Dark-field X-ray microscopy reveals mosaicity and strain gradients across sub-surface TiC and TiN particles in steel matrix composites

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Metal-matrix composites (MMCs) based on steel reinforced with ceramic particles represent an attractive class of structural materials [1]. They are primarily used in wear and transportation applications due to high strength, elastic modulus and toughness, excellent wear resistance as well as low production costs combined with reduced weight [2]. The MMCs reinforced with high strength ceramics such as SiC, B₄C, Al₂O₃, TiB₂, TiC and TiN have been extensively investigated for their mechanical, thermal, microstructural and residual stress properties [3]. Numerous experimental and theoretical studies deal with the correlation of the internal microstructure, properties of interfaces, the load transfer between the matrix and the particles and the overall mechanical response of MMCs, as reviewed in Refs. [4–6]. A special attention was devoted to the analysis of the metal–ceramic interface reactions and their influence on through particle cracking, interfacial decohesion and the metal-particle load transfer [7–9]. Additionally, ex-situ and in-situ studies during or after high-temperature treatments have been used to characterize volume-averaged hydrostatic stresses, coefficients of thermal expansion (CTEs), chemical reactivity between the matrix and the particles, microstructure and transformation kinetics at temperatures up to 900°C or even higher [10–13].

A strain analysis was performed in the matrix phases and in the particles at the ambient temperature and during transformation sequences, indicating linear and non-linear strain evolutions in the phases as a result of CTEs mismatch and α–γ phase transformation, respectively [14–16]. Especially in-situ high-energy X-ray diffraction (HE-XRD) was applied to assess volume-averaged changes in lattice parameters and CTEs. In particular, X-ray elastic strains were investigated via the full width at half maximum (FWHM) analysis of Bragg peaks of α, α′, γ and cementite phases of the matrix as well as of the ceramics [14, 16]. The diffraction studies indicated pronounced changes in FWHM of steel α′ and γ phases during thermal cycling, as a result of thermal and transformation stress heterogeneities as well as martensitic formation. Comparably small FWHMs of TiC 220 reflections in exemplary steel–TiC system, however, were reported by Geandier et al. [16]. This was interpreted as evidence for negligible stress gradients within the particles, whereby a small FWHM magnitude decrease was observed when the temperature was increased in the range of 200–900°C [16]. To date, however, all HE-XRD studies have been devoted exclusively to the characterization of volume-averaged properties of the steel matrix and the embedded ceramics particles.

Although the technology and the scientific understanding of MMCs are at mature level, little is known about the spatial distribution of gradients of residual strains and crystallographic orientation (mosaicty) within sub-surface particles as a function of particular thermal treatments. These parameters are, however, needed to quantitatively assess the stress transfer in MMCs during macroscopic loading, for instance in view of micromechanical modelling in order to predict overall mechanical response of MMCs [17–19].

Recently, a novel synchrotron technique, dark-field X-ray microscopy (DFXM) using refractive optics, was introduced [20], which allows for the characterization of strain and crystal orientation gradients within sub-surface crystallites. By applying HE-XRD, the approach allows for 3D non-destructive mapping of local mosaicity and strain within embedded crystalline regions with ~100 nm spatial resolution [21].
In this work, DFXM was applied to characterize nanoscale gradients of crystallographic orientation and strain within TiN and TiC particles embedded in a steel matrix after two thermal treatments \[10, 12, 13, 22\]. The twofold motivation resides (i) in proving the feasibility of evaluating the orientation and strain gradients within exemplary ceramic particles and (ii) in comparing the results with conventional laboratory electron backscatter diffraction (EBSD) data collected from the same samples.

Two types of MMCs based on TiN and TiC particles were produced from a gas atomized steel powder (specified with \[0.46, 0.57, 0.42, 4.37, 2.06, 0.63, 1.6, 4.68\] mass % of C, Si, Mn, Cr, Mo, V, W, Co, respectively, and) with 10 volume % of ceramics, which were sourced from Sigma Aldrich. The d50 sizes of steel and ceramic particles were \(~19.9\) and \(~27\) \(\mu\)m, respectively. The two blended powder mixtures were consolidated by hot isostatic pressing at a temperature of \(~1140\)°C for 6 h, then austenitized in a vacuum furnace for 30 minutes, and then quenched with a quenching rate of \(\lambda = 0.4\) erg cm\(^{-3}\)s\(^{-1}\). Finally, two groups of MMCs with TiN and TiC particles each were tempered for 2 hours at 480 or 650°C, respectively. Then a water-cooled diamond saw was used to cut out four needle-like samples with dimensions of \(~0.3\times0.3\times20\) mm\(^3\). Based on our previous experimental work \[23\], we assume that the cutting did not significantly influence the stress state of particles located more than \(~30\) \(\mu\)m below the surface. These samples, differing in the type of the ceramic particles and the tempering temperatures, will be further denoted as TiN 480, TiN 600, TiC 480 and TiC 600. They were then char-acterized at the beamline ID06 of the European Synchrotron Radiation Facility (ESRF) using DFXM \[20\]. Complementary, EBSD analysis on equivalent electro-chemically polished samples was performed using an AU-RIGA CrossBeam systems (from Carl Zeiss) to obtain data on orientation spread across the ceramic particles. For the DFXM experiments, an X-ray beam with an energy of 33 keV selected by a Si(111) double monochromator was used. The incident beam was focused by means of two dimensional (2D) transfocators with Be compound refractive lenses (CRLs) having 14 lenslets, resulting in a 200 \(\times\) 200 \(\mu\)m (horizontal \(\times\) vertical) spot size on the samples. Diffraction intensities of TiN 111 or TiC 111 reflections (Fig. 1a) collected from (randomly selected) embedded grains within the samples were magnified by a X-ray objective comprising a SU-8 resist CRLs \[24\]. The projections of the grains were captured by two dimensional indirect detector with an effective pixel size of 1.4 \(\mu\)m (Fig. 1a). The magnification of the X-ray lens of \(~14.15\times\) provided an effective pixel size at the sample of \(~95\) nm and resolution of \(~190\) nm \[20, 25\]. The recorded images are projections, integrated along the beam path through the diffracting particles.

