

Sample cutting-free pole figure measuring method for centreless diffractometers in modified X mode

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ABSTRACT

Conventional X-ray diffraction-based pole figure measurements are realized with dedicated equipment which are accompanied with certain limitations such as sample size, geometry, and no option for on-site measurements. These restrictions limit the availability of texture measurements in cases when sample cutting from the object of interest is not allowed. Within this manuscript, a novel method is described with which pole figure measurements can be realized using centreless diffractometers originally developed for residual stress measurements. The theoretical background of the proposed method is presented in detail. Instances of pole figures obtained with the described method are presented and compared to conventional ones. It was concluded that the presented method is applicable to measure pole figures in a reliable manner with all the benefits of centreless diffractometers: no need for sample cutting, large geometrical freedom and the possibility for on-site measurements.

1. Introduction

The isotropy of macroscopic properties (yield strength, elongation, electric and magnetic properties, etc.) of polycrystalline materials strongly depend on the orientation of individual crystals (or unit cells) [1,2]. Therefore, knowing the orientation of crystals, i.e. crystallographic texture, is essential in the field of materials science [3–6]. The semi-quantitative description of texture is realized through pole figure measurements. Pole figures give the spatial intensity distribution of the beam diffracted by a given $\{hkl\}$ lattice plane series, relative to the coordinate system of the sample. On the polar coordinate system of pole figures, the reference direction (RD) - which is the rolling direction in case of rolling - points to the 12 o' clock direction. χ (chi) is the angle of sample tilting (more precisely, the angle between the normal of the sample surface and the normal of the diffracting plane series) increases radially. The ϕ (phi) coordinate gives the clockwise rotation around the normal direction (ND) of the sample (more precisely, the angle between RD and the perpendicular projection of the normal of the diffracting plane series on sample surface), which increases clockwise [1,2,7–9]. Conventional X-ray diffractometers developed for pole figure measurements rotate and tilt the sample relative to the beam path while recording the intensity of the selected $\{hkl\}$ reflection. To realize these movements, measurable samples are restricted in size [10]. Also, for most equipment, the measured surface must be flat. These sample

restrictions limit the applicability of conventional texture measurements.

Centreless diffractometers have been developed during the past decades for residual stress measurements even on large-scale objects with complex geometry without the need of sample cutting. Avoiding sampling is essential in order not to disturb the original stress field of the object. Some of these devices are also portable; therefore, they are suitable for on-site measurements. In contrast to conventional diffractometers, the measured objects are stationary here, while centreless diffractometers themselves are tilted around the objects. During tilting, the intensity of a given $\{hkl\}$ reflection is measured from different directions. Since centreless diffractometers had been developed for residual stress measurements, most of them are capable of measuring reflections only in the large 2θ range (typically, in the $2\theta = 120^\circ$ – 170° range). However, with proper tube selection, the $\{220\}$, $\{222\}$, $\{311\}$, and $\{400\}$ or $\{200\}$ reflections - which represent the deformation and recrystallization texture of most common FCC and BCC metals (Al, Cu, austenite, ferrite) [11] - is within the measurable 2θ range. The prominent advantage of centreless diffractometers over conventional devices is that they can also be used in situations where sample cutting is not allowed. The most obvious examples for such cases arise in the field of archaeometry, where artefacts have relatively large dimensions and sample cutting is strictly forbidden, yet, information regarding the manufacturing technology of the artefacts is strongly desired. Pole

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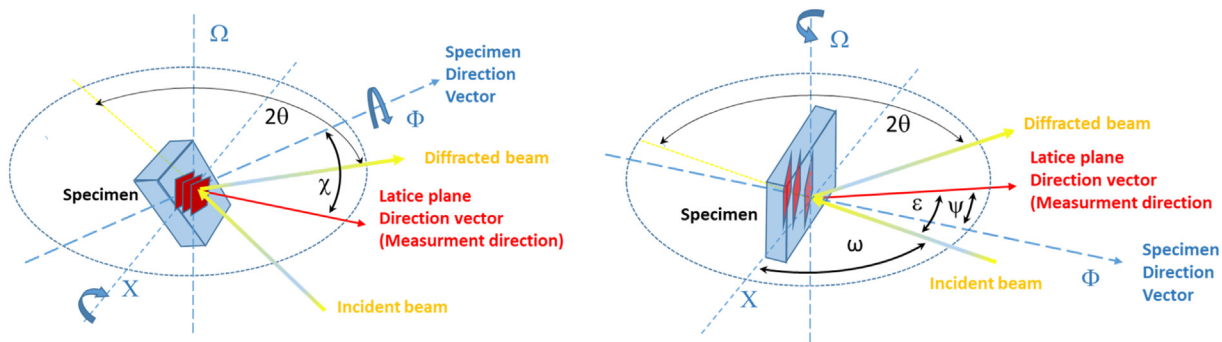


