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Mechanical properties of metal-ceramic nanolaminates: Effect of constraint and temperature

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ABSTRACT

Al/SiC nanolaminates with equal nominal thicknesses of the Al and SiC layers (10, 25, 50 and 100 nm) were manufactured by magnetron sputtering. The mechanical properties were measured at 25 °C and 100 °C by means of nanoindentation and micropillar compression tests and the deformation mechanisms were analyzed by *in situ* micropillar compression tests in the transmission electron microscope. In addition, finite element simulations of both tests were carried out to ascertain the role played by the strength of the Al layers and by the elastic constraint of the ceramic layers on the plastic flow of Al in the mechanical response. It was found that the mechanical response was mainly controlled by the constraint during nanoindentation or micropillar compression tests of very thin layered ($\approx$10 nm) laminates, while the influence of the strength of Al layers was not as critical. This behavior was reversed, however, for thick layered laminates (100 nm). These mechanisms point to the different effects of layer thickness during nanoindentation and micropillar compression, at both temperatures, and showed the critical role played by constraint on the mechanical response of nanolaminates made of materials with a very large difference in the elasto-plastic properties.

1. Introduction

Metal-ceramic composites were designed to combine the ductility and toughness of metals with the hardness and high temperature strength of ceramics [1]. Early studies showed that the elastic constraint imposed by the stiff ceramic reinforcement on the plastic deformation of metals introduced high hydrostatic stresses, which increased the strain hardening rate of the metal and improved the strength [2]. However, the presence of tensile triaxial stresses triggered early damage by void nucleation and growth in the metal, as well as brittle tensile fracture in the ceramic [2–4], leading to low ductility and toughness, so the full potential of these materials could not be achieved.

These limitations can be overcome with the development of metal-ceramic laminates with layer thicknesses in the range of a few nm to 100–200 nm [5–15]. Metal-ceramic nanolaminates exhibit very high hardness, which has been related to the plastic deformation mechanisms in the metallic layers [11]. For the thicker layers ($\gtrsim$ 50 nm), the yield strength of the metallic layers appears to be controlled by dislocation pile-ups, while confined layer slip might dominate the deformation for thinner layers ($\lesssim$ 50 nm), which are too small to accommodate dislocation pile-ups [11]. Due to their larger intrinsic strength, the ceramic layers are expected to undergo either elastic deformation or fracture during deformation, but further strengthening is also expected from the layer thickness reduction due to the Griffith effect that results from the reduction in the size of pre-existing flaws. For very thin layers, the stresses can become so high that might activate plastic co-deformation of the metallic and ceramic layers, leading to a transition from a brittle to a ductile behavior. This behavior was found in Al/TiN nanolaminates when the bilayer thickness was reduced to a few nanometers [11].

Even though these strengthening effects are expected to operate in most metal-ceramic nanolaminates, the dependence of hardness

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with the layer thickness has been shown to vary from system to system. For instance, the hardness of Al/TiN nanolaminates [11,12] showed a noticeable dependence with layer thickness, while it was rather weak in Cu/TiN [13] or Al/SiC nanolaminates [8,10]. Some of these differences have been attributed to the characteristic length scale controlling plasticity in each case, which depends on the grain size of the individual layers [13]. For instance, both Al and TiN layers in Al/TiN nanolaminates presented a columnar grain structure with in-plane grain sizes that were relatively larger than the individual layer thickness. Hence, the hardness was controlled by the interaction of dislocations with the metal-ceramic interfaces. In the case of Cu/TiN [13], both Cu and TiN layers were nanocrystalline with a grain size that was much smaller than the layer thickness (due to their lower chemical affinity) and dislocation/grain boundary interactions seemed to control the plastic deformation.

All in all, the hardness of metal-ceramic nanolaminates has shown a relatively weak dependence on layer thickness (even when microstructural features, like the grain size in each layer, are considered) and this behavior was particularly obvious when the mismatch in mechanical properties between the soft metal and the hard ceramic layers was very large. Al/SiC nanolaminates, which have been profusely studied in literature [8,10], are a paradigmatic example. The SiC layers are amorphous and, therefore, they do not introduce an internal length scale, like the grain size, that can compete with the layer thickness. It was shown that the hardness was strongly influenced by the volume fraction of the ceramic constituent [8], and that the thickness of the Al layers had little effect on the hardness, even though their yield strength was layer thickness dependent [10]. This was attributed to the large constraint imposed on the deformation of the metal-ceramic nanolaminate during the nanoindentation test. Under these circumstances, the plastic deformation of the metallic layers is not only confined by the ceramic layers, but there is an additional source of elastic constraint imposed by the undeformed material surrounding the hardness impression. These highly constraint conditions, in the case of nanoindentation, might conceal the actual role played by the active deformation mechanisms in the metallic layers. The motivation of this work was therefore, to use an alternative micromechanical testing technique, like micropillar compression, to get a better understanding of the active deformation mechanisms and of the role of constraint in Al/SiC nanolaminates as a function of layer thickness and temperature.

