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Residual stress gradients in α-Al_2O_3 hard coatings determined by pencil-beam X-ray nanodiffraction: The influence of blasting media

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ABSTRACT

Post-deposition blasting treatments are widely used to introduce compressive residual stress into CVD hard coatings, which are typically in a tensile stress state after deposition on cemented carbide substrates. Within this work, α-Al_2O_3 coatings grown by CVD on TiCN base-layers were dry-blasted using a globular as well as an edged blasting medium and subsequently annealed at 900 °C. The as-deposited, blasted and annealed samples were characterized using cross-sectional synchrotron X-ray nanodiffraction using a pencil X-ray beam of 10 μm × 100 nm in size as well as complementary synchrotron energy dispersive and laboratory monochromatic X-ray diffraction. The results document that the maximum compressive stress of 4 GPa in the samples blasted with the edged medium is significantly higher compared to the samples blasted with the globular medium, which showed maximum compressive stress of 2 GPa. The stress gradient obtained after blasting with the edged medium is steeper, while the zone with compressive stress reaches deeper into the coating for the samples blasted with the globular medium. In the substrate, significantly increased compressive stress of 400 ± 60 MPa compared to 90 ± 30 MPa in the as-deposited state was observed only after blasting with the globular medium and relaxed fully after annealing. In addition, the observed stress gradients were corroborated by the particle impact using a finite element contact mechanics approach.

1. Introduction

Al_2O_3 hard coatings deposited by chemical vapor deposition (CVD) on cemented carbide are widely used in cutting applications due to their high wear resistance and beneficial high temperature properties [1–3]. However, one major drawback of CVD is the high deposition temperature around 1000 °C, which result in the evolution of tensile residual stress due to the mismatch of the thermal expansion coefficients of the coating and the substrate material [4–6]. Consequently, crack networks are formed during cooling after deposition which negatively affect the tool's lifetime as they decrease the mechanical stability of the coating. These crack networks also provide diffusion paths and consequently foster diffusion wear and oxidation [7,8]. In the last years, special attention has been paid to overcome the negative effects of these tensile residual stresses. By applying post-deposition treatments like wet- or dry-blasting, compressive residual stress can be introduced into the coating [9,10], which positively affects its mechanical properties [11–13]. In addition, it has been reported that carefully chosen blasting parameters can result in a change of the stress state of the substrate material as well [9]. That might further increase the tool's lifetime if the formation of comb-cracks, which is reported to correlate with the formation of tensile residual stress in the substrate [14], can be delayed.

In a previous work, Schalk et al. have shown that the magnitude of compressive stresses introduced by blasting can be significantly influenced by the chosen blasting medium and blasting pressure. Full relaxation of the introduced stress was observed after annealing at 900 °C [15]. Bartosik et al. demonstrated that annealing of blasted TiN hard coatings results in stress relaxation already at 200 °C and higher annealing temperatures result in the buildup of even higher tensile stresses after cooling down [16]. However, the stress characterizations within those works were performed using the conventional $\sin^2\psi$ method; thus, no depth resolved information about the stress gradients after blasting or annealing was obtained.

The aim of this work is to perform a detailed characterization of residual stress and microstructure gradients in blasted α-Al_2O_3 hard coatings using the recently developed synchrotron cross-sectional

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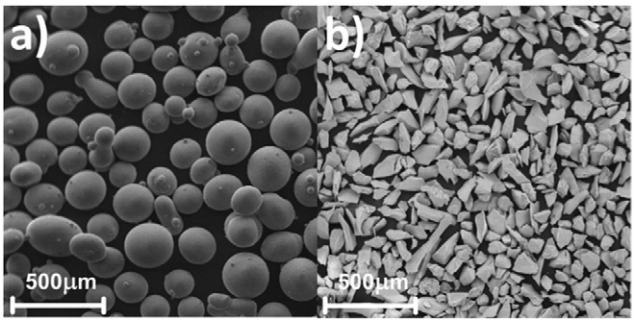

Fig. 1. SEM micrographs of the globular (a) and the edged (b) blasting medium.

X-ray nanodiffraction [17]. Since this approach operates with a point X-ray beam of about 100 nm in diameter [17], it was modified in order to achieve sufficient diffraction statistics for coarse grained materials like CVD α-Al_2O_3 coatings with a grain size in the µm range. For this purpose, an X-ray beam with a pencil-like shape similar to Vaxelaire et al. [18] was applied to as-deposited, blasted and annealed α-Al_2O_3 coatings. Complementary, laboratory monochromatic X-ray diffraction (XRD) and energy dispersive synchrotron polychromatic XRD were used to obtain volume-averaged data from the coatings and substrates. To analyze the effect of the blasting media on the stress formation in the coating surface, finite element (FE) simulations were performed using the software package *Abaqus/Explicite 6.13-3*.

