Influence of Cross Rolling and Heat Treatment on Texture and Forming Properties of Molybdenum Sheets


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Abstract

To produce molybdenum sheets or ribbons the metal has to be thermomechanically processed leading to characteristic deformation and recrystallization textures depending on the deformation and/or annealing conditions. The so produced textures have an impact on certain properties of the metal. The present work concentrates on the influence of different rolling processes and the last step of heat treatment on microstructure, texture and the mechanical properties of molybdenum sheets. Usually, unidirectional rolling leads to a strengthening of the main texture component, which for molybdenum is a weak $\alpha$-fibre with a maximum at the rotated cube component $\{100\}<110>$. This component leads to a strong anisotropy of the mechanical properties in the sheet plane. Cross rolling steps during the thermomechanical process increases the rotated cube component. To decrease the intensity of the rotated cube component and simultaneously increase the intensity of the $\gamma$-fibre, specific annealing stages of the final heat treatment were tested. The texture development during these annealing stages is discussed with regard to microstructural changes. The strong differences in the textures measured are reflected in the plastic anisotropy characterized by the Lankford parameters. The Taylor-Bishop-Hill theory is used to qualitatively explain the plastic anisotropy.

Keywords

Molybdenum, rolling, microstructure, texture, tensile tests

Introduction

Molybdenum is a refractory metal with wide use. The high melting point, high temperature strength, low vapour pressure, low coefficient of thermal expansion, as well as good thermal and electrical conductivity are the basis for application in lighting, glass, furnace, nuclear industry and electronics. The weak oxidation behaviour at temperatures above 400-500°C makes an oxygen free atmosphere necessary for applications at elevated temperatures.

For industrial production of molybdenum parts sheet rolling and deep drawing are important processes. Preforms of compacted and sintered molybdenum powder have to be hot and cold rolled with annealing treatments generally necessary between the rolling passes. As a consequence, specific crystallographic
Textures and microstructures are produced depending on the thermomechanical process applied. The present investigations concentrate on the influence of different rolling processes and the last step of heat treatment on microstructure, texture and the resulting mechanical properties of molybdenum sheets.

The relaxed constraints (RC) Taylor model is used to calculate the Lankford parameters $r_i$ of the sheets for different directions as well as the technically relevant normal and planar anisotropies, $\langle r \rangle$ and $\Delta r$, respectively [1].

**Experimental**

**Samples**

All molybdenum sheets were produced in the following way: Plates of pressed and sintered Mo powder were reduced in thickness and porosity by means of hot rolling. After that, they were heated up to about 1000°C - 1400°C and rolled to an intermediate stage. If cross rolling was applied it was also done under warm condition. The final rolling steps took place at room temperature (cold rolling). Between the steps annealing treatments in hydrogen atmosphere were necessary to decrease the hardness for further deformation and preserve the ductility of the sheets.

<table>
<thead>
<tr>
<th>sheet</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
</tr>
</thead>
<tbody>
<tr>
<td>cross rolling</td>
<td>at early stage</td>
<td>at late stage</td>
<td>no</td>
<td>complex changes in RD</td>
<td></td>
</tr>
</tbody>
</table>

In table 1 the production route for different samples is given. The rolling direction (RD) was changed by 90° for sheets A and B. For sheet A the change was at an earlier stage during the hot rolling, whereas it was done at a later stage of hot rolling for sheet B. Sheet C was unidirectionally rolled. The main texture component of rolled molybdenum sheets is the rotated cube component. To break the symmetry of the rolling process a complex route including 45°, 90° and 135° changes in RD was used for D and E. Sheet E was additionally annealed at the end of rolling. All sheets had a final thickness of 0.2 mm. The limited width of sheet B did not allow a complete mechanical characterization.

