NUCLEATION AND FORMATION OF OXIDE FILM UNDER A MAGNETIC FIELD

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Abstract— Nucleation and formation of an oxide film in contact with sliding unidirectional dry were studied in a vacuum chamber with and without the application of a magnetic field to identify the role of the intensity of magnetic field on the oxidation wear. The wear tests of AISI 1045 steel / steel AISI 1045 Were are carried in a tribometer pin on disc in three different gas environments: in ambient air, in oxygen at $10^5$ Pa and under vacuum at $5.10^{-5}$ Pa. The formation of the oxide film depends strongly on the partial pressure of oxygen and the intensity of the magnetic field. After delamination of the first wear debris, these wear debris compacted to form the oxide film.

Keywords—friction; wear; oxidation; environment gas.

I. INTRODUCTION

Reduction in friction and wear loss presents many interests for tribologists. The associated researches were carried out mainly by experiments [1-2], where the presented results come mainly from pin-on-disc.

Furthermore, the magnetic field is generated in several mechanical and electrical sectors such as generators, electric motors... It is very important to understand the influence of the magnetic field on tribological behaviour of metal contacts, in order to prevent failure mechanisms and obtain greater energy efficiency. The objective of this study is to investigate mechanisms and effects by which the magnetic field changes the dry sliding contact of steel ferromagnetic/steel ferromagnetic couple.

The effect of magnetic field on friction and wear mode has been studied by some researchers [3-10]. After all works realised on this study, it is difficult to draw definitive conclusions of a fundamental nature. In 1970 Muju [3] was possibly the first to observe the effects of magnetic field on wear mode of different metallic materials. These results have shown that the magnetic field increases the hardness of the sliding surface and magnetic permeability.

Hiratsuka and Sasada [6] have observed a severe-mild wear transition and reduction of wear rate and friction coefficient in air due to the magnetic field during pin on disc experiments carried out for Ni/Ni and Fe/Fe contacts. Paulmier and Zaidi [7-11] have observed a decrease in the wear rate and friction coefficient of a steel/steel couple in ambient air.

The effect of magnetic field has a direct relationship with the atmosphere around the rubbing surface. Moreover, the application of magnetic field affects the contact junctions and the wear debris. In our study, we determine the constitution of oxide films as their kinetic growth, their role on the tribological behaviour and the temperature contact of ferromagnetic/ferromagnetic couple. Besides, it applied a magnetic field perpendicular to the contact surface.

The main objective of this study is to investigate mechanisms and effects by which the magnetic field affects the sliding contact. Another objective is to evaluate the role of partial pressure of oxygen. In this case, the contact was realised in a controlled atmosphere under a magnetic field to identify the physic-chemical phenomenon, and mechanical characteristics of the oxidation process.

II. EXPERIMENTAL CONDITIONS

A multi-specimen friction test was used as the wear test rig on pin-on-disc tribometer located in a stainless steel vacuum chamber; the pin specimen was kept stationary while the circular disc was rotating.

The pin has a cylindrical shape of 15 mm in length and 5 mm in diameter, the disc has 70 mm of diameter and 7 mm of thickness, with a plane face of roughness 0.3 µm. Radius of contact track on disc is 25 mm. In order to get the same conditions of sliding surface in successive tests, the pin and the disc are replaced before each experiment. The sliding speed is fixed at a constant value $V = 0.5$ m/s. The material studied is the steel AISI 1045, the chemical composition in weight percent for both the pin and the disc is presented in table 1.

The tests are carried out under three types of gas environment, in both ambient air and oxygen at a pressure of $105$ Pa and in vacuum at a pressure of $5.10^{-5}$ Pa. The ambient temperature is approximately 21°C, the relative humidity varies between 30 and 40%. The tests were carried out under four normal loads applied on the top end of the pin: 9.25 N, 18.5 N, 27.75 N and 37 N, other important loads were used to compare and validate results obtained. The duration of each test is 30 minutes; this time interval was chosen because friction and wear stabilize before 30 min.

