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# Deformation of Hard Coatings at Elevated Temperatures

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ABSTRACT

The elevated temperature performance of a wide range of chromium nitride-based hard coatings was evaluated using *in situ* micro-compression in the scanning electron microscope. This allows the first direct measurement of the high temperature compressive strength, rather than the hardness, of such coatings. The microstructure of the coatings was analyzed using X-ray diffraction and focused ion beam cross sectioning followed by electron microscopy. Micropillars were examined using electron microscopy before, during and after compression. Trends in deformation behavior and failure stress with temperature are discussed in relation to the coatings' microstructures and their room temperature mechanical properties.



## 1. Introduction

Thin chromium aluminum nitride based hard tool coatings deposited by the cathodic arc PVD method represent the industrial state of the art for use in a wide range of metal machining applications, especially in high-performance, and interrupted cutting operations such as milling or hobbing [1–3]. These cubic ceramics offer a variety of attractive properties, such as a reasonable high temperature stability and a high resistance to abrasive wear and oxidation [4–11]. In the optimization of such coatings and the selection of candidate elements to be added, such as transition metals or silicon, one major goal is to optimize strength and fracture toughness at high temperatures, as those are crucial to improve wear resistance. A well-known fact is that the grain size, crystal textures, and nanolayer or nanocomposite nanostructure of these coatings significantly influence their mechanical properties.

In order to optimize the performance of these hard coating materials used in modern machining processes, it is necessary to investigate the material properties between at high temperatures generated in service, typically several hundreds of degrees above room temperature. However, the small length scale or thickness of the coatings limits the utility of conventional high temperature testing techniques. The recent extension of nanoscale techniques such as nanoindentation and nanoimpact to elevated temperatures has recently made this a possibility, and recent work using these techniques has demonstrated the utility of these techniques [3,12–16]. The development of vacuum techniques for high temperature nanoindentation [17–19] has recently also allowed measurement at temperatures where oxidation of the indenter or sample may be problematic.

The advent of focused ion beam (FIB) machining techniques for micro-scale specimen preparation [20] has enabled several novel test geometries to determine various properties of thin coatings. The most widely used of these geometries is the micropillar [21], which allows uniaxial compressive stress-strain behavior to be determined for micro-scale specimens. However, several other geometries have also been developed for testing tension [22], cantilever bending [23,24], and shear [25]. As toughness is a key parameter for hard coating performance, several studies have investigated micro-scale toughness tests using cantilevers [23,24,26], double cantilevered bridges [27], and double cantilevered beams [28]. However, the design of these test geometries requires prior knowledge of the yield strength to prevent plastic yielding from contributing to the apparent fracture energy, so high temperature compression testing is required before such test geometries can be implemented to determine high temperature fracture toughness of hard coatings.

Micropillar compression at high temperature offers several attractive advantages over hardness testing [21]. Indentation is highly dependent on tip geometry for accurate measurements, and indenter geometry variation due to erosion by oxidation and abrasive wear or plastic deformation of the tip is a serious concern at high temperatures [29]. Using a flat punch indenter avoids any such geometric variation. Also, the micropillar geometry offers a uniaxial stress state in contrast to the triaxial stress state of indentation. This provides a direct measurement of the compressive yield or failure stress, rather than an indirect method such as indentation where the relationship between the yield stress and hardness is complex [30]. Lastly, *in situ* micro-pillar compression allows direct visualization of the deformation mechanism changes at elevated temperature [31], so that buckling, fracture, or plastic deformation can be immediately observed. In this work, the benefits of *in situ* high temperature micro-compression testing are utilized to characterize the

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uniaxial compressive strength of a range of chromium aluminum nitride coatings at near service temperatures.

## 2. Materials and methods

### 2.1. Sample production and characterization

All coating samples were produced at a coating temperature of 480 °C using a $\pi^{411}$ industrial PVD unit (PLATIT AG, Switzerland), using a combination of lateral (LARC®) and central (CERC®) cylindrical rotating arc cathodes, a process pressure of 3.5 to 4 Pa and a bias voltage during the main coating step of −40 V. The targets were selected from highly pure Cr, Ti, AlCr, Al, and AlSi materials (PLANSEE AG, Germany) depending on the coating type. An overall coating thickness of approx. 7 μm was deposited on the substrates, which were either double side polished cemented carbide strips 5 × 40 × 0.5 mm (for micro-compression testing) or single side polished cemented carbide disks Ø 32 × 4 mm (for XRD and nanoindentation), both made from ultrafine grade CTU24L with 12% Co by mass (Ceratizit, Germany). In case of the CrN and AlCrN layers, the thick coatings were applied in two consequent batches with intermediate surface polishing. All samples were coated in triple rotation mode and were mechanically polished before FIB preparation.

Tescan Vela and Lyra dual-beam focused ion beam (FIB) stations were used to mill rectangular trenches using $Ga^+$ at 30 kV and currents of ~6 nA to obtain cross sections of all the coatings. The cross sections were fine milled and polished using lower currents ranging from 1 nA to 300 pA and imaged using a Hitachi S4800 high-resolution scanning electron microscope (HRSEM).

