THE EFFECT OF SWIFT HEAVY ION IRRADIATION ON THE MICROSTRUCTURES OF Si_3N_4 AND AlN

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The development of novel materials with future nuclear applications is of ongoing concern. As part of this investigation we address two major issues concerning such applications. Firstly, improving the performance of structural materials in the reactor core for new and existing reactors and also the processing and storage of nuclear waste. The focus of this investigation is on Si_3N_4 and AlN which are under consideration as candidate materials for inert matrix fuel hosts. These materials will experience high levels of different types of radiation in the reactor core such as fission fragments and α-radiation. Due to the convenient similarities in mass and energy the effects of fission fragments are simulated by means of swift heavy ion implantation and α-radiation by He implantation. The combined effects of SHIs and He on these materials is also studied, since there is some indication from literature that SHI irradiation may significantly modulate the behaviour of gaseous species in some materials. To simulate the effects if SHIs; Si_3N_4 and AlN samples were irradiated with Xe and Bi ions, with energies ranging from 167 MeV to 2.6 GeV and temperatures ranging from LNT to 700 °C. The effects of α-particles were simulated with 20 keV He. The He irradiated samples were also subsequently annealed at 600 °C for 20 minutes to facilitate diffusion and agglomeration. The microstructure of the annealed samples was investigated by scanning electron microscopy (SEM) and transmission electron microscopy (TEM) before and after SHI implantation. In general, the results showed that latent ion tracks are formed in Si_3N_4, but not in AlN.

Introduction

Recent studies have shown that swift heavy ion irradiation may significantly modulate hydrogen and helium behaviour in certain materials [1, 2]. This phenomenon is of considerable practical interest for various ceramics and semiconductors, specifically for candidate materials for use as inert matrix (IMs) fuel hosts and for coatings for structural materials in the nuclear reactor.

Inert matrix (IM) fuel hosts have been suggested as a means of processing transuranic waste products resulting from the nuclear fuel cycle [3].

These materials accumulate helium via (n, α) reactions and will also be subjected to irradiation by high energy fission fragments in the nuclear reactor environment [3].

Two materials which are part of a larger group of nitride ceramics which are currently considered as candidate materials for IMs are Si_3N_4 and AlN. [4, 5]. The physical properties of these materials make them well suited to reactor conditions [4, 6]. However, to prove the viability of these materials for nuclear applications their radiation stability must be tested. In this investigation, swift heavy ions (SHIs) are therefore used to simulate the effects of fission fragments (FFs) on the microstructure of these ceramics. The effects of α-particles irradiation is simulated by means of He ion irradiation.

In addition to the analysis of the radiation stability of these nitride ceramics it is also advantageous to study the latent tracks, if present. The mechanisms involved in the process of latent track formation, through electronic energy deposition, in radiation resistant materials are still unconfirmed it is therefore also important, from a fundamental point of view, to obtain experimental data from a wide range of materials so that the true mechanisms involved in track formation can be determined.

Experimental

Commercially available polycrystalline Si_3N_4 and AlN were used as target materials in this investigation.

To simulate the effects of SHIs the samples were irradiated with Xe and Bi ions, with energies ranging from 167 MeV to 2.6 GeV and temperatures ranging from LNT to 700 °C. These irradiation parameters also allow for the determination of electronic energy deposition effects within the aforementioned materials.

To simulate the effects of α-particles the samples where irradiated with 20 keV He to a maximum fluence of $2 \cdot 10^{16}$ cm^{-2}. To determine whether SHI irradiation can modulate the agglomeration of He in these ceramics the He irradiated samples were subsequently irradiated with 167 MeV Xe to varying fluences. The He and He + Xe samples were also annealed at 600°C for 30 min under flowing argon.

TEM lamellae were produced by means of a FEI Helios Nanolab FIB-SEM, which was also used for SEM analysis. The lamellae were investigated with either a JEOL 2100 LaB_6 or a JEOL ARM 200F TEM both operated at 200 kV.

Results and Discussion

Latent ion tracks were not observed in any of the AlN analysed in this study. The polycrystalline AlN samples used in this study do however contain Y-Al-O based impurities, latent tracks were observed in these impurities where present. The AlN crystallites directly neighbouring these impurities are however unaffected. Latent ion tracks are observed in Si_3N_4 at all ion energies (i.e. at high and low levels of electronic energy transfer. The track diameters vary slightly with temperature and electronic energy loss (See Fig. 1–5).

