

ANL/ET/CP--88594  
CONF-960629--2

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ENGINE APPLICATIONS\***

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February 1996

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For presentation at the 29th International Symposium on Automotive Technology and Automation, June 3-7, 1996, Florence, Italy

\*Work supported by the U.S. Department of Energy, Office of Transportation Technologies, under Contract W-31-109-Eng-38.

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## DEVELOPMENT OF LUBRICIOUS COATINGS FOR ADVANCED TURBINE ENGINE APPLICATIONS

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### ABSTRACT

Development of durable and low-cost high-temperature lubricants for use in gas-turbine regenerator cores is critically important for achieving improved performance and fuel - efficiency in future automotive and hybrid-electric vehicles. Successful development and implementation of such coatings could have substantial technological and economic impacts on energy conservation and could reduce dependence on imported oil. Furthermore, this technology may also contribute to a cleaner environment by reducing emissions.

Argonne National Laboratory is exploring new oxide-based lubricious coatings that can meet the stringent tribological conditions of advanced regenerator cores and seals. One of the key elements of this project involves the development of new ceramic and alloy coatings that will reduce friction and wear at temperatures up to 2000°F. This paper will highlight our recent research in high-temperature lubrication and in the development of new and improved carbon-carbon and carbon-polyimide based composites that can be used at temperatures up to 1100°F.

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## INTRODUCTION

Development of high-performance gas turbine engines is a practical approach for achieving higher fuel efficiency and lower emissions in future automobiles and hybrid vehicles. A major obstacle in this endeavor is the design and development of reliable and low-cost regenerator cores and seals that can safely operate at temperatures to 2000°F. A properly designed regenerator core and seal assembly can increase the fuel efficiency of turbine engines by as much as 30%.

Current regenerator cores are made of tape-wrapped metallic or ceramic foils. The cores serve as heat exchangers, transferring heat from hot exhaust gases to incoming air that is burned during combustion. Figure 1 shows schematic diagram of the "hot" and "cold" sides of a regenerator core with seals attached. In actual operation, hot exhaust gases pass through the narrow openings in the cores and transfer heat to the ceramic matrix before leaving the system. As the heated core segment rotates to the inlet section, it transfers heat / energy to incoming air. When the incoming air exits the hot side of the core, its temperature has increased to 1650°F. The cores are approximately 1 ft in diameter and rotate at speeds of <100 rpm.

The regenerator seals are either graphite (stabilized to reduce oxidation) in both low- and high-temperature regions, or a plasma-sprayed NiO/CaF<sub>2</sub> coating in very-high-temperature regions. The graphite seals are limited to 1150°F and the NiO/CaF<sub>2</sub> to 1800°F. At these temperatures, the seals are limited by oxidation, high wear, and friction, as well as by chemical reactivity with the core and plasma-bond-coating materials. Previous work with various NiO/CaF<sub>2</sub> coating compositions at temperatures of 1600 to 1800°F demonstrated that the desired friction and wear properties of the NiO/CaF<sub>2</sub> compositions considered will not meet the design criteria of the advanced core-seal assembly.

In an effort to improve the performance, reliability, and durability of the seals, we have initiated a research program with Allison Engine Co. to explore durable, low-cost oxide-based lubricious coatings that can meet the stringent tribological conditions of advanced regenerator cores and seals. Furthermore, the project will explore new alloys and coating compositions that can promote

the in-situ formation of wear-resistant and lubricious oxides, thus eliminating the use of time-consuming and expensive plasma-spray coatings.

In this paper, we present the results of a series of preliminary tests performed in our laboratory using both traditional and new lubricious coatings. High-temperature tribological properties of a series of graphite, carbon-carbon, and polyimide-base materials were also examined and are reported here.

## EXPERIMENTAL PROCEDURES

### Test Materials:

Inconel 625, high-temperature superalloy, was chosen as the pin material in our study. Pin diameter was 1 in. (25.4 mm) and the roughness of their flat ends was 0.3 to 0.5  $\mu\text{m}$  centerline average (CLA). The plasma spray coatings evaluated in this study included an NiO/CaF<sub>2</sub> and an I112 composition made of ZnO, SnO, and CaF<sub>2</sub>. Both coatings were applied over an NiCrAlY bond -coat. Table 1 lists the major components of each coating.

The alloy coatings evaluated in this study consisted of a number of transition metals with some boron and carbon additions. Selection of particular elements was based on previous studies [1,2] that suggested that they can form in-situ wear-resistant and lubricious oxides during sliding at high temperatures. These coatings were applied on Inconel 625 pins by a high-velocity oxyfuel and plasma-transferred arc method. Table 1 gives the details of alloy coatings evaluated in this study.

Honeycombed disks were used as the counterfaces in friction and wear tests. They were fabricated from tape-wrapped lithium-aluminum-silicate (LAS) and extruded magnesium-aluminum-silicate (MAS) materials. The cell densities of the LAS and MAS disks were 1400 and 1100/in.<sup>2</sup>, respectively. Figure 2 is a low-magnification scanning electron microscopy image of an

MAS disk. The honeycombed disks were 1 to 3 in (25.4 to 76.2 mm) thick and 6 in. (152.4 mm) in diameter. The sliding contact surfaces of all disks were ground sequentially by first 100-grit and then 320-grit diamond grinding wheels to ensure flatness and consistent surface finish.

