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Letter to the Editor

Hardening of Al–Cu–Mg alloy by energetic ion irradiation

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ABSTRACT

We irradiated Al–Cu–Mg alloy with 10 MeV iodine ions at room temperature and measured the surface microhardness. We analyzed the microstructure using a three-dimensional atom probe. Irradiation for 3.5 h led to an increase in hardness comparable to that obtained after 4 days of aging at 423 K. Precipitates of about 2.9 nm in diameter were distributed homogeneously over the irradiated region. The nanometer-sized precipitates produced by the irradiation caused a remarkable increase in hardness.

1. Introduction

There have been various investigations of the hardening method of aluminum-based alloys. Above all, a well-known method for hardening is the aging treatment of supersaturated alloys [1–3]. Typical precipitation for age-hardening of Al–Cu alloy progresses through the GP1 and GP2 zones that form during an early aging stage. This precipitation behavior greatly influences the increase in hardness but to obtain sufficient hardness by this method, the materials need to be kept at elevated temperatures for long periods of time.

On the other hand, when supersaturated alloys are irradiated with energetic particles, point defects (interstitials and vacancies) are produced and they enhance the diffusion of supersaturated atoms and their segregation. This phenomenon, which is called the radiation enhanced segregation, is caused by the interaction between point defects and supersaturated atoms. So far, the research of the radiation enhanced segregation has been promoted in the research field of hardening and embrittlement of reactor pressure vessels (RPV) in nuclear power plants [4]. In the study of RPV embrittlement, nanometer-sized precipitates of Cu-rich phase have been observed by irradiation with energetic particles, which cause the material hardening and embrittlement because precipitates of Cu-rich phase act as strong obstacles against the dislocation motion [5,6].

In this study, we applied the irradiation enhanced segregation caused by ion irradiation to control the hardness of a supersaturated Al–Cu–Mg alloy, which is generally known (or commercially used) as an age-hardenable "duralumin" alloy. Aluminum alloys have often been used as structural materials especially for test reactors. They are also candidate materials for fusion reactors because of their low induced radioactivity. Therefore, the study on hardness change of aluminum alloys by irradiation is important also in the field of the nuclear power technology. We report on the results of micro Vickers hardness measurements and three-dimensional atom probe observations for the Al–Cu–Mg alloy irradiated with 10 MeV iodine ions at room temperature.

2. Experimental procedure

Supersaturated Al–Cu–Mg alloy was selected for our experiments. The chemical composition of the specimens is listed in Table 1. Specimens were cut to $10 \times 10 \times 1\ mm^3$ in size using a micro-cutter. They were solution-annealed at 813 K in air and then quenched in water at 273 K. Specimen surfaces were polished using emery paper and buffing compounds just before irradiation.

The specimens were irradiated with 10 MeV iodine ions (I^{3+}) using a tandem accelerator at the JAEA–Takasaki. All irradiation was performed at room temperature. The beam current was about 150 particle nA/cm^2. The rise in temperature because of beam heating was negligibly small. To compare the hardness of the ion-irradiated region with that of the unirradiated region in the same specimen, about half the area of each specimen was irradiated and the other half remained unirradiated.

After irradiation, the Vickers hardness was measured near the boundary of the irradiated and unirradiated regions. The applied loads were 25, 50, 100 and 200 gf and the indentation time was 10 s for each indentation. We measured the indent-depth dependence of the change in Vickers hardness for each specimen. The indentation was performed at regular intervals of 0.5 mm on a straight line perpendicular to the boundary of the irradiated and unirradiated regions. The hardness was determined by averaging the results of more than 30 measurements.

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Table 1
Chemical composition of the Al–Cu–Mg alloy (wt.%).

Cu	Mg	Mn	Si	Fe	Zn	Cr	Al
3.5–4.5	0.4–0.8	0.4–1.0	0.2–0.8	0.7	0.25	0.1	Balance

The microstructure was analyzed using an atom probe at the Central Research Institute of Electric Power Industry. The specimens used for the atom probe analysis were the solution-annealed specimen, the specimen irradiated with iodine ions at a fluence of $1 \times 10^{15}/cm^2$ and the specimen aged at 423 K for 4 days. For the ion-irradiated specimen, an extremely small piece was sampled from the irradiated surface region and it was processed into a sharp needle like specimen using a focused ion beam (FIB) system. The solution-annealed specimen and the aged specimen were cut into rods and their tips were sharpened by electropolishing. The atom probe analyses were performed at 50 K with a pulse fraction of 15%.

