A New Specimen Preparation Method for Three-Dimensional Atom Probe

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In our previous study, we introduced a new specimen preparation method for the three-dimensional atom probe in which the materials were pared off from oblique back by Ga-FIB. It was suggested that there was a possibility of reducing the gallium implantation in the analyzable regions by using the method. In this study, the gallium implantation level of the method was evaluated with better statistical accuracy by extending the analyzed volume and improving the mass resolution. The specimens were fabricated by means of the conventional and our new methods, and analyzed by atom probe with voltage or laser pulses. The mass spectra with the larger analyzed volume or counts of detected ions and the higher mass resolution than those of our previous study were obtained. Then, the ratio of gallium ions to all detected ions was calculated with better statistical accuracy and it was confirmed that the gallium ion concentration at the surface area was reduced close to or less than the detection limits of our instrument by our new method. [DOI: 10.1380/ejssnt.2010.141]

Keywords: Atom probe; Focused ion beam; Implantation

I. INTRODUCTION

As the coming of nanotechnology, the scale of electronic devices has been getting smaller and smaller. For example, commercialized devices are expected to be designed within 1-nm rules in the next decade. In analyzing these devices, existing analytical tools reach their performance limits. The three dimensional atom probe (3DAP) has attracted attention as one of the most powerful methods for three dimensional analysis on an atomic scale [1]. The atoms are field-evaporated from the specimen apex as ions when high voltages and voltage or laser pulses are applied to the specimen. In this case, the voltage or laser pulse works as a trigger for the field evaporation of atoms. The elemental identity of ions is determined by the time-of-flight (TOF). The lateral position of each detected ion is determined with a position sensitive detector (PSD) and the depth positions are inferred from the sequences of detected ions. An atomic scale image of the specimen is reconstructed from these data in three dimensional virtual space.

However, there are some problems. For 3DAP analysis, the needle-shaped specimens with an end radius of typically <100 nm are required. To fabricate these specimens, the focused ion beam (FIB) is often used. There are two specimen preparation methods using FIB: annular milling and linear cut [2]. In the former method, the ion beam is irradiated to the apex of the specimen in a circular pattern. In the latter method, a series of inclined line cuts are made across the end of the material at equal increments of rotation around the specimen axis. In these methods, FIB is irradiated from the direction of the specimen apex. The gallium ions are implanted into the apex of the specimen and the surface structure of the specimen is disarranged. 30 keV gallium ions penetrate at least 20 nm in depth [3]. For 3DAP analysis, the analyzable region is a few hundreds of nm in depth. Then this gallium implantation has a great effect on reconstructing an atomic scale image of the specimen. To avoid this gallium implantation, we proposed a new specimen preparation method in this study.
which the materials were pared off from oblique back by FIB [4]. It was suggested that there was a possibility of reducing the gallium implantation in the analyzable regions by using the present method. However, the gallium implantation level was not evaluated with sufficient statistical accuracy, because the analyzed volume or the counts of detected ions were too small. In addition, the specimens were analyzed with only voltage-pulsed 3DAP in our laboratory which had relatively low mass resolution, then gallium isotopes cannot be resolved [4].

In this study, to compare the gallium implantation level for the conventional method with that for the present method, the specimens were fabricated by these two methods and analyzed with improved 3DAP in our laboratory [5]. From the results, the gallium implantation level was evaluated.

II. SPECIMEN PREPARATION METHOD

A. Instrumentation

The schematic diagram of the assembly in a present method is shown in Fig. 1. A commercial FIB instrument (SMI-3050SE, SII NanoTechnology Inc.) was chosen as the base instrument. To rotate the specimen, a small two phase stepping motor (dimension: φ6 mm, total length: 12.2 mm, step angle: 18°, rated current: 100 mA/phase) was used.

The block diagram of the system to control the stepping motor is shown in Fig. 2. Since the step angle of the motor was 18°, the computer controlled current drive circuits (1 mA resolution) were employed to get smaller step angle. DA converter (PCI-3347, Interface Corporation) and the homemade V-I converter (20 mA/V) were used.

The typical data sets of phase currents were input to the computer and the rotation angles were measured. The stepping motor rotates about 72° for these data sets. By repeating these data sets, the stepping motor rotates 360°. Figure 3 shows the phase currents corresponding to the rotation angle read from the measured results. The rotation in increments of about 1° was achieved.

When the proper data set of phase currents is input to the computer, the stepping motor rotates certain degrees. By repeating the process of rotating the stepping motor some degrees and irradiating FIB to the specimen, the specimen is fabricated in the shape of multi-sided pyramid.

B. Procedure

The electropolished specimen was fixed on the stepping motor with the homemade jig after the positions of the specimen, the jig and the motor were adjusted so that the rotation wobble of the specimen apex was reduced to <100 µm [4]. The specimen fixed on the motor was set to FIB stage holder parallel to the holder surface. Then the stage holder was introduced to FIB instrument and tilted. The taper angle of the specimen was determined by the tilt angle. FIB was irradiated from oblique back of the specimen and stepping motor was rotated some degrees. This process was repeated until the rotation angle reached 360°.

III. EXPERIMENTS

A. Specimens

Three specimens A, B and C were prepared from a 99.95% pure tungsten wire (diameter 0.1 mm; NILACO #W-461167). The wire was cut into 10-15 mm long and fixed in the Cu-tube. The specimens were electropolished in a 5% sodium hydroxide solution to the end radius <100 nm, and fabricated using FIB as follows. The specimens A and B were fabricated by the annular milling method [2]. The specimen was set parallel to the beam axis of FIB and FIB was irradiated in a circular pattern from the direction of the specimen apex. The specimen C was fabricated by a present method [4]. The specimen was set parallel to the stage holder surface, the stage holder was tilted to 45°, and therefore FIB was irradiated from oblique back of the specimen. Then, the motor rotated about 72°. By repeating the process of irradiating FIB and rotating the motor, the five-sided pyramidal specimen was fabricated.