The test disks used against the stabilized graphite, carbon-carbon composites, and polyimide materials were made of MAS material. These carbon-base materials are intended for use in the cold side of regenerator cores. The MAS disks used in tests were 0.25 in. (6.35 mm) thick and 3 in. (76.2 mm) in diameter. In actual tests, the carbon-base materials were cut into 0.5-in.-diameter rods or disks and used as the test pins.

#### **Friction and Wear Test:**

The friction and wear testing of ceramic and alloy coatings and carbon-base materials was carried out with two high-temperature tribotest machines. The ceramic- and alloy-coated Inconel 625 samples were evaluated in a machine with a three pin-on-flat geometry, whereas the carbon-base materials were tested in a tribometer with a single-pin-on-disk geometry. Both tribometers were instrumented with high-precision load cells, temperature controllers, and high-capacity data acquisition and control systems. Tests were run in open air of 20 to 50% relative humidity, under loads of 19.2 lb (86 N) (for three pin-on-disk) and 3.8 lb (17.4 N) (for single-pin-on-disk). The test duration ranged from 6 to 16 hr for ceramic and alloy coatings, but was up to 175 hr for graphite and carbon-base materials. Frictional torque was monitored by a load cell and collected by the data acquisition system throughout the tests. Rotational velocity ranged from 20 to 40 rpm for tests evaluating ceramic and alloy coatings, but 212 rpm for carbon-base materials. To establish a baseline, few tests were run to assess the tribological performance and durability of plain Inconel 625 material. Wear of the Inconel 625 pins was rather insignificant during these screening tests. The wear of carbon pins varied substantially, depending on material type, and was assessed from the height reduction as monitored by a linear-voltage displacement transducer and a dial indicator.

## RESULTS AND DISCUSSION

### Microstructure:

Figure 3 is a cross-sectional SEM photomicrograph of a plasma-spray NiO/CaF<sub>2</sub> coating used in our test program. At the magnification shown, bonding between the NiO/CaF<sub>2</sub> coating and the Inconel 625 substrate appears to be good. EDAX analysis of the regions near the interface confirmed the presence of a bond layer consisting of Y, Al, and Cr. These coatings remained intact during sliding tests at temperatures up to 1800°F.

### Friction and Wear:

#### Ceramic and Metallic Coatings

Figure 4 shows the frictional performance of uncoated, NiO/CaF<sub>2</sub>-coated, and I112-coated Inconel 625 test specimens during sliding against the LAS disks. The friction coefficients of uncoated Inconel were 0.7 to 0.65 at temperatures to 1000°F, but increased to 0.95 during sliding at 1200 to 1600°F. At 1800°F, the friction coefficient decreases 0.8. These results suggest that bare Inconel 625 causes high friction and thus cannot be used for seal applications. The high-friction nature of this material can be attributed to its inability to form a lubricious oxide layer during sliding at elevated temperatures.

Compared to that of the bare Inconel 625, the frictional behavior of the NiO/CaF<sub>2</sub> coating appears to be somewhat better. Its friction coefficients fluctuated between 0.6 and 0.75 throughout the tests. As for the I112 coating, its frictional performance was much better, providing a friction coefficient of 0.42 at 800°F. At higher temperatures, the coefficient increased steadily, reaching 0.75 at 1600°F. At 1800°F, the coefficient decreased slightly to 0.66.

The results of these tests indicate that the frictional performance of Inconel 625 can be improved

by applying a lubricious coating such as I112. However, it is important to point out that friction is very much dependent on temperature and fluctuates substantially with increasing temperature. We believe that the compositions of these coatings must be further optimized, or new formulations developed, to achieve lower friction coefficients at high temperatures.

Figure 5 shows the results of friction tests with extruded MAS material. The frictional performance of Inconel 625 against this material was very poor. The friction coefficient at 800°F was quite high (0.8), and wear of the MAS disk was substantial. Therefore, we limited tests with bare Inconel 625 to 800°F.

The frictional performance of both the NiO/CaF<sub>2</sub> and I112 coatings against MAS appeared quite impressive. The friction coefficient of NiO/CaF<sub>2</sub> was 0.4 to 0.5 at temperatures up to 1400°F. The frictional performance of I112 was even better, with its coefficient ranging from 0.36 to 0.5 at up to 1400°F (see Fig. 5). However, at 1600°F, the friction coefficients of both coatings increased to 0.7 and 0.8, respectively.

These results demonstrate that the overall frictional performance of NiO/CaF<sub>2</sub> and I112 was much better against the MAS than the LAS material. This observation suggests that the type of core material makes a significant difference in frictional performance. Although MAS seems to be a better match in terms of friction, field tests have indicated that this material may lose its strength after long-duration use and rapid thermal cycling.

Figure 6 presents the results of friction tests with three alloy coatings; both the LAS and aluminum silicate (AS) disks were used. Coatings were applied on Inconel 625 by a plasma-transferred-arc method. Average friction coefficient of Alloy I was <sup>a</sup>0.5 during sliding at 800 to 1200°F, but increased to 0.6 and beyond as the temperature rose. The frictional performance of Alloys II and III was not as good as that of Alloy I; coefficients were relatively high and ranged from 0.6 to 0.73, depending on test temperature.

These results indicate that the friction coefficients of alloy coatings are higher than those of the



ceramic coatings. Apparently, the kinds of oxides forming at sliding surfaces are not especially lubricious or are reacting with the disk material to form adhesive bonds that translate into higher friction. Overall, it seems that in order to achieve low friction coefficients with alloy coatings, we must further optimize their compositions.

