



## EFFECTS OF ETHANOL ON FILM THICKNESS AND FRICTION OF GROUP ONE ENGINE OILS\*

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### Abstract

This paper aims to investigate the effects of a possible incorporation of ethanol to the lubricant during engine use on friction and lubricant film thickness, by adding small amounts of ethanol to Group I base (with and without an organic friction modifier) and to formulated oils. Film thickness was measured using ultrathin film interferometry. Friction tests were carried out using a mini-traction machine to obtain Stribeck curves. The film thickness measurements showed that in the region of low speeds, where the elastohydrodynamic film is very thin and therefore boundary lubrication may prevail, the addition of ethanol produced some boundary film, which was not present for the base oils. For the base oil with an organic friction modifier, the presence of ethanol increased the thickness of the low friction boundary film. For the formulated oil, the addition of ethanol reduced film thickness in all lubrication regimes. The friction tests showed some friction reduction due to addition of ethanol to the base oil, in particular at low speeds. For the formulated oil, the addition of ethanol reduced friction at high speeds, which was associated to a reduction in the viscosity of the lubricant, but at low speeds, the presence of ethanol reduced the formation of a boundary layer, increasing friction. For the base oil containing an organic friction modifier, the presence of ethanol increased the range of speeds for which low boundary friction occurred.

**Keywords:** Ethanol; Lubricant; Engine oil; Film thickness; Stribeck curves.

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## 1 INTRODUCTION

Engines that use any proportion of ethanol or gasoline as fuels, named flex-fuel engines, face today great tribological challenges, which seem to be related to inadequate lubrication and tribochemical wear. Severe wear has been frequently reported by users of different sizes and models of flex-fuel engines.

The use of ethanol as fuel, despite having started in Brazil in the 1930's, was propelled by a national ethanol programme called PROALCOOL, launched by the Brazilian government in 1975, which aimed to implement a Brazilian technology for ethanol production from sugar cane and for adaptation of car engines to use ethanol as fuel. In 1981, the production of ethanol-run automobiles already represented 30% of the Brazilian production, and peaked in 1986 at 96% of the Brazilian production [1]

Brazil was, in the 1970s, the global pioneer in promoting ethanol at a large scale as a vehicle fuel, and is today the second world producer (38.2% of global production and 30.4% of demand in 2008). Brazil stands apart because of the matchless competitiveness of its ethanol as an automobile fuel and reduced emissions over this fuel's life cycle [2].

From 1987, policies adopted by the Brazilian government resulted in reduction of sugar cane production, and therefore in shortages of ethanol throughout the country. Towards the end of the 1980s, Brazil was importing methanol for internal consumption, both in ethanol-run engines and in gasoline-run engines, since 5% of ethanol was added to gasoline in Brazil at that time. The shortage of ethanol in petrol stations resulted in a serious credibility crisis among Brazilian consumers and therefore in a temporary collapse of the PROALCOOL program [3].

At the beginning of the 2000s, facing the difficulties of reliable ethanol supply throughout the year, the Brazilian automobile industry decided to develop engines capable of using gasoline and/or ethanol in any proportion. This range of proportions varies from 22% of anhydrous ethanol (99.5% of pure ethanol and 0.5% water), which is then added to gasoline according to more recent Brazilian legislation, to pure, hydrated ethanol. This new family of engines was named *flex-fuel* engines.

This flexibility is possible due to the use of the electronic injection technology, which was not available in a large scale in the 1980s and 1990s in Brazil. An electronic system monitors the engine to choose the fuel mixture. This system uses the lambda probe signal to monitor the composition of the exhaust gases in order to identify the fuel present in the tank. However, a great deal of additional adaptation is necessary to allow the engine to work properly when ethanol is used. Various changes in flex-fuel engines are associated with tribological problems due to less efficient lubrication, such as changes in piston rings, valves and valve lifters [1].

The proportion of vehicles using flex-fuel engines commercialized in Brazil increased to 85.7% in 2014, compared with 83.3% in April 2013, according to information published by the Brazilian national association of automobile vehicle manufactures (Anfavea) [4]. It is estimated that nowadays half of the amount of combustible used in Brazilian automobiles is renewable [5]. Since the introduction of the flex-fuel engines, the use of fuel hydrated ethanol in Brazil increased by 60% [2].

Hydrated ethyl alcohol fuel, or simply ethanol, is produced in Brazil via sugar cane sugar fermentation. When added to gasoline, it is necessary to use anhydrous ethanol, since the water used in hydrated ethyl alcohol is not soluble in gasoline [1]. Hydrated ethanol produced in Brazil presents high detonation resistance and low energy per volume. Therefore, the use of ethanol fuel allows the use of higher

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compression rates in the engine, but the mixture air/fuel needs to be richer than for gasoline, i.e., higher power and thrust are obtained, but more fuel is used [1]. Despite the large success of the use of flex-fuel engines in Brazil, the use of ethanol fuel represents an enormous tribological challenge both to car manufacturers and to fuel producing companies. Failures frequently reported for components of flex-fuel engines are associated with severe wear, which is still not very well understood [6]. This paper aims to investigate the extent to which the tribological problems frequently reported for flex-fuel engines are related to the effects of ethanol on film formation and friction. To do this, elastohydrodynamic (EHL) film thicknesses were measured for lubricants contaminated with ethanol over a wide range of speeds, to span lubrication regimes ranging from boundary to EHL. These results were complemented by measurement of Stribeck curves, to help understand the mechanisms by which the presence of ethanol can affect friction. In order to separate the interaction of ethanol with the base oil from that with other additives, both base oils and formulated oils without friction modifiers were investigated. In addition, the interaction of ethanol with organic friction modifiers was investigated for blends where an organic friction modifier was dissolved in the base oil.

