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# **Tribological Characteristics of Sputtered Au/Cr Films on Alumina Substrates at Elevated Temperatures**

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Work performed for  
**U.S. DEPARTMENT OF ENERGY**  
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Prepared for the  
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ABSTRACT

This paper describes research to evaluate the tribological properties of alumina pins sliding against thin sputtered gold films deposited on alumina disk substrates. A 250 Å thick chromium interlayer was first deposited onto the alumina test disks to enhance adhesion and high temperature wetting of the gold films. The Au/Cr films were tribotested in pure sliding in a pin-on-disk tribometer under a 4.9 N load at 1 m/s. The test atmosphere was room air at temperatures of 25, 500, and 800 °C and the test duration varied from 60 to 540 min.

The use of the Au/Cr films reduced friction by about a factor of two compared to the unlubricated alumina sliding couple. The coatings prevented wear of the alumina substrate disks and reduced pin wear by one to two orders of magnitude. In addition, wear lives in excess of 200 000 sliding passes (9 hr) were observed during sliding at 800 °C. The results suggest that these films show promise for the practical lubrication of many high temperature sliding components.

INTRODUCTION

The development of mechanical components and devices that operate at ever increasing temperatures demands the concurrent development of material-lubricant combinations that can survive such extreme environments. High-temperature tribology has been called "the single greatest problem facing the adiabatic engine" where temperatures of 560 °C are expected at top ring reversal [1,2]. Even higher temperatures, up to 1000 °C, are expected in sliding engine and control surface seals for proposed



hypersonic vehicles [3]. Ceramic materials, because of their light weight, high temperature stability and low thermal conductivity, are promising candidates for the above and other applications. However, unlubricated ceramics present unacceptably high friction and wear rates [4,5].

Solid lubrication, in the form of thin, soft metallic films, such as gold or silver, appears to be an effective method for lubricating ceramics [6,7]. The beneficial value of thin soft films in lubricating hard substrates has long been documented [8]. Friction is reduced in the contact zone due to the low shear strength of the soft coating coupled with the high load carrying capability of the hard substrate [9]. The coatings must of course adhere to the substrate during sliding. Unfortunately, relatively inert metals such as Ag and Au typically adhere poorly to ceramic substrates [10].

The interfacial bond can be improved by applying the coatings with a high energy technique such as ion-beam assisted deposition (IBAD) [6,7,11]. Bare alumina pins sliding against alumina disks coated with IBAD Au and Ag films reduced friction and wear significantly compared to unlubricated disks in tests run at temperatures up to 400 °C. However, metallic films such as Ag, still dewet ceramic substrates at moderate temperatures (about 500 °C) [11,12].

Sputter deposition of Au and Ag coatings is another and less costly method of application, but debonding and dewetting at high temperature remains a problem. A more adherent sputtered film can be produced by introducing a thin bond layer of a more reactive metal between the inert Au or Ag coating and the ceramic substrate [12]. The "binder" or bond metal reacts with the substrate forming a tenacious interlayer that still presents a metallic surface to the nonreactive solid lubricant. The Au or Ag then wets and adheres to the bond metal. A recent paper by one of the author's shows that alumina disks sputter deposited with Ag and a Ti binder layer do not dewet after being subjected to heat treating at 850 °C as do disks sputtered only with Ag [12]. They also retain their good tribological properties when tested at room temperature after heat treating. More recent testing by the authors has shown that the Ag/Ti composite coating performs well at temperatures up to about 400 °C. At higher temperatures, coating delamination occurs resulting in high friction and wear and excessive transfer to the pin.

Au/Cr is another coating combination that has shown promise in other ceramic applications. As examples, chromium has been used in the electronics industry to bind gold contacts to ceramics and has

also been used in the brazing of ceramics [10,13,14]. For tribological purposes, gold's low shear strength and excellent thermal and chemical stability make it a potentially good solid lubricant over a wide temperature range. In the Au/Cr system, a thin layer of chromium would act as the interfacial bond between the ceramic substrate and the inert gold solid lubricating layer.