**Texture measurements**

Three pole figures (110, 200 and 211) measured by X-ray diffraction using a $\theta$ - $2\theta$ goniometer (HZG 4, FPM Freiberg, Cu K$_\alpha$-radiation) with Euler cradle in back reflection mode [2] were used as input for calculating the orientation distribution function (ODF) with a computer program developed by Dahms [3]. The degree of series expansion used for the calculation is 22, the Euler angles used are in the Bunge notation [4]. The pole figure measurements were carried out at the surface and in the central layer of the sheets. The preparation of the samples was done by mechanical grinding. The main texture components of rolled bcc metals are given in the $\varphi_2 = 45°$ -section of the ODF (see fig. 1).

**Microstructure**

The microstructure of the sheets was investigated by backscattered electron (BSE) orientation contrast imaging in a scanning electron microscope (SEM, Zeiss DSM 962). The samples were grinded,
mechanically polished and finally electropolished at about -10°C and a voltage of 18 V. The electrolyte consisted of 20 vol% HCl, 20 vol% H₂SO₄ and 60 vol% CH₃OH.

Tensile tests / Lankford parameters

The tensile tests were done with a Zwick/Roell Z250 testing machine in combination with the optical system ARAMIS (GOM company). The sample geometry is shown in fig. 2. For calculation of the local strains, changes in a pattern sprayed on the sample surface were analyzed.

Fig. 1: Location of the main texture components of rolled bcc metals in the 2 = 45° ODF-section

Fig. 2: Sample geometry for tensile tests

The Lankford parameter \( r_\alpha \) (eq. 1) is the ratio of logarithmic reduction of width to logarithmic reduction in thickness during tensile test parallel to the direction \( \alpha \) within the sheet plane. For calculation of the Lankford parameters constant volume was assumed. In the following the mean Lankford parameters analyzed over the whole sample area are presented.

\[
r_\alpha = \frac{d \varepsilon_{\text{width }, \alpha}}{d \varepsilon_{\text{thickness }, \alpha}}
\]

The value of \( r \) is zero for plane strain deformation with elongation in tensile direction while it is one for isotropic reduction in thickness and width. For ideal deep drawing of sheets \( r \) should be larger than one. For better characterizing the deep drawing behaviour the normal Lankford parameters (eq. 2) and their planar anisotropies (eq. 3), \( <r> \) and \( \Delta r \), respectively, are used. These parameters are derived from tensile tests in three directions (RD, 45° to RD and TD = transverse direction) as follows:

\[
<r> = \frac{1}{3}(r_{RD} + 2r_{45°} + r_{TD})
\]
\[ \Delta r = \frac{1}{2}(r_{RD} - 2r_{45^\circ} + r_{TD}) \]  

Deep drawing

Discs with a diameter of 55 mm were cut from the sheets by spark erosion. The final diameter of the deep drawn cup was 33 mm. In fig. 3 the deep drawing tool is sketched schematically.

![Deep drawing tool](image)

The disc was deformed by lowering the plunger with the edge hold down by a blank holder. The force applied was small enough to guarantee slip of the disc between drawing die and blank holder. The cups deformed in this way were analyzed with respect to earing defined by the parameter \( Z \) (eq. 4) according to DIN EN 50155

\[ Z = \frac{\langle h_e \rangle - \langle h_v \rangle}{\langle h_v \rangle}, \]  

with \( \langle h_e \rangle = \) mean height of ears, \( \langle h_v \rangle = \) mean depth of ears.

Modelling of the plastic anisotropy

For calculation of the Lankford parameters the RC Taylor model and the Taylor-Bishop-Hill theory of crystal plasticity was used [5-8]. First, the textures measured at different sheet positions were summed up and discretized in 1000 single orientations using a Monte Carlo code. This set of orientations served as input for the RC Taylor program. By relaxing two shear strains \( \varepsilon_{zx} \) and \( \varepsilon_{zy} \) in the sheet plane the flat grain shape was taken into account. Next, the Taylor factor \( M \) was calculated as a function of strain mode by modifying the deformation tensor systematically. The strain in tensile direction was kept fixed, while the ratio of strain in width to thickness direction, i.e. \( r \) was varied. The \( r \) value at minimum Taylor factor describes the deformation condition requiring the least amount of plastic work. The extraction of \( \langle r \rangle_{\text{calc}} \) and \( \Delta r_{\text{calc}} \) from \( r_\alpha \) yields information on the deep drawing behaviour based on the texture alone.

