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THE ROLE OF THE MICRO ENVIRONMENT ON THE TRIBOLOGICAL BEHAVIOR OF MATERIALS

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ABSTRACT

Tribological systems consist of three components: The material surfaces to be lubricated, the lubricants, and the environment. Very frequently, the environment can play a dominant role in the behavior of materials in solid state contact. In the absence of surface oxides, adhesion and complete welding of solid surfaces occurs.

Differences in molecular species adsorbed on the solid surface also influence the behavior in adhesion, friction and wear studies. The presence of surface films can be affected by the environment where certain surface species which provide lubricating films can be displaced by environmental constituents. In liquid environments, the presence of surface active agents such as organic acids and other materials can cause changes in mechanical properties of those solid surfaces and thereby influence tribological properties. Even with lubricants, such as solid films such as molybdenum disulfide and graphite the environment plays a very dominant role in the friction behavior of these materials. Altering ambient pressure for example can produce marked differences in the friction characteristics, for molybdenum disulfide and graphite.

INTRODUCTION

The environment surrounding practical tribological components can play a very strong role in the adhesion, friction and wear behavior of these materials in solid state contact. It is for this reason that frequently the lubrication or tribological system is referred to as consisting of essentially three components. One, is the material surfaces to be lubricated whether it be a bearing, gear or seal or some other mechanical device; secondly, there is the lubricant be it a liquid or solid that provides the friction and wear reducing protective surface films and lastly there is the environment. The environment in many practical tribological systems can have a very strong influence upon the behavior exhibited and in some instances, the environmental constituents when present on surfaces can completely mask other effects such as that of the lubricants.

The objective of the present paper is to review the role of the micro-environment, that is, the environment on the surface of solids in solid state contact with other solids and to demonstrate the properties of the environment which exert an influence upon adhesion, friction, wear and lubrication of materials in contact. Further, the effect of the environment upon lubricants and lubricant properties will also be discussed as they relate to the interaction of the lubricant with the material surfaces in contact and the ability of lubricants to provide protective surface films.

ADSORBATES AND OXIDES

In a conventional atmospheric environment, the oxygen present in the air interacts with freshly generated metal and alloy surfaces to produce surface films; namely, oxides. These oxides play a very strong role in the adhesion, friction and wear behavior of metals and alloys. In the absence of these oxides, very strong adhesion, high friction coefficients and ultimately cold welding of materials from one surface to another is observed. If, for example, two normal metal surfaces are taken and placed inside a vacuum environment with a system capable of achieving pressures to 10^{-10} torr and the surfaces are very carefully cleaned with argon ion bombardment and then brought into touch contact, adhesion will immediately occur of one surface to the other. Attempts at tangential motion will cause a growth in the area adhered at the interface, that is, in the real contact area with an ultimate complete seizure of the surfaces one to another. With this occurence, the surfaces are generally severely disrupted (1,2).

When adhered surfaces are separated, the adhesion at the interface between the two dissimilar solid surfaces is sufficiently strong so that fracture does not occur at the interface but generally in the cohesively weaker of the two materials in contact (3). An example of such behavior is shown in the photomicrograph of figure 1. In figure 1 are presented the results of an experiment conducted in a vacuum chamber where two solid surfaces were brought into contact and the surfaces had been cleaned in the vacuum environment. Adhesion occurred at the interface, and when separation of the solid surface was attempted, fracture occurred in one of the two materials leaving material transferred to the opposite surface. The actual area of real contact at the interface can be seen in figure 1 by a careful examination of the interface region. There are what appear to be voids in the interface region where complete and intimate solid state contact across the interface did not occur. However, a great portion of the interfacial area does reflect solid to solid contact in adhesive bonding. This adhesive bond, that of figure 1, was generated as a result of attempting to slide one surface over the other. This resulted in growth in the adhered junctions at the interface leaving only a small area wherein there was no intimate contact of the two solid surfaces. On separation, the adhesive bond at the interface remained intact and fracture occurred in one of the solids as indicated in figure 1 by the rough piece which remained on the solid surface.

This type of adhesion and transfer behavior is observed for all clean metal surfaces and alloys in solid state contact. That is, when the environmentally contributed surface films, namely the oxides are removed, such adhesion is observed with strong adhesion and friction forces measured in excess of 100 under such circumstances. The surfaces of solids such as metals and alloys are so sensitive to the microenvironment in the contact region that the admission of very small concentrations of adsorbates from the environment to the solid surface are sufficient to markedly reduce adhesion and friction. For example, fractions of a monolayer on the solid surface will produce a marked reduction in the adhesion and corresponding static friction coefficients for metals in contact (4).

