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Thermal conductivity of titanium nitride/titanium aluminum nitride multilayer coatings deposited by lateral rotating cathode arc

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ABSTRACT

A series of $[\mathrm{TiN/TiAlN}]_n$ multilayer coatings with different bilayer numbers $n = 5, 10, 25, 50$, and 100 were deposited on stainless steel substrate AISI 304 by a lateral rotating cathode arc technique in a flowing nitrogen atmosphere. The composition and microstructure of the coatings have been analyzed by using energy dispersive X-ray spectroscopy, X-ray diffraction (XRD), and conventional and high-resolution transmission electron microscopy (HRTEM). XRD analysis shows that the preferential orientation growth along the (111) direction is reduced in the multilayer coatings. TEM analysis reveals that the grain size of the coatings decreases with increasing bilayer number. HRTEM imaging of the multilayer coatings shows a high density misfit dislocation between the TiN and TiAlN layers. The cross-plane thermal conductivity of the coatings was measured by a pulsed photothermal reflectance technique. With increasing bilayer number, the multilayer coatings' thermal conductivity decreases gradually. This reduction of thermal conductivity can be ascribed to increased phonon scattering due to the disruption of columnar structure, reduced preferential orientation, decreased grain size of the coatings and present misfit dislocations at the interfaces.

1. Introduction

In order to improve the performance and service life of cutting and forming tools, nitrides of binary metal alloy coatings are coated on them resulting in a higher hardness and toughness. Ti-based hard coatings (e.g. TiN and TiAlN) are well known hard coatings in industry applications, typically deposited by physical vapor deposition [1,2]. TiAlN shows excellent mechanical properties, such as high hardness, oxidation resistance and excellent thermal stability [3,4]. Controlled deposition of the thin film allows producing multilayer materials with unique mechanical and physical properties. Knutsson et al. reported that the hardness of TiAlN/TiN multilayer coatings was higher than the single layer and increases with decreasing multilayer period [5]. It has been shown that multilayer TiN/TiAlN coatings are superior to single layer TiN and TiAlN coatings in many mechanical properties [6].

Besides the protection requirement against wear and oxidation, the thermal properties, and especially the thermal conductivity of hard coatings play an important role in their performance. For instance, materials with high thermal conductivity are required for electronic devices in order to dissipate heat, and act as a heat sink. Materials with low thermal conductivity are required for hard coatings and barrier coated elements in order to reduce the heat transport from the hot to the cold sides, and act as heat insulation. In high-speed machining, hot forging and casting, the temperature at the tool–chip interface moves up and this affects various tooling parameters (e.g. materials softening [7], friction condition and tool deformation [8]) and coating formation and properties (e.g. oxidation rate, hardness and thermal conductivity). A lower thermal conductivity of the hard coating is desired as a barrier layer to delay the temperature rising on the tool substrate materials and redirects heat flow into the chip and to dissipate the heat from the chip. This effect helps to reduce heat transport to the metallic components and thereby protects the tools against thermal overload [9–11]. Recently, it was reported that nano-multilayered TiAlN/Cu coatings have better cutting performance due to the lower thermal conductivity of nano-multilayered AlTiN/Cu than single layer TiAlN [12].

In our previous works [13,14], room temperature thermal conductivity values of a series of TiAlN coatings with a different Al/Ti atomic ratio and TiAlSiN nanocomposite coatings with different (Al + Si)/Ti atomic ratios on an AISI 304 substrate have been measured by pulsed

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Table 1
Ti and Al content in the TiN and TiAlN layers in the multilayer coatings.

Layer	Ti content (at.%)	Al content (at.%)
TiN	44.8 ± 2.8	–
TiAlN	20.5 ± 2.7	30.4 ± 3.0

photothermal reflectance technique (PPR). The results showed that the thermal conductivity of the TiN coatings decreases rapidly with Al incorporation and also it was found that the nanocomposite TiAlSiN coatings' thermal conductivity decreases with increasing (Al + Si)/Ti atomic ratio. In the present work, we investigate the thermal conductivity of TiN/TiAlN multilayer coatings, and demonstrate that it is lower than the thermal conductivity of TiN and TiAlN coatings. Moreover, it will be shown that the thermal conductivity depends on the coatings' microstructure and number of layers.

