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Pressure and temperature effects on the decomposition of arc evaporated $\text{Ti}_{0.6}\text{Al}_{0.4}\text{N}$ coatings in continuous turning

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Abstract

The isostructural decomposition of arc evaporated $\text{Ti}_{0.6}\text{Al}_{0.4}\text{N}$ coatings at the elevated temperatures and high stresses occurring during metal cutting have been studied.

Comparisons are made with short time ($t=10$ min) anneals at temperatures typical for steel turning operations. The evolution of the spinodal wavelengths are studied by analytical transmission electron microscopy from samples originating from the rake face. Temperature and force measurements during turning allowed for separation of the effects of the temperature and stresses on wavelength evolution. The results show a peak temperature of around 900 °C and a peak normal stress of around 2 GPa during cutting. The overall wavelength is longer after cutting compared to the annealed sample at the same temperature. The results suggest that pressures generated during cutting promote coherent isostructural decomposition which is in line with theoretical studies but for considerably higher pressures.

Keywords: Spinodal decomposition, Metal cutting, High stresses

1. Introduction

Industrially important since the 1990s [1,2], $\text{Ti}_{1-x}\text{Al}_x\text{N}$ is a widely used physical vapor deposited hard coating with superior high temperature properties compared to its predecessor TiN. A contributor to this is the observed age hardening during annealing, which is an effect of the decomposition of the as-deposited unstable cubic (*c*)- $\text{Ti}_{1-x}\text{Al}_x\text{N}$ (B1) structure into nanometer sized *c*-TiN rich and *c*-AlN rich coherent domains [3,4]. The lattice mismatch between the coherent domains introduces microstresses at the domain borders which in combination with evolving elastic property differences hinder dislocation motion [5]. As *ab initio* calculations [6] are showing a miscibility gap with a negative second derivative of the Gibbs free energy for the $\text{Ti}_{1-x}\text{Al}_x\text{N}$ solid solution, this step is considered a spinodal decomposition. Upon further annealing, the TiN domains are enriched while *c*-AlN transforms to its stable wurtzite (*w*) phase (B4, *w*-AlN), which detrimentally reduces the mechanical properties of the coating [7].

Recent theoretical studies show that an external pressure will thermodynamically promote the spinodal decomposition [8] and suppress the *c*-AlN to *w*-AlN transformation [8,9]. Reasons for this include the deviation from Vegard's law and the pressure dependent stability of *c*-AlN. Based on this study [8], the effect of pressure is believed to be more distinct at Al compositions around 0.4 due to a shoulder of the spinodal region in the isostructural phase diagram of $\text{Ti}_{1-x}\text{Al}_x\text{N}$. Since not only high temperatures [10,11] but also high stresses prevail at the rake face [12,13] of an insert during cutting, it is suspected that cutting operations are likely to give an even more pronounced spinodal decomposition compared to isothermal anneals.

Although several studies previously reported on the decomposition of post annealed $\text{Ti}_{1-x}\text{Al}_x\text{N}$ [3,7,14], a detailed study of the $\text{Ti}_{1-x}\text{Al}_x\text{N}$ decomposition at the cutting edge is still

lacking. This is the motive of the present study where the microstructure evolution of $\text{Ti}_{0.6}\text{Al}_{0.4}\text{N}$ after cutting was compared to reference heat treatments performed with isothermal steps of 10 min. Microstructure and local chemistry at different positions along the rake face of the cutting insert, i.e., at sample positions exposed to different stresses and temperatures during the cutting process were investigated by analytical transmission electron microscopy (TEM). The evolution of the spinodal decomposition is later discussed in terms of the temperature and stress distributions.

2. Experimental details

A commercial Sulzer Metaplas MZR323 reactive cathodic arc evaporation system was used to deposit the coatings in a 4.5 Pa N_2 atmosphere, substrate temperature $\sim 500^\circ\text{C}$ with a bias of -40 V. Polished WC-Co cutting inserts (ISO geometry TPUN160308) and blanks (ISO SNUN120408) were used as substrates that were ultrasonically cleaned and Ar-ion etched prior to the coating process. The substrates were mounted on a rotating drum (substrate holder) facing the cathodes placed in line and at different heights on the side wall of the deposition chamber. With this configuration, coatings with different chemical compositions can be deposited by combining different cathodes. In this work, we deposited a $\text{Ti}_{0.6}\text{Al}_{0.4}\text{N}$ composition by using a pure Ti cathode in combination with a compound $\text{Ti}_{0.50}\text{Al}_{0.50}$ cathode mounted in two positions at different heights in the chamber. Energy dispersive x-ray spectroscopy (EDS) was employed to verify the coating composition using a Leo 1550 Gemini scanning electron microscope operated at a working distance of 10 mm and using 20 kV.