## 2 MATERIAL AND METHODS

### 2.1 Materials

The effect of the presence of ethanol was investigated when it was added to a base oil, in order to separate the effect of ethanol on the base oil from its interactions with additives present in fully formulated oils. A group I base oil was chosen, since this is the base oil type currently most commonly used in Brazil for the production of lubricants.

The effect of the addition of ethanol was first investigated for a SL group I formulated oil, but without any friction modifiers, in order to separate the interaction of ethanol with the other additives in the oil from the interaction of ethanol with friction modifiers. This oil was called formulated oil SLB.

Finally, the interaction of ethanol with organic friction modifiers was investigated for blends where an organic friction modifier was dissolved in the base oil. This organic friction modifier was called FM01. The amount of FM01 friction modifier added to the base oil (1.2% wt.) followed recommendation from the supplier and the oil samples containing additives were heated at 50°C in a magnetic stirrer until total dissolution occurred.

Viscosities and densities were measured for the oils and their blends with ethanol using a SVM3000 Stabinger viscometer at the three test temperatures of 40°C, 70°C and 100°C. Table 1 shows that the addition of HE reduced viscosity of the Group I and II base oils and of the formulated oil at 40°C and 70°C. At 100°C, it was not possible to measure the viscosity of the mixtures containing ethanol due to the formation of a large number of bubbles in the measurement tube, probably resulting from evaporation of ethanol. AE, when added to the oils, reduced viscosity more than HE.

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**Table 1.** Viscosity measurements for the Group I base oil, where  $\rho$  is the density,  $\eta$  is the dynamic viscosity, VI is the viscosity index, HE is hydrated ethanol, AE is absolute ethanol; FM01 is an organic friction modifier; N.M. is not measurable

Oil	$\rho$ at 15°C (g/cm <sup>3</sup> )	$\eta$ at 40°C (mm <sup>2</sup> /s)	$\eta$ at 70°C (mm <sup>2</sup> /s)	$\eta$ at 100°C (mm <sup>2</sup> /s)	VI
Base	0.874	28.698	10.515	5.092	104.8
Base+2% AE	0.871	23.773	8.7069	N.M.	N.M.
Base+2% HE	0.873	27.218	9.6789	N.M.	N.M.
Base + FM01	0.877	30.258	10.709	5.281	102.9
Base + FM01+2% HE	0.871	25.650	8.130	N.M.	N.M.
Base + FM01+2% AE	0.871	21.51	7.703	N.M.	N.M.
SLB	0.8701	97.16	30.809	13.621	141
SLB+5% HE	0.866	89.237	24.803	N.M.	N.M.
SLB+5%AE	0.865	70.259	20.732	N.M.	N.M.

Although the viscosity of pure ethanol is higher than that of water, it was hypothesized that when both water and ethanol are present the true solubility of the ethanol in hydrocarbon is reduced, resulting in some of the ethanol being present as a microemulsion, thereby having little influence on viscosity [7].

## 2.2. Methods

The rig used to measure film thickness in this work is able to measure film thickness down to 1 nm for different loads, rolling speeds and temperatures in a ball-disc contact. The technique is fully described in [8].

Both ball and disc can be independently driven, but in this work the disc was rotated and drove the ball in nominally pure rolling. 19 mm diameter, AISI 52100 steel balls of root mean square surface roughness ( $R_q$ ) between 10 and 13 nm were used. The load was 20 N, which produces a maximum Hertzian contact pressure of 0.52 GPa. A new steel ball was used for each test. Disc and test chamber were thoroughly rinsed using Analar toluene followed by Analar isopropanol prior to each test.

One test was carried out at 40°C, followed by a test at 100°C within the same test sequence, *i.e.*, the balls and disk were not cleaned between the tests at 40°C and 100°C, and the lubricant sample was not changed. At least three repetitions were carried out for all the conditions tested in this work.

Friction tests were carried out using the MTM rig to obtain Stribeck curves. Stribeck curves for the base oil and mixtures containing HE and AE were obtained, where friction is measured over a range of entrainment speeds (mean rolling speed) for a fixed slide roll ratio (*SRR*). To achieve this, the ball and disc are driven by two independent DC motors. The disc is fully-immersed in lubricant and the temperature of the lubricant and contact are controlled to a set value within  $\pm 0.5$  °C [9]. *SRR*, which in our tests was held constant at a value of 50%, is defined as  $(UB-UD)/U$  where  $UB-UD$  is the sliding speed and  $U$  is the entrainment or mean rolling speed, given by  $U = (UB+UD)/2$ . The tests started at a mean rolling speed of 1000 mm/s, which was continuously reduced (31 values logarithmically spaced) down to 1 mm/s. The balls were 19 mm AISI 52100 bearing steel with Young's modulus of 210 GPa and  $R_q$  between 10 and 13 nm and the discs were AISI 52100 discs with  $R_q$  between 25 and 30 nm. The load applied was 20 N, corresponding to a maximum Hertz pressure of 0.82 GPa. Fresh balls and disks were used in each test and were ultrasonically cleaned in toluene followed by acetone prior to a test. The rig was cleaned using toluene followed by Analar isopropanol before each test. The choice of

