TRIBOLOGICAL AND RHEOLOGICAL BEHAVIOUR OF LUBRICATING OILS ADDITIVED WITH POLYMERS

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SUMMARY
In this paper, the effect of the polyethylene additive on the tribological and rheological behaviour in EHD lubricated contacts was investigated. The tribological behaviour was studied from two point of view: EHD film-forming properties (in a rolling bearing) and scuffing resistance (on four-ball machine). A semi-empirical method is described to calculate the rheological parameters in EHD contacts by using the traction measurements performed with a two-roller machine. The Eyring stress and shear modulus of the test oils were examined by curve-fitting procedure based on the non-linear viscoelastic model introduced by Johnson and Tevaarwerk. All tests shown that polyethylene in small concentration apparently increases the film thickness, maximum traction coefficient and scuffing resistance.

Keywords: lubricant oils, polymer additives, rheological characteristics, EHD traction

1 INTRODUCTION
The primary functions of a liquid lubricant are to control friction, wear and surface damage over the intended life of a system that contains machine elements. The lubricant choice depends on the tribological system, cost, as well as on health, safety and environmental conditions. Lubricant properties depend on the base oil (or blends of base oils) and on the used additives.

The polymers represent an important category of additives incorporated into the base oils [1], increasing the viscosity and therefore the lubricant film thickness [2]. The polymer additives can be also introduced as extreme pressure agents, oxidation or corrosion inhibitors, pour-point depressants, etc. [1]. The macromolecular chains dispersed into the oil adopt a progressively more open molecular conformation as temperature increases, so that it makes a larger contribution to the overall viscosity of the blend at high than at low temperatures. On the other hand, at very high shear rates, which are present in rubbing components (bearings, cams, pistons, etc.), polymers can exhibit a shear thinning behaviour. As a consequence, the additived oils form a much thinner hydrodynamic films than predicted from the low shear rate viscosities of their blends.

The problems of fluid film lubrication and wear were previously investigated for a series of base oils additived with polyethylene [2, 3]. We observed that for each polymer – lubricant oil system there is an optimum value of polymer concentration that assures a maximum film thickness and a minimum wear in a tribomechanical system. This concentration belongs to the dilute regime, below the critical concentration at which the polymer coils begin to overlap each other [2]. In this paper, the problems of fluid film lubrication for polymer-thickened lubricants are discussed by following their performances in some operating conditions in a mechanical system, i.e. in a tapered roller bearing.

2 TRIBOLOGICAL CONSIDERATIONS
There is a growing interest in the lubrication of mechanical systems subjected to severe operating conditions of load, speed and temperature. The most advanced engineering applications require high reliability and precision (such as: aerospace, military, gas turbine engine, automobile industry, etc). In these domains of activities, the safety of different machines depends on the reliability of elementary mechanisms that are incorporated (Figure 1): (a) rolling bearings, (b) cam - follower systems, (c) gears transmissions and (d) journal bearings.

The primary function of the gears is to transmit the power and motion from one shaft to another. The friction energy, which is dissipated as heat in the gear materials, is added to the output energy to determine the total energy. The sliding action also causes the gear tooth to wear. Gear tooth wear results in a change in the motion transfer characteristics from the design conditions. Thus, one criterion in the system design is to choose materials and environments to reduce friction and wear.

The cam and follower is another system in which motion and power transfer are from one shaft to another. The sliding and rolling bearings are devices that locate a rotating shaft with respect to a non-rotating support. The
material loss by wear can contribute to the increasing of
the internal clearance determining the loss in stability
and precision and thus the level of vibrations and noise
become higher. In these conditions, the bearing is
considered failed.

The lubricant film (having the thickness denoted by "h",
see Figure 1) transmits the loading from one surface to
other.

In these tribological systems, the base oils are not
always able to separate totally the contact surfaces. The
additives to base oils are required to enhance the lubricity by forming different sorts of protective films
on the rubbing surfaces, adsorbed films, interaction
films or chemically deposited films.