2. Experimental and theoretical methods

The coatings investigated within this work were deposited using a SuCoTec SCT600TH industrial-scale CVD plant. Cemented carbide inserts in SNUN 120412 geometry (according to ISO 1832) with a chemical composition of 77 wt.% tungsten carbide, 12 wt.% mixed carbides and 11 wt.% cobalt were used as substrates. A TiCN base-layer [19] with a thickness of ~9 µm was deposited using $TiCl_4$–CH_3CN–H_2–N_2–CO precursors at a temperature of 900 °C and a pressure of 100 mbar. Afterwards, the ~8 µm thick α-Al_2O_3 layer was grown using $AlCl_3$–CO_2–H_2–H_2S precursors at a temperature of 1000 °C and a pressure of 75 mbar [20]. Subsequently, the samples were dry blasted using (i) a globular medium provided by Kuhmichel Abrasiv shown in Fig. 1a and (ii) an edged medium provided by Treibacher Schleifmittel shown in Fig. 1b. The globular medium has a particle diameter of 125–250 µm, a hardness of ~700 HV and a phase composition of ~62 % ZrO_2, ~28 % SiO_2 and ~5 % Al_2O_3 (remaining fraction ceramic impurities). The mesh grit size of the edged medium is 180/220, the hardness ~2000 HV and the phase composition is ~55 % Al_2O_3 and ~42 % ZrO_2 (remaining fraction ceramic impurities). The chosen blasting pressure was 1.5 bar, applied under a working angle of 15° for a time of 14 s.

The blasted samples were tempered at 900 °C for 15 min in a HTM Reetz vacuum furnace (base pressure < 5×10^{-4} Pa), using a heating rate of 20 K/min and a cooling rate of 60 K/min. The laboratory XRD investigations of the coatings were performed using a Rigaku SmartLab five-axis X-ray diffractometer equipped with Cu-K_α radiation, a parabolic multilayer mirror in the primary beam and a secondary graphite monochromator. The stress characterization was performed using the conventional $\sin^2\psi$ method [21]. All scanning electron microscopy (SEM) investigations were performed using a Zeiss Auriga Crossbeam field emission gun SEM. The electron backscatter diffraction (EBSD) measurement was performed with an EDAX DigiView IV EBSD detector. Prior to the EBSD measurement, the surface of the fracture cross section was polished with an Orsay Physics Cobra Z-05 focused ion beam (FIB) extension.

For the cross-sectional X-ray nanodiffraction experiments, 100–150 µm thick lamellae were cut out from the samples using a Struers Accutom precision saw equipped with a diamond cutting wheel. Subsequently, the lamellae were manually polished down to a thickness of approximately 30 to 50 µm. The synchrotron measurements were performed at the nano-focus extension of the ID13 beamline of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France using a monochromatic X-ray beam with an energy of 14.9 keV. A conventional cross-sectional X-ray nanodiffraction setup operating with a beam diameter of 100 nm did not provide sufficient diffraction statistics as demonstrated by the spotty diffraction pattern from the charge-coupled device (CCD) in Fig. 2a. Therefore, a pencil-shaped X-ray nanobeam with dimensions of 10 µm × 100 nm was implemented for this study using dedicated focusing optics [22]. The diffraction experiment using the pencil beam aligned parallel to the coating-substrate interface resulted in a significant improvement of the diffraction statistics, as demonstrated in Fig. 2b.

The lamellar samples were scanned in transmission geometry along the coating depth in steps of 100 nm and the two dimensional data (2D) from the CCD were evaluated using the software package Fit2D [23]. To achieve even better diffraction statistics, for each coating depth five measurements were performed at different positions and the 2D patterns were summed. The residual X-ray elastic strain at different coating depths was evaluated from the elliptical distortion of the Debye–Scherrer rings of TiCN (111) and α-Al_2O_3 (104) reflections according to Refs. [17,24]. To derive the residual stress from the residual strain, the X-ray elastic constants (XEC) were used which were determined from the single crystal elastic constants [25,26] using the Kröner-model [27].

The residual stress state in the surface region of the cemented carbide substrates was determined at the materials science synchrotron-beamline EDDI (Energy Dispersive Diffraction) of the synchrotron source BESSY in Berlin, Germany using energy dispersive XRD as specified in Refs. [28,29]. Volume-averaged residual stresses were calculated from the measured X-ray elastic strains, determined from the shift of substrate WC (001) reflections by applying XEC from Ref. [30]. The experimental conditions corresponded to an information depth of approximately 500 nm.

The FE simulations were performed using the software package *Abaqus/Explicite 6.13-3*. An axisymmetric model was defined with a spherical body similar to the globular blasting medium and a sample

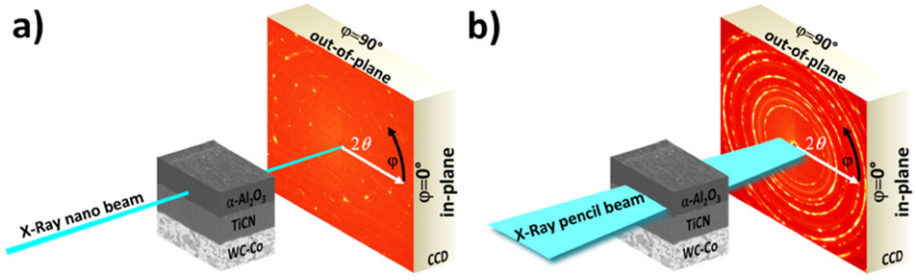

Fig. 2. Schematic description of the cross-sectional X-ray nanodiffraction setup according to Ref. [17] (a) and the setup using a pencil-like X-ray nano-beam (b). The two dimensional images indicate an improved diffraction statistics of the Debye–Scherrer rings in (b).