The machining experiments consisted of dry longitudinal turning in a carbon steel, C45E (yield stress of 280 MPa, rupture stress of 590-740 MPa and hardness of 165-220 HB), with a cutting speed (v_c) of 200 m/min, feed (f) 0.3 mm/rev, depth of cut (a_p) 3 mm and time

in cut of 10 min. Additionally, dry orthogonal cutting tests were performed to measure the cutting forces using a standard three-force component dynamometer and the tool temperature distribution was measured using an IR-CCD camera as described by M'Saoubi *et al* [15]. The temperature measurements were performed using a cutting time of 10-15 s. The information of the cutting forces together with cutting parameters including chip thickness, work material shear strength and contact length are further introduced in an analytical model [16] for calculation of the normal and tangential stress distribution along the rake face.

For comparison, part of the coatings were annealed in vacuum (base pressure $<3 \cdot 10^{-5}$ Torr) with a heating rate of 20 °C/min up to $T_{\max}$ (900 and 1000 °C) where the coatings were held isothermally for 10 min before a cool down with 50 °C/min. Prior to annealing, samples were cut in small pieces, $1.7 \times 0.5 \times 0.5 \text{ mm}^3$ in order to minimize thermal gradients.

Structural characterizations were performed with a Fei Tecnai G² TF 20 UT analytical transmission electron microscope (TEM and STEM) operated at an acceleration voltage of 200 kV and equipped with an energy dispersive x-ray spectrometer (EDS). STEM imaging was performed with a high angle annular detector at a camera length of 170 mm.

TEM sample preparation on the rake face of the coated insert, after machining, was performed using a Carl Zeiss CrossBeam 1540 EsB focused ion beam (FIB) to create cross sectional samples with the lift out technique described in [17]. Cross sectional TEM sample preparation for annealed samples was performed by mechanical grinding and polishing followed by thinning to electron transparency with a Gatan Precision Ion Polishing System.

The spinodal wavelengths have been extracted from STEM micrograph using the line profile tool in the Gatan Digital Micrograph. Several line profiles in the micrographs have

been performed from where an average of the sinusoidal gray scale intensity variations has been calculated.

3. Results and discussion

3.1 Temperature and stress distribution

Figure 1 (a) shows the temperature distribution of a cutting tool insert during turning at $v_c=200$ m/min. The insert is viewed from the side with the chip sliding along the rake face on the left side where the tool-chip contact length is marked with an arrow. Figure 1(b) shows the temperature profile along the contact length, with temperatures between 700 °C and 900 °C. The moderate temperature at the cutting edge increases sharply to a nearly constant value of 850-900 °C across a 0.5 mm wide region. Further away the temperature decreases. The highest temperature is observed at about half the contact length, hereinafter referred to as the hot zone where the maximum temperature, T_{max} , is measured to 890 °C. Both the distribution and the magnitude of the temperature profile are consistent with previous studies [15,18,19].

Figure 1. a) Side view temperature distribution as measured by the IR-CCD camera during cutting with cutting speed of 200 m/min. The arrow indicates the distance from the cutting edge and the position of the temperature extraction shown in b).

Figure 2 shows the normal and tangential stress distribution along the rake face at $v_c=200$ m/min. The normal stress has its maximum of about 2 GPa at the cutting edge and monotonically decreases with increasing distance from the cutting edge. The tangential stress distribution starts with a low value of about 0.4 GPa at the cutting edge and increases up to its maximum value of about 0.8 GPa at half the contact length after which it decreases further away from the cutting edge. Both the magnitude and relative values of the normal and tangential stresses are in reasonable agreement with previous studies which show peak normal stresses of ~ 2 GPa and peak tangential stresses of ~ 0.7 GPa [11]. An error margin is estimated to be 15%, taking into account the standard deviation of the cutting forces and measurement errors. In addition, the inset shows the rake face of a worn cutting insert with an arrow indicating the line along which the relative distance from the cutting edge was measured. The letters A-D describe the relative positions along this line from which TEM samples have been obtained.