Literature shows that the polymer-thickened lubricants
have been used since the mid-1950s for increasing the
viscosity and lubricant film thickness [4]. As oil-in-oil
eumasions or polymer-in-oil dispersions, they have
demonstrated highly efficiency as viscosity index
improvers (VII) with a view to transport, storage and
processing. The most popular polymers used as VII are
the polymethacrylates, olefin polymers and copolymers
and hydrogenated styrene diene copolymers. A new
variant as concentrated mixed polymers was also lanced
on the market [5].

Generally, the chemical structure of the polymers used,
as additives are very closed to the base oils, for
obtaining a well dispersed lubricating system.

3 MATERIALS

Three mineral oils - BO1, BO2 and BO3 - were used as
base oils (Table 1), containing mainly paraffins and a
low content of iso-paraffins, cycloparaffins or
aromatics. As additive for the base oils was used low-
density polyethylene (PE) with a branched structure.

The additivation of the mineral oils with polyethylene
was realised by mixing the polymer solid particles with
the base oils under continuous magnetic stirring at
60°C. The maximum packing fraction was attained
around 1% PE for BO1, 3% PE for BO3 and 5% PE for
BO2 (Table 1). The polymer additive and the base oils
are completely miscible in the range of studied
temperature.

4 VISCOSITY-TEMPERATURE AND
VISCOITY-PRESSURE
RELATIONSHIPS

The variation of the lubricant viscosity as a function of
the temperature can be expressed by the Roelands’
relationship:

\[ \log[\log(\eta_T + 4.2)] = \log A - B \cdot \log \left( 1 + \frac{T}{135} \right) \]  
(1)

where \( \eta_T \) is lubricant absolute viscosity (Pa·s) at
temperature T (°C), A and B are constants determined
starting from the viscosity values obtained at different
temperatures.

The influence of pressure upon lubricant viscosity is
given by Barus exponential low:

\[ \eta = \eta_T \cdot \exp(\alpha p) \]  
(2)

where \( \alpha \) is the viscosity-pressure index (Pa⁻¹) and it is
assumed to be of 1.15 \times 10^{-8} \text{ Pa}^{-1}.

5 RESULTS AND DISCUSSION

5.1 Tribological behaviour

The film-forming properties were analyzed on the
tapered roller bearing No. 30206. During the tests, the
bearing was subjected to an axial load of 450N and the
rotational speed was of 2030rev/min. The minimum
film thickness was determined by electrical resistance
method and using the calibration curves for each
concentration of PE in base oil [2]. Figure 2a gives the
dependence of minimum film thickness at the roller-
raceway contact versus PE concentration in the BO1
base oil at different temperatures. The decrease of the
film thickness with increasing polymer concentration
above 0.5% can be attributed to the entanglement
formation. Concentration values of 0.1% and 0.5%
corresponds to the dilute regime, while the
concentration of 1% belongs to semi dilute regime. In
the dilute regime, some macromolecular coils dispersed
into the oil are trapped on the surface leading to a rough
layer of polymer with a thickness corresponding to one
coil. When entanglements are formed, it is more
difficult to achieve adsorption on the solid surface.

To analysis the effect of polymer concentration on
scuffing resistance a classical four-balls machine was
used. The balls diameter of test is of 12mm and its were
made of AISI 52100steel; the balls surface roughness
was 0.04μm; the upper ball was rotated with 1500rpm
and the axial load of 450N generated o Hertzian
pressure of 2.28GPa. During the experimental tests the
minimum lubricant film thickness between balls was of
0.018μm. In these conditions, Figure 2b gives the wear
path diameter as a function of PE concentration in BO2.
The smallest wear path on the balls was found with
polymer concentration of 1%. Around this concentration
scuffing probability is minimum, and 1%PE can be
considered as optimum concentration value for this
lubricant system. The beneficial effect of polyethylene
on scuffing resistance is attributed to the formation of
boundary viscous films on the contacting surface that
resist to the contact pressure and the sliding speed.