Because of the short test duration (i.e., 6 h), wear on both the disks and the coated and uncoated Inconel 625 pins was difficult to measure after sliding tests. We used a dial indicator and a digital micrometer to monitor the wear losses during and after the tests. However, the dial indicator showed mostly the height displacements due to thermal expansion while readings from the digital micrometer yielded no significant reduction in pin heights; this suggests that wear was minimal. In certain alloy coatings, we even observed a slight increase in pin height, which may have been due to oxidation.

### **Tests with Carbon-Base Materials**

The friction and wear performance of a series of graphite-base materials is shown in Fig. 7. The disks used in these tests were made of MAS material. Note that the bars in Fig. 7 show amount of wear, while the values above them give average friction coefficients. As is clear, the overall friction and wear performance of stabilized graphite appears to be the best. Compared to the other graphitic materials, it provided the lowest friction coefficients at each temperature and its wear losses remained very low even at 900°F. The friction and wear of both the high-density and the impregnated graphite increased markedly with increasing temperature.

The friction coefficients of various polyimide and carbon-carbon materials are shown in Fig. 8. These materials are intended for use in the cold rim sections of the regenerator cores. Polyimide I exhibited friction coefficients ranging from 0.1 at 400°F to 0.48 at 625°F. Its wear performance deteriorated with increasing temperature. The frictional performance of Polyimide II was essentially similar to that of the Polyimide I material, but its wear performance was slightly better. As shown in Fig. 8, at 400°F, it had a friction coefficient of 0.09; but at much higher temperatures, its friction coefficient increased substantially, reaching 0.53 at 625°F. Despite such

high friction, its wear losses remained relatively low.

The friction and wear performance of two carbon-carbon materials are shown in Fig. 8. The standard carbon-carbon material suffered significant wear during sliding at 400°F. Its friction coefficient was also relatively high (0.06 to 0.14); however, at and above 450°F, friction and wear performance was relatively good. The friction coefficient was 0.05 at 450°F and tended to decrease further as the temperature was increased. Average pin wear was also very low at elevated temperatures. Compared to the polyimides, this carbon-carbon material provided the best overall performance.

The frictional performance of inhibited carbon-carbon material was not as good as that of the standard carbon-carbon material (see Fig. 8). Its friction coefficient was substantially higher and erratic, often ranging from 0.06 to 0.2 during sliding at 400 and 450°F. At 500°F, it began to suffer high wear losses despite relatively low friction coefficients of 0.08 to 0.1. Because of high wear, we terminated this test at 500°F.

We also attempted to run a test with a fully graphitized carbon-carbon material. Its friction coefficient was the highest, i.e., 0.84 to start with, and remained relatively high (0.42) even after three hours of sliding at 400°F. Furthermore, it suffered one of the worse wear losses (0.079 in. in 3 hr). Because of high friction and severe wear, we did not pursue this test beyond 400°F.

## SUMMARY

The results of preliminary tests reported here indicate that the friction and wear of most ceramic and alloy coatings, as well as of the carbon-base materials, are very sensitive to temperature. Specifically, in most cases, increasing the test temperature resulted in high friction. Moreover, the wear of most of carbon-base, graphite, and polyimide materials increased with temperature. The frictional performance of base Inconel 625 was rather poor; it exhibited friction coefficients of 0.7 to 0.95 during sliding against LAS and MAS materials. With the use of lubricious NiO/CaF<sub>2</sub> and

I112 coatings at the sliding surfaces, frictional performance was markedly improved, especially at lower temperatures. The tests revealed that the I112 was more lubricious than the NiO/CaF<sub>2</sub> coating, especially when rubbed against MAS material. Its friction coefficients ranged from 0.36 to 0.5 against MAS at temperatures up to 1400°F.

The frictional performance of alloy coatings was better than that of the base Inconel, but was not as good as those of the NiO/CaF<sub>2</sub> and I112 coatings. Further improvements in their compositions are needed in order to achieve lower friction. In general, Alloy I appeared to provide better frictional performance than Alloy II and III.

The stabilized graphite afforded the best overall friction and wear performance of all the graphitic materials. The friction and wear performance of high-density or impregnated graphite was also reasonably good at lower test temperatures, but deteriorated as temperature was increased. The wear performance of Polyimide II was better than that of Polyimide I, but frictional performance was similar in both cases. The nongraphitized carbon-carbon exhibited the best overall friction and wear performance, whereas the inhibited carbon material suffered severe wear losses. The fully graphitized carbon-carbon material was the worse in terms of friction and wear.

#### ACKNOWLEDGMENT

This work was supported by the Office of Transportation Technologies, U.S. Department of Energy, under Contract W-31-109-Eng-38.

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1. M. B. Peterson, S. Z. Li, and S. F. Murray, "Wear-resisting oxide films for 900°C Applications," Final Report to Argonne National Laboratory, ANL/OTM/CR-5, 1994.
2. H. E. Sliney, "Solid Lubricants," Metals Handbook, Vol. 18, pp. 114-122.

Table 1. Major components of ceramic and alloy coatings.