Figure 2. Stress distribution for normal and tangential stress during orthogonal cutting at cutting speed 200 m/min. The inset shows a top view SEM micrograph of the rake face with the distance from cutting edge indicated by the arrow. The horizontal arrows indicate to which y-axis the plot belongs, note the different scale on the y-axes. The letters A-D denote the position of the TEM samples in Figure 5.

3.2 Microstructure evolution

Figure 3 (a-c) shows cross sectional overview TEM micrographs of $\text{Ti}_{0.6}\text{Al}_{0.4}\text{N}$ coatings in its as-deposited state, after cutting and after annealing, respectively. For the as-deposited coating, the micrograph reveals a dense columnar structure with a column width around 500 nm and a high defect density typical for arc evaporated $\text{Ti}_{0.6}\text{Al}_{0.4}\text{N}$. The defect density is clearly reduced after cutting and annealing, suggesting that defect annihilation has taken place despite

the relatively short exposure time of 10 min during either the annealing or the cutting experiments.

Figure 3. TEM overview for the a) as-deposited coating, b) after cutting and c) after annealing at 1000 °C.

Figure 4 (a-c) shows cross sectional STEM, Z-contrast, micrographs of the $\text{Ti}_{0.6}\text{Al}_{0.4}\text{N}$ layers in its as-deposited state and after annealing to 900 and 1000 °C. The *c*-TiN-rich areas appear with a brighter contrast and *c*-AlN-rich areas with darker contrast (confirmed by EDS mapping but not shown). In its as-deposited state, Figure 4 (a), the Z-contrast image reveals a small layered modulation in composition. This layering effect stems from the rotation of the inserts during the deposition process as described by Eriksson *et al* [20]. After annealing, Figure 4 (b-c) reveals diffuse nanostructured TiN- and AlN-rich domains where the spinodal wavelength is notably larger after annealing to 1000 °C.

A detailed interpretation of the effect of the above mentioned as-deposited compositional modulation on the decomposition to *c*-TiN and *c*-AlN rich domains is beyond the scope of this paper. However, it can be speculated that the as-deposited compositional gradients are further enhanced by the up-hill diffusion, characteristic for spinodal

decomposition, as the free energy is generally lowered with increased compositional fluctuations [21]. Hence, it is possible that the layering acts as a weak template during the spinodal decomposition, thus to some extent the decomposition pathway is influenced by the layers.

Figure 4. STEM micrographs for coatings a) in as-deposited state, b) after annealing with an isothermal step for 10 min at 900 °C and c) at 1000 °C.

Figure 5 (a-d) shows cross sectional STEM micrographs of the $\text{Ti}_{0.6}\text{Al}_{0.4}\text{N}$ layers after metal cutting. Here, the samples were obtained close to the cutting edge at the positions A-D which indicate its position along the rake face, i.e., along a line where both the normal stress and temperature decreases, c.f. Figure 1 and Figure 2. As it is apparent in the micrographs, the spinodal wavelength is dependent on the position from where the TEM-samples were obtained.

The coherency of the decomposed regions (consisting of *c*-AlN and *c*-TiN rich domains) after cutting at position B is shown in Figure 6. Here, high resolution TEM (HRTEM) micrographs along the $\langle 110 \rangle$ zone axis with corresponding Fast Fourier transforms (FFT) are shown. The micrograph covers approximately $20 \times 20 \text{ nm}^2$, whereas the domain size is roughly

$5 \times 5 \text{ nm}^2$ as seen in Figure 5 (b) and hence several domains are covered. The coherency between the decomposition proves the existence of an isostructural decomposition of *c*-(Ti,Al)N into *c*-TiN and *c*-AlN. This is then the underlying mechanism for the previously mentioned age hardening due to the mismatch in lattice parameters and elastic properties between the domains. Similar coherency is seen for the other samples in the series but not shown.

Figure 5. STEM micrographs for coatings after cutting at a) position A, b) position B, c) position C and d) position D. See Figure 2 for approximate stress distributions at positions A-D.

Figure 6. HRTEM micrographs with corresponding FFT of the coating after cutting at position B.

The measured wavelengths from Figure 4 and Figure 5 as well as the temperature profile from Figure 1 (b) are plotted in Figure 7. The upper part recapitulates the temperature profile and the lower part shows the different wavelengths at positions A-D as evaluated from the STEM micrographs. As evidenced, the average wavelength after cutting decreases along the rake face in the direction away from the cutting edge from around 11 nm at positions A and B to 8.1 nm at position C and finally down to 4.8 nm at position D. The average wavelengths after annealing are also shown in the graph, plotted as horizontal dotted lines.