The polymer action upon the results presented in
Figures 2 can be explained according to lubrication
regime developed. As a function of the film thickness
magnitude, the following lubrication regimes can be
defined (Figure 3): (a) thick films (h ≥ 100 nm) and (b)
thin films (h < 100 nm). Thick-film lubrication refers to
the total separation of asperities by a lubricant film
thickness many times larger than the size of the
lubricant molecules. In this case the polymer used plays
the role of IV-improver viscosity. In the case of thin-
film lubrication (such us developed during the scuffing
tests, Figure 2b), the order of magnitude of film
thickness is greater than the diameter of the average
polymer molecule; the critical film thickness is
comparable with the diameter of polymer coils. Using a
force balance microscope, it was shown [6-8] that
polymers could be adsorbed on solid surface to form immobile viscous layers. Thus, low molecular weight polymers can form surface layers more than 10 nm thick. The viscosity of these adsorbed layers is much higher than the bulk solution. The polymer film formed between the solid surfaces prevents the scuffing phenomenon.

5.2 Tractional and Rheological Behaviour in EHD Contacts

The conventional way of studying the traction behaviour in EHL lubrication experimentally is to use a rolling-sliding two-discs machine apparatus, which combines rolling and sliding action found in practical applications. The resultant force of the shearing action in lubricant is the traction force that can be measured under different operating conditions of load, speed and temperature. The discs were made of AISI 52100 steel, the discs hardness is of 60-62 Rc, and the surface roughness is of less than 0.05μm C.L.A.

The influence of the polymer concentration in BO3 on the traction behaviour at 30°C is presented in Figure 4. The applied load generates a Hertzian pressure of 1.75GPa and the rolling velocity is of 5m/s. By increasing the polymer concentration from 0.0% to 2.0%, one obtains an increase of the traction coefficient. The results from Figure 4 show that the polymer used as additive (polyethylene) contributes to the increasing of traction coefficient with increasing the polymer concentration from 0.0% to 2.0%, one obtains an increase of the traction coefficient.

The viscosity of these adsorbed layers is much higher than the bulk solution. Thus, some useful practical data can’t be obtained from rolling/sliding contact experiments. Thus, its value may be determined most convenient for the linear slope of a simple traction-slide/rolling ratio.

5.2.1 Newtonian limit stress \( \tau_o \)

The methodology to predict shear stress \( \tau_o \) is based on the cognition of traction curve. In the linear region of the traction curve, the elastic effects can be neglected and for \( \tau \geq \tau_o \) the rheological model of Johnson and Tevaarwerk [9] (Eq. (3)), can be written as:

\[
\tau = \tau_o \ln \left( \frac{2\eta \gamma}{\tau_o} \right)
\]

Finally, the Newtonian limit stress was found to be:

\[
\tau_o = 2\eta \dot{\gamma}_o.
\]

5.2.2 Lubricant shear modulus \( G \)

Actually, reliable independent measurements of the shear modulus \( G \) of traction fluid are not available, so some useful practical data can’t be obtained from rolling/sliding contact experiments. Thus, its value may be determined most convenient for the linear slope of a simple traction-slide/rolling ratio.

\[
G = \frac{pB^*h_{cen}}{2 \ln \left( 1 - \frac{pB^*h_{cen}}{U} \right)^{-1}}
\]

Johnson and Cameron [10] have demonstrated that, in linear region of the traction curve, where the sliding velocity is very small, it is possible to extract the apparent viscosity.

\[
\eta_{app} = \frac{h_{cen} \cdot p}{U}
\]

At small deformations, the force required to extend the polymer chain is linearly proportional to the amount of extension, i.e., the polymer acts according to the Hooke’s law. However, as deformations become larger, the required forces increase exponentially. A simple conceptual model can be used to explain the lubricant behaviour passing through the Hertzian contact (Figure 6). The extendable non-linear elastic spring–bead model consists of two rigid spheres (accounting for hydrodynamic forces acting on the polymer molecules) connected by an unbendable spring that simulates elastic forces acting on the polymer chain. The solvated polymer molecules are able to impart elasticity to the bulk fluid (base oil) when the polymer molecules are deformed. After the deformation, the polymer relaxes to its previous coiled conformation with the energy dissipated as heat to the surrounding fluid.

