Precession electron diffraction assisted orientation mapping of gradient nanostructure in a Ni-based superalloy

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Abstract. Surface mechanical grinding of a Ni-based superalloy can introduce a gradient microstructure in the surface layer with a grain size from nanoscale to microscale. In-depth investigation of the crystal orientation distribution of the surface nanostructured layer is more often, however, not an easy work by using the scanning electron microscope (SEM) based electron backscatter diffraction (EBSD) method due to its sensitivity to lattice distortions and spatial resolution limitation. Here we use a newly developed precession electron diffraction (PED) technique coupled with transmission electron microscopy (TEM) to investigate the microstructural and crystallographic characteristics of the surface gradient nanostructure, with particular emphasis on the topmost nanocrystalline layer. A strong shear texture and a minor Copper texture were identified according to orientation analyses of the 1.6 \( \mu \text{m} \) thick near-surface nanocrystalline layer. The PED technique is proved to be practical for two dimensional orientation mapping of severely deformed microstructures at the nanoscale.

1. Introduction
Surface integrity becomes increasingly vital for mechanical processing of structural components since the chemical, microstructural and crystallographic characteristics of the surface layers can crucially determine various kinds of mechanical properties in these regions, and greatly affect the final performance of these components [1-5]. Many efforts have been made in recent decades to optimize surface microstructure and properties, of which surface nanocrystallization is an effective and prevalent approach [4-10]. Through a surface mechanical attrition treatment, surface mechanical grinding treatment (SMGT), shot peening, laser shock processing as well as other machining treatments [1,2,4-10], the former coarse grains in surface-adjacent regions transform into a gradient microstructure with a grain size ranging from a few nanometers at the topmost surface to coarse grains in the bulk interior. The extent of the grain refinement depends on processing conditions and original microstructural features. The advent of such a surface gradient microstructure significantly and simultaneously improves tensile strength, fracture toughness, fatigue lifetime as well as other mechanical properties of the sample [3-6]. In-depth investigation of the surface nanocrystalline layer in some face centered cubic (FCC) metals to explore the origin of property optimization from a crystallographic perspective shows that a typical shear texture \{001\}<110> generally forms in the SMGT samples, based on the scanning electron microscope (SEM) based electron backscattered diffraction (EBSD) analyses of large amounts of nanocrystallines, ultrafine grains and coarse grains within a nearly half millimeter thick layer adjacent to the topmost surface [4]. Specifically, the texture determination of the nanocrystalline layer actually
results from grains larger than 30-50 nm in diameter or spacing, and located 3-10 μm or more depth from the topmost surface, mainly because of the spatial resolution limitation of the SEM electron beam and sensitivity of EBSD to lattice distortion arising from severe plastic deformation (SPD) [11,12]. Such a situation indicates that grains smaller than 30-50 nm and located in the top 3-10 μm nanocrystalline layer actually do not contribute to the global texture analysis. Therefore, the individual texture information is missing in the texture analysis, leaving a question as to whether the top nanocrystalline layer of a few micrometers thick possesses a similar crystallographic feature as shown in large scale EBSD texture analyses. An effective texture analysis method with a higher spatial resolution and less sensitivity to lattice distortion, therefore, becomes an expanding demand for crystallographic analyses of SPD induced nanocrystallines, especially those in the topmost surface layer, with grain size smaller than 30-50 nm.

The electron beam in transmission electron microscope (TEM) generally has a high spatial resolution and its interaction with grains of polycrystalline materials can generate different oriented diffraction patterns at the back focal plane of the objective lens. Thus it is a good choice for nanoscale characterization of crystallographic orientations. However, typical dynamical effects, such as double diffraction, bending and thickness effects, may substantially reduce the sensitivity of a diffraction pattern to orientation variations [11-14]. While fast precessing focused incident electron beam of a small angle around the optical axis, reflections from zero and high order Laue zones and tilted reciprocal lattice may overlap, complement and finally present quasi-kinematical diffraction patterns [13-15]. By precisely and ultrafast template matching of these patterns with precalculated kinematical diffraction patterns corresponding to a series of orientations, the crystal orientation then can be determined. The precession electron diffraction (PED) currently used shows good prospect for phase and orientation mapping of polycrystalline materials [13-17].