<b>Coating</b>	<b>Major Ingredients</b>
NiO/CaF <sub>2</sub>	NiO and CaF <sub>2</sub>
I112	ZnO, SnO, CaF <sub>2</sub>
Alloy I	Ni, Fe, Cr, B
Alloy II	Cr, Mo, Ni, Si, Co
Alloy III	Cr, W, Co, C

## FIGURE CAPTIONS

Figure 1. Schematic illustration of (a) hot and (b) cold sides of regenerator core assembly.

Figure 2. Scanning electron photomicrograph of honeycombed MAS disk.

Figure 3. Scanning electron photomicrograph of cross-section of NiO/CaF<sub>2</sub>-coated Inconel 625 sample.

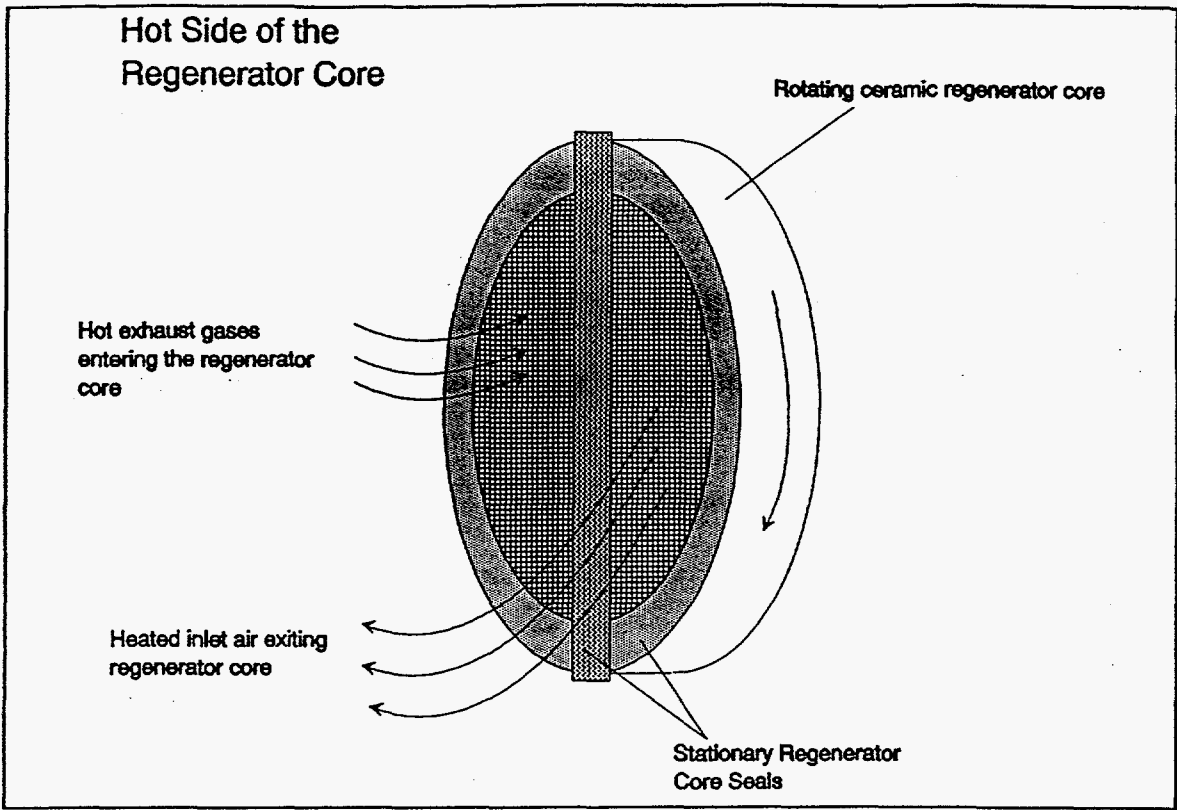
Figure 4. Variation of friction coefficients of Inconel 625, NiO/CaF<sub>2</sub>, and I112 coatings during sliding against LAS material as a function of increasing temperature.

Figure 5. Variation of friction coefficients of Inconel 625, NiO/CaF<sub>2</sub>, and I112 coatings during sliding against MAS material as a function of increasing temperature.