In addition to metals and alloys, non-metallic materials are also markedly influenced by the presence of the environment in their adhesion, friction and wear behavior. The presence of adsorbates on ceramic materials such as aluminum oxide have a pronounced influence on the friction coefficients measured for aluminum oxide (5).

Polymeric materials are also affected by the presence of environmental constituents on their surfaces. For example, nylon is a material which serves as a good solid self-lubricating material in certain mechanical applications. Nylon, however, depends upon the presence of adsorbed moisture for its effective lubrication; that is, for its low friction and wear properties. In the absence of moisture, nylon will not lubricate effectively and it becomes a poor tribological material (5).

Carbon materials are heavily used in mechanical devices such as dynamic seals and are extremely sensitive to environment and environmental constituents. It was established during the second World War that aircraft flying at high altitude resulted in excessive wear in carbon generator brush materials. Careful analysis of the surfaces revealed that the excessive wear of carbon materials, carbon bodies at high altitudes, was due to a reduction in the ambient pressure and more particularly with the reduction in the presence of moisture in the environment. Careful controlled experiments in the laboratory subsequently demonstrated that in the presence of moisture, carbon lubricates effectively exhibiting low friction, low wear and little tendency to adhere. In the absence of moisture, however, the carbon exhibits extremely heavy wear and becomes a very poor friction and wear material. In fact, by simply reducing the ambient pressure from 760 torr of air to an ambient pressure of approximately 1 torr, a 1,000 fold change in wear properties is observed. The wear increases 1,000 fold with a reduction in ambient pressure and a loss of moisture from the environment on the wear properties of carbon. Thus, moisture is needed on these surfaces and at the interface between two carbon bodies in relative contact in motion or between the carbon body and some other material in solid state contact (5).

LUBRICANT STRUCTURE

If one considers the environment as being something other than an ordinary air containing principally oxygen and nitrogen with some water vapor, but also considers vapors of hydrocarbons as constituents of the environment then the particular hydrocarbon molecular structure that may be present in the environment can have a very pronounced influence upon the adhesion, friction and wear behavior of materials in contact. For example, careful cleaning of iron surfaces in a vacuum environment will result in the generation of surfaces that are extremely highly energetic and will adhere one to another when brought into contact with cold welding occurring readily. If, however, a small amount of hydrocarbon gas is admitted to the vacuum chamber and allowed to adsorb on the clean iron surface, a structure will develop on the solid surface that will provide that surface with a protective film. Further, clean iron surfaces will chemibsorb nearly all hydrocarbons (6).

The presence of the hydrocarbon film will reduce adhesion, friction and wear because the surface energy has been reduced by the presence of the hydrocarbon molecules on the surface. The surface energy on the clean iron surface available for bonding across an interface to another solid surface has been taken up in the interaction of the clean iron surface with the lubricating molecules absorbing on the surface. The particular molecular structure, however, of the adsorbing hydrocarbon will also make a difference upon the tribological behavior. That is, a slight modification in the molecule will produce sensitivities in adhesion friction, and wear. These slight differences in molecular structure producing a change in adhesive friction and wear indicate extreme sensitivity in the tribological behavior of materials to environment and environmental constituents.

This effect can be demonstrated by the adsorption of a simple hydrocarbon such as ethylene oxide on to an iron surface and exposing that same surface to a different simple hydrocarbon with a slightly modified molecular structure, something such as ethylene chloride or as is commonly called vinyl chloride. If clean iron single crystal surfaces of the same orientation are exposed to equivalent concentrations of these two different simple hydrocarbons, namely ethylene oxide and vinyl chloride, entirely different surface structures result. These differences can be seen in the LEED (Low Energy Electron Diffraction) patterns presented in figure 2. LEED (Low Energy Electron Diffraction) is a device which allows one to examine the structural arrangement of atoms in the outermost atomic layer of the solid surface. Thus, in figure 2, we see the molecular arrangement in the diffraction pattern for the adsorbed ethylene oxide and vinyl chloride on the iron surface in the two patterns. Equivalent concentrations of each specie was provided. Thus, everything is constant except the particular molecular structure.

