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Effect of relative humidity on the tribological performance and tribofilm characteristic of boundary lubricated systems with ZDDP containing oil; Part II: Numerical

Pourya Parsaeian [a,b*], Ali Ghanbarzadeh [a], Marcel C.P. Van Eijk [b], Ileana Nedelcu [b], Ardian Morina [a], Anne Neville [a]

[a] University of Leeds, School of Mechanical Engineering, institute of Functional Surfaces, Leeds, UK

[b] SKF Engineering and Research Centre, The Netherlands

Corresponding author:

E-mail address: *p.parsaeian@leeds.ac.uk (Pourya Parsaeian)

Abstract

Relative humidity and the effect on tribochemistry and wear of boundary lubricated systems are examined experimentally in part one of this study. In part II of this study the tribofilm thickness and wear results obtained experimentally are used to develop a semi-deterministic approach to implement the effect of humidity in wear prediction of boundary lubrication for the first time. Two approaches were used for this purpose; firstly, a modification factor was found to be suitable for Archard's wear equation to be able to account for the effect of relative humidity. This factor is found to be good for engineering designers to be able to predict the lifetime of machine parts. Secondly, the effect of humidity on the tribofilm growth on the surfaces was captured in the model and its effect on wear was tested based on a recent model developed by the authors. It is shown that, as expected, if the tribofilm growth is captured effectively, wear can be predicted. The prediction results were validated with the experimental results showing good agreement. Calibration of the tribochemical model at different levels of relative humidity suggest that the maximum tribofilm formation factor (h_{max}) is varying linearly with the humidity percentage. This led to the further development of the tribochemical model of the authors to adapt to the humid environments.

Key words: *ZDDP, Boundary lubrication, Relative humidity, Wear*

1 Introduction

Previous investigations revealed that water which comes from the environment plays a critical role in the damage process of oil-lubricated tribological systems [1-7]. Corrosion or hydrogen embrittlement could occur in the presence of water in oil which plays a significant role in causing accelerated wear [8-14].

1.1 Effect of water on lubrication

Additives present in the lubricant can be decomposed by small amounts of water and produce sludge and acid which are harmful for every tribological system [11]. Rounds et al [15] found that the presence of water in the oil changes the decomposition rate of the ZDDP. Water molecules can also prevent the additive molecules reacting with the substrate to form a protective reaction layer on the contacting surfaces due to the fact that the non-polar head of water molecules points out to the surrounding oil while the hydrophilic polar heads assemble in the centre and surround the polar additive molecules [3, 11, 12, 16].

It was also proposed by Faut et al [17] that water delays the formation of the tribofilm on the surface which can affect the wear process of the system. The same trend was observed by Parsaeian et al [12]. Water has a big influence on the additive-containing oils because of the hydrophobicity of these oils [11, 18-20]. Another way that water can influence the lubricant properties is to change the solubilisation characteristics of the lubricant which results in the de-solubilisation of the additive in the lubricant [11]. It was shown that contacting surfaces can be affected in humid conditions by mechanochemical reactions involving the adsorbed water which can be attributed to hydrolysis reactions enhanced by interfacial shear [21].

It was reported that thinner tribofilms are formed on the surface at higher levels of relative humidity and higher water concentration which can strongly affect the wear performance [11, 12, 16, 22]. Parsaeian et al [12, 16] found that water can affect the performance of ZDDP by altering the polyphosphates chain length. This effect manifests itself in the formation of shorter polyphosphate chain at higher levels of relative humidity [16, 23]. The results are in a good agreement with the study done by Nedelcu et al [24].

1.2 Wear prediction

Wear prediction has been the subject of many research papers in the past few decades in order to predict the lifetime of machine elements. Because of the complicated nature of the processes occurring, a universal wear model that is able to fully predict the whole process considering the physical, chemical and mechanical aspects is still lacking. However there are numerous wear models in the literature [25] which can predict wear in specific working conditions or are valid for particular mechanism of wear [26-29].

Archard's wear equation was widely used in the literature to predict wear in lubricated and unlubricated conditions in different contact configurations and also different length scales [30, 31]

. It is clear that the chemistry of lubricants and the chemical and mechanical interactions at the interfaces are significantly important in the wear of boundary lubricated contacts. Hence it is important to develop models that can capture these important properties of interfaces.

In some recent studies, tribochemistry was considered in modelling of boundary lubrication. Andersson et al. [32] developed a mathematical model using an Arrhenius-type equation for the formation of the tribofilm on contacting asperities and used mechanical properties of the tribofilm to calculate plastic deformations. Bosman et al. [33, 34] used diffusion reactions to capture the growth of tribofilm on the surfaces. They proposed a new wear model that considers partial removal of the tribofilm responsible for mild wear of boundary lubricated contacts.

