

## A Rheological Model Based on Primary Laboratory Data of Lubricants with Polymers Additives

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**Summary:** The present work reports some experimental data consisting in traction behavior for lubricants with polymer additives. The rheological response of lubricants without polymer additives and with additives as a function of polymer concentration, pressure and temperature is discussed. Based on the laboratory data, the paper presents a quasi-empirical approach which predicts elastic shear modulus, limiting shear stress and characteristic shear stress of a lubricant with polymer additives.

### Introduction

There is an increasing interest that polymer systems, in particular, and a number of other rheological fluids, in general, can be characterized in different flow conditions by considering a reliable constitutive equation. Sometimes, it is difficult to obtain complete information concerning the rheological behavior in some severe conditions of exploitation of lubricants in engineering applications.

The polymers are incorporated as additives into the base oils, increasing the viscosity and therefore the lubricant film thickness.<sup>[1-3]</sup> In most cases, the macromolecular chains dispersed into the oil adopt a progressively more open conformation as temperature increases, so that it makes a larger contribution to the overall viscosity of the blend at high versus at low temperatures.

Friction between two surfaces is one of the most important interaction which controls the behavior of mechanical components in a wide range of applications. In concentrated contacts, the relative sliding velocity is superposed on nominal rolling velocity and the resulting friction is often referred to as traction. By using polymers as additives it is possible to modify the traction behavior of the lubricant oils.<sup>[4]</sup> The modelling of the

traction behavior for such systems is a complex problem because of mechanical, thermal and physico-chemical interactions.

The present paper discusses the influence of polymer content on traction behavior of a mineral oil additived with polyethylene at different pressures and temperatures. A semi-empirical approach for determining the effect of the operating conditions (load, temperature and polymer concentration) upon the effective rheological parameters of the lubricant in an elastohydrodynamic contact (EHL) is also presented.

## Experimental

A paraffinic base oil (with the dynamic viscosity of 0.081 Pa·s at 40 °C and 0.008 Pa·s at 100 °C) was additived with low density polyethylene (PE),  $M_w = 1.2 \cdot 10^5$  g/mol. The additivation was realized by mixing the polymer solid particles with the base oil under continuous magnetic stirring at 60 °C. Samples of 0.0 % (base oil) to 2.0 % (the concentration is expressed in grams of polymer dispersed into 100 ml of oil) were investigated in the range of temperature 30 °C - 75 °C.

Rheological measurements were performed by using a rheometer with coaxial cylinders.

Traction behavior of lubricant was studied at different temperatures with a rolling-sliding two-disk machine apparatus<sup>[4]</sup> by which the combined rolling and sliding action found in practical applications can be simulated.

## Primary Laboratory Data Obtained for a Lubricant Oil Additived with Polyethylene

The traction curves are plotted in the form of traction coefficient ( $\mu$ ) versus slide-to-roll ratio ( $\xi = \Delta U/U$ ;  $\Delta U$  and  $U$  are the sliding and rolling velocity, respectively) for different pressures, temperatures and polymer concentrations. The influence of pressure on the EHL traction behavior is exemplified in Figure 1. The presence of the macromolecular additive does not alter this behavior (continuous increase in traction with increasing the pressure).

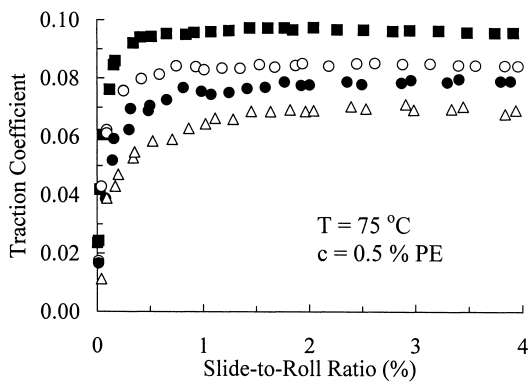


Figure 1. Traction curves obtained for 0.5 % PE at 75 °C and different pressures  $p_{\text{max}}$ : (Δ) 1.00 GPa; (●) 1.25 GPa; (○) 1.50 GPa; (■) 1.75 GPa.

