X ray analysis of the mechanical state of a Nickel based multicrystal on the mesoscopic scale: Role of the grain orientation and its boundary

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ABSTRACT

The use of the well known sin^2ψ-method allows to determine the macroscopic internal stresses in an isotropic, small grained and untextured material. Second order stresses at the scale of the grain can be determined by using a single grain measurement method. Depending on the relation between the dimensions of the grain and the beam on the sample, an average of the grain stress state can be determined. Still using a single grain measurement but inside the grain allows to investigate the influence of the grain boundaries on the grain stress state.

In order to combine an experimental and a numerical approach, a large grain size is necessary (between one and four millimeters). In finite element calculations, the grains are meshed and therefore the number of grains has to be known to introduce the multicrystal. The grains are supposed to cross the sample and the grain boundaries can be approximated by perpendicular walls.

After determination of the grain orientation it is possible to measure the peak shift of several crystallographic planes compared to the non deformed material. The calculation of the elastic strain tensor based on the peak shift measurements leads to the determination of the stress tensor at the scale of the irradiated volume in one single grain. Using a large collimated beam, the average stress state of one grain in a polycrystalline matrix is determined. The analysis of the stress heterogeneities in one grain needs smaller beams. The use of fiber optics allows beam spots of 100 to 200 μm.

A tensile device is mounted on a four circle goniometer, the second order and the mesoscopic stress state are determined under successive loading in several neighbouring grains. The elastic and plastic anisotropy of the "single crystal" in the polycrystalline matrix is the source of the inhomogeneous behaviour of the whole grain. Finite element simulations calculate the elastic and plastic strain and confirm heterogeneous behaviour on the scale of the grain.

From the local stress values, the resolved shear stress of each slip system can be calculated. The evolution of the local stress tensor is linked to the evolution of the resolved shear stresses of the slip systems, which allows to conclude on the hardening law of the grain average and the grain boundary.

Complementary analysis confirm the slip activity of the grain such as the profile broadening of Rocking curves.

INTRODUCTION

So far, crystallographic models such as the Sachs, Taylor or selfconsistent model [1,2] have been able to take crystallographic slip into account, but without any aspects of morphology of the polycrystalline matrix.

More powerful computers, like the sp2 at the Ecole des Mines in Paris, allow now to take the grain structure into account by discretizing the grains using finite elements. In this way, it is possible to investigate the local stress and strain fields inside the grain.

The adapted experimental techniques to validate the local stress field is the single grain measurement method by x-ray diffraction. Microscopic observations are done to characterise the plastic slip.

New types of beam collimation and better optics allow finer measurements which will be confronted to the numerical simulations. Structural calculations which take into account the
microstructure of the polycrystal will justify polycrystalline approaches like selfconsistent modelling or pure phenomenological approaches like Hill or Von Mises [3].

The x-ray diffraction technique was already applied by different authors:

Reimers and al. measured the crystallite-crystallite interactions of a coarse grained (Ø≈1mm up to Ø≈5mm) Ni-base alloy IN 939 under applied load in the elastic range using neutron diffraction. Strain heterogeneities inside one grain could be shown using synchrotron radiation [4].

Gergaud and al. used the single grain measurement technique to investigate the radial evolution of the residual planar stresses in a coarse grained silicon billet elaborated by an electromagnetic cold crucible using a continuus casting process. The crystal quality of the photovoltaic silicon was very good compared to a metallic material, so that optimisation algorithms were not applied [5].

Lebrun and al. introduced this measurement technique for two phase coarse grained material like a cast duplex stainless steel [2, 6].

In the present study a Ni-base alloy IN 600 close to a welding zone was used. The microstructure in the heat affected zone is characterised by the strong heterogeneity in the grain sizes (Fig. 1). A simplification of the microstructure was necessary to allow finite element calculations of a whole structure with a reasonable grain number. The recrystallisation process initialised by the temperature gradient introduced by the welding process was continued until the grain growing stage. A long heat treatment of 140 hours at 1570°K gave a homogeneous grain distribution of grains in the range of Ø≈1mm up to Ø≈5mm. Unavoidable side effects of such a heat treatment is the thermal twinning of face centered cubic materials. Depending on the size of the twins, they were considered like independent grains or they were neglected.

