RESIDUAL STRESSES IN NOVEL METAL/CERAMIC COMPOSITES EXHIBITING A LAMELLAR MICROSTRUCTURE

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ABSTRACT

The aim of this study is to analyze the mechanics of a new class of metal/ceramic composite on a mesoscopic length scale. These composites are produced by melt infiltration of porous ceramic preforms produced by freeze-casting and subsequent sintering. This production route has a high application potential since it offers a cost-effective way to obtain composites with ceramic content in the range 30-70 vol%. The as-produced material exhibits a hierarchical domain structure with each domain composed of alternating layers of metallic and ceramic lamellae. Residual stresses present in all phases of the composite produced by infiltrating alumina preforms with a eutectic aluminium-silicon alloy have been measured. Integral as well as spatially resolved measurements were carried out on single domain samples at the high-energy, energy-dispersive diffraction (EDDI) beamline at the synchrotron radiation source BESSY (Berlin, Germany). Results show that strongly fluctuating residual stresses are introduced by the production process, which can be rationalized taking into account the thermal expansion mismatch of alloy and preform.

INTRODUCTION

Metal matrix composites (MMC) are technically important because of their high specific stiffness and strength, high wear and fatigue resistance and enhanced high temperature properties [1]. Conventional routes for MMC fabrication are reviewed in [2]. However, research is still going on to fabricate composites having novel property profiles and more efficient and economic processing routes. A new processing route has recently been opened by the availability of ceramic preforms processed by freeze-casting of ceramic suspensions. Details about the freeze casting process can be found in [3]. Ceramic preforms produced by freezing of water-based suspensions (slurries) have a typical hierarchical lamellar domain structure. The size and internal structure of these domains are controlled by the freeze-casting parameters [3, 4]. Preforms produced this way have excellent permeability for liquids and gases along with acceptable mechanical strengths and they are suitable for the fabrication of metal/ceramic composites by infiltration of liquid metal [3, 5]. This way it is possible to fabricate composites with ceramic contents in the range of about 30 to 70 vol%. This intermediate range is of particular interest since conventional particle- or fiber-reinforced composites typically contain either a relatively low (5–30 vol. %, e.g. [2]) or a fairly high (50–80 vol. %, e.g. [6]) reinforcement content due to processing constraints. Because of the difference in thermal expansion coefficients of the metallic and the ceramic phases, considerable thermal residual stresses are developed during processing of MMCs [7]. These thermal residual stresses may significantly affect the subsequent mechanical behavior of the composite material [8, 9].

The current study is aimed at gaining more insight about the mechanics of individual domains. Previous elastic [10, 11] as well as elastic-plastic [12] analysis have shown that the

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domains show pronounced anisotropy in their mechanical behavior. In the present study the lattice strain distributions were measured in a single-domain sample using energy dispersive synchrotron X-ray diffraction. Integral as well as spatially resolved measurements are carried out to determine the three principal stress differences in all three phases of the composite using the well known $\sin^2 \psi$– method [13]. Lattice strain fluctuations along and across the lamellae are determined by stepwise translation of the gauge volume within the sample. Thus a comprehensive picture of the residual stress state on single-domain level is obtained.

**MEASUREMENT PRINCIPLE**

High-energy synchrotron X-rays offer a unique combination of flux and penetration depth and thus allow stress analysis in the bulk work with fairly small gauge volumes [14, 15]. In conventional approaches for X-ray stress analysis, angle dispersive methods are applied using monochromatic X-rays to measure typically one single line profile. In contrast, a white beam is used in the energy dispersive approach to record a multitude of reflections over a broad energy range. The positions of both the detector and the sample are fixed, with the detector position being controlled by the choice of the fixed scattering angle $2\theta$. The value of $2\theta$ is chosen with respect to the appearance of the diffracting spectra in order to be able to separate the individual diffraction lines of the contributing crystalline phases by appropriate mathematical fit functions. The dimension of the resulting nominal gauge volume also depends upon the choice of the angle $2\theta$. Normally this angle is chosen in the range between 5 and 15°. In residual stress analysis the selection of the gauge volume is of prime importance because all the information is limited to this volume. The shape and dimensions of the gauge volume are controlled by slits placed in the path of the incident and the diffracted beams (see Figure 2). Because of the rather small scattering angle the gauge volume has the shape of an elongated diamond. The dimensions of the gauge volume can be calculated using simple geometrical relations using the value of the scattering angle, dimensions of the slits, distances of the slits to the sample etc. [16].