In the present paper, sputter deposited Au/Cr coatings on alumina substrates were tested in sliding against bare alumina pins at 25, 500, and 800 °C. Baseline comparative tests were run at the same temperatures with unlubricated alumina disks and also with disks coated only with Au, sliding against unlubricated alumina pins. During preliminary testing it was observed that heat treating the Au/Cr coated disks prior to testing significantly improved coating adhesion. Therefore, a 6 hr heat treatment at 725 °C was instituted. Following tribotesting, Scanning Electron Microscopy and Energy Dispersive Spectroscopy (SEM/EDS) analyses were conducted to assess the performance and utility of these coatings.

## EXPERIMENTAL

### Materials/Coatings

Disks of 99.4 percent pure alumina, with a nominal diameter of 6.35 cm and surface polished to approximately 0.4  $\mu\text{m}$  rms, were used in all of the tests. The pins, also of 99.4 percent  $\text{Al}_2\text{O}_3$ , were 0.953 cm diameter cylindrical rods with both ends finished to a 2.54 cm radius of curvature. Pin and disk property data and fabrication information can be found in a previous publication [12]. Targets of 99.999 percent Au and 99.95 percent Cr were used for sputter deposition of the solid lubricating films.

The pins were cleaned with the following five step process; Rinse with ethyl alcohol, polish with 0.3  $\mu\text{m}$  alumina powder, rinse with tap water, rinse with deionized water, and finally, blow dry with lab air. Prior to sputtering, the disks were ultrasonically cleaned for 15 min each in acetone then methanol, after which they were dried with nitrogen. The disks were backscatter etched for 5 min at 0.500 kW, 20 mtorr, with ionized argon atoms in a final cleaning process before deposition of the surface coatings.

A 250 Å layer of chromium was sputter deposited onto each alumina disk, followed by a 2  $\mu\text{m}$  layer of gold. Sputter times were; 25 sec at 0.500 kW, 8 mtorr for the chromium and 282 sec at 1.00 kW, 8 mtorr for the gold coating. The sputtering times necessary to achieve the desired thicknesses were determined

experimentally and verified using surface profilometry on a quartz standard. Figure 1 shows an SEM micrograph of an as deposited Au/Cr film.

Prior to tribotesting, the disks were annealed in air at 725 °C. Heat treating for 6 hr appears, based upon initial tribotest results, to form a more tenacious film. Various intermediate times and temperatures were tried before adopting this heat treatment which may not yet be optimal.

#### Apparatus

Pin on disk friction and wear tests were carried out in a high temperature tribotester. This equipment has been described fully in a previous publication [15]. Briefly; the pin and disk apparatus is enclosed in a resistance heated furnace capable of attaining and maintaining temperatures of up to 1200 °C. A dead weight system loads the pin on the disk with a force of 4.9 N. The disks are rotated at 370 rpm, resulting in a nominal linear velocity of 1 m/s. Prior to testing, the disk is carefully aligned to reduce the total indicated runout to less than 0.025 mm. The friction force and the load force are continuously recorded via a strip chart. In addition discrete data, including load, friction force, temperature, and speed, are sampled and stored every 30 sec by a computer acquisition system.

Wear tests were run at temperatures of 25, 500, and 800 °C. Tribotests were initially run for 30 min. Test length was increased to 60 min when it became apparent that the Au/Cr coating was maintaining its integrity for the duration of the shorter tests. Even longer tests, up to 9 hr in length, were run to establish the durability of the films. All of the testing was done in atmospheric air with a relative humidity of 50 to 75 percent at room temperature.

A SEM/EDS was used to image the test specimens and conduct elemental analyses of the disks before and after wear testing. The  $\text{Al}_2\text{O}_3$  pins were examined only after testing because they had to be coated with a conductive film in order to prevent "charging" in the SEM. Pin wear volume was calculated using an optical microscope to measure wear scar diameter. Disk wear volume was determined using a surface profilometer. Wear factors ( $k$ ), in  $\text{mm}^3/\text{N}\cdot\text{m}$ , were then calculated by dividing the wear volume by the product of the load and sliding distance.



