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Investigating fracture of nanoscale metal–ceramic multilayers in the transmission electron microscope

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ABSTRACT

In-situ transmission electron microscopy is used to investigate crack propagation parallel to the interfaces of a Ti/ZrO_2 multilayer. The cracks propagate along the middle of the 100 nm thick polycrystalline Ti layers, causing extensive dislocation activity, void coalescence, and crack bridging. The plastic zone size has been determined from the range of dislocation activity and agrees well with estimates of the toughness obtained from the measured crack tip opening displacement. The toughness is much smaller than in bulk Ti, which we attribute to the constraint on dislocation activity and cropping of the plastic zone by the small Ti layer dimensions.

Fracture toughness often sets the limit for the use of materials in applications. Unfortunately, the factors controlling crack initiation and propagation are not well understood, particularly for nanocomposites, which have nanoscale dimensions and many interfaces. The activity of dislocations in nanostructured materials is primarily hampered by the small dimensions and by the presence of interfaces, thus affecting ductile crack propagation which proceeds via dislocation processes near the crack tip [1]. The structure of the material, such as grain boundaries, precipitates, inclusions, fiber reinforcement, etc., may lead to crack deflection and meandering, crack bridging, and branching into multiple cracks contribute towards a higher toughness [2]. Further, the cracks can be stopped by the interfaces [3], or delamination may take place depending on the interface strength. Many natural materials exploit these toughness-enhancing processes by combining ductile but weak materials with strong but brittle materials to achieve both high strength and high toughness in nanocomposites [4].

Technology copies nature by using nanocomposite design to achieve strength and toughness for many different applications. However, technological developments to achieve a sustainable future will not only demand that we make strong, tough and reliable nanocomposites but will also require that we can take them apart on demand, so that the materials can be reused or recycled. This will necessitate knowledge of the controlling mechanisms of fracture for physical separation of the constituent materials when chemical reclamation is not possible. Fracture behavior is not only affected by materials, interface structure and chemistry, and composite geometry [5], it is also fundamentally influenced by material and structural length scales in ways that depend on whether the bulk material is ductile or brittle. Flaw insensitivity [6–9], Griffith's criterion effects [10,11], and crack tip shielding [12,13] from dislocation pileups [14] may all contribute to toughening of brittle materials at small length sizes. In contrast, in the case of ductile materials nanostructuring and nanoscale features are widely observed to lead to a loss of toughness [15–18]. Although the stress required for dislocation motion increases in small volumes ("smaller is stronger"), thus contributing to an increase in the energy required to propagate a crack, the plastic zone size is greatly reduced by both the increased flow stress and by cropping at the sample boundaries. Since it is widely observed that the energy required for crack propagation is decreased in nanostructured and nanograined metals [7], the second effect is assumed to dominate.

The effects of constrained plasticity on crack propagation have been investigated in a number of layered metal/ceramic systems. In metal/ceramic systems under Mode I loading (opening), it was reported that a crack propagating parallel to the interfaces will be deflected into the layer with the lower modulus [19] which is most often the metal layer. Several studies on metal/ceramic systems have also reported crack propagation through the metallic layer as well as interface debonding [17]. Although it is generally expected that the process with the lowest fracture energy will dominate [17,18], no clear experimental trend has been found for the preferred crack path as the fracture energy is changed by varying the metal layer thickness [20–23]. Thus, there is no clear experimental confirmation for the expected behavior until now.

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Here, we attempt to understand the mechanisms controlling fracture in a metal/ceramic nanocomposite by conducting in-situ transmission electron microscope (TEM) experiments on Ti/ZrO_2 multilayers. We aim to identify the roles of length scales and interfaces on the crack propagation and crack tip plasticity and to understand how they control the toughness. We present studies of crack propagation parallel to the interfaces to illustrate the knowledge that can be gained from in-situ studies.

Multilayer films of titanium and zirconium oxide were prepared by pulsed laser deposition from a 99.99% pure Ti target and a 3 mol% Y_2O_3-doped ZrO_2 target onto oxidized single-crystalline Si (111) wafers. The individual layer thicknesses for both Ti and ZrO_2 are approximately 100 nm and the total film thickness is around 3 μm. The ZrO_2 is amorphous while the Ti layers have a columnar microstructure with an in-plane grain size of roughly half of the layer thickness (ca. 50 nm) and a strong (1010) out-of-plane texture [24,25]. Investigations with x-ray reflectometry and TEM show that the interfaces between the layers are smooth and sharp with a mean roughness of around 1 nm [26]. A more detailed description of the deposition process and multilayer microstructure is published elsewhere [24]. A final 3 μm thick capping layer of Si, which is necessary to prepare a geometry for in-situ fracture tests, was deposited on top of the films using ion beam sputtering.

For sample preparation, 200 μm wide strips were cut from the deposited wafers using a wafer saw and then glued onto 3 mm diameter copper TEM half-grids for further preparation with the focused ion beam (FIB, Fig. 1a). The FIB machining process includes milling at high ion beam currents (ion beam parallel to y-axis) to form a 10 μm wide (x-direction), 5 μm thick (z-direction), and 20 μm high (y-direction) block (Fig. 1a). Starting from this block, the region with the multilayers is thinned with decreasing ion beam currents to about 100 nm. Finally, a 1 pA ion beam parallel to the z-axis is used to form a notch parallel to the interfaces in the multilayer film, in order to have a preferred site for crack initiation. The geometry is designed for Mode I testing.