6 CONCLUSIONS

In this paper, lubricating oils containing different concentrations of polyethylene were examined in different tribological systems. The minimum film thickness, wear resistance and traction coefficient were examined. From tribological point of view, the conditions of optimum (resistance to wear and separation) depend both on the polymer concentration and type of the fluid used as base oil. The rheological properties of the lubricant were correlated with the traction behaviour. A semi-empirical method was briefly described to calculate the rheological parameters.
in EHD contacts. Thus, the Eyring stress and shear modulus of the test oils were examined by curve-fitting procedure based on the non-linear viscoelastic model introduced by Johnson and Tevaarwerk.

All tests shown that polyethylene in small concentration apparently contributes to increasing the film thickness, maximum traction coefficient and scuffing resistance of the concentrated contacts.

7 ACKNOWLEDGMENT
The authors wish to thank Dr. Fabrice Ville and Dipl. Viorel Paleu for their help to perform the traction experiments in the Laboratory of Contact Mechanics, INSA Lyon (France)

8 REFERENCES

9 NOMENCLATURE
b = half width of an elliptical contact in the rolling direction, m
\( B' \) = slope of traction curve in the linear region
G = elastic shear modulus of lubricant, N/m²
\( h_{min} \) = thermally minimum film thickness, m
N = normal load contact, N
\( \bar{p} \) = mean Hertzian pressure, N/m²
\( p_{max} \) = maximum Hertzian pressure, N/m²
T = lubricant temperature, °C
U = rolling velocity = (U_2+U_1)/2, m/s
\( \Delta U \) = sliding velocity = U_1-U_2, m/s
\( \alpha = \text{pressure-viscosity coefficient, Pa}^{-1} \)
\( \dot{\gamma} = \text{shear rate, s}^{-1} \)
\( \eta = \text{lubricant absolute viscosity, Pa\cdot s} \)
\( \lambda = \text{roughness parameter} = h_{min}/\sigma \)
\( \mu = \text{traction coefficient} \)
\( \tau = \text{lubricant shear stress, Pa} \)
\( \sigma = \text{the composite roughness of the contacting surfaces, m} \)
\( \tau_0 = \text{Newtonian limit shear stress of lubricant, Pa} \)
(\( \xi = \text{slide - to - roll ratio} = \Delta U/U \)

Table 1: Physical properties of the base oils

<table>
<thead>
<tr>
<th>Parameter</th>
<th>BO1</th>
<th>BO2</th>
<th>BO3</th>
</tr>
</thead>
<tbody>
<tr>
<td>density @ 20°C (g/cm³)</td>
<td>0.910</td>
<td>0.850</td>
<td>0.890</td>
</tr>
<tr>
<td>dynamic viscosity (Pa\cdot s)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>@ 40°C</td>
<td>0.16</td>
<td>0.060</td>
<td>0.081</td>
</tr>
<tr>
<td>@ 100°C</td>
<td>0.015</td>
<td>0.003</td>
<td>0.008</td>
</tr>
<tr>
<td>Flash point (°C)</td>
<td>210</td>
<td>65</td>
<td>140</td>
</tr>
<tr>
<td>Pour point (°C)</td>
<td>-20</td>
<td>-18</td>
<td>-19</td>
</tr>
<tr>
<td>Maximum concentration in PE (% wt)</td>
<td>1.0</td>
<td>5.0</td>
<td>3.0</td>
</tr>
</tbody>
</table>
Figure 1: Elementary mechanisms used in current applications of mechanical engineering

Figure 2: Tribological behaviour for polyethylene blended mineral oil: (a) minimum film thickness vs. concentration of polyethylene in BO1 oil at different temperatures and (b) wear surfaces diameter measured on four-ball machine versus polymer concentration of the suspensions of PE in BO2 (at 25°C)

Figure 3: Typical lubricant flow through a rolling Hertzian conjunction: (a) - thick-film lubrication and (b) - thin-film lubrication
Figure 4: Traction curves obtained for $p_{\text{max}} = 1.75$ GPa at 30°C: (●) base oil; (◊) 0.5% PE; (x) 1.0% PE; ( ) 1.5% PE and (▲) 2.0% PE.

Figure 5: Shear modulus and Eyring shear stress vs. polymer concentration for $p_{\text{max}} = 1.75$ GPa at 30°C.

Figure 6: Response of polymer to fluid break-up.