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Residual stress depth profiling in complex hard coating systems by X-ray diffraction

M. Klaus [a], Ch. Genzel [b,*], H. Holzschuh [c]

[a] Technische Universität Berlin, Institut für Werkstoffwissenschaften und-technologien, Sekr. BH 18, Ernst-Reuter-Platz 1, 10587 Berlin, Germany
[b] Hahn-Meitner-Institut Berlin, Bereich Strukturforschung, Albert-Einstein-Straße 15, 12489 Berlin, Germany
[c] Walter AG, Derendinger Straße 53, 72072 Tübingen, Germany

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Abstract

X-ray residual stress analysis on multilayered coating systems is a quite difficult and demanding procedure. To obtain information on both, the individual sublayers the coating consists of and the interfacial substrate region, it is necessary to apply different methods which are complementary with respect to the accessible information depth. Based on the concept of an 'equivalent thickness' for describing angle-dispersive diffraction in multilayer structures, a method is proposed that allows for the evaluation of steep intra — as well as interlayer stress gradients within the upper sublayers of multilayer coating systems. Furthermore, energy-dispersive diffraction is shown suitable to detect the residual stress distribution in the near interface substrate zone beneath the coatings. The applicability of the approaches introduced here is demonstrated by the example of cemented carbide WC/Co cutting tools being coated by chemical vapor deposition with sequences of Al_2O_3/TiCN sublayers.

Keywords: X-ray diffraction; Residual stress analysis; Stress gradients; Multilayer systems

1. Introduction

Cutting tools and other highly stressed technical components are coated by chemical and physical vapor deposition methods (CVD/PVD) to protect them from wear and to increase lifetime. To combine different advantageous properties, the coatings are usually not uniform but consist of stacks of alternating sublayers with different thickness, chemical structure and crystallographic texture. By means of 'residual stress engineering' beneficial compressive stresses can be generated within the coating systems either during the deposition process itself or by subsequent mechanical surface treatment like grit blasting. These stresses usually occur in form of steep intra- or long-range inter-layer gradients, the former being balanced within the topmost sublayers and the latter between the sublayer stack and the substrate.

A non-destructive and phase-selective analysis of residual stresses is possible by means of diffraction methods [1]. For X-ray stress analysis (XSA) in thin films, various approaches have been developed which take into account the special problems related to thin film diffraction such as texture, small scattering volume and residual stress gradients [2]. Whereas most of these methods focus on the study of single films, only little has been reported so far on the depth-resolved residual stress analysis in multilayer coatings. In [3] real space strain depth profiling performed in the energy-dispersive (ED) diffraction mode was shown suitable for detecting residual stress differences between the topmost and near interface sublayer of multilayer coatings. However, the procedure turned out to be very time-consuming and does in general not allow for the evaluation of steep stress gradients within the sublayers.

The present work aims at the study of the residual stress depth distribution in individual sublayers as well as in the interfacial substrate zone of multilayered coating systems. Based on the $\sin^2\psi$-measuring technique [4] two advanced methods which are sensitive in different depth zones are introduced and applied for the first time to Al_2O_3/TiCN multilayer hard coatings deposited by CVD on WC/Co substrates. With respect to the upper sublayers which are most affected by the blasting process applied to generate compressive

* Corresponding author.
E-mail address: genzel@hmi.de (C. Genzel).

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stresses, use is made of angle-dispersive (AD) diffraction. The 'equivalent thickness' concept will be shown suitable for analyzing steep intra- as well as interlayer stress gradients occurring within and between different sublayers. To get information on the interfacial substrate zone beneath the multilayer stack, energy-dispersive diffraction within an energy range between about 20 keV and 80 keV is applied. By evaluating the various diffraction lines detected simultaneously in one ED diffractogram, the 'modified multi-wavelength method' [5] allows for the evaluation of the in-plane residual stress depth gradients.

2. Depth-resolved stress analysis on multilayer systems

2.1. Angle-dispersive approach: the 'equivalent thickness' concept

Assuming the residual stress state in thin films to be biaxial and of rotational symmetry, i.e. $\sigma_{11}=\sigma_{22}=\sigma_{//}$, $\sigma_{12}=\sigma_{13}=\sigma_{23}=\sigma_{33}\equiv 0$, the fundamental equation of X-ray stress analysis in its depth dependent forms reads [1,2]

$$\varepsilon_{\psi}^{\mathrm{hkl}}(\tau) = \left(\frac{1}{2}s_2^{\mathrm{hkl}}\sin^2\psi + 2s_1^{\mathrm{hkl}}\right)\sigma_{//}(\tau) \quad (1)$$