SINGLE GRAIN MEASUREMENT METHOD

Polycrystalline measurement techniques, like the sin²θ-method [2], average over a large number of grains in the diffracting volume, so that the measurement directions are isotropically distributed in the space. Second order stress measurements on the scale of the grain are confronted to the single crystal anisotropy and the measurement directions have to be determined depending on the grain orientation in the sample. An orientation matrix defines the relationship between the crystal and sample reference system. The acquisition of a polefigure using a four-circle goniometer set-up allows the determination of the orientation matrix in an automatic, but quite time consuming way. Using a Scanning Electronic Microscope (SEM) equipped with an electron sensitive camera, the exploitation of the Kikuchi diagrams gives the orientation matrix of several grains in a very fast way (Electronic BackScattering Patterns, EBSP). In keeping the
same macroscopic reference system, the sample is mounted on a four circle goniometer and geometrical calculations determine the spherical co-ordinates of the investigated (hkl) crystallographic planes.

The electron beam size is in the order of microns, so that the determination of the crystal orientation is very local. In general the X-ray beam is much larger, hence the analysed volume averages over a larger part of the crystal. The calculated orientation matrix is not very precise and an optimisation algorithm is necessary for each analysed plane [8]. Even the use of a pole figure acquisition makes the matrix determination not very precise and an optimisation process is also necessary. Reimers showed [8], that two approaches of (hkl) measurements are possible, which is either the refinement of the orientation matrix or the optimisation of each hkl -pole individually. Comparable measurements of the two approaches showed, that the second one is more precise.

In figure 2 the (hkl) reflection of a grain is schematised by an ellipsoid in the degrees of liberty of the laboratory reference system. The broadening is due to the experimental broadening and especially to the crystal quality in the diffracting volume. The inaccurate position calculated by the orientation matrix is somewhere in the ellipsoid and provokes offsets in omega-2theta positions and leads therefore to undesired pseudo stresses. The searched position is the center of gravity of the ellipsoid.

A series of omega, omega-2theta and psi scans is repeated until the peak shift of each scan stays in the experimental resolution error (Fig. 3).

The measurements allow to determine six unknowns of the strain tensor, which makes at least six measurements necessary. Using between 12 and 15 (hkl)-planes reduces statistically the experimental error bars on the strain and stress values. The elastic strain tensor is calculated by using the metric tensor formalism [5]. Applying the Hook law determines finally the stress tensor on the diffracting volume.

A uniaxial tensile device was mounted on a four axial goniometer to investigate the accommodation of the strain between and within grains under load. The local stress state was
measured at different loading stages. The local mechanical behaviour of the grain can be compared with the macroscopic behaviour of the sample using strain gages covering the whole strained zone and a piezoelectric force measurement device.

Discussion

The experimental difficulties cover especially the crystalline quality in the diffracting volume. During plastic strain the mosaicity or the lattice misorientation, is getting more important. The omega profiles (or Rocking-curves) of the optimisation scans are broadened and even peak splitting can appear (Fig. 18). Due to the anisotropic mosaicity evolution, the measured (hkl) planes used for the strain measurement are more or less sensitive on the peak broadening. The broadening of the Rocking-curves depends strongly on the crystallographic orientation of the rotation axis. For the (hkl) family measured, the quality of the signal is getting poorer due to the plastic strain, so that the peak positioning is determined with different accuracy depending on the peak quality. Finally the error bars of the stress determination at higher strain levels are getting more important. Very small beam sizes in the order of microns would minimise the averaging volume and would also avoid the average over several mosaics.

Long range internal stresses [7] corresponding to third order stresses, are responsible for a non symmetric broadening of the omega-2theta peaks, which raises the error bars for the correct determination of the peak position, as well.

