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Application of the whole powder pattern decomposition procedure in the residual stress analysis of layers and coatings

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ABSTRACT

The X-ray investigation of stress states in materials, based on the determination of elastic lattice strains which are converted to stresses by means of theory of elasticity, is a necessity in quality control of thin layers and coatings for optimizing manufacturing steps and process parameters.
This work introduces the evaluation of residual stress from complex and overlapping diffraction patterns using a whole-powder pattern decomposition procedure defining a 2θ-offset caused by residual stresses. Furthermore corrections for sample displacement and refraction are directly implemented in the calculation procedure. The correlation matrices of the least square fitting routines have been analyzed for parameter interactions and obvious interdependencies have been decoupled by the introduction of an internal standard within the diffraction experiment. This decomposition based evaluation has been developed on tungsten as a model material system and its efficiency was demonstrated by X-ray diffraction analysis of a solid oxide fuel cell multilayer system. The results are compared with those obtained by the classical $\sin^2\Psi$-method.

1. Introduction

With increasing application of thin films and surface treated structures such as high-temperature barrier-layers, wear-resistant and low-friction coatings as well as ion conducting layers in solid oxide fuel cells (SOFC), there is a growing demand for improved methods to determine the residual stresses within these layers.

The reliability of these coatings, when subjected to external loading and/or temperature depends on the stress state and the knowledge of its magnitude. Using this information the optimization of process parameters and manufacturing routes to improve layer-adhesion can be undertaken.

The determination of residual stresses in layers and coatings by the well-established classical $\sin^2\Psi$ method is a complex task and often limited in those cases where stress gradients or texture effects appear, leading to typical non-linear ε-$\sin^2\Psi$ functions.

An alternative to the classical technique is the application of a multiple (hkl)-method based on datasets obtained at constant incident angles (α) of the primary X-ray beam and the analysis of the diffraction pattern by a whole powder pattern decomposition procedure (wppd). The decomposition of diffraction patterns with the main focus on determination of residual stresses and separation of peak-shifts caused by sample displacement/height errors and refraction, is the main goal of this work.

The applicability of the decomposition based evaluation has been developed and tested on a tungsten layer, which has the advantage of being elastically isotropic, making all corrections concerning the orientation anisotropy of the X-ray elastic constants zero (Zener's ratio $A = 1$).

Furthermore it is easy to induce significant residual stresses (in the GPa range) in tungsten coatings and layers by control of the deposition parameters. This ensures that the measured peak shifts caused by residual stresses are much larger than the detection limits.

2. Experimental details

2.1. X-ray diffraction

All X-ray diffraction investigations have been carried out on a Bruker D4 Endeavor diffractometer, equipped with a Goebel mirror, θ–2θ goniometer, Sol-X energy dispersive detector and autosampler using Cu radiation ($\lambda_{K\alpha} = 1.5406$ Å, X-ray tube Siemens KFL Cu 2K, long fine focus).

The diffractometer has been aligned with high accuracy to determine the lattice constant of LaB_6 (NIST SRM 660a, line position and line profile standard) with a relative precision of ± 0.0001.

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The determination of the residual stress was realized using two different approaches [1]:

1. In accordance with the classical method [2,3], the determination was performed in so-called ω-tilt (iso-inclination) [1] on a single reflection with high multiplicity in 2θ range 120° to 150° (step size 0.1°, time/step 10 s). These measurements were carried out in three different Φ-directions (0°, 45°, 90°) and at 17 Ψ-inclinations (positive and negative tilts). The ω-angle describes the angle between the incident X-ray beam and the sample surface.
2. Alternatively the diffraction experiments were carried out in asymmetric diffraction geometry (fixed incident beam- to sample angle, step size 0.02°, time/step 1 s) at several incidence angles (α) in two Φ-directions (0°, 90°) and are analyzed by a whole powder pattern decomposition procedure [4]. The modification of the incidence angle allows the determination of the stress depth-profile within the coating due to an alteration of the penetration depth of the X-ray beam as function of α. Since a sample displacement also leads to a significant 2θ-offset at decreased incidence angles, this effect must also be considered in the calculation procedure.

When using the classical $\sin^2\Psi$ method all diffraction peaks were smoothed and corrected for background and CuKα2. The correction of the sample displacement (height error) was based on a reference measurement of a tungsten powder (6 μm grain size), which can be assumed to be stress-free. The sliding gravity method was used for calculating the peak positions (the evaluation procedure is implemented in software Diffracplus Leptos S, version 7.03, Bruker AXS).

The data analysis of the measurements obtained in asymmetric diffraction geometry was performed with the software Diffracplus TOPAS version 4.2 (Bruker AXS), used for profile analysis, whole powder-pattern-modeling and decomposition, Rietveld analysis and ab-initio structure solution. The mathematics of these methods and the procedure to extract instrumental- and sample-broadening have been described elsewhere [5–7].

