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Size and plasticity effects in zirconia micropillars compression

Erik Camposilvan [a, b, *], Marc Anglada [a, b]

[a] Department of Materials Science and Metallurgical Engineering, Universitat Politècnica de Catalunya, Av. Diagonal 647, 08028 Barcelona, Spain
[b] Center for Research in NanoEngineering CRnE, Universitat Politècnica de Catalunya, C/ Pascual i Vila 15, 08028 Barcelona, Spain

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ABSTRACT

The micropillar compression technique has shown the potential for activating the brittle-to-ductile transition in ceramic monocrystals when testing reduced volumes. In this work, the role of size is studied by comparing the mechanical response of polycrystalline tetragonal zirconia micropillars and macroscopic specimens under compression. In micropillars, the absence of the natural defect population typical of bulk zirconia increases considerably the strength, allowing the activation of plastic deformation mechanisms and their study, showing in this way that the brittle-to-ductile transition is not limited to ceramic monocrystals only. The main mechanism of plastic deformation is transformation-induced plasticity, which is shown to be size dependent. The deformation behavior is studied in detail by loading-unloading tests at constant and increasing peak stresses, while the microstructure evolution is revealed by FIB cross-sections, TEM and STEM observations performed on lamellas extracted from pillars retrieved before failure. Finally, a failure mechanism is proposed, based on the damage induced by phase transformation.

1. Introduction

Intrinsic mechanical properties of materials, such as yield stress and strength, may exhibit an extrinsic behavior when their volume is greatly reduced. This tendency has been observed in monocrystals, where increasing yield stress is obtained by reducing the sample size. The existence of size effects has been highlighted by testing in different modes, such as micro- and nanoindentation [1,2], torsion tests [3], bending [4,5], tension [6] and compression [7].

The micropillar compression approach has received special attention due to several advantages. Firstly, the absence of important strain gradients that are usually responsible for substantial extrinsic contributions and can mask the effect of the sole volume and free surfaces [3,8]. Secondly, the relative simplicity of sample preparation and testing is another merit of micro-compression, which can be performed either ex-situ or in-situ under SEM or TEM observation [9], as well as coupled with other electrical and acoustical [10], spectroscopic [11] and diffraction [12] techniques.

The limitations of this approach are mainly related to the micropillar taper angle, unavoidable in most of the sample preparation routes, and the confinement at the base of the specimens imposed by the surrounding material. Other limitations are related to the possible misalignment and the presence of friction forces between the micropillar top surface and the flat-punch compression tip. All these effects contribute to the non-uniformity of the micropillar stress state, making difficult to compare samples of different size and taper angle [13–15]. Misaligning of the system leads to underestimation of the elastic modulus, stress concentration at the pillar top edge, and the possibility of failure by buckling instead of compression [15]. Thermal and mechanical drift during testing may also be a source of errors in strain measurement [16].

By testing single-crystal micropillars of soft metals in compression one can study the activation of individual slip systems and the yield stress as a function of pillar size. An inverse power-law has been found between the critical resolved shear stress along the slip plane normalized by the shear modulus and the pillar diameter [14,17]. This relationship holds for FCC single crystals, for which the exponent is close to 0.6, while in BCC single crystals the behavior is somehow erratic and very sensitive to the initial dislocation density, with the yield stress also scaling with micropillar size trough an inverse power law, but now with lower exponents [14]. At the same time, both the extent of the size effect and the length scale where the transition between bulk and small-scale behavior takes place appear to depend on the bulk shear yield

* Corresponding author. Department of Materials Science and Metallurgical Engineering, Universitat Politècnica de Catalunya, Av. Diagonal 647, 08028 Barcelona, Spain.
E-mail addresses: erik.camposilvan@upc.edu, erik.camposilvan@gmail.com (E. Camposilvan).

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stress [18]. Thus, soft materials show more pronounced size effects and the transition occurs in bigger samples.

At present, full understanding of the behavior of polycrystalline micropillars is still lacking. In these materials a combination of extrinsic and intrinsic length scales seems to affect the overall behavior. Different and conflicting deviations from the Hall–Petch relation have been found by independent studies in polycrystalline wires, sheets and micro-pillars of FCC metals [19]. In particular, in nanocrystalline Nickel it has been shown that the "smaller being stronger" trend can be inverted depending on the ratio thickness/grain size [20]. As a result of dislocation dynamics simulations, two types of behavior have been deduced for metallic systems, depending on sample size, grain size, and dislocation density [21]. One is dominated by strain-hardening and follows the Hall–Petch relation; in the other type, plasticity is source-limited and the "smaller being stronger" behavior is expected, reflecting in part the tendency previously observed by Janssen et al. for Aluminum [22].

With reference to structural ceramics, testing of single-crystal micropillars has received much less attention. Nevertheless, this technique may be of particular interest to investigate the transition from brittle to ductile behavior in ceramics when the volume becomes small enough [23,24]. In this sense, Korte and Clegg [18] have shown that hard brittle materials like MgO can be deformed plastically at room temperature by micropillar compression. Once plasticity was activated, the size effect was similar to BCC metals for MgO soft slip systems, while hard slip systems showed a behavior rather similar to GaAs [25] and Si micropillars [26]. Deformation by dislocation glide during micropillar compression could also be activated at room temperature in high strength ceramic monocrystals like sapphire [27], silicon carbide [28] and silicon nitride [29]. With respect to zirconia, Lai and co-workers [30] have recently shown that single-crystal or oligocrystalline micropillars of zirconia highly doped with Ce and Y can display shape memory and superelastic effects in association with phase transformation under compression, while the presence of other plastic phenomena has not been discussed.

