Residual stress measurement in textured thin film by grazing-incidence X-ray diffraction

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Abstract

Measurements of residual stresses in textured thin films have always been problematic. In this article, a new experimental method using grazing-incidence X-ray diffraction is presented with its principles based upon the conventional sin$^2$ψ method. Instead of using the Bragg–Brentano (B-B) or Seemann–Bohlin geometry, the proposed method utilizes an asymmetrical diffraction geometry for which the X-ray beam is incident at a grazing angle γ to the sample surface, while the angle ψ is the tilt angle of the sample surface as defined by the conventional sin$^2$ψ method. Basic equations involved in the X-ray residual stress analysis are described, along with exemplified experimental data. Analysis shows that, for an isotropic medium, strain measured using this grazing-incidence geometry assumes a linear relationship with the geometrical parameter cos$^2$α sin$^2$ψ, where the angle α is a constant and is defined as the Bragg angle at ψ=0°, $\theta_o$, minus the grazing incidence angle $\gamma$, i.e. $\alpha = \theta_o - \gamma$. The grazing-incidence diffraction geometry effectively increases the irradiation volume from a thin-film specimen, thereby giving rise to higher intensity for high-angle Bragg peaks than the conventional B-B geometry. The proposed analysis has another advantage, in that the inhomogeneous sample casts little effect on the residual stress results when compared to the traditional sin$^2$ψ method.

Keywords: Textured thin film; Residual stress; cos$^2$α sin$^2$ψ method

1. Introduction

Residual stress plays an important role in the microstructural details and associated properties that thin films exhibit. Two non-destructive techniques have been commonly used for residual stress measurements, one of which measures the bending radius of the substrate and the other utilizes X-ray diffraction (XRD). The former technique measures the curvature of the substrate, before and after coating, and the residual stress is calculated using Stoney’s equation [1]. It requires the use of a relatively large sample, and the localized stress distribution in the plane of the thin film sample cannot be conveniently determined due to poor spatial resolution. For the X-ray diffraction method, spatial resolution below 1 μm is attainable with the use of modern techniques, such as X-ray microprobe utilizing synchrotron X-rays. Nevertheless, difficulties still exist in X-ray residual stress measurements, especially in thin films with strong texture.

The traditional XRD method for residual stress measurement is known as the sin$^2$ψ method, which is based on the measurement of the shift of a diffraction peak position recorded for different ψ angles [2,3]. In this approach, a specific diffraction plane is selected and the interplanar spacing is measured from a coupled $\theta$–$2\theta$ scan [standard Bragg–Brentano (B-B) geometry] of the
specimen at different specimen tilt angle $\psi$—the angle between the diffracting plane normal and the specimen surface normal. Ideally, a high-$\theta$ diffraction peak is chosen to ensure higher sensitivity to strain. Thereafter the residual strain can be derived from the slope of a linear plot between the fractional change of the plane spacing (i.e. strain) and $\sin^2\psi$. In most cases a bi-axial stress model is then used to convert the strain measured to the stress.

Unfortunately, for a highly textured or single crystal-like film under the symmetric B-B diffraction geometry, only a few specific $(hkl)$ peaks show up, while other Bragg peaks can hardly be observed. In addition, the diffraction volume in thin films under the B-B diffraction geometry is proportional to the film thickness, so that, for thin film samples, high-$\theta$ diffraction peaks might be too weak to be conveniently measured even if they are present. The above conditions make the traditional residual stress measurements using symmetric B-B geometry unsuitable for highly textured thin-film specimens.

In an attempt to overcome the above problems, Perry and his co-workers [4] proposed a modified $\sin^2\psi$ method using the Seemann–Bohlin (S-B) focusing geometry [5,6]. In their method, the interplanar spacings of all measurable diffraction peaks with different Miller indices were determined using the grazing-incidence X-ray diffraction geometry. Diffraction peaks of different $(hkl)$ planes were collected in a single $2\theta$ scan with a fixed incident-beam angle to the specimen. Since diffraction planes make different angles to the sample surface normal in Perry’s approach, the sample tilting $\psi$ is not necessary. In fact, Perry et al. pointed out that the angle $\psi$ actually corresponds to the Bragg angle $\theta$ minus the grazing angle $\gamma$ ($\psi = \theta - \gamma$). Therefore, in a single $2\theta$ scan, a range of $\psi$ angles is automatically selected when a number of Bragg peaks with different Miller indices are measured at different $2\theta$ angles. The residual stress was then derived from a plot of the lattice parameters calculated from different peaks vs. $\sin^2\psi$.

