany.

Implantation Parameter:

- Ion: Selenium (Se), Energy = 150 KeV, Fluence = 1×10^{16} cm^{-2}, Temperature = 23 °C and 200 °C.

Vacuum Annealing (Webb 77 graphite furnace**)**

Isochronal annealing with temperature range: 1000 °C to 1200 °C (in steps of 100 °C), Time: 5h.

Analytical Techniques:

- **SIMS:** 10 keV O^{2+}, scan area (150 x 150 µm^2), sputtered crater depth measurement using a DEKTAK 8 stylus profilometer
- **Raman spectroscopy**: Argon laser (532 nm wavelength), 50 × objective lens, Double subtractive mode:

Results

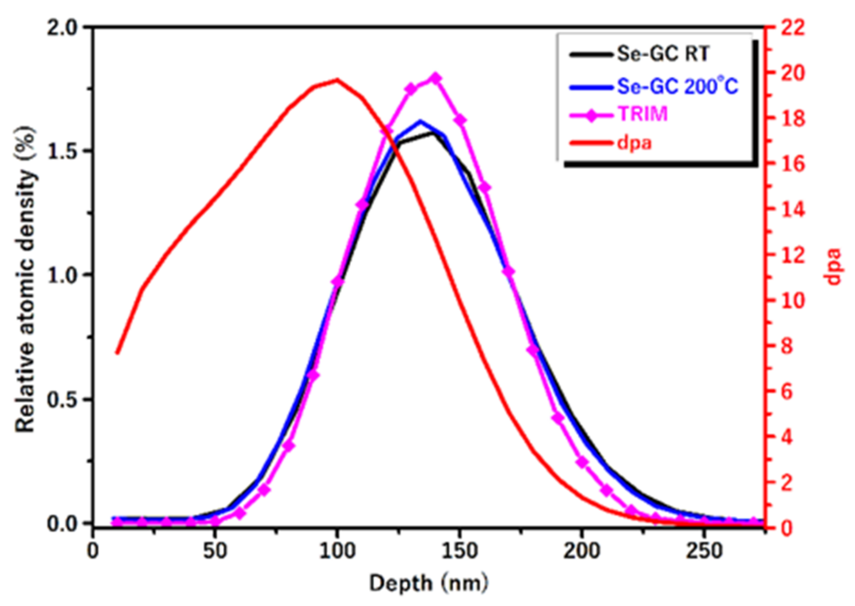


Figure 1: Depth profile of 150 keV Se ions implanted in GC at RT and 200 °C: Experiment and simulation

Table 1.0. The first four moments of Se ion distribution in GC

	Simulation	Experiment	
Moments	TRIM	RT	200 °C
R_p	132.80	129.56	130.10
ΔR_p	30.40	35.40	35.07
γ	0.18	0.29	0.28
β	2.86	2.95	2.94

$$dpa = \frac{{}^{v_{ac}}/_{ion(A)} x\, 10^8}{\rho_C(atomscm^3)} \times \varphi(ionscm^{-2}) \quad (1)$$