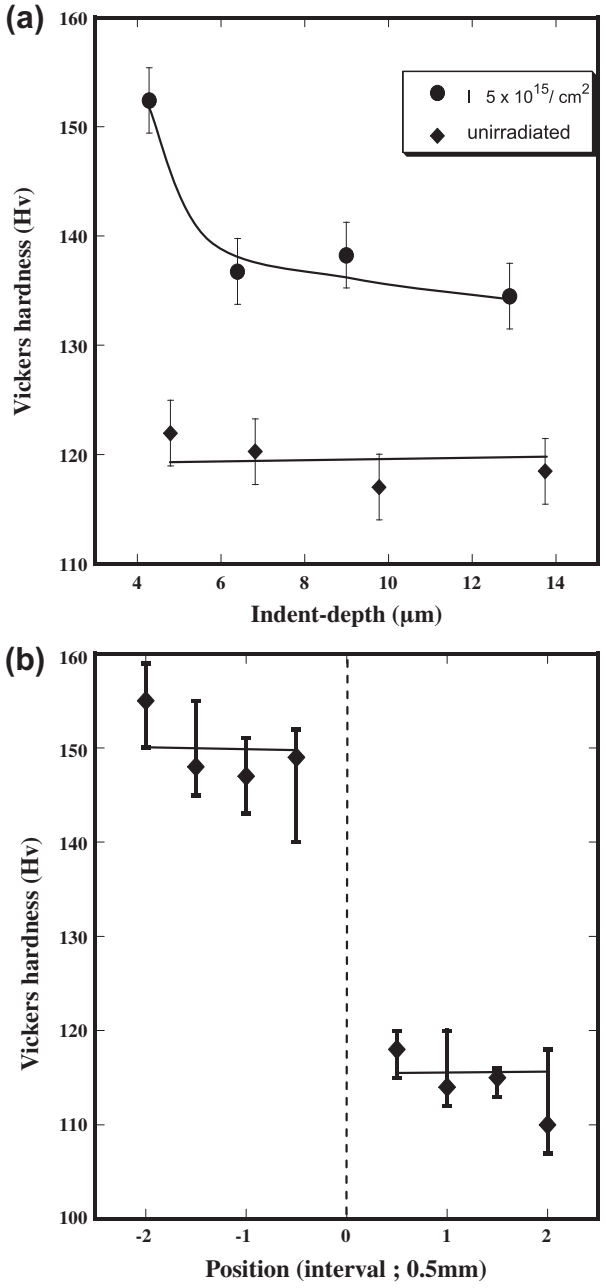

Fig. 1. (a) Indent-depth dependence of the Vickers hardness for the specimen irradiated with 10 MeV iodine ions. For comparison, the data for an unirradiated specimen are also plotted. (b) Relationship between Vickers hardness and measuring positions near the boundary of irradiated and unirradiated regions in specimens irradiated at a fluence of $5 \times 10^{15}/cm^2$.

3. Result and discussion

Fig. 1a shows the indent-depth dependence of the change in Vickers hardness for the specimen irradiated at a fluence of $5 \times 10^{15}/cm^2$. Data for the unirradiated specimen are also plotted. The Vickers hardness for the unirradiated specimen is not influenced much by the indent-depth, which indicates that the hardness is nearly constant over the observed depth. For the irradiated specimen, the value for the hardness is higher than that for the unirradiated specimen. The hardness is, however, not constant but decreases gradually with an increase in indent-depth. The depth dependence of hardness for the irradiated specimen is attributed to the fact that the energy deposited by 10 MeV iodine ions in the specimen distributes only to a depth of 4 μm below the surface. This has been calculated using SRIM 2009 software [7].

Fig. 1b shows the relationship between the Vickers hardness with an applied load of 25 gf and the indentation position near the boundary of the irradiated and unirradiated regions for the specimen irradiated at a fluence of $5 \times 10^{15}/cm^2$. The applied load of 25 gf corresponds to an indentation depth of 4 μm, where the elastic energy deposited by the ions reached a maximum value. The dashed line at the center of the figure represents the boundary of irradiated (left) and unirradiated (right) regions. It is clear that the Vickers hardness increases by 40 only in the irradiated region.