To investigate the effect of the magnetic field on the sliding contact, a magnetic field is created by electric current.
applied around the pin with a coil of 300 spires (Fig. 1) being able to deliver a field intensity from $H = 0$ to 40 kA/m.

III. RESULTS

A. Friction

Figure 2 illustrates the effect of gas environment and magnetic field on the evolution of friction coefficient with sliding distance. The application of magnetic field in ambient air changes completely the tribological behaviour of rubbing surface. So, in ambient air, the friction coefficient decreases from 0.5 to 0.43 for a normal applied load $N = 37$ and a magnetic intensity $H = 20$ kA.m$^{-1}$. However, the low magnetic intensity ($H < 5$ kA.m$^{-1}$) has not a great influence on friction and wear in ambient air.

Under pure oxygen, the friction coefficient decreases from 0.42 to 0.3 for a normal load

$N = 37$ N in the presence of magnetic field ($H = 20$ kA.m$^{-1}$). Besides, the fluctuations friction coefficient are reduced in this atmosphere around the average value. Therefore, under oxygen, a severe-mild wear transition was observed at the beginning of sliding contact and for a few seconds, when the oxide layer was nucleated.

In vacuum, the magnetic field has no effect on the friction coefficient, it remains almost stable around an average value of 0.38 for $N = 37$ N, and the wear track remains ductile without formation of oxide layer.

The evolution of friction coefficient has two distinct phases in different gases:

- The first concerns the transitional phase, which corresponds to the running-in period during which the friction coefficient $\mu$ evolves between the static friction coefficients $\mu_s$ and the dynamic friction coefficient $\mu_d$.
- After transitional phase, $\mu$ remains almost stable and fluctuates around its average value.

B. Wear

Fig. 3, reports the measurements of wear rate with normal load in different atmospheres without magnetic field. It shows that the lowest wear rate is obtained in oxygen and the most important one is obtained in ambient air. The effect of the magnetic field on the wear rate is significant in ambient air, noting that a sufficient intensity of magnetic field ($H = 10$ kA/m) reduces the wear rate by 8 to 10 times (Fig. 4). The thickness of the oxide layer increases until the rubbing surface was covered; this layer is extremely thin and porous. Furthermore, the increase of magnetic field intensity up to 20 kA/m in ambient air decreases more the wear rate.

The effect of the magnetic field on contact tribology in ambient air remains valid up to a limit intensity $H = 40$ kA/m. Beyond this limit, other effects appear: increase of the magnetic attraction force and contact temperature. When magnetic intensity exceeds 40 kA/m, no-oxidized wear particles result only at the beginning of sliding.

In oxygen, the wear rate is very low. So, the ratio between the wear rate obtained in ambient air and in oxygen is respectively, from 8 to 10 times with magnetic field and about 30 to 40 times without magnetic field (Fig. 5). Except that in

### TABLE 1 COMPOSITION OF STEEL AISI 1045

<table>
<thead>
<tr>
<th>Element</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>98.80</td>
</tr>
<tr>
<td>C</td>
<td>0.45-0.51</td>
</tr>
<tr>
<td>Mn</td>
<td>0.50-0.80</td>
</tr>
<tr>
<td>Si</td>
<td>0.10-0.40</td>
</tr>
<tr>
<td>S</td>
<td>$\leq 0.035$</td>
</tr>
<tr>
<td>P</td>
<td>$\leq 0.035$</td>
</tr>
</tbody>
</table>

With the increasing of the normal load, the friction coefficient of the steel/steel couple tends towards a stable value. The friction coefficient becomes stable with the increasing of normal load up to 75 N, it remains unchanged ($\mu = 0.38$) for each gas environment.
oxygen, the magnetic field intensity cannot exceed 20kA/m, because it has a degassing in this atmosphere.