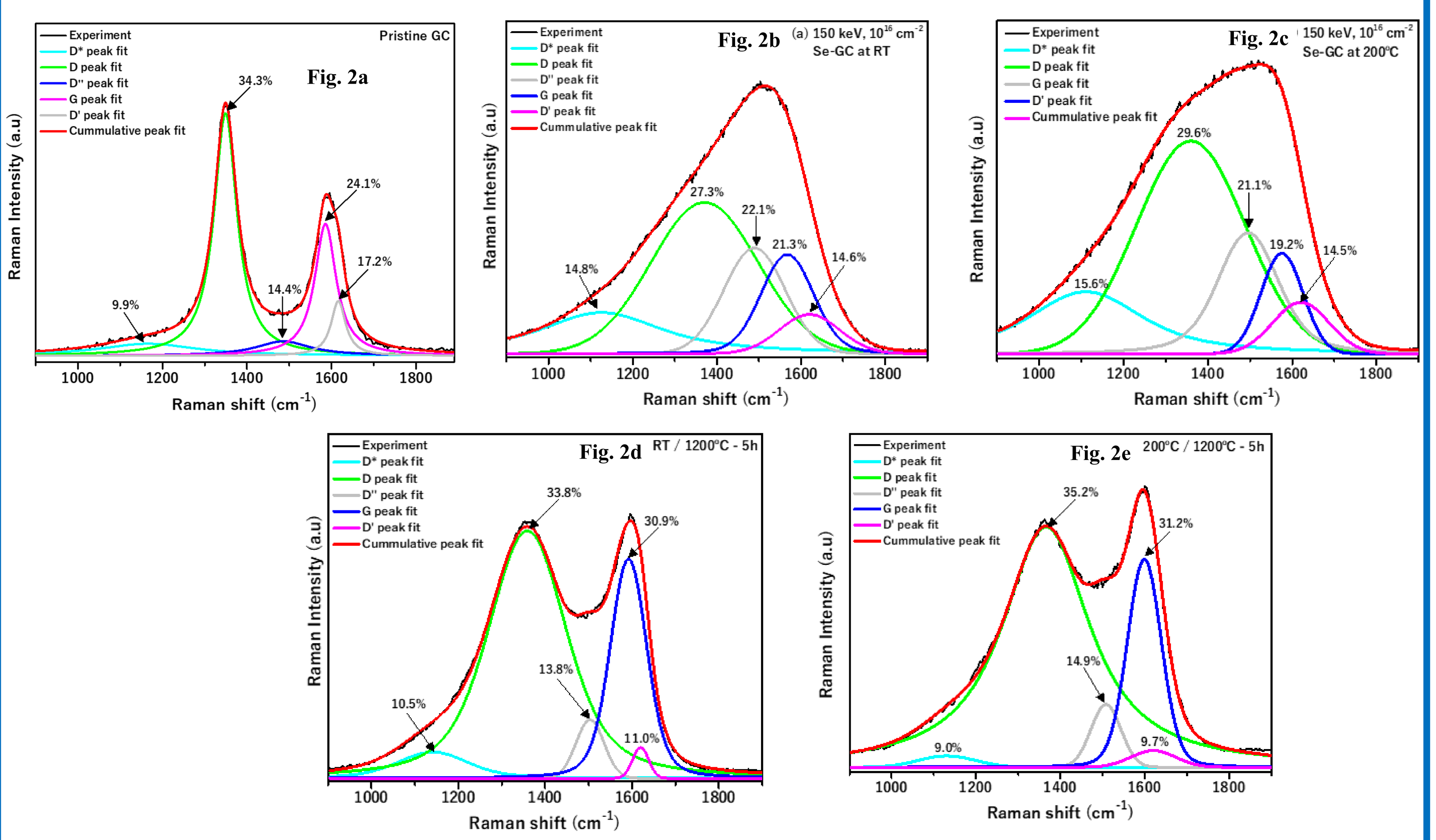
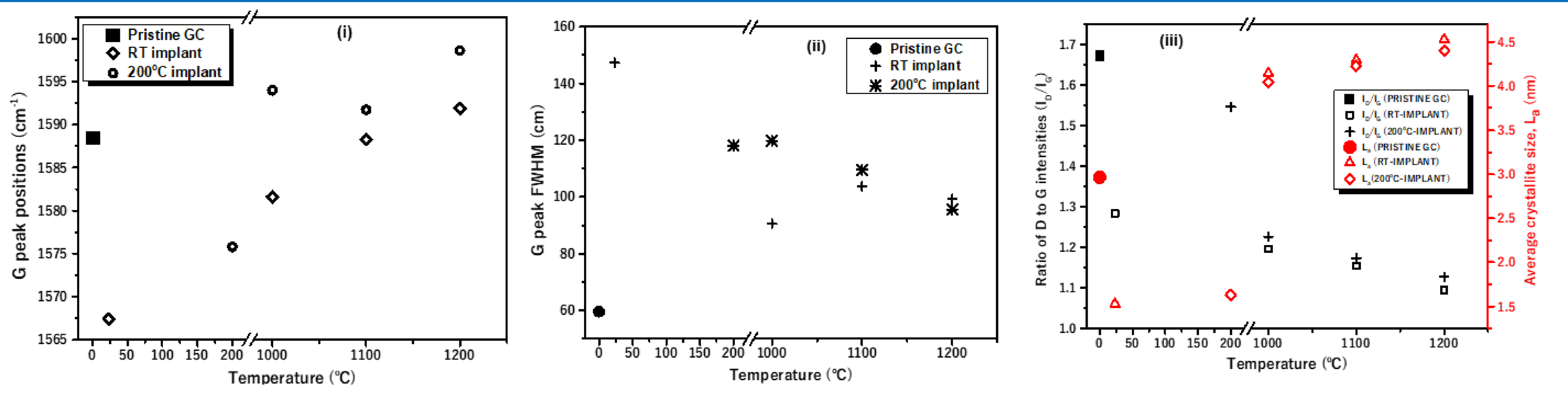