Fig. 1. General scheme of beam path, tilting axes and relevant angles of X-ray goniometers.

figures represent the type (rolled, recrystallized, cast, isotropic, etc.) and magnitude of texture. Therefore, if pole figure measurements can be realized on an artefact without doing any harm, it can provide reliable information about its texture, from which the manufacturing technology can be deduced. Theoretically, in such situations, electron backscattered diffraction (EBSD), neutron diffraction, or synchrotron can also be a choice [12–15]. However, these methods also have object size limitations and the latter is rather rare; therefore, none of them provide applicable solution. In addition, centreless diffractometers use small, variable spot sizes (typically $\sim 0.5\text{ mm}$ – 5 mm in diameter). By selecting the proper spot size, objects with curved surfaces can also be measured with negligible defocusing error (resulting from tilting) [16]. Thus, the examination point is not restricted to flat surfaces. The benefits of portable centreless diffractometers could be utilized in the field of metal sheet manufacturing as well. With portable centreless diffractometers, pole figure measurements could be carried out on coils even with industrial dimensions. Thus, there would be no need to rip up large-scale coils to investigate its texture. This is much favoured if texture variation along the length of the coil is of the question. Finally, if pole figure sections are sufficient instead of pole figures, measurement time can be significantly reduced. These benefits clearly point out that the development of sample cutting-free (non-destructive) pole figure measurement methods is reasonable and can bring texture measurements to areas where it has not been applied before.

In this manuscript, a new method is presented with which pole figure measurements can be realized with centreless diffractometers in modified X (CHI) mode (also referred to as modified ψ mode). The method is based on finding the correlation between pole figure coordinates (χ and ϕ), the tilting and rotation angles of centreless diffractometers and Bragg-angle of the examined $\{hkl\}$ reflection. This is obtained through mathematically connecting the measurement directions of conventional pole figure measurements and those of centreless diffractometers. The described method is applied on cold rolled 3003 type aluminium sheets with different production histories. The obtained pole figures are compared with conventionally measured pole figures.

2. Experimental

For validation, one of the most commonly used formable alloys, 3003 type cold rolled aluminium sheets were investigated. In the case of formable alloys, such as the examined 3003 type, knowing the crystallographic texture is crucial since the rolled sheets can undergo notable plastic deformation such as deep drawing. The cold rollings were performed with a VonRoll experimental roll stand having 220 mm roll diameter, using 50 m/min rolling velocity and commercial Palmerol (mineral oil-based liquid) lubricant. The examined sheets were cold rolled from 4.8 mm thickness to 1 in one step, or to 0.2 mm in one or two steps with total average strains (in plain strain conditions) of 1.8 and 3.7, respectively. The average strains of rolling were calculated according to Eq. (1)

$$\bar{\epsilon} = \frac{2}{\sqrt{3}} \ln \frac{h_0}{h} \quad (1)$$

where $\bar{\epsilon}$ is the average strain, h_0 is the initial and h is the final sheet thickness. Conventional pole figure measurements were carried out with a Bruker D8 Advance type powder diffractometer equipped with an Eulerian cradle and $\text{CoK}\alpha$ radiation source (wavelength: 179.02 nm) operating with 40 kV tube voltage and 40 mA tube current. Pole figures were measured up to 75° tilting with 5° increment and visualized by the own TexEVAL software of the equipment. The new method was introduced by using a Stresstech XStress 3000 G3R type centreless diffractometer with $\text{CrK}\alpha$ source operating with 30 kV tube voltage and 8 mA tube current. The absorption corrected pole figures were visualized by Origin software. For both equipment, absorption correction was performed on Al powder.

2.1. Measurement method

In general, three different modes can be distinguished based on goniometer tilting relative to sample: Ω (OMEGA), X (CHI) or Ψ (PSI) and modified X or modified Ψ . The term “ Ψ mode” is widespread in Europe, while “X mode” is more commonly used in North America [17]. The names refer to the axis around which the goniometer circle (holding the source and detectors) is tilted. Here, a universal nomenclature system (Fig. 1.) is applied in which the three different modes can be univocally distinguished. Axes are represented with Greek uppercase letters while tilting/rotation angles with Greek lowercase letters. The X axis is the intersection of the plane of sample surface and the plane of the goniometer circle. Tilting around X axis is indicated by the angle χ . It is the angle between the normal of the sample surface and the normal of the diffracting plane series (i.e., the measuring direction). The Ω axis lies within the plane of sample surface and it is perpendicular to the goniometer circle. The Ω axis is thus perpendicular to the plane of primary (incident) and diffracted beam. The ω angle is interpreted as the angle between the primary beam and the sample surface (or the X axis). While rotating/tilting around Ω axis, ψ angle forms between the measuring direction and the normal of the surface of the sample. The Φ (PHI) axis is normal to the sample surface. Rotation around Φ axis is marked with ϕ , which increases clockwise.