According to Bragg’s law, the energy of the diffracted peaks is related to the spacing of the corresponding planes according to:

$$E_{\text{hlk}} = \frac{h \cdot c}{\lambda} \cdot \frac{1}{2 \sin \theta} \cdot \frac{d_{\text{hlk}}}{d_{\text{hlk}} \cdot \sin \theta}$$

where $E_{\text{hlk}}$ is the energy of the \{hkl\} reflection in keV, $h$ is Planck’s constant, $\lambda$ is the wavelength, $c$ is the speed of light and $d_{\text{hlk}}$ is the spacing of the \{hkl\} planes in Å. The lattice strain is related to the shift in the peak energy according to:

$$\varepsilon = -\frac{\Delta d}{d_{\text{hlk}}} = \frac{\Delta E}{E_{\text{hlk}}}$$

**EXPERIMENTAL PROCEDURE**

Specimen material
Alumina preforms with open porosities of about 56 vol. % were produced at Institut für Keramik im Maschinenbau (IKM) at Universität Karlsruhe, Karlsruhe, Germany, via freeze-casting of a ceramic suspension and subsequent sintering. Water was used as the liquid vehicle during freeze-casting and the suspension contained 22 vol. % of alumina powder (CT3000SG from Almatis with nominal alumina content of 99.8 %, powder particle size 2.5 µm/D90 Cilas and a fired density of 3.90 Mg/m³.), 0.5 wt% Dolapix CE64 as dispersant and 10 wt% Optapix PAF60 as a binder. The freeze-casting temperature was -10° C. Freeze-cast
ceramic preforms were freeze dried for 48 hours and then sintered at 1550 °C for 1 hour. Afterwards they were brought to room temperature at a cooling rate of 4°C/min. Preforms with nominal dimensions of $10 \times 44 \times 66 \text{ mm}^3$ were infiltrated with a eutectic aluminium–silicon alloy (Al–12Si) at the Casting Technology Centre at Aalen University of Applied Sciences, Aalen, Germany, using a squeeze casting technique. Before infiltration the preforms were preheated to 800 °C and the press was heated up to 400 °C. After squeeze casting, the infiltrated samples were heated to 450 °C, held at that temperature for 2 hours and then subsequently furnace cooled. Figure 1a shows a typical microstructure of a poly-domain sample sectioned perpendicular to the freezing direction.

![Figure 1a](image)

![Figure 1b](image)

**Figure1:** Cross sections perpendicular to the freezing direction (= coordinate axis 1). In these optical micrographs the metallic alloy appears bright and ceramic dark (a) poly domain sample and (b) actual microstructure of the sample on which measurement was carried out. The lamellae are oriented parallel to coordinate axis 2.

The domain structure is clearly visible, with individual domains composed of alternating metallic and ceramic lamellae. To analyze the mechanics of the individual domains, small rectangular parallelepiped samples with dimensions between 1.8 to 2.6 mm were produced from the composite plates via wire cutting using 220 µm thick, diamond-coated steel wire. The edges of the final parallelepipeds were always aligned parallel to the edges of the composite plates and they had arbitrary domain orientations. Figure 1b shows the face perpendicular to the freezing direction for the actual sample used in this analysis. The lamellae in this sample are fairly parallel over the whole sample and their orientation with coordinate axis 2 is roughly 0° (this orientation scheme has been described in [10]).

**Energy dispersive synchrotron X-ray diffraction**

The diffraction experiments were carried out at beamline EDDI at the Berlin synchrotron storage ring BESSY, Berlin, Germany. A schematic of the beamline components is shown in Figure 2 while detail specifications of the beamline can be found in [17]. For the present study, an energy range between 20–90 keV was selected for analysis and a scattering angle $2\theta = 7^\circ$ was chosen as it gave good energy separation as well as sufficient peak intensities. For translation scans the slit size S2 was maintained at 200 µm×200 µm while slit system S3 and S4 had dimensions 30 µm×5 mm. Slit system S2 was increased to 1 mm×1 mm for $\sin^2 \psi$ measurements. A solid state germanium detector coupled with a multi channel analyser (MCA) was used to record the diffraction spectra.
Diffraction peaks were fitted by a “Pseudo-Voigt” function to determine the positions of the energy peaks.

RESULTS AND DISCUSSIONS

Figure 3 shows a typical energy-dispersive diffraction spectrum, where the individual diffraction peaks for each of the three contributing phases are marked. 8 diffraction peaks of alumina, 4 of aluminium and 3 of silicon were indexed for analysis.