Due to the optimisation process and the precise omega-2theta scan (0.01° step size, between 5s and 10s per step) the measurements in one grain can take several hours which introduces a slight relaxation of the sample, so that the applied local stress level at the beginning of the measurement is slightly higher than at the end.

In x-ray analysis for stress measurement applications, the penetration depth of the x-rays in the material is supposed to be low, so that the stress normal in the sample reference system can be taken equal to zero. This hypothesis allows the calculation of the stress free lattice parameter and then the metric tensor $G_0$. Figure 5 shows the calculated lattice parameter at different measurement points and at different loadings of one grain. The oscillations lower than 0.0005 Å show the good quality of the measurements and confirm that the use of the hypothesis $\sigma_{33,\text{sample}}=0$ is permitted. The influence on the stress values of the experimental fluctuations are shown in [6].

HETEROGENEOUS 2nd ORDER STRESS STATES IN A NI-BASED MULTICRYSTAL

Like already previously mentioned, different orders of stresses are distinguished. Second order stresses average the stress behaviour of the whole grain. The measurement of volume fractions inside the grain using smaller beams, could be defined as intermediate between second and third order stresses, which characterise the stress field on the dislocation scale.
Two samples are presented, the first is measured by a divergent beam averaging over the whole grain and the second by a collimated beam of 100μm beam size measuring inside the grain.

Figure 6 shows the microstructure of the flat multicrystalline tensile sample. The marked grains 3, 8 and 13 are the grains which will be discussed more closely. The sample was deformed until 1.5% macroscopic strain and the macroscopic stress raised until about 120MPa. The macroscopic deformation was kept lower than two per cent to stay in the approximation of small perturbations used in the finite element calculations. Another reason is the fast evolution of the mosaicity at higher plastic strain which makes the measurements more difficult, as discussed before.

Results of the local stress analysis

Single grain stress measurements were done at different loading stages. The (22)-component of the analysed stress tensors is the component in tensile direction. Figure 7 compares the macroscopic and the local mechanical behaviour as a function of the macroscopic strain, which is of course not representative for the local strain of the grain. Microgrid measurements would have been necessary to get the characteristic strain of one single grain. In loading direction grain 8 and 13 have a very close behaviour, lower than the applied stress. Grain 3 shows slightly higher stress values, which presents already a heterogeneity at the grain scale looking only at the tensile direction.

Regarding on the results of the stress tensors at 1.0% macroscopic strain, three different stress states can be observed. Grain 3 is characterised by a uniaxial stress state with a even higher stress value in loading direction. The lateral direction and the shear stresses are close to zero. Grain 8 shows high compressive values in the lateral direction, but still very low values concerning the shear stresses, which is not true for grain 13. Strong compressive shear stresses in the order of the lateral stress are found.

Grain 3: \[ \begin{pmatrix} -13 & -11 & -6 \\ -11 & 159 & -5 \\ -6 & -5 & 0 \end{pmatrix} \]
Grain 8: \[ \begin{pmatrix} -64 & -1 & -25 \\ -1 & 91 & 14 \\ -25 & 14 & 0 \end{pmatrix} \]
Grain 13: \[ \begin{pmatrix} -55 & -45 & 12 \\ -45 & 95 & 2 \\ 12 & 2 & 0 \end{pmatrix} \]

Fig. 8: Stress tensors in three grains at 1% macroscopic strain

The introduction of multiaxial stresses are the effects of the polycrystalline matrix and the boundary effects. Due to their orientation, the grains have different slip directions corresponding to their slip systems, each characterised by its own critical stress. The yield stress depends on the
actual hardening and stress state of the grain, so that in more complex stress field the grain behaviour can be quite different compared to the behaviour of a single crystal under uniaxial loading.