However, there are difficulties in the determination of lattice parameters from high-index peaks in a highly textured thin film. This is so because the Bragg peak intensity normally decreases as $2\theta$ increases, which makes peak position determination difficult for a weak and broad peak profile. In Perry’s method, each peak corresponds to a specific sample-tilting angle $\psi$. If high-angle peaks cannot be included, the $\psi$ range used to calculate residual stress would be reduced. This will lead to errors in residual stress measurements. Moreover, the nature of the Perry method utilizes lattice parameters determined from Bragg peaks of different $(hkl)$ planes. Therefore, for any material showing sufficient degree of anisotropy, the assumption of isotropic elastic behavior adopted in Perry’s method could result in appreciable error.

Another approach to X-ray residual stress measurements in epitaxial thin films was suggested by Uchida et al. [7], which follows the $\sin^2\psi$ method as well. This approach basically measures as many accessible Bragg peaks as possible using asymmetrical X-ray diffraction, so that variation in $\psi$ angles is automatically obtained for the same reasons as in the Perry method. A coupled $\theta-2\theta$ scan as in the traditional $\sin^2\psi$ method is used. The drawbacks of this approach include the assumption of isotropic stress, the use of non-constant incident angle so that the irradiation volume within the sample can be different, and the inability to obtain the crystallographic dependence of stress.

Recognizing the advantages as well as the shortcomings of the above three methods, we propose a new XRD approach for the residual stress measurements in textured thin films. Grazing incident X-ray diffraction geometry is used and a fixed $(hkl)$ peak is measured for different $\psi$ tilt in a way similar to, but uniquely different from, the traditional $\sin^2\psi$ method. The use of a specific $(hkl)$ peak removes the concern of elastic anisotropy. The use of asymmetric diffraction geometry in the grazing incidence mode maximizes the diffraction volume and allows more peaks to be measured. It is believed that the method proposed will provide a more convenient and accurate approach to the measurements of residual stresses in textured thin films.

2. Theoretical basis

The underlining principle of the proposed X-ray residual stress measurement method is exactly the same as the traditional $\sin^2\psi$ method. It is based on the measurements of the lattice parameters determined from a fixed $(hkl)$ Bragg reflection but at different tilt angle $\psi$. Unlike the traditional $\sin^2\psi$ method, the method proposed employs an asymmetric B-B XRD geometry, with the incident X-ray beam making a grazing angle $\gamma$ with respect to the specimen surface-tilting axis $S_1$ (Fig. 1) where the angle $\gamma$ is the $\omega$ angle in traditional four-circle diffractometer geometry. The orthogonal coordinate systems used in the following discussion are shown in Fig. 1a. The axes $S_i$ define the co-ordinate system of the thin-film specimen, with $S_1$, $S_2$ contained in the sample surface plane, and $S_3$ being the specimen surface normal. The laboratory system $L_i$ is defined such that $L_1$ is the normal of diffraction planes $(hkl)$ for which spacing is measured by X-rays. $L_1$ and $L_2$ are the two orthogonal axes lying on the $(hkl)$ plane. The $S_1$, $S_2$, and $S_3$ are the new specimen axes when the specimen is rotated about the $S_1$ axis for an angle $\psi$. As observed in Fig. 1b, the $L_2$ axis makes an angle $\psi$ with respect to $S'_2$ after the tilting. (In a four-circle geometry, rotation along the $S_1$ axis is known as the $\chi$ rotation.) At the same time, $L_1$ is kept at a constant angle $\alpha$ with $S_1$, where the angle $\alpha$ is equal to a referenced Bragg angle...
of the observed plane \((hkl)\) at \(\psi = 0^\circ\), \(\theta_0\), minus the grazing angle \(\gamma\), i.e. \(\alpha = \theta_0 - \gamma\), as shown in Fig. 1c. The strain from the residual stress on the plane \((hkl)\) along \(L_3\) can be obtained when the lattice spacing \(d_{\alpha\phi}\) is derived from the position of the diffraction peak for a given reflection \((hkl)\); it is shown as the following formula:

\[
(e'_{33})_{\alpha\phi} = \frac{d_{\alpha\phi} - d_0}{d_0}
\]