The in-situ fracture tests were performed using a TEM-STM holder (nano-Factory) inside an FEI Titan TEM with image correction at 300 kV. The prepared specimen is mounted in the holder opposite to a wedge-shaped tungsten tip that can be moved in all three directions using piezo actuators with sub-nanometer resolution. The tip was milled into a wedge shape with an opening angle of approximately 120° using the FIB. Once mounted in the TEM, the wedge was aligned to the notch in the fracture specimen (Fig. 1b) using the coarse movement controllers. From this point, the fine movement mode of the y-axis piezo actuator was used to achieve contact of the wedge tip with the Si frame of the fracture specimen and to pry the frame apart by displacing the wedge in the negative y-direction, which initiates a crack at the predefined notch and drives propagation of the crack. TEM images and movies were taken between and during loading.

Altogether, seven in-situ fracture tests were performed on the samples. Two of the tests led to unstable cracking and loss of the specimen. This was attributed to poor alignment of the tip with the specimen or to imperfections in the specimen geometry. The other tests (5 out of 7) showed the stable crack growth reported here.

Fig. 2 shows TEM micrographs of a crack during loading (online supplementary material available). The crack initiates at the apex of the FIB-prepared notch located within a single ZrO_2 layer (Fig. 2a) and immediately moves diagonally across the ZrO_2 layer and into an adjacent Ti layer, where it propagates through the middle of the Ti layer (Fig. 2b). The reconstructed overview of the crack at the end of loading (Fig. 2c) shows that the fracture surface in the Ti layer is rough, with features on the length scale of the grain size. In fact, it is quite difficult to locate the exact location of the crack tip because of the heterogeneous deformation and grains that bridge the crack. Although the contrast changes in TEM images are not always straightforward to interpret, there appears to be void formation and coalescence ahead of the crack tip — a characteristic of ductile fracture. To gain a better insight of the deformation along the crack, we define an opening displacement (*OD*) along the crack and ahead of the crack tip as $OD = d_{int} - t_{Ti}$, where d_{int} is the distance between the two neighboring Ti/ZrO_2 interfaces and t_{Ti} is the Ti layer thickness prior to crack initiation (Fig. 2d). The ZrO_2 layer thicknesses do not measurably change during crack propagation. The *OD* decreases linearly towards the position of the crack tip, but extends approximately 1 μm beyond the tip (Fig. 2d). The strains perpendicular to the Ti layer in front of the crack tip can be estimated as OD/t_{Ti} (right hand axis of Fig. 2d). They are much larger than the elastic limit, supporting the TEM observations of crack bridging and extended plasticity.

The movement of the crack tip is accompanied by "flickering" contrast changes in the Ti layer ahead of the crack tip as well as in the surrounding Ti layers. The flickering is attributed to dislocation activity, which is supported by the fact that no such contrast changes are observed in the amorphous ZrO_2 layers. The contrast changes occur within a distance of 750 nm of the crack tip, which we associate with the plastic zone size. There is no measurable trend indicating a change in the zone size as the crack progresses. In fact, the 1 μm extent of the plastic region beyond the blunted crack tip (Fig. 2d) is in reasonable agreement with the extent of dislocation activity (750 nm). The flickering continues

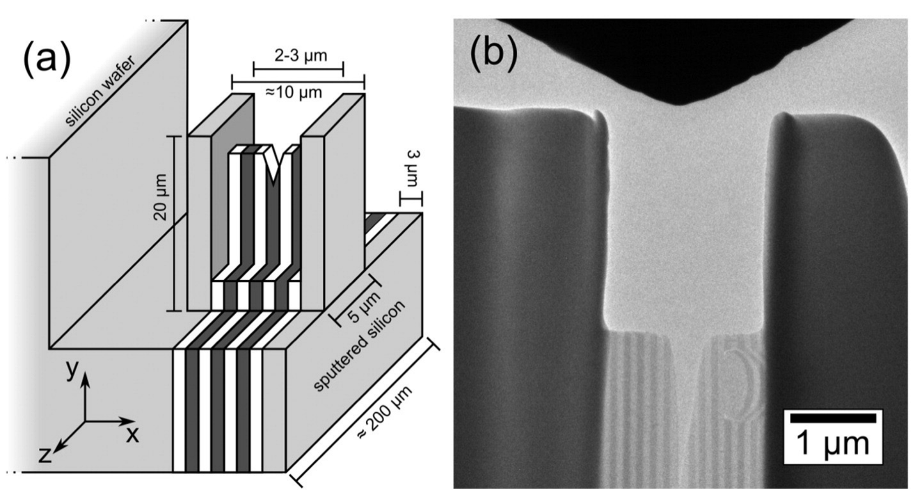

Fig. 1. (a) Schematic overview of the in-situ fracture specimen geometry (not to scale). The crack is loaded by prying the Si frame apart with a wedge tip while TEM imaging is performed with an electron beam incident along the z-direction. (b) Overview TEM micrograph showing the wedge tip approaching the lamella from the y-direction. The Ti droplet on the right part of the lamella results from the PLD preparation processes and does not affect the fracture tests.