Application of three-dimensional electron tomography using bright-field imaging—Two types of Si-phases in Al–Si alloy

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Abstract

When a dilute amount of Si is added to Al, it results in the precipitation of Si-phases, either planar- and/or rod-type, depending on the ageing conditions. Observation of these phases had been carried out by TEM two dimensionally so far; nevertheless information of the thickness as well as the distribution had been neglected in the past. In this paper, a combination of electron diffraction, high-resolution transmission electron microscopy, and three-dimensional electron tomography was applied to characterize the morphologies and the orientation relationship of the Si-phases in an Al–Si alloy.

Keywords: TEM; Electron tomography; Si-phases; Al-Si alloy

1. Introduction

Conventional (two-dimensional) transmission electron microscopy (TEM) has been an essential technique to characterize materials in the nano-order for a few decades \cite{1,2}, and has been playing a key role is understand structures of dislocations, grain sizes, orientations, interfaces, and compositions. Although TEM has advantages of observing the internal structures of the specimen, it has difficulties in observing the structural details from the different depths. Recently, electron tomography (ET) has been applied for 3D structural characterization of a variety of materials \cite{3–12}. 3D-ET is a method to reconstruct a 3D object from a series of 2D projections acquired by tilting a TEM specimen within the column, in which higher tilts with smaller angular increment improve the spatial resolution of the reconstructed volume further \cite{7,8}.

In the case of materials science applications, care has to be taken due to their complex contrast mechanisms. Usually, in the case of bright-field TEM (BF-TEM) micrographs of thin amorphous materials, the incident electrons exhibit single-scattering processes, so that the contrasts within the micrographs vary monotonically with mass-thickness. However, in the case of crystalline materials, where the diffraction contrast typically dominates the image formation, the contrast within the micrographs does not vary monotonically. This makes it difficult to obtain a suitable tilt series of TEM images of crystalline specimens for the tomographic reconstructions. It has therefore been assumed that BF-TEM tomography is inappropriate for crystalline materials, and combination of tomographic technique with other imaging methods, such as energy-filtering TEM and/or high-angle annular dark-field-STEM (HAADF-STEM), \cite{7,8} are desirable as the solution for the 3D characterization of crystalline materials. These imaging techniques would strongly reduce the diffraction effects and fulfill the projection criterion even in the presence of highly crystalline materials.

The mechanical properties of materials strongly depend on their inner structures. Development of Al-based alloys with low weight, high stiffness and high tensile strength has been desired in the last decade because of the expectation of the improvement of structural efficiency of transportation vehicles and the reduction of fuel consumption. Within these Al-based alloys, Al–Si alloys have attracted much attention for fundamental studies on nucleation in solids \cite{13–18}. The system is a simple binary eutectic with limited
solubility of Si in Al. In fact, the solubility of Si in Al reaches a maximum 1.5 at% at the eutectic temperature (850 K), and it decreases to 0.05 at% at 573 K [19]. The equilibrium phase diagram of the binary Al–Si system shows that the mutual solubility of these elements is negligibly small at room temperature. Ageing of a supersaturated Al–Si alloy leads to precipitation of both plate- and rod-type pure Si-phases without formation of any intermediate metastable phase.

In this paper, 3D morphologies and orientation relationships of Si-phases in the Al–Si system are carefully examined by both high-resolution TEM and 3D-ET.

2. Experimental methods

2.1. Specimen

An Al–1.5 at% Si alloy was prepared from high-purity (99.99%) Al and high purity (99.9%) Si. The alloy was melted in air, cast into an ingot with dimensions of $17 \times 55 \times 120 \text{mm}^3$ and then homogenized at 843 K for 24 h. The ingot was solution treated at 850 K for 1 h and aged at 473 K for 5 h for the precipitation of Si-phases. For TEM studies, 3 mm diameter samples were punched from thin slices with 0.3 mm thickness and ground to approximately 0.1 mm thickness. A Gatan Model 656 Dimple Grinder and a Model 691 Precision Ion Polishing System were used to produce thin foils.