## RESULTS AND DISCUSSION

### Tribotesting

Friction.—Friction coefficients for the tests conducted, averaged over the first 60 min of testing, are given in Table I. The friction for the Au-Cr coated disks was approximately 50 to 60 percent less than on unlubricated disks at all tested temperatures. Friction coefficient versus time is plotted for the Au-Cr coated disks and uncoated alumina disks at each tested temperature in Fig. 2. One possible reason the friction is lower at elevated temperatures is that the shear strength of the gold film is reduced as temperature is increased. This reason does not explain why the friction at 500 °C is slightly lower than at 800 °C. However, since the same trends are observed for the unlubricated sliding case the alumina surface may also have an effect on friction.

Although several long term durability tests (up to 9 hr or 32.4 Km) were run on the Au-Cr disks only the initial 60 min of testing is shown. The longer tests merely exhibited a continuation of the same frictional behavior. For example; 60 min into one of the long duration tests at 800 °C, the friction coefficient was 0.32, after 9 hr of sliding, the friction coefficient had gradually increased to only 0.35. Therefore, the additional data points associated with the longer tests were omitted from the plot for clarity.

Because of the preliminary nature of this work and the long (greater than 60 min) life of these films, it is difficult to assign a definite film wear life. Only a limited number of long term durability tests have been performed. At 800 °C, three separate tests ran without failure for 3, 5, and 9 hr. Another endurance test at the same temperature failed after approximately 6 hr. Coating failure was deduced by rising and fluctuating friction. At 500 °C, coating failure began after 5 hr of sliding while a disk tested at 25 °C continued to perform for 9 hr. Based upon these limited tests under these conditions, the useful coating life is estimated to be between 5 and 10 hr of sliding (18 to 36 km)

Wear.—Wear factors for pins tested against uncoated disks and disks sputter coated with Au and Au/Cr films are summarized in Table I and plotted in Fig. 3. Pins run on Au/Cr coated disks exhibited substantially less wear at all temperatures than pins tested on unlubricated disks. At 800 °C pin wear is 30 times lower on heat treated Au/Cr coated disks than on unlubricated disks.



At room temperature, Au films with no Cr interfacial layer also produced substantial reductions in pin wear; approximately 23 times less wear than that of pins tested on uncoated disks. However, when tested at 800 °C, these simple Au coatings failed by complete delamination in the wear track. Excessive coating transfer from disk to pin precluded accurate and meaningful wear measurements for these tests.

Disk wear for the coated specimens is more difficult to quantify, and possibly less meaningful. Post tribotest surface profilometry indicates, that the maximum wear depth is  $\approx 2 \mu\text{m}$  which corresponds to the thickness of the gold lubricating layer. Thus for the coated disks, the wear volume consists primarily of the Au/Cr films. Therefore, direct comparison of the lubricated and unlubricated disk wear data cannot be made.

#### Surface Analysis

SEM and EDS analyses of the wear specimens helps elucidate some potential reasons for the long (up to 36 Km), high temperature wear lives of the Au/Cr films. Figure 4 shows a photomicrograph of a pin wear scar after sliding for 3 hr against a Au/Cr coated  $\text{Al}_2\text{O}_3$  disk at 800 °C without failure. A thin transfer film of gold and chromium (as determined with EDS) is observed on the wear surface. Figure 5 shows photomicrographs of the disk wear track from this test. Figure 6 shows the corresponding EDS analysis of the features observed in the wear track. Higher magnifications reveal small ( $<1 \mu\text{m}$ ) rounded patches of gold which have apparently been removed from adjacent larger regions of the coating. These small gold patches are deposited on the chromium layer which endures even after high temperature testing.