Results and Discussion

Texture

The main texture components of the sheets are the rotated cube \{100\} \(<110>\) and \{112\} \(<110>\) component, both located on the \( \alpha \)-fibre (\(<110>\ || \ \text{RD})\). As shown in fig. 1, the \( \alpha \)-fibre contains all
orientations with <110> parallel to RD, including the two main components {100}<110> and {112}<110>. The \(\gamma\)-fibre orientations with \{111\} \(\parallel\) the rolling plane are less pronounced. All orientations with \{111\} parallel to the rolling plane are located on the \(\gamma\)-fibre. For deep drawing the rotated cube component produces a high anisotropy in the sheet plane leading to earing. High texture intensities on the \(\gamma\)-fibre are generally important for good deep drawing properties of bcc metals [9, 10]. This situation results in an in-plane mechanical isotropy and the Lankford parameter becomes much larger than 1 [11]. The textures of the samples A, B, C, D and E at the surface and in the centre of each sheet are given in fig. 4 by \(\phi_2 = 45^\circ\) ODF sections. Intensity plots along the \(\alpha\)-fibre are presented in fig. 5 for the surface and centre of the sheets.

The rotated cube component with a spread along the \(\alpha\)-fibre characterizes the texture of sheet A. The through-thickness gradient of the texture in this sheet is small. The texture is much stronger in sheet B and consists of the rotated cube as the main component. Sheet C has a weaker texture with an incomplete \(\alpha\)-fibre. The maximum position changes from near {115}<110> at the surface to an orientation near {112}<110> in the centre. There is also a change on the \(\gamma\)-fibre from {111}<112> at the surface to {111}<110> near the centre. The maximum ODF intensities are lower than in sheets A and B. The textures of D and E are quite similar. However, the maximum intensities are the weakest compared to A, B and C. The main component is near the rotated cube orientation and the relative strength of \(\gamma\)-fibre orientations is higher than in the other sheets. The through-thickness gradient of texture is negligible in D and E.

The textures observed are in agreement with those reported in the literature [12-14]. Low straight rolling reductions produce an incomplete \(\alpha\)-fibre with the maximum at the rotated cube component {100} <110>. The maximum shifts to {112} <110> at higher degrees of rolling. Cross rolling shifts the stable {112} <110> component from the \(\alpha\)-fibre to \{112\} <111>. This component is not stable during rolling leading to a texture dominated by the rotated cube component. The through-thickness texture gradient results from differences in the deformation mode. Near the surface additional shear deformation influences the texture components. Towards the centre the share of shearing decreases leading to an almost ideal plain strain deformation.
Fig. 4: Textures of samples A, B, C, D and E represented by $\phi_2 = 45^\circ$ ODF sections in the surface and centre of the sheets (maxima are given in multiples of a random distribution m.r.d.)
Microstructure

The microstructures of the three sheets (A, B and C) after the standard final heat treatment procedure differ in the amount of recrystallization, see fig. 6. The microstructure of sheets A and C shows recrystallized grains, the recrystallized volume fraction being small. The recrystallization has a through-thickness gradient. The size of the recrystallized grains in the central area is larger than near the surface. The first recrystallized grains form colonies with an elongation in RD [15]. The deformed grains have pancake shape and are strongly elongated in RD. The thickness is about 1 µm. Sheets B and E are recovered while D (without final annealing) shows a deformation structure typical for high rolling deformation. The (sub)-grain structure is highly elongated in RD and TD [8, 15].

In molybdenum recrystallization and grain growth during the final heat treatment do not change the type of texture. Three different temperature regimes have been distinguished during final heat treatment. At low temperatures, (regime I, up to 700°C) extended recovery takes place. The medium temperature regime II (750°C - 800°C) is characterized by partial recrystallization by strain induced boundary migration. In the high temperature regime III (above about 850°C) the sheets recrystallize totally. Simultaneously, grain growth takes place. In all regimes the type of texture does not change drastically, but the relative intensities of the different texture components are shifted. The heat treatments applied did not produce a strong and homogeneous γ-fibre favourable for good deep drawing [8, 15, 16].