More recently, Ghanbarzadeh et al. [35] used a mechano-chemical approach to develop a tribochemical model for the formation and removal of the tribofilm and modified the Archard's wear equation to account for the effect of the tribofilm. The wear model was validated against experimental wear measurements for different temperatures and showed good agreement [36]. The model suggests that a good prediction of the tribofilm will result in a fairly good prediction of wear in the case of antiwear tribofilm formed on steel substrate. The same model was used in another work of the authors to predict the tribocorrosive wear of boundary lubricated systems in the presence of water [12]. Two numerical approaches were used in the paper to predict tribochemical wear in the presence of water. In the first approach, the Archard wear coefficient was modified with a factor to account for the effect of water. In the second approach, coefficient of wear was assumed to be constant and the effect of tribofilm was considered for all different levels of water content. It was shown that if the growth of the tribofilm can be captured, the wear can be predicted using the model reported in Refs [35, 36].

This study indicates incremental improvement to a recently developed tribochemistry model which is capable of predicting wear and tribofilm growth in humid environment. For this purpose, a set of experimental results reported in another paper of the authors [16] (first part of this study) was employed to calibrate and validate the model. The experimental results reported in Ref [16] include the tribofilm thickness measurements at different levels of relative humidity as well as wear measurements on the samples. The results are then used to predict wear in two different numerical approaches that will be explained in detail in Section 4. The discussion on the numerical approaches is reported in Section 5.

2 Experimental results

To investigate the effect of relative humidity on tribofilm formation and wear of the system, a set of experiments was carried out by using Mini Traction Machine (MTM) with the Spacer Layer Interferometry Method (SLIM) attachment. A humidity control system was designed to monitor relative humidity during the experiments. The experimental results reported in detail in the first part of this study [16].

Figure 1 and Figure 2 illustrate the effect of different levels of relative humidity on tribofilm thickness. The results show that the higher tribofilm thickness was observed at lower relative humidity for both temperatures while the thickness of the tribofilm reduced by increasing the relative humidity. The lowest tribofilm thickness was found at the highest value of the humidity for both temperatures. Figure 3 indicates that the average wear depth for both temperatures increased by increasing the humidity but it is more noticeable for lower temperature and higher humidity due to the higher water concentration in the oil.

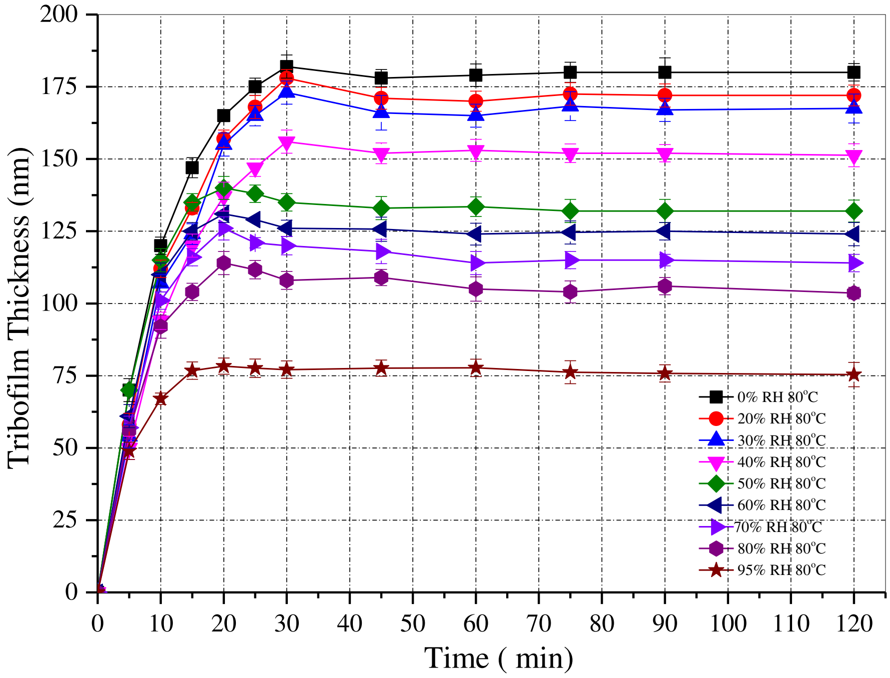

Figure 1 Tribofilm thickness measurement results at 80°C for different levels of relative humidity