In the ethylene oxide there is the basic ethylene structure with oxygen present in the molecule and with vinyl chloride, essentially the same type of structure that is the ethylene structure but instead of having oxygen, chlorine is substituted in the structure for oxygen. With this subtle difference in the structures, however, marked differences in surface coverage are observed in the LEED patterns of figure 2. With the ethylene oxide, the six diffraction spots in an hexagonal array indicate that the ethylene oxide molecule was completely masked or covered with the iron surface. There are no diffraction spots seen from the iron in diffraction pattern of figure 2. There is a close packing, as it were, of the molecules of ethylene oxide on the iron surface providing a very effective and continuous surface film.

In contrast, however, with the vinyl chloride, the structure is a much more open structure with less than complete surface coverage and bonding of vinyl chloride to the surface. The four bright diffraction spots seen in a rectangular array in figure 2 with vinyl chloride absorption are associated with the iron. Thus, there is not complete coverage by vinyl chloride and nascent iron is still exposed at the surface. As one might anticipate, differences in adhesion and friction behavior are observed with these two films present. With the ethylene oxide, the adhesive forces are appreciably reduced between two clean iron surfaces in contact. Further the friction forces are less for the ethylene oxide on the iron surface than is observed with the vinyl chloride present on the surface. Thus, slight differences in the molecular structure of hydrocarbons present in the microenvironment of solid surfaces in contact can influence the tribological behavior of those surfaces.

LUBRICANT-ENVIRONMENT INTERACTIONS

Once a lubricating film has been formed on a solid surface as a result of interactions with the environment or by the deliberate application of lubricating films to solid surfaces, the presence of a lubricant on a surface can be altered or modified by interactions of environmental constituents with the surface in the asperity contact regions. Sulfur is an element frequently used as an anti-wear and anti-seizure surface film material. When sulfur is present in organic molecules or organo-metallics, it can interact at the solid surface in metallic systems to form metal sulfides which provide a minimum of adhesion, friction and wear for lubricated systems. In fact, many practical tribological devices rely very heavily upon sulfur for the formation of protective surface films.

Metal sulfides, however, that may exist on the surfaces of solids as a result of the interaction of lubricating species with the solid surface are extremely sensitive to microenvironmental constituents such as oxygen. The presence of oxygen at a rubbing interface of two surfaces in sliding, rolling or rubbing contact can bring about a change in the nature of the surface chemistry as a result of the interaction of the oxygen with the lubricated surface to produce a reduction, as it were, of the metal surface sulfide with the formation of metal surface oxides. In fact, if a carefully controlled metal sulfide surface is exposed to oxygen, the oxygen can completely displace or remove the sulfur from the solid surface with an oxide replacing it. This effect is demonstrated in the data of figure 3.

In figure 3, is plotted the relative Auger peak intensities where an Auger spectrometer is used for monitoring the presence of sulfur and oxygen on a iron surface. Auger electron spectroscopy is a surface sensitive analytical tool that allows one to analyze all the chemistry of a solid surface to a depth of four or five atomic layers with a sensitivity to 1/100th of a monolayer. It can detect all elements present on the solid surface except hydrogen and helium. The data of figure 3 were obtained by generating a sulfide film on an iron surface. The sulfide film representing basically a lubricating protective film. The surface was then exposed in a clean vacuum system to various concentrations of oxygen. As the concentration of oxygen in the system continuously increases as represented in figure 3 by the exposure in Langmuirs of oxygen, the Auger peak intensity for oxygen increased. With increasing exposures of oxygen the sulfur peak intensity decreased as indicated in the data of the figure. Ultimately, at some exposure, the sulfur present on the iron surface was completely displaced from the solid surface by oxygen and was replaced by an oxide film. There is a gradual decrease in the concentration of sulfur on the surface and a gradual increase in the concentration of oxygen on the surface with continued exposures to oxygen. Thus, microenvironmental constituents such as oxygen at the interface between two surfaces in contact with sulfide films can completely erase, as it were, the lubricating film that may be present.

