

NASA TECHNICAL NOTE



NASA TN D-3261

NASA TN D-3261

LOAN COPY: R  
AFWL (WL)  
KIRTLAND AFB

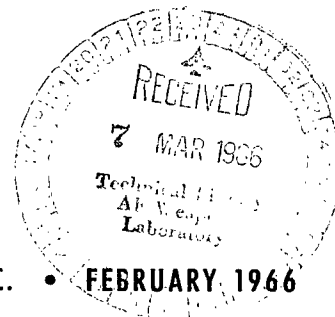


FRICITION AND WEAR CHARACTERISTICS  
OF POLYIMIDE AND FILLED POLYIMIDE  
COMPOSITIONS IN VACUUM ( $10^{-10}$  mm Hg)

*by Donald H. Buckley*

*Lewis Research Center*

*Cleveland, Ohio*



NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • FEBRUARY 1966



0079824

FRICITION AND WEAR CHARACTERISTICS OF POLYIMIDE AND FILLED  
POLYIMIDE COMPOSITIONS IN VACUUM ( $10^{-10}$  mm Hg)

By Donald H. Buckley

Lewis Research Center  
Cleveland, Ohio

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

---

For sale by the Clearinghouse for Federal Scientific and Technical Information  
Springfield, Virginia 22151 - Price \$1.00

# FRICION AND WEAR CHARACTERISTICS OF POLYIMIDE AND FILLED POLYIMIDE COMPOSITIONS IN VACUUM ( $10^{-10}$ mm Hg)

by Donald H. Buckley  
Lewis Research Center

## SUMMARY

The friction and wear characteristics of polyimide and filled polyimide compositions were examined in a vacuum environment ( $10^{-10}$  mm Hg). Copper-filled (to 50 weight percent) compositions as well as graphite- and molybdenum-disulfide-filled compositions were studied.

Friction and wear experiments were conducted with a 3/16-inch-radius hemispherical rider sliding on a flat disk, which was rotated to produce sliding speeds to 1600 feet per minute. The polyimide was loaded against the disk (various metals) under a load of 1000 grams.

The results of the investigation indicate that, to obtain effective lubrication (low friction and wear), a transfer of the polyimide film to the mating surface must be achieved. In its absence, high friction occurred with all polyimide materials. With copper addition, a minimum in wear rate was obtained at 30 weight percent copper. Both friction and wear increased with the addition of graphite as a filler to the polyimide. With the addition of molybdenum disulfide, marked decreases in both friction and wear occurred.

## INTRODUCTION

In many space applications, solid polymer compositions are very attractive for self-lubricated parts in mechanical systems. These polymers have an advantage over thin-solid-film lubricants in that, in many instances, they can be fabricated into components of the lubrication system and the problem of limited endurance life of thin films is overcome. Polytetrafluoroethylene (PTFE) compositions, for example, have been successfully used in vacuum (refs. 1 and 2).

A relatively new type solid polymer composition, polyimides, which appears to have the desired stability needed for vacuum applications has been developed (refs. 1 and 3 to 7). Further, it has shown some promise in bearing lubrication at high temperature in air (ref. 8).

The polyimide has better mechanical properties than PTFE (ref. 8). The desirable

characteristic of PTFE is its ability to transfer, under sufficient load, to the components to be lubricated. The ability to form a transfer film and to exhibit low friction and wear in a vacuum has yet to be demonstrated for the polyimide.

Limited previous friction and wear studies with the polyimide in vacuum indicate that it may be useful for vacuum and space lubrication systems. Further, these studies indicated that the presence of copper as a filler to the polyimide markedly improves its friction and wear characteristics (ref. 1). In order to gain a better understanding of the mechanism of lubrication with the polyimide, experiments on substrates other than those cited in reference 1 must be performed and the influence of fillers explored.

This investigation was therefore conducted to determine the vacuum lubricating characteristics of polyimide: (1) its ability to lubricate various kinds of materials, (2) its tendency to decompose at the sliding interface, and (3) the effect of various fillers on its friction and wear characteristics. Friction and wear experiments were conducted with a 3/16-inch-radius hemisphere sliding on a flat disk surface at various loads and speeds in a vacuum of  $10^{-10}$  millimeter of mercury.