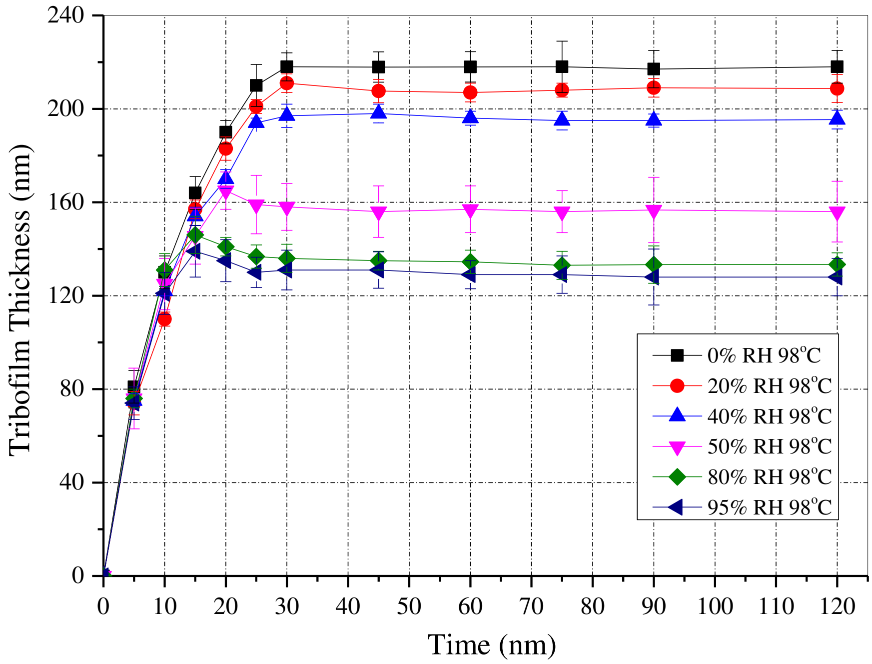

Figure 2 Tribofilm thickness measurement results at 98°C for different levels of relative humidity

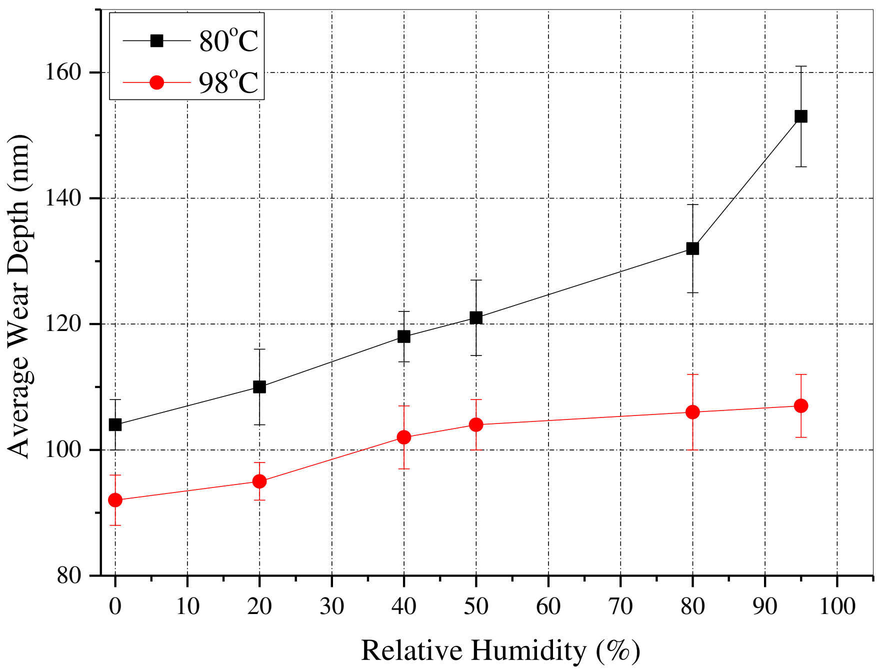

Figure 3 Effect of relative humidity on the average wear depth at two different temperatures of 80°C and 98°C

3 Numerical model

The model used in this work is the semi-deterministic approach developed by authors in Refs [36, 37]. The model was modified to account for the effect of water and its tribochemistry on the wear performance of boundary lubricated contacts in the presence of ZDDP as an anti-wear additive [12]. The details of the numerical model and its validations are reported in detail in Refs [36, 37]. However, the components of the model are presented here. The model consists of three important parts:

I. A contact mechanics code for rough surfaces assuming an elastic-perfectly plastic material response;

II. A semi-analytical tribofilm growth model which includes both tribofilm formation and partial removal; and

III. A modified Archard's wear equation which accounts for the local thickness of the ZDDP tribofilm.

The tribochemical model of the tribofilm growth developed is as the following:

$$h(t) = h_{max}\left(1 - e^{\left(-\frac{k_1 T}{h'} . x_{tribo} . t\right)}\right) - C_3(1 - e^{-C_4 t}) \qquad (1)$$

where k_1 and h' are the Boltzmann and the Planck constants respectively, T is the flash temperature and C_3 and C_4 are constants accounting for the continuous partial removal of the tribofilm. $.x_{tribo}$ is the term introduced in the earlier works of the authors [35, 36] that stands for the effect of entropy change or mechanical rubbing on the induction of tribochemical reactions.

The mathematical foundation of the model can be found in Ref [35], the wear model details and the effect of tribochemistry in modifying Archard's wear equation is available in Ref [36] and numerical approaches to account for the effect of water and its tribochemistry is presented in Ref [12].

4 Numerical analysis

The wear in this work is predicted in two different numerical approaches. The experimental results of Section 2 are used to both calibrate and validate the model. Numerical approaches used in this study are similar to the approaches presented in a recent study of the authors in Ref [12]. Details and the rationale for both numerical approaches can be found in that work. However a brief explanation is reported here as well as the discussion related to the effect of humidity.

