THE EFFECT OF SWIFT HEAVY ION IRRADIATION ON THE MICROSTRUCTURES OF YAP, YAG AND YIG

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The radiation stability of yttrium-based oxides as important ingredients of oxide dispersion strengthened alloys is a subject of extensive study. Oxide particles containing yttrium have been shown to be very stable at elevated temperatures. Of particular interest to investigations in this field is the radiation damage induced by swift heavy ion (SHI), which serves to simulate fission fragment impact. The aim of this investigation is to assess the effects of SHI irradiation on the microstructure of single crystalline YAP (Y-Al-Perovskite, $YAlO_3$), YAG (Y-Al-Garnet, $Y_3Al_5O_{12}$) and YIG (Y-Fe-Garnet, $Y_3Fe_5O_{12}$). The samples used in this investigation were irradiated with Ar, Bi, Kr and Xe ions with energies ranging from 46 MeV to 2.6 GeV to fluences in the range of 10^{10} - 10^{14} cm^{-2}. A subset of these samples was irradiated at temperatures ranging from LNT to 700 °C. The overall intention of these experiments was to determine the threshold stopping power for latent track formation and the influence of increasing stopping power and irradiation temperature on track morphology, employing Transmission Electron Microscopy techniques. Since these materials are similar in elemental composition but differ in crystal structure, the differences in microstructure of latent tracks is very informative when considered within the framework of the thermal spike model.

Keywords: electron microscopy; swift heavy ions; inelastic thermal spike; YAP; YAG; YIG.

Introduction

ODS steels are novel material consisting of a steel matrix with oxide particles dispersed throughput the material. The oxide particles in ODS steels may consist of various oxides such as: Al_2O_3, ThO_2, SiO_2, TiO_2, etc [1]. However, oxide particles containing Yttrium (Y_2O_3, $Y_2Ti_2O_7$, $Y_2Al_4O_9$, $YAlO_3$, $Y_3Al_5O_{12}$, $Y_3Fe_5O_{12}$) have been shown to be more stable than other oxides at elevated temperatures [2]. ODS steels are expected to have superior radiation resistance, superior strength (including at elevated temperatures) and good creep resistance due to dislocation pinning [3]. However, there is still uncertainty concerning the overall irradiation performance of ODS steels, specifically the effect of radiation on the oxide particles. Understanding the effects of radiation on the microstructure, which affect the mechanical properties of the particles and therefore also the overall properties of the steel is very important. If the properties and behaviour of the oxide particles are well understood the selection of the most stable oxide can be made with greater confidence than is currently possible. It is envisioned that ODS steels will be used in both fission and fusion reactors in future as structural materials in the reactor core [4,5]. These materials will be subject to large irradiation doses of different types, such as fission fragments, neutrons, electrons, protons, γ-rays, X-rays etc.

The focus of this investigation is on three yttrium-based oxides namely: YAP (yttrium aluminium perovskite, $YAlO_3$), YAG (yttrium aluminium garnet, $Y_3Al_5O_{12}$) and YIG (*yttrium iron garnet, $Y_3Fe_5O_{12}$*).

The accurate measurement of latent track parameters, in these materials, such as microstructural changes, diameter and depth using TEM techniques allows for the calculation of parameters such as the threshold electronic stopping power S_{et} for latent track formation to a high degree of accuracy. The experimentally obtained results are compared to what is predicted by the inelastic thermal spike model of *Toulemonde et al.* [6] and the analytical thermal spike model of *Szenes* [7]. The aim being the improvement of the current models, whilst gaining knowledge on the radiation performance of said materials.

Experimental

Bulk single crystalline $YAlO_3$, $Y_3Al_5O_{12}$ and $Y_3Fe_5O_{12}$ crystals were irradiated with Ar, Kr, Xe, and Bi ions with energies ranging from 46 MeV to 2.6 GeV to fluences in the range of 10^{10} - 10^{14} cm^{-2} close to room temperature. In some cases, an Al foil "degrader" is used to alter the stopping power of the incident ion. A subset of these samples was irradiated at temperatures ranging from LNT to 700 °C. Irradiation was conducted at the FLNR, JINR in Dubna, Russia with the IC-100 cyclotron and the DC-60 cyclotron at the IRC in Astana, Kazakhstan. TEM lamellas were prepared using a FEI Helios Nanolab FIB. Specimens were analysed with either a JEOL JEM 2100 LaB_6 or a JEOL ARM200F TEMs operated at 200 kV.