In this study, we use PED coupled TEM to investigate microstructural and crystallographic characteristics of the surface nanostructured layer in the SMGT Ni-based superalloy, with particular emphasis on the topmost nanocrystalline layer. The texture feature is identified and the problems related with orientation indexing are further discussed.

2. Experimental

A commercial Ni-based superalloy (with nominal composition of Ni-18.9Cr-Fe-3.1Mo-5.3Nb-1.1Ti-0.5Al (wt. %)) plate (100 × 80 ×50 mm³ in size) is used in the present investigation. Before SMGT, the plate surfaces were polished with silicon carbide papers, then aged at 720 °C for 8 h and furnace-cooled at the rate of 50 °C/h to 620 °C, further held at 620 °C for 8 h and air cooled. The microstructure of the annealed Ni-based superalloy is characterized by a coarse grain γ matrix with a high density of strengthening phase γ′ and γ” as well as randomly distributed δ particles. The average grain size of the γ matrix is 4-10 μm.

The precise SMGT of the pretreated plate was accomplished at ambient temperature on MM 7120A horizontal surface grinder under wet condition. The processing parameters were employed as follows: wheel speed 25 m/s, table speed 720 m/s, grinding depth 0.005 mm and feed rate 1.0 mm/rev. The feed direction was defined as SD, while the direction perpendicular to the grinding surface is defined as ND.

After SMGT, the cross-sectional TEM samples were prepared via an improved transverse TEM sample preparation method, to show microstructural evolution along the depth direction. Much attention has been paid to the sample preparation processes such as cutting, grinding, dimpling and ion milling to ensure the topmost surface of the SMGT sample was well protected. TEM microstructural characterization was performed on a 300kV field emission transmission electron microscope (TEM) Tecnai F30 G2, equipped with a commercial PED system NanoMEGAS ASTAR (Brussels, Belgium). The crystal orientation analysis of the surface nanostructured layer was obtained through continuous scanning of the fast precessed electron beam on an area of interest with a precession angle of 0.6°, beam size of 5 nm and step size of 4 nm, simultaneously fast acquisition of PED patterns (Fig. 1a) and offline ultrafast matching of these obtained PED patterns with the precalculated kinematical diffraction patterns (Fig. 1b and 1c). Further orientation mapping analysis and visualization were carried out using
ASTAR MapViewer, the information about orientation, index, reliability, misorientation and their combinations can be revealed in detail. To obtain more crystallographic information, the orientation data were also processed and visualized using a Channel 5 software package.

Fig. 1. (a) Schematic illustration of the precessed and focused electron beam and its continuous scanning on an area of interest near the sample surface, resulting in PED patterns corresponding to the grains with different orientations; (b) The simulated kinematical diffraction patterns in the crystal databank; (c) Template matching between the obtained PED patterns and their precalculated counterparts.

3. Results

Figure 2 shows the cross-sectional TEM image of the surface layer of the SMGT Ni-based superalloy. From low magnification TEM image of the top 6 \( \mu m \) thick surface deformation layer (Fig. 2a), the microstructural evolution along the depth direction from the sample surface can be clearly observed, i.e. the top tiny grain layer with thickness ranging from 150 to 600 nm (see red dotted line), a thin lamellar layer with irregularly elongated grains, and equiaxed coarse grain layer with increasing grain size with the depth from the sample surface. The coarse grain layer is characterized by intersecting slip traces on two typical slip planes, indicating the activation of multiple slip systems. Close observation of the top tiny grain layer distinctly demonstrates two sublayers (Fig. 2b, see yellow dotted line), one adjoining to the topmost surface is composed of equiaxed nanocrystallines of 4-10 nm in diameter, the other mainly consists of laminated grains with a boundary spacing of about 15-60 nm, as well as some scattered nanocrystallines with various sizes and morphologies. On the whole, such complicated but universal hierarchical nanostructures are a typical surface microstructure of the SMGT Ni-based superalloy as well as other materials after various kinds of surface mechanical treatments [4-10].