No loose debris was observed on or near the disk wear track or pin wear scar further suggesting that the gold that is removed from the large coating areas is redeposited elsewhere on the wear surfaces and available for continued lubrication. Considering the relatively low thickness of these films,  $\approx 2 \mu\text{m}$ , their unexpectedly long wear lives also provide additional support for this type of redeposition lubrication mechanism.

Prior to testing and heating, no chromium is discernible during EDS analysis of the Au-Cr sputter coated disks. This is to be expected since the penetration depth of the electron beam is only about  $1 \mu\text{m}$  while the gold layer is  $2 \mu\text{m}$  thick. After annealing for 6 hr at 725 °C, a clear chromium peak was

observed in the spectra of unworn areas of the disks. These effects suggest that the chromium is diffusing through the gold. EDS examination of alumina pins after sliding against Au/Cr coated disks also reveals distinct chromium transfer. Although the exact role the chromium plays in film adhesion and wear life is not known, its presence may be enhancing the performance of the gold-alumina sliding contact system.

#### CONCLUDING REMARKS

The results presented in this paper indicate that the sputtered Au/Cr films effectively lubricate the alumina specimens over a wide temperature range (25 to 800 °C). Compared to unlubricated alumina sliding couples, both friction and pin wear were substantially reduced at all temperatures tested. Although sputtered gold films without a chromium interfacial bond layer performed adequately at room temperature, coating delamination occurred during sliding at elevated temperatures suggesting that chromium can enhance the adherence and performance of gold lubricant films. Based upon these results, sputtered Au/Cr films may be appropriate and effective lubricants for advanced high temperature sliding applications.

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TABLE I.—FRICTION AND WEAR SUMMARY

[Test conditions: 4.9 N load, 1 m/s sliding velocity, air atmosphere, 60 min test.]

Disk specimen	Friction coefficient			Pin wear factor, $\text{mm}^3/\text{N}\cdot\text{m}\cdot 10^{-7}$		
	25 °C	500 °C	800 °C	25 °C	500 °C	800 °C
Unlubricated	$0.85\pm.03$	$0.69\pm.05$	$0.76\pm.02$	$23.2\pm.7$	$140\pm 15$	$45\pm 5$
Au coated	$0.24\pm.05$	(a)	(b)	$1.0\pm.2$	(a)	(c)
Au/Cr coated	$0.40\pm.05$	$0.30\pm.03$	$0.34\pm.02$	$0.60\pm.20$	$0.16\pm.06$	$1.40\pm.40$

## Notes:

-Uncertainties represent one standard deviation of the data. At least six repeat tests were run for each data point given.

<sup>a</sup>Test not run.

<sup>b</sup>Friction was 0.35 until coating delaminated prior to end of 60 min test period.

<sup>c</sup>Immeasurable due to excessive coating transfer from disk to pin.

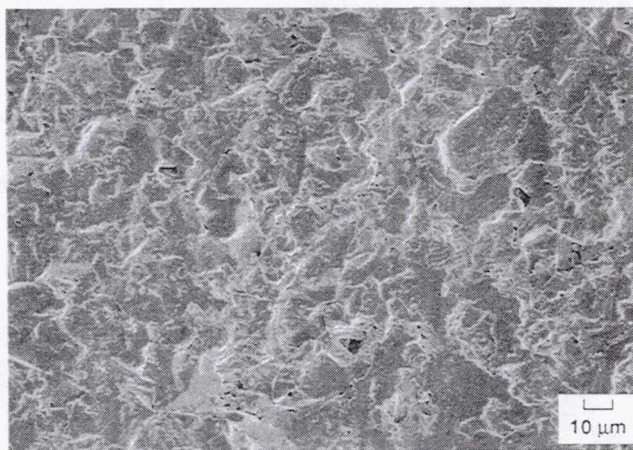


Figure 1.—SEM photomicrograph of an as-deposited Au/Cr coating on an  $\text{Al}_2\text{O}_3$  substrate.