Analysis of the plastic slip

Starting the interpretation of the grain behaviour, it is easier to compare with a free single crystal, what is done in table 1. The Schmid factors are calculated for the 12 slip systems in a face centered cubic material. The projection of the slip plane and the slip direction define the Schmid factor (Fig.9).

The three grains show quite high Schmid factors (bolded, table 1) which leads to a quite strong plastic activity of the concerned slip systems.

<table>
<thead>
<tr>
<th>Grain</th>
<th>(111)</th>
<th>(-111)</th>
<th>(1-11)</th>
<th>(11-1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>system</td>
<td>-[110]</td>
<td>-[101]</td>
<td>[01-1]</td>
<td>[110]</td>
</tr>
<tr>
<td>3</td>
<td>0.07</td>
<td>0.46</td>
<td>0.40</td>
<td>0.17</td>
</tr>
<tr>
<td>8</td>
<td>0.13</td>
<td>0.33</td>
<td>0.45</td>
<td>0.00</td>
</tr>
<tr>
<td>13</td>
<td>0.18</td>
<td>0.31</td>
<td>0.13</td>
<td>0.50</td>
</tr>
</tbody>
</table>

Table 1: Classification of the slip systems

Grain 3 and 13 represent two extreme stress states, one almost single crystalline behaviour, the other in a complex multiaxial stress state. Microscopic observations of projections of the activated slip planes confirmed the activity of the slip planes with the highest Schmid factors. A good agreement between the calculated and measured angles confirm the primary slip of the three grains. The micrographs of grain 3 and 13 are represented in figure 10.

The determination of the local stress state at different loadings allows the representation of the shear stress evolution of each slip system. The shear stress is the projection of the stress tensor on the slip plane and the slip direction: \( \tau = m \cdot \sigma \cdot n \) (\( m \) = slip plane, \( n \) = slip direction, \( \sigma \) = local stress tensor, fig. 9)
In figure 11 the shear stresses of grain 3 and 13 are shown with a characteristic behaviour of a more uniaxial (grain 3) or multiaxial stress state (grain 13).

For grain 3 only the well orientated systems with the highest Schmid factors (figure 11) get off the zero level.

The more complicated morphology of grain 13 is one reason for the multiaxial stress state which could lead to a multiple slip if the critical stresses for the concerned systems are reached.

Still, the microscopic observations confirm only the slip of the best orientated slip system (-111)[110], which shows that the critical stresses of the other slip systems could not be overwhelmed even in this complex stress state.

Fig. 11: Evolution of the shear stresses of grain 3 and 13 (the numeration of the slip system corresponds to table 1 from the left to the right)

EXPERIMENTAL AND NUMERICAL APPROACH FOR A MULTICRYSTAL UNDER APPLIED LOADING

Next step is the confrontation of the experimental results with the numerical simulations. A second sample with a known number of grains is discretised. Optimising the experimental data, the sample was chosen with a comparable microstructure on each side of the sample. Figure 12 shows the microstructure of the two faces of a real multicrystal and the modelled one. The verification of the grain nominations were done by Electronic Backscattering Patterns. Compared to the former experiment, a micro focus was used. A pipe collimator of 100μm beam size was installed and local measurements inside the grain were done. A motorised sample holder allowed the sample movements in a reproducible way.

Figure 12 shows an example of two polefigures of the same grain at each side of the sample. If it is the same grain the polefigures (here presented for the (220) family) are only distinguished by a symmetry around the (11) axis. A look on the side of the flat tensile sample shows that the majority of the grains are crossing through the whole thickness of the sample. Grains which do not pass the whole sample thickness are approximated due to their importance of the whole microstructure of the multicrystal.
Results on the local stress behaviour

The determined stress tensors inside grain 6 and 7 show a quite homogeneous stress state in the error interval of the measurement. Remarkable are the shear stresses, which have the tendency to get more important close to the grain boundary.

Only three measurements were done in the grains 4 and 5 because of the important plastic strains very localised in this zone (figure 16 b). Quite important diffraction peak splitting was observed, so that the measurement were getting quite difficult.

