Ultramicroscopy ■ (■■■■) ■■■–■■■

Contents lists available at ScienceDirect

Ultramicroscopy

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

Probing the crystallography of ordered Phases by coupling of orientation microscopy with atom probe tomography

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ARTICLE INFO

Article history:
Received 17 March 2014
Received in revised form
19 July 2014
Accepted 8 September 2014

Keywords:
Superalloys
Ordered phases
APT
EBSD
Field evaporation
SDM

ABSTRACT

The determination of atomic scale structural and compositional information using atom probe tomography is currently limited to elemental solids and dilute alloys. In the present article, a unique coupling of orientation microscopy and atom probe tomography successfully facilitates the crystallographic study of non-dilute alloy systems, with high evaporation fields. This reproducible methodology affords a new perspective to the conventional atom probe tomography of ordered precipitate strengthened superalloys. The high accuracy in crystallographic site-specific sample preparation results in high spatial resolution in APT, which has been demonstrated in Co-base superalloys. The practical applications of this technique can be extended to accurately characterize the nature of buried order/disorder interfaces at the atomic scale, as well as the site occupancies associated with different solute atoms in multi-component superalloys.

1. Introduction

Advancements in transmission electron microscopy, such as three-dimensional (3D) tomography and aberration corrected techniques have enabled the extraction of structural and compositional information in materials with very high resolution (in some cases approaching sub-angstrom resolution). However, techniques like high angle annular dark field scanning transmission electron microscope (HAADF-STEM) has mass sensitivity for beam diffraction, and hence have limited light element detectability [1]. Along with that, these techniques are restricted in the characterization of embedded nano-scale features, within the sample thickness, as the beam passes through multiple phases [2]. On the other hand, atom probe tomography (APT) works on the principle of evaporating materials in a controlled, atomic layer by layer manner, to generate three dimensional (3D) compositional maps with higher spatial resolution as compared to that in other tomography tools and does not suffer from the above mentioned limitations. However, it should be noted that the limited efficiency of detectors used in present generation APT instruments, results in a substantial loss of information, consequently resulting in a reduced spatial resolution. There are numerous efforts currently underway to both improve the efficiency of detectors [3], as well as model the positions of the missing atoms using advanced computational tools [4].

Superalloys are a class of materials that possess stable microstructures at elevated temperatures due to the presence of coherent $L1_2$ γ' precipitates spatially aligned along elastically soft directions in a face centered cubic (FCC) γ matrix [5]. Nickel-base superalloys are used in a variety of applications such as in turbine blades of aircraft engines and land-based turbine engine [5]. The recent discovery of novel Co-base alloys [6], which also form γ–γ' microstructures, similar to nickel base superalloys, has led to rapid research in these alloys as potential next generation superalloys [7–9]. The structural and compositional information obtained by APT has given insights to the atomistic behavior of nickel and cobalt-base superalloys [10–13]. The observation of lattice planes in the APT reconstruction allows for characterization of the ordered precipitates in terms of site occupancy of solute atoms within the precipitates [14–17]. Recent studies on quantification of the compositional width of γ/γ' interfaces by APT have associated the energetics of the interface with the coarsening of γ' precipitates [11,18–21]. Apart from these studies, APT in conjunction with modeling tools has accurately quantified and rationalized segregation of elements at interphase interfaces in various systems [22–24].

There is an increasing effort towards obtaining crystallographic information at the highest possible resolution using APT [15,25–27]. Novel techniques developed towards this end, are being employed for three-dimensional (3D) orientation mapping of grain boundaries in nano-crystalline materials and sub-lattice occupancy of trace and

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http://dx.doi.org/10.1016/j.ultramic.2014.09.001
0304-3991/

solute atoms [25,28]. The hemispherical surface of the atom probe tip cuts multiple planes of different crystallographic orientations, and hence the surface is not atomically smooth and it contains a series of concentric terraces, better known as crystallographic poles. The atoms present at the kink site of the terrace get ionized first, and subsequently they leave behind the low evaporating region, the crystallographic pole. The intersections of different terraces on the hemispherical surface form a pattern of zone lines. Thus, the two-dimensional (2-D) desorption image, formed by accumulation of ions traveling to the detector, reveals the crystallography of the atom probe tip. The desorption image obtained in APT is analogous to that obtained in field ion microscopy (FIM) and ion desorption microscopy [29,30]. The arrangement of poles in a 2D desorption image is used to understand the 3D crystallography of the materials where individual lattice planes are resolved in both metals and semiconductors [16,31]. Due to trajectory aberrations associated with field evaporation of ions, the crystallographic poles appear to be joined by zone lines. However, to a large extent, crystallographic studies using APT have been limited to pure metals or dilute alloys where individual poles and zone lines are clearly visible even within a limited field of view [28,32]. The algorithms like Fourier transforms and Hough transforms have been used to index the poles in the desorption image [25,33]. However, highly alloyed materials, typically yield low contrast in 2D-desorption images due to the large differences in the evaporation fields associated with multiple elements. Consequently, the identification of multiple crystallographic poles in these desorption images becomes rather challenging. Hence, crystallographic studies using the present generation techniques have been very difficult for such multi-component systems [34].