Note that unlike ambient air and oxygen atmosphere, the wear process in vacuum is completely different. A severe adhesive wear is generated in vacuum which causes a transfer of particles from the pin to the disc. However, the disc loses almost the same weight as that lost in ambient air without magnetic field, because in vacuum, the magnetic field does not generate an oxide layer. The small decrease of the wear rate generated in vacuum with magnetic field (Fig. 6) can be explained by the magnetic attraction of wear particles on the interface and the magnetostrictive changes of the worn surface [4].

C. Contact temperature

Fig. 7, illustrates the evolution of contact temperature obtained in the three environments with the sliding distance. It is observed that the contact temperature increases abruptly during 2 to 3 minutes at the beginning of sliding contact to about 60 °C. After this phase, the temperature evolution remains low ascending. In ambient air, when the magnetic intensity increases beyond 10 kA.m⁻¹, the impact of joule effect due to eddy currents is shown on the evolution of contact temperature.

The average temperature in oxygen is less than that obtained in ambient air, this reduction is due to the mild wear. Because the increase in local temperature results from the friction energy dissipated in the contact area. Thus, the oxide layer minimizes plastic deformations that generate even more flash temperatures at contact junctions [2]. So, the average contact temperature obtained under oxygen remains stable after the running-in period. In oxygen and without magnetic field, the temperature remains almost unchanged after the abrasive phase and throughout the rubbing contact. The temperature recorded in the presence of the magnetic field increases as the magnetic intensity increases, this contact temperature is about 92 °C for H = 20 kA.m⁻¹ and N = 37 N. The increase of temperature induced by the magnetic field was low; it can not affect the activation of the steel oxidation.

The average temperature obtained in vacuum is more important compared to that recorded in ambient air and in
oxygen. It reached up to 140 °C for a normal load of 37 N. The transfer particles adhered on the pin generates the delamination on the volume of sliding surface of the disc. This delamination produces rough particles which were adhered to the pin surface (Fig. 8). The transfer particles lead to a small real contact area between the pin and the disc. According to the Archard law [13], when the contact area decreases, the contact temperature increases. In addition, in vacuum, several processes occur, which increase the contact temperature, these process are: the important roughness of the interface and the strong adhesion of contact. In the presence of the magnetic field, the pin temperature increases from 3 to 22 °C in proportion to the magnetic intensity.

**D. Evaluation of contact temperature**

According to Archard [13] method, the total heat quantity released by friction and Joule effect in the interface is:

\[ Q = Q_d + Q_p \]  

Where \( Q_d \) is the heat quantity per unit of time conducted in the rotating disc; \( Q_p \) is the heat quantity conducted in the pin. \( Q \) is shared between the two surfaces, so the rise in temperature \( \Delta \theta_m \) is the same on each side.

When the sliding speed \( v \) is low (\( v < 1 \text{ m.s}^{-1} \)), the temperature variation \( \Delta \theta_m \) of the pin contact surface is given by the following formula:

\[ \Delta \theta_m = \frac{Q_p}{4a\lambda_p} \]  

On the disc contact surface the rise of the temperature is given by:

\[ \Delta \theta_m = \frac{Q_d}{4a\lambda_d} \]

Where \( a \) is the real contact radius given by the following relationship:

\[ a = \left( \frac{N}{\pi H_v} \right)^{1/2} \]

Where \( H_v \) is the hardness of the softer material and \( N \) is the normal load.

\( \lambda_p \) and \( \lambda_d \) are the thermal conductivities of the pin and the disc respectively; as the steel pin and the steel disc are the same one, so \( \lambda_p = \lambda_d = \lambda \)

\[ \lambda = 46 \text{ W.m}^{-1}.\text{K}^{-1} \]

In the other hand, the heat quantity generated by the friction energy \( Q \) is done by the next relationship:

\[ Q = \mu N N \]  

Where, \( \mu \) : the friction coefficient , \( N \) : the normal load and \( v \): the relative speed

the heat conduction and dynamic moment are characterized by the Peclet number \( L \). The speed limit is determined from the values of the Peclet number \( L \):

\[ L = \frac{v a}{2 \chi_t} \]

and \( \chi_t = \frac{\lambda}{\rho c_p} \) thermal diffusivity,
where $\rho$: charge density of the disc, $c_p$: specific heat.