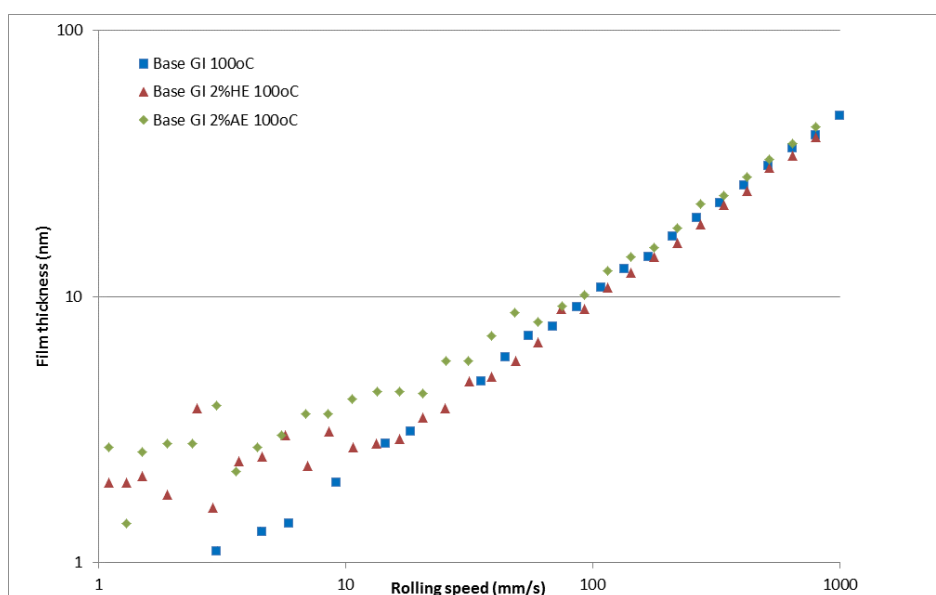
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temperatures for the tests tried to cover a range that was relevant to car engines, but that could still allow some ethanol to be retained in the lubricant.

After the tests, the amount of ethanol present in the lubricant was measured using refractive index measured of a sample of distilled water used to extract ethanol from the lubricant. This new technique has been proposed by Costa and Spikes and is fully described in [7]. For the EHD tests there was some loss of ethanol at 100°C but more than half of the ethanol content remained to the end of the tests. This is because the EHD test apparatus had a good seal and the volume of oil used during the tests was relatively large (around 150 ml). The reduction in the amount of ethanol due to evaporation during the MTM tests at 100°C was much higher than for the EHD tests, so that the final amount was below 0.5% wt.

### 3 RESULTS AND DISCUSSION

Figure 1 analyses the effect of adding 2% of hydrated ethanol (HE) to a group I base oil at 100°C on film thickness for a large range of entrainment speeds. The base oil shows an approximately linear relationship between log (film thickness) and log (entrainment speed) for the whole range of speeds tested, in accord with EHD theory. Both AE and HE generate a thin boundary film, between 2 and 5 nm thick, which is not present with the ethanol-free base oil.

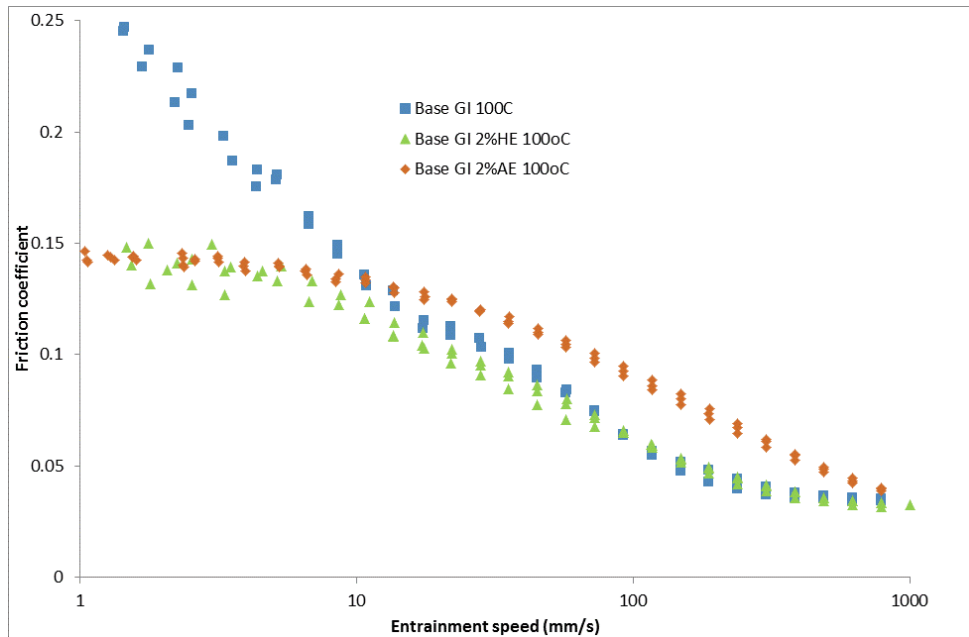


**Figure 1.** Effect of addition of hydrated ethanol (HE) and anhydrous ethanol (AE) on film thickness for Group I base oil at 100°C.

Stribeck curves obtained at 100°C for the base oil are shown in Figure 2. At high speeds, the curve for the base oil approaches a plateau, corresponding to full film EHD lubrication. As the speed is reduced, friction coefficient increased, indicating mixed lubrication. At high speeds, the addition of HE has no effect on friction coefficient. For the addition of AE, a plateau was no longer detected for the highest speeds. It appears that the transition from full film EHD lubrication to mixed lubrication occurs at a higher speed for the blend with AE than for the base oil or the HE blend. This probably originates from the lower viscosity of the AE blend, which leads to thinner films and therefore more asperity contact. At low speeds, the formation of a boundary film that stabilizes friction coefficient at around 0.15 is very clear for both HE and AE, and is not present for the base oil.