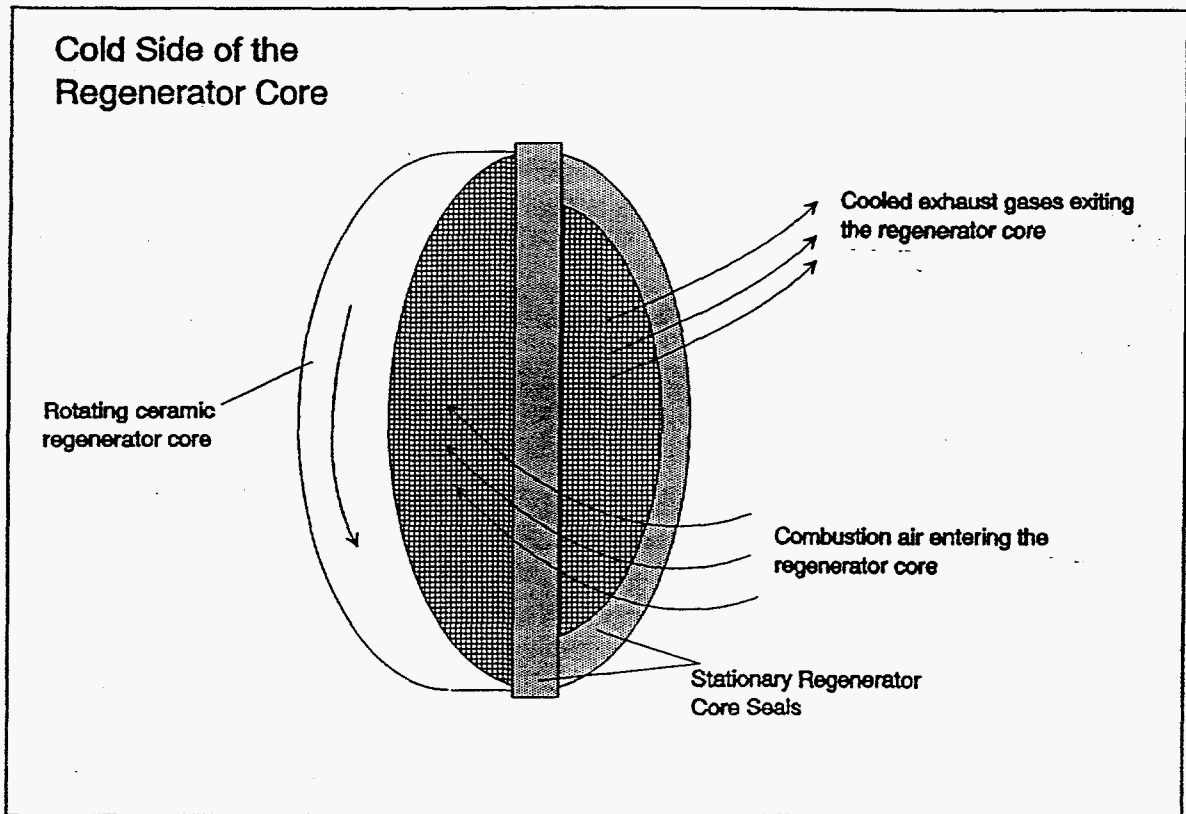
Figure 6. Variation of friction coefficients of various alloy coatings during sliding against LAS and AS materials as a function of increasing temperature.

Figure 7. Friction and wear performance of stabilized, high-density, and impregnated graphite against MAS material as a function of increasing temperature.

Figure 8. Friction and wear performance of polyimide and carbon-carbon-base materials against MAS material as a function of increasing temperature.



(a)



(b)  
Figure 1.

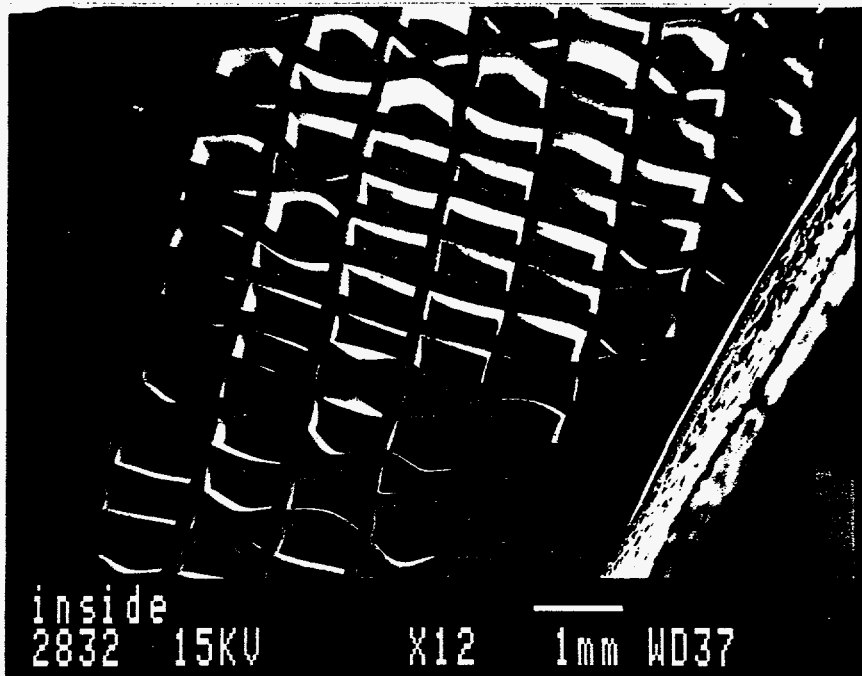


Figure 2.

