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Radiation defects in nano-structured materials

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Abstract. We investigated defect formation within nanostructured materials using a transmission electron microscope (TEM) and ion accelerators. Controlling diffusion coefficient of the defects we have first observed changes in pattern of defect clusters, namely, the diffusion-limited reaction of defects in nanocrystals.

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1 Introduction

Among many new properties of the nanostructured materials we are interested in the behavior of defects in nanoparticles. When the size of the structural particle is comparable to or less than the defect free zone [1] annihilation would be superior to the production of defects in the particles. In this context, studies have been made putting an interest in mechanical properties of nanocrystalline metals [2]. As to the radiation induced defects, Rose *et al.* [3] have shown a correlation between grain size and defect density (size effect) in a statistical sense. In this study, we intend to reveal the defect formation in a single particle under irradiation. One of the aims of this study is to obtain damage free material against radiation damage utilizing the nanostructure. The experimental setup and the results are described in the following section. In order to support the understanding of the result we have developed a simulation program. That is presented in section 3. In the end we will realize the role of the diffusion process of point defects.

2 Experiments

2.1 Experimental setup

In order to introduce defects into the nanoparticles we used both low energy and high energy ion accelerators. The observation of the defects was carried out by using a transmission electron microscopy (TEM), JEOL JEM2000F, operating at 200 kV. The TEM is equipped with a low energy ion source, which enables us an in situ observation of the radiation effects [4]. Gaseous ions up to 35 kV are available to irradiate onto the TEM specimens at elevated temperatures. Another ion source is the JAERI tandem accelerator that can supply high energy heavy ions. Gold was used as the specimen in the present experiments because of its chemical stability. Nanoparticles were produced by the gas deposition method [5]. Au was evaporated by resistive heating from an alumina-coated boat under an Ar gas pressure up to 20 kPa. Evaporated Au was guided through a thin transfer tube, being carried by the gas flow, and deposited directly onto a TEM substrate, carbon coated micro-grids. This method, we consider, may minimize the stress in the sample during the sample preparation.

2.2 Experimental results

In the low energy irradiation experiments bubble formation in the particles was a characteristic phenomenon for the TEM observation as shown in Figure 1. Figure 1-a shows bubbles, shown as small white spots, produced by 15 keV nitrogen ion irradiation at room temperature. The ion fluence was 1.5×10^{16} ions/cm^2 and estimated ion range by the TRIM code was 18 nm. Small bubbles of about 1.5 nm in diameter are distributed uniformly over the particles. On the contrary, Figure 1-b shows a result when the irradiation temperature was raised to 673 K, at nearly equal ion fluence to that of Figure 1-a. Obvious difference of bubble formation can be seen concerning both the size and the distribution. In this case large bubbles of about 6 nm in diameter appeared and these exist rather in the central region than in the edge region. Observation by tilting the sample indicated that the bubbles lie not near the surface but the core side of the particle. Black spots appeared in the TEM observation were a feature of the high energy ion irradiation as shown in Figure 3. Among those spots triangle shaped ones are the stacking-fault tetrahedra (SFT). The irradiation was carried out with 100 MeV iodine ions at room temperature. The calculated dpa was about 2. When the irradiation temperature

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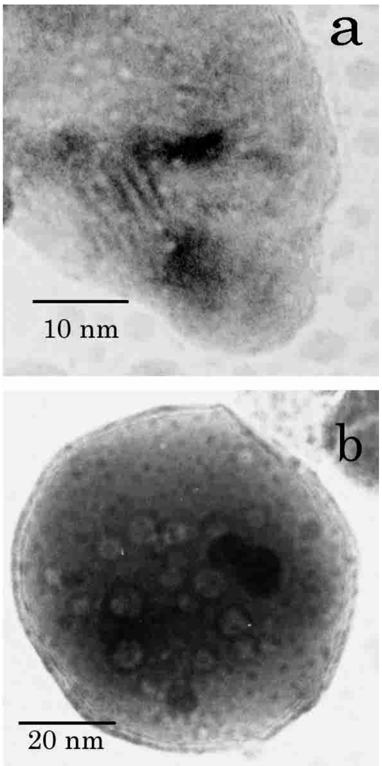

Fig. 1. TEM observation of bubbles in Au nanoparticles under N ion irradiation: -a at room temperature and -b at 673 K.

was increased to 473 K the black spots still appeared in large particles (typically 100 nm in diameter), but hardly appeared in small ones (less than 50 nm). These results imply that the diffusion limited phenomenon took place in the particles. Hence, in the next section we will develop a simulation program for the cluster formation and compare the results with the experimental observation.