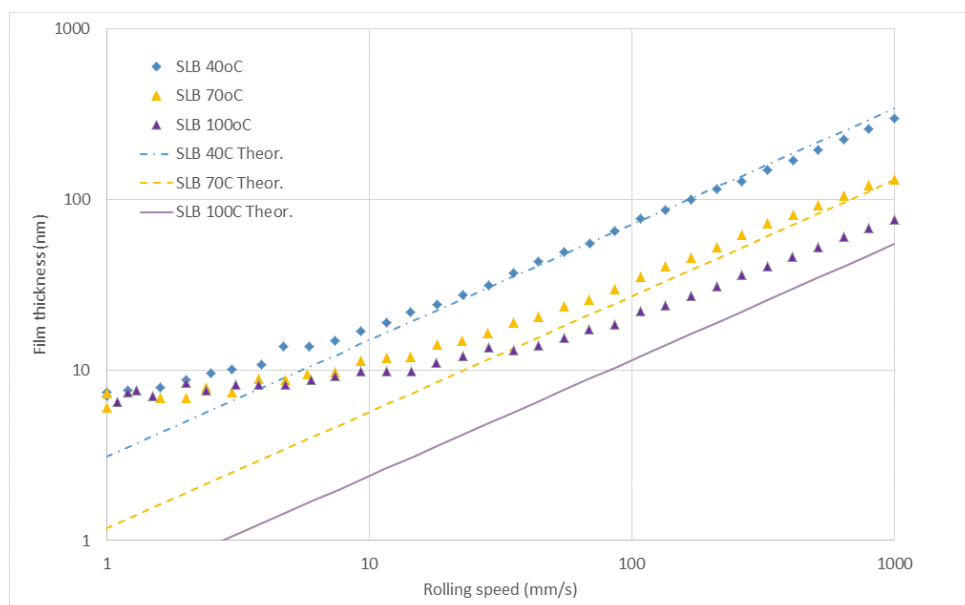
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**Figure 2.** Effect of ethanol on Stribeck curves for Group I base oil, 100°C.

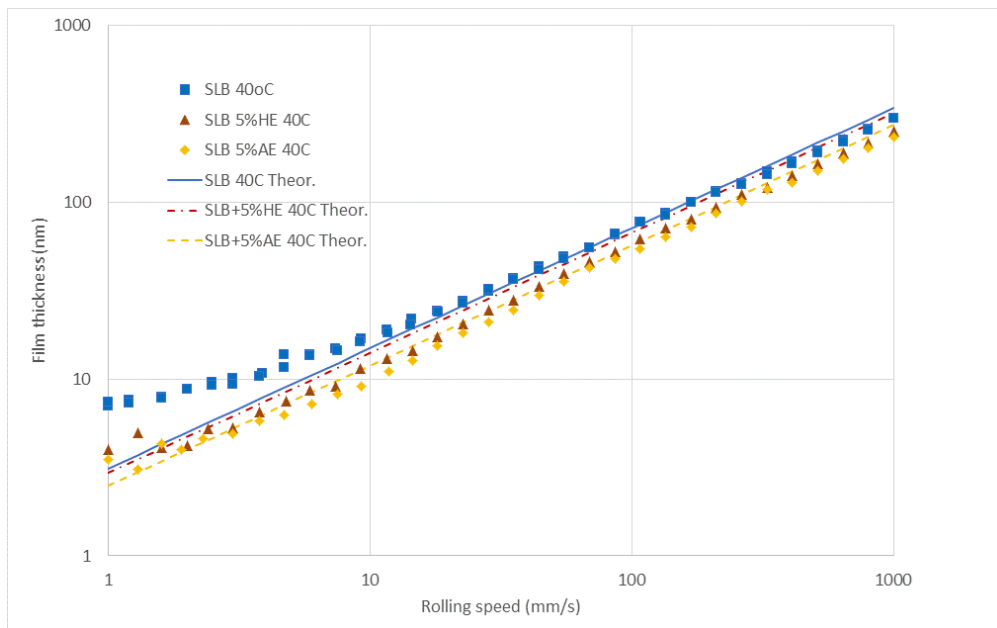
For the formulated oil, film thickness measurements at temperatures of 40°C, 70°C and 100°C are shown initially, in order to understand the behaviour of the formulated oil without ethanol contamination (Figure 3). This figure also shows plots of the predicted film thickness for each temperature based on the Hamrock and Dowson equation for an EHL elliptical contact [10]. The values for the pressure-viscosity coefficients were estimated from [11], while  $E_{steel} = 203$  GPa,  $E_{silica} = 72$  GPa,  $\nu_{steel} = 0.3$ ,  $\nu_{silica} = 0.2$  [9]. The experimental results suggest the formation of a boundary film of thickness *ca* 8-9 nm at all temperatures tested. Comparison with the theoretical curves shows that at 40°C the measured values agree well with the predictions for the higher speeds. When the temperature decreases and therefore film thickness decreases, the measured values are higher than the predicted, but the difference reduces as the speed increases and remains approximately 10 nm. This suggests that the boundary film remains on the sliding surfaces at relatively high speeds.