As a first step, it is necessary to assess the actual radius of contact $a$. A reasonable order of magnitude is obtained using the traditional relationship:

$$N = \pi a^2 H_d \quad (8)$$

($H_d$: hardness of the steel after rabbing).

The Archard method shows that the contact temperature of the steel/steel couple increases with the increasing friction and reducing the actual area of surface contact. Indeed, the actual area of contact gradually decreases when the hardness of the interface increases. These two major parameters for calculating the contact temperature show that the method presents a gap which varies from 40 to 48 °C on oxygen and ambient air; this is because the actual area of contact is a parameter that remains difficult to determine. In vacuum, the temperature difference between the calculated and experimental temperature is around 25 °C. This small difference is justified by the absence of convection in this environment. Temperatures obtained in vacuum are due to the strong bond that generates the transfer of particles, and promotes delamination on the volume of material by shearing fast.

II. DISCUSSION

A. Formation of oxide layer

During sliding contact of the steel / steel couple, the oxide film is formed gradually on the worn surface. This oxide film is formed in the following conditions: ambient air with magnetic intensity $H > 5$ kA / m, oxygen with and without magnetic field. The wear track in these conditions is partially or completely covered by an oxide film. Indeed, under magnetic field, the worn surface of the disc undergoes an abrasive wear, and then the formation of oxide particles takes place on the worn surface. If the applied normal load and the magnetic intensity are low ($P = 9.25$ N, $H = 5$ kA / m), abrasive wear at the beginning of the sliding persists a long time, and the nucleation of oxide film starts after about 4 min. When the normal load increases with a lower magnetic intensity ($P > N 9.25$, $H = 5$ kA/m), abrasive wear increases and generates large wear particles. So, the normal load has practically no effect on the nucleation of oxide film.

However, when the magnetic field increases between 5 and 40 kA/m, the abrasive phase reduces and the formation of the oxide film takes place quickly on the worn surface. The oxide film is formed as the following mechanism: at the beginning environments. According to the Archard law, the rise in temperature is written as:

$$\Delta \theta_m = \alpha \frac{Q}{4a\lambda} \quad (9)$$

and $\alpha = -0,11L + 0,85$

Our measurements show that in oxygen, the hardness of wear track is higher to that measured in ambient air, $H_d = 550$. However, in vacuum, the hardness grows up to 700 Hv. The values of the temperature rise are given in the following table:

<table>
<thead>
<tr>
<th>Environment</th>
<th>$\mu$</th>
<th>$P$ (N)</th>
<th>$H d$ (Pa)</th>
<th>$\alpha$ (µm)</th>
<th>$L$</th>
<th>$\alpha$</th>
<th>$Q_d$ (W)</th>
<th>$T m$ (°C)</th>
<th>$\Delta T$exp(°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient air</td>
<td>0.51</td>
<td>18.5</td>
<td>25,108</td>
<td>48.5</td>
<td>0.96</td>
<td>0.74</td>
<td>4.62</td>
<td>108</td>
<td>60</td>
</tr>
<tr>
<td>Oxygen</td>
<td>0.47</td>
<td>18.5</td>
<td>29.2,108</td>
<td>45</td>
<td>0.93</td>
<td>0.74</td>
<td>4.16</td>
<td>98</td>
<td>58</td>
</tr>
<tr>
<td>Vacuum</td>
<td>0.42</td>
<td>18.5</td>
<td>37,108</td>
<td>39.9</td>
<td>0.9</td>
<td>0.75</td>
<td>3.9</td>
<td>125</td>
<td>100</td>
</tr>
</tbody>
</table>

Note that, the size of wear particles generated during the latter steps increases with the load and decreases when the magnetic intensity increases up to 40 kA/m.

