



Evaluation of fracture toughness of ZrN hard coatings by internal energy induced cracking method



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ABSTRACT

The objectives of this study were to evaluate the fracture toughness of ZrN hard coatings and to investigate the effects of preferred orientation and stress gradient on the fracture toughness. A recently developed energy-based method named internal energy induced cracking (IEIC) method was employed to measure the fracture toughness of ZrN hard coatings. ZrN film was continuously deposited until reaching the thickness at which the film was fractured due to stored energy accumulation. The residual stress before crack initiation was used to calculate the stored energy (G_s), from which fracture toughness (G_c) could be derived. The results showed that for a ZrN coating with (111) texture coefficient of 0.71, the fracture toughness was estimated to be ranged from 23.9 to 29.9 J/m². Fracture toughness of ZrN coatings was related to both residual stress and preferred orientation, which were controlled by deposition parameters. With lower substrate bias, the residual stress and accompanying stored energy were lower and the texture remained strong (111), and therefore thicker ZrN coating could be deposited. As the film thickness was above 4.22 μm, the (200) orientation increased in the strongly (111) textured ZrN coatings, which could be associated with the release of stored energy. The stored energy in the ZrN coating was partly released, when the energy was higher than 31.6 J/m², accompanying with the stress relief. The release of stored energy was considered to be the driving force of texture inversion. Stress gradient may play an important role in the fracture mode. The fracture could be initiated at a position of local maximum stress gradient along with defects. The fluctuating behavior in stress gradient was possibly owing to the accumulation and relaxation of residual stress during film growth, which may serve as a stress tuning mechanism in depositing thick coatings.

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1. Introduction

Fracture toughness is one of the crucial mechanical properties in the engineering application of structural materials. For bulk materials, fracture toughness can be routinely measured following ASTM E399 and E1820 standard procedures. Using fracture toughness as a key index, the probability of catastrophic failure for structural materials has been greatly decreased in the last few decades [1]. On the other hand, the measurement of fracture toughness for thin films or hard coatings is still problematic due to the thickness limitation and lack of reliable test procedures.

Numerous techniques have been proposed to evaluate the fracture toughness of thin films, such as tension [2–7], bending [8–10], buckling [11] and indentation [12–16] tests, which can be categorized into stress-based and energy-based methods [17]. For the stress-based methods, the toughness is obtained by measuring stress intensity factor following the ASTM standards. However, the design of reliable testing apparatus

for thin film specimens is still challenging; moreover, owing to the thickness limitation of thin film specimens, the specific substrate shape and micro-precrack are difficult to be routinely produced. For the energy-based methods, the fracture toughness is determined by the energy difference in a system caused by crack propagation, where the formation of fracture surface is solely related to the critical energy release rate. Consequently, the testing techniques are simpler compared with those of stress-based methods due to no requirements of pre-cracks and special stages [17]. In both methods, external stress is commonly required to apply on the specimen, which increases the difficulties in both stress distribution measurement and fracture toughness assessment, thus greatly complicating the experimental setup.

Recently, we proposed an energy-based technique named internal energy induced cracking (IEIC) method, by which the fracture toughness of TiN hard coatings was measured without externally applied stress and the influence of substrate effect [18]. The IEIC method used the intrinsic residual stress in the coatings to replace the external stress, where the thickness of TiN coating was continually increased by increasing deposition time until a major crack occurred from one edge of the rectangular specimen. The experimental method involved the measurements of residual stress using laser curvature method, Young's modulus by nanoindentation and film thickness from cross-sectional

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SEM images. The stored energy G_S in the coating with different thickness was calculated by the equation

$$G_S = \frac{(1-\nu_f^2)}{2E_f} \sigma_m^2 h_f \quad (1)$$

where σ_m is the residual stress, E_f is the Young's modulus, ν_f is the Poisson's ratio, and h_f is the thickness of the coating. When crack growth initiated, the minimum G_S in the film was taken to be the fracture toughness G_C of the coating. The theoretical basis and derivation of Eq. (1) were detailed in our previous paper [18]. The results showed that the fracture toughness of TiN hard coatings was $2.7 \text{ MPa}\cdot\text{m}^{1/2}$ which was close to the literature values [10,14,16]. In our previous study, TiN hard coating was selected as the model system owing to the well-characterized mechanical properties and appropriate elastic isotropy (Zener factor = 0.86) [19]. Furthermore, the residual stress of TiN coatings deposited by magnetron sputtering could be controlled by adjusting deposition parameters and measured nondestructively.

ZrN coatings have similar golden color but better mechanical, electronic, and optical properties than TiN coatings [20–23]; hence, ZrN has been replacing TiN to serve as the protective coatings in tool industry and diffusion barrier in microelectronic devices. In addition, since ZrN is a biocompatible material, ZrN coatings also have potential for biomedical applications.

Similar to TiN, most of the mechanical properties of ZrN have been well-characterized, but there is little information available on the fracture toughness of ZrN coatings. The fracture toughness of bulk ZrN was reported by Alexandre et al. [24]. They measured the fracture toughness of sintered ZrN bulk specimens and found that the fracture toughness of the specimens increased with decreasing porosity. Their results showed that fracture toughness of ZrN was mostly ranged from 35.3 to 48.1 J/m^2 except for the specimen with the lowest porosity ratio of 1.4% whose fracture toughness was about 94.5 J/m^2 . In addition, the reported Young's moduli ranging from 300 to 380 GPa were fairly consistent with the recent theoretical calculation value 350 GPa [25].