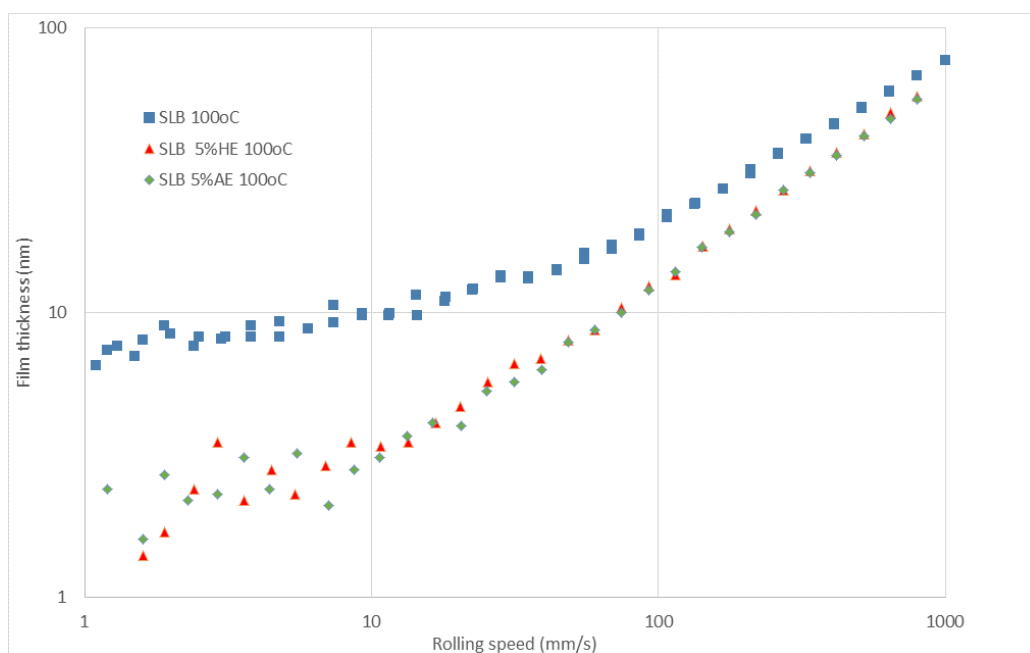
**Figure 3.** Film thickness measurements for the formulated oil SLB at 40°C, 70°C and 100°C.

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Figure 4 shows that the addition of ethanol reduces film thickness in the mixed and full film lubrication regimes, which is slightly larger for AE than for HE, which can be explained by the effects of ethanol on viscosity (Table 1). In the boundary regime, both AE and HE reduce the thickness of the boundary layer. Comparison of the measured values with predicted EHD film thicknesses using the measured viscosities suggests that when ethanol is added, the measured values only deviate from a straight line at very low speeds, below 3 mm/s, where a thin boundary film of around 3 to 5 nm is seen, similar to the behaviour found for the base oil. These results suggest that the presence of ethanol inhibits the formation of the thick boundary film otherwise formed for the formulated oil, but still allows a thin boundary layer. At 100°C (Figure 5), both AE and HE reduce film thickness for the whole range of speeds investigated, but no difference was detected between HE and AE.



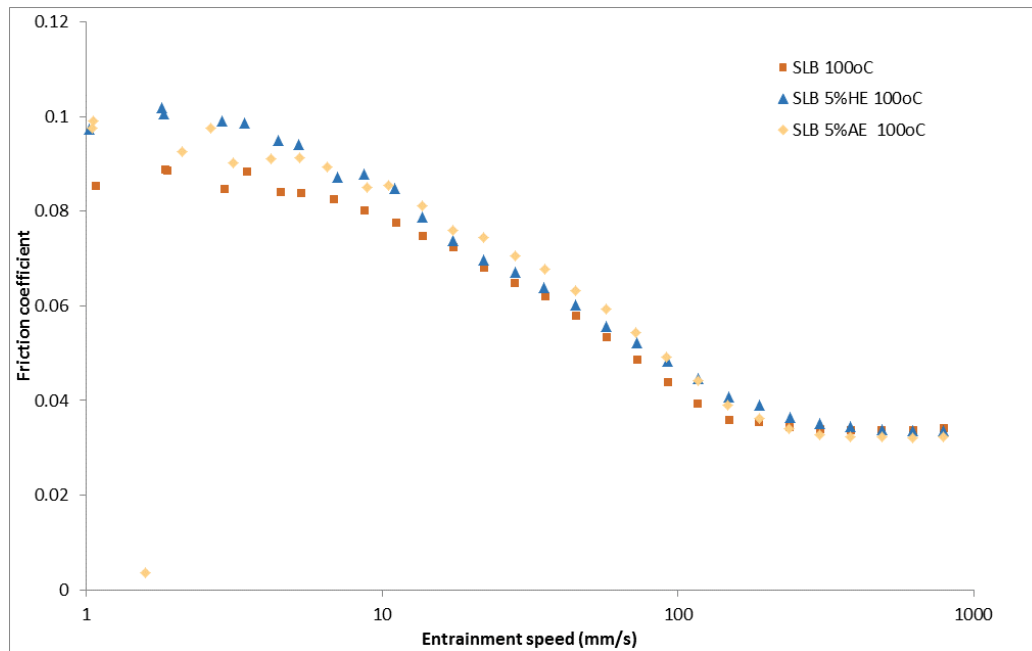
**Figure 4.** Effect of addition of ethanol to formulated oil SLB on film thickness at 40°C.



**Figure 5.** Effect of addition of ethanol to formulated oil SLB on film thickness at 100°C.

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The effects of ethanol contamination on friction for the formulated oils were similar to those observed for the base oils under conditions where full film or mixed lubrication prevailed, but different in the boundary lubrication regime. At 100°C (Figure 6), in the region of mixed lubrication, the addition of ethanol increases friction and the increase is slightly higher for AE than for HE. This can be attributed to the reduction in viscosity caused by the presence of AE and HE, because thinner films enable more metal contact. Both AE and HE appear to suppress some boundary film formation, slightly increasing friction coefficient.

