Phase Transitions and Recrystallization in a Ti-46at%Al-9at%Nb Alloy as Observed by In-Situ High-Energy X-Ray Diffraction

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ABSTRACT

High-energy synchrotron X-ray diffraction is a powerful tool for bulk studies of materials. In this investigation, it is applied to the investigation of an intermetallic γ-TiAl based alloy with a composition of Ti-46Al-9Nb. The morphology of the reflections on the Debye-Scherrer rings is evaluated in order to approach grain sizes as well as crystallographic correlations. An in-situ heating cycle from room temperature to a temperature above the α-transus temperature has been conducted starting from a massively transformed sample. With increasing temperature the occurrence of strain relaxation, chemical and phase separation, domain orientations, phase transitions, recrystallization processes, and subsequent grain growth can be observed. During cooling to room temperature, crystallographic correlations between the re-appearing γ-phase and the host α-phase, known as the Blackburn correlation, are observed in the reciprocal lattice, which splits into different twinning and domain orientation relationships present in the fully lamellar microstructure.

INTRODUCTION

The main benefit of high-energy X-rays around 100 keV is the deep penetration into matter which makes them a probe for bulk samples in physics and materials science. Scattering angles are small and diffraction is directed forward allowing for simple two-dimensional detector set-ups in transmission geometry [1]. The brilliance of modern 3rd generation synchrotron radiation facilities have boosted real space, reciprocal space and time resolution of this technique and, thus, allow for in-situ studies in complex systems.

We have employed this kind of radiation for in-situ two-dimensional powder diffraction on the intermetallic alloy Ti-46Al-9Nb (composition in atomic percent). Intermetallic γ-TiAl based alloys bear the advantage of their light weight combined with mechanical strength at very high temperature which makes this material a strong candidate for heat-exposed mechanical parts, such as turbine blades or turbo chargers in the aerospace and transportation industry [2]. The mechanical properties depend strongly on the prevailing microstructure, which is not only a function of composition, but largely depends on the thermo-mechanical history of the material. Therefore, the detailed understanding of the physical and chemical processes during thermo-mechanical processing or annealing are of great importance. Traditionally, samples are heated to

the temperature condition of interest, then quenched and subsequently investigated at room temperature. Here we present a novel in-situ study performed by high-energy X-rays.

EXPERIMENT

The high-energy beamline ID15b at the European Synchrotron Radiation Facility (ESRF) has been used for the present study. A monochromatic beam with an X-ray energy of 94.4 keV ($k = 47.8$ Å^{-1}, $\lambda = 0.131$ Å) and a beam size of about 0.1 x 0.1 mm^2 was used for a diffraction setup in transmission, allowing to integrate over the bulk of the sample. The diffraction angles of this short wavelength are typical smaller than 10°, allowing Debye-Scherrer rings to be recorded in forward direction with a flat MAR345 image plate detector [3]. Reflections of the γ-phase are indexed according to the tetragonal lattice. Both α and α_2-Ti$_3$Al are hexagonal with a distinguished superstructure in the ordered phase. For the purpose of minimal ambiguity, α and α_2 are indexed according to the larger unit cell of the ordered phase α_2. Further and detailed information concerning data analysis is given in [4].