Fig. 2 shows the dependence of the change in Vickers hardness on the ion fluence with an applied load of 25 gf. The Vickers hardness increases significantly with an ion irradiation of only $3 \times 10^{14}/cm^2$ and then tends to be saturated at higher fluence.

Fig. 3 shows the change in Vickers hardness as a function of processing time for the specimens irradiated with iodine ions and for the specimens aged at 423 K and 453 K. For the specimen aged at 423 K, the hardness reaches a maximum value after 4 days and then decreases. For the specimens aged at 453 K, the time required to reach a maximum hardness is shorter than that for the specimen

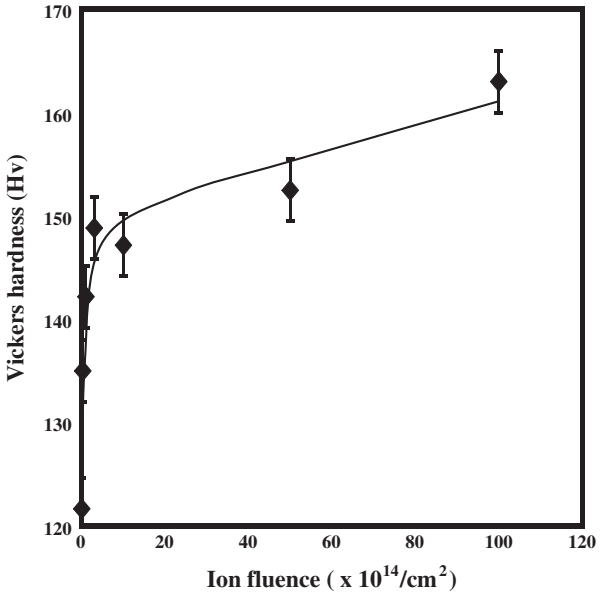

Fig. 2. Ion fluence dependence of Vickers hardness. The indent-depth is 4 μm.

aged at 423 K. The maximum value of the hardness, however, decreases with an increase in the aging temperature. Meanwhile for the specimens irradiated at room temperature, the surface hardness increases far more rapidly than that for the thermally-aged specimens and only 3.5 h are required to reach a Vickers hardness of 160, which is about the same as the maximum hardness value obtained when aged at 423 K.

The microstructure of the irradiated specimen was observed using transmission electron microscopy (TEM). We did not, however, find any morphology that corresponds to the GP zone, although this zone has previously been clearly observed in thermally-aged specimens. We then used the three-dimensional atom probe method to study the microstructure of the irradiated specimens at the atomic scale. The result of the atom probe is shown in Fig. 4. This figure shows distributions of highly concentrated regions of Cu, Mg and Si atoms in the solution-annealed specimen, the specimen irradiated at a fluence of $1 \times 10^{15}/cm^2$ and the specimen aged at 423 K for 4 days. Irradiation-produced precipitates were scarcely observed in the normal atom probe elemental map and, therefore, we used cluster analysis to extract the state of solute atom clustering. The parameters used for the cluster analysis are as follows: d is the maximum separation distance between solute atoms and N is the minimum number of solute atoms associated with a cluster. d and N were determined using the combined nearest neighbor distance distribution of the solute atoms and the Al atoms in the solution-annealed state, which gives a matrix with randomly distributed solute atoms. In the present analysis, the chosen parameters for d and N were 0.6 nm and 20, respectively.

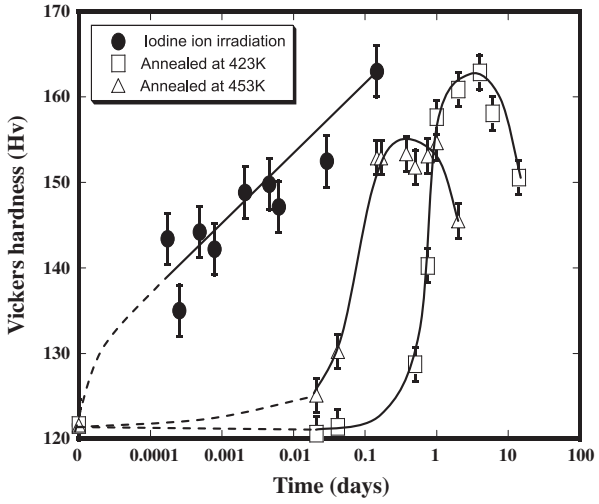

Fig. 3. Comparison of the change in Vickers hardness between irradiated specimens and specimens thermally aged at 423 K and 453 K as a function of processing time.