2. Experimental methods

$[TiN/TiAlN]_n$ multilayer coatings with a thickness of about 1.2 μm were grown onto a mirror-finished AISI 304 stainless steel substrate (with a dimension of 25 × 25 × 1 mm^3 and surface roughness of 10 nm) using a lateral rotating cathode arc (LARC) technique, which has been described in detail elsewhere [15]. For deposition of the multi-layer coatings, one elemental Ti and Al cathodes were employed in this work. These two cathodes are laterally rotating during the coating deposition. At first, the substrates were ultrasonically cleaned in a series of alkaline solutions, washed in deionized water, and dried by nitrogen gas blowing and further dried in an oven at 100 °C. Then the precleaned substrates were mounted on a carrousel substrate holder which rotated continuously around the vertical central axis at a speed of 12 rpm. The coating deposition was conducted in a flowing pure nitrogen atmosphere with a working pressure controlled at 1.5 Pa. During the deposition, a negative bias of −70 V was applied to the substrate, and the substrate temperature was controlled at 480 °C. In order to control the composition of the as-deposited coatings, the direct current applied on the two cathodes, I_{Al} and I_{Ti} was fixed at 120 A and 50 A, respectively. For depositing the $[TiAlN/TiN]_n$ multilayer coatings, the DC current applied on the Al cathode was turned on and off periodically. The total deposition time is fixed at 60 min, and the time to deposit each layer is $30/n$ min. We have prepared a series of five $[TiN/TiAlN]_n$ multilayer coatings with bilayer number n equal to 5, 10, 25, 50, and 100. The corresponding bilayer thickness ranges then from about 240 nm to 12 nm. Two TiN and TiAlN single layer coatings were deposited under identical conditions on the AISI 304 substrate as well. The DC current of the Al cathode I_{Al} for TiN deposition was fixed at 0 A.

The crystalline structure of the coatings is analyzed by X-ray diffraction (XRD). The XRD measurements were performed on a Burker AXS D8 Advance X-ray diffractometer by using a Cu Kα radiation (λ = 1.54178 Å, 40 kV, 40 mA) under θ–2θ scan mode in a 2θ range from 30° to 80°.

Sample preparation for cross sectional transmission electron microscopy (TEM) and scanning transmission electron microscopy (STEM) observation was performed using a focused ion beam DA300 system from FEI. TEM images were taken on the cross-section microstructure of the coatings using a Tecnai X-TWIN system from FEI, working at 200 kV. The dark field STEM images were taken using a Fischione high angle annular dark field detector. The chemical composition was measured by energy dispersive X-ray spectroscopy (EDX) analysis together with STEM imaging. Full quantification EDX analysis was performed using TEM Imaging and Analysis software.

HRTEM imaging was used to identify the misfit dislocation between the layers in the multilayer coating based on fast Fourier transformation (FFT) and geometric phase analysis (GPA). FFT and GPA of the high resolution micrographs were analyzed with Digital Micrograph software (version 3.11.1 Gatan). Bragg filtered image is generated by applying a spot mask to spots in the FFT of the HRTEM images. Afterwards, the FFT image was inverse Fourier filtered.

In order to characterize the thermal conductivity and thermal diffusivity of the coatings, the PPR method was used. First, a 1.0 μm thick gold film was deposited on the surface of the samples to enhance thermal absorption. A Nd-YAG laser (532 nm), operated in pulsed mode, with 7 ns full width at half maximum pulse duration, spot size (diameter of the laser beam on the gold layer) of 3 mm and pulse energy of 5 mJ was used to strike the sample and induce a temperature change at the surface of the samples. A 1 mW HeNe laser with 632.8 nm wavelength focused at the center of the pump spot with a 20 μm spot size to monitor

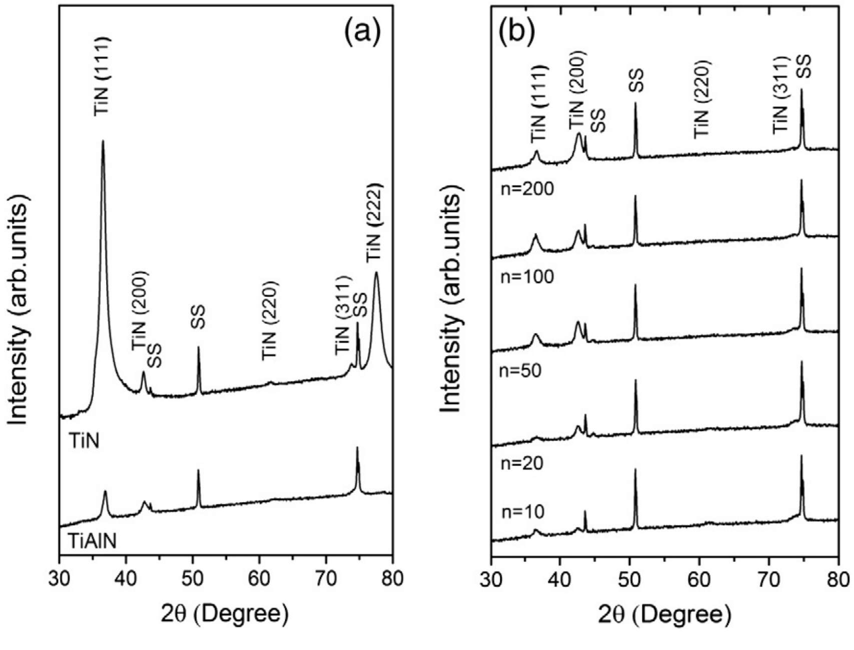

Fig. 1. X-ray diffraction pattern of the as-deposited (a) TiN and TiAlN single layer coatings and (b) $[TiN/TiAlN]_n$ multilayer coatings with different bilayer numbers n.