## APPARATUS AND PROCEDURE

The apparatus used in this investigation is described in detail in reference 1 and is shown in figure 1. The basic elements of the apparatus were the specimens (a  $2\frac{1}{2}$ -in. - diam flat disk and a 3/16-in. -rad rider) mounted in a vacuum chamber. The disk spec-

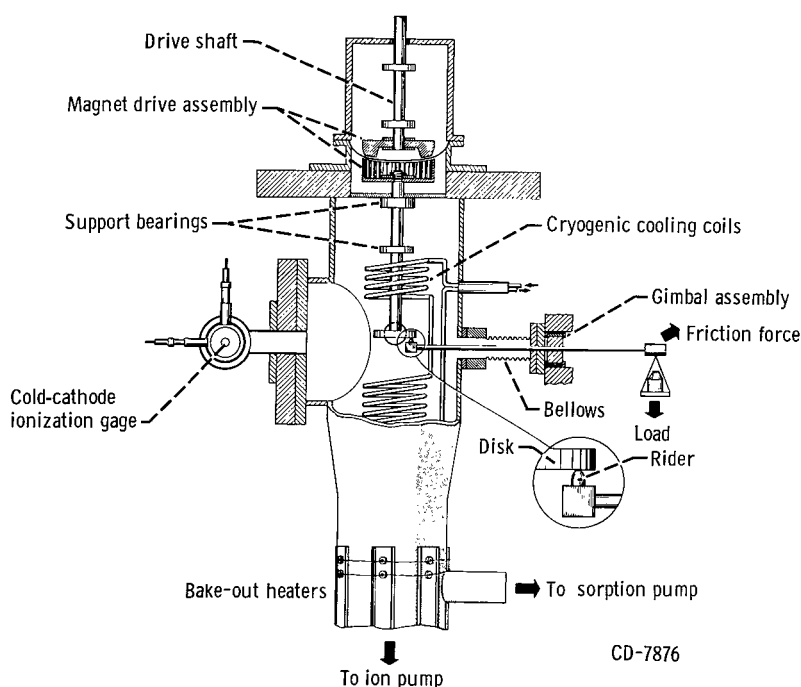


Figure 1. - High-vacuum friction and wear apparatus.

imen was driven by a magnetic drive coupling. The coupling had two 20-pole magnets 0.150 inch apart with a 0.030-inch diaphragm between magnet faces. The driver magnet that was outside the vacuum chamber was coupled to a hydraulic motor. This magnet was shrouded completely with a nickel-alloy housing (cutaway in fig. 1) and was mounted on one end of the shaft within the chamber. The other end of the shaft supported the disk specimen.

The rider specimen was supported in the specimen

chamber by an arm that was mounted from a gimbal and sealed to the chamber with a bellows. A linkage at the end of the restraining arm farthest from the rider specimen was connected to a strain-gage assembly that was used to measure frictional force. Load was applied through a deadweight loading system.

Attached to the lower end of the specimen chamber was a 400-liter-per-second ionization pump and a mechanical forepump with liquid-nitrogen cold traps. The pressure in the chamber was measured adjacent to the specimen with a cold-cathode ionization gage. In the same plane as the specimens and ionization gage was a diatron-type mass spectrometer (not shown in fig. 1) for determination of gases present in the vacuum system. A 20-foot, 3/16-inch-diameter stainless-steel coil was used for liquid-nitrogen and liquid-helium cryopumping of the vacuum system.

The polymer-composition specimens used in the friction and wear experiments were machined to size. The metal disks were circumferentially finish ground to a surface roughness of 4 to 8 microinches. Before each experiment the metal disks were given the same preparatory treatment: (1) a thorough rinsing with acetone to remove oil and grease, (2) a polishing with moist levigated alumina on a soft polishing cloth, and (3) a thorough rinsing with tap water followed by distilled water. The polymer specimens were rinsed with acetone followed by ethyl alcohol. For each experiment a new set of specimens was used.

## RESULTS AND DISCUSSION

The polyimide is made by the reaction of pyromellitic dianhydride and an aromatic diamine to form polyimide acid. The acid is then heated to remove water and the polyimide composition is thus formed (ref. 8). The molecular structure of polyimide is shown in figure 2. The presence of acid groups in the molecular structure could provide for surface reaction in contact with metals.