Lattice strain fluctuations in the alumina phase for two different orientations were measured. As stated before, the domain has about 0° orientation with the specimen coordinate axis 2, while axis 1 is the freezing direction. As shown schematically in Figures 4a and b, the incident beam in both cases was parallel to axis 2, with the scattering vector being parallel to axis 1 in the first case and it being parallel to axis 3 in the second case. This was carried out by translating the gauge volume through the sample by moving the sample with a step width of 50 µm, while keeping the gauge volume fixed. Strain in the alumina phase was calculated using Eqn. 2, where the energy positions of the stress free material were determined from diffraction measurements on an uninfiltrated alumina preform. This was done by carrying out
sin²ψ measurements on a sintered alumina block cut from an uninfiltrated preform at 9 different ψ-tilts between 0-63.43°. Data analysis showed that the residual stresses in the preform range between ±10 MPa and hence the sintered preform was considered to be stress free. Hence, the average of the energy peak positions over the whole ψ-tilt for each alumina diffracting line was taken as the energy positions for the stress free state. Figures 4a and b show the schematics of the orientation of the scattering vector with respect to the freezing direction in the above mentioned two cases. Because of the chosen set-up the scattering vector is tilted 3.5° with respect to the sample co-ordinate system. In both cases the scanning direction was along specimen coordinate axis 3 (corresponding to 90° to lamellae direction; see Figure 1b).

Corresponding strain distributions are shown in Figures 4c and d. In these diagrams the individual lines correspond to individual diffracting lattice planes of alumina while the bold lines represent the continuum mechanics average micro strain. This average micro strain was calculated according to the method proposed by Daymond [18]:

\[ \varepsilon = \frac{\sum_{hkl} \alpha_{hkl} \varepsilon_{hkl}}{\sum_{hkl} \alpha_{hkl}} \]  

(3)

where \( \varepsilon_{hkl} \) are the individual plane strains and \( \alpha_{hkl} \) are weighting factors defined as:

\[ \alpha_{hkl} = \frac{T_{hkl} m_{hkl} E_{hkl}}{E} \]  

(4)
where $T_{hkl}$ is the texture factor, $m_{hkl}$ is the multiplicity factor, $E_{hkl}$ is the Young’s modulus of individual planes and $E$ is the macroscopic Young’s modulus of polycrystalline material. As a first approximation, the individual phases were assumed to be texture free (texture factor taken equal to unity). The shift in the lattice strain distribution for individual diffracting planes is due to the interplanar elastic anisotropy. There are significant strain fluctuations along the direction transverse to the lamellae which can be explained by the constraint imposed during the infiltration and solidification of liquid Al-12Si. Along the freezing direction the ceramic lamellae would be free from such constraints and accordingly less fluctuations of strain are observed here. The individual lamellae in the composite had thicknesses in the range of 20-100 µm, while the slit system for the incoming beam had an opening width of 200 µm. In order to monitor the local fluctuation of the lattice strains more accurately, a gauge volume having dimensions smaller than the lamellae would be ideal. However, this approach was not practicable due to insufficient grain statistics evoked by the small gauge volume. The chosen approach for using a larger gauge volume is practicable to monitor the local strain distributions in different directions.

Processing-induced (thermal) residual stresses in all three phases of the composite (alumina, silicon and aluminium solid solution) were determined using the \( \sin^2 \psi \) - method. To calculate the absolute phase-specific stresses it is a prerequisite to determine the lattice spacings of the stress free material for all contributing phases. This requires additional measurements on stress free materials having the same process history. However, this approach is not feasible for the material under investigation with justifiable effort. To overcome this shortcoming the three principal stress differences \((\sigma_2-\sigma_1)\), \((\sigma_3-\sigma_1)\) and \((\sigma_3-\sigma_2)\) were determined for the alumina and for the silicon phase, respectively. This was done according to the \( \sin^2 \psi \) - method for stress analysis by tilting the sample stepwise from 0°-90° for the respective planes (see Fig. 4 a, b). From these deviatoric stress components the von Mises equivalent stress was calculated using the following expression:

\[
\sigma_{VM} = \frac{1}{\sqrt{2}} \sqrt{(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2}
\]  

(5)