Figure 2. Raman spectra of (a) pristine GC (b) GC implanted with 150 keV Se^+ at RT (c) GC implanted with 150 keV Se^+ at 200 °C (d) RT GC sample annealed at 1200 °C & (e) 200°C GC sample annealed at 1200 °C, for 5h.



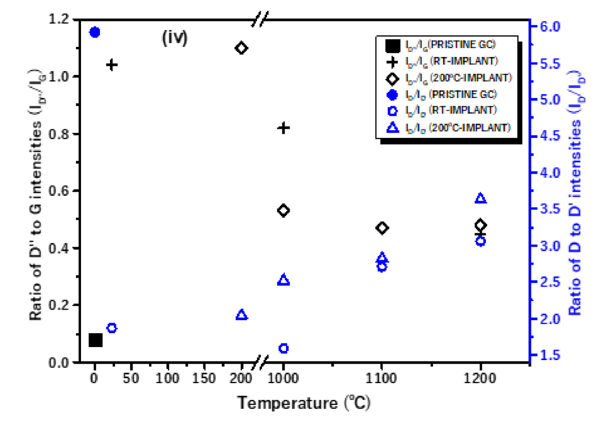
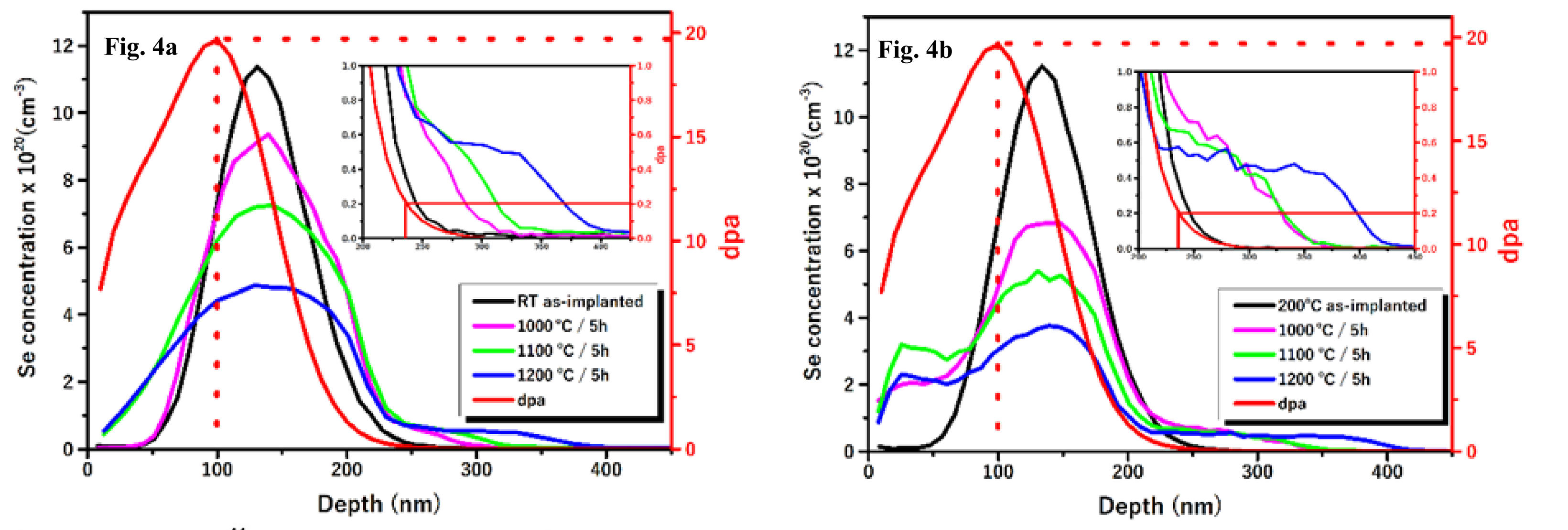


