Ultramicroscopy 144 (2014) 9–18

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

A novel approach for site-specific atom probe specimen preparation by focused ion beam and transmission electron backscatter diffraction

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ARTICLE INFO

Article history:
Received 22 January 2014
Received in revised form
4 April 2014
Accepted 11 April 2014
Available online 21 April 2014

Keywords:
Focused ion beam (FIB)
Grain boundary segregation
Transmission electron backscatter diffraction (t-EBSD)
Transmission Kikuchi diffraction (TKD)
Atom probe tomography (APT)
Transmission electron forward scatter diffraction (t-EFSD)

ABSTRACT

Atom probe tomography (APT) is a suitable technique for chemical analyses with almost atomic resolution. However, the time-consuming site-specific specimen preparation can be improved. Recently, transmission electron backscatter diffraction (t-EBSD) has been established for high resolution crystallographic analyses of thin foils. In this paper we present the first successful application of a combined focused ion beam (FIB)/t-EBSD preparation of site-specific APT specimens using the example of grain boundary segregation in technically pure molybdenum.

It will be shown that the preparation of a grain boundary can be substantially accelerated by t-EBSD analyses in-between the annular milling FIB procedure in the same microscope. With this combined method, a grain boundary can easily be recognized and positioned in the first 220 nm of an APT sample much faster than e.g. with complementary investigations in a transmission electron microscope. Even more, the high resolution technique of t-EBSD gives the opportunity to get crystallographic information of the mapped area and, therefore, an analysis of the grain boundary character to support the interpretation of the APT data files. To optimize this newly developed technique for the application on needle-shaped APT specimens, a parameter study on enhanced background correction, acceleration voltage, and tilt angle was carried out. An acceleration voltage of 30 kV at specimen surface tilt angles between -45° and -35° from horizontal plane leads to the best results. Even for molybdenum the observation of crystal orientation data up to about 200 nm specimen thickness is possible.

1. Introduction

Atom probe tomography (APT) is a highly sensitive tool to detect individual atoms within a needle-shaped specimen [1–3]. Therefore, it is used to study multiphase materials, interfaces in multilayer films, segregation at dislocations or grain boundaries as well as precipitates with almost atomic resolution [4]. Due to the small amounts of impurities, APT is a very suitable method to study grain boundary segregation in technically pure metals [5,6]. However, a limitation of APT is for example the complex specimen preparation [4]. Conventional APT specimen preparation techniques by electro-polishing limit the field of application for grain boundary segregation studies due to the small volume probed by APT.

Nowadays, it is possible to produce specimens out of site-specific regions of interest employing focused ion beam (FIB) systems [7,8]. With this technique, the site-specific specimen preparation of features as grain boundaries is possible for further APT analyses [4].

One of the first site-specific methods to study grain boundaries in the atom probe was applied by Miller et al. [9]. A small wedge or lamella containing a single grain boundary is lifted out of a bulk material with a micromanipulator and is attached to a post-material by welding with e.g. platinum [10–12]. Another preparation method is the use of electro-polished APT specimens with re-sharpening them with a FIB in the region of interest [6,13]. Due to the fact that the visibility in the FIB/scanning electron microscope (SEM) for radii lower than ~500 nm is poor, additional studies in the transmission electron microscope (TEM) have been applied to analyze the location of a grain boundary [6,10,14]. However, a time-consuming repeated exchange between TEM and FIB/SEM is necessary for this technique [6]. Furthermore, to get information about the characteristics of the grain boundary, diffraction patterns have to be recorded. These processes contaminate the specimen and influence the APT analyses [15]. Sha et al. [16] instead used the major zone lines and crystallographic poles of two neighbor grains in a desorption map of an atom probe analysis to investigate the crystallographic structure of the grains.

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http://dx.doi.org/10.1016/j.ultramic.2014.04.003
0304-3991/

This technique was recently developed by Yao at al. [17]. However, this kind of investigation is complex and time-consuming.

Due to the obvious drawbacks of these methods, we propose a new characterization method for the site-specific preparation of grain boundaries in APT specimens based on transmission electron backscatter diffraction (t-EBSD) in this study. Our technique decisively simplifies the preparation process and gives detailed information about the crystallographic grain boundary properties.

The application of t-EBSD expands the field of conventional electron backscatter diffraction (EBSD) in the SEM. It is possible to quantitatively analyze the microstructure of materials based on crystallographic orientations with higher spatial resolution than standard EBSD by using a thin specimen in conventional EBSD systems [18]. In transmission mode, the orientation information contained in the diffraction patterns is derived from only the last 10–20 nm of material at the lower surface before the electrons exit the sample, which enables the higher spatial resolution [18]. Keller et al. [19] firstly proposed to position a thin TEM foil above a commercial EBSD camera to acquire Kikuchi patterns by collecting transmitted forward scattered electrons to improve the spatial resolution of conventional EBSD. This technique is easily applicable because a standard EBSD detector and a commercial software is used.

Keller et al. [19] initially termed the method "transmission-EBSD" (t-EBSD) in 2012, but Trimby [20] and Suzuki [18] who did parameter studies proposed the term "transmission Kikuchi diffraction" (TKD). Another acronym was recommended by Brodusch et al. [21]. He defined the method as "transmission electron forward scatter diffraction" (t-EFSD) due to the fact that the appearing process is physically related to electron forward scattering. In this study, we decided to use the more easily recognizable name "t-EBSD", as specified by Keller et al., although this definition is perhaps not exactly correct.