As already discussed in the chapter ‘single grain measurement’, the relaxation of the sample played an important role. The maximal applied load was around 100 MPa but was decreasing during the whole measurement of about 10% which explains the relatively low stress values in tensile direction of the grains 5, 6 and 10. The large plastic deformation of grain 4 leads to strong stress heterogeneities inside the grain, so that higher and lower stress values in tensile direction could be found. The shear stresses are still staying quite high.

As already mentioned, the plastic deformation was very localised in the top of the sample due to the orientation distribution of the grains in the multicrystal. A zoom on the deformed microstructure in figure 14 shows the grain boundary of grain 4, 5, 6 and 7. A clear frontier of plastic slip can be observed. Grain 4 and 5 have got a twinning orientation relationship, so that the projections of the activated slip systems on the surface of the sample are parallel.

The orientation of the grains determines the distribution of the Schmid factors (tab. 3).

<table>
<thead>
<tr>
<th>Grain</th>
<th>(111)</th>
<th>(1-11)</th>
<th>(-111)</th>
<th>(111)</th>
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<td>[110]</td>
<td>[101]</td>
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<tr>
<td>4</td>
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<td>0.18</td>
<td>0.30</td>
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<td>5</td>
<td>0.14</td>
<td>0.12</td>
<td>0.03</td>
<td>0.27</td>
</tr>
<tr>
<td>6</td>
<td>0.25</td>
<td>0.31</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>7</td>
<td>0.21</td>
<td>0.29</td>
<td>0.08</td>
<td>0.12</td>
</tr>
<tr>
<td>10</td>
<td>0.17</td>
<td>0.18</td>
<td>0.02</td>
<td>0.15</td>
</tr>
</tbody>
</table>

Table 3: Schmid values of several grains in sample 2
The two systems close to 0.5 explain the single slip of the two grains 4 and 5. The systems could be identified in figure 14. In the grains 6, 7 and 10 the Schmid factors are smaller than 0.42 and no slip systems have been observed on optical micrographs.

A confirmation of the plastic activity of the grains can be done by a qualitative analysis of the broadening of omega-2theta peaks. Averaging over the Full Width of Half Maximum (FWHM) of the considered (hkl) planes for each stress measurement and for each measurement point, a distribution of the local hardening state can be observed (figure 15). The important plastic strain of the grain 4 and 5 can be confirmed by the highest dislocation density corresponding to a higher 2 theta peak broadening. Grain 7 and 10 stayed more or less in the elastic domain, so that they can be considered as the reference width. Grain 6 was plastically active (much less than grain 4 and 5). This difference between grain 6 and 7 is not fully coherent with the Schmid factor calculation. Grain 7 shows a higher Schmid factor than grain 6, but looking on the numerical results in figure 17, the plastic activity was lower. The strong plastic activity of grain 4 and 5 in the close neighbourhood of grain 6 introduces shear stresses in form of strain bands, which activated a slip system with a relatively low Schmid factor of 0.36. Numerically the system could be identified, but this has not been confirmed by optical observations.

**Numerical results**

The applied single crystal model was developed in a viscoplastic frame with threshold for plastic slip [9]. The only introduced plastic deformation process are the 12 octahedral slip systems in face centered cubic materials already presented. The Schmid law is verified at each calculated Gauss point with its proper orientation. The shear stress for each of the 12 slip systems is calculated (1) and compared to the actual threshold depending on the hardening state of the grain (4). The viscoplastic shear strain is then calculated and finally the viscoplastic strain tensor using the Schmid law for each slip system is determined (3).

The kinematic ($\dot{\gamma}$) and isotropic ($\dot{\gamma}$) hardening (2) depend on the variables $\alpha^\beta$ and $\dot{\gamma}^\beta$ (3, 4). The formulation of the hardening laws in this way allows saturation of the hardening. The matrix $h_{rs}$ takes into account the interactions of the different slip systems.