Data obtained in reference 1 with the polyimide sliding on itself and on 440-C stainless steel in vacuum indicated that the polyimide was worthy of further consideration for lubrication applications. For comparative purposes, the friction and wear properties of the polyimide are presented with those of PTFE (ref. 1) in figure 3. Examination of figure 3

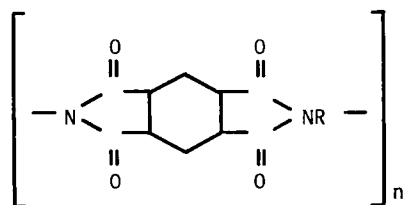


Figure 2. - Polyimide structure.

indicates that for the unfilled composition both friction and wear for the polyimide are less than were obtained for PTFE. This observation was also made for copper-filled polyimide and PTFE compositions (fig. 3). These data were sufficiently encouraging to warrant further studies with polyimide.

In order to gain a better understanding of the me-

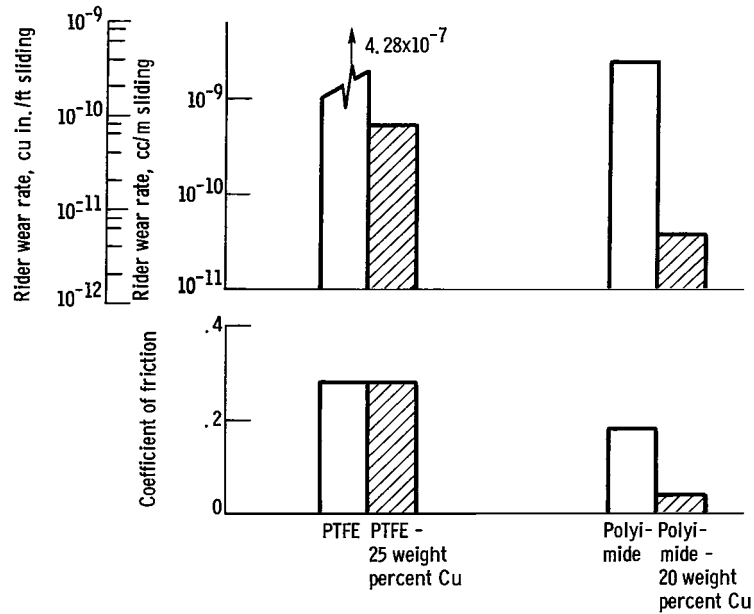


Figure 3. - Comparison of coefficient of friction and rider wear for polytetrafluoroethylene and polyimide compositions sliding on 440-C stainless steel in vacuum ( $10^{-10}$  mm Hg). Sliding velocity, 197 centimeters per second; load, 1000 grams; duration of run, 1 hour; no external specimen heating. (PTFE data from ref. 1).

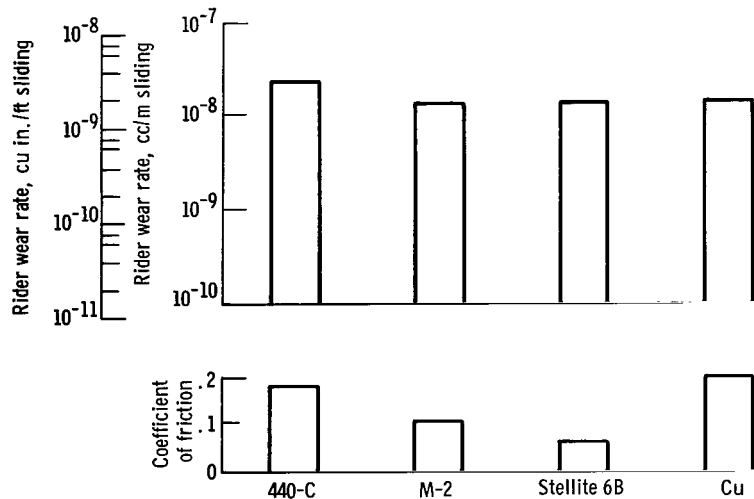


Figure 4. - Coefficient of friction and rider wear rate for polyimide sliding on various substrates in vacuum ( $10^{-10}$  mm Hg). Sliding velocity, 197 centimeters per second; load, 1000 grams; duration of run, 1 hour; no external specimen heating.