On the other hand, the influence of substrate bias on fracture toughness of ZrN thin films was systematically studied by Chen et al. [26] by direct crack measurement using Vickers indentation. The reported values of fracture toughness, ranging from 0.22 to 6.6 J/m^2 , were much smaller than those of bulk ZrN, and showing large data variation. The uncertainty may come from the difficulties in fulfilling the requirements of the indentation techniques, where the radial crack length (c) should be longer than the diagonal length ($2a$) of the indent and the indentation depth should be less than $1/10$ of the film thickness [12]. Furthermore, the reported Young's moduli ranging from 166.4 to 197.8 GPa were generally lower than the theoretical value [25]. However, the fracture toughness measured from bulk ZrN specimens cannot be directly compared with that obtained from thin films and coatings, because the density of thin films is normally lower than that of bulk counterpart; moreover, the density of thin films may be closely associated with the deposition method and deposition parameters.

Other factors that should be considered are the elastic anisotropy of ZrN and in-depth stress gradient in the coating, which may affect the measurement of fracture toughness and the fracture mode of the coating. Unlike TiN, ZrN is an elastically anisotropic material (Zener factor = 0.52) [19], and thus the fracture toughness of ZrN coatings may be more dependent on the preferred orientation. There have been two major theories for the formation and transition of texture in transition-metal nitride thin films. Pelleg et al. [27] suggested that for the transition-metal nitride with NaCl structure, (200) plane has the lowest surface energy while (111) plane has the lowest strain energy. With increasing film thickness, the overall energy will be dominant by strain energy and thereby the texture may switch from (200) to (111). Gall et al. [28] proposed a competitive growth theory to explain the texture transition in TiN. Since the topography of (111) is rough like

sawtooth while (200) is relatively smooth, the diffusion length for the Ti adatoms on (200) is much longer than on (111). Consequently, Ti adatoms are more likely to be trapped on (111) plane and thereby enhancing the growth of grain with (111) orientation. The (111) preferred conditions can be changed by increasing the N_2^+ ions on (200) plane to hinder the surface diffusion of Ti adatoms and facilitate the growth of (200) grains.

In addition, the coatings are commonly recognized to be under equibiaxial compressive stress state, where the crack initiation and propagation in a film/substrate system is usually along the interface of film/substrate or perpendicular to the film surface. However, it was observed in our previous study that cracks were formed inside the TiN coatings and fractured by loading mode II [18]. We conjectured that the in-depth stress gradient in the coating may initiate a crack at local defects and induce crack growth inside the film, driven by the release of elastic energy; yet, in-depth stress measurement was not performed to confirm the hypothesis. One common explanation for the stress gradient is that the gradient originates from two kinetically competitive stress generation mechanisms: growth-induced point defects by ion/atomic peening (compressive stress) and void formation due to surface roughness and shadowing effects generating attractive forces at the columnar boundaries (tensile stress) [29–31]. Shull and Spaepen [32] suggested that the fluctuation of stress between surface and film/substrate interface may be related to stress relaxation produced by the reconstruction of film structure. With increasing film thickness, the compressive stress caused by ion/atomic peening becomes difficult to release, because the amount of slip planes in the column is less than that in the equiaxed grain formed in the early stage of deposition [33]. If the film grows without stress relaxation, the accumulation of elastic strain energy may cause fracture or delamination. The purposes of this study were to measure the fracture toughness of ZrN hard coatings using IEC method and to investigate the effects of preferred orientation and stress gradient on the fracture toughness.

2. Experimental procedures

Deposition of ZrN film was carried out using dc unbalanced magnetron sputtering (UBMS). The substrate material was P-type Si(100) with dimensions of $35 \times 35 \times 0.525 \text{ mm}^3$. The substrates were ultrasonically cleaned progressively in acetone and methanol, each for 5 min, and then were dried by blowing nitrogen gas and immediately placed into the deposition chamber. Afterwards, the chamber was evacuated to a base pressure below $6.7 \times 10^{-4} \text{ Pa}$ ($5 \times 10^{-6} \text{ Torr}$) and the substrates were simultaneously heated to $400 \text{ }^\circ\text{C}$.

Before deposition, the substrates were pre-sputtered by Ar ion at a bias of -1000 V under an Ar (99.9995% in purity) flow rate of 50 sccm for 5 min to eliminate the surface oxide. Then, the working gas was introduced into the chamber, consisting of Ar and N_2 (99.9995% in purity) with flow rates of 30 sccm and 1.5 sccm, respectively. The working pressure in the chamber was maintained at $1.5 \times 10^{-1} \text{ Pa}$ ($1.1 \times 10^{-3} \text{ Torr}$). Single target was used in the sputtering system, and the target material was pure Zr (99.5 wt.% in purity). During deposition, the target current was fixed at 0.34 A and two substrate bias voltages, -70 V and -75 V , were applied to control the magnitude of residual stress. The distance between target and substrate was 10 cm. The rotating speed of the substrate holder was 18 rpm and the deposition temperature was $400 \text{ }^\circ\text{C}$. The film thickness was controlled by increasing deposition time ranging from 60 to 510 min.

X-ray photoelectron spectroscopy (XPS) was used to measure the N/Zr ratio of the ZrN coatings using an X-ray source of Mg K α (1253.6 eV) operating at 15 kV. Prior to analysis, the specimen was pre-sputtered by 3 keV Ar^+ ion for 5 min to remove the contamination on the surface. The deconvolution and fitting of the XPS spectra was used a computer program XPSPEAK 4.1, where the background was subtracted by the Shirley's method [34]. The uncertainty of the reported values of coating composition was within 10%. The

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