Conventional pole figure measurements are carried out in X mode. In a traditional equipment, the sample is tilted around the X axis (Fig. 2 a, b). Centreless X-ray diffractometers typically operate in modified X or Ω modes. Here, only the modified X mode (marked as X') is discussed. (Axes, angles, directions regarding a centreless diffractometer system are distinguished by apostrophe (') from the conventional ones.) In this mode, the equipment is tilted around its X' axis (Fig. 2 c, d). However, the main difference between the X and X' modes, is not the tilting choice of sample or goniometer, but the different beam paths. This is more easily seen in Fig. 3, which represents the schematic beam paths of a conventional diffractometer in X mode (Fig. 3 a) and a centreless equipment in X' mode (Fig. 3 b, c). In X mode, the measurement

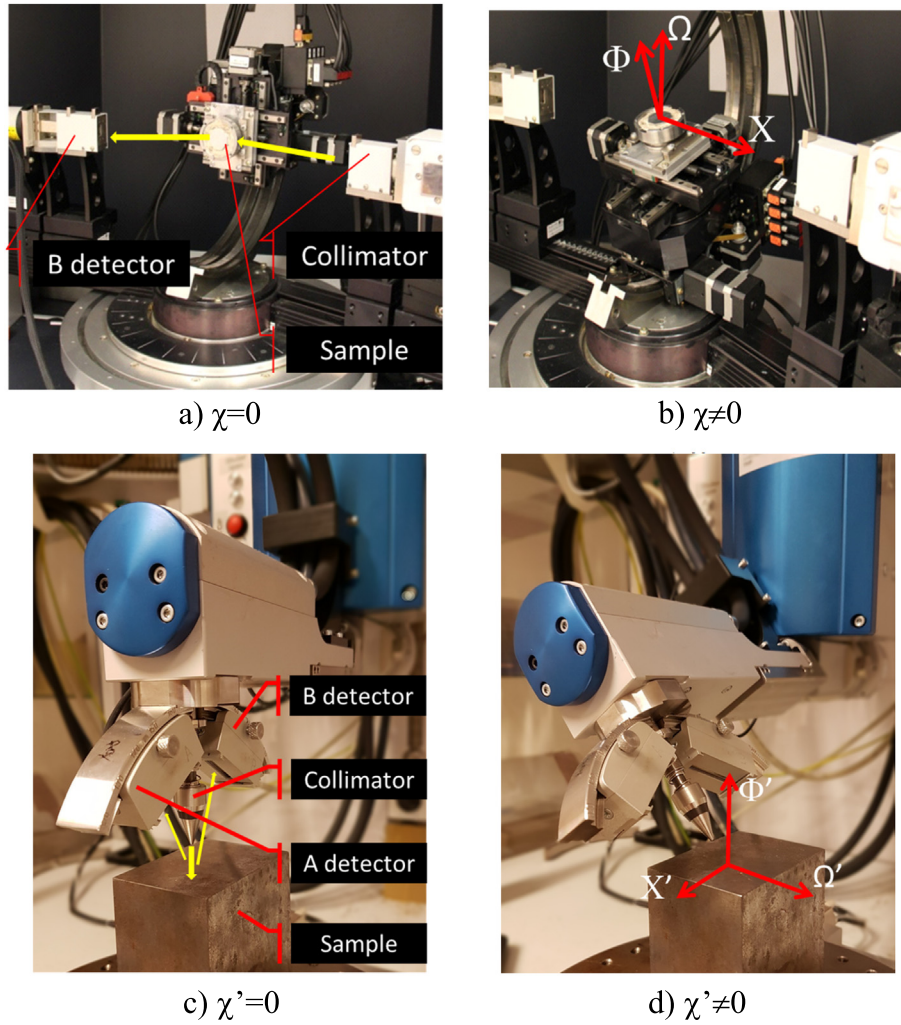


Fig. 2. Beam path of different diffractometer set-ups. a), b) conventional diffractometer in X mode, c), d) centreless diffractometer in X' mode.

direction (m_A) is perpendicular to X, while in X' mode, it is the incident beam (S_0') that is perpendicular to X'. The measurement directions (m_A' and m_B' , in the case of a two-detector set-up) close the angle ψ' with S_0' , since $2\psi'$ is the complementary angle of $2\theta'$. Thus, ψ' can be calculated from the Bragg angle ($2\theta'$), which is determined at 0° tilting, by Eq. (2).