chanism by which polyimide might lubricate, some friction and wear experiments were conducted in vacuum with the polyimide sliding on various substrate materials. With different substrate materials it was hoped that the influence of the substrate surface, if any, could be determined. The results obtained in some of these experiments are presented in figure 4. Changing substrate material (fig. 4) appears to have very little influence upon the wear properties of the polyimide. The friction characteristics do, however, appear to be affected by the substrate. In all these experiments, an initial high coefficient of friction was obtained; however, it decreased very rapidly to the values reported in figure 4. The differences in friction data of figure 4 cannot be attributed to substrate hardness because the Stellite 6B, which exhibited the lowest friction coefficient, had a lower hardness than either the M-2 tool steel or the 440-C stainless steel. Further, the copper is considerably softer than 440-C stainless steel yet exhibited very nearly the same friction coefficient. Severe scoring of the copper by the polyimide was noted upon completion of the copper experiments although a transfer film of polyimide was present on the copper surface. The scoring may have developed during or prior to the formation of the transfer film.

The differences in friction properties for the polyimide sliding on the different substrates may be related to the type of film on the metal substrate. Since the experiments are conducted in vacuum ( $10^{-10}$  mm Hg), the influence of oxygen (to form metallic oxides) can be discounted. The reason for differences in friction would appear to lie in the continuity of the transfer film and the degree of contact (metal to polyimide) through the thin polymer transfer film.

It was mentioned earlier that the coefficients of friction for all substrates were initially high and then decreased with the formation of a transfer film. Friction results from one such experiment are presented in figure 5. At high sliding speeds, an initially high coefficient of friction, which decreased very rapidly to a very low value (fig. 5(a)) was observed. If, however, the sliding speed was decreased from 197 to 0.013 centimeter per second, a much more gradual decrease in friction was noted. The decrease in friction is believed to be due to the formation of a transfer film of polyimide on the metal substrate. In order to obtain acceptable friction coefficients with the polyimides, therefore, the formation of a transfer film seems imperative. Although an appreciable reduction in friction coefficient is observed with the formation of a thin transfer film to the metal substrate, it is reasonable to assume that only partial rider to thin polyimide film contact does occur. That is, the friction coefficients of figure 4 can be assumed to be the sum of two friction forces (between the polyimide and the transfer film and between the polyimide and the metal). Further, the ability of the various substrates to form a continuous surface film will differ, particularly if a reaction takes place between the polyimide and the metal substrate to form the thin surface film.

The importance of transfer films to the friction characteristics of materials in

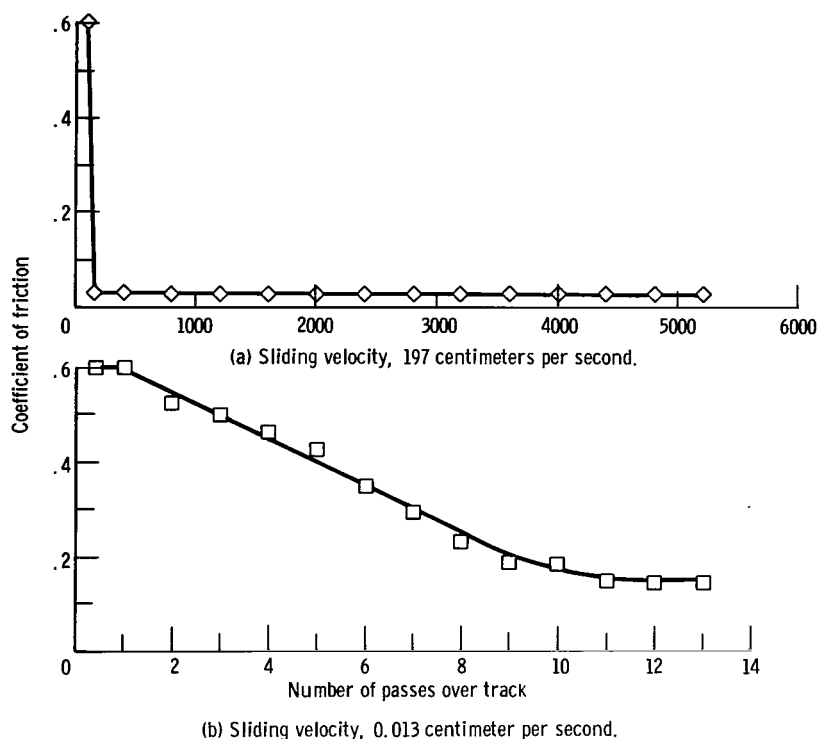


Figure 5. - Coefficient of friction for 20-weight-percent-copper-powder-filled polyimide sliding on 440-C stainless steel in vacuum ( $10^{-10}$  mm Hg). Load, 1000 grams; ambient temperature, 25° C.

sliding contact has been observed with materials other than polyimide. With mechanical carbons sliding on metal substrates, high friction coefficients were observed (ref. 9) in the absence of carbon transfer films, and the formation of these films was dependent on the thermodynamic stability of the substrate metal oxides. That is, the formation of a transfer film with carbon was a function of metal-surface chemistry.