MECHANICAL-ENVIRONMENTAL EFFECTS

In tribological systems, surfaces of two solids are generally in contact with either sliding, rolling or some type of relative motion between the two surfaces. In addition, there is some velocity or some rate of motion associated with the movement of the surfaces one relative to the other. Further, there is generally some mechanical loading applied to the surfaces in contact. The presence of these factors, namely, relative motion between the two surfaces and the imposition of loads on the surfaces in contact, can bring about marked changes in the nature of the surface chemistry in the presence of certain environments and the environment can affect the wear behavior of such systems. For example, for two solid iron or steel surfaces in contact in the presence of a liquid lubricant containing a sulfur additive, rubbing of the surfaces under relatively light loads results in the formation of surface films which are generally identified with XPS (X-ray Photoelectron Spectroscopy) as being principally surface oxides. With the mechanical activity of the surfaces relatively low wear is observed in the presence of these surface oxides. If, however, the load is increased to the point where the additive, the sulfur containing additive in the lubricant can interact with the nascent iron or steel surfaces as a result of disruption of surface oxides then with XPS analysis, sulfides are found to be present on the surface. In the presence of these sulfides, the friction, adhesion and wear behavior are much higher than they are in the presence of the oxides. This effect is demonstrated in the data of figure 4 where XPS data are presented for surfaces containing oxides and sulfides under three sets of conditions, (1) in the absence of rubbing, (2) under mild wear conditions, and (3) under severe wear conditions. It can be seen that under mild wear conditions, one type of surface film is present while under severe wear conditions, a completely different type of surface film is present. If the sulfur containing surface film observed in the severe wear conditions is exposed to oxygen for either sufficently long periods of time or at sufficiently high concentrations of oxygen, data analogous to that obtained in figure 3 will be observed for these particular films (7).

In the 1930's, Roscoe observed that the interaction of the environment with solid surfaces, particularly metal surfaces that produced metal oxides, altered the mechanical behavior of the metals. In some experiments with cadmium crystals, Roscoe found that when cadmium oxide was present on the surface, as a result of interaction of the cadmium single crystal with oxygen, that the mechanical properties were altered. The crystal became much less prone to plastic deformation increasing in its hardness (8).

In contrast to the observations of Roscoe, Rehbinder and his colleagues in the 1940's in Russia observed that in the presence of certain surface active organic molecules such as organic acids on solid surfaces made those surfaces much more prone to deformation. The surfaces strained more readily and at much lower stress levels than they did in the absence of the surface active organic species (9).

The Rehbinder work as well as that of his colleagues have established that surface softening, can occur for a number of classes of materials which include metals as well as non-metals. These surface interactions from environmental constituents with the solid surface not only influence mechanical properties, but also accordingly influence tribological behavior and this is demonstrated in the data of figure 5. In figure 5(a) is a schematic illustration of these surface effects (10). It is a plot of stress versus strain for materials under normal conditions and with the presence of surface films manifesting the Roscoe effect as well as with the presence of surface active liquids representing or demonstrating the Rehbinder effect. It can be observed from the data of figure 5(a) that in a stress/strain relationship with a surface film present from interaction of the environment with a solid surface, producing or manifesting the Roscoe or surface hardening effect, the stress/strain curve shows an increase in the strength of the material or reduction in plastic deformation with applied stress. In contrast, however, where a surface active specie is present, which produces a surface softening or a Rehbinder effect, a reduction in the stress required to produce strain is observed for materials when compared to the normal or the Roscoe effect in figure 5(a).

A sensitivity of tribological behavior to such environment surface interactions are seen in the data of figure 5(a). In figure 5(b), both a wear track width and friction coefficient are plotted for a zinc crystal surface, the basal orientation (0001) surface where a ruby ball slid on that surface in a $[10\bar{1}0]$ direction. The sliding experiments were conducted in three different environments. Dry sliding which represents the equivalent of the normal condition of figure 5(a), sliding with an oxidized surface which is analogous to the Roscoe effect in figure 5(a), bearing in mind that Roscoe observed the formation of oxides on cadmium while in figure 5(b) the oxides are present on zinc. The third environment was the sliding, friction and wear behavior of the zinc surface in contact with the aluminum oxide in a 5 percent hydrocholoric acid solution. The 5 percent hydrochloric acid solution representing a surface active liquid situation which would produce the Rehbinder effect.

The data of figure 5(b) indicate that the normal dry surface, gives values in track width which would be equivalent to the wear of the surfaces which are immediate between the surface which has been oxidized to produce a surface hardening or Roscoe effect and that of the surface softening or Rehbinder effect. The tribological results being very analogous to the stress/strain data of figure 5(a).

With the oxide present on the surface in the sliding friction experiment, the track width is much smaller in size than it is in the absence of the surface hardening effect of the oxide. One observes a smaller track width because the surface has been hardened by the presence of the oxide as was observed by Roscoe and that produces a reduction in wear to the surface. In contrast, in the presence of the surface active liquid, the surface becomes softer or more prone to deformation plastically under a fixed load. As a consequence, thereof, the wear track generated in the solid surface is much larger than it is in the normal case or in the case where the oxide is present on the solid surface. Thus, there is a distinct and definite relationship between the track widths observed in figure 5(b) and the stress/strain behavior observed in figure 5(a).