The crystallographic orientation (grain mosaicity) was assessed by scanning one (\(\omega\)) or two (\(\omega, \eta\)) sample axes while the scattering angle \(2\theta\) was fixed to the TiN 111 or TiC 111 reflection. The orientation scans were recorded with a step size of 0.02°over a maximum range of 0.6°, depending on the grain. The strain scans comprised a collective motion of the sample tilt \(\omega\), the detector and the objective to capture a \(\omega-2\theta\) mesh, at a constant \(\eta\) angle (cf. Fig. 1a). The results from the strain (\(\omega-2\theta\)) scans represented the local variations of the lattice spacing \(i.e\), axial strain) of TiN or TiC (111) crystallographic planes, whereas the orientation (\(\omega-\eta\)) scans at the constant \(2\theta\) angle indicate the spatial variation of the (111) plane normals’ orientation. The latter may be caused by either tilt or twist of the crystallographic lattice or due to the presence of shear strains. To distinguish between these two contributions, however, one would have to perform additional scans of other non-collinear Bragg reflections.
FIG. 2. Relative axial strain $\Delta 2\theta /2\theta_0$ distributions obtained from $\omega$–$2\theta$ scans in TiN and TiC particles annealed at 480 and 600°C (a, b, d, e) together with histograms showing the relative axial strain distribution spreads (c, f). The separate areas like in a, c, d, h, i and j may represent a single grain or two different grains that by coincidence have the same orientation within the gauge volume illuminated by the box beam.

This task was not performed in the present synchrotron experiment due to limitations in time and the goniometer geometry. For the data evaluation, we considered the intensity at a pixel coordinate as function of the goniometer angles across the scan. The centre-of-mass positions form 2D maps of strain and orientation. The integrated intensity over the scan provides reference images of the particle shape. Details of the generation of these maps can be found in Ref. [26]. Due to limited measurement time, however, only one ceramic particle was characterized by a series of scans in every sample and only 2D projections were evaluated.

In Figs. 1b–e, exemplary results obtained from a subsurface TiN particle in a TiN 600 sample are presented (from blue to yellow). The four DFXM 2D distributions express particle properties averaged along the scattering vector $Q$ directions being perpendicular on (111) crystallographic plane. The integrated intensity map from a $(\omega$–$\eta)$ scan in Fig. 1b shows strong and weak diffraction intensities from the grain interior and edges, respectively, as expected for a globular particle shape. Within the interior of the grain, interference fringes due to dynamic diffraction are observed, indicating the grain’s high crystal quality (Fig. 1b). A continuous variation of centre-of-mass $\omega$ values (from blue to red) in Fig. 1c obtained as a result of rocking curve analysis (by scanning the $\omega$ axis) indicates curvature of the crystal lattice across the particle. Additionally, regions with distinct colour code may represent sub-grains [27] of different crystallographic orientation and an orientation mismatch up to $\sim 0.01^\circ$. In Fig. 1d, a normalized axial strain $\Delta 2\theta /2\theta_0$ distribution obtained from a $\omega$–$2\theta$ scan show a relative variation in the Bragg’s angle position with pronounced negative and positive blue and red near-edge regions, which can be interpreted by the presence of respective tensile and compressive strain concentrations within the particle’s outer region. In Fig. 1e, the data from two dimensional orientation $(\omega$–$\eta)$ scans indicate again a continuous variation of crystal lattice curvature across the TiN particle along with the presence of distinct subgrains at the grain edges, in obvious correlation with the data in Fig. 1c. In general, the distributions in Figs. 1b–e (i) document gradual orientation changes across the particle and the probable formation of subgrains separated by ultra-low-angle boundaries [27] at the edges as well as (ii) indirectly suggest a presence of an outer envelope with a different strain state compared to the particle interior (cf. Figs. 1b, d).