In reference 1, the addition of 20 weight percent copper fiber to the polyimide appreciably reduced the friction and wear of the polyimide. This reduction was believed to be due to the presence of a thin film of the polyimide between the metal substrate and the copper fibers. Examination of the wear area indicated a high concentration of copper. Etching with acids revealed no reaction of the copper and indicated that a polyimide film was present over the copper seen on the wear scar. The copper served to conduct frictional heat away from the sliding interface. The dissipation of frictional heat from the sliding interface is extremely critical with polymeric materials because of their poor heat conduction characteristics. Thus, while wear for the unfilled polyimide may be due in part to a degradation (thermal) mechanism at the sliding interface, the addition of copper (a good heat conductor) reduces this effect.

In order to determine the optimum in copper-fiber concentration in polyimide for a minimum in friction and wear, experiments were conducted in vacuum with various per-



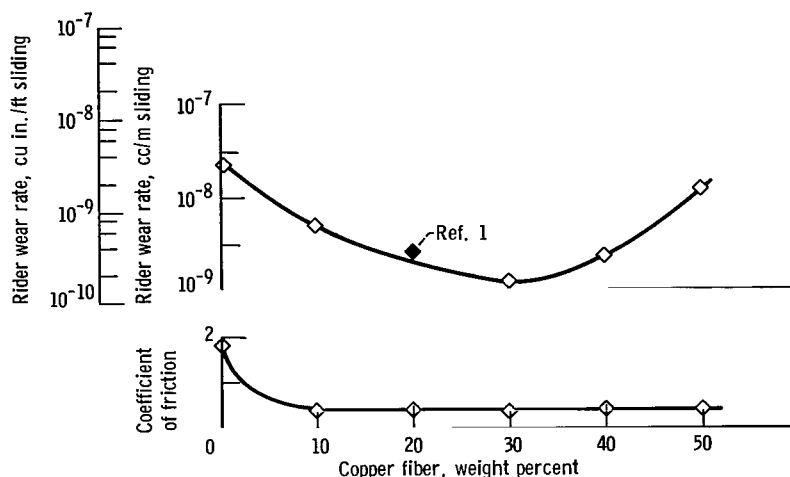


Figure 6. - Influence of copper-fiber filler in polyimide on friction and wear for polyimide sliding on 440-C stainless steel in vacuum ( $10^{-9}$  to  $10^{-10}$  mm Hg). Sliding velocity, 197 centimeters per second; load, 1000 grams; duration of run, 1 hour.

centages of copper fibers present in polyimide (fig. 6). With the addition of as little as 10 percent copper fiber to the polyimide, the friction coefficient decreased from 0.18 to 0.04. Further copper additions (beyond 10 percent) have little effect upon friction coefficient. Wear, however, was markedly influenced by the addition of various percentages of copper fiber with an optimum concentration appearing at about 30 weight percent copper fiber. The addition of 30 weight percent copper fiber to the polyimide reduced wear to one-tenth that obtained for the unfilled polyimide.

It is interesting to note that the wear rate for the polyimide decreased with increasing copper fiber to 30 weight percent. This reduction could be attributed to a decrease in thermal degradation of polyimide at the sliding interface. That is, frictional heat dissipation from the sliding interface by the copper fibers in the polyimide matrix and near the interface is optimum at 30 weight percent. Beyond 30 weight percent, the wear rate increases; this increase may be attributed to excess copper. Examination of the wear track on the stainless-steel disk after experiments with the 40- and 50- weight-percent-copper polyimides revealed the presence of metallic copper in the polyimide transfer film. Further, deposits of a carbonaceous debris were also noted. These deposits are evidence of thermal degradation of the polyimide at the sliding interface. The presence of copper in the transfer film would indicate that the polyimide rider is no longer contacting only a thin polyimide film but is also contacting some metallic copper. At higher copper concentrations (greater than 50 percent), an increase in friction coefficient could be anticipated to accompany the increasing trend in wear rate.