(1)

where \(d_0\) is the initial lattice spacing when \(\psi = 0\). The strain \((e'_{33})_{\alpha\phi}\) may be expressed in terms of the strain \(\varepsilon_{ij}\) in the co-ordinate system of the film by a tensor transformation:

\[
(e'_{33})_{\alpha\phi} = a_{3i}a_{3j}\varepsilon_{ij}
\]

(2)

where \(a_{3i}\), \(a_{3j}\) are the direction cosines between \(L_3\) and \(S_i\), \(S_j\), respectively. The direction cosine matrix for this case is:

\[
a_{ik} = \begin{vmatrix}
\cos\alpha & 0 & -\sin\alpha \\
\sin\alpha\sin\psi & \cos\psi & \cos\alpha\sin\psi \\
\sin\alpha\cos\psi & -\sin\psi & \cos\alpha\cos\psi
\end{vmatrix}
\]

(3)

Substituting for \(a_{3i}\), \(a_{3j}\) in Eq. (2) gives:

\[
(e'_{33})_{\alpha\phi} = \frac{d_{\alpha\phi} - d_0}{d_0} = \frac{1 + \nu}{E} \cdot \sigma_{kk} - \frac{1 + \nu}{E} \cdot \frac{\nu}{\sigma_{kk}}
\]

(4)

If the material is isotropic (i.e. the Poisson ratio, \(\nu_{hkl} = \nu\), and Young’s modulus \(E_{hkl} = E\)), the strain becomes:

\[
\varepsilon_{ij} = \frac{1 + \nu}{E} \cdot \frac{\sigma_{ij}}{E} - \frac{1}{2} \cdot \frac{\nu}{E} \cdot \sigma_{kk}
\]

(5)

which, upon substitution into Eq. (4), yields:

\[
\frac{d_{\alpha\phi} - d_0}{d_0} = \frac{1 + \nu}{E} \cdot \frac{\sigma_{ij}}{E} - \frac{1}{2} \cdot \frac{\nu}{E} \cdot \sigma_{kk}
\]

(6)

It is evident from Eq. (6) that, if the stress existing in the films is biaxial, the stress tensor assumes the following form:

\[
\begin{pmatrix}
\sigma_{11} & \sigma_{12} & 0 \\
\sigma_{21} & \sigma_{22} & 0 \\
0 & 0 & 0
\end{pmatrix}
\]

(7)

Then Eq. (6) becomes:

\[
\frac{d_{\alpha\phi} - d_0}{d_0} = \frac{1 + \nu}{E} \cdot \sigma_{11} \sin^2\alpha - \sigma_{12} \cos^2\alpha \sin^2\psi + \frac{1 + \nu}{E} \cdot \frac{\sigma_{11} \sin^2\alpha + \sigma_{22} \cos^2\alpha}{E} - \frac{1 + \nu}{E} \cdot \frac{\sigma_{12} \sin\alpha \sin2\psi}{E}
\]

(8)

For \(\sigma_{11} = \sigma_{22}\) and \(\sigma_{12} = 0\), the stress tensor is as follows:

\[
\begin{pmatrix}
\sigma & 0 & 0 \\
0 & \sigma & 0 \\
0 & 0 & 0
\end{pmatrix}
\]

(9)

Subsequently, Eq. (8) reduces to:
\[
\frac{d_{\alpha\beta} - d_0}{d_0} = \frac{1 + \nu}{E} \sigma \cos^2 \alpha \sin^2 \psi + \frac{1 + \nu}{E} \sigma \sin^2 \alpha - \frac{2 \nu}{E} \sigma
\]

The \((d_{\alpha\beta} - d_0)/d_0\) vs. \(\cos^2 \alpha \sin^2 \psi\) plot will be linear according to Eq. (10). The residual stress can be obtained by determining the slope of the linear fitting between the fractional change in lattice spacing and the term \(\cos^2 \alpha \sin^2 \psi\). Therefore, the slope is equal to 
\[
\frac{1 + \nu}{E} \sigma, \text{ and the intercept, when } \cos^2 \alpha \sin^2 \psi = 0, \text{ is equal to } \frac{1 + \nu}{E} \sigma \sin^2 \alpha - \frac{2 \nu}{E} \sigma.
\]

The resulting Eq. (10) has a linear relationship with respect to \(\cos^2 \alpha \sin^2 \psi\) similar to the linear relationship with \(\sin^2 \psi\) obtained in the traditional \(\sin^2 \psi\) method. The difference lies in the geometrical correction factor, \(\cos^2 \alpha\) that sets the proposed new method, utilizing a grazing incidence angle, apart from the traditional B-B diffraction geometry.