For the specimen aged over 4 days, needle-like Mg_2Si compounds and plate-like Al_2Cu compounds of several tens of nanometer in size are present. For the ion-irradiated specimen, many small precipitates of about 2.9 nm in diameter are distributed homogeneously in the irradiated region. The difference in precipitation between ion-irradiated specimen and thermally-aged specimen can be tentatively explained as follows; in thermally-aged specimen, the precipitation occurs through the long-range thermal diffusion of solute atom, resulting in coarse precipitates. On the other hand, in ion-irradiated specimen, the precipitates were produced by the radiation enhanced segregation. As a result, the precipitation occurs through the short-range diffusion, causing nanometer-sized precipitates. These precipitates act as effective obstacles for dislocation motion, causing a remarkable increase in hardness. More detailed analysis will, however, be needed for a complete understanding of the generation of the fine precipitates by the irradiation.

From these experimental results, we conclude that nanometer-sized precipitates, which are produced by short-time energetic ion irradiation at room temperature, cause a remarkable increase in the hardness of supersaturated Al–Cu–Mg alloys. These homogeneously distributed nanometer-sized precipitates have rarely been produced by conventional thermal processing. The present result

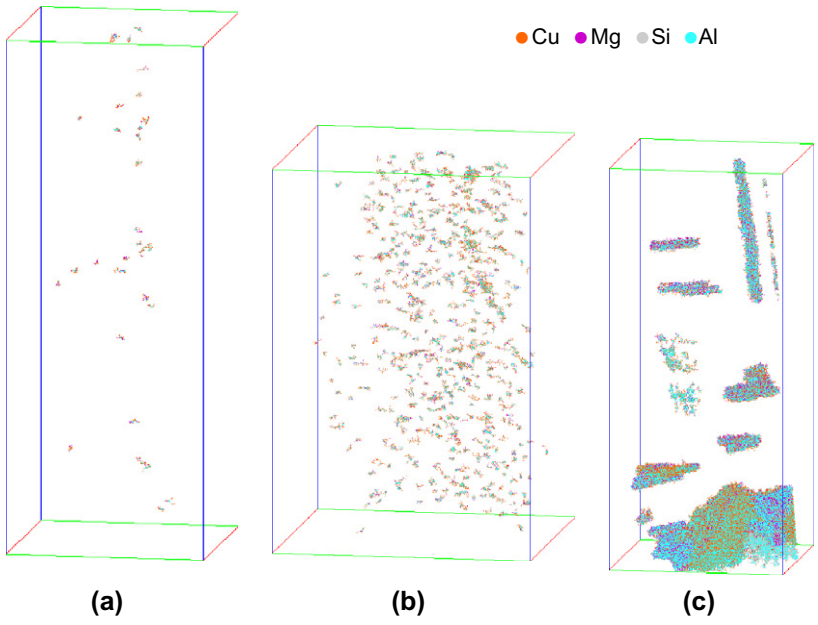

Fig. 4. Atom probe elemental mapping for (a) the specimen solution-annealed at 813 K for 4 h, (b) the specimen irradiated with iodine ions at a fluence of $1 \times 10^{15}/cm^2$, and (c) the specimen aged at 423 K for 4 days. Box sizes are: (a) $73 \times 73 \times 190$ nm^3, (b) $98 \times 98 \times 150$ nm^3 and (c) $65 \times 65 \times 150$ nm^3.

suggests that irradiation with energetic particles can be used as a tool for the effective control of the hardness of Al–Cu–Mg alloy.

4. Summary

Precipitation of solute atoms and the related change in hardness of the Al–Cu–Mg alloy using 10 MeV iodine ion irradiation was investigated using Vickers hardness measurements and three-dimensional atom probe analysis. Irradiation over 3.5 h at room temperature leads to an increase in hardness, which is comparable to that after 4 days of aging at 423 K. Atom probe analysis reveals that many irradiation-induced precipitates of about 2.9 nm in diameter mainly contribute to a remarkably rapid increase in hardness.

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