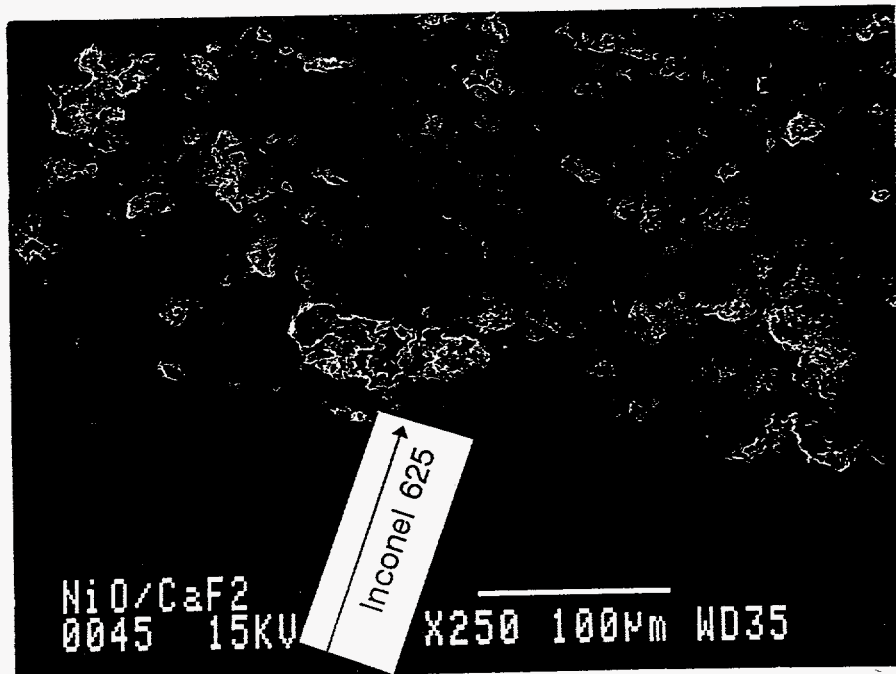


Figure 3.

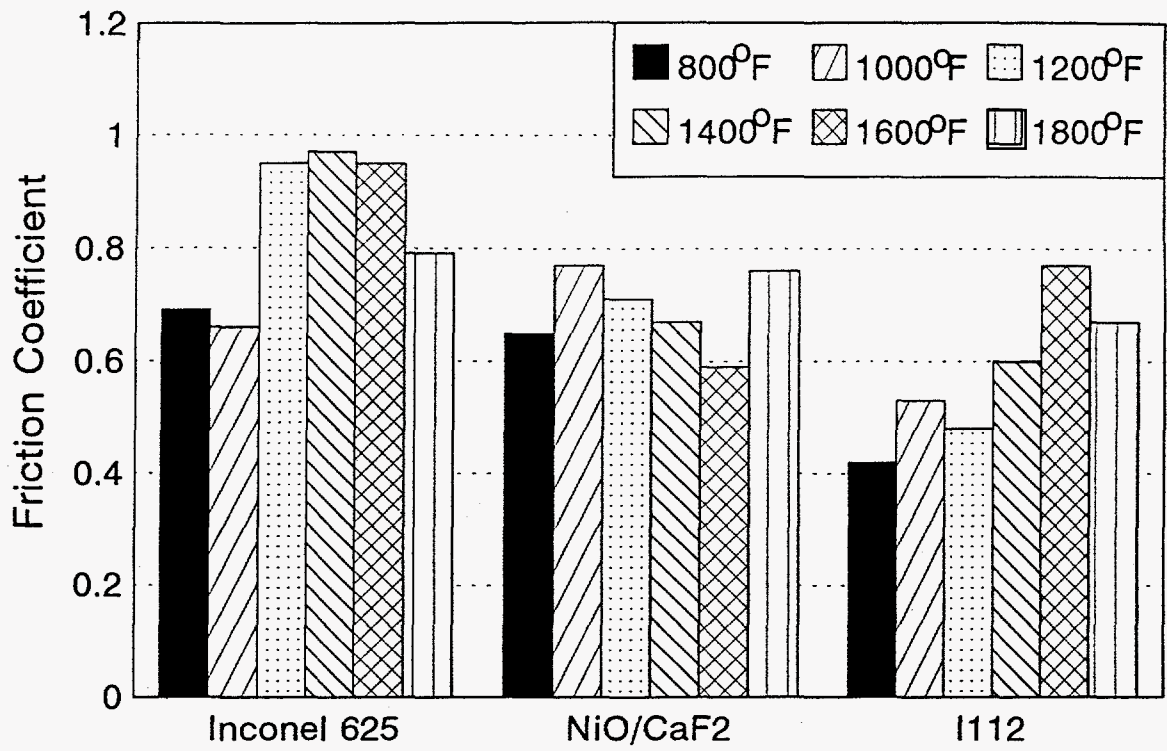


Figure 4.