To further unravel the microstructural feature of the gradient nanostructure and its formation mechanism from a crystallographic perspective, the crystal orientation was analyzed on a PED platform integrated into a 300kV TEM. The operation procedures and parameters were detailed in Section 2. Through fast scanning of the focused precessing electron beam on a two dimensional area (1600 \( \times \) 800 nm\(^2\), as shown in the virtual bright field (VBF) image Fig. 3a) within ~1.6 \( \mu m \) thick near-surface layer, a series of PED patterns were acquired (Figs. 3b-d) and further indexed via ultrafast template matching. The obtained inverse pole figure (IPF) orientation map of the surface SPD layer is shown in Figs. 3e and 3f, from which both the morphological and crystallographic features of nanocrystallines can be distinguished, better than from the corresponding VBF image (Fig. 3a). The reliability of orientation indexing was roughly evaluated through point-to-point examination of template matching between the obtained PED patterns and probable precalculated patterns. Most of the PED patterns, such as diffraction pattern obtained from site A (Fig. 3b), are proved to be single crystal diffraction patterns and match well with corresponding orientation indexing results shown in the IPF orientation map on Fig. 3e. While the
Fig. 2. (a,b) Cross-sectional TEM images showing surface gradient nanostructures in SMGT Ni-based superalloy sample. Note that the topmost surface of the samples are on the right.

...rest are superimposed patterns, which gradually increase in number with decreasing grain size and thus reducing reliability of orientation indexing (Fig. 3g). Take the diffraction pattern (Fig. 3d) obtained from site C as an example, the superimposed PED pattern was actually constituted by four inclined diffraction patterns <011>, <214>, <215> and <114>, but only <215> was recognized as the most reliable orientation of the nanograin. This unique choice may probably cause misinterpretation of the true orientation of nanocrystallines.

Grain boundary misorientation analysis further demonstrates that some adjoining grains with planar boundaries (see arrows) show a misorientation of about 60° <111> (Fig. 3h), indicating the formation of nanoscale deformation twins in the surface SPD layer. In specific, nanotwin lamella (see dotted line in Fig. 3f) with a boundary spacing as small as 8 nm (Fig. 3h) can be clearly distinguished, implying a higher spatial resolution in PED assisted orientation mapping than that in commercial EBSD [11,12]. Based on orientation analyses, the texture components of the 1.6 μm thick near-surface SPD layer are identified as a major shear texture {001}<110> and a minor Copper texture {112}<111> (Fig. 3i).

4. Discussion

The IPF orientation maps from SD and ND directions (Figs. 3e and 3f) collectively revealed the microstructural and crystallographic features of the surface SPD layer, but some details of the same structure can also be detected different from part to part. As shown in the left part of Fig. 3e, the adjacent laminated grains show similar orientations when revealing by IPF in SD. However, these orientations show differences when revealing by IPF in ND (Fig. 3f). This implies that these laminated grains shared a common SD, forming a fiber texture.