B. Atom probe analyses

These specimens were analyzed by our homemade atom probe instrument [5] with a pressure <1×10^-7 Pa triggered with voltage or laser pulse. The specimen A was analyzed with voltage pulse under the condition of 1 kV pulse height, <10 ns pulse width and 1 kHz pulse repetition rate. The specimens B and C were analyzed with laser pulse under the condition of 1064 nm wavelength, 3.7 nJ/pulse power (for specimen B), 0.8 (for specimen C) nJ/pulse power,
300 fs pulse width and 5 kHz pulse repetition rate. The analyses were performed at room temperature.

IV. RESULTS AND DISCUSSIONS

Figure 4 shows the mass spectrum of the specimen A fabricated by annular milling method and analyzed with improved voltage pulse. Ga$^{2+}$, W$^{4+}$ and W$^{3+}$ were detected (Ga$^{2+}$: 54343, W$^{4+}$: 52750, W$^{3+}$: 663851 at counts). The ratio of W$^{4+}$ to W$^{3+}$ was about 8%. In our previous study, Ga$^{3+}$, W$^{4+}$ and W$^{3+}$ were detected (Ga$^{3+}$: 16, W$^{4+}$: 24, W$^{3+}$: 223 for the specimen fabricated by annular milling method and analyzed with voltage pulse) and the ratio of W$^{4+}$ to W$^{3+}$ was about 11%. The ratio of detected ions depends on the electric field at the specimen apex [6], and then the approximately the same electric fields arose at the specimen apaxes.

Figure 5 shows the mass spectrum at the surface area (cumulated detected ions was about 50000 at counts) of the specimen B fabricated by annular milling method and analyzed with laser pulse. Ga$^{+}$, WO$_2^{2+}$, WO$_3^{2+}$ and WO$_3^{3+}$ were detected (Ga$^{+}$: 1031, WO$_2^{2+}$: 1502, WO$_3^{2+}$: 7058, WO$_3^{3+}$:6211 at counts). Figure 6 shows the mass spectrum at the surface area of the specimen C fabricated by a present method and analyzed with laser pulse. W$^{3+}$, W$^{2+}$, WO$_2^{2+}$, WO$_3^{2+}$ and WO$_3^{3+}$ were detected (W$^{3+}$: 526, W$^{2+}$: 513, WO$_2^{2+}$: 391, WO$_3^{2+}$: 991, WO$_3^{3+}$: 744). In Figs. 5 and 6, two peaks assigned to $^{69}$Ga and $^{71}$Ga were observed and the ratios of gallium ions to all detected ions were calculated more accurately to be 5.9 and 6.5%, respectively. On the other hand, there was no peak at $m/z = 71$ in Fig. 6. Therefore, the peak at $m/z = 69$ might not be gallium. Even if the peak at $m/z = 69$ is gallium, the ratio of gallium ions to all detected ions was calculated to be <1%.

Barofsky and Muller reported that on the field evaporation of the iron, the abundance of the pure iron decreased and the abundance of iron oxide increased as the temperature of the specimen was raised because of the surface migration of oxygen from the shank [7]. In this study, there was a possibility that the temperature of the analyzable regions was raised by the laser pulse.

In this study, the counts of detected ions were larger (all detected ions were about 800000, 15000, 300 at counts for the specimen A, B, C, respectively) and our improved instrument had higher mass resolution (FWHM: 40, 75, 300 for old voltage pulse, improved voltage pulse, laser pulse [5], respectively) than those of our previous study. Then gallium isotopes can be resolved. In Figs. 4 and 5, two peaks assigned to $^{69}$Ga and $^{71}$Ga were observed and the ratios of gallium ions to all detected ions were calculated more accurately to be 5.9 and 6.5%, respectively. On the other hand, there was no peak at $m/z = 71$ in Fig. 6. Therefore, the peak at $m/z = 69$ might not be gallium. Even if the peak at $m/z = 69$ is gallium, the ratio of gallium ions to all detected ions was calculated to be <1%.

Figure 7 shows the gallium atom density distribution against the cumulated number of detected ions of the specimen B. It is clear that implanted gallium ratio reduced from surface to interior of the specimen. The ratio of gallium ions at the interior area was about 0.5%. The ratio of gallium ions at the surface area of the specimen fabricated by the present method was <1%, then it was suggested that the present method reduced the gallium
concentration at the surface area to the same level as that at the interior area.

The gallium atom density distribution against the cumulated number of detected ions from surface to interior of the specimen B was observed. The gallium ion concentration at the surface area for the specimen C was close to or less than the detection limits of our instrument, and it was confirmed strictly that the gallium implantation was suppressed by our new method as it had been shown in our previous study (The ratios of gallium ions to all detected ions in our previous study were 6% and 1%) [4]

V. CONCLUSION

The multi-sided pyramidal specimen was fabricated by using the stepping motor. The gallium atom density distribution against the cumulated number of detected ions from surface to interior of the specimen was determined from the specimen fabricated by the conventional annular milling method. It was confirmed with good statistical accuracy that FIB fabricating from oblique back of the specimen reduce the gallium concentration at the surface area in the analyzable regions close to or less than the detection limits of our instrument compared to the conventional head-on annular milling.

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