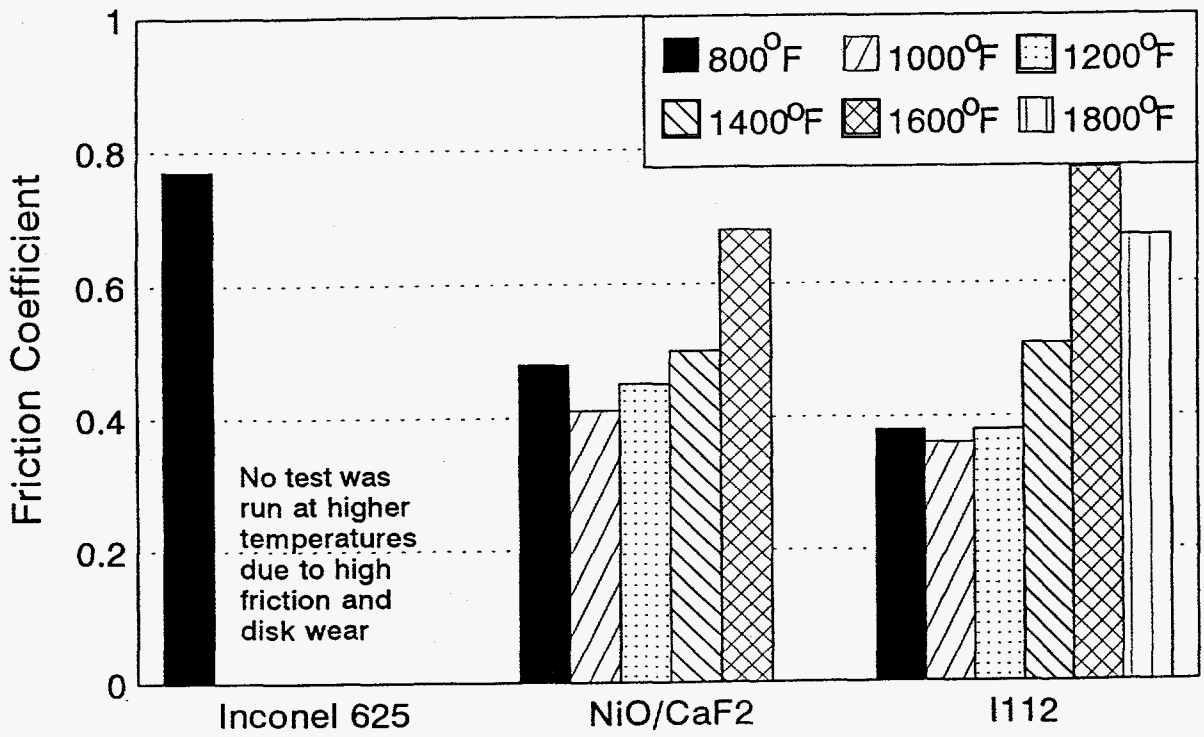


Figure 5.



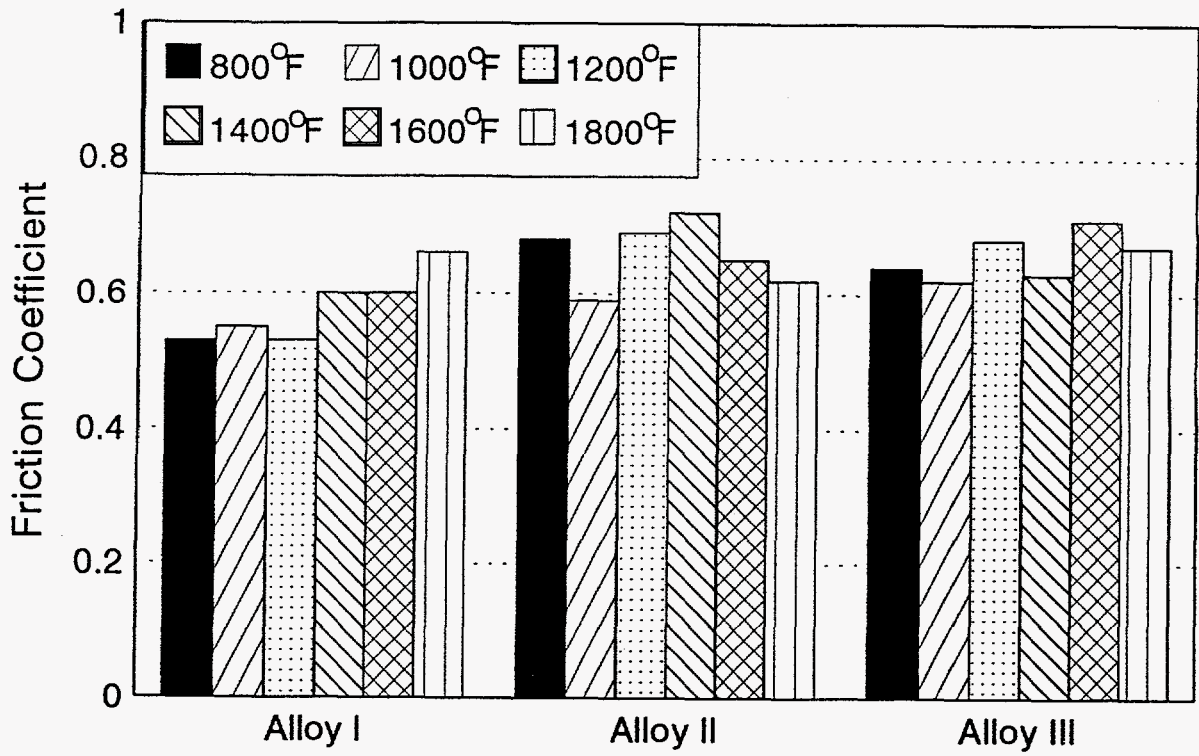


Figure 6.

