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Length-scale effects on fracture of multilayers

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Fracture of Ti/TiN multilayer specimens was studied in situ inside a transmission electron microscope. Fracture toughness for cracks propagating perpendicular to the multilayers is found to be thickness dependent, varying from 1.45 to 2.45 MPa-m$^{0.5}$ as the specimen thickness increased from 150 to 300 nm. Single-mode crack renucleation was observed in the metals layers, which is anomalous to the continuum-based elastic–plastic multilayer fracture model predictions. This is explained by the ultrafine columnar grain structure in the metal layers.

Keywords: Transmission electron microscopy; Toughness testing; Multilayer thin films; Nanocrystalline microstructure

Mechanical properties of metal–ceramic multilayers are important because these systems are prevalent in electronic devices [1], biomedical implants [2] and advanced coatings [3,4], where mutually exclusive features such as hardness and toughness are contributed by the ceramic and metal layers, respectively. As a result, fracture mechanics of metal–ceramic multilayers has been extensively studied in the literature [5–8]. In these studies, the metal and ceramic layers are assumed to exhibit isotropic and bulk (where continuum assumption holds good) properties. It is commonly accepted that stable propagation of a crack along a direction normal to that of the multilayer film is dependent upon the ratios of thickness and Young's modulus of metal and ceramic layers [9]. The stress at the crack tip is primarily relaxed by the plastic deformation in the metal layer. Upon further loading, the ceramic layers in front of the metal layer also relax some stress by developing microcracks ahead of the propagating crack tip – a process known as crack renucleation. Depending upon the extent of plasticity in the metal layers, renucleation in the ceramic layers can take place as one dominant crack in the same plane of the macroscale crack or as multiple cracks in different planes [5,6]. Another strengthening mechanism is the deflection and/or termination of the cracks at the interfaces. The most critical element of the metal ceramic multilayers reliability is therefore the plasticity of the metal layers, thickness of which has been decreasing from the microscale [10] to the nanoscale [11,12]. This trend is motivated by the Hall–Petch relation in multilayers, which predicts increase in strength as layer thickness decreases [13]. However, the reduction in thickness puts more scrutiny on the plasticity in metal films, which is known to be a strong function of layer thickness and grain size [14]. It is well known that nanoscale plasticity is a complex interplay of microstructure, lattice and grain boundary dislocations as well as diffusion [15–17]. Further, the microstructures of the metal and of the ceramic layer also dictate the coherency of the interface. A coherent interface may allow dislocation motion across it, whereas incoherent interface results in defects that may help pin the dislocation [18]. Another novel way to achieve a nanocomposite/multilayer structure has been reported [19] that uses diamond-like carbon to achieve the required deformation characteristics.

To understand the role of layer thickness and microstructure on the fracture properties of metal–ceramic multilayers, we adopt a unique experimental approach. We use a focused ion beam (FIB) to mill nanoscale specimens from bulk substrates. These specimens are then integrated with micro-electro-mechanical systems (MEMS) based mechanical testing tool to drastically miniaturize the overall fracture testing setup to about 3 mm × 5 mm. This allows the experiments to be

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performed in situ inside a transmission electron microscope [20], which visualizes dislocations, grain boundaries, cracks and other defects in real time with very high resolution. The motivation is to obtain quantitative (stress, strain, crack length) and qualitative (microstructural visualization) data simultaneously for a fundamental understanding of the physical mechanisms of multilayer fracture. We used magnetron sputtering to deposit alternate layers of titanium and titanium nitride on a silicon substrate. The thicknesses of the individual Ti and TiN layers were 850 and 450 nm, respectively. Single-edge fracture specimens about 20 μm long, 5 μm wide and 150–300 nm thick were prepared using a transmission electron microscopy (TEM) lift-out technique [21] inside a FEI Quanta 3D FIB scanning electron microscope. The fracture testing device used micromachined thermal actuators for loading and was equipped with force and displacement sensors. The operating principle [22] and an example application [23] of this setup are already described in the literature. Figure 1a and b shows a specimen coupon being lifted off the substrate and the micromachined fracture testing device, respectively. A magnified view of the specimen mounted on the force and displacement sensors is given in Figure 1c, while Figure 1d shows a further magnified view of the multilayer specimen.