2.2. Transmission electron microscopy

TEM was performed on two different microscopes. Structures and morphologies of Si-phases were observed by a high-resolution TEM (JEM-4000EX, JEOL, Japan) and 3D-ET was carried out by a computer-controlled fully-digitized TEM (TECNAI-G20, FEI, USA) equipped with a specially designed high-tilt holder (Fischione Instruments Inc., USA). A large number of TEM parameters were controlled during the acquisition of the tilted series of projections by a cooled slow-scan charge-coupled device (CCD) camera: the defocus, the beam shift, the beam tilt, the angular step of tilts. In general, this consists of not only on the image resolution but also on the angular range and the quality of reconstructed volume are dependent not only on the image resolution but also on the angular range and the angular step of tilts. In general, this consists of collecting images at 1–2° angular intervals over an angular range of $-70°$ to $+70°$ [8,11]. It should be noted that the shape of the projected structure changes with the direction of projection. Therefore, 3D-ET requires fully automated and fully digitized TEM with an accurate tilt stage and a specially designed high-tilt specimen holder. In addition, it is necessary to consider that the increase of thickness contrast, i.e., the path length of the electron beam through the specimen, is twice the specimen thickness at $60°$ and three times the specimen thickness at $70°$. Although energy-filtering TEM tomography (EF-TEM-tomography) using Si-K or Si-L edges was expected to provide reconstructed Si phases in 3D volumes, 2D investigation of studying the EF-TEM images using these edges over the Al matrix showed weak contrast, and longer exposure time was necessary.

Once the acquisition of the tilt series was completed, the data sets were transferred to a UNIX workstation for fine tuning of the alignments using IMOD software [20]. 3D reconstruction was achieved using a weighted back-projection of consecutive 2D slices, by AMIRA 3.0 on a PC.

3. Results and discussion

3.1. TEM

Fig. 1 shows a typical set of TEM images of the Si-phases in the Al–Si alloy, in which a large number of Si-phases, both planar- and rod-type Si-phases, are finely and evenly distributed within the field of view, as shown in the bright-field image, Fig. 1(a). A selected-area diffraction pattern of the same region, Fig. 1(b), indicates that the specimen was positioned with the beam direction parallel to $[001]$. Furthermore, a typical dark-field image, Fig. 1(c) was also achieved from the same field of view as in the case of the bright-field image, with the diffraction spot indicated by a small white arrow in Fig. 1(b). Within the selected-area diffraction pattern, large spots correspond to diffracted beams from the Al matrix and small spots represent those from the diamond structure of Si-phases with the lattice parameter $d_{Si} = 0.5431 \text{nm}$. The other weaker spots appearing are due to double diffraction, as schematically shown in Fig. 1(d). From these results, it can easily be concluded that Si-phases have an orientation relationship with the matrix, as cube(fcc)—cube(diamond), $[001]_{Al}/[001]_{Si}$ as well as $[110]_{Al}/[110]_{Si}$. The average size of Si-phases was approximately 50 nm.

Fig. 2 shows a typical HRTEM image of the rod-type Si-phase, with a clear orientation relationship between the Si-phase and the matrix. The presence of dislocations caused by the lattice mismatch are also seen at the interfaces, indicated by thin white arrows. In addition, the occurrence of a twin boundary was confirmed as indicated by a thick arrow.

A typical HR-TEM image of the plate-type Si-phase with again a clear cube–cube orientation relationship is shown in Fig. 3. The presence of dislocations caused by the lattice mismatch are also seen at the interfaces, as in the
case of rod-type Si-phase. The bottom left hand corner of the precipitate shows a different periodicity to that of the rest of the precipitate, although it is simply a thin part of the precipitate with less dynamical interaction.

Careful observation by conventional-TEM and HRTEM revealed that the morphology of Si-phases was classified into two types:

1. the rod-type Si-phase with approximately 4–5 nm diameter and 200 nm length;
2. the planar-like Si-phases with approximately 20–30 nm width; both types of Si-phase have a clear cube-cube orientation relationship with the matrix.

3.2. 3D-ET

Although the 2D distributions and the microstructures of the Si-phases in the Al–Si alloy could be observed by conventional TEM with the combination of 2D images and selected-area electron diffraction (SAED) patterns, the information of depth was still unknown. Therefore, 3D-ET was required for further understanding the 3D distribution of Si-phases.