4.1 Semi-deterministic coefficient of wear (approach one)

In the first approach it has been decided to modify the Archard wear equation by adding a modification parameter for design purposes. In this case, the model does not deal with changes in the thickness of the tribofilm at different levels of humidity and only changes in the *initial* wear coefficient is considered.

In the wear equation, the parameter which is affected by humidity is considered to be the *initial* coefficient of wear (K_{steel}). The experimental values reported in Section 2 were used to compare with the simulation results at different levels of humidity. Simulations were conducted for each experimental case with different *initial* coefficients of wear. K_{steel} values that result in the wear predictions close to the experimental results were recorded. The accuracy of the wear calculation was set to be 0.1 nm. It means that simulation results corresponding to the *initial* coefficients of wear was set to be close to the values measured experimentally. The *initial* wear coefficients calculated from the simulations are reported in Table 1 for different levels of humidity at 80 °C and 98 °C. Similar to Ref [12] the recent proposed wear model reported in the tribochemical model [36] can be modified using a modification parameter φ to account for the effect of humidity on tribocorrosive wear of the system. These values for φ are reported in Table 2. Therefore, the new wear model that contains the effect of humidity is as the following:

$$K(h) = \varphi . K_{steel} - (\varphi . K_{steel} - K_{min}) . \frac{h}{h_{max}} \qquad (2)$$

in which φ is the modification parameter used to take into account the effect of humidity in Archard's wear equation. It should be noted that the value for relative humidity of 0% has been used as the reference and the φ value for 1 has been assigned to it. This approach is a good approach for designers of machine elements to include the tribochemical effects of humidity on the wear of boundary lubricated systems without any detailed chemical analysis only by modifying Archard's wear equation. By applying the proper value for the φ parameter, wear can be successfully predicted in the numerical simulation.

Table 1 Dimensionless *initial* wear coefficients in the numerical simulations in the first approach

Relative Humidity	Temperature 80ºC	Temperature 98ºC
0	2.2×10^{-8}	1.2×10^{-8}
20	2.5×10^{-8}	1.3×10^{-8}
30	2.6×10^{-8}	-------------
40	2.95×10^{-8}	2×10^{-8}
50	3.1×10^{-8}	2.2×10^{-8}
60	3.5×10^{-8}	-------------
70	4×10^{-8}	-------------
80	4.2×10^{-8}	2.3×10^{-8}
95	6×10^{-8}	2.35×10^{-8}

Table 2 φ factors for modifying the Archard equation for different levels of relative humidity

Relative Humidity	Temperature 80ºC	Temperature 98ºC
0	1	1
20	1.13	1.08
30	1.18	-------------
40	1.34	1.66

50	1.41	1.83
60	1.6	-------------
70	1.81	-------------
80	1.90	1.91
95	2.72	1.95

4.2 Effect of tribochemistry (approach two)

In the second method, a different approach is used to consider the effect of humidity on the tribochemical wear. The *initial* coefficient of wear (K_{steel}) is set to be constant in all cases and the dynamic growth of the tribofilm on the surfaces and its effect on changing the wear coefficient is analysed. The tribofilm growth results reported in Figure 1 and Figure 2 have been used to capture the behaviour in the model. In the wear model used in this paper (Equation 2) and reported in one of the earlier works [36], the thickness of the tribofilm changes the local coefficient of wear at asperity-scale. Therefore, capturing the dynamics of the tribofilm in the numerical model results in changing the spatial coefficient of wear over time and hence the overall wear of the system.

The parameters (x_{tribo}, h_{max}, C_3 and C_4) of the model of Equation 1 and reported in Refs [12, 36, 37] are obtained by fitting the mathematical expression of the tribochemical model to experimental tribofilm thickness results in Figure 1 and Figure 2. The calibration parameters are reported in Table 3 and

Table 4 for different levels of humidity. The accumulative wear is computed in the numerical model with respect to the tribofilm growth behaviour.

Simulation of the tribofilm has been carried out in the model and its dynamic growth over time has been shown in Figure 4 and Figure 5 for 80ºC and 98ºC respectively. The wear predicted based on the growth behaviour is plotted in Figure 6 and Figure 7 as well as the experimental wear measurements for comparison purposes. It is clear that the different growth behaviour of the tribofilm on contacting asperities at various levels of relative humidity, affect the mild wear of boundary lubricated contacts. Similar to the results previously reported in Refs [12, 36], it can be interpreted that a good prediction of the wear of boundary lubricated contacts can be obtained by successful capturing of the tribofilm dynamic behaviour on the surfaces.