To this end, the mechanical behavior of Al/SiC nanolaminates with equal layer thicknesses (in the range 10 nm–100 nm) was determined by means of nanoindentation and micropillar compression tests at ambient and elevated temperature. Nanoindentation and micropillar compression tests allow the exploration of the effect of constraint for different layer thicknesses, while the strength of nanoscale metallic layers decreases rapidly with temperature. In addition, numerical simulations of both tests were carried out by means of the finite element method in order to understand the experimental observations.

2. Materials and experimental techniques

2.1. Materials

The Al/SiC nanoscale multilayers were deposited by magnetron sputtering. Details on the sputter unit and the processing conditions can be found elsewhere [8,10]. Four different nanolaminates were manufactured with equal nominal values of the thickness of the Al and SiC layers: 10, 25, 50 and 100 nm, all with a nominal volume fraction of each constituent of 50%. The following notation is used to refer to each nanolaminate: AlxSiCy, where x and y represent the Al and the SiC nominal layer thickness, respectively, in nanometers. All the nanolaminates were deposited to a total film thickness above 10 μm. The nanolaminates were characterized by TEM using a JEOL JEM 2100 microscope (JEOL Ltd., Tokyo, Japan). The cross-sectional TEM specimens were prepared by FIB using a dual beam Field Emission Gun SEM (FEI Helios 600i, Hillsboro, USA).

2.2. Mechanical characterization

The mechanical properties of the nanolaminates at 25 °C and 100 °C were measured by means of nanoindentation and micropillar compression tests, using a NanoTest™ platform III (Micro Materials, Wrexham, UK). This nanoindentation platform uses independent heating of the specimen and the indentation tip, which is the best strategy to achieve thermal equilibrium during indentation in order to minimize thermal drift. The indentations were carried out with a Berkovich tip to a maximum load of 100 mN at a loading rate of 10 mN/s, a holding time of 5 s at maximum load, followed by unloading at 20 mN/s. The maximum penetration was always below 10% of the total film thickness, thus avoiding substrate effects on the indentation curves. The load-displacement curves were analyzed using the Oliver and Pharr method [17].

Micropillars with 2 μm in diameter and 4 μm in height were fabricated following an annular milling procedure using the same dual beam FIB. The milling was carried out in several steps by employing ion currents from 9.3 nA down to 80 pA in the final polishing step, using a procedure that ensures minimum FIB induced damage [7]. The final micropillars were slightly tapered, but the tapper angle was generally <2°. The micropillars were compressed at 25 °C and 100 °C using a 10 μm diamond flat punch. The thermal drift rates at both temperatures were carefully controlled within 0.1 nm/s. The compression tests were performed in displacement-control mode at a nominal strain rate of 1.25×10^{-3} s^{-1} up to a maximum engineering strain of 0.12. Engineering stresses were computed using the top area of the micropillars.

2.3. In situ mechanical tests within the transmission electron microscope

In order to ascertain the deformation micromechanisms, Al100SiC100 and Al10SiC10 micropillars were also tested *in situ* within a Tecnai F20 TEM (FEI Co., Hillsboro, USA) using a PI95 picoindenter (Hysitron, Inc., Minneapolis, USA). To this end, electron transparent micropillars, with a height of ≈1 μm and a width of ≈500 nm, were also milled using the FIB and compressed *in situ* using a flat punch.

3. Numerical model

Nanoindentation and micropillar compression tests were simulated using the finite element method to ascertain the role of the yield stress of the Al layers, of the layer thickness and of the constraint effect imposed by the SiC layers on the mechanical response. The geometrical model for the simulation of the nanoindentation test is equal to the one in Ref. [10] and it is depicted in Fig. 1a for the sake of completion. The indentation model includes the Si substrate, all the nanolaminate layers (up to a total thickness of 10 μm) and the rigid conical indenter with a semi-angle of 70.3°. Four models with equal Al and SiC nominal layer thicknesses of 10, 25, 50 and 100 nm were built. The geometrical models were discretized using two-dimensional four-node linear axisymmetric elements (CAX4). The lateral dimensions of the models were large enough to avoid any boundary effects. The bottom of the model was