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ScienceDirect
Acta Materialia 88 (2015) 252–259
Acta MATERIALIA
www.elsevier.com/locate/actamat

Sample size induced brittle-to-ductile transition of single-crystal aluminum nitride

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Received 9 August 2014; revised 13 January 2015; accepted 16 January 2015

Abstract—Ceramics are known to be mechanically hard, chemically inert and electrically insulating for many important applications. However, they usually suffer from brittleness and have moderate strength that strongly depends on their microscopic structure. In this study, we report a size induced brittle-to-ductile transition in single-crystal aluminum nitride (AlN). When the specimen diameters are smaller than ~3–4 μm, AlN micropillars show metal-like plastic flow under room-temperature uniaxial compression. The unprecedented plastic strain of ~5–10% together with the ultrahigh strength of ~6.7 GPa has never been achieved before. Transmission electron microscopy demonstrates that dislocations play a dominant role in the plasticity of the micro-sized AlN.

Keywords: Ceramics; Micropillar; Plastic deformation; Size effect; Transmission electron microscopy

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1. Introduction

Covalently bonded ceramics exhibit many distinctive physical and mechanical properties, compared to metallic and polymeric materials, but the propensity toward brittle fracture has limited their applications especially in forming and load bearing operations [1–6]. As exceptions, only a handful of ceramics show room-temperature plasticity via anomalous deformation processes, such as martensitic transformation [1,2], kink bands [3] and grain boundary sliding [4,7], or subjected to extreme loading conditions, such as shock [6,8], high pressure [9–11] and strain confinement [11–13]. Nevertheless, large dislocation plasticity is rarely seen in high strength ceramics during room-temperature uniaxial deformation [14].

Aluminum nitride (AlN) is a high-performance covalently bonded ceramic material and possesses high hardness and strength with relatively low specific density. It has a stable wurtzite-type structure with a smaller c_0/a_0 ratio of ~1.60 than the formal hexagonal close-packed lattice (1.633) [11]. AlN has been widely used in electronics and as a structural ceramic material. In the past decades, particular attention has been paid to its mechanical stability. Wilkins et al. first pointed out the importance of inelastic deformation in the impact performance of AlN [15]. Moreover, it has been reported that hydrostatic confinement facilitates dislocation plasticity of AlN [11,13]. However, the room temperature plasticity cannot be retained in confinement-free AlN, which always fails in a brittle manner without any plastic strain under quasi-static uniaxial deformation [11,15–18].

Since the advent and development of microsystems and nanotechnology, the understanding of micro-scale deformation and failure mechanisms of advanced materials has been gaining importance. It was recently observed that the deformation mechanisms of metallic alloys have shown a consistent difference in strength and plasticity between micro-sized and bulk specimens [19–21]. These findings strongly suggest that specimen size may influence the activation and motion of dislocations, and hence the mechanical response of ductile metals. Compared to metallic materials, the mechanical response of ceramics is expected to be strongly size-dependent because of their rigid ionic and covalent bonding which is highly sensitive to defects and flaws. However, for strong and relatively brittle ceramics, the size dependence of their strength and ductility has not been well explored [4,22–24]. Recently, the size induced plasticity has been observed in covalent and ionic crystals, such as sapphire [25], GaAs [26], SiC [27] and MgO [28], during room-temperature microcompression. These studies indicate that decreasing sample sizes to the order of micrometer scale can suppress cracking and leads to the change in the deformation behavior from brittle to ductile [29,30]. In this work, we report the deformation behavior

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http://dx.doi.org/10.1016/j.actamat.2015.01.043
1359-6462/

of single crystal AlN micropillars subjected to uniaxial compression along the [0001] and $[10\bar{1}0]$ axes. The microcompression experiments at a small length scale reveal unprecedented large dislocation plasticity in AlN that has been conventionally characterized as a brittle ceramic.

2. Experimental details

2.1. Materials and microcompression testing

AlN single crystals used in this study were commercially obtained from Fairfield Crystal Technology. The single crystals appear in the form of a hexagonal pillar as shown in the inset of Fig. 1. Micropillars for compression testing have the top surfaces parallel to the basal plane (0001) and side plane $(10\bar{1}0)$, respectively. The microcompression technique developed by Uchic et al. [20] has proven itself as a reliable way to explore the mechanical behavior of micro-size specimens. Here we have used a focus ion beam (FIB) system to fabricate columnar micropillars with diameters ranging from ∼2.5 to 20 μm and the aspect ratio approximately 2.5:1 for diameters below 10 μm and 2:1 for above 10 μm samples. The micropillars located in the center of large trenches which can guarantee that a flat punch is in contact only with the pillars during compression testing. The uniaxial compression tests were carried out by nanoindentation device (Shimadzu W201S) and the tests were performed using flat punch indenters with diameters of 10 μm and 40 μm, respectively. All the single-crystal micropillars were compressed to a preset depth of 10% to 15% of the initial heights of the pillars at a strain rate of $\sim 10^{-4}$ 1/s. The engineering stress–strain curves were calculated based on instantaneous measurements of load–displacement data and the specimen dimensions were determined precisely by scanning electron microscopy (SEM) after FIB fabrication. All the tests were conducted at room temperature. The active slip systems were determined on the basis of crystallographic and geometric analyses with the help of Schmid's law.