Mechanical anisotropy

The mechanical properties measured show a high anisotropy. The Lankford parameter $r$ in the case of sample A for tension parallel RD and TD is about 0.1 while for 45° it is 1.25. This means that for tension in RD and TD the deformation in direction of the thickness is larger than the deformation in direction of the width. Tensile tests under 45° show opposite results. The strong differences lead to disadvantageous deep drawing properties, i.e. the cups show strong earing in this case.
B and C sheets were too small to perform tensile tests in all three directions. In the case of sheet B, it was possible to cut samples for tension in RD and under 30° to RD. The Lankford parameters for 45° and TD are 0.77 and 0.32 for sheet D and 0.62 and 0.27 for sheet E, respectively. The anisotropy of the Lankford parameter is smaller than for sheet A and all three measured values of the Lankford parameter are smaller than one. For this texture a lower development of ears is expected, but the sheet will become thinner during deep drawing. These assumptions are supported by the calculated normal and planar anisotropies.
To quantify the deep drawing behaviour, the mean height and depth of the ears was measured and $Z$ calculated (table II). The results of modelling and experiment are compared in table II. For the Lankford parameters there is a quantitative agreement for most of the samples. Moreover, $Z$ almost linearly increases with increasing $\Delta r$. The deviation of the calculated and measured mechanical parameters may in part be attributed to the effect of grain shape, latent hardening and dislocation structure which has not been taken into account in this work [17, 18].

<table>
<thead>
<tr>
<th></th>
<th>$r_i$</th>
<th>$&lt;r&gt;$</th>
<th>$\Delta r$</th>
<th>$Z$</th>
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<tr>
<td></td>
<td>exp.</td>
<td>calc.</td>
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<td>A</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>RD</td>
<td>0.11</td>
<td>0.11</td>
<td>0.67</td>
<td>0.79</td>
</tr>
<tr>
<td>TD</td>
<td>0.08</td>
<td>0.05</td>
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<tr>
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</tr>
<tr>
<td>RD</td>
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<td>0.05</td>
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<tr>
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<td>45°</td>
<td>1.00</td>
<td>1.00</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TD</td>
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<tr>
<td>C</td>
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<tr>
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<tr>
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<tr>
<td>D</td>
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<tr>
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<td>E</td>
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<td>TD</td>
<td>0.27</td>
<td>0.18</td>
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</table>

**Conclusions**

(i) The rolling process independent of cross rolling steps produces an incomplete $\alpha$-fibre with the maximum intensity located at the rotated cube component $\{100\} <110>$. The maximum shifts to $\{112\} <110>$ at higher degrees of rolling. Cross rolling shifts the stable $\{112\} <110>$ component from the $\alpha$-fibre to $\{112\} <111>$ which is not stable during further rolling. This finally leads to a texture dominated by the rotated cube component. The textures of samples with a complex rolling procedure (e.g. changes under 45° to RD) are quite similar and the maximum intensities are weaker compared to the other samples. The relative strength of $\gamma$-fibre orientations is a little bit higher than in the other sheets.

(ii) The microstructures of the three sheets produced in accordance to standard production (rolling direction changed only under 90°) differ in the volume fraction which is recrystallized. The microstructure after the final heat treatment is characterized by three different temperature regimes marked by different portions of recrystallized volume, respectively grain growth. In all regimes the type of texture does not change drastically during the final heat treatment, but the relative intensities of the different texture
components are shifted. The heat treatments applied did not produce a strong and homogeneous \( \gamma \)-fibre favourable for good deep drawing.

(iii) Earing of the sheets, observed during deep drawing experiments is in good accordance to experimental determination and calculations of the Lankford parameter and its planar anisotropy.

(iii) The RC Taylor theory qualitatively explains the plastic anisotropy.

Acknowledgments

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References