3 Simulation

3.1 Simulation of the cluster formation

A tow-dimensional lattice program on which point defects walk randomly and make clusters was developed applying a cellular automata model [6] using Mathematica on IBM PC. The model and the rules of the motion of vacancy and the cluster formation are as follows.

A square shape of 100×100 lattice is the model of a particle, where at a time interval a point defect (*i.e.* vacancy) is created randomly on the lattice. All single vacancies move 1 step at an another time interval. When a vacancy meets with another ones, namely, takes a position of the von Neumann neighborhood, they combine with each other. Di-vacancy can move but tri-vacancy can not and be a nucleus of the succeeding cluster formation. Once clusters are formed, they do not separate any more. When they touch the boundary, they are annihilated. The cluster is formed artificially in square shape in the procedure otherwise it would be the tree-shape one that opposes the observation.

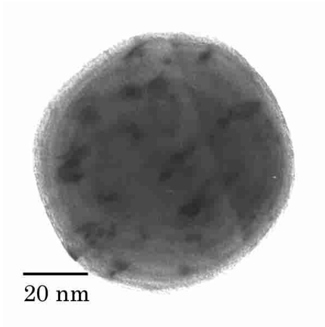

Fig. 2. TEM observation of defect clusters caused by high energy heavy ion irradiation. Triangle shaped black spots are the stacking-fault tetrahedra (SFT).

3.2 Results of the simulation

Figure 3 shows dependence of the pattern of clusters on diffusion coefficient of the point defect. In both cases 1200 point defects were created. The "relative" diffusion coefficient of lower case is 4000 times larger than that of upper case where the "relative" diffusion coefficient is determined by step frequency of the random walk during the interval of the defect creation, that is the time base in this model. In Figure 3-upper small clusters distributed uniformly over the lattice (particle), which resembles Figure 1-a, and in Figure 3-lower also resembles Figure 1-b. Hence, we see good agreement between Figure 1 and Figure 3, namely the experiments and the simulation, concerning the pattern of the clusters. In Figure 4 partitions (*i.e.* boundaries) are introduced in the original lattice, which is intended to simulate small size particles as compared to the previous ones. Other conditions are the same as those of Figure 3-lower. The result shows that at raised temperature cluster formation hardly takes place in small particles. This tendency has been also observed at high energy irradiation case as described in the previous section.

4 Concluding summary

Within a preliminary experiment, we have first observed a diffusion-limited reaction of defects in nanoparticles. Although a simplified model we found good qualitative agreements between the simulation and the experimental observation. This suggests that the simulation model with further improvements will be able to use to predict operation condition for defect free materials against irradiation. The radiation resistant materials will be one of the fruits of the nano-technology.

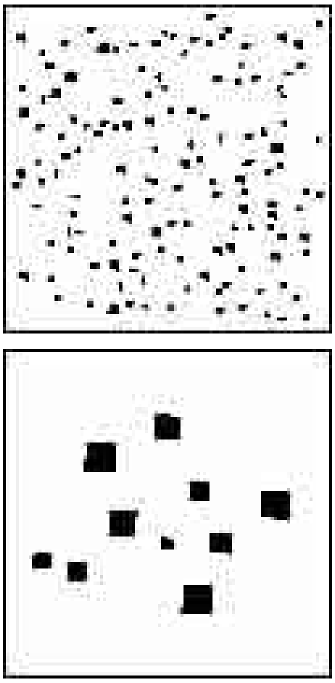

Fig. 3. Simulation of cluster formation using random walk model. The square frame represents the boundary of the particle. Clusters are shown in black spots. Upper/lower case correspond to small/large diffusion coefficient case, respectively.

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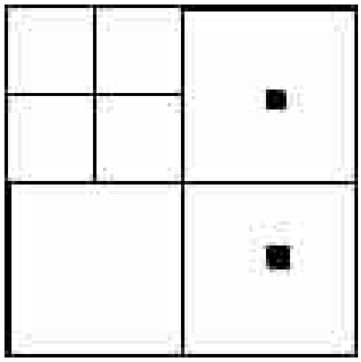

Fig. 4. Simulation of cluster formation for small sized particles by introducing partitions. The sink effect of the boundary for the cluster formation is exhibited at large diffusion coefficient case.

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