2.2. Microstructure characterization

The single crystal nature of the AlN specimens was verified by X-ray diffraction (XRD, RINT 2200, Rigaku X-ray diffractometer) and Raman Microscopy (Renishaw, U.K.). The cross-sectional TEM specimens of compressed micropillars were prepared by a lift-out FIB technique using a dual-beam FIB/SEM system (JEOL, JIB-4600F). The deformation regions of micropillars were characterized by TEM (JEOL JEM-2100F) operated at 200 kV.

3. Results

3.1. Orientation determination of single-crystal AlN

The single-crystal nature of the AlN specimens used in this study was determined by Raman spectroscopy. Raman spectra taken from polished basal plane (0001) and prismatic plane $(10\bar{1}0)$ present an obvious crystallographic orientation dependence of Raman scattering as shown in Fig. 1. Along the direction perpendicular to the (0001) plane, only the E_2^1, E_2^2 and A_1(LO) phonons, centered at 249.1, 656.4, and 888.2 cm^{-1}, respectively, are observed in the backscattering Raman spectrum. In the direction perpendicular to the $(10\bar{1}0)$ plane, the TO component of A_1(TO) at 611.3 cm^{-1} and E_1(TO) at 669.8 cm^{-1} are detected while the A_1(LO) becomes inactive. These stress free phonon modes of (0001) and $(10\bar{1}0)$ planes are fully consistent with the selection rules for the backscattering geometry of single-crystal AlN [31,32]. Separate X-ray diffraction further confirms the single-crystal nature and crystallographic orientations of the samples (data not shown here). Both diffraction spectra from basal and prismatic planes show a single-crystal feature and the diffraction peaks are fully consistent with the prediction from the theoretical structure model of wurtzite AlN.

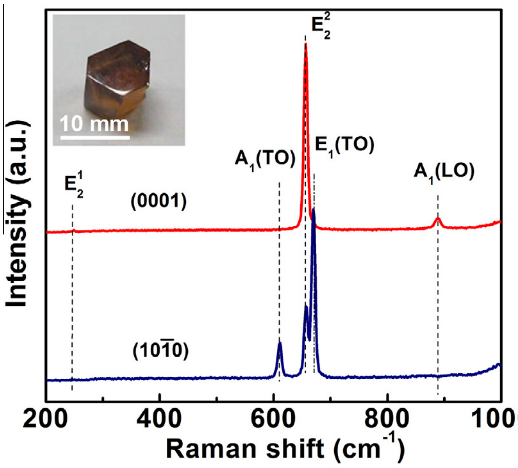

Fig. 1. Raman spectroscopy of single-crystal AlN. Raman spectra were taken from the basal plane (0001) and prismatic plane $(10\bar{1}0)$ of single crystal AlN. Inset photograph shows an as-received AlN single crystal.

3.2. AlN micropillar compression along [0001] direction

A representative engineering stress–strain curve of a (0001) micropillar with a diameter of 4 μm is shown in Fig. 2a. The compressive deformation response is characterized by linear elastic behavior up to ∼6 GPa where macroscopic yielding initiates, followed by ∼7% plastic flow and 6.7 GPa ultimate strength before failure. One important aspect of the stress–strain curve is the work hardening behavior, which is frequently observed in ductile metals but rarely in ceramics. Since work hardening usually originates from the interaction of dislocations from different slip systems, it indicates that multiple slip systems are activated in the (0001) single crystal. Fig. 2b and c shows the SEM images of the (0001) micropillar before and after uniaxial compression. Corresponding to the large plastic strain, the SEM micrograph reveals the presence of multiple shearing traces along two intersecting slip planes on the pillar surface (Fig. 2c). These observations indicate that at least two independent slip systems are activated during the plastic deformation. The slip planes have an angle of ∼55° with respect to the (0001) plane and could correspond to either $\{2\bar{1}\bar{1}2\}$ or $\{1\bar{1}01\}$ planes. The slip directions for each of these planes are $\langle 11\bar{2}3\rangle$ ($c + a$ type). Both of the slip systems have been frequently observed in the wurtzite structure [33], which are expected to provide five independent slip modes to satisfy von Mises criterion for continuous