Here, the wavelength after annealing is increasing from 5.6 nm to 10 nm with an increasing annealing temperature from 900 °C to 1000 °C, respectively.

Figure 7. The upper part shows the temperature profile along the rake face and the lower part the measured spinodal wavelength after cutting (data points) and after annealing (horizontal lines).

The wavelengths in Figure 7 are of the same order as previously seen for isothermally annealed coatings [22,23]. There is a clear increase of the wavelength after annealing at 1000 °C compared to 900 °C, which can be explained by the fact that an increased temperature favors the kinetics during the coarsening process [24]. By comparing the

temperature profile with the measured wavelengths relative to each other, a clear temperature effect is also seen after cutting. Both the plateau around the hot zone and the decrease in wavelength with decreasing temperature can be distinguished. Although the temperature dependence is very pronounced after cutting, there also exists a large difference between the wavelengths during cutting and heat treatments despite a similar temperature.

As the measured temperature during cutting is below 900 °C at all positions, an expected wavelength would have been around 5.5 nm at the plateau instead of the measured wavelength above 10 nm. Hence, the progression of the decomposition process is driven farther after cutting at around 900 °C compared to annealing at the same temperature. One explanation for this can be the difficulty to measure correct temperatures or the difference between longitudinal and orthogonal turning. Though, the fact that the difference is rather large and earlier temperature measurements have shown a total error in temperature of 25-30 °C [15,19] using an identical set-up by invoking the uncertainty of the emissivity value with an IR-camera in the error analysis. In further support of the accuracy of our temperature measurement we note that the recorded temperature is consistent with FEM-simulations of the temperature distribution on a cutting insert using a similar set up [18]. We therefore conclude that the temperature effect alone does not fully explain the large wavelength difference.

Instead we suggest that the external stress acting on the rake face promotes the spinodal decomposition as is discussed by Alling *et al* [8]. In the initial stage of the spinodal decomposition the fastest growing wavelength is determined by the second derivative of the free energy [21]. A more negative second derivative yields the shorter fastest growing wavelength for the coating used in the turning operation. This wavelength is then maintained whilst the amplitude in composition fluctuations increases until the second derivative of the

free energy is zero. At this point, a decrease in energy is only gained by minimizing the gradient energy, i.e. by coarsening of the domains [25,26]. As the driving force for separation is larger with the applied stress during cutting, this system will begin the coarsening stage earlier than the annealed system given that all other parameters are similar, e.g. temperature or diffusion coefficients. Despite the initial wavelength being shorter during cutting, its effect is overruled by the earlier start of the coarsening step. Based on our stress calculations however, the stress affecting position D is zero. Still, there is an influence in wavelength similar to what is seen at the other positions which seems contrary. We believe that this may be explained by the variations in terms of chip flow direction between orthogonal turning and longitudinal turning, the latter resulting in an additional stress contribution which results in higher stresses at all positions and a nonzero stress component at D.

The hydrostatic pressures (5-20 GPa) in the calculations by Alling *et al* [8] are however higher than those during cutting, but what is of interest is merely the trends rather than absolute figures. Also, there exist studies using finite element method reporting larger local contact stresses during longitudinal cutting [12,13] and hence it is possible that the stresses achieved during cutting is large enough to give notable differences in the evolution of the spinodal decomposition.

Based on the results we conclude that the microstructural evolution during cutting is affected by both the temperature and the stress distribution. However, whether it is beneficial or not for the tool life with highest normal stresses as possible remains to be proven. Most likely though, there is a limit where the positive influences on the $\text{Ti}_{1-x}\text{Al}_x\text{N}$ layer, e.g., the age hardening effects caused by the isostructural decomposition into coherent c-TiN and c-AlN domains, are dominated by the negative aspects of a too high yield stress resulting in

plastic deformation and eventually a catastrophic failure of the cutting insert by, e.g., a cutting edge break down.

4. Conclusions

Based on microstructural investigations with electron microscopy combined with thermal and mechanical analysis of the tool-chip contact, the following conclusions can be drawn:

- An ongoing spinodal decomposition of $\text{Ti}_{0.6}\text{Al}_{0.4}\text{N}$ is observed during metal cutting.
- The decomposition process is varied along the rake face due to an inhomogeneous temperature and applied stress distribution.

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