$$\psi' = 90^\circ - \theta' \quad (2)$$

In order to deduce the mathematical correlation between X and X' modes, let's examine the realized measurement directions of the two cases. During conventional pole figure measurements, the sample is tilted with a defined χ value around X axis and is rotated around the Φ

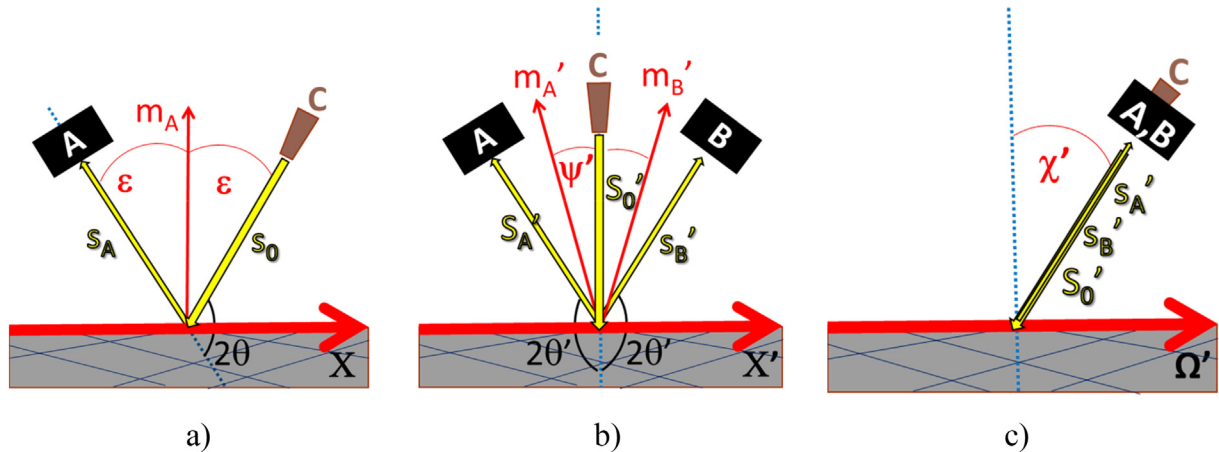


Fig. 3. The scheme of beam paths of different diffractometer set-ups. a) conventional equipment in X mode at 0° tilting viewed from the Ω axis, b) centreless diffractometer in X' mode at 0° tilting, viewed from the Ω' axis, c) at a general tilting, viewed from the χ' axis (C-collimator, A, B detectors, S_0 , S_0' incident beam, S_A , S_B , S_A' , S_B' diffracted beams, m_A , m_B , m_A' , m_B' -measurement directions).

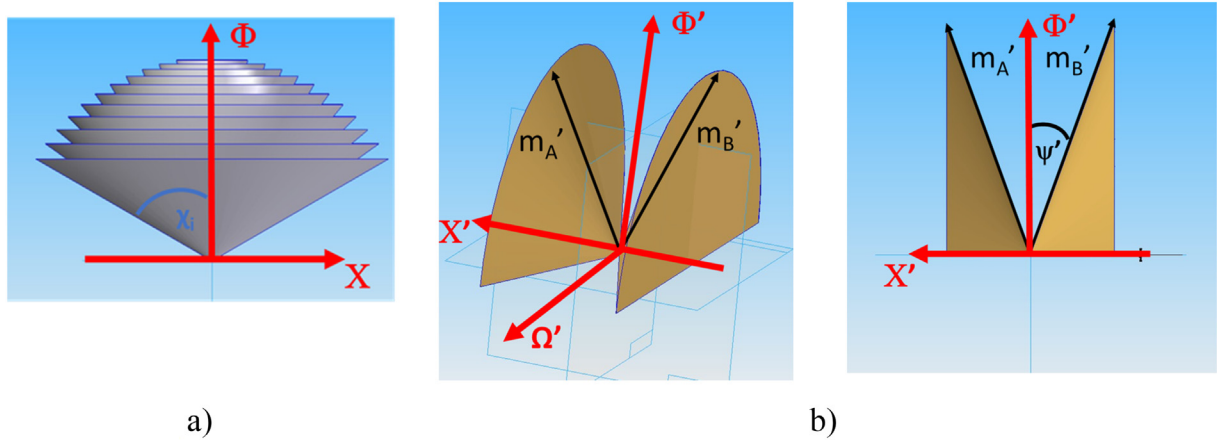


Fig. 4. Theoretical view of the paths of measurement directions a) during X mode pole figure measurement b) and X' mode up to $\pm 90^\circ \chi'$ tilting.

axis. During the full rotation around Φ , the measurement directions form a measurement cone with half apex of χ . The rotations performed at every χ_i tilting result a series of measurement cones (Fig. 4a).