The experimental errors associated with the X-ray measurement of residual stress are the same as the traditional \(\sin^2 \psi\) method. Readers can refer to Chapter 6 of the book by Noyan and Cohen [3] for a detailed discussion.

3. Experimental method

The method proposed has been applied to ZrN and TiN films with two different thickness values. The details of the film preparation utilizing an ion plating method have been described elsewhere [8]. The pole figure results show the specimens have (111) fiber texture. The ZrN sample has a film thickness of 0.62 \(\mu\)m deposited on the Si(100) substrate. This film showed sufficiently strong high-2\(\theta\) peaks using the standard B-B geometry, so that a traditional \(\sin^2 \psi\) method can be applied (coupled 0–2\(\theta\) scan). It serves as a reference measurement with which the residual stress measurement resulting from the new method can be compared. Another film of 0.22-\(\mu\)m-thick TiN was deposited on a stainless steel substrate. This sample has very weak high-2\(\theta\) diffraction peaks that overlap with the substrate noise if using the standard B-B geometry. It is shown that in this case the new method has much greater advantage for residual stress measurements.

All XRD scans were carried out using the Philips X’Pert system equipped with a Cu target. The operation voltage and current were 45 kV and 40 mA, respectively. The size of the beam-defining collimating slit was 2 \(\text{mm}\times2 \text{mm}\). The samples were mounted on a four-circle goniometer, where the film tilted angle \(\psi\) ranges from \(0^\circ\) to \(\pm50^\circ\). A low grazing angle of X-ray incidence was set at \(\gamma\), and the detector scanned in the 2\(\theta\) ranges from \(50^\circ\) to \(63^\circ\) and from \(56^\circ\) to \(66^\circ\) to measure the ZrN(220) and TiN(220) peaks (2\(\theta\) = 56.833\(^\circ\) for ZrN from JCPDS#35-753, and 61.812\(^\circ\) for TiN from JCPDS#38-1420), respectively. Naturally, higher 2\(\theta\) peaks are preferred for higher strain sensitivity. However, when high-2\(\theta\) peaks are not accessible or are too weak to result in accurate measurements, our method can also be used for intermediate 2\(\theta\) peaks. For example, the (220) plane was mostly used due to its relatively strong intensity, plus the fact that higher 2\(\theta\) peaks have much lower intensities and irregular shapes.

A standard B-B geometry was also used with a 20-detector that scanned in the range from \(50^\circ\) to \(63^\circ\) for comparison. The center of each Bragg peak was identified using the Gaussian curve-fitting method.

4. Results and discussion

The lattice spacing vs. \(\cos^2 \alpha \sin^2 \psi\) plot for the 0.62-\(\mu\)m-thick ZrN film with a grazing incidence angle of 2\(^\circ\) is shown in Fig. 2. The slope of the linear fitting curve is \(-0.0096\) with a correlation (R) factor equal to \(-0.95\). The \(\sin^2 \psi\) plot taken from the same sample but using the traditional B-B XRD method is shown in Fig. 3. The slope of the linear fitting curve is \(-0.0103\) with an R-value of \(-0.95\) when the first data point at \(\psi = 0^\circ\) is ignored in the fitting process. Realistically speaking, a linear fitting to the data as shown in Fig. 3 is not appropriate without further analysis [3]. As pointed out by Noyan and Cohen [3], it is not unexpected to see an oscillatory \(\sin^2 \psi\) plot for an inhomogeneous sample. The strain field near the film surface could be different from that near the film substrate interface. In comparison, data obtained using the grazing method show little or no such effect. This could be because the new method keeps a very shallow and near-constant observation depth [9] for each given \(\psi\) angle, so that the inhomogeneity of the sample does not affect the measurement.
in the grazing angle geometry. The oscillatory pattern has been observed elsewhere [10]. It complicates data analysis, which again shows the disadvantage of the traditional \( \sin^2 \psi \) method. Nevertheless, residual stresses derived from the \( \cos^2 \alpha \sin^2 \psi \) plot and the \( \sin^2 \psi \) plot (linear fitting with \( \nu = 0.186 \), and \( E = 460 \) GPa [11,12]) is \( -3.73 \) and \( -4.01 \) GPa, respectively. The difference is 7.5%.