Figure 2 shows the effect of the temperature on the traction behavior for 1.0 % PE and maximum Hertzian pressure ( $p_{\text{max}}$ ) of 1.50 GPa. Also, the presence of the macromolecular additive does not change the typical evolution of the traction coefficient (continuously decreasing when the temperature increases).

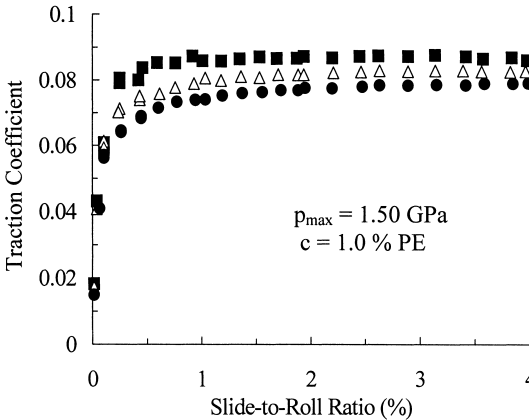


Figure 2. Traction curves obtained for 1.0 % PE ( $p_{\text{max}} = 1.50\text{ GPa}$ ) at different temperatures: (■) 30 °C; (Δ) 50 °C; (●) 75 °C.

The influence of the concentration on the traction curves is presented in Figure 3. By increasing the polymer concentration from 0.0 % to 2.0 %, one obtains an increase of the

traction coefficient. It is difficult to predict the effect of shear thinning of polymer on EHL film formation. A number of explanations have been advanced as to why polymer solutions give thinner EHL films than predicted from their low shear rate viscosities. These include shear thinning of polymer (temporary viscosity loss) in EHL contact inlet, polymer fragmentation (permanent viscosity loss) under EHL conditions and inability of large polymer coils to enter in EHL contacts.<sup>[2,5]</sup>

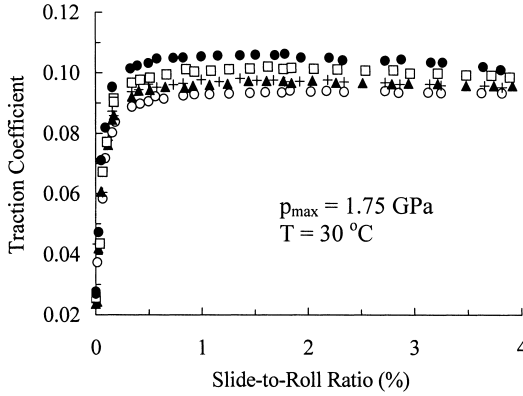


Figure 3. Traction curves obtained for  $p_{\max}=1.75 \text{ GPa}$  at  $30 \text{ }^{\circ}\text{C}$ : (o) base oil; ( $\square$ ) 0.5 % PE; (+) 1.0 % PE; ( $\blacktriangle$ ) 1.5 % PE and ( $\bullet$ ) 2.0 % PE.

The maximum traction coefficient ( $\mu_{\max}$ ) and the slope of the traction versus slip curve are very important parameters. The curve obtained for the base oil used in this study (Figure 3) shows a maximum value for the traction coefficient of 0.085 for  $p_{\max} = 1.75 \text{ GPa}$  at  $30 \text{ }^{\circ}\text{C}$ . The maximum traction coefficient and the slope of the traction depend on polymer concentration.

### Analytical Approach

It is now accepted that the rheological behavior of a lubricant in an EHL contact is described by the Maxwell model, which superposes an elastic shear rate  $\dot{\gamma}_E$  and a viscous shear rate  $\dot{\gamma}_V$ :

$$\dot{\gamma} = \dot{\gamma}_E + \dot{\gamma}_V = \frac{1}{G} \frac{d\tau}{dt} + F(\tau) \quad (1)$$

In the equation (1)  $G$  is the elastic shear modulus of the lubricant,  $\tau$  is the shear stress and

$F(\tau)$  is the dissipative function. Using the theory of Eyring, Johnson and Tevaarwerk<sup>[6,7]</sup> proposed a rheological model with "sinh" law for dissipative function:

$$\dot{\gamma}_{J-T} = \frac{1}{G} \frac{d\tau}{dt} + \frac{\tau_0}{\eta} \sinh\left(\frac{\tau}{\tau_0}\right) \quad (2)$$

where  $\tau_0$  is the Newtonian limit stress and represents the shear stress which characterizes the departure from the linear to non-linear part of traction curve. In accordance with the non-Newtonian fluid model of Bair and Winer,<sup>[8]</sup> the constitutive rheological equation is:

$$F(\tau) = \frac{1}{G} \frac{d\tau}{dt} + \frac{\tau_L}{\eta} \ln\left(1 + \frac{\tau}{\tau_L}\right) \quad (3)$$

where  $\tau_L$  represents the limiting shear stress of lubricant.