During the operation of centreless diffractometers in X' mode, the measuring head with the collimator and detectors is tilted around the X' axis with defined χ' angles. Thus, the paths of measurement directions (of detectors A and B) form half cone pairs with half apex of $(90-\chi')$. In the figure, these cone pairs are drawn up to $\pm 90^\circ \chi'$ tilting. If the cone pairs of the centreless diffractometer are inserted into the measurement cone series of the conventional pole figure measurement, the intersections of the two cone systems represent the measurement directions that can be realized with both equipment. Determination of the coordinates of the intersections is the crucial point in measuring pole figures with centreless diffractometers.

For simplicity, let's consider the length of cone generatrix unity and deal with only one cone (m_B') of the pair (Fig. 5). At $\chi' = 0^\circ$ angle tilting, P is the intersection of m_B' and one of the measurement cones of X mode. M' is the perpendicular projection of m_B' on the tilting axis (X) and can be calculated according to Eq. (3) (Fig. 5):

$$M' = \sin(\psi') \quad (3)$$

R_i , the perpendicular projection of m_B' on the sample surface can be calculated according to Eq. (4). (In Fig. 5, R is rotated around the centre of the cones to show its true length and the referring triangle).

$$R_i = \sin \chi_i \quad (4)$$

Note that at $\chi' = 0^\circ$, M' and R are equal (Fig. 5). This is because at $\chi' = 0^\circ$, m_B' intersects the measurement cone of which the half apex (χ) is ψ' . (This will not be the case at $\chi' \neq 0^\circ$ tilting.)

U' , the distance of P from X can be calculated according to Eq. (5) (Fig. 5).

$$U' = \cos \psi' \quad (5)$$

H_i , the distance of P from the sample surface can be calculated according to Eq. (6) (Fig. 5).

$$H_i = \cos \chi_i \quad (6)$$

Again, at $\chi' = 0^\circ$, U' and H are equal (Fig. 5), for the reason above. (And again, this will not be true at $\chi' \neq 0^\circ$.)

At $\chi' \neq 0^\circ$ tilting, Q marks the intersection of m_B' and one measurement cone (Fig. 6). In this case, U' is the distance between Q and X and H is the distance of Q from the sample surface. In this position, the value of M' and U' are the same as were at 0° tilting, since the measurement head is rotated around X' axis; ψ' is also constant; and R and H can be calculated similarly as in the case of $\chi' = 0^\circ$ tilting angle Eqs. (4) and (6), respectively (In Fig. 6, R again is rotated around the centre of the cones to show its true length and the referring triangle). Now, m_B'

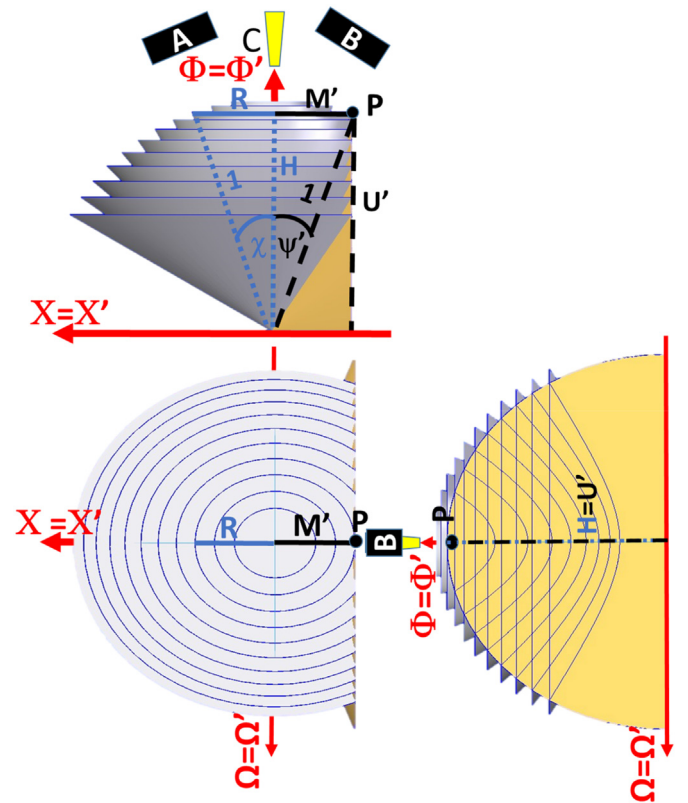


Fig. 5. Theoretical view of measurement directions of X and X' modes at $\chi' = 0^\circ$ tilting. a) (front) view from the direction perpendicular to the tilting axis (X'), b) (top) view from the primary beam, c) (side) view from the tilting axis (X').

intersects a different measurement cone of X mode than previously, thus, $M' \neq R$ and $U' \neq H$. This is clearly shown in Fig. 6.