Table 3 Simulation inputs and calibration parameters at 80ºC

Relative Humidity %	**h $_{max}$ (nm)**	**X $_{tribo}$**
0	300	1.88×10^{-16}
20	257	1.74×10^{-16}
30	218	1.75×10^{-16}
40	184	1.75×10^{-16}
50	179	1.83×10^{-16}
60	140	1.89×10^{-16}
70	121	1.84×10^{-16}
80	126	1.9×10^{-16}
95	83	1.93×10^{-16}

Table 4 Simulation inputs and calibration parameters at 98ºC

Relative Humidity %	**h $_{max}$ (nm)**	**X $_{tribo}$**
0	365	1.54×10^{-16}
20	323	1.47×10^{-16}
40	246	1.75×10^{-16}
50	225	1.86×10^{-16}
80	183	1.93×10^{-16}
95	171	1.85×10^{-16}

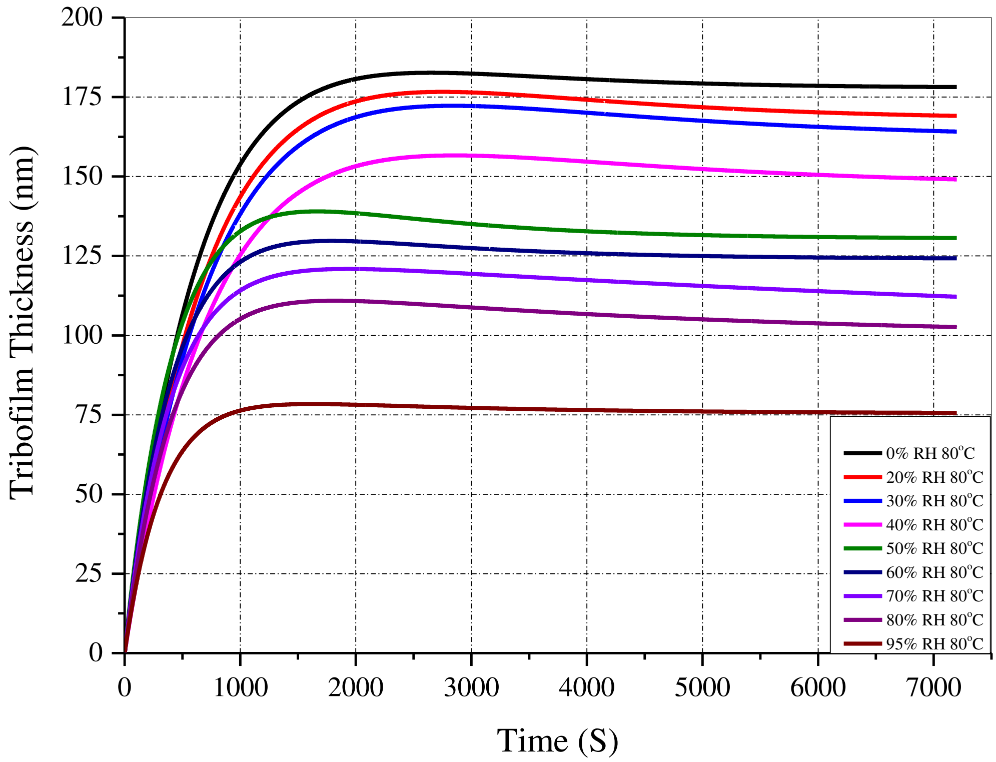

Figure 4 Tribofilm growth simulations for different values of relative humidity at 80°C

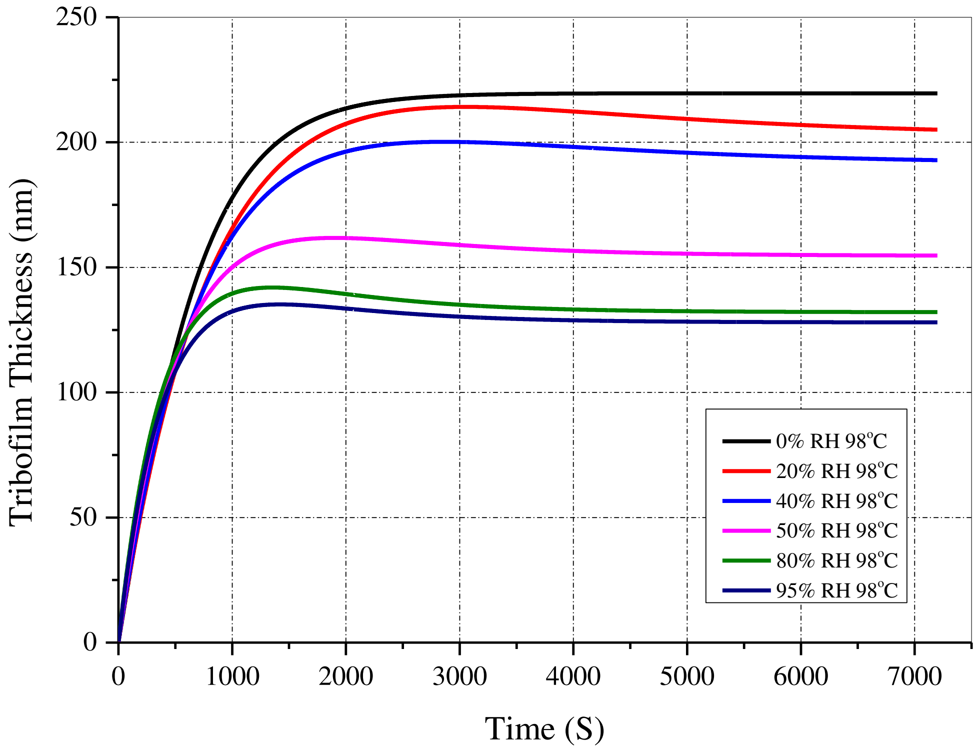

Figure 5 Tribofilm growth simulations for different values of relative humidity at 98°C