In the first part of this article, a novel reproducible approach has been presented to extract crystallographic information using APT from highly alloyed materials, by synergistic coupling of orientation microscopy with APT, which has been demonstrated for cobalt-base superalloys. In this approach, as a first step, orientation microscopy is used to determine the site-specific crystallographic orientation of the region from which the atom probe samples are subsequently extracted. Hence, the identification of poles in the 2D desorption image becomes possible in spite of its poor visibility, which in turn permits crystallographic information to be determined from the APT reconstructions. Some recent articles have also discussed the coupling of orientation microscopy with atom probe tomography for microstructural characterization [35–37].

In the second part, how the accuracy of site-specific sample preparation affects the spatial resolution in the atom probe reconstruction has been demonstrated. The anomalous dependence of spatial resolution with respect to the miller indices of the specific crystallographic plane has also been demonstrated and possible practical applicability of this characterization method to ordered structures has been discussed.

2. Materials and methods

The as-cast Co–10Al–10W (at%) and Co–10Al–10W–2Ta (at%) alloys were encapsulated in a quartz tube which was vacuumed and backfilled with argon. The alloys were supersolvus solution treated in a vacuum furnace at 1250 °C in the single γ phase field for 12 h. to dissolve any existing γ', followed by water quenching. Then, quartz encapsulated homogenized sample with argon atmosphere was aged for 256 h. at 900 °C and finally water quenched. The annealed samples were then prepared by conventional metallographic studies for orientation microscopy studies using electron backscatter diffraction (EBSD). Electron backscatter diffraction studies were carried out in a dual-beam focused ion beam (dual-beam FIB) instrument, the FEI FESEM NOVA 200, retrofitted with EDAX DigiView IV EBSD camera. TSL OIM Data Collection and TSL OIM Data Analysis 5.3 software were used for data acquisition and analyses respectively. The microscope stage was tilted to 61° for EBSD data collection. The angle of tilt was limited by the allowed tilt in the microscope. The sample surface was then scanned in a systematic manner from one edge to the other, with each scan covering an approximate area of 500 μm × 500 μm with a step size of 10 μm. Once the desired {001} plane was obtained in the scan, another high resolution scan with a step size of 0.5 μm was carried out in the interested grain, and pole figures were plotted to confirm the grain orientation. Once identified, the boundaries of the grain were marked by trenches cut using the ion beam by tilting the stage back to 52° at the same time keeping the scanned region in focus. This tilt was also aided by the hydrocarbon deposits on the sample surface that occur during the EBSD scan. The stage was then tilted back to 22°, and site specific APT samples were prepared using the dual-beam FIB lift-out process. Experiments were carried out using a local electrode atom probe (LEAP™ 3000X) system in the voltage evaporation mode at a temperature range of 40–60 K, with the evaporation rate at 0.5% and pulse frequency of 200 kHz. Data analysis was performed using the CAMECA™ 3.6.6 software.

3. Technique of coupling OM with APT

Atom probe tomography works on the principle of field evaporation where atoms from the surface of a nano-scale hemispherical apex are ionized under a high pulsating electric field [34]. The ions are desorbed and subsequently detected by a multichannel plate based detector atomic layer by layer, which results in the depth resolution of this tomography tool better than an inter-planar spacing. On the other hand, the lateral resolution is lower due to the trajectory aberration being one of the primary reasons [38]. Due to the atomically uneven surface of the atom probe tip, the distribution of electric field on the surface varies. There are local variations in the electric field at the intersections of different terraces on the atom probe tip that result in deviation in the ionic trajectories. These trajectory aberrations are reflected in the desorption image as high and low hit-density. The difference in evaporation fields due to different atoms in multi-component alloys aggravates the trajectory aberration. As the APT works on layer-by-layer evaporation, the depth resolution is highly unlikely to get affected by the trajectory aberrations. In addition to trajectory aberrations, the surface migration of atoms due to electric field gradients on the atom probe tip has also been observed [39].This directional walk of solute atoms can lower lateral resolution in atom probe tomography. Although the best depth and lateral resolution in APT have been evaluated as 0.06 nm and 0.2 nm respectively in pure W [40], the degradation of spatial resolution in practical systems makes the imaging of a complete lattice still a challenge. But, the high depth resolution has allowed for exploration of structural information in a wide variety of materials. Recently, Moody et al. have discussed that the highest resolution of atomic planes is achieved when the normal to the atomic planes is exactly parallel to the tip axis [32]. On the other hand, when the normal to the atomic planes is at a solid angle to the tip axis, there will be a loss of spatial resolution along the direction normal to the atomic planes.

Fig. 1 shows a schematic of the hemispherical apex of an atom probe tip, which describes the geometric sensitivity of the spatial resolution based on a simple arithmetic treatment, similar to previously reported methods [41]. The schematic shows two cases: (1) when the normal to a certain set of crystallographic planes is exactly parallel to the tip axis, and (2) when the same set of crystallographic planes are at an angle to the tip axis. It has been

Please cite this article as: S. Meher, et al., Probing the crystallography of ordered Phases by coupling of orientation microscopy with atom probe tomography, Ultramicroscopy (2014), http://dx.doi.org/10.1016/j.ultramic.2014.09.001