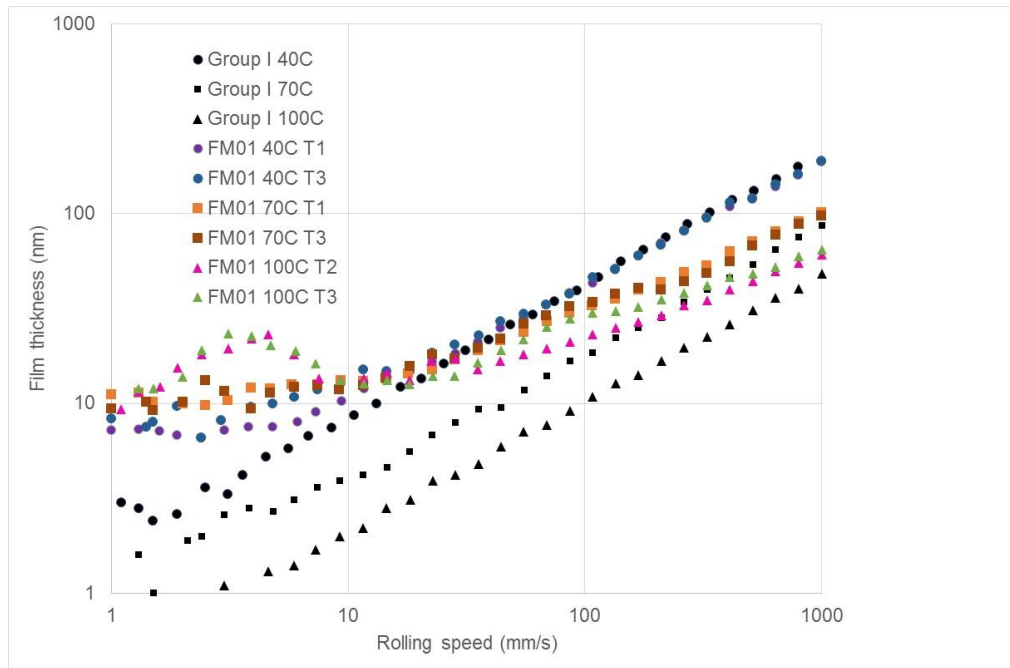


**Figure 6.** Effect of ethanol on Stribeck curves for the formulated oil SLB, 100°C.

The behaviour of the base oil with a dissolved organic friction modifier is shown in Figure 7. It is clear that relatively thick boundary films are formed at all temperatures tested. For comparison, measurements for the base oil at the different test temperatures are also presented. At 40°C, the boundary film is slightly thinner than for 70°C for the lowest speeds. Some authors have suggested that weaker adsorption of organic friction modifiers can occur as the temperature is reduced [12]. For the region of high speeds, where full film lubrication occurs, there is no difference between the base oil and the oil with additive at 40°C. This suggests that any adsorbed film is completely removed at high speeds for this temperature. On the other hand, for 70°C, the film thickness is larger for the oil with additive even at high speeds. For 100°C, the film thickness in the boundary region starts at similar values for those measured for 70°C at the lowest speeds. However, the boundary film thickness increases to around 20 nm with speed and then decreases again. The reason why boundary film thickness initially increases but then decreases with sliding speed for 100°C is unclear. In the region of high speeds, the film thickness remains substantially larger than that measured for the base oil.

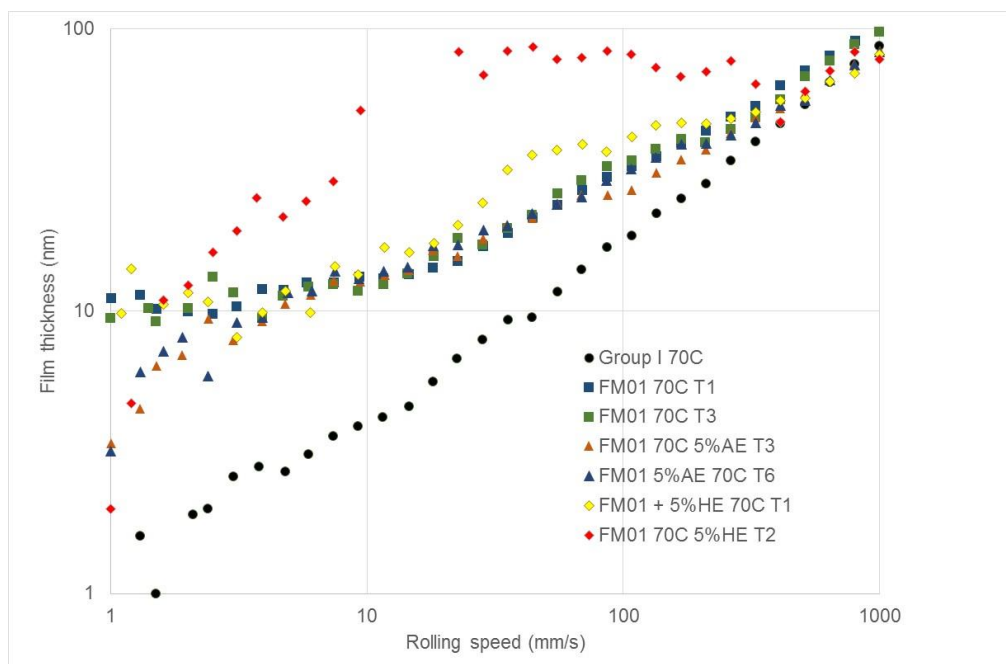
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**Figure 7.** Film thickness measurements for the addition of FM01 to the base oil at different temperatures.