The present PED assisted crystal orientation analysis of the surface nanostructures clearly identified a strong shear texture, {001}<110>, and a minor Copper texture. The shear texture was previously reported in the nanolaminated structure of SMGT pure Ni by large scale EBSD texture analyses [4]. It is reasonable that the shear texture was produced by the strong shear deformation during the SMGT process. While the present results show that there is a minor Copper texture within topmost layer. It was reported that Copper component can be produced by lattice rotation of {001}<110> orientation.
Fig. 3. (a) The VBF image of a scanned area in the surface nanostructure layer; (b-d) PED patterns obtained from sites A, B and C, respectively; (e,f) Orientation maps constructed by IPF from SD and ND directions, respectively; (g) Reliability map; (h) The misorientation of grain boundary variation across nanotwin lamella along the dotted line shown in (f); (i) {111} pole figure corresponding to (e). Note that (a,e-g) share the same scale bar, and the topmost surface is on the right side.
during plastic deformation [18]. In the IPF orientation map, nanotwin lamella with boundary spacing of 8 nm can be well distinguished in the surface SPD layer. PED coupled TEM orientation mapping is thus proved to be insensitive to severe lattice distortions and capable of suppressing various dynamical effects. Despite the success, additional attention should also be paid to orientation indexing of nanocrystallines near the topmost surface. As shown in Fig. 3d, a superimposed PED pattern constituted by four possible diffraction patterns was obtained from site C (Fig. 3a). Three most possible diffraction patterns, including <214>, <215> and <114>, show similar reflection intensity and correlation index value, but only <215> was recognized as the most reliable orientation of a nanograin. This pattern indexing method, despite insufficient evaluation of indexing reliability, can still not avoid probable misinterpretations of superimposed PED patterns arising from grain boundaries and junctions, or overlapped grains. The typical example given by Wu and Zaefferer [19] vividly showed that two overlapped grains with equal thickness but different orientation can coincidentally generate superimposed diffraction pattern as the same as another single crystal diffraction pattern. For electron beam transparent polycrystalline sample, the smaller the grain size, the higher the overlapping probability of nanocrystallines along the incident beam direction, and the easier the mis-indexing of superimposed PED patterns. Similarly, as the focused precessing electron beam is often highly distorted, the equivalent spot size becomes several times larger than its counterpart without precession. Thus the spatial resolution would be dramatically reduced, along with an increasing probability of superimposed PED patterns resulting from adjoining grains. Specifically, as the average size of nanocrystallines is approximated to the spot size of the focused precessing electron beam, the reliability of pattern indexing of superimposed PED pattern becomes even worse. Considering the experimental parameters used in this study, i.e. an electron beam with a precession angle of 0.6°, equivalent beam size of 5 nm and scanning step size of 4 nm, conservatively we can estimate that orientation indexing of nanocrystallines in IPF orientation map with diameters or spacings larger than 8 nm can be trusted, since at least 2 × 2 pixel or 4 independent pattern indexing has the same orientation. This estimation was fully proved to be reasonable and reliable by orientation indexing of nanotwin lamella with boundary spacing of 8 nm in Figs. 3e and 3g. Last but not least, we should point out that this critical grain size for orientation indexing reliability is not constant but more often changes with local sample thickness, overlapped grain number and percentage thickness, diffraction condition and so on. Pattern indexing results, especially those for nanocrystallines with a grain size in the same order of magnitude of the beam size, need careful reevaluation to maximally avoid probable misinterpretations. The recently developed TEM based three dimensional orientation mapping technique [20], however, may be a powerful way to solve this problem and precisely reveal the crystallographic feature of the overlapped tiny nanocrystallines near the topmost surface. Anyway, the high spatial resolution, together with high efficiency of pattern acquisition and indexing, fully indicates that the PED assisted TEM orientation mapping is practical and promising for two dimensional crystal orientation mapping of severely deformed structures at the nanoscale.

5. Conclusions
With the help of a TEM based PED technique, the orientation mapping of a surface nanostructure in SMGT Ni-based superalloy was carried out in addition to conventional TEM characterization. Dislocation activity and deformation twinning play distinct but cooperative roles in grain refinement during SMGT, while dislocation based plasticity is still in effect for nanocrystalline as small as 5-10 nm. A strong shear texture was identified in the surface nanocrystalline layer, which is in agreement with large scale SEM-EBSD analysis results. However, a minor Copper texture was also detected in the layer. Despite the deficiency to resolve the superimposed diffraction patterns, PED assisted orientation mapping is proved to be capable of mapping nanocrystalline metals with grain size down to a few nanometers in diameter, far smaller than what can be resolved in the commercial EBSD technique.
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