A polycrystalline, massively transformed Ti-46Al-9Nb sample has been prepared by annealing at 1330°C for 4 min followed by oil quenching [5]. A piece of the massively transformed sample with a cross section of about 1 mm^2 was mounted on a ceramic holder of an ESRF-designed diffraction furnace. The latter device operates in air, and consists of a vertical boron nitride tube of 10 mm diameter and 100 mm height. The mantle contains a KANTHAL heating element attaining temperatures up to 1400˚C. The temperature measurement had a significant systematic error, in particular when running a ramp, and had been recalibrated to the α-transus temperature $T_\alpha = 1330°C$, as obtained from other measurements. The exposure time was 45 s and together with data recording and an additional erasing cycle of the image plate, full Debye-Scherrer rings were recorded in 16-bit gray levels continuously in time intervals of 275 s while running the temperature profile as given in figure 1. The initially massively transformed sample completely recrystallized during the experiment, ending up in a fully lamellar microstructure [4].
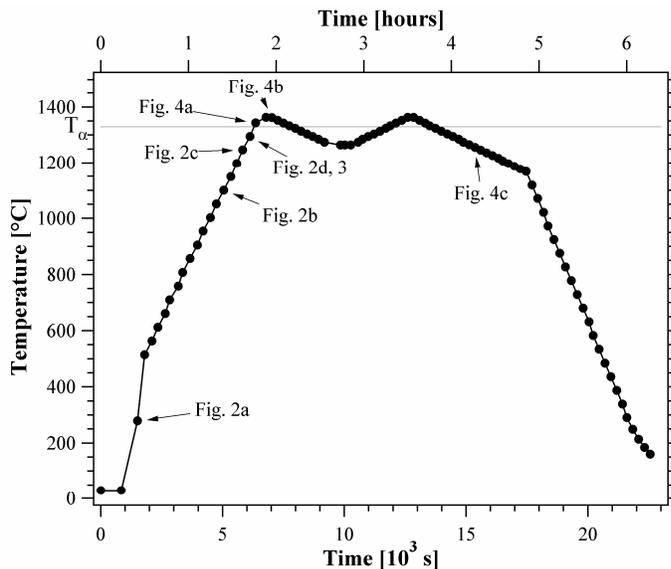

Figure 1. Temperature profile as used for the in-situ experiment with data acquisition points (full circles). Indicated is where figures 2 and 4 were taken. T_α: marks the α-transus transition temperature of the Ti-46Al-9Nb alloy.

RESULTS AND DISCUSSION

Measurements below the α-transus temperature

Typical sections of the fully recorded Debye-Scherrer patterns are shown in figure 2 for various temperatures below T_α. At 280°C, figure 2a, the pattern is exclusively composed by a contribution of the γ-phase and looks very blurred. Particularly, the γ-002 and γ-200 reflections should feature a double ring because of the slight difference in lattice spacing along the c and the a axis of the γ-TiAl cell (ordered tetragonal $L1_0$ structure). This splitting is washed out, not to a single sharp ring as in a cubic structure, but to a broad distribution between the reciprocal c and a values. It attests to huge internal stresses, microstructural and chemical disorder stemming from the nucleation and growth process of the massively transformed material [4,5] when γ-domains grew spontaneously all over a parent α-grain, each with the c-axis oriented arbitrary in one of the preferred directions. After the whole material had transformed, domains touched each other and the misfit stores a large amount of mechanical energy. Since disorder is high, domains are small such that the stress cannot relax. The micro-stresses relax with increasing temperature and the expected double-ring feature of the γ-002 and γ-200 reflexes appears which we attribute to the re-orientation of smaller domains in favor of a larger. This recovery process starts above 700 ˚C and both rings are already well distinguished in figure 2b. With increasing temperature, re-arrangement of the spots on the Debye-Scherrer rings occurs and, in particular, weaker reflections disappear proofing for γ-grain growth, precipitation of α_2/α-phase and related change in phase ratio [4], which is seen particularly in figures 2c and d, where a major amount of α-phase is present. From 1100˚C to 1295˚C, the morphology of the ring-patterns changes completely and gives evidence for an entirely re-arranged microstructure, still entangled by the coexistence of two phases. The quantitative evaluation of ring diameters and thus lattice parameters relating to thermal expansion as well as changes in chemistry upon segregation and
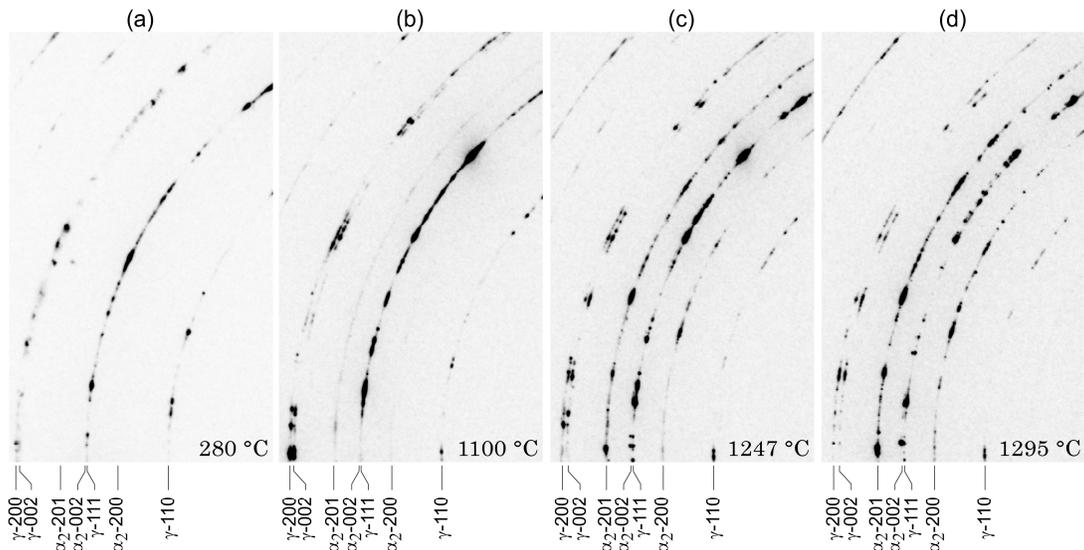