The effect of the addition of ethanol to the base oil containing FM01 is exemplified for the tests at 70°C (Figure 8). The behaviour of the contaminated oils seems quite complex. When AE is added, the thickness of the boundary film is reduced for very low speeds, but it increases with speed. This might suggest the occurrence of a viscous boundary film whose structure is modified by the presence of ethanol. When HE was added, significantly thicker films were measured. However, the film thickness varied largely, both within one test and between different tests. This suggests that the presence of water interferes strongly with the structure of the boundary film formed. At the highest speeds, this very thick film seems to disappear.

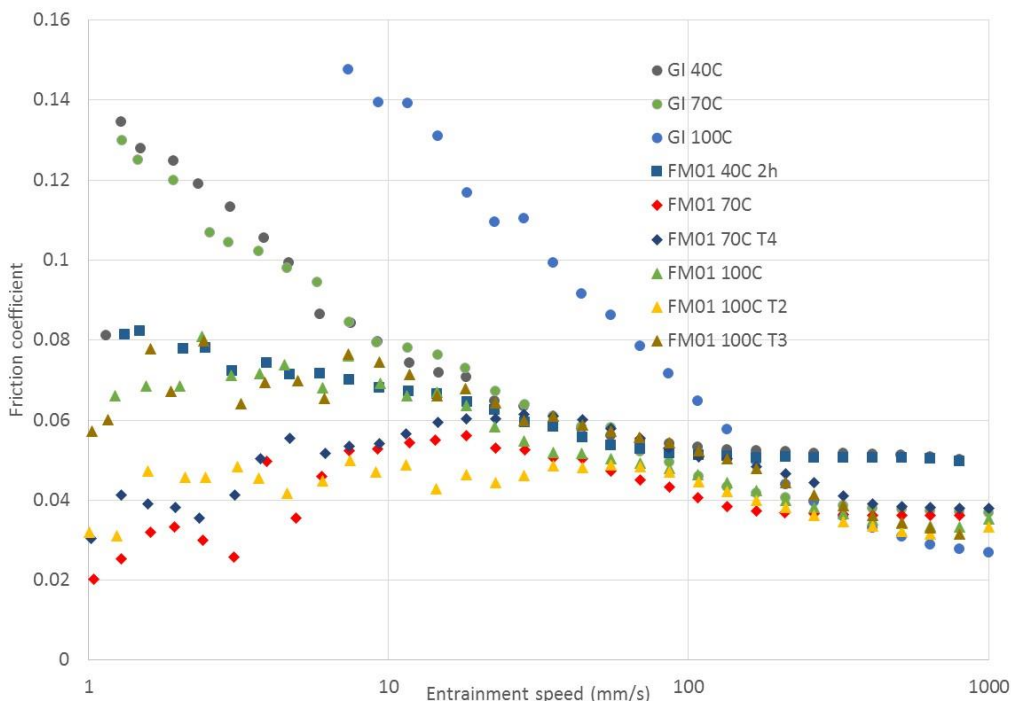


**Figure 8.** Effect of addition of ethanol to mixtures of Group I base oil + 1.2%FM01 on film thickness at 70°C.

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Figure 9 shows Stribeck curves for the Group I base oil containing FM01 at different temperatures. For comparison, Stribeck curves for the base oil are also presented. Since organic friction modifiers are expected to operate via adsorption of surfactants, which is diffusion controlled, similar tests were carried out leaving the samples immersed in oil after stabilization of the test temperatures during both 10 minutes and 2 hours before the Stribeck curve was obtained, but no significant difference was observed. This figure shows some friction reduction at 40°C due to the addition of the friction modifier and friction coefficients around 0.08 are achieved in the low speed regime. For temperatures of 70°C and 100°C, friction coefficients dropped to around 0.06 or lower, but this varied between repetitions for the same test temperature. At high speeds, the friction coefficient restores to the values that was obtained for the base oil for the tests at 40°C and 70°C, suggesting removal of the boundary film. For the tests at 100°C, friction was slightly higher for the mixtures containing additive in the region of high speeds.



**Figure 9.** Stribeck curves for Group I base oil + FM01 at 40°C, 70°C and 100°C.

The effect of ethanol contamination on the base oil containing an organic friction modifier is depicted for tests at 70°C (Figure 10). The addition of both HE and AE reduced friction significantly in the region of intermediate speeds, suggesting an increase in the range of speeds for which a boundary film still exists on the surfaces. For the lowest speeds, all blends showed very low friction, independent of the presence of ethanol.

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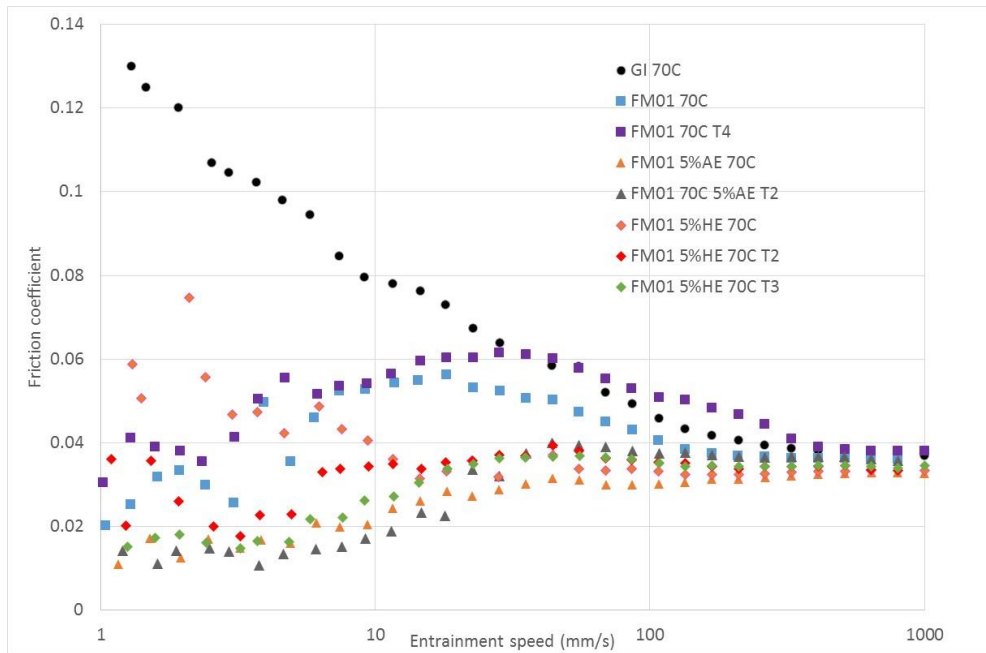