It was observed during measurement that \(2\theta\) changed slightly with time. This effect can be attributed to the fill level of the liquid nitrogen Dewar vessel necessary for the cooling of the solid state detector. By carrying out measurements on a reference sample before and after filling the Dewar it was observed that in the range of 20-90 keV the shift in the angle \(2\theta\) ranges between 0.0042-0.0058°. This caused a strain change \(\Delta \varepsilon = 1.2-1.6 \times 10^{-3}\). Since this effect is a long term effect, where the changes will only be seen on a time scale extending over some days, the slight changes in the \(2\theta\) angle have a negligible effect on the individual scans but may have a strong effect on the calculated absolute d-spacings. To overcome this, only changes in d-spacing as a function of \(\sin^2 \psi\) are shown in Figure 5. The measured d vs. \(\sin^2 \psi\) - results were fitted by a straight line and the intersection with \(\psi = 0^\circ\) was chosen as a reference. This procedure can be followed since only deviatoric stress components in different orientations are of interest and not the absolute d-spacing. Among the three phases, the results of aluminium always showed a particularly strong scatter, which can be attributed to the relatively large grain size of this phase. Hence, the further analysis is based on the results for alumina and silicon only. In Figure 5, the results obtained for the (214) reflections of alumina and the (311) reflections of silicon are shown. The continuum mechanics average deviatoric stresses were calculated for silicon and alumina using Eqns (3) and (4). These as well as the von Mises equivalent stress calculated there from using Eqn. 5 are shown in Table 1.
Figure 5: Change in d-spacing vs. $\sin^2 \psi$ plot for single-domain sample: (a) (311) planes of silicon and (b) (214) planes of alumina

Table 1: Continuum mechanics equivalent deviatoric stresses and the von Mises equivalent stress in alumina and the silicon phases of the single domain sample

<table>
<thead>
<tr>
<th></th>
<th>Alumina</th>
<th>Silicon</th>
</tr>
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<tbody>
<tr>
<td>$\sigma_3-\sigma_1$ (MPa)</td>
<td>74</td>
<td>-154</td>
</tr>
<tr>
<td>$\sigma_2-\sigma_1$ (MPa)</td>
<td>-7</td>
<td>-17</td>
</tr>
<tr>
<td>$\sigma_3-\sigma_2$ (MPa)</td>
<td>98</td>
<td>-179</td>
</tr>
<tr>
<td>von Mises equivalent stress (MPa)</td>
<td>87</td>
<td>-167</td>
</tr>
</tbody>
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Table 1 clearly illustrates that in both alumina and silicon, the absolute values of the components ($\sigma_3-\sigma_1$) and ($\sigma_3-\sigma_2$) are similar and also significantly higher than the amounts of ($\sigma_2-\sigma_1$), which are near zero in both of the phases. Although from these results it is not possible to determine the absolute values of the individual stress components, they definitely suggest that in both alumina and silicon $\sigma_{22} \approx \sigma_{11}$. This is due to the fact that the sample makes 0° orientation with axis 2 and hence the processing induced stresses along axes 1 and 2 are almost similar. Also, the signs of the stress differences are opposite in silicon and alumina. As the silicon phase is dispersed in the form of very fine particles in aluminum phase, it can be assumed that its behavior images that of the Al-12Si alloy. The opposing signs of the stresses in the alumina and silicon phases are thus due to the different thermal expansion coefficients of the alumina preform and the infiltrated metallic alloy. The results are thus in agreement with the necessary condition that the overall composite as a whole is free of macroscopic residual stresses.

CONCLUSIONS

Energy-dispersive X-ray diffraction has been shown to be a viable technique for analyzing the deviatoric stresses as well as the local strain fluctuations in a novel metal/ceramic composite having a lamellar microstructure. For the first time the process-induced residual stresses of this kind of composite have been assessed. The results clearly indicate that strongly fluctuating local phase specific (micro) residual stresses are present in the as-produced state which can be explained taking into account the thermal expansion mismatch of the alloy and the ceramic preform. Analysis of the deviatoric residual stress components show that only small amounts of phase specific stresses are induced by the squeeze casting process. The
results indicate that these phase specific residual stresses show directionality with respect to the freezing direction of the composite. Taking into account the phase contents of the contributing phases the material emerges to be nearly free of macroscopic residual stresses within the measurement uncertainties as expected for the applied manufacturing process.

The chosen stress analysis approach appears ideal for successive studies on the internal load transfer from the soft and compliant metallic alloy to the hard and stiff ceramic occurring when the composite is subjected to external loads. In the next analysis step, in-situ diffraction experiments will be performed for single domain samples being subjected to uni-axial compressive loading. Results of this analysis will be presented in a subsequent work.

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REFERENCES