The metallic fibers in polymer materials are known to improve their mechanical strength. Based upon the wear mechanism proposed herein for the polyimide, the heat

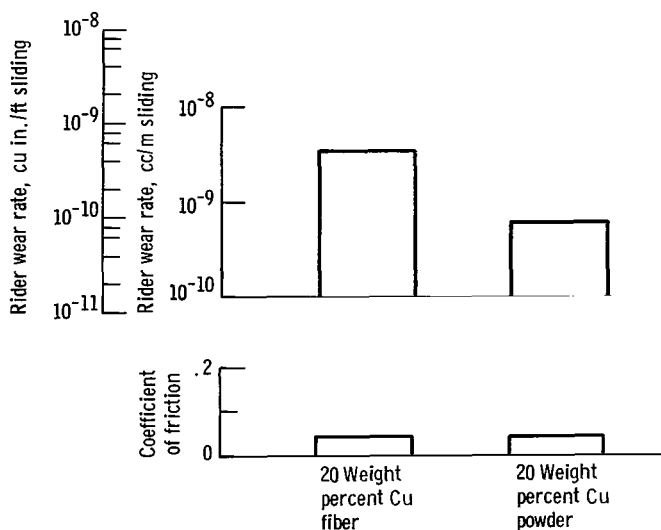


Figure 7. - Coefficient of friction and rider wear for copper-filled polyimide sliding on 440-C stainless steel in vacuum ( $10^{-10}$  mm Hg). Sliding velocity, 197 centimeters per second; load, 1000 grams; duration of run, 1 hour; no external specimen heating.

capacity of the metal is the important factor in the improvement. Therefore, the physical form of the copper should not be critical. Friction experiments were therefore conducted with a 20-weight-percent-copper-powder-filled polyimide to determine whether the physical form of the copper was critical. The results obtained together with that for the 20-weight-percent-copper fiber are presented in figure 7. The difference in the form of the copper did not affect the friction results; rider wear was somewhat reduced with the copper powder. The decrease in wear is believed to be due to a simple increase in surface area for equivalent quantities of copper. This theory was

substantiated by examination of the wear scar for the two copper-filled compositions. The amount of copper exposed surface area was greater for the powder-filled composition.

In addition to metallic fillers, various other materials can be added to solid polymers such as the polyimide to improve their friction and wear characteristics. The polyimides containing two solid lubricants, graphite and molybdenum disulfide, were examined in vacuum. The friction and wear results obtained are presented in figure 8. With the addition of 15 weight percent graphite, an increase in both friction and wear was observed. The friction coefficient for electrographitized carbon sliding on 440-C stainless steel in vacuum is about 0.4. The friction for graphite-filled polyimide is between that for polyimide and for graphitized carbon. It is well known that graphite will not lubricate in a vacuum environment, and the results obtained with the graphite-filled composition seem to confirm the graphite results.

Results obtained with the molybdenum-disulfide-filled polyimide are also presented in figure 8. The addition of molybdenum disulfide to the polyimide markedly reduced both the friction and the wear of the polyimide. In soft materials like PTFE, however, the addition of a solid lubricant such as molybdenum disulfide will not markedly affect friction and wear because the solid lubricant particles become isolated and coated with PTFE. With a brittle polymer such as the polyimide, this does not occur. Here the individual molybdenum disulfide particles are exposed in the wear process at the sliding interface between the steel substrate and the polyimide. The shear process appears not