The tilting angle of the centreless equipment (χ') as a function of χ pole figure coordinate and ψ' (derived from the Bragg-angle) can be calculated according to Eq. (7).

$$|\chi'| = \arccos\left(\frac{H_i}{U'}\right) = \arccos\left(\frac{\cos \chi_i}{\cos \psi'}\right) \quad (7)$$

The desired ϕ coordinate can be set by aligning the sample relative to the centreless diffractometer according to the following: α is the angle between R and Ω axis and can be calculated according to Eq. (8) (Fig. 5).

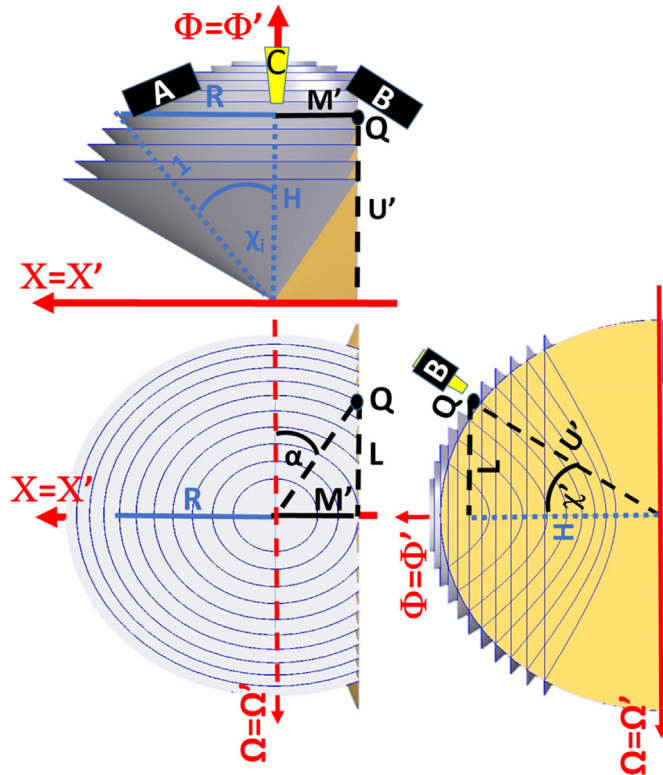


Fig. 6. Theoretical view of measurement directions of X and X' modes at $\chi' \neq 0^\circ$ tilting. a) from the direction perpendicular to the tilting axis (X'), b) from the primary beam, c) from the tilting axis (X').

$$\alpha_i = \arcsin\left(\frac{M'}{R_i}\right) \quad (8)$$

Then, φ can be calculated according to Eqs. (9), (10), (11) and (12) within the I., II., III. and IV. quarter of the current coordinate system, respectively (Fig. 7), where β is the angle between Ω and Ω' or the physical rotation of the centreless diffractometer around Φ' axis.

$$\varphi_i = \alpha_i + \beta_j \quad (9)$$

$$\varphi_i = (180 - \alpha_i) + \beta_j \quad (10)$$

$$\varphi_i = (180 + \alpha_i) + \beta_j \quad (11)$$

$$\varphi_i = (360 - \alpha_i) + \beta_j \quad (12)$$

After absorption (defocusing) correction, the measured intensity values are available as a function of χ and φ , from which the pole figure can be constructed.

3. Results and validation of the new method

Fig. 8 shows the {222} pole figures of a 3003 type aluminium sheet cold rolled in one step from 4.8 mm to 0.2 mm thickness (average strain: 3.7) measured with a conventional, texture measurement dedicated diffractometer in X mode and a centreless diffractometer in modified X mode, using the described method. Pole figures were measured up to 75° tilting with the conventional method, while up to 60° with the centreless diffractometer. As seen in the figure, the shape of the obtained pole figures is identical, both represent the typical {hkl} pole figures of cold rolled FCC metals. In modified X mode, the middle section of the pole figure was not measurable due to geometrical limits. This section can be measured in Ω mode, though, if needed. However, since the present manuscript focuses solely on modified X mode (and the middle section does not have relevant information of rolled sheets),

