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Load-dependent high temperature tribological properties of silver tantalate coatings

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ABSTRACT

Silver tantalate coatings were produced by reactive unbalanced magnetron sputtering from silver and tantalum sources as potential high temperature solid lubricants. The films were wear-tested at 750 °C under normal loads of 1, 2, 5, or 10 N against a Si_3N_4 counterface. These sliding tests revealed that the friction monotonically increased as the load was increased. A systematic investigation of the surface and sub-surface regions of the wear track, using techniques such as Scanning Auger Nanoprobe, Atom Probe Tomography, and cross-sectional transmission electron microscopy, revealed the following trends with increasing load: (1) a decrease in the amount of Ag on the surface of the wear track; (2) a decrease in the thickness of the mechanically mixed layer that forms as a result of the reconstruction of $AgTaO_3$ to form Ta_2O_5 and Ag; and (3) the formation of a porous structure throughout the tribofilm as a result of the segregation and migration of Ag from the original $AgTaO_3$ matrix. These results were complemented by molecular dynamics simulations, which confirmed the increase of friction with load. Further, the simulations support the hypothesis that this trend can be explained in terms of decreased presence of Ag clusters near the sliding surface and the associated decreased porosity.

1. Introduction

Oxides exhibit an unparalleled spectrum of physical properties, which gives them the potential to be incorporated into next-generation devices used in optics, magnetism, and electronics [1]. Among oxides, ternary oxides (more specifically, those that consist of two metals in addition to oxygen) that possess layered crystal structure were very recently shown to reduce friction at high temperatures ($T > 500$ °C) [2]. These oxides may be used as effective solid lubricants that are applied between sliding surfaces that operate under extreme conditions [3]. Currently, the most commonly used solid lubricants at high temperatures are soft metals and a few binary oxides or fluorides [4,5]. Ternary oxides were recently shown to be more effective than these other materials since they exhibit friction coefficients < 0.3 at high temperatures. Examples of ternary oxides that have been recently investigated include silver, copper, and cesium molybdates [6–9], silver and copper vanadates [10,11], silver niobates [12], and cesium silicates [13]. Erdemir [14] formulated the concept of crystal chemical model that stipulates that there is a correlation between the difference in ionic potentials and the high-temperature friction coefficients for ternary oxides.

Silver tantalate is a ternary oxide with perovskite crystal structure whose tribological properties were recently investigated by the authors as a potential lubricious high temperature material [15]. This material was created (i) in powder form using the solid-state synthesis technique and (ii) as a coating grown on Inconel 718 substrates using unbalanced magnetron sputtering. $AgTaO_3$, grown by sputtering, displayed a coefficient of friction (CoF) of 0.06 when tribotested at 750 °C using a 2 N load, which is a very low value at such elevated temperatures. Cross-sectional TEM studies after tribotesting revealed that $AgTaO_3$ is the main phase that was deposited and that the sliding created a mechanically mixed layer (MML) on the surface of the wear track. A tribofilm of silver clusters and surrounding Ta_2O_5 were observed on the surface suggesting the decomposition of $AgTaO_3$ into nanocrystalline Ag and Ta_2O_5. These features were reproduced in molecular dynamics simulations [16]. It was determined that the reduced friction coefficient at high temperatures was due to the existence of the MML near the surface, with the potential for continued re-construction of the Ag and Ta_2O_5 tribofilm from silver tantalate through the wear process.

The objective of the current research is to use experimental measurements and molecular dynamics simulations to investigate load dependency on the tribological properties of sputter-deposited $AgTaO_3$ coatings. Specifically, the elemental and phase composition and the crystal structure in the wear track are investigated to understand the changes that occur in these coatings as a result of the application of various loads during the sliding process. This study significantly extends

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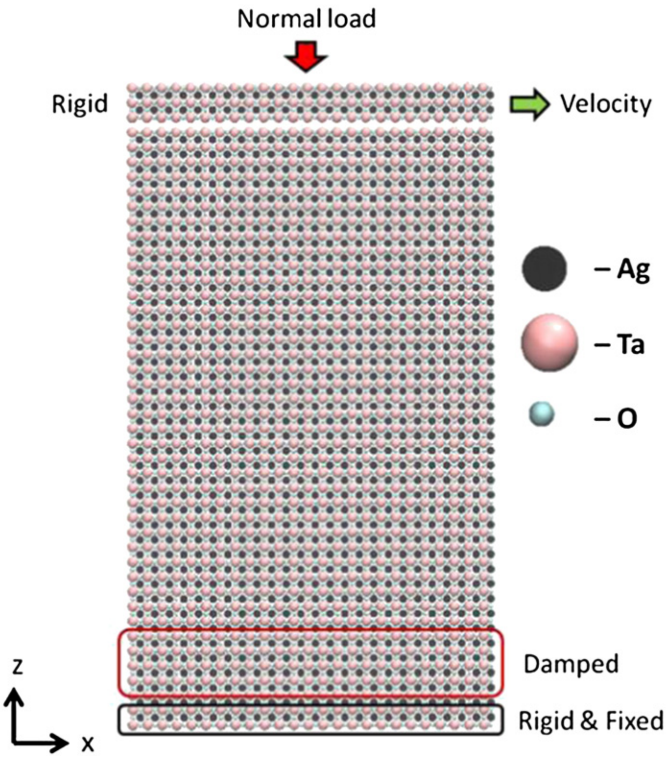

Fig. 1. Snapshot of $AgTaO_3$ tribofilm setup.

previous work on a promising high temperature solid lubricant by characterizing and providing mechanistic insight into its behavior under different operating conditions.