To initiate sharp cracks for fracture testing, a U-shaped notch was milled on one edge of the specimen using FIB (Fig. 1d). The Ti/TiN specimen along with the device is then positioned on a custom designed single-tilt TEM holder for a JEOL 2010 transmission electron microscope with electrical feedthroughs. Figure 2 shows a TEM micrograph of one of the Ti–TiN specimens before loading in a mode I crack opening. The TiN layers are of NaCl B1 cubic crystal structure, whereas the Ti layers show a hexagonal closed-pack structure [24]. It should be noted that the both metal and ceramic layers had columnar grain structures that extend throughout the layer thickness. However, the

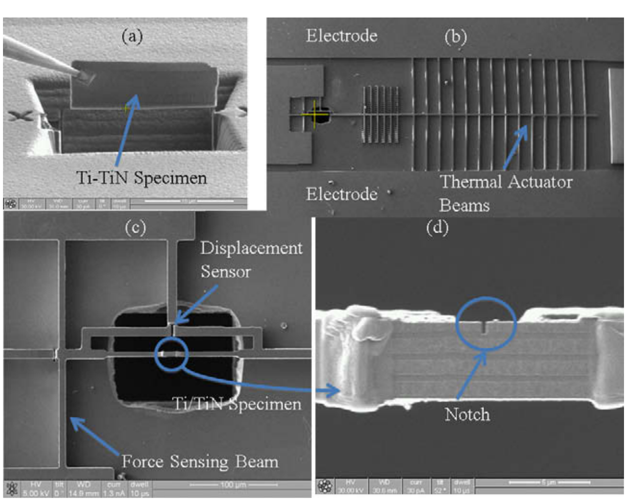

Figure 1. In situ TEM fracture testing setup: (a) Ti/TiN multilayer specimen picked using a nanomanipulator; (b) micromachined thermal actuator integrated with force and displacement sensors; (c) magnified view of the force and displacement sensors; and (d) magnified view of the specimen with a focus ion beam milled notch for crack initiation.

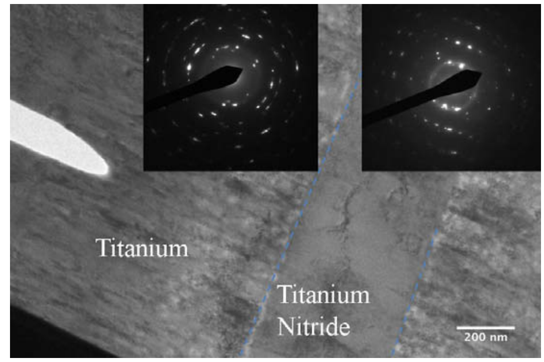

Figure 2. TEM micrograph of a Ti–TiN multilayer specimen (inset: selected area diffraction pattern).

average column diameter for the metals layers was very small (about 25 nm) compared to the layer thickness (850 nm). The TiN grains were also columnar, with grain diameter varying from 0.1 to 1 μm. Selected area diffraction patterns (insets in Fig. 2) confirm the strong 001 texture along the layer thickness direction. Such a columnar grain structure results in enhanced strain tolerance, lower thermal conductivity and higher thermal reflectivity [25], which are desired features for advanced multifunctional coatings. The Ti layers are seen to contain forests of dislocations, probably due to the large residual stress developed during the multilayer deposition. Dislocations are also seen in the TiN layer, albeit with lower density.

In a typical experiment, stress is applied on the specimen quasi-statically by supplying voltage to the MEMS actuator in steps of 0.1 V. It is expected that stress concentration at the notch will lead to a sharp crack that will propagate across the multilayers. Therefore, the notch was kept at the center of the screen while recording the video for each loading step. The force, displacement and crack length are measured for each loading step until catastrophic fracture of the specimen. For a 150 nm thick specimen the nominal fracture stress was found to be about 700 MPa, whereas a 300 nm thick specimen had a fracture stress of about 1.45 GPa. The average critical stress intensities for the 150 and 300 nm thick specimens were found to be 1.45 and 2.45 MPa-m$^{0.5}$, respectively. Such low toughness (bulk TiN value is about 5 MPa-m$^{0.5}$) and the strong thickness dependence in the critical stress intensity factor can be explained by the prevailing plane stress nature of the loading. Unfortunately, the Ti–TiN multilayer literature only reports elasticity [26], hardness [27] and wear [28] properties; and very little is known about the bulk fracture toughness [29]. It is expected that the bulk toughness values will be slightly higher than the thin film values for the present experimental setup. This is because the bulk specimens contain large compressive residual stress, which enables them to withstand large stresses before fracture. Also, bulk specimens are generally tested while still on substrates, which also increases the multilayer fracture stress because of the stress relaxation due to plasticity in the substrate [3] in addition to the multilayers. In comparison, our specimens were free from the substrate, and also partially relieved from the