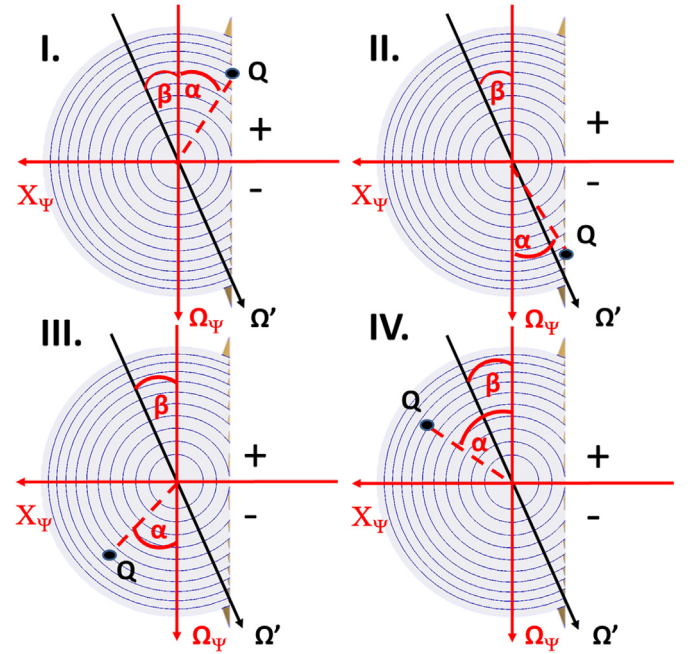


Fig. 7. Normal projections of the intersections (Q) of measurement directions of X' mode and the series of X mode pole figure measurement directions. Intersection is a) in the I., b) in the II., c) in the III., d) in the IV. quarter.

the middle section was left unmeasured.

Fig. 9 shows further instances of pole figures obtained with the conventional and the presented method. Fig. 9 a, b and c correspond to the sheet cold rolled from 4.8 mm to 1 mm thickness in one step (average strain: 1.8), while Fig. 9 d, e and f to the sheet subjected to cold rolling from 4.8 mm to 0.5 mm then to 0.2 mm thickness (total average strain: 1.8). The texture numbers (T_{MAX}) are also indicated. {111} and {222} pole figures represent the shape typical to cold rolling. The difference in degree of deformation of the two sheets is clearly visible if one compares the {111}, {222} or {311} pole figure pairs. The differences in texture are evident in texture numbers as well.

4. Discussion

Comparing the pole figures obtained with the presented method to those of the conventional method it shows that the difference in their appearance solely originates from the different representation mode of measured data. The relevant information of the pole figures of the two methods is the same. Consequently, pole figures of cold rolled aluminium sheets have been successfully measured using the presented method. The measurement method is the same for other materials as well, even if they have multiple phases. The only variable is the Bragg-angle - determined at the beginning - which determines the required tilting angles. Thus, it can be safely stated, that the proposed method is applicable to measure pole figures in a reliable manner on various crystalline materials with centreless diffractometers with all their benefits inherited. First, there is no need for sample cutting, which is a clear advantage over the conventional methods, since those are limited in sample size [10,12,13,15]. Second, the proposed method is not limited to flat surfaces [16]. Third, using portable centreless diffractometers, pole figure measurements can be realized as on-site measurements [18]. These advantages can be strongly utilized in the field of archaeometry, where high value artefacts are the subject of interest and sampling is strictly forbidden. Also, most artefacts have rather complex shape with embossments and other decorations. Therefore, the ability to measure non-flat surfaces is another evident benefit [19].

Besides its benefits, the proposed method has some disadvantages.

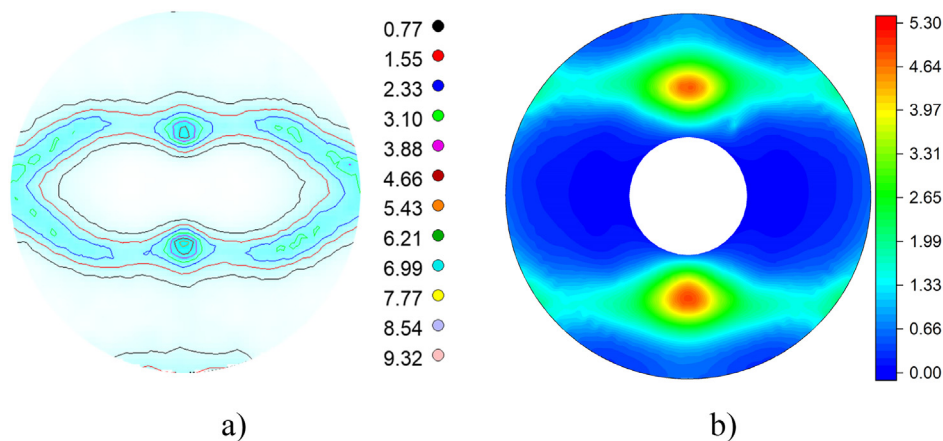


Fig. 8. {222} pole figures and pole density numbers of a cold rolled 3003 type aluminium sheet measured with a) conventional equipment (max $\chi = 75^\circ$), b) centreless diffractometer (max $\chi' = 40^\circ$) in modified X mode.