2. Material and methods

Silver tantalate coatings were deposited on Inconel 718 substrates having been mechanically polished to a mirror finish. The substrates were cleaned ultrasonically in acetone and methanol for 15 min, rinsed in deionized water, and dried using compressed nitrogen prior to insertion into an ATC 1500 unbalanced magnetron sputtering chamber (AJA international, North Scituate, MA). Elemental targets (Ø = 5 cm) of Ta (99.98%), and Ag (99.997% purity) were acquired from Plasmaterials, Inc. (Livermore, CA) and used as the source material. After the system was evacuated to a base pressure below 10^{-4} Pa, growth of the thin films was executed in a mixed atmosphere of Ar (99.999%) and O_2 (99.99%) with partial pressures of 0.67 and 0.16 Pa, respectively. Substrates were held at a temperature of 550 °C (to facilitate formation of tantalate phases) and a bias potential of − 120 V while being rotated about their polar axis at a speed of 50 rpm. A thin Ta layer was deposited for the initial 5 min to enhance adhesion of the coating to the substrate. Thin films of $AgTaO_3$/Ag were then deposited for 3 h, which corresponds to a total thickness of 2 μm.

XRD patterns before and after tribotesting were acquired using a GBC MMA diffractometer equipped with a Cu Kα radiation source to evaluate the structural properties of the films. Wear testing of these films was carried out under a load of 1, 2, 5, and 10 N using a Nanovea ball on disk tribotester (Microphotonics Inc., Irving, CA) at 750 °C (heating stage temperature) in atmospheric conditions (relative humidity of 60 ± 5%) with a sliding speed of 0.11 m/s and a total sliding distance of 10 m. The tests were performed at 3.33 Hz using a 6 mm diameter Si_3N_4 ball, since it is the material of choice in high temperature hybrid bearing assemblies. Under these conditions, the corresponding initial mean Hertzian contact stresses were 0.5, 0.6, 0.8, and 1.0 GPa, respectively. Three tests were performed for each load. The elemental composition for as-deposited and after tribotesting inside and outside the wear track was determined using a Model 670xi Scanning Auger Nanoprobe (SAN). The composition of the as-deposited film was found to be 22 at.% Ag, 17 at.% Ta, and 61 at.% O. Also, phase identification inside and outside of the wear tracks was evaluated using a Thermo Scientific Nicolet Almega XR Raman spectrometer with 532 nm laser excitation. In addition, site selective cross-sectional transmission electron microscopy (TEM) studies inside wear tracks were performed with high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM), bright field TEM (BFTEM), energy dispersive X-ray spectroscopy (EDS), and selected area electron diffraction (SAED) using an FEI Tecnai G2 F20 S-twin TEM operated at 200 keV. Finally, site selective specimens compatible with 3D Atom Probe Tomography (APT) were analyzed using a Cameca LEAP 3000X-HR™, operating at a specimen base temperature of 35 K and equipped with a 10 ps 532 nm pulsed laser with a pulse frequency of 160 kHz and a pulse energy of 0.2–0.3 nJ. Site specific TEM and APT specimens were prepared using a FEI Nova 200 NanoLab dual-beam focused ion beam (FIB)/scanning electron microscope (SEM).

3. Theory/calculation

To complement the experimental measurements, molecular dynamics (MD) simulations were carried out using the open source code LAMMPS. The simulation process was similar to our previous work [16] and can be simply described as a rigid plate sliding over a $AgTaO_3$ tribofilm. The top plate consisted of $AgTaO_3$ atoms that acted as a rigid body. This rigidity assumption means that the plate does not represent a real material and we found that model-predicted trends were unaffected by its atomic composition. The dimensions of the film were $9.7 \times 1.94 \times 15.52$ nm^3 and it contained 25,375 atoms. The top rigid plate moved laterally at a constant speed of 5 m/s for 8 ns with a

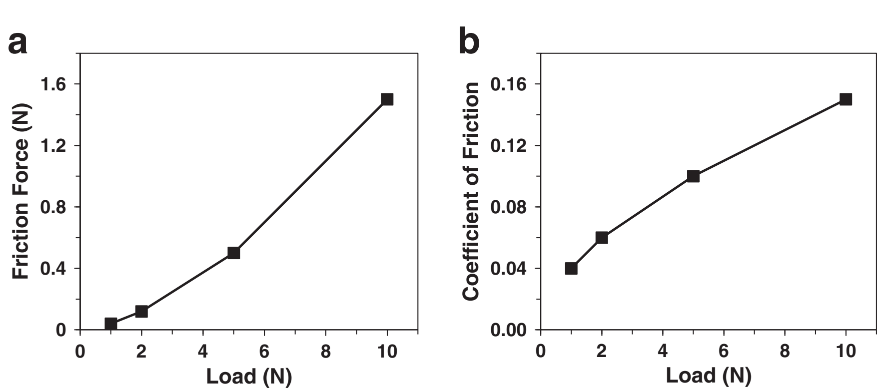

Fig. 2. (a) Friction force and (b) steady-state CoF for silver tantalate as a function of load tested at 750 °C.