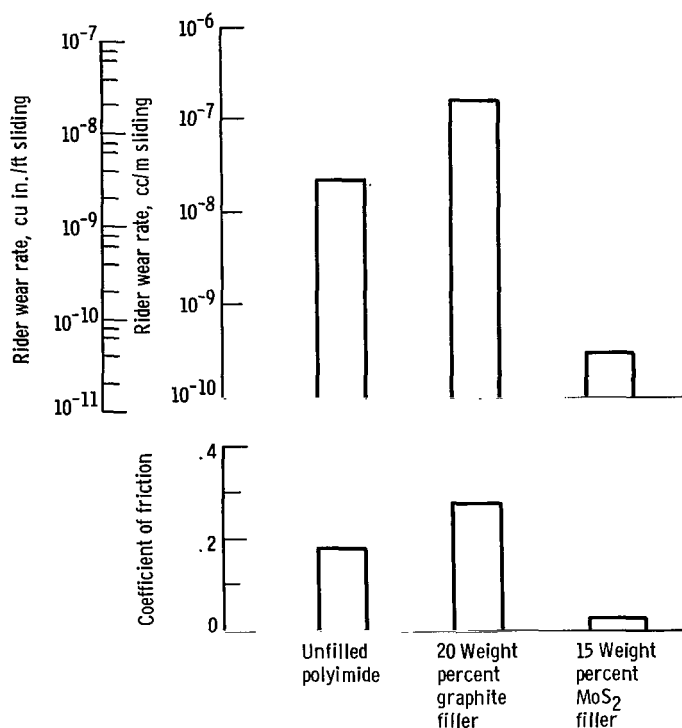


Figure 8. - Friction coefficient and rider wear for solid-lubricant-filled polyimide compositions sliding on 440-C stainless steel in vacuum ( $10^{-10}$  mm Hg). Sliding velocity, 197 centimeters per second; load, 1000 grams; duration of run, 1 hour; no external specimen heating.

to involve shearing of the polyimide but rather shearing between molybdenum disulfide lamellae. The friction coefficient is therefore representative of what would be experienced with molybdenum disulfide present on a surface as a solid-film lubricant.

A very important consideration in the use of polymers in a vacuum environment is their outgassing characteristics. The polyimides in this regard appear superior to materials such as PTFE. Figure 9 represents mass spectrometer data obtained for the polyimide sliding on 440-C. For the three higher mass-to-charge ratios  $M/e$  of 28, 30, and 50, essentially no increase in concentration was observed with increase in sliding velocity (interface temperature).

With a  $M/e$  of 28, however, a decrease in concentration did occur. This decrease may be attributed to possible surface reaction at the increased interface temperatures which result at higher sliding velocities. That is, the material is a component involved in the surface reaction. This is probably what was occurring since, on decreasing the sliding velocity, the concentration of  $M/e$  of 18 again increased. It should be emphasized that the concentrations of gaseous products for the polyimide are appreciably less than noted for PTFE in reference 1. These data indicate the vacuum stability of the polyimide.

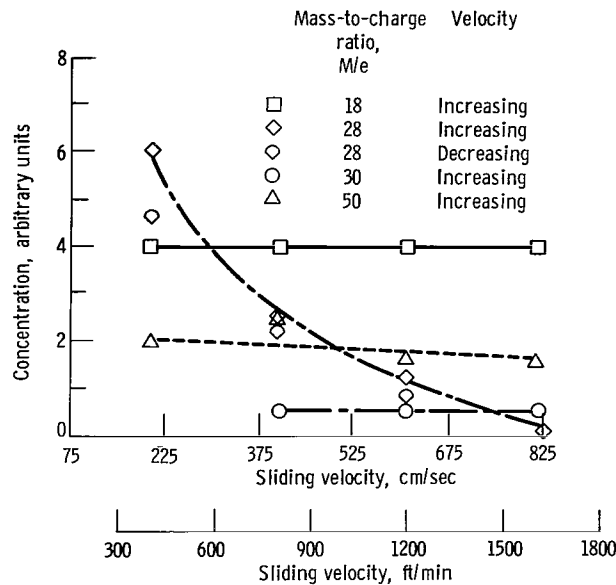


Figure 9. - Mass-to-charge ratios at various sliding velocities for polyimide sliding on 440-C stainless steel in vacuum ( $11^{-10}$  mm Hg). Load, 1000 grams; no external specimen heating.

## SUMMARY OF RESULTS

From an investigation of the friction and wear characteristics of polyimide compositions in vacuum ( $10^{-10}$  mm Hg), the following results were obtained:

1. For effective boundary lubrication with polyimide compositions (and polyimide with fillers) sliding on metal, a transfer film must form; that is, the polyimide must be sliding on a thin film of polyimide. In the absence of this film, high friction characteristics were observed for all polyimide compositions examined.
2. The addition of copper as a filler to polyimide reduced the friction and wear of polyimide appreciably. The addition of copper fiber (from 0 to 50 weight percent) to polyimide indicated an optimum in fiber concentration at 30 weight percent for a minimum in friction and wear for the conditions of this investigation.
3. The addition of molybdenum disulfide (15 weight percent) reduced the friction and wear characteristics of the polyimide.
4. The polyimide exhibited superior friction and wear characteristics to polytetrafluoroethylene in vacuum, once a transfer film of the polyimide to the mating surface was achieved.
5. Although some differences in friction were observed for polyimide sliding on various metal substrates, the nature of the metal substrates had very little effect upon wear.