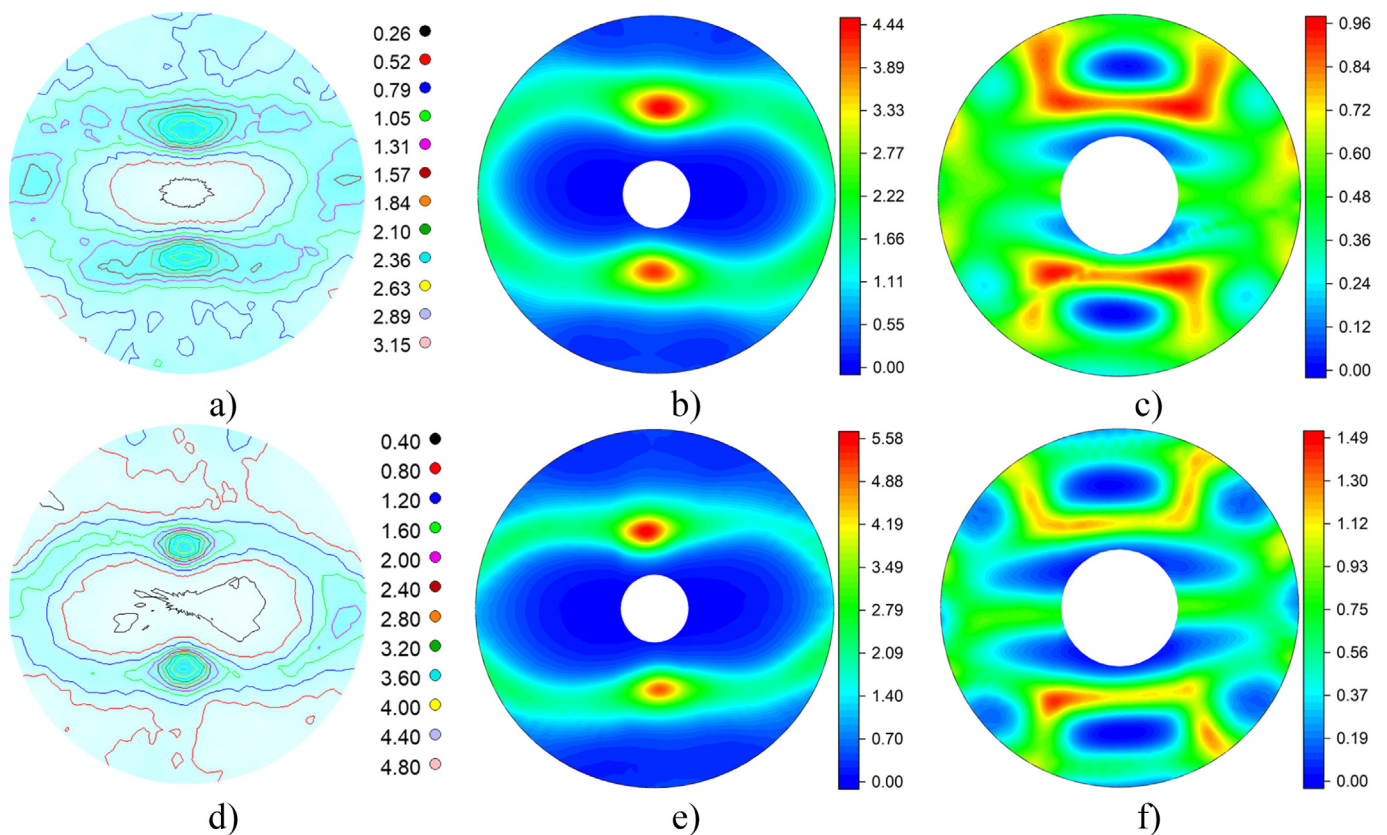


Fig. 9. Pole figures and pole density numbers of cold rolled 3003 type aluminium sheets after a), b), c) 89% deformation; d), e), f) 95% deformation; a), d) {111} pole figures measured with a conventional diffractometer (max $\chi = 75^\circ$); b), e) {222} pole figures; c), f) {311} pole figures measured with a centreless diffractometer (max $\chi' = 60^\circ$) in modified X mode.

Centreless diffractometers have been developed especially for residual stress measurements. It is true, that some of them are capable for retained austenite measurements as well, but it is the residual stress measurement that was in the focus during their design. Thus, currently there is no user-friendly measurement protocol for pole figure measurements [18]. In more details, managing the input data (requested tilting positions) and output data is rather cumbersome. Another drawback is that centreless equipment can handle the high Bragg-angle range ($\sim 130^\circ$ – 160°), since residual stress is usually measured in that region [16–18]. Therefore, the applied X-ray wavelength must be chosen in a way that the {hkl} reflection of interest would lie within that range. However, the authors would like to point out that these weaknesses can

clearly be overcome if the manufacturers put some effort in product developments.

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Data availability statement

The raw/processed data required to reproduce these findings cannot be shared at this time due to technical or time limitations. Data will be made available on request. Corresponding author: femvali@uni-miskolc.hu.

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