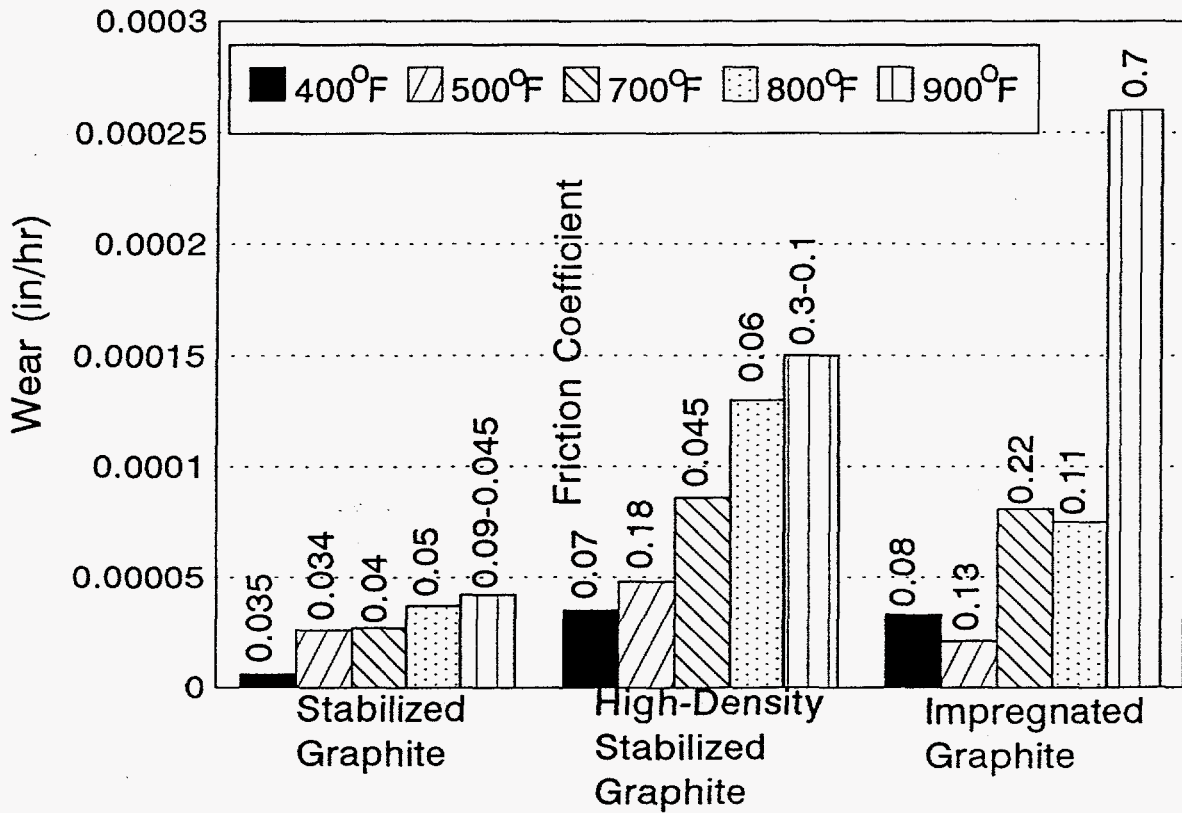


Figure 7.

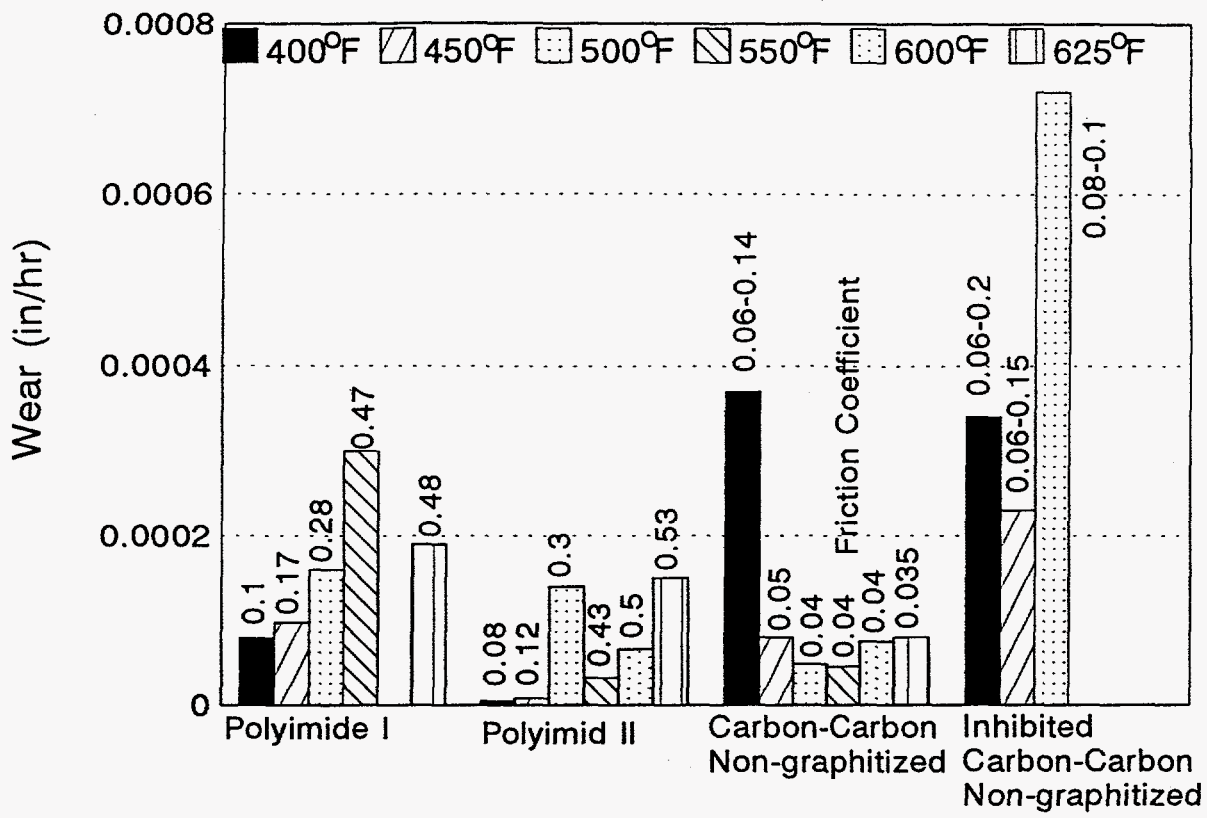


Figure 8.