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Characterization of Radiation-Induced Clustering using Atom Probe Tomography in Nuclear Structural Materials

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1. Introduction

Various structural materials are used in a nuclear power plant, and Fe-based metals are mainly applied to the nuclear reactor components. For the reactor pressure vessel, low alloy steels are used due to the high integrity and economic feasibility, and reactor internals are basically composed of stainless steels with corrosion resistibility. Such structural metals suffer degradations originating from neutron irradiation during operation. The degradations include the change in mechanical properties, which are related to the microstructure evolution caused by irradiation.

The most widely used tool for the imaging irradiated microstructure is transmission electron microscopy (TEM) [1]. The composition of irradiation defects can be analyzed using X-ray spectroscopy (EDS) equipped in the TEM. However, composition characterization of the nano-sized irradiation defects in the matrix is limited due to the beam broadening of TEM and the overlapping of the probed volume during EDS analysis.

Recently, Atom probe tomography (APT) has been introduced to the characterization of irradiation defects [2,3]. APT provides sub-nano scale position of atoms and the chemical composition of a selected volume. This is very useful for the characterization of irradiation defects. In this work, we introduced APT technique to characterize the segregation of atoms in irradiated nuclear structural materials. The shapes and compositions of clusters were compared between the samples. This work can provide an application of APT on the defect characterization in irradiated materials in nuclear applications.

2. Methods and Results

2.1 Experiment

Two types of samples were prepared for APT analysis. The first sample is commercial-grade SS316 which were ion-irradiated at various temperatures. The ion source was Fe^{4+}, and the acceleration voltage was 2 MeV. The total ion fluence was estimated to be 5.6 × 10^{15} ions/cm^2. The region at a depth of 1 μm was used because of low penetration depth of ion irradiation. The SRIM code estimated that the dose was 4 dpa. The APT specimens were prepared using FIB and APT measurements were carried out in Pohang nano fab center.

The second sample was SA508 grade 4 low alloy steel which were neutron-irradiated in HANARO research reactor. The total fluence was 5.8 × 10^{19} n/cm^2 and the temperature was 288°C. The shape of the specimen was circular disk with the diameter of 3 mm.

The samples were prepared using FIB and APT measurements were carried out in Tohoku university lab in Oarai, Japan. because of its radioactivity by neutron irradiation

2.2 Characterization of irradiation defects

The atom positions within the 3D volume were reconstructed using the reference program from APT manufacturer. The solute clustering analysis was performed with a program developed at the Korea Atomic Energy Research Institute, which used the density-based spatial clustering of applications with noise (DBSCAN) [4] algorithm. Basically, DBSCAN defines a solute atom as a cluster if its k-th (K) nearest-neighbor solute atom is within a maximum distance (d_{max}). The application of DBSCAN to the cluster search in APT is well-described in the work of Stephenson et al. [5].

2.3 Si Clustering in Ion-irradiated SS

Fig. 1 shows the Si clusters that were identified in the APT samples. The values of d_{max} and K for DBSCAN were fixed to 1 nm and 5, respectively. The temperature effect on Si clustering is evident. The 400°C specimen has a number of large clusters composed by many Si atoms and the shapes of the clusters exhibit a wide variety. In the 300°C specimen, the majority of the clusters are small and their shapes are rather spherical. The 200°C specimen has few clusters with a very small size. Note that there is a difference between the number of clusters and the number of clustered atoms. Although the 400°C specimen has a larger number of clustered atoms, the total number density of clusters in the 300°C specimen is higher than that in the 400℃ specimen. The number of clusters is drastically affected by the scan parameters; thus, special attention must be paid to the comparison of the cluster analysis results when different parameters are applied.

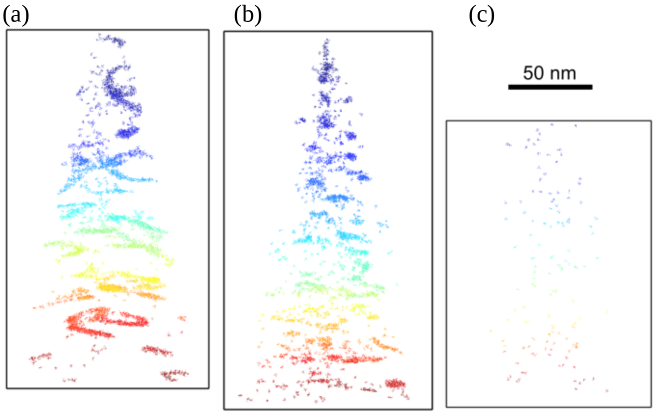

Fig. 1. Si clusters of the (a) 400℃, (b) 300℃, and (c) 200℃ specimens of ion-irradiated SS316. The colors indicate individual defects.

2.3 Si Clustering in neutron-irradiated SA508G4

Fig. 2 shows the neutron-irradiated SA508G4. The sample showed the Si clustering. Si atoms were localized at linear features which were expected to be dislocations. Majority of the clusters are spherical, and distributed evenly in the matrix, which is similar to the result of ion irradiation of SS316.

The composition analysis showed that clusters are mainly composed of Ni and Si. Because of low concentration of Mn, there was no clear evidence of clustering of Mn. The detailed analysis of composition is under progress.

Fig. 2. Si clusters of SA508G4 low alloy steels which was neutron irradiated. Black dots represent Si atoms.

3. Summary

In this study, microstructure analysis of clustering in irradiated nuclear structural materials was performed by APT analysis. In order to quantify the clustering, a method based on the DBSCAN algorithm was implemented. SS316 irradiated with Fe ions at above 300℃ caused significant clustering and segregation of Si and Ni at defect sinks. The neutron irradiated low alloy steel showed similar clustering of Ni and Si. The approach of using APT was demonstrated to be well suited for discovering the structure of irradiation defects and performing quantitative analysis in nuclear materials irradiated at high temperature.

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