Figure 2. Selected extracts of Debye-Scherrer patterns obtained below the alpha transus temperature. The starting material was a massively transformed Ti-46Al-9Nb alloy. The anisotropic intensity distribution along the rings stems from reflections of individual γ-TiAl crystallites and evolves with temperature, such as the appearance of the α_2/α-phase.
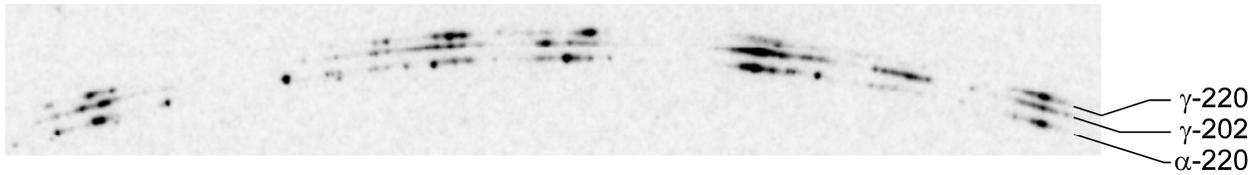
γ-220
γ-202
α-220

Figure 3. Ring triplet measured at 1295 °C showing correlations between the γ- and α-phases which are coherently aligned to each other by the Blackburn relationship and domain orientations. Features from one of the rings appear on the others, either face-to-face or shifted by a small angle grain (domain) boundary, depending on misfit and projection.

the corresponding c/a ratio of the unit cell are given elsewhere [4].

The detailed look to the α-002 γ-111 pair gives similar evidence of face-to-face reflections which relate directly to the Blackburn relation [6] between the two phases. Accordingly, the ring triplet α-220, γ-202, γ-220 taken at 1295°C is shown magnified in figure 3 and demonstrates coherent relations in a second orientation between the two phases and the domains in the γ-phase which are perpendicular to the hexagonal base plane and the common {111} planes, respectively. The longitudinal variation of the scattering reciprocal lattice vectors, i.e. the radii of the Debye-Scherrer rings in figure 3, can be evaluated by azimuthal integration over a representative sector of the rings which leads to a classical powder diffraction pattern.

Measurements above the α-transus temperature

Rapid nucleation and grain growth kinetics are observed above $T > T_\alpha$, where the Ti-46Al-9Nb alloy transforms to pure α-phase. A set of three typical diffraction patterns is shown in figures 4a-c. The typical ring pattern of the initial material fades rapidly when heating above T_α. First, all the γ-phase disappears, leaving spots only on the α-rings. Extensive grain coarsening leaves only a few groups of sharp spots such as around the α-002, 201, $20\bar{1}$ and $00\bar{2}$ positions in figure 4a. Figure 4b shows a regular pattern of diffuse spots. They stem from one large, single grain. Cooling below T_α gives rise of γ-reflections which are regularly arranged and well correlated with the α-orientations as seen in figure 4c. The residual α-pattern can be indexed as given in figure 4b. By fortunate incident, the crystallite was aligned in a way that we observe a plane in reciprocal space, spanned by the α-002 and α-200 reflections and with the α-$\bar{1}20$ normal.