Figure 1. Effect of ethanol addition on Stribeck curves for Group I + OFM at 70°C.

When ethanol is added to the base oil and to the formulated oil, it reduces friction at high speeds in the full film EHD lubrication regime characterised by the friction levelling out at a relatively low value ( $\mu_L$ ) [13, 14]. Under these conditions, friction depends only on the molecular nature of the lubricant and its free volume. In general, the reduction in  $\mu_L$  was found to be higher for AE than for HE, while measurements also showed lower viscosities for blends with AE than with HE. This suggests that possibly the addition of ethanol confers a greater free volume, enabling lubricant molecules slide more easily in relation to each other, reducing both viscosity and  $\mu_L$  and that this effect is more significant for AE than for HE.

In the MTM tests, as speed and thus EHD film thickness is reduced, friction increased for the fluids without friction modifiers, as the contact enters the mixed lubrication regime. In this region, the addition of ethanol increases friction coefficient compared to the ethanol-free fluids, both for the base oil and for the formulated oil. Also, a slight reduction in film thickness in pure rolling was detected due to the addition of ethanol to the base oils and to the formulated oil for tests conducted at lower temperature and higher rolling speeds. This film thickness reduction seems then to be related to the reduction in the viscosity of the lubricant due to the presence of ethanol, which was more significant for AE than for HE. Thinner films may cause more contact between the asperities of the surfaces, which may explain the increase in friction coefficient observed, in particular, with AE.

At very low speeds, especially when combined with higher temperatures and therefore very thin EHL films, the presence of ethanol seems to promote the formation of a boundary layer that is not present for the ethanol-free base oils under pure rolling. Also, Stribeck curves showed a small reduction of friction in the regions of very thin EHL film thickness when ethanol is present in the base oil, also suggesting the formation of a boundary film. Contrarily, when present in the formulated oil, ethanol reduces the thickness of the boundary film formed under pure rolling and increases friction in the boundary lubrication regime.

For the base oils, one possible explanation of the formation of a boundary layer due to the addition of ethanol could be the fractionation of ethanol to the metallic

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surfaces, due to its higher polarity when compared with the base oil lubricant. However, since ethanol is less viscous than the oil, the boundary layer should be thinner. Therefore, the hypothesis of fractionation of ethanol molecules to the metal surface due to their higher polarity must be discarded. Another possibility could be the occurrence of oxidation reactions of the ethanol when in contact with the hot metal surface. This seems to be the case, as reported in [7].

For the ethanol-free formulated oil, both the film thickness measurements under pure rolling and the Stribeck curves suggested the formation of a quite thick boundary film. Chemical composition of the oil showed the presence of Zn, S and P and Ca. It was suggested in [7] that this film results from the formation of a  $\text{CaCO}_3$  film from an overbased detergent, as proposed in [15].

For the case of an organic friction modifier dissolved in base oil, friction coefficients below 0.06 were achieved for the region of low speeds. In this case, the presence of ethanol was not detrimental to friction and boundary film formation. On the contrary, the presence of ethanol increased the range of speeds over which low boundary friction prevailed and the boundary films were thicker than for the non-contaminated oils. This was not surprising, since organic friction modifiers are generally long chain surfactants, which are easily adsorbed to the metal surfaces [16]. It is believed that neither ethanol nor water will compete with them for adsorption. The fact that such thick boundary films form when ethanol is present might suggest the formation of viscous boundary films. These phenomena need to be further investigated.

De Silva et al. [17] also detected a small reduction of mean friction coefficient due to the presence of ethanol and water in a fully formulated oil. However, it was not possible to identify if this reduction occurred for full film, mixed and/or boundary lubrication. Also, it was not possible to identify the extent to which the interaction of ethanol occurred with the base oil or with additives present in fully formulated oil, such as detergents, anti-wear additives and friction modifiers, since only formulated oil was studied.

#### **4 CONCLUSIONS**

The addition of quite small proportions of ethanol slightly reduces EHD film thickness and friction and causes the shift from full film to mixed lubrication to occur at lower entrainment speeds, which can be associated with a reduction in viscosity.

Ethanol contamination of the base oil promotes the formation of a boundary layer that was not present with the ethanol-free base oils, which seems to result from oxidation of ethanol when in contact with a hot, rubbing metal surface.

However, when present in a formulated engine oil, the presence of ethanol interferes with the formation of a boundary film, reducing its thickness from ca 9 nm to ca 2-3 nm.

The presence of ethanol does not seem reduce the efficiency of friction modifiers. In fact, it increases the thickness of the low friction boundary film and therefore increases the range of speeds over which very low friction, below 0.06, can be achieved.

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