In sliding friction, there are two components to the friction force or resistance to tangential motion which results in the measure of friction coefficients. This is the friction associated with the real area of contact in shear as well as the amount of plowing that must take place of the surface. With the sapphire or ruby ball sliding on the zinc surface, there is a considerable amount of deformation that occurs to the zinc surface under the very hard ruby ball. The presence of the oxide, however, while it reduces the amount of plastic deformation that may occur on the solid surface, it also increases the strength of the surficial layers which must be plowed increasing their resistance to tangential motion and causing an increase in fricton force. In contrast, in the presence of the surface active liquid, namely the 5 percent hydrochloric acid in figure 5(b), the increase plasticity of the surface reduces the resistance to tangential motion and the plowing of the zinc by the ruby ball. As a consequence, a lower friction coefficient is measured in the presence of the surface active liquid manifesting the Rehbinder effect than is observed for the oxidized case representing the Roscoe effect. This difference was observed over a range of loads as indicated in the data of figure 5(b). Thus, from the data of figure 5, it is apparent that interaction of environmental constituents with solid surfaces can produce changes in the mechanical behavior of the solid surfaces and those changes in mechanical behavior of the solid surface are reflected in changes in tribological behavior such as friction and wear.

EFFECT OF ENVIRONMENT ON SOLID FILM LUBRICATION

In addition to the environment influencing or altering the chemistry of solid surfaces and their adhesion, friction, wear or mechanial behavior, the environmental constituents can interact again with lubricants particularly with solid film lubricants that may be present on surfaces to alter their observed behavior.

Molybdenum disulfide and graphite are two of the most commonly used solid film lubricants used to reduce adhesion, friction and wear of metals in solid state contact. Yet, both of these materials are extremely sensitive to the microenvironment in their tribological performance. For example, molybdenum disulfide is a much better lubricant in a vacuum environment than it is in air. In contrast, graphite which lubricates very effectively in air at atmospheric pressure is completely ineffective as a lubricant in a vacuum environment. These differences are due to environmental constituents that are present on the surface of the solids; namely, water vapor.

The effect of atmospheric pressure upon the friction performance of molybdenum disulfide and graphite are indicated in the data of figure 6. In figure 6, a friction coefficient is plotted as a function of ambient pressure from molybdenum disulfide and graphite. It can be seen from the data of figure 6 that as the ambient pressure is reduced toward that of a vacuum of 10^{-10} torr, the molybdenum disulfide goes through a decrease or a marked reduction in friction coefficient as one achieves levels of high vacuum. The presence of adsorbates which have been pumped off the surface and identified by mass spectrometry indicate that the water vapor that is present on the molybdenum disulfide crystallites is deterimental to its lubricating characteristics. The friction coefficients are markedly superior in the absence of the water vapor than they are in its presence.

In contrast to the behavior of molybdenum disulfide in Figure 6, it is seen that the friction coefficient for graphite increases as the ambient pressure is reduced from atmospheric to that of a good vacuum. Again, water vapor plays a very strong role in the behavior of the solid film lubricant. In this instance, the water vapor is extremely beneficial for the lubrication of solid surfaces with graphite. Physically adsorbed water on the graphite platelets improves friction characteristics. In a vacuum system, again by mass spectrometry, water vapor is observed to be liberated by the graphite and the friction coefficient of the graphite increases markedly with a loss of the water from the solid surface. Ultimately, in a vacuum environment, very high friction coefficients comparable to those obtained for drying metal sliding are observed for the graphite. The graphite becomes ineffective as a solid film lubricant in the vacuum environment and is never used as a lubricant for vacuum applications for this reason. Thus, from the data of figure 6, it is apparent that the lubricating properties of solid film lubricants can be altered by a change in the microenvironment at the surface of solids in contact. Alterations in the atmospheric pressure and atmospheric constituents can produce a destruction of the lubricating properties of graphite and can result in an improvement in the lubricating properties of molybdenum disulfide. The data of figure 6 again indicating the extreme importance of microenvironment on the behavior of tribological systems.

CONCLUSIONS

Based upon the results obtained and presented in this review, it can be seen that the microenvironment plays a very strong role in tribological behavior of materials in contact.