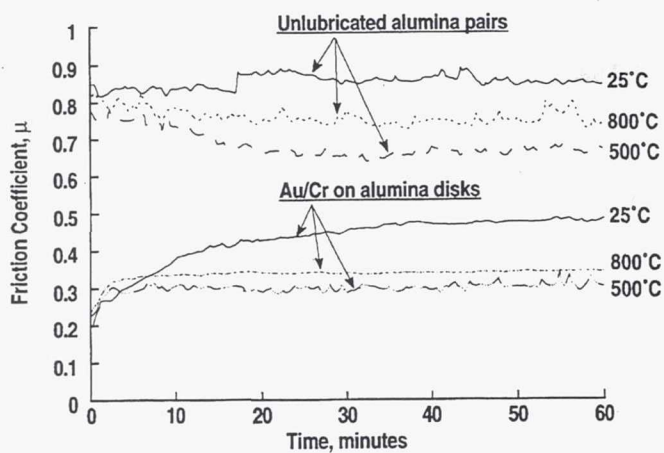


Figure 2.—Friction coefficient vs. time for unlubricated and Au/Cr lubricated alumina specimens. 4.9N load, 1m/s sliding velocity, air atmosphere.

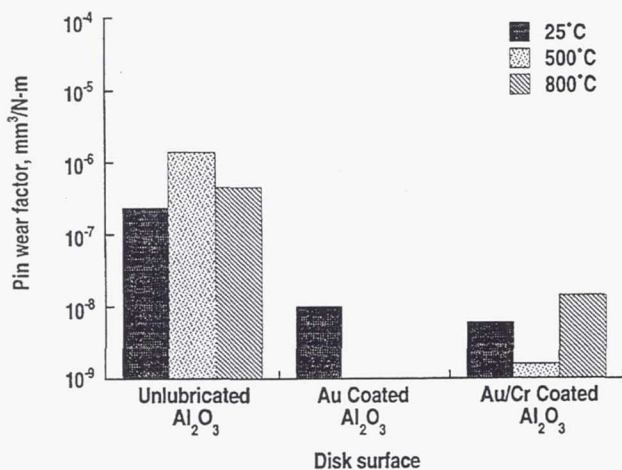


Figure 3.—Pin wear factors (k) for  $\text{Al}_2\text{O}_3$  pins sliding against various disk surfaces.

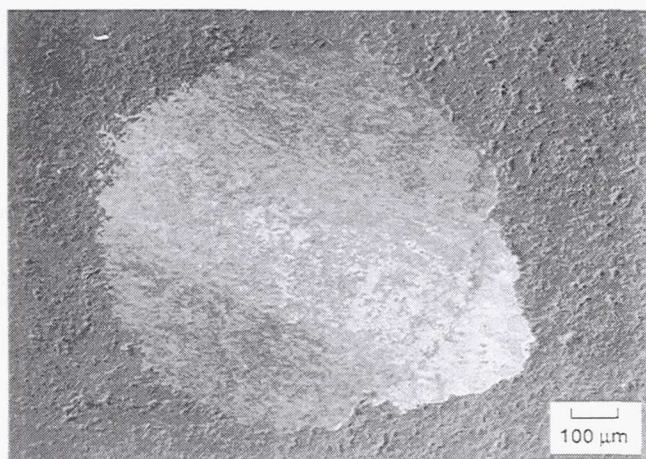


Figure 4.—SEM photomicrograph of pin wear scar after sliding against Au/Cr film at 800 °C.