6. With changing sliding velocities (interface temperatures), no marked evidence for polymer decomposition was observed by mass spectrometry.

Lewis Research Center,  
National Aeronautics and Space Administration,  
Cleveland, Ohio, November 30, 1965.

## REFERENCES

1. Buckley, D.H.; and Johnson, R. L.: Degradation of Polymeric Compositions in Vacuum  $10^{-9}$  mm Hg in Evaporation and Sliding Friction Experiments. SPE Trans., Vol. 4, No. 4, Oct. 1964, pp. 306-314.
2. Brown, R. D.; Burton, R. A.; and Ku, P. M.: Long-Duration Lubrication Studies in Simulated Space Vacuum. ASLE Trans., vol. 7, no. 3, July 1964, pp. 236-248.
3. Hendrix, William Robert: Process for Preparing Polyimides by Treating Polyamide-Acids with Aromatic Monocarboxylic Acid Anhydrides. Patent No. 3,179,632, United States, Apr. 1965.
4. Edwards, Walter Murray: Polyamide-Acids, Compositions Thereof, and Process for Their Preparation. Patent No. 3,179,614, United States, Apr. 1965.
5. Endrey, Andrew Laszlo: Process for Preparing Polyimides by Treating Polyamide-Acids with Lower Fatty Monocarboxylic Acid Anhydrides. Patent No. 3,179,630, United States, Apr. 1965.
6. Endrey, Andrew Laszlo: Aromatic Polyimides from Meta-phenylene Diamine and Para-phenylene Diamine. Patent No. 3,179,633, United States, Apr. 1965.
7. Endrey, Andrew Laszlo: Aromatic Polyimide Particles from Polycyclic Diamines. Patent No. 3,179,631, United States, Apr. 1965.
8. Devine, M. J.; and Kroll, A. E.: Aromatic Polyimide Compositions for Solid Lubrication. Lub. Eng., vol. 20, no. 6, June 1964, pp. 225-230.
9. Buckley, Donald H.; and Johnson, Robert L.: Mechanism of Lubrication for Solid Carbon Materials in Vacuum to  $10^{-9}$  Millimeter of Mercury. ASLE Trans., vol. 7, no. 1, Jan. 1964, pp. 91-100.

*"The aeronautical and space activities of the United States shall be conducted so as to contribute . . . to the expansion of human knowledge of phenomena in the atmosphere and space. The Administration shall provide for the widest practicable and appropriate dissemination of information concerning its activities and the results thereof."*

—NATIONAL AERONAUTICS AND SPACE ACT OF 1958

## NASA SCIENTIFIC AND TECHNICAL PUBLICATIONS

**TECHNICAL REPORTS:** Scientific and technical information considered important, complete, and a lasting contribution to existing knowledge.

**TECHNICAL NOTES:** Information less broad in scope but nevertheless of importance as a contribution to existing knowledge.

**TECHNICAL MEMORANDUMS:** Information receiving limited distribution because of preliminary data, security classification, or other reasons.

**CONTRACTOR REPORTS:** Technical information generated in connection with a NASA contract or grant and released under NASA auspices.

**TECHNICAL TRANSLATIONS:** Information published in a foreign language considered to merit NASA distribution in English.

**TECHNICAL REPRINTS:** Information derived from NASA activities and initially published in the form of journal articles.

**SPECIAL PUBLICATIONS:** Information derived from or of value to NASA activities but not necessarily reporting the results of individual NASA-programmed scientific efforts. Publications include conference proceedings, monographs, data compilations, handbooks, sourcebooks, and special bibliographies.

*Details on the availability of these publications may be obtained from:*

SCIENTIFIC AND TECHNICAL INFORMATION DIVISION  
NATIONAL AERONAUTICS AND SPACE ADMINISTRATION  
Washington, D.C. 20546