**Constitutive equations:**
Some results of the mechanical behaviour simulation of the sample are given in figure 16. Resuming the experimental analysis by the numerically calculated mechanical behaviour, the individual grain behaviour in the multicrystalline matrix can be confirmed in figure 16 a. Grain 4 shows the most important strain compared to grain 10 which stays in the elastic regime. Considering grain 4 and 10 in a single crystal calculation conserving their orientation, the soft orientation of grain 4 and the harder one of grain 10 could be confirmed. Grain 10 itself is even harder than the whole multicrystal. The mean strains in grain 4 (respectively 10) are much higher (respectively lower) if the grains are considered within the polycrystal. Comparing the numerical and experimental results of the macroscopic behaviour of the multicrystal, a good agreement could be found.

In figure 16 b and c the total strain in loading direction is presented and the Von Mises stresses. The most important strain is localised in grain 4, creating a band until grain 6 where the strongest deformations are observed at the edge of the sample. The largest strain of grain 4 is localised in the center of the grain, far from any boundary conditions. The stress state, in contrary, shows the highest values close to the grain boundaries. These high stress gradients are found very close to the grain boundaries, so that very small beam sizes would have been necessary to show the tendency.

Comparing the stress components in tensile direction, the model and the experiment show a quite acceptable agreement. Only grain 10 shows experimentally a quite low value, although it stayed in an elastic regime, but a possible explanation could come from the relaxation of the sample.

<table>
<thead>
<tr>
<th>Stress in tensile direction</th>
<th>grain 4</th>
<th>grain 5</th>
<th>grain 6</th>
<th>grain 7</th>
<th>grain 10</th>
</tr>
</thead>
<tbody>
<tr>
<td>experiment</td>
<td>114±43</td>
<td>59±38</td>
<td>/0±4/</td>
<td>69±32</td>
<td>98±26</td>
</tr>
<tr>
<td>model</td>
<td>115</td>
<td>92</td>
<td>86</td>
<td>108</td>
<td>111</td>
</tr>
</tbody>
</table>

Table 4: Experimental and numerical results of the stress component in tensile direction

Concerning the other components of the stress tensor, experimentally the shear stresses are much more important than numerically, where the multiaxial stress state remains concentrated at the grain boundary. One reason is surely the heterogeneous mosaic distribution which will be discussed in the following.
Local evolution of the mosaicity

The plastic strain inside one grain is very heterogeneous due to the grain boundaries. This observation can be confirmed by presenting only one active slip system in the grain (fig. 17). As the total strain, the highest slip activity is concentrated at the center of the grain. This leads to a heterogeneous distribution of crystallographic misorientations, which have been neglected in the numerical simulations, so far.

Local measurements inside the grains showed very heterogeneous Rocking curve distributions which prove this local disorientation inside the grain by broadening of the Rocking curves or even splitting of peaks which reveals the creation of subgrains.

Figure 18 shows an example of two separated measurement points in grain 6 which was even less strained than grain 4. The beginning of a subgrain structure can be observed in point 1.

Fig. 16: Comparison of the microscopic and macroscopic behaviour (a), Local strain: component in tensile direction (b), von Mises stress (c)

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Fig. 17: Primary slip of grain 4

Fig. 18: Rocking curve profiles of two measurement points in grain 6
CONCLUSION

A multicrystalline approach was presented to investigate the stress and strain heterogeneities in a polycrystalline material. The single grain measurement technique was applied to determine second order stresses over the whole grain volume and local measurements inside the grain. Rocking curve analysis confirmed the strong heterogeneities concerning the lattice rotations. Finite element calculations using a semi phenomenological approach were able to simulate the stress and strain field in the measured multicrystal. Comparing the numerical and experimental results, the diffracting volume still is not sufficiently small to observe the strong stress heterogeneities close to the grain boundary, so that synchrotron radiation is necessary. The local mosaicity evolution showed the necessity of the introduction of the lattice rotations in the finite element code, even in small strain regimes.

REFERENCES