In the present work, the compression behavior of yttria-stabilized zirconia (3Y-TZP) micropillars has been studied and compared with the one of macroscopic samples. This material is composed of tetragonal-metastable polycrystals with ~10 vol.% of stable cubic phase. It is well known that the transformation from the tetragonal phase into the stable monoclinic phase can be locally triggered, either mechanically by the presence of high stresses [31] or chemically by the diffusion of water species from the environment [31,32]. Here, 3Y-TZP micro- and nano-pillars of different sizes were milled by focused ion beam (FIB) and tested by monotonic and cyclic compression tests in order to investigate the role of size, phase transformation, plastic deformation and damage. All these aspects could be studied thanks to the considerable increase in strength of micropillars as compared to macroscopic specimens.

2. Materials and methods

Commercial spray-dried zirconia powder (TZ-3YSB-E, Tosoh Corp.) was isostatically pressed at 200 MPa in a rod shape and sintered at 1450 °C in air inside a tubular furnace for 2 h, obtaining a ceramic piece with density of 6.06 ± 0.02 g/cm^3 (99.5 ± 0.3% of the theoretical value) as measured by the Archimedes' method. The rod was cut into disks of approx. 1.5 mm thickness, which were ground and polished with diamond pastes down to less than 20 nm Ra. The disks were cut along the diameter and the resulting cross-section was polished using diamond films on a tripod fixture (Struers A/S) to reduce blunting of the cross-sectional edge. The disk halves were mounted on inclinable SEM holders with hard conductive adhesive and coated with a thin carbon layer of ~10 nm. The holder was tilted conveniently to facilitate both the parallel and normal visualization of the sample surface by the electron and ion beams employed during micropillars milling in a Zeiss Neon 40 dual beam station. The surface was visualized tangentially by the ion beam to set the reference angle for milling and then the stage was tilted and orientated accordingly for starting the milling procedures. The micropillars were milled directly into the sample polished surfaces, close to the edge of the cross-section, in order to allow better visualization after testing. Milling was performed with a two-steps procedure that had been optimized for each pillar size in order to minimize the taper angle and maintain similar aspect ratios and reasonable milling times. In the first step a large well, ~34 μm in diameter, was attained with a relatively high ion current. The size of the well had been adapted, on one hand, to facilitate the compression experiments avoiding contact between the indenter tip and the surface and, on the other hand, to allow locating the pillar by the built-in optical microscopes mounted on the two nanoindenters employed. In the second step, lower currents were used and the milling profile, i.e. the dwell time associated to each pixel as a function of the distance from the pillar axis, was tuned for each pillar size. Four nominal sizes were selected: 3.3 μm, 1 μm, 0.65 μm and 0.30 μm in diameter. The aspect ratio was between 1:2 and 1:4 in all the pillars. A thin cyanoacrylate strip was manually deposited on the half disk from where the samples were milled, approximately 500 μm away from the micropillars edge, serving as a soft surface for detecting the shape and location of the mark left by the flat-punch tip before testing. The two nanoindentation systems employed for testing were an MTS XP Nanoindenter (now Agilent, Oak Ridge, TN) and a CSM Ultra Nano Hardness Tester (CSM Instruments, Peseux, Switzerland). In both systems, the site of the indentation was first selected by imaging the surface with the optical microscope and then the indenter (or the sample, depending on the system) was moved to perform the indentation at the selected location. Therefore, the distance between the microscope and the indenter tip had to be properly calibrated. Prior to testing, the SEM holder was tilted to orientate the surface normally to the loading axis and mounted on supports designed for the indenter. Once inside the nanoindentation system, additional adjustments on the tilting angle were performed, if needed, by indenting the cyanoacrylate strip and visualizing the mark with the microscope until a regular shape was observed also at small indentation depths. The loading rates were adjusted to obtain similar strain rates ($1–3 \times 10^{-3}$ s^{-1}) for all the conditions and tests duration of approximately 40 s (monotonic tests to failure). Accurate microscope-to-indenter calibrations were performed by indenting on the cyanoacrylate strip before each single test to guarantee the correct positioning of the flat-punch tip over the pillar head. This procedure was also useful for cleaning the indenter tip from residual debris from broken samples. Compression testing at constant loading rate was used in all the tests. Repeated loading/unloading tests until rupture were also performed by loading between a constant low load and either constant or increasing peak loads.

The pillars were visualized before and after testing. The actual diameter, height and taper angle of each pillar were measured from SEM micrographs. Since the maximum stress occurs at the pillar top, the initial diameter measured at this location was used in the calculation of stress. Load-displacement data were converted into stress-strain curves to compare different sample sizes. Some pillars were unloaded before breakage to observe their appearance by SEM and, in selected cases, to observe a FIB cross-section or to lift out a thin lamella from the pillar cross-section. The latter was observed first by Scanning Transmission Electron Microscopy (STEM) inside a Zeiss Neon 40 dual-beam station equipped with a STEM module at 30 KV, obtaining bright field and annular dark field images. Conventional Transmission Electron Microscopy (TEM)