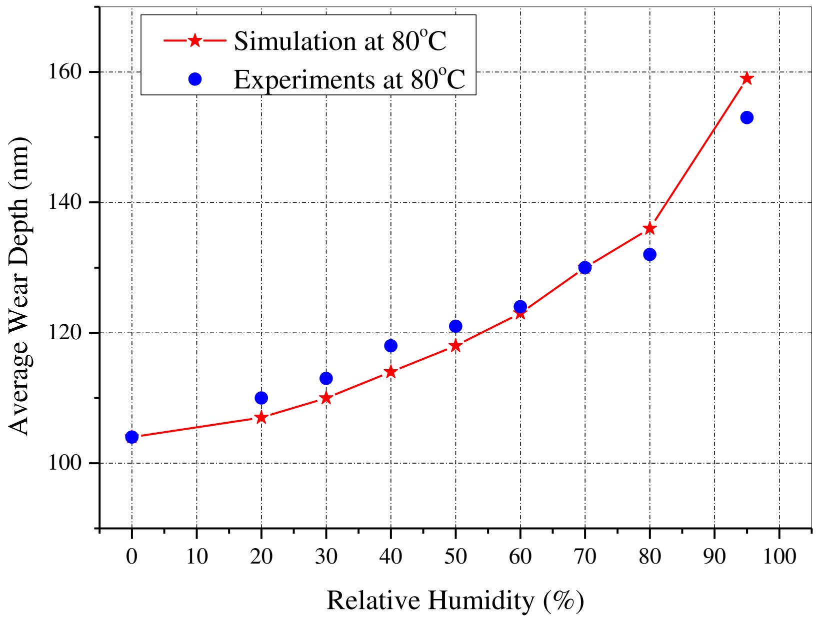

Figure 6 Numerical wear calculation in comparison with experimental measurements at 80°C (calculated from second approach)

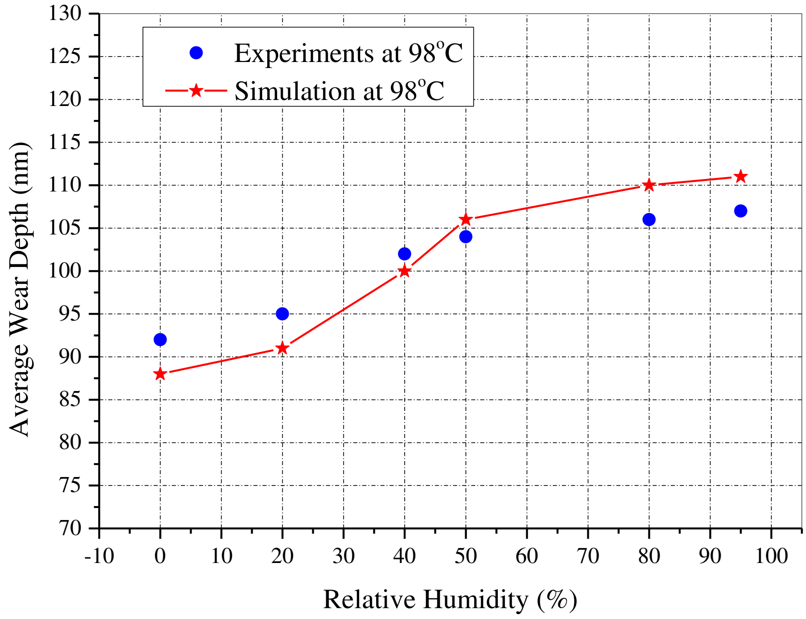

Figure 7 Numerical wear calculation in comparison with experimental measurements at 98°C (calculated from second approach)

5 Discussion

The wear of boundary lubricated system in a tribocorrosive environment in the presence of ZDDP as an antiwear additive has been modelled in this work by considering the tribochemistry for different levels of relative humidity. The wear in this condition was modelled using two numerical approaches. As explained is Section 4.1 a modification to

Archard's wear equation that accounts for the effect of relative humidity can successfully predict wear in boundary lubrication. This approach can be simply used in design of machine elements working in humid environments. The modification parameter was reported to be between 1 and 2.8 for both temperatures and is similar to the factors introduced in the recent reports of authors in Ref [12]. The second approach accounts for the effect of tribofilm and its thickness at different relative humidity levels. The effect of relative humidity on the growth of ZDDP tribofilm was investigated in this approach. The same *initial* coefficient of wear was used in all simulations and the effect of tribofilm thickness in reducing the coefficient of wear was modelled. Wear was then predicted based on Equation 2 and fairly good agreement was found between experimental and numerical results (Figure 6 and Figure 7).