1. Oxygen from a normal air environment interacts with solid metal surfaces, to provide a continuously protective surface film and to keep metal surfaces from adhering one to another. Naturally occurring oxides are probably the best natural solid film lubricants we have. They are contributed to us by the environment.
2. Hydrocarbons adsorbed from the environment on solid surfaces alter adhesion, friction and wear behavior and minor differences in molecular structure produce marked differences in tribological behavior.
3. Environmental constituents can completely displace lubricating films from solid surfaces altering adhesion, friction and wear behavior.
4. Mechanical activity of solid surfaces in contact can produce interactions with environmental constituents to alter surface chemistry and mechanical behavior. This alteration in surface chemistry and in mechanical behavior will alter tribological performance.
5. The presence of environmental constituents such as water vapor can markedly alter the lubricating characteristics of of solid film lubricant materials. Molybdenum disulfide can be improved in it lubricating characters by reduction in ambient pressures and elimination of moisture from the environment whereas graphite becomes ineffective as a solid film lubricant once moisture from the environment has been removed.

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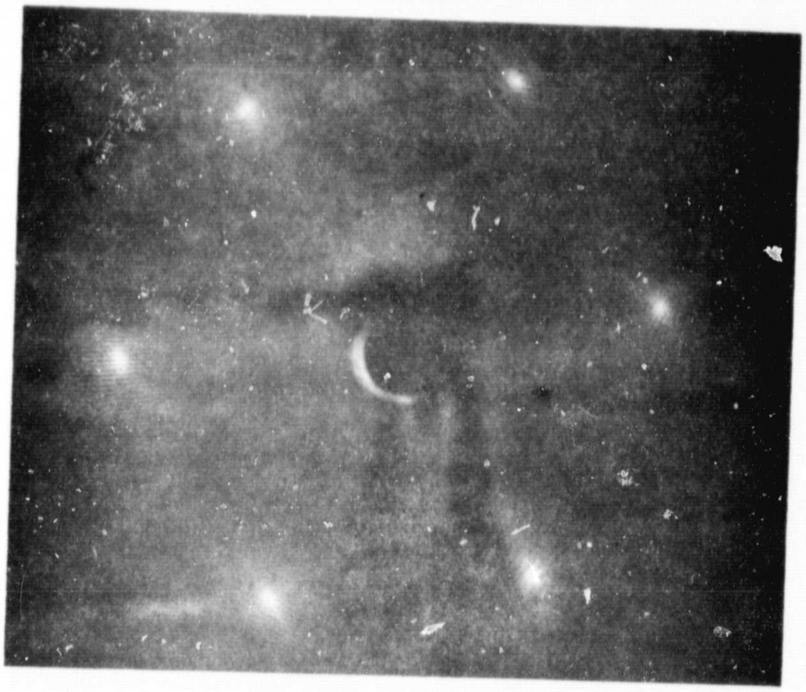
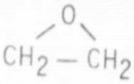
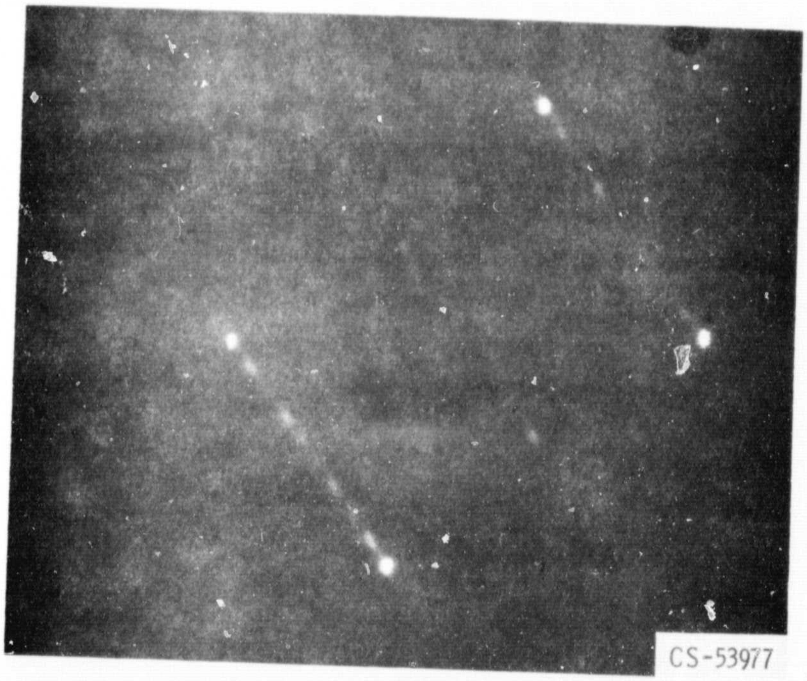
CS-32218

Figure 1. - Severe surface welding resulting from unlubricated sliding. (2% Al-Ni alloy from 10^{-9} mmHg vacuum experiment.)





$$\overset{O}{CH_2 - CH_2}$$



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$CH_2 = CHCl$

Figure 2. - Leed patterns obtained with two polymer forming hydrocarbons on iron (011) surface. 100 L exp.