For the calculations, the following assumptions are adopted: (i) pressure distribution is the same as for a Hertzian contact; (ii) the shear stress is considered only in the parallel region and the flow caused by pressure gradient is ignored; (iii) oil and roller surface temperatures are constant; (iv) the variation of the lubricant viscosity as a function of the temperature can be expressed by the Roelands' relationship:

$$\log[\log \eta_T + 4.2] = \log K1 - K2 \cdot \log\left[1 + \frac{T}{135}\right] \quad (4)$$

where  $\eta_T$  is lubricant absolute viscosity (Pa·s) at temperature  $T$  (°C),  $K1$  and  $K2$  are constants determined starting from the viscosities obtained for each lubricant at different temperatures; (v) the influence of pressure upon lubricant viscosity is given by:

$$\eta = \eta_T \cdot \exp(\alpha \cdot p) \quad (5)$$

where  $\eta$  represents the lubricant viscosity at a given pressure,  $p$  and  $\alpha$  is the viscosity-pressure index ( $\text{Pa}^{-1}$ ).

The traction data can be correlated with the rheological behavior of the lubricant oil.<sup>[4,9,10]</sup> For the most liquid and some solid lubricants, the general traction behavior is a function of sliding velocity  $\Delta U$  or slide-to roll ratio  $\xi$ , as schematically presented in Figure 4. According to Kragelski,<sup>[11]</sup> this traction behavior can be simulated by:

$$\mu = (A + B \cdot \xi) \exp(-C \cdot \xi) + D \quad (6)$$

where  $A$ ,  $B$ ,  $C$  and  $D$  are the coefficients of the model which depend of operating conditions and are determined from the experimental investigations.

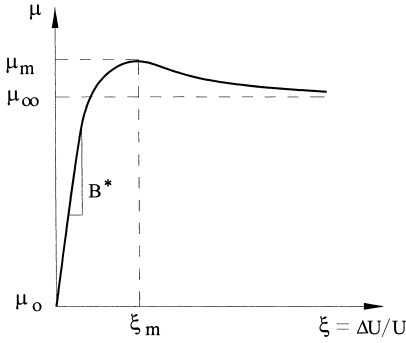


Figure 4. Typical form of a traction

### Limiting Shear Stress

If the operating conditions are confined to large strains and large stress, that is, to eliminate the viscous and elastic effects, it is reasonable to assume that the shear stress reaches the maximum value, namely, the limiting shear stress state, in the whole Hertzian contact zone, which corresponds to the maximum point in the traction curve. Wu and Cheng<sup>[12]</sup> assumed that in comparison with the limiting shear stress distribution to Hertzian contact pressure profile, certain similarities can be drawn.

$$p = p_{\max} \sqrt{1 - \frac{x^2}{a^2} - \frac{y^2}{b^2}} \quad (7)$$

where  $p_{\max}$  is the maximum or Hertzian pressure,  $a$  and  $b$  are the semi-major and semi-minor axis of elliptical contact, respectively. According to Wu and Cheng, the limiting shear stress distribution over the contact area is:

$$\tau_L = \tau_{\max} \left( 1 - \frac{x^2}{a^2} - \frac{y^2}{b^2} \right)^m \quad (8)$$

where  $\tau_{\max}$  is the maximum limiting shear stress in an EHL contact and  $m$  is an index number. The traction force can be determined by integrating the expression above over the contact area:

$$F = \tau_{\max} \frac{\pi ab}{m+1} \quad (9)$$

By combining Eqs. (7)-(9), the limiting shear stress is:

$$\tau_L = (m+1) \frac{F_{\max}}{\pi ab} \left( \frac{2}{3} \right)^{2m} \quad (10)$$

or

$$\tau_L = (m+1) \mu_{\max} \bar{p} \left( \frac{2}{3} \right)^{2m} \quad (11)$$

where  $\mu_{\max}$  is the maximum traction coefficient that can be obtained from traction curves obtained through a two-disc machine test. The average value for the index number  $m$  taken in this study was of 0.75.