Tribofilm thickness in this case follows Equation 1 in all levels of humidity and the model of Equation 1 was fitted into the experimental tribofilm thickness results reported in Figure 4 and Figure 5 for 80ºC and 98ºC respectively. The fitting parameters are reported in Table 3 and

Table 4. These parameters were then used to run the simulations and predict wear based on Equation 2. It is presented that capturing the growth of tribofilm can be the first step for predicting the wear of boundary lubricated systems. Once the tribofilm behaviour is captured, the wear is successfully predicted using Equation 2.

It is clear from the experimental results of tribofilm growth, that the humidity hinders the further growth of tribofilm on the contacting asperities. The rate of growth of the tribofilm on the surfaces is almost similar for all humidity levels and this can be seen from the initial stages of the tribofilm growth in Figure 4 and Figure 5. On the other hand, the steady-state film thickness that tribofilm reaches, is significantly affected by relative humidity. This is an interesting finding. This is perhaps because, it takes time for the humidity to dissolve in the lubricating oil and move to the contact and adsorb on the surface. Comparing the results with a recently published paper of the authors in Ref [12] indicates that if the water is added to the lubricant, it prevents the formation of the tribofilm from the beginning. This is because the water is already in dissolve form in the oil. On the other hand, in the case of humidity, it takes time for humidity to convert to dissolved water and the rate of the growth of the tribofilm does not change much in the beginning.

In the tribochemistry model of Equation 1, steady-state tribofilm thickness is a combined effect of steady-state tribofilm formation (h_{max}) and the steady-state tribofilm removal (C_3) terms. Extracting these information from fitting Equation 1 into experimental tribofilm growth results, show that the tribofilm formation rate term (x_{tribo}) is not significantly dependant on the relative humidity as was expected from the experimental results (the rate of growth seems to be similar for different levels of relative humidity). On the other hand, the term h_{max} in the model which is a representation of maximum tribofilm *formation* (not growth) in the absence of tribofilm removal, is significantly affected by relative humidity. In this regard, the calibrated h_{max} reported in Table 3 and

Table 4 are plotted for different levels of relative humidity in Figure 8 and Figure 9 for 80°C and 98°C respectively. It can be seen that the term h_{max} is clearly reducing linearly with relative humidity values.

The mathematical model representing this behaviour for both temperatures are presented below:

$$h_{max} = -2.27 \times RH + 291.31 \qquad T = 80^oC$$

$$h_{max} = -2.10 \times RH + 352.05 \qquad T = 98^oC$$

In which RH is the relative humidity percentage ranging from 0 to 100.

In order to modify the tribochemistry model of Equation 1 it has been decided to see the effect of relative humidity on the maximum film formation form and averaging the above mentioned equations. The variation of h_{max} with relative humidity can be expressed as:

$$h_{max} = -2.18 \times RH + 321.68 \qquad (3)$$

Equation 3 can be used to predict the term h_{max} in Equation 1 in humid environments which then can lead to good prediction of tribofilm growth on the surfaces and result in good prediction of wear in boundary lubricated systems in the presence of anti-wear additives. The tribochemical model of Equation 1 is then converted to:

$$h(t) = (-2.18 \times RH + 321.68)\left(1 - e^{\left(-\frac{k_1 T}{h'}.x_{tribo}.t\right)}\right) - C_3(1 - e^{-C_4 t}) \qquad (4)$$