A series of tilted images was acquired over $-60^\circ$ to $60^\circ$ with an image taken every $2^\circ$ interval (only a part of the images is shown in Fig. 4(a)). Projections from different orientation of the specimen, Fig. 4(b), show the three-dimensional distributions of both plate-type and that of rod-type Si-phases within the section, from different orientations; more than 30 Si-phases are apparent within the volume rendered image. Furthermore, the cross-sectional magnified images of Fig. 4 were obtained by the orthogonal-slice method, visualizing scalar data fields defined on uniform Cartesian grids. Square-like cross-section of rod-type, and plate-type Si-phases are shown in Fig. 5. Whichever way the plate-type Si-phases were oriented, the cross-sectional image would be different from the case of rod-type Si-phases.
Fig. 3. An HRTEM image of the triangular plate Si-phase recorded parallel to the $\langle110\rangle$ zone axis of the matrix. The periodicity of precipitate at the bottom left-hand corner is different from the rest of the precipitate due to the thickness changes, where less dynamical interaction is present.

Fig. 4. (a) A tilted series of the nanocomposite particle (partly shown), (b) a reconstructed volume-rendered image viewing from different orientation of a Si-phase showing a clear morphology of both plate- and rod-type Si-phases.
Empty volumes and striations (indicated by white arrows) are occasionally seen within the plate-type Si-phases. These phenomena are probably due to the lack of information by the limited angular range. Relatively large apertures were used to include both diffraction patterns of matrix and Si-phases, but to avoid strong diffraction contrast within the projections during the acquisition of each tilted series. In fact, prior to the acquisition of tilted series, the diffraction patterns were carefully studied and the specimen was manually rotated to avoid strong diffraction. However, the diffraction contrasts of Si-phases caused by the different scattering characteristics were maintained within the tilted series.

Currently, there are several contrast mechanisms available in the TEM imaging methods: the thickness contrast, the diffraction contrast, the mass contrast and the phase contrast. Therefore, the cause of contrast is usually not monotonic, and involves further simulation and modification of images. In fact, when applying 3D-ET to crystalline materials, the diffraction contrast plays a major role, which sometimes causes difficulties in achieving reconstructed 3D volumes with good quality. Therefore, when the crystalline specimens are examined by 3D-ET, the specimen should be carefully observed by incoherently scattered electrons using a high-angle annular dark-field detector, running under scanning conditions (HAADF-STEM) [7,8]. However, in the case of the Al–Si system, the effect of thickness-, mass- and phase- contrasts could be ignored from the cause of contrast. For example, the tilt series were acquired from the region of the same thickness, and the atomic numbers of both Al (Z = 13) and Si (Z = 14) are next to each other. Within this experiment, the diffraction contrast of Si-phases with different diffraction characteristics from the matrix were still remaining and a BF-TEM 3D-ET could be achieved to reconstruct the 3D volumes. BF-TEM 3D-ET optimized the morphology and the distribution of the rod- and planar-type Si-phases within the Al–Si system.

In addition, the accuracy of the alignment was measured by the cross-correlation method. The mean error in the specimen position during alignment of tilt projection was less than one pixel for 10 times in this case, as shown in Fig. 6.

4. Conclusion

The morphologies and the distributions of those precipitated Si-phases become apparent, by application of HRTEM, electron diffraction and 3D-ET. Two types of Si-phases, rod- and plate-type, were clearly observed, whose orientations were cube-cube, [1 1 0]_Si//[1 1 0]_Al and [1 0 0]_Si//[1 0 0]_Al.

Due to the presence of the double diffraction, 3D-ET by DF-TEM was not as straightforward as expected. Therefore, 3D-ET by BF-TEM imaging was carried out for this Al–Si alloy case, which was still capable of visualizing not only the morphologies of Si-phases but also the distributions of Si-phases.

Fig. 5. A magnified image of Fig. 4, in which cross-sectional images of both plate- and rod-type Si-phases are easily observed.

Fig. 6. Mean error in specimen position incurred during alignment of tilt projections for 3-D reconstructions.
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