### Eyring Shear Stress

For determining the limiting shear stress  $\tau_L$ , the equality between the total shear rate, from Johnson and Tevaarwerk model ( $\dot{\gamma}_{J-T}$ ) and the total shear rate described by Bair and Winer's rheological model ( $\dot{\gamma}_{B-W}$ ) is imposed. The elastic component of shear rate must be equal in these two rheological models. This involves that viscous components must be equals.

$$F(\tau)_{J-T} = F(\tau)_{B-W} \quad (12)$$

or

$$\frac{\tau_0}{\eta} \sinh\left(\frac{\tau}{\tau_0}\right) = \frac{\tau_L}{\eta} \ln\left(1 - \frac{\tau}{\tau_L}\right) \quad (13)$$

Equation (13) must be valid only for  $\tau = \tau_0$  when the following equation of variable  $\chi$  ( $\chi = \tau_0/\tau_L$ ) is obtained:

$$\chi \cdot (e^2 - 1) + 2 \cdot e \cdot \ln(1 - \chi) = 0 \quad (14)$$

The equation (14) shows that the ratio between the Eyring shear stress and limiting shear stress have a constant value of  $\chi = 0.283$ . Therefore, the Eyring shear stress can be easily determined.

$$\tau_0 = \chi \cdot \tau_L \quad (15)$$

### Lubricant Shear Modulus

Independent measurements of the shear modulus ( $G$ ) of the traction fluid are not available, thus, some useful practical data can not be obtained experimentally. Its value may be determined knowing the slope around the origin of the traction curve. In the linear part of lubricant traction curve, the model of Johnson and Tevaarwerk<sup>[10]</sup> is reduced to:

$$\dot{\gamma} = \frac{1}{G} \cdot \frac{d\tau}{dt} \quad (16)$$

The experimental traction curve provides only average shear stress, so one can write:

$$\tau = \mu \cdot \bar{p} \quad (17)$$

Algebraically, the variation in traction of the Newtonian behavior region, may be written as:

$$\mu = B^* \cdot \xi \quad (18)$$

where the constant  $B^*$  represents the slope of the traction curve.

Therefore, the elastic component of the Johnson and Tevaarwerk model becomes:

$$\dot{\gamma} = \frac{1}{G} \cdot \frac{\mu \cdot \bar{p} \cdot U}{b} \quad (19)$$

where  $b$  is half width of disk-to-disk contact in rolling direction.

The limiting shear modulus  $G$  takes a form which depends on traction measurements:

$$G = \frac{\bar{p} \cdot B^* \cdot h_{cen}}{b} \quad (20)$$

The semi-empirical results about lubricant shear modulus ( $G$ ), limiting shear stress ( $\tau_L$ ) and Eyring shear stress ( $\tau_0$ ) are studied in several ways which include: (a) the effect of pressure; (b) the effect of temperature and (c) the effect of polymer concentration.

#### (a) Effect of Pressure

Figure 5 presents the limiting and Eyring shear stresses (a) and the shear modulus (b) as a function of maximum Hertzian pressure at a given temperature and polymer concentration.

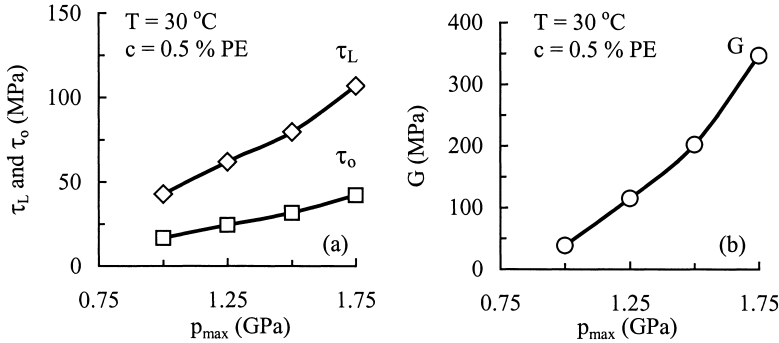


Figure 5. (a) Eyring shear stress ( $\tau_0$ ), limiting shear stress ( $\tau_L$ ) and (b) shear modulus ( $G$ ) vs. maximum Hertzian pressure ( $p_{max}$ ) for 0.5 % PE at 30°C.