Si_3N_4 has a much lower thermal conductivity compared to AlN. It is thought that thermal conductivity is one of the main parameters which determine

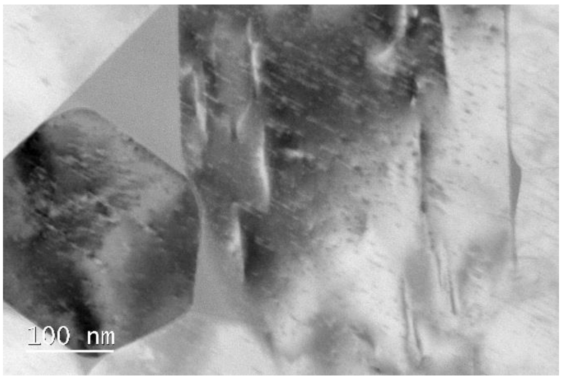

Fig. 1. Si_3N_4 irradiated with 167 MeV Xe to a fluence of $1{\cdot}10^{12}$ cm^{-2} at room temperature

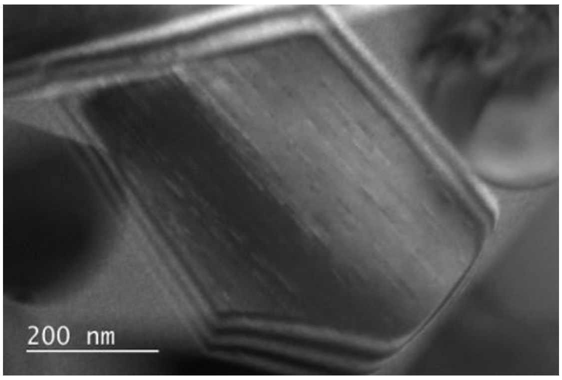

Fig. 2. Si_3N_4 irradiated with 220 MeV Xe to a fluence of $1.09{\cdot}10^{11}$ cm^{-2} at LN temperatures

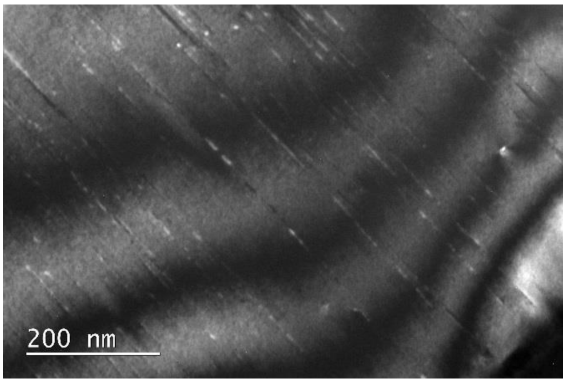

Fig. 3. Si_3N_4 irradiated with 220 MeV Xe to a fluence of $1.09{\cdot}10^{11}$ cm^{-2} at a temperature of 700°C

whether tracks will form in a material. The results of this study seem to support this conjecture since tracks are observed in the one nitride and not the other.

Other factors should however still be eliminated before a final conclusion can be made.

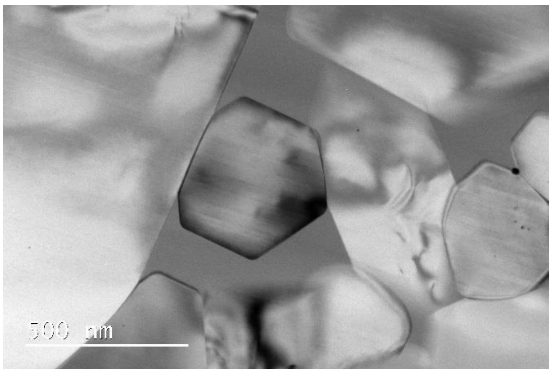

Fig. 4. Si_3N_4 irradiated with 500 MeV Xe to fluence of $4{\cdot}10^{10}$ cm^{-2} at room temperature

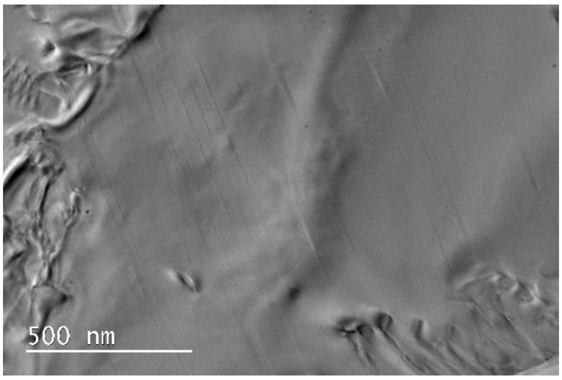

Fig. 5. Si_3N_4 irradiated with 700 MeV Bi to fluence of $1{\cdot}10^{10}$ cm^{-2} at room temperature

Conclusion

Latent tracks were observed in Si_3N_4 for all ions and energies, opposed to this no indication of latent tracks were observed in AlN. Preliminary results obtained from the samples irradiated at varying temperatures also suggest that temperature has little to no effect on the formation of latent tracks in Si_3N_4 and AlN.

The behaviour He in Si_3N_4 and AlN after SHI irradiation is as of yet still undetermined.

References

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