So far, t-EBSD has only been used to analyze ultrafine-grained materials [22–24] and nanosized particles [25]. These authors used thin foils for all studies. In our work, we apply for the first time t-EBSD on needle-shaped APT specimens with a tip radius of about 100 nm for site-specific APT specimen preparation. Thus, in this investigation we present the first successful application of a combined FIB/t-EBSD preparation process for APT site-specific specimens and a t-EBSD parameter study on APT tips to optimize the new technique.

2. Material and methods

For this study technically pure molybdenum was used to investigate grain boundary segregation which is not well understood in this material. The same material has been studied by TEM and APT in a previous study [6] which can be used as reference. An industrially processed hot-rolled sheet of molybdenum in the as-deformed and recrystallized condition was investigated. The production procedure of this sheet (termed sheet "1" in [6]) as well as the chemical composition is described in detail in [6]. In the present study, the recrystallized state (grain size ~48 μm parallel to the rolling direction) was selected for the demonstration of the APT specimen preparation process by FIB/t-EBSD. The parameter study was carried out with both, the recrystallized and as-deformed state (with a recovered subgrain structure) of the same technically pure molybdenum sheet.

Prior to the preparation process in the FIB combined with t-EBSD, APT specimens were produced by the standard electro-polishing method. Small rods ($10 \times 0.3 \times 0.3\ mm^3$) were cut out of the sheet with their largest dimension parallel to the normal direction. The electrolyte for polishing of the APT specimen was 12.5 vol% H_2SO_4 in ethanol in a gold ring (microloop) as counter electrode. A detailed description of the electro-polishing process is given in [26].

All preparation work was done in a FEI Versa 3D DualBeam (FIB/SEM) workstation equipped with an EDAX Hikari XP EBSD system; therefore, no exchange between different microscopes was necessary. The EDAX OIM Data Collection 7 software was used for the measurements and the EDAX OIM Analysis 7 for the evaluation of the EBSD data files.

For a successful APT measurement, the region of interest has to be located in the first few hundreds of nanometers below the apex. Thus, annular milling in the FIB was performed to position the grain boundary close to the apex of the needle-shaped specimen. All preparation steps were carried out with an acceleration voltage (Acc. V.) of 30 kV. To keep the gallium implantation low, the last annular milling steps were performed at an Acc. V. of 5 kV. However, it is always difficult to deduce if the grain boundary is in the volume for radii lower than ~500 nm due to the poor visibility in the FIB/SEM [6]. Therefore, the last preparation steps were supported by t-EBSD analyses to clearly identify the position of the grain boundary in the specimen. The t-EBSD analyses during the preparation process were carried out at an Acc. V. of 20 kV with a spot size of 2.5. The step size was set to 10 nm and for the last scan prior to the APT measurement to 5 nm. A 4×4 binning of the EBSD camera (i.e. a resolution of 160×120) was used. The scan speed was between 30 and 60 frames per second. All FIB preparation and t-EBSD work was done at a tilt angle of 52° which is also the position of the ion column and a working distance of 10 mm which is the intersection point. Fig. 1 shows the schematic set-up at 0° (a) and 52° (b) stage tilt in the microscope chamber. The stage

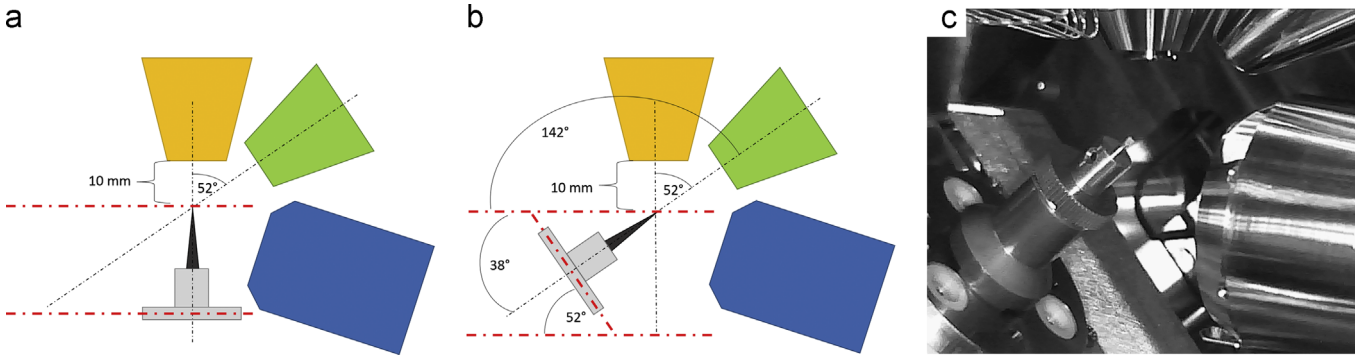

Fig. 1. (a) Schematic set-up in the microscope chamber at a stage tilt of 0°. The APT specimen is placed vertically, parallel to the optical axis of the electron column (yellow). The ion source (green) is located in an angle of 52° to the electron source. The beams have their intersection point at a working distance of 10 mm. The EBSD detector (blue) is located in the bottom right corner of the chamber. Due to the fact that the CCD camera is positioned at the back side of the microscope, the entire set-up is mirror-inverted if examined in the front view of the microscope. (b) The schematic set-up in the microscope chamber at a stage tilt of 52°. The specimen surface tilt angle is -38° (or 142°) from horizontal plane, therefore, the OIM data collection software is set to a −38° tilt angle. (c) The actual set-up in the chamber at a stage tilt angle of 52°. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)