B. Worn surface in ambient air

In ambient air and without magnetic field, some wear particles remain trapped on the sliding surface. These particles reduce the real contact surface and increase the pressure at the contact asperities. This process increases the shear stresses at the asperities and generates a repetitive plastic deformation during sliding contact, which is unfavourable to the dry sliding, and leads to a severe abrasion and a significant wear rate. As for the effect of the magnetic field on the chemisorption of oxygen, EDX analysis confirms that in ambient air without magnetic field, the worn surface retains the same chemical composition as steel before friction and the contact interface remains metal/metal.

When the magnetic field is applied, the oxidation of the worn surface is accelerated, which decreases the mutual adhesion of contact surfaces. Finally, the oxide film had an almost homogeneous form; it covers the most rubbing surface (Fig. 9). The agglomeration of oxide particles generates the third body; this one becomes increasingly thin when the intensity of magnetic field increases. The oxide layer plays a role of solid lubricant which is favourable to the reduction of wear rate and friction. The EDX analysis show that the oxygen rate adsorbed on the worn surface of the disc varies with the applied magnetic intensity. Figure 9 shows the difference in the oxygen rate absorbed during the sliding contact, for $H = 0$ and 20 kA / m.

Indeed, the oxygen rate adsorbed on the worn surface in ambient air without magnetic field is almost the same throughout the contact surface. The intensity of oxygen varies between 100 and 200 coups (Fig. 9-a); it is virtually the same
as that of a fresh steel surface. However, the application of magnetic field increases the chemisorption of oxygen. So the fine oxide particles are compacted particularly in the grooves of sliding surface. In this case, the oxygen intensity rises to 2200 coups (Fig. 9-b).

Indeed, the dislocations induced by the magnetic field accelerate and generate defects of contact asperities [3]. Warren [5] has shown that the stress concentrator on the worn surface has a beneficial effect on increasing dislocations movement. Remind that, the accumulation of dislocations on interface harden the worn surface and the contact junctions.

C. Worn surface in oxygen

Most theories of wear that exist refer to the generation of wear particles delaminated from that the friction and wear of dry contact are affected by the presence of fine particles in the sliding surface [14]. Hiratsuka and Pan [6, 15] found that the magnetic field applied to ferromagnetic materials increases the oxygen chemisorption on the surface magnetized. Indeed, the no-magnetised sliding contact in oxygen shows that a part of particles are ejected as wear debris. The formation mechanism of oxide layer in oxygen is the same one with or without magnetic field. Therefore, the oxide layer in magnetised contact is more compact.

The relationship between the wear rate and the partial pressure of oxygen can be explained by the size of wear debris. Increasing the partial pressure of oxygen leads to decrease the size of wear particles, which facilitates the continued development of the oxide layers and promotes the compaction of particles trapped on the sliding surface.

The application of magnetic field accelerates the oxygen chemisorption in the vicinity of contact junctions, which oxidize faster these junctions. Then, repeated contacts shear the oxidized junctions to be agglomerated and compacted in the interface. Figure 10 shows respectively, the wear track in oxygen with and without magnetic field and their EDX analysis. The EDX analysis of the worn surface shows the difference in oxygen rate at the peaks and the grooves of rubbing surface. It is noted that the wear track with magnetic field is less rough, and the oxygen rate absorbed on the wear track is larger than that produced without magnetic field.

The thickness of the oxide film in oxygen is based on the oxygen rate adsorbed. So, the oxygen rate had an intensity of about 6500 coups in magnetized contact. In the non-magnetized contact, the intensity of oxygen is about 3200 coups. The report of oxygen rate adsorbed between the non-magnetized contact and magnetized contact is approximately 2 times.

According to the results, it finds that:

Increasing the partial pressure of oxygen is fundamental to generate the formation of oxide film.

The application of magnetic field in oxygen reduces the wear rate because the size of the particles trapped at the interface of contact becomes increasingly fine when the magnetic field increases.