A quantitative statistical analysis was established to evaluate strain and orientation distributions within the four analysed particles TiN 480, TiN 600, TiC 480 and TiC 600 (Figs. 2, 3). In Fig. 2, axial strain distributions (like in Fig. 1d) are presented together with histograms showing the strain spreads. In order to evaluate the strain distributions, reference $2\theta_0$ values were chosen at the grain positions with the highest intensities, in agreement with our previous studies [28, 29]. The re-
sults indicate a complex variation of the strain magnitude across the individual particles, with localized strain concentrations and/or relaxations. There are, however, three remarkable features visible in Fig. 2: (i) the TiN and TiC particles annealed at 600 °C (Fig. 2b, e) show the presence of envelope-like outer regions with different strain state than the particle interior. (ii) The strain spread in TiN particles is significantly smaller (by ≈75%) than in TiC (cf. Fig. 2c, f), and (iii) the strain spreads in both ceramics (TiN or TiC) do not change significantly after heating at 480 °C compared to 600 °C (Fig. 2c, f).

In Fig. 3, distributions of crystal lattice orientations obtained by ω and η rocking curve measurements together with histograms are presented. The results from TiN (Fig. 3a, b, f, g) indicate a presence of gradual orientation changes across the particles with a relatively large orientation spread, whereas a mosaicity (i.e. more localized changes between regions with approximately constant orientation) can be observed within the two TiC particles, indicating a presence of subgrains [27]. The histograms in Figs. 3c, f suggest that the annealing at 600 °C decreased the orientation spreads in both types of ceramics, a recovery effect which is common for nanoceramics synthesized far from thermodynamic equilibrium [30]. An interesting result can be seen in the TiN 480 grain upon a comparison of the strain and mosaicity maps in Fig. 2a, 3a and 3e. The strain map of the TiN 480 grain (Fig. 2a) shows no data for the lower half of the grain, while the orientation maps of the same grain (Fig 3a, 3e) shows the full shape of the grain with a vertical angular gradient. This is due to the decoupling of the lattice parameter variation and the lattice curvature. In fact, the curvature effect is reflected in the ω and η distributions, spanning a wide angular range. Consequently, the lower half of the grain fall outside of the η resolution window and is therefore not imaged in the (ω–2θ) scan.

The surfaces of the four investigated samples were electro-mechanically polished and further examined using EBSD in order to map local crystal orientation distributions across randomly selected representative grains. EBSD data were used to evaluate pole figures and inverse pole figures, which are presented in supplementary data, and relative local misorientation across the grains with respect to the average crystal orientation of every particular grain, which are presented in Fig. 4. The EBSD data indicated that (i) the TiN and TiC grains are actually single crystalline (cf. supplementary data), (ii) TiN grains appear to possess an envelope with slightly different crystallographic orientation compared to the grain interior (Fig. 4a,b) and (iii) all four analysed grains contain small misoriented regions appearing as lines (Fig. 4). Both latter features were attributed to the sample polishing procedure. In general, the EBSD analysis indicated that the grains in Fig. 4 are single crystalline and the observed variability in the orientation is very small, practically beyond the resolution limit of the used EBSD approach. We do not expect that the absence of the orientation gradients in Figs. 4 was caused by the sample surface polishing procedure, which has been routinely used to prepare ceramic samples exhibiting large mosaicity, like in Ref. [31].

The main difference between the DFXM (Figs. 1c, e and 3) and the EBSD (Fig. 4) orientation data is the fact that the DFXM revealed gradual changes in the crystallographic orientation and the presence of sub-grains, which were not visible in the EBSD maps. This is possible because of the better resolution function of DFXM, for both the orientation and strain with sensitivities of
FIG. 4. Relative orientation maps obtained using EBSD display the local crystallographic misorientation across electrochemically polished surfaces of four particles with respect to the average orientation of the particular grains (cf. supplementary data for the complete data).

0.1 mrad and $10^{-4}$ [20], respectively, compared to the EBSD angular resolution of 5–10 mrad [32], depending on the electron beam convergence.

The strain data in Figs. 1d and 2 show gradual changes in strain concentrations, which however do not always correlate with the orientation data (Figs. 1 and 3). In other words, the formation of subgrains with a different crystallographic orientation is not always correlated with the elastic strain gradients, whose origin could be therefore attributed to different strain states of grains’ interiors and borders. This effect is visible especially in the case of both particles heated at 600°C (Fig. 2c, f) and is in contradiction with results from metallic materials like in Ref. [28].

In summary, DFXM was used to characterize the crystallographic orientation and strain spread across four ceramic grains embedded in a steel matrix. It revealed significant strain and mosaicity gradients, which could not be observed using the complementary EBSD analysis. Even though the DFXM data represent only projected data along the diffraction beam path (Fig. 1a), they unambiguously indicate the complex elastic strain and mosaicity variations, which have been inaccessible using other techniques. In order to make representative quantitative conclusions on the state of TiN and TiC particles, further investigations on a larger number of particles will be necessary.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at https://ars.els-cdn.com/content/image/1-s2.0-S135964622030419X-mmcl.pdf.