The evaluation of residual stresses from the whole diffraction pattern is based on the multiple (hkl)-method [8]. By the introduction of a user defined equation and the definition of a 2θ-offset, expressed by the general stress equation (Eq. (1)) [9,10], the peak-shifts due to macroscopic stresses are taken into account.

$$\begin{aligned}\Delta 2\theta = -2\tan\theta\{&\frac{1}{2}S_2\left[\sigma_{11}\cos^2\Phi + \sigma_{12}\sin(2\Phi) + \sigma_{22}\sin^2\Phi\right]\sin^2\Psi \\ &+\frac{1}{2}S_2[\sigma_{13}\cos\Phi\sin(2\Psi) + \sigma_{23}\sin\Phi\sin(2\Psi) + \sigma_{33}\cos^2\Psi] \\ &+S_1(\sigma_{11}+\sigma_{22}+\sigma_{33})\}\end{aligned} \quad (1)$$

Table 1
Comparison of the normalized correlation matrices (in percent) of TOPAS (limited to the relevant interacting parameters) for a simultaneous refinement in two Φ-directions on residual stress and displacement (argon sputtered tungsten coating, $\alpha = 25°$), (a) internal standard added, (b) without internal standard.

a

		1	2	3	4	5
σ_{11}	**1**	100	0	5	54	73
σ_{22}	**2**	0	100	5	72	53
Displacement z	**3**	5	5	100	−1	−1
Lattice constant a in $\Phi = 0°$	**4**	54	72	−1	100	78
Lattice constant a in $\Phi = 90°$	**5**	73	53	−1	78	100

b

		1	2	3	4	5
σ_{11}	***1***	100	86	93	−24	−17
σ_{22}	***2***	86	100	93	−15	−23
Displacement z	***3***	93	93	100	−44	−44
Lattice constant a in $\Phi = 0°$	***4***	−24	−15	−44	100	80
Lattice constant a in $\Phi = 90°$	***5***	−17	−23	−44	80	100

Table 2
Summary of the 2θ-offsets caused by refraction, displacement ($z = +0.003$ mm).and residual stress ($\sigma_{11} = \sigma_{22} = -2000$ MPa) for the main reflections of tungsten in asymmetric diffraction geometry at an incidence angle $\alpha = 2°$.

hkl	2Θ	Refraction $\Delta 2\Theta$	Displacement $\Delta 2\Theta$	Residual stress $\Delta 2\Theta$
{1 1 0}	40.265°	0.0811°	−0.0158°	−0.0893°
{2 0 0}	58.276°	0.0805°	−0.0209°	−0.0915°
{2 1 1}	73.197°	0.0806°	−0.0235°	−0.0611°
{2 2 0}	87.024°	0.0810°	−0.0245°	0.0012°
{3 1 0}	100.651°	0.0817°	−0.0241°	0.1039°
{2 2 2}	114.928°	0.0829°	−0.0222°	0.2704°
{3 2 1}	131.184°	0.0853°	−0.0185°	0.5750°

$$\Psi = (\theta - \alpha) \quad \text{for asymmetric diffraction} \quad (2)$$

with

σ_{ii}, σ_{ij}	normal and shear stresses in MPa
Φ	azimuthal angle (angle between the projected diffracting planes normal to the sample's surface with respect to the x-axis of the sample) (°)
Ψ	tilt angle between the normal of the diffracting plane and the normal of the sample's surface (°)
θ	Bragg angle (°)
α	incidence angle of the primary X-ray beam (°)
$S_1, \frac{1}{2}S_2$	diffraction elastic constants in MPa^{-1}.

The emission profile of the X-ray source has been described by the CuKα5 Berger profile [11].

Aberration broadening of the parallel beam optics for the applied diffraction geometry (fixed α) has been determined on LaB_6 (NIST SRM 660a) and expressed by a measured instrument function in an empirical way.

Due to the fact, that in grazing incidence diffraction sample displacement and positioning errors cause increasing peak shifts at decreased incidence angles (α), all calculations were corrected by introducing a second 2θ-offset, expressed by the following Eq. (3), where θ = diffraction angle (°), α = incidence angle (°), R = goniometer-radius (mm) and z = displacement (mm) [12]:

$$\Delta 2\theta = -z\sin(2\theta)/(R\sin\alpha). \quad (3)$$

In general there exist two basic strategies to implement the sample displacement/positioning error in the calculation:

1. The determination of the instrumental positioning error by an additional reference measurement on LaB_6 (NIST SRM 660a, line position and line profile standard) and a refinement of the displacement

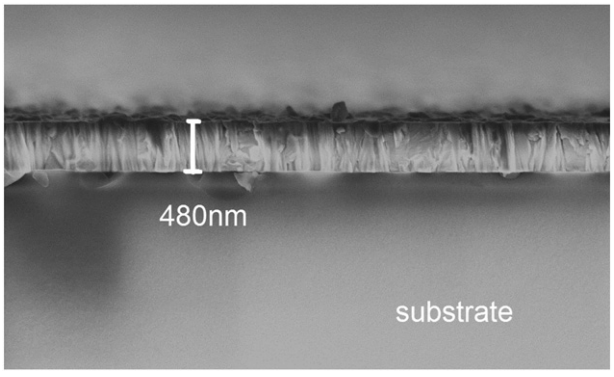

Fig. 1. Fracture cross section of krypton sputtered tungsten coating on display glass Corning Eagle XG (SEM picture Plansee).