Results and Discussion

Transmission electron microscope (TEM) analysis of cross-sectional and planar YIG, YAG and YAP samples revealed the presence of latent ion tracks due to swift heavy ion irradiation. The appearance of the tracks in YAP and YAG/YIG differs to some degree, which suggests a difference in the nature of the tracks in these crystals. High angle annular dark field (HAADF) scanning transmission electron microscope (STEM) micrographs of planar sanples show that latent tracks in YIG (Fig 1.) and YAG (Fig. 2) are similar in size, but noticeably smaller in YAP (Fig. 3) In general it can be said that track diameters are largest in YIG and smallest in YAP at the same stopping powers with tracks in YAG closer in size to those in YIG as revealed by the measurements of track radii from a number of different ions (stopping powers) as shown in Figure 4.

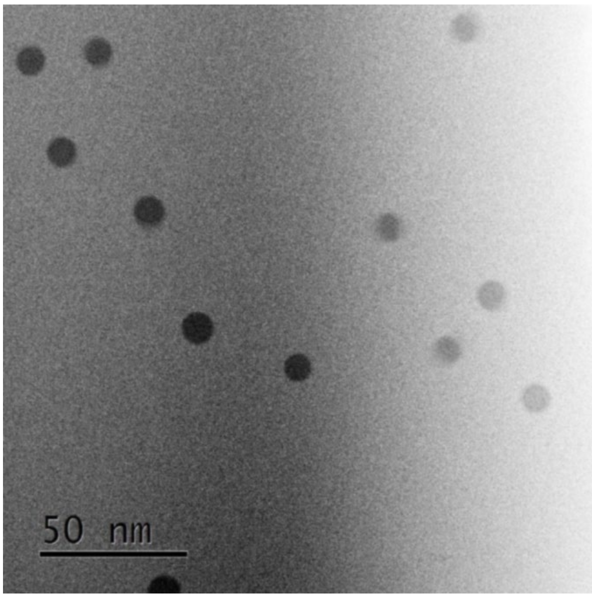

Fig. 1. High angle annular dark field STEM micrograph of YIG irradiated with 167 MeV Xe to a fluence of 5×10^{10} cm^{-2}

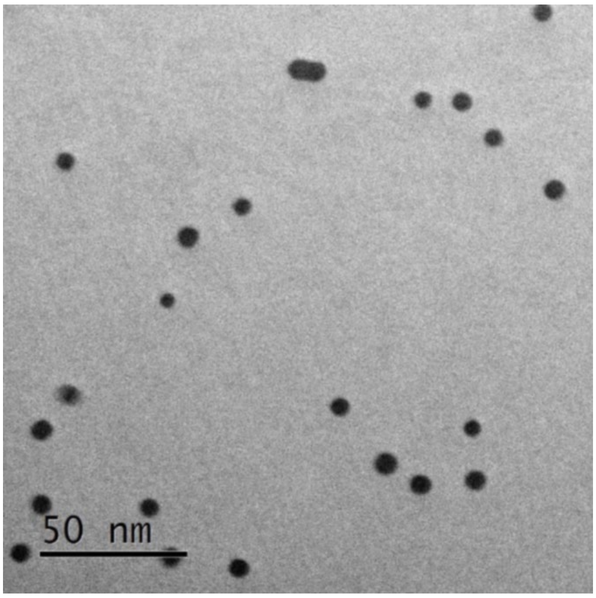

Fig. 2. High angle annular dark field STEM micrograph of YAG irradiated with 167 MeV Xe to a fluence of 5×10^{10} cm^{-2}

A significant number of track radii were measured at each data point. Some data points clearly have much larger errors in both the actual stopping power and track radius. This uncertainty is a direct result of using an Al foil as a stopping power degrader which introduces a larger energy spread and consequently a larger variation in track radii. The overall trend in track radii however is reasonably clear and clearly shows a difference in tracks sizes in the different materials.

The results of this investigation have shown some clear evidence that the complexity of the crystal structure plays a significant role in the recrystallisation

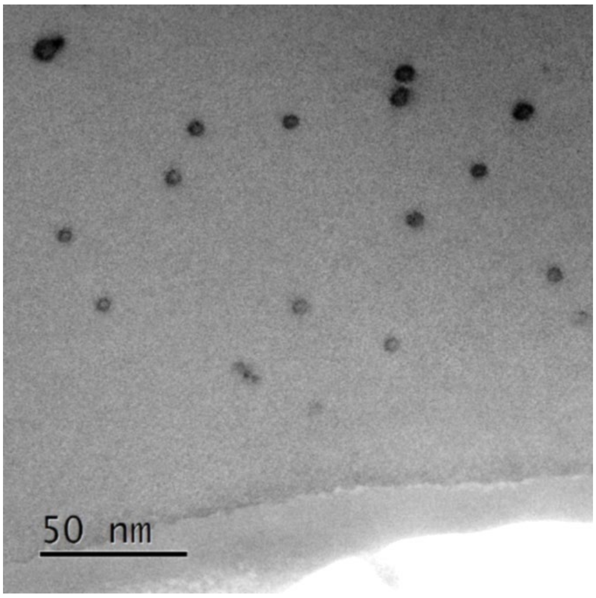

Fig. 3. High angle annular dark field STEM micrograph of YAP irradiated with 167 MeV Xe to a fluence of 5×10^{10} cm^{-2}