Grazing incidence X-ray diffraction (Bruker AXS, D8 Discover) measurements were made by using Cu$K\alpha$ ($\lambda = 1.5418$ Å) radiation to characterize the coatings. The excitation voltage and current were set at 40 kV and 40 mA, respectively, in the diffractometer. The angle of incidence was kept constant at 2°. The step size and the scan range used were 0.02° and from 20 to 90°, respectively. The grain size of the coatings was estimated from the Scherrer formula [32], as given in Eq. (1),

$$t = \frac{0.9\lambda}{B\cos(\theta)} \tag{1}$$

where $B$ is the full width at half maximum (FWHM) of a Bragg peak, λ is the X-ray wavelength, and $\theta$ is the Bragg angle.

Micropillars were fabricated with equivalent diameters of ~1.5 μm with aspect ratios of ~3 using a Ga ion beam with an accelerating voltage of 30 kV in Tescan Vela and Lyra FIB instruments. Initially, coarse milling using high currents of ~6 nA was used to mill large craters around the micropillars to allow visualization during compression. Fine polishing to the desired micropillar dimensions was achieved using lower currents ranging from 1 nA to 300 pA, which in turn minimizes irradiation damage. The micropillars were imaged after FIB machining and after compression at elevated temperatures using a Hitachi S4800 high-resolution scanning electron microscope (HRSEM) and SEM of the Tescan Lyra FIB.

### 2.2. Micro-mechanical testing

Room temperature nanoindentation testing of the coatings was performed using a CSM Instruments NanoHardness Tester (NHT). This was calibrated using a fused silica reference with a calibrated modulus, $E_{fs}$, of 73 ± 0.2 GPa immediately previous to testing due to the low indentation depths and high hardness and modulus of the coatings to be tested. A minimum of 20 indentations each was performed at maximum loads of 20, 30, 50 and 70 mN at loading rates of 20/30/50/70 mN/min, respectively, and a holding time at peak load of 1 s. The Young's modulus was extracted from the load–displacement curves using the Oliver and Pharr method [33] assuming a Poisson's ratio of $\nu = 0.3$ for the coatings. The results for hardness and modulus were averaged from the four partial results.

Elevated temperature micro-compression was performed *in situ* in a Zeiss DSM 962 SEM using an Alemnis *In-situ* Indenter modified for high temperature operation [19]. This system allowed testing to be performed at precisely measured and calibrated surface temperatures [34] with near-negligible thermal drift. Micropillars were compressed using displacement control at a constant displacement rate appropriate to generate a $1 \times 10^{-3}$ $s^{-1}$ strain rate for the heights of the pillars. At least 4 pillars were compressed for each test condition. The intrinsic displacement control of the system prevented the indenter tip from crushing the pillar samples upon a yield or fracture event, so that the pillars could be examined after failure.

## 3. Results and discussion

### 3.1. Microstructural characterization

In order to provide context to the mechanical testing measurements, the microstructures of the coating systems were characterized using several methods: HRSEM, XRD and FIB cross-sectioning.

X-ray diffraction was used to determine the crystal structure, crystallite size and texture of the coatings. The results from the various coatings, shown in Fig. 1, show broadly similar trends. Peak intensities and locations show slight differences between the different coatings, but the coatings all exhibit the typical peaks of the FCC phase occurring at 37.5°, 43.7°, 63.6° and 76.6°. No traces of soft hexagonal Al(Cr)N phase could be detected. The average crystallite size of the coatings, calculated from the half width of the three most intense peaks using Eq. 1, was determined to be ~16 nm for the CrN coating and ~11 nm for all of the aluminum-containing coatings. All of the coatings are only weakly textured with a slight preference for the (111) orientation.

XRD measurements can provide a general picture of the coating's crystallographic structure, but this yields relatively little data about the coating's morphology in terms of layer thicknesses, inclusions, and grain shape/orientation. Traditionally, this information was acquired using metallographic sectioning and polishing followed by HRSEM observation. Currently, cross-sections provided by FIB trench milling and polishing provide a more rapid alternative, which also reduces the risk of an abrasive alteration of the microstructure, such as removal of inclusions, and edge rounding typical of conventional polishing techniques. These FIB cross-sections are highly instructive for investigating coating microstructures, and they can be performed at specific locations without the need for mounting and polishing. Cross-sections of the various coating systems are shown in Fig. 2.

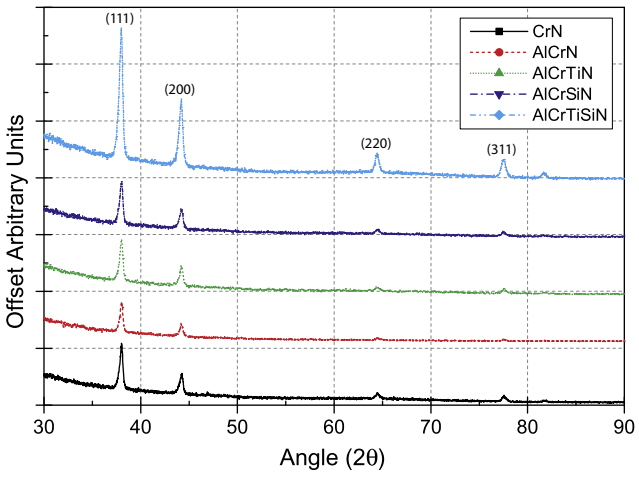


**Fig. 1.** Grazing incidence X-ray diffraction results for the investigated coatings.