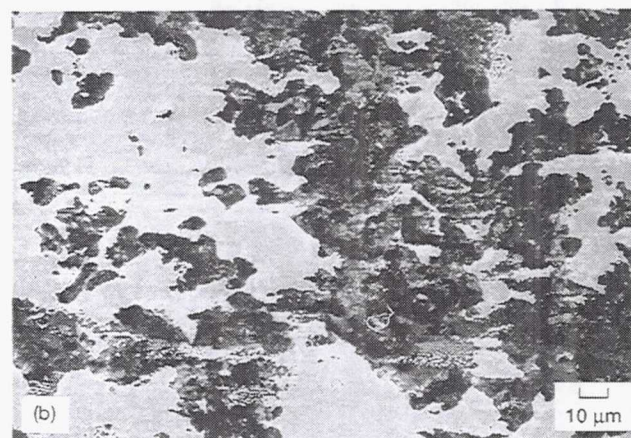
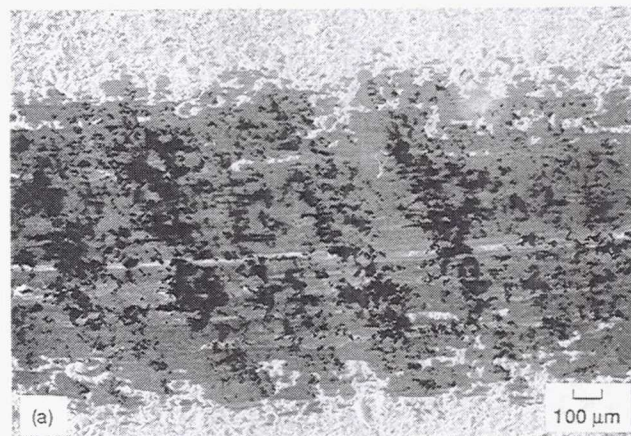
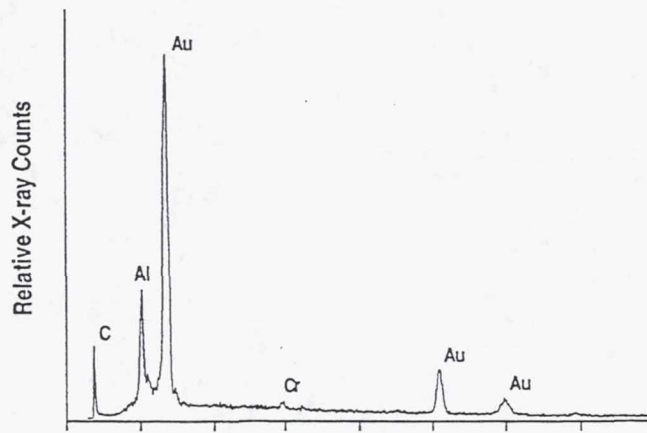
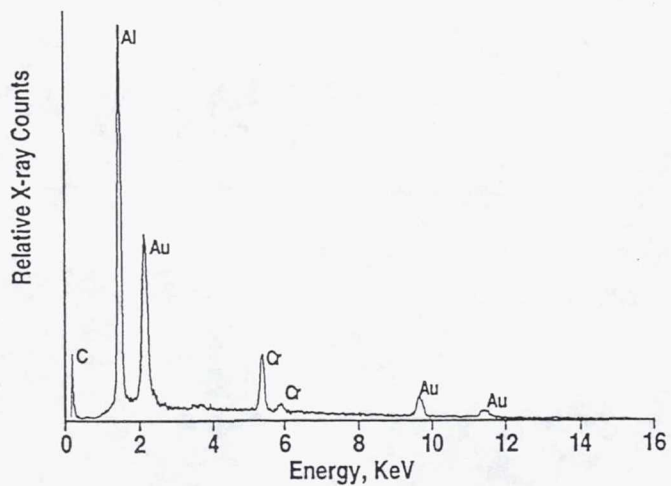


Figure 5.—SEM photomicrograph of disk wear track of Au/Cr coated  $\text{Al}_2\text{O}_3$  specimen after sliding at 800 °C. Au (bright regions) appear to migrate from larger regions as small patches. Higher magnification (c) shows migrating gold patches.



(a) Small gold patch.



(b) Surrounding darker area showing persistence of Cr layer.

Figure 6.—Corresponding EDS X-ray spectra.



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