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Reply

Reply to Bair, S. Comment on "Neupert, T.; Bartel, D. Evaluation of Various Shear-Thinning Models for Squalane Using Traction Measurements, TEHD and NEMD Simulations. *Lubricants* 2023, *11*, 178"

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1. Introduction

After the publication of our paper, we received a comment [1] to which we would like to respond here. First of all, we would like to thank the commentator for his constructive criticism of our paper. The comment primarily criticises the selected pressure viscosity model according to Rodermund (Ref. [44] of the paper), which was used for three of four fluid models and cannot represent an inflection point. Furthermore, it is pointed out that the Eyring shear thinning model (Ref. [45] of the paper) is not able to adequately describe the shear thinning measured in a Couette viscometer and only apparently leads to good results in traction calculations due to incorrect pressure viscosity modelling.

First, we would like to point out that we consider fluid modelling from the perspective of its application in TEHD simulations. That means, from our point of view, the main objective of viscosity modelling is the usage for the prediction of the occurring friction and film thickness of machine elements. For this purpose, the interaction of the temperature, pressure and shear dependence of the viscosity within the contact is essential.

2. Fitting Challenge

It should be noted that all equations describing the rheometry of a liquid are fit functions that attempt to describe complex molecular dynamic phenomena on a macroscopic level. This means that the aforementioned "fitting challenge" must be carried out not only in investigations using traction measurements but also in rheometer experiments. For example, for the development of the Tait-Doolittle and shifted Carreau model of squalane proposed by the commentator in Ref. [8] of the paper, the parameters of the Tait equation were fitted from density measurements (Ref. [8] of the paper), the parameters of the Doolittle equation were derived from low shear viscosity measurements (Ref. [8] of the paper) and the parameters of the shifted Carreau model were determined by fitting Couette viscometer measurements combined with NEMD simulations (Refs. [8,23] of the paper).

All in all, the objective is to identify model equations that ideally provide high performance for rheology, molecular dynamics and TEHD simulation compared with traction measurements. As described in detail in the paper, all validation methods have advantages and limitations.

3. Validation of the Assumed Flow Behaviour with Tribometers

We agree with the commentator that a test rig is not a rheometer [2], as the measured friction is only an integral response of extremely different states in the contact. This problem is pointed out several times in our paper. However, a good fluid model must be able to represent the friction that occurs in real contact (assuming a reliable numerical TEHD model). Otherwise, the objective of the lubricant modelling is only partially achieved. Validation with traction measurements is, therefore, an appropriate method for verifying a chosen modelling. If certain assumptions about fluid behaviour exist, the parameters of the model can be optimised by comparing the calculation results and the traction measurements. However, this requires a small number of variable model parameters.

The commentator states that "The prediction of friction should not require a measurement of friction". To be precise, the metrological determination of fluid behaviour always requires a measurement of friction. Rheometers also measure the friction response of the fluid, whether directly in a Couette gap or indirectly via the oscillation frequency of a quartz. However, the advantage of rheometers over traction measurements is that the boundary conditions of the measurements can be specifically defined. Of course, it would be desirable to derive all rheometric data from such experiments and to be able to apply them directly in TEHD simulations. Unfortunately, rheometric measurements currently still have their limits in terms of pressure, shear rates and the thermal influence on the measurement due to shear heating (Ref. [10] of the paper).

4. Pressure Viscosity Model

Rheometric measurements, especially the pressure viscosity behaviour, are an essential prerequisite for successful TEHD simulation. For this reason, they are presented first in the paper. We agree with the commentator that it is impossible to reliably determine such curves from TEHD measurements [3].

If the focus is on an exact representation of the measured pressure viscosity dependence, degressive-progressive models (such as Tait-Doolittle or hybrid models) are preferable, as shown by the commentator in a large number of studies. For the determination of the central film thickness, which is mainly influenced in the inlet area of the TEHD contact with moderate pressures [4], this modelling can be directly used. However, for the calculation of friction, which is largely determined by the high-pressure region in the middle of the contact, users usually have the problem that the curves measured in the rheometer have to be extrapolated into the high-pressure range. Extrapolation is generally subject to uncertainties and is based on the assumption that the behaviour continues as specified by the underlying equation. Unfortunately, degressive-progressive models, along with the available shear-thinning models, often lead to worse results in friction calculations. This is the reason why degressive models are often used in the TEHD community despite better knowledge of rheometry. It is therefore necessary to compare this approach in studies, as it is also done in two other recent papers for squalane [5,6].

Actually, we are surprised at the criticism regarding our representations. The Comment suggests that we have completely ignored the degressive-progressive models. In fact, however, we have deliberately presented and discussed the Tait-Doolittle equation for squalane proposed by the commentator as model 4 and included it unchanged in all our investigations.

In the discussion of the curves in the paper, it is explicitly pointed out that "the measurement in rheometers indicates solidification for very high pressures" and that the curves of the Tait-Doolittle equation "at very high pressures $p > 1500$ MPa, [...] show an incipient glass transition and, thus, increasingly larger viscosity values [than the Rodermund equation]".

Using the sentence "On the basis of the available measurement values, an interpolation fitting was carried out so that all data are matched as well as possible." was not the authors' intention to claim that the Rodermund equation was chosen because it can best represent the measurement data. It should only be pointed out that the parameters of the equation (A, B, C, D, E, p_0) have been determined in such a way that the degressive curve represents the measured values (not ideally but) in the best possible way.

5. Shear Thinning Models

Figure 3 of the comment shows that the Rodermund and Eyring model, which is one of the four fluid models investigated, does not follow the results from thin film Couette viscometer measurements for high shear rates. It is shown that the sinh curve progression of the Eyring model is not correct.

However, Spikes et al. have frequently shown that traction measurements can be attributed to sinh behaviour (Refs. [10,12,14] of the paper). MD simulations can also be well-fitted with Eyring models. For instance, Jadhao et al. (Ref. [25] of the paper) and our own MD simulations show good agreement with this equation. Simultaneously with our paper, Xu et al. [7] published a study in which squalane was modelled on the basis of rheometric measurements (degressive-progressive modelling) and MD simulations using an Eyring approach. As a result, good agreements were found between TEHD simulations and traction measurements. Findings from traction measurements and MD simulations, therefore, do not contradict the Eyring model.

Again, the Comment overlooks the fact that our paper also considers other models than that of Rodermund and Eyring. Thus, the Tait-Doolittle and shifted Carreau model proposed by the commentator is applied with unchanged parameters. This is based solely on rheometric and molecular dynamics considerations, as requested. However, it leads to large deviations when used in TEHD simulations. To improve agreements, a simple pressure-dependent limiting shear stress $\tau_{\text{lim}} = 0.075 \times p$ was introduced by the commentator in calculations (Ref. [39] of the paper). However, this assumption was not derived from a rheometer but "was found from a traction measurement at $p_h = 1.93$ GPa where the traction coefficient reached a plateau at 0.075". This assumption was repeatedly adopted in many subsequent publications (Refs. [19,20] of the paper and Refs. [4,5]) and also tested in our investigations. Nevertheless, even with this approach, there are only unsatisfactory results.

Overall, it can be stated that it is currently not possible to completely represent all observations of rheometers, MD simulations and TEHD simulations with the help of a single model. Depending on the modelling approach, one of the disciplines usually suffers, as evidenced by the long-standing discourse, our paper and the Comment. As is usual in science, we will, of course, incorporate the latest findings into our work in order to come closer to the objective of a model based entirely on physical principles.

Conflicts of Interest: The authors declare no conflict of interest.

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