One observes that  $G$  and  $\tau_0$  increase nearly linear with the contact pressure. The values obtained for the shear modulus are of the same order of magnitude to those reported in literature for the mineral oils.<sup>[12]</sup>

### (b) Effect of Temperature

Figure 6 shows the evolution of the lubricant shear modulus and Eyring shear stress as a function of temperature for maximum Hertzian pressure of 1.50 GPa and 1.0 % PE. One observes that  $G$  decreases whereas  $\tau_0$  remains almost constant with increasing the temperature.

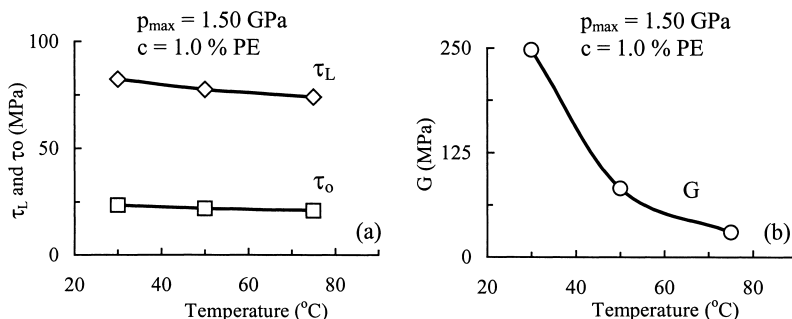


Figure 6. (a) Eyring shear stress ( $\tau_0$ ), limiting shear stress ( $\tau_L$ ) and (b) shear modulus ( $G$ ) vs. temperature for 1.0 % PE,  $p_{\max} = 1.50$  GPa.

### (c) Effect of Polymer Concentration

Figure 7 gives the variation of the lubricant shear modulus and Eyring shear stress as a function of polymer concentration for maximum Hertzian pressure of 1.75 GPa at 30 °C. With increasing the temperature, one observes that  $G$  increases and  $\tau_0$  varies insignificantly. The traction data obtained for the base oil at high value of  $p_{\max}$  (respectively 1.75 GPa in Figure 3) are in agreement with those reported by Muraki and Dong<sup>[13]</sup> for  $p_{\max}$  varying from 0.5 to 0.7 GPa. In this study, a small polymer concentration (up to 2 % PE) increases the maximum traction coefficient of the additived lubricant (Figures 1-3). The high initial slope is due, on the hand, to the presence of the macromolecules into the lubricant oil and, on the other hand, to the high pressure used in our measurements.

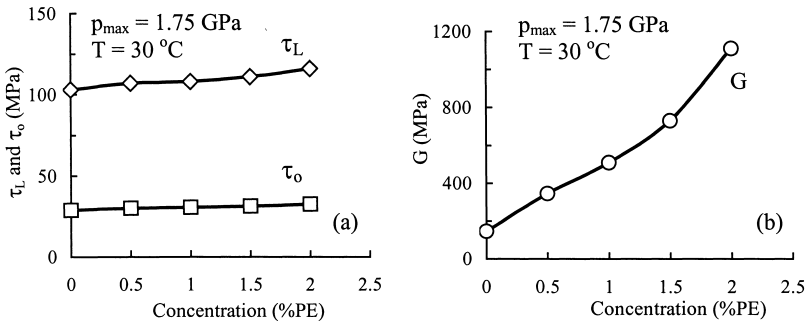


Figure 7. (a) Eyring shear stress, limiting shear stress and (b) shear modulus and vs. polymer concentration for  $p_{\max} = 1.75$  GPa at 30°C.

## Conclusion

The experimental traction data demonstrates that polyethylene can be used as additive to obtain an inexpensive traction fluid. Starting from the experiment, a semi-empirical approach is developed for determining the effective lubricant rheological parameters (the elastic shear modulus, the Eyring shear stress, the limiting shear stress) in an EHL contact.

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