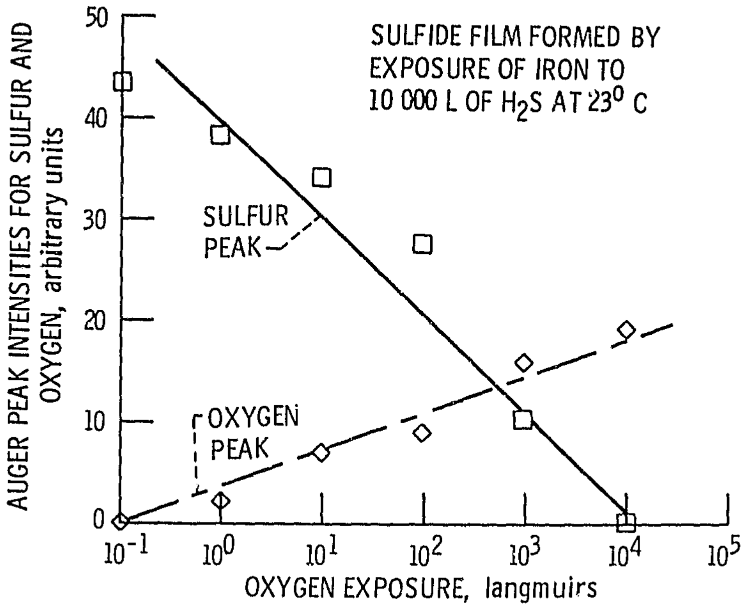


Figure 3. - AES evidence for displacement of sulfur from iron surface by oxygen.

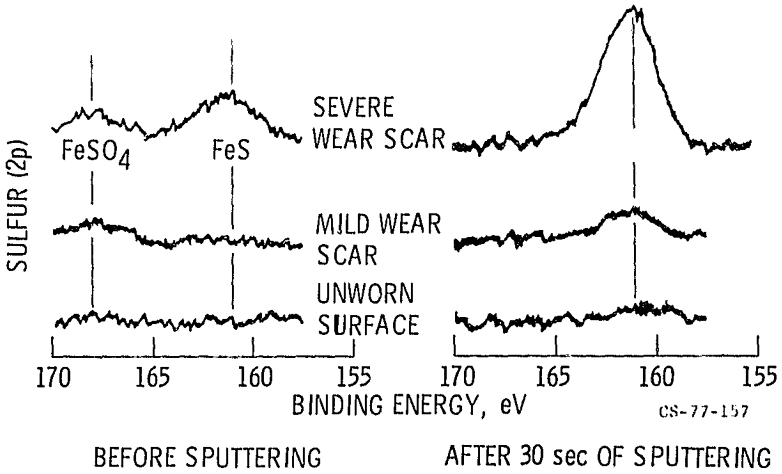
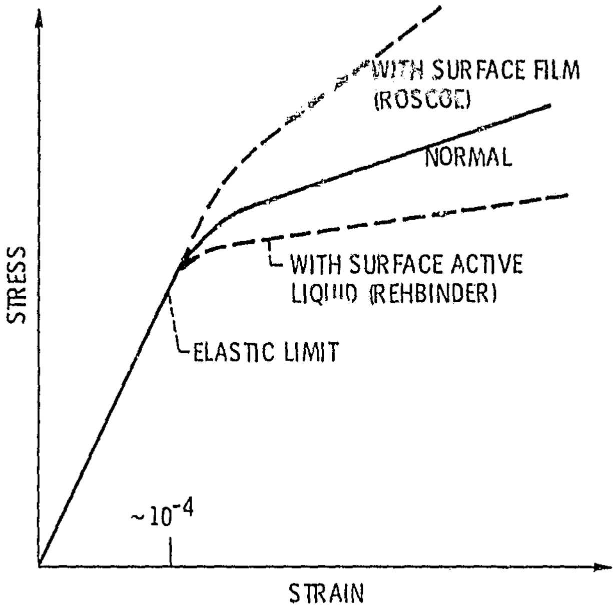
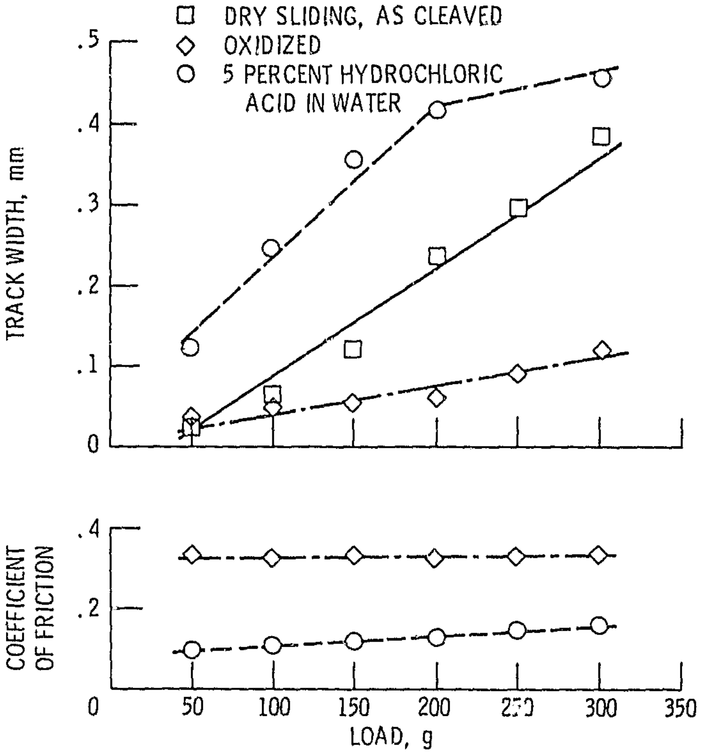