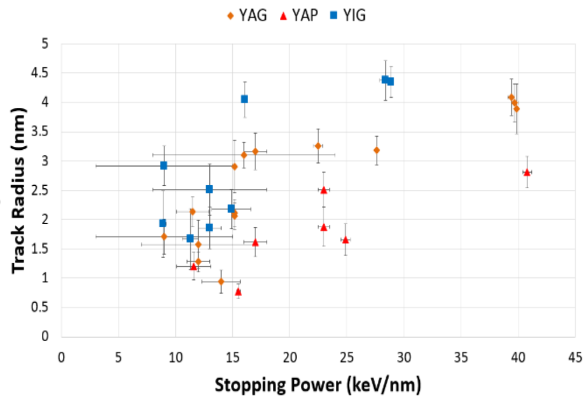

Fig. 4. Track radii vs. stopping power for YIG, YAG and YAP

behaviour of the specific crystal. The role of crystal complexity on the ability of a material to anneal out defects or recrystallise has been observed before in other materials [8, 9]. YIG and YAG have a body centred cubic crystal structures with the unit cell containing 160 atoms whereas YAP has a much less complex orthorhombic crystal structure [10, 11, 12]. YIG has a lattice parameter *a* = 12.37 Å and YAG a slightly smaller one at *a* = 11.95 Å. To overwhelm this ability of YAP, which is less complex, to recrystalise at a faster rate more energy must be added to the electron/lattice subsystems to induce a larger amount of remaining disorder. The results from this investigation shows a much smaller rate of diameter increase with stopping power when compared with other results for YAG and prediction of the thermal spike model [13, 14].

The results from the samples irradiated at different temperatures show that there is only a significant increase in track diameter at 1000 K for YAP, YAG and YIG. At 80 K and 300 K the increase in temperature required to reach the melting temperature is similar. At 1000 K the required increase is much smaller and therefore easier to attain. The relative increase in track size, with temperature, in all three materials is similar. Simulation with the thermal spike software shows the same trend as the experimental results. The thermal conductivities of YAP and YAG are very similar at higher temperatures, however the track diameters do not differ significantly, even though the stopping power in YAP is greater than that in YAG. This suggests that the thermal conductivity is not the main parameter determining the latent track size, at least at 1000 K. If the difference between experimental and iTS calculated values for YAG and YAP is due to some recrystallisation processes which occur in the quenched molten phase within the incident ion path, it can be concluded that YAP is more inclined to damage recrystallisation than YAG, thus the smaller track sizes at identical stopping powers. This result leans the argument towards the idea that crystal complexity or rather the, material specific, rate of recrystallisation plays a role in the resulting track diameter.

Conclusions

The results of this investigation have shown that amorphous latent ion tracks form YIG, YAG and YAP at stopping powers in the range from 11 to 41 keV/nm. Track diameters also increase with increasing stopping power and fit well with results from literature where available. YAP also consistently has latent ion tracks smaller in size than those in YAG and YIG at similar stopping powers. The results from samples irradiated at different temperatures suggest that approximately 10 keV/nm more energy is required to produce tracks, in YAP, that are similar in size to those in YAG and YIG.

There is clear indication from the results of this investigation that thermal conductivity plays a role in determining the tracks sizes in these materials. However, at the same time the results indicate that the complexity of the crystal structure seems to also play a significant role. At elevated temperatures (1000 K) the thermal conductivities of YAP and YAG become near identical, however the disparity in track sizes at identical stopping powers remains. YAG having a more complex unit cell presents larger tracks, whereas YAP with a much simpler unit cell presents significantly smaller tracks.

In terms of the use of these materials in ODS steels it is possible that structural modification such as, amorphization, dissolution and dispersion of the oxide particles may lead to detrimental stress accumulation in the metal matrix of ODS steels. The structural modification of the oxide particles in ODS steel may in turn lead to deterioration in the overall physical properties of the steel. It is therefore important to understand and predict the possible changes in these particles and to select the best oxide material based on experimental results. The results of this investigation also add valuable knowledge to the still developing field of latent ion track modelling in materials.

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