It relates the measured lattice strain profile $\varepsilon_{\psi}^{hkl}(\tau)=(d_{\psi}^{hkl}(\tau)-d_0^{hkl})/d_0^{hkl}$ (d_0^{hkl}-strain-free lattice parameter, τ −information depth) to the in-plane stress depth profile $\sigma_{//}(\tau)$. s_1^{hkl} and $\frac{1}{2}s_2^{\mathrm{hkl}}$ are the diffraction elastic constants (DEC) being valid for quasi-isotropic polycrystalline material. For a multilayer system consisting of a sequence of diffracting sublayers D_i of thickness $t_i^D=z_{2i}-z_{2i-1}$, which are separated by solely absorbing sublayers A_i of thickness t_i^A with a structure different from that of D_i, the relation between the total residual stress depth profile $\sigma_{//}(\tau)$ obtained experimentally by means of Eq. (1) and the actual residual stress distribution $\sigma_{//}(z)$ within the individual D_i sublayers is given by

$$\sigma_{//}(\tau)=\sigma_{//}\left(\frac{1}{\mu k}\right) = \frac{e^{-\mu k t_1^{A,\mathrm{eq.}}}\int_{z_1}^{z_2}\sigma_{//}(z)e^{-\mu k z}dz + \mathrm{e}^{-\mu k t_2^{A,\mathrm{eq.}}}\int_{z_3}^{z_4}\sigma_{//}(z)e^{-\mu k z}dz + \ldots}{e^{-\mu k t_1^{A,\mathrm{eq.}}}\int_{z_1}^{z_2}e^{-\mu k z}dz + e^{-\mu k t_2^{A,\mathrm{eq.}}}\int_{z_3}^{z_4}e^{-\mu k z}dz + \ldots} \quad (2)$$

To make sure $\tau=1/\mu k$ (μ − linear absorption coefficient, k − geometry factor) to be the only variable in Eq. (2), the actual thickness t_i^A of the solely absorbing sublayers was replaced by the 'equivalence thickness' $t_i^{A,eq.}$, which is defined by the condition that the respective sublayer has the same attenuation as the sublayer A_i of thickness t_i^A, but with the linear absorption coefficient being valid for the D_i sublayers. By applying appropriate models for the residual stress distribution, Eq. (2) can be used to evaluate the contribution of each sublayer from the total $\sin^2\psi$-strain distribution obtained by Eq. (1). Hence, Eqs. (1) and (2) provide the basis for both, simulating and evaluating AD-XSA experiments on multilayer systems.

2.2. Energy-dispersive approach: the modified multi-wavelength method

Due to the absorption by the coating above, residual stress analysis in the buried interfacial substrate zone of multilayer systems requires the application of ED diffraction with high energy photons. ED-diffraction is based on the measurement of complete diffraction spectra under fixed geometrical conditions. The position of the individual diffraction lines E^{hkl} is related to the lattice spacings by

$$E^{\mathrm{hkl}}[\mathrm{keV}] = \frac{0.6199}{\sin\theta}\frac{1}{d^{\mathrm{hkl}}[\mathrm{nm}]}. \quad (3)$$

The diffraction angle θ in Eq. (3) is an arbitrary parameter that can be chosen freely to adjust the optimum conditions for the maximum information depth τ^{hkl} related to an individual reflection E^{hkl} [5].

Depth-resolved information on the residual stress state by means of ED diffraction can be obtained with the 'modified multi-wavelength method'[5]. The procedure is based on the idea that the in-plane residual stresses $\sigma_{//}^{hkl}$ to be calculated for each reflection E^{hkl} by linear regression from the $d_{\psi}^{hkl}-\sin^2\psi$-distributions can be plotted versus an average information depth, which is defined for the symmetrical Ψ-mode of the XSA [1] by

$$\langle\tau^{\mathrm{hkl}}\rangle = \left\langle\frac{\sin\theta}{2\mu(E^{\mathrm{hkl}})}\cos\psi\right\rangle = \frac{1}{2}\left(\tau_{\psi_{\mathrm{min}}}^{\mathrm{hkl}} + \tau_{\psi_{\mathrm{max}}}^{\mathrm{hkl}}\right). \quad (4)$$

In Eq. (4) $\tau_{\psi\mathrm{min}}^{hkl}$ and $\tau_{\psi\mathrm{max}}^{hkl}$ are the penetration depths for the minimum and maximum inclination angle ψ, respectively, and $\mu(E^{hkl})$ is the energy-dependent absorption coefficient.

3. Experimental

3.1. X-ray diffraction

The AD diffraction experiments for coating stress analysis were carried out on the 5-circle diffractometer ETA built by GE Inspection Technologies, which is equipped with a special thin film attachment for grazing incidence diffraction (polycapillary semi-lens in the primary beam, soller slit+001 LiF monochromator in the diffracted beam) [6]. CoKα- and CuKα-radiation were used to record $\sin^2\psi$-diagrams in steps of $\Delta\sin^2\psi=0.05$ up to $\psi=89.5°$ for the 116- and the 024-Al_2O_3 and the 220-TiCN reflections, respectively. The DEC s_1^{hkl} and $\frac{1}{2}s_2^{\mathrm{hkl}}$ were calculated by means of the Eshelby/Kröner model [7] using the single crystal elastic constants for Al_2O_3 and TiN, respectively, which were taken from [8].

The ED experiments to evaluate the substrate stresses were carried out at the materials science synchrotron-beamline EDDI [9] at BESSY. The primary beam cross-section was defined by 0.5×0.5 mm^2, the angular divergence in the diffracted beam was restricted by a double slit system with apertures of 0.03×5 mm^2 to $\Delta\theta\leq0.005°$. To achieve an optimum situation with respect to the information depth within the WC-substrate, a