Figure 4. - Sulfur ESCA peaks from wear scars and unworn surface.



(a) SCHEMATIC ILLUSTRATION OF THE PRINCIPAL EXTRINSIC SURFACE EFFECTS (REF. 55).



(b) WIDTH OF WEAR TRACK AND COEFFICIENT OF FRICTION PRODUCED WITH RUBY BALL SLIDING ON ZINC SINGLE CRYSTAL (1001) SURFACE IN [1010] DIRECTION. (SLIDING VELOCITY, 1.4 mm/min; TEMPERATURE, 23° C; DRY ARGON).

Figure 5. - Influence of surface active species on mechanical and tribilogical behavior.

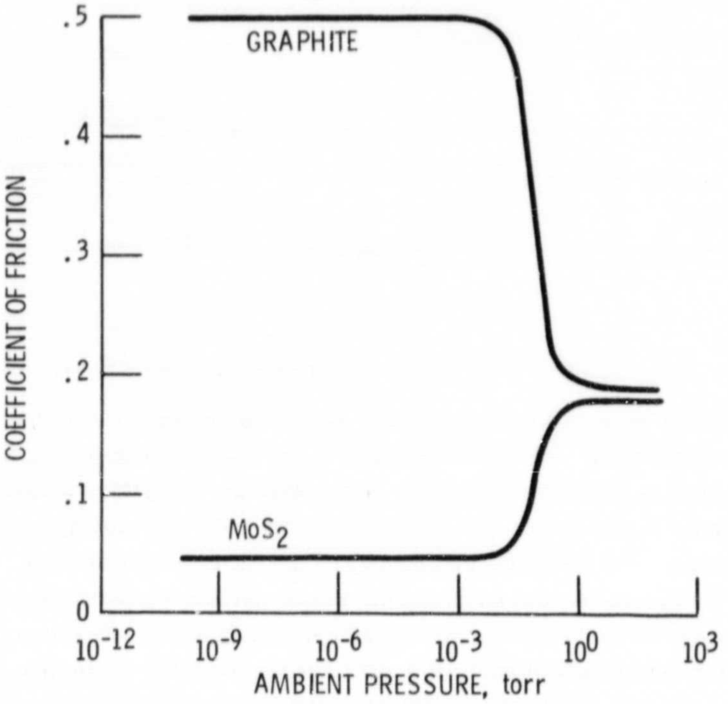


Figure 6. - Effect of ambient pressure on friction of graphite and MoS_2.