Figure 3. Raman fitting parameters (i) G peak positions (cm^{-1}) (ii) G peak FWHM (cm) (iii) I_D/I_G ratio and L_a (nm) (iv) $I_{D''}/I_G$ ratio and $I_D/I_{D'}$ ratio, of the acquired Raman spectra of glassy carbon implanted with 150 keV Se ions at RT and 200°C after isochronal annealing at 1000, 1100, and 1200 for 5h.



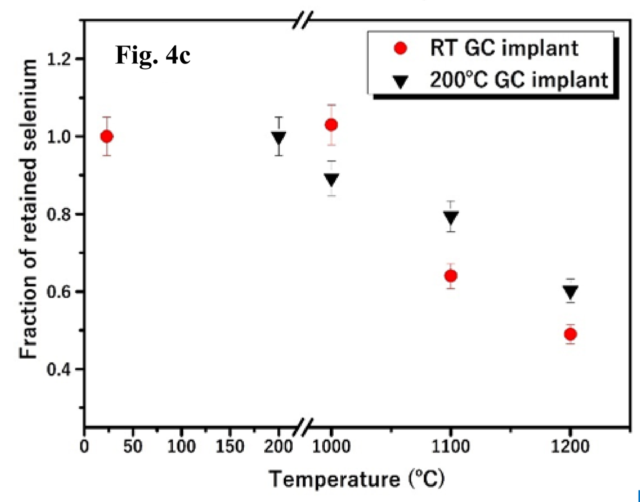


Fig.4. SIMS Se depth profiles for (a) RT as-implanted glassy carbon samples and after isochronal annealing at 1000-1200 °C (b) 200 °C as-implanted glassy carbon samples and after isochronal annealing at 1000 -1200 °C (b) Retained ratio of Se in the annealed RT and 200°C GC implanted samples.

Summary

- Implantation of Se resulted in defect accumulation in the GC structures of both RT and 200 °C samples, as shown by the Raman spectra in Fig. 2b & 2c.
- Annealing caused progressive healing of the defects, but the GC structure has not fully recovered after annealing at 1200 °C (see Figs. 2(d&e)) and Figs. 3 (i-iv).
- Boundary-like defects in the as-implanted samples were retained after annealing both sample types up to 1200 °C (See Fig. 3iv).
- Annealing the samples caused the migration of the Se atoms towards the surface and deeper into the bulk of the GC substrate in both RT and 200 °C GC implants, accompanied by a strong release of Se after annealing at 1100 and 1200 °C, as shown in Fig. 4(a&b).
- The migration towards the surface in the annealed 200 °C GC samples, accompanied by the segregation of some Se atoms at the surface, was attributed to the presence of a high concentration of defects traps, vacancies, and interstitials in the surface region the segregation was observed.
- The migration of Se atoms into the bulk of the GC substrate in both RT and 200 °C GC occurred following the annealing of the defects at the maximum implantation depth. The Se atoms that have migrated inside the bulk could get trapped in the pores in GC. A Se atom has an atomic radius of 0.19 nm and would successfully fit into a pore size of 0.3 nm.
- The difference in the migration behaviour of Se as exhibited in both annealed GC implant types would depend on the significance of the radiation damage introduced during the implantation process.

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