D. Worn surface in vacuum

Figure 11 shows that the wear track on the disc is very rough in the presence and the absence of magnetic field. Similarly, the EDX analysis of the wear track under vacuum with or without magnetic field shows that the chemical components of the worn surface is almost the same one as that of original steel. Indeed, the number of oxygen molecules is insignificant in vacuum, which prevents the formation of the oxide layer.

The wear debris obtained under vacuum are metallic and relatively large with and without magnetic field. The strong adhesion at contact interface increases the hardness of wear track from 20 to 30%, which generates the delamination of large particles in the disc volume. Theses particles are
loosened as wear debris, or transferred to another zone in the wear track of the disc or the pin. By reducing the pressure on the vacuum chamber from 10−2 Pa to 10−4 Pa, the wear debris size is multiplied by 2 and the wear track becomes more hardened. Secondly, the wear mode in vacuum changes completely the morphology of the wear track. At the beginning of sliding, the pin loses some particles, and after a few cycles when the hardness of wear track increases, another layer of particles adheres to the pin surface. The transfer of particles to the pin ceases when the transferred layer reaches a thickness of about 0.5 mm.

In addition, the magnetic field has almost no effect on the coefficient of friction and wear rate. This stability is due to the no formation of the oxide layer in this environment. Indeed the negligible impact of magnetic field in vacuum leads to two essential conclusions:

- Low partial pressure of oxygen in the environment does not allow the formation of the oxide layer,
- The magnetic field accelerates the chemisorption of oxygen by iron.

E. Oxidation wear process

Under atmospheric condition, magnetic field and partial pressure of oxygen cause oxidation of sliding surface of steel/steel couple. Additionally plastic deformation of worn surface accelerates oxidation process [16].

The formation of oxide film on worn surface requires a period of time. After a short running-in period (more than 500 cycles) an oxidational wear mechanism was established for all the steels tested under magnetic field in both ambient air and oxygen. In running-in period, the thickness of oxide film is too low, thus severe wear occurs. After running-in period, oxygen content increases enough to form thick oxide film and spread over the whole area of the contacting surface, thus wear rate reduced. But when oxide film reaches a critical thickness, oxide film becomes unstable and breaks up to form oxide wear debris due to the brittleness and internal stress of oxide. A steady oxidation wear continues with alternating process of oxide-film formation and delamination of oxide particles from contacting asperities. It can be noticed that the variation of wear rates corresponds to variation of magnetic intensity and partial pression of oxygen (Fig. 4-5). When the oxide film reaches about 2 µm in thickness, sliding surface state was relatively smooth and friction coefficient decreases.

III. CONCLUSION

A parametric study was carried out on the influence of gas environment and magnetic field on tribological behaviour of the sliding contact of ferromagnetic steel AISI 1045 / ferromagnetic steel AISI 1045 couple, which leads to the following conclusions:

1. The magnetic field has a great influence on the wear rate in ambient air, its presence divides the wear rate by 8 times. Thus, it changes the wear mode from a severe abrasive wear to a mild wear.
2. Oxygen is the best environment for gaseous tribological behaviour of the steel / steel couple. The wear rate decreases from 30 to about 35 times compared to the one obtained in ambient air.
3. In vacuum, the adhesive wear generates the transfer particulate from the disc to the pin. Therefore, the application of magnetic field in vacuum had a negligible effect.
4. The EDX analysis shows that the oxygen rate adsorbed on the worn surface of the disc increases with the magnetic intensity and the partial pressure of oxygen.
5. The oxidation of the worn surface is accelerated by the magnetic field, which decreases the mutual adhesion of surfaces in contact. The third body becomes increasingly thin in the presence of a magnetic field, and plays a role of solid lubricant which is favourable to the reduction of wear and friction. The oxidation of ferromagnetic steel / ferromagnetic steel couple is mainly due to the chemisorption of oxygen.
6. In addition, the magnetic field accelerates the oxidation of contact surfaces and allows the transition from severe to mild wear after the nucleation of oxide film on the wear track.

References