The \( \cos^2 \alpha \sin^2 \psi \) plot for the 0.22-\( \mu \)m-thick TiN film on stainless steel substrate with a grazing angle of 0.5° is shown in Fig. 4. The slope of the linear fit is \( -0.0211 \) with an \( R \)-value of \( -0.98 \). The residual stress calculated is \( -8.18 \) GPa (with \( \nu = 0.2 \) [13] and \( E = 447 \) GPa [14]).

A \( \phi \) study, which measured the residual stress distribution in the sample along different \( \phi \), was also carried out. The sample was rotated along the \( S_3 \) axis with an angle \( \phi \) and the residual stress was measured using the new method. The \( \phi \) range was chosen between 0° and 45° with a step size of 15°. The residual stress measured \( (\sigma_{\phi}) \) includes three terms under the biaxial assumption, where \( \sigma_{\phi} = \sigma_{11} \cos^2 \phi + \sigma_{12} \sin 2 \phi + \sigma_{22} \sin^2 \phi \) [3]. However, \( \sigma_{\phi} \) is equal to \( \sigma \) when \( \sigma_{11} = \sigma_{22} = \sigma \) and \( \sigma_{12} = 0 \).

The difference in residual stresses measured from different \( \phi \) was within 4.5%, and all the \( R \)-values of the curve-fitting of the \( \cos^2 \alpha \sin^2 \psi \) plot were near \( -0.98 \pm 0.1 \). These results indicate the uniformity of the residual stress of the sample along various \( \phi \) directions.

When the strain is smaller than 0.1%, we can simply use the initial point \( d \) (\( \psi = 0^\circ \)) as \( d_0 \) [15]. However, we suggest the use of the intercept of the linear fitting of \( d \) vs. \( \cos^2 \alpha \sin^2 \psi \) plot when \( \psi = 0^\circ \). This is because the initial point might have more error than other points and \( d_0 \) is a multiplier in the slope, so the error will propagate directly into the calculation.

Two factors should be considered in the selection of \( \alpha \), namely, grazing incidence angle \( \gamma \) and diffraction angle \( \theta_o \) at \( \psi = 0^\circ \). The selection of grazing angle is mostly dependent on the film thickness; the grazing angle is decreased to increase the diffraction volume as the film thickness decreases. The diffraction angle \( \theta \) corresponds to the diffraction plane. Theoretically, high-2θ peaks are better for measurement of the lattice parameter; however, the peak intensity may be too low, or too broad or irregular in shape to enable accurate stress analysis. Besides, a higher 2θ would narrow the \( \cos^2 \alpha \sin^2 \psi \) range due to the decrease in \( \cos^2 \alpha \) with increasing \( \theta \), thereby decreasing the precision of the residual stress measurement. Therefore, intermediate 2θ angles ranging from 60° to 80° would be ideal for this new approach. For (220) peaks adopted in the above examples and a grazing angle of 2°, the uncertainty of \( \theta_o \) angle within a range of \( \pm 0.6^\circ \) would result in an error of less than 0.006% in the residual stress calculated.

5. Conclusions

As elaborated in the paper, the new XRD method using a grazing-incidence geometry greatly simplifies non-destructive X-ray residual stress measurements. Analysis has been presented to show that the strain from the residual stress yields a similar linear relationship with \( \sin^2 \psi \) to the traditional XRD method, except for a correcting factor \( \cos^2 \alpha \), in which \( \alpha = \theta_o - \gamma \), where \( \gamma \) is the grazing incidence angle and \( \theta_o \) is the Bragg angle of the observed plane at \( \psi = 0^\circ \). With a grazing incident diffraction mode, the sample volume irradiated by X-rays is greater than traditional B-B diffraction, thereby yielding higher peak intensity, which facilitates residual stress measurements in thin films with strong texture. Second, it was demonstrated by examples that the inhomogeneous residual stress casts little effect on the proposed method of analysis, which is another advantage over the traditional \( \sin^2 \psi \) method. Finally, the approach proposed can be applied to bulk samples as well. By
varying the grazing incidence angle $\gamma$, it is possible to measure the depth dependence of residual strain.

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