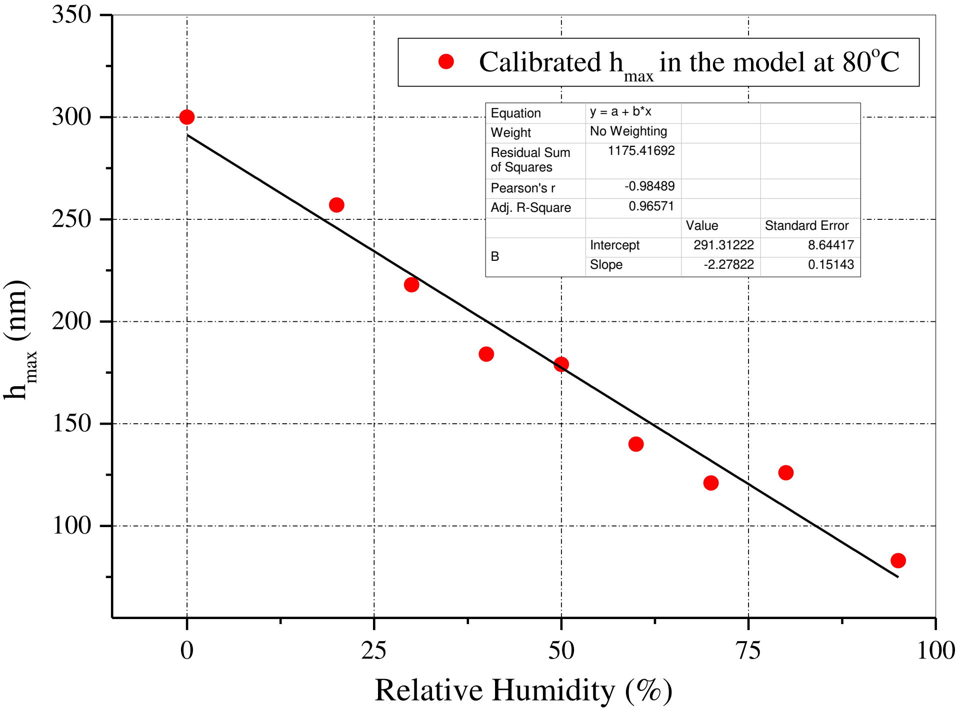

Figure 8 Variation of h_{max} by relative humidity at 80°C

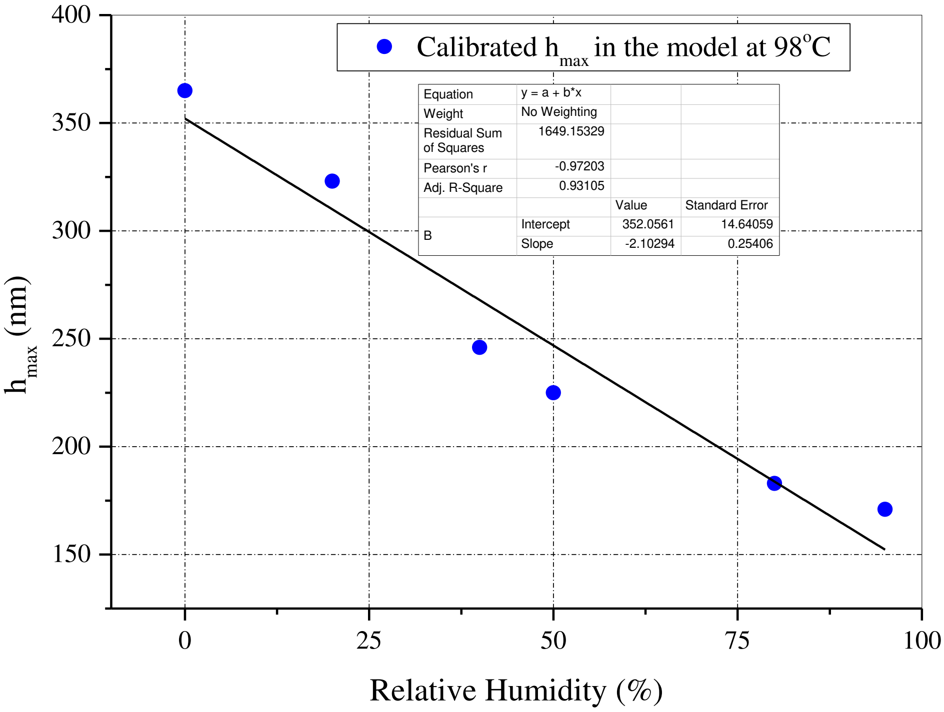

Figure 9 Variation of h_{max} by relative humidity at 98°C

According to the calibration of Equation 1 at different humidity levels, the term x_{tribo} which in principle is responsible for the formation rate of tribofilm, is not significantly affected by altering the humidity. This is because the humidity does not affect the growth rate of the ZDDP tribofilm considerably as shown in experimental results of Figure 1 and Figure 2 in comparison with the effect of mixed-water in the oil which can significantly affect the growth rate of the tribofilm [12].

6 Conclusions

A semi-analytical approach was applied to model the effect of relative humidity on tribofilm growth and wear in boundary lubricated system in presence of ZDDP as an anti-wear additive. Two approaches were employed to implement the effect of relative humidity in predictive wear model and the following conclusions can be drawn:

- In regards to the first approach, Archard's wear coefficient is semi-deterministically obtained for different levels of relative humidity from the simulations. This can lead to the modification of Archard's wear equation with a modification factor of φ. This approach can be used by designers of tribological parts to take in to account the effect of humid environment on the durability.
- In addition, the modification factor φ is increasing while the humidity increases.
- The second numerical approach was applied to consider the effect of humidity on the tribofilm growth and the corresponding wear behaviour in boundary lubrication. It is shown that successfully capturing the tribofilm growth behaviour leads to predicting the tribochemical wear in boundary lubricated conditions.
- It was the effect of relative humidity on tribofilm growth behaviour and wear is different from the effect of mixed-water in the oil which was the subject of a recent study by the authors. In the latter case, the maximum film thickness was found the same for different levels of water concentration while the tribofilm growth rate found to be significantly affected which led to different wear rates in running-in stage. In the case of relative humidity, the maximum film thickness is influenced considerably.
- Calibration of the numerical-tribochemical model suggests a linear variation of h_{max} (maximum tribofilm formation) with relative humidity and the result is a modification to the tribochemical model to adapt it to the tribocorrosion conditions in humid environment (Equation 4).

7 Acknowledgement

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