The coarsening of the α-grains from 1344 °C to 1362 °C occurs in such a way that groups of many spots, around e.g. the α-002 position or the α-$20\bar{1}$ position, rearrange to form the new and large single spot. This may happen when two neighboring spots on the same ring stem from regions inclined by a small-angle boundary which has been broken up. However, neighboring spots may even merge together when they originally lie on different rings, i.e. neighboring grains have different crystallographic orientations by a large angle. For example, the lattice leading to a 201-type reflection may rearrange to a 002-type. Both of these reflections correspond to lattice spacings which only differ by a few percent, and the atoms have to rearrange only a little amount to swap the crystallographic orientation by a large angle, rather than to rotate the whole volume by this value. In addition, almost symmetric hexagons can be recognized in the rectangular diffraction pattern, such as the points (000, 201, 203, 004, $\bar{2}03$, $\bar{2}01$), which are distorted by a

few percent only to match the hexagon of the basal plane. Such almost-twin relationships are another argument for correlated mechanism of grain growth [4].
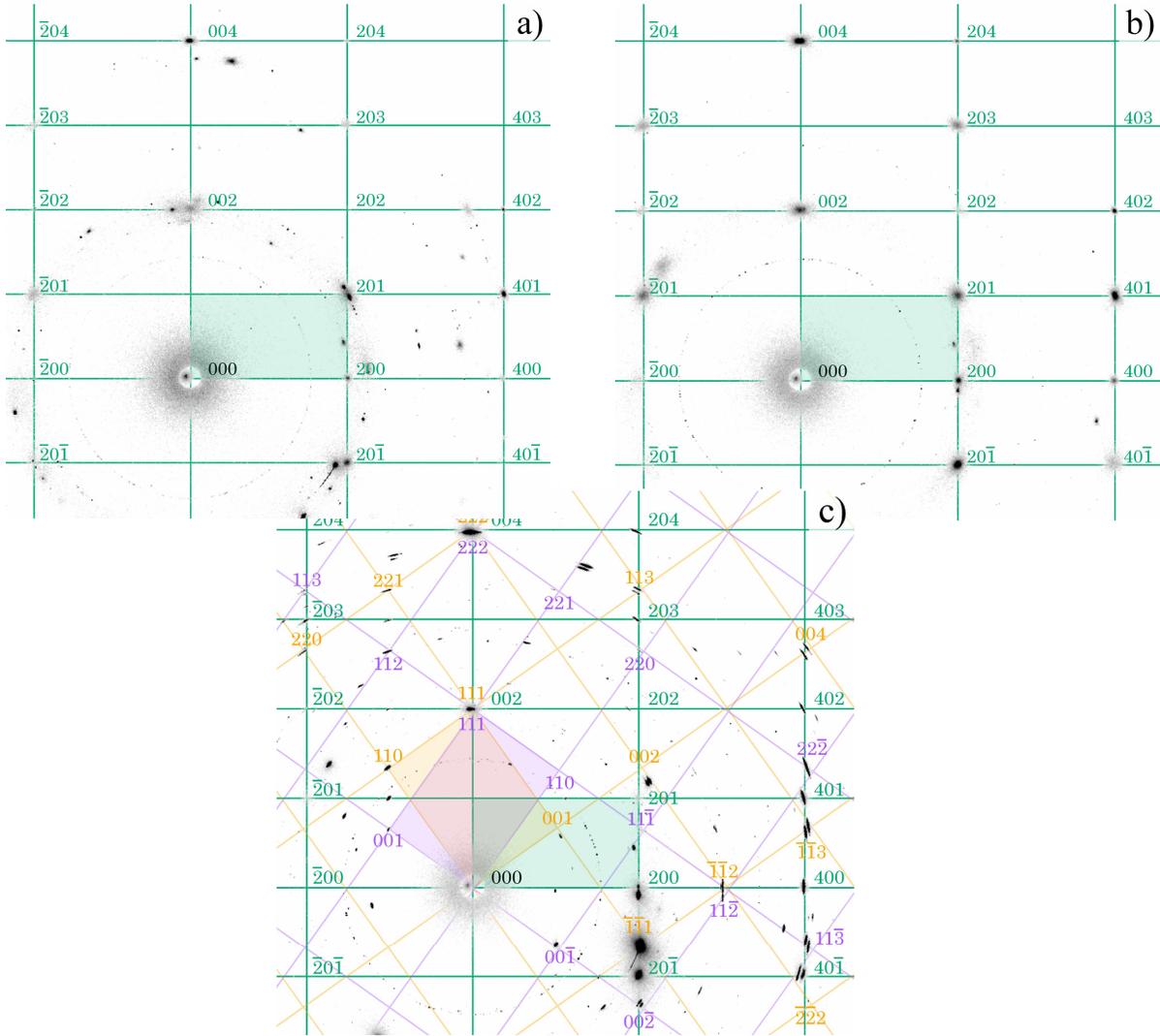

Figure 4. (a) Diffraction patterns taken in-situ from the α-phase of the Ti-46Al-9Nb alloy at 1344°C immediately after crossing the α+γ → α transition temperature, (b) 660 s later at 1362 °C and (c) after re-entering the (α + γ) phase region and subsequent cooling to 1256 °C (figure 1). A clear reciprocal lattice stemming from one large crystallite shows up in (b) which is drawn by the green lines and indexed. The indication has been taken back to (a) and reveals reflections grouping already around these lattice positions, i.e. around 002, 201 and 20$\bar{1}$ reflections. The reappearance of the γ-phase in (c) is crystallographically well oriented and reflections appear regularly between the α-reflections. They can be indexed by two separated γ-lattices drawn in orange and purple colors, coupled by a twin relationship and both sharing the γ-111 with the α-002 reflection. The γ-indexing is given for a cubic approach of the tetragonal lattice.

Re-entrance into the α+γ-phase region

The γ-phase and the α-phase after re-entering the α+γ-phase region are connected through the Blackburn-relationship, in which α-002 and γ-111 fall together (figure 4c). As already discussed above, there exists an additional correlation where the α-$\bar{2}$20 and, e.g., the γ-$\bar{2}$20 reflections fall together. Then the γ-$\bar{1}\bar{1}$2 spot can be indexed, which lies 90° to the γ-111 reflection and 3/4 to the α-400 vector. These two reciprocal lattice vectors span one plane in reciprocal space and many spots can be identified falling onto the rectangular pattern spanned by the reciprocal γ-001 and γ-110 vectors. Other spots can be identified by a second, simultaneously occurring reciprocal lattice, which is mirrored at the line from 000 to 111. It is the crystallographic twin with the common points $111 \rightarrow 111_T$ and $\bar{1}\bar{1}2 \rightarrow 11\bar{2}_T$. All these considerations have been indexed with the notation for a cubic γ-lattice. The tetragonal lattice with lower symmetry gives rise to three different domains, depending in which cubic direction the c-axis is aligned. It expresses the splitting of reciprocal lattice spots like the $00\bar{2}$ double spot on the first γ-lattice of figure 4c, which at least stems from two domains.

The reciprocal plane normal to the α-$\bar{2}$20 reflection can be described by three coherent reciprocal lattices, namely the α-lattice and the two twin γ-lattices. They correspond well to the observed fully lamellar microstructure which shows large grains made of coherent lamellae, i.e. regions with γ-laths of three domains and two twin orientations intersected by very fine α_2-lamellae.

SUMMARY

An in-situ 2D high-energy X-ray powder-diffraction experiment has been conducted on a massively transformed Ti-46Al-9Nb polycrystalline sample revealing details in the relationships between grains and phases. Debye-Scherrer rings, which are extremely rich in information, were recorded with a two-dimensional detector. Reflections from individual grains or colonies can be followed as a function of temperature in real time and allow to analyze their evolution. The Blackburn relation reigns the phase transition between the α_2/α and the γ-phases, and there is great evidence that crystallite growth occurs preferably through an ordered rearrangement process of atoms. In particular, neighbored crystallites with a large crystallographic misalignment may be aligned with lattice planes of similar spacing (within 10%) and close orientation (up to 10°) to each other, which then rearrange into one major grain.

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