Review of: "On the rheology of thixotropic and rheopexic suspensions: accounting for the formation of trimers"

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Potential competing interests: No potential competing interests to declare.

This paper is about the modeling of non-newtonian fluid through multi parameter semi-empirical equations. The paper takes into account the formation of doublets and triplets in a suspension and their effect on the suspension viscosity and shear stress, trying to give insight on the importance of triplets towards the correct evaluation of these quantities.

While the idea is of interest the general subject of thixotropy, plasticity, rheopexy etc. as well as the particular results achieved in this paper are poorly presented.

Abstract and introduction needs rewriting adding more literature citations. The model equations should be better presented and explained. Particular attention should be given to the “Results and discussion” section because wrong results are shown here (see the details below).

You can find a detailed analysis of your paper below.

Abstract:

From the Abstract it is not clear what are the purposes and main goals of this article. I suggest rewriting it in order to make it clear addressing their impact on the scientific field topic. The correct spelling is Krieger-Dougherty I suggest you correct it throughout the paper.

Introduction:

I suggest adding here a literature review which will benefit the reader, guiding him from the general topic to the more specific one analyzed in this paper. In particular the paper will benefit from a review of the paper where the Krieger-Dougherty formula was used together with a brief description of the system analyzed and from a review of other methods and models used to describe thixotropic and rheopexic suspensions. I also suggest adding the reasons you chose to use this specific model instead of another with all its pros and cons when compared to other models.

I think that a clear definition of dimers and trimers should be introduced here. I understand that they come from the aggregations of singlets into doublets and triplets but through some sort of chemical reaction but I think that this part should be made clear.

What do the parameters k_1 and k_2, which appear in the formula for k, represent?

I suggest rewriting the last two introduction paragraphs in order to clarify what are the purposes of this work.

Theory of calculations:

It is not clear how the volume fractions (and the number of particles per unit volume) are related between them and to the reactions constants. I think that some more explanation on this point is needed to better explain how you derive eq. 4.

As you state at the end of this section, this model depends on 12 parameters. Since this is a good amount of parameters to use in a model and other reviewers already pointed out possible problems related to them, could you clarify how to select them: do you find them in literature or can you derive them by fitting experimental data?

In eq. 7 you also introduce a yield stress while the Krieger-Dougherty model you start from does not have it. Can you comment on the necessity of this introduction?

It could also be wise to present here the viscosity equation derived from eq. 7 since at this point you are not using eq. 1 anymore.

Results and discussion:

I suggest adding a legend on every plot of this section describing what the dots and lines represent. It is not clear from the text nor the captions what the error bars do represent. If they are the std of your curve from the empirical data I suggest adding points on your curve, corresponding to the experimental one, and placing the error bars on the theoretical points and not the experimental ones.

It is not clear from the text how the experiment was performed. From fig. 1 I understand that the shear rate was first increased (blue arrow) in steps, for every step the steady state was achieved and then the value of the shear stress was measured. The same was done after reaching the maximum shear rate of about 1000 while decreasing the sear rate (red arrow). If this is the case I think these details should be added in the text.

I don’t understand how you chose the parameters you used to derive the continuous curves reported in fig. 1. At this stage, without better explanation, the only thing that one may think is that you have a good agreement between experimental and theoretical data because you chose ad hoc parameters and one may argue that this could be always possible because you have 12 adjustable parameters in your model. So I really think you should better explain how you select the values for all your parameters in order to make the model and its explanation consistent and to avoid such objections.

Fig. 2 presents a wrong result. The suspension viscosity should go to infinity for both the blue and the red curves when using the equation for the shear stress eq. 7 to calculate it.

In fact you can obtain the viscosity expression dividing eq. 7 by the shear rate, in this case the first term of the expression becomes tau_0/gamma which becomes infinite when the shear rate goes to zero. From what I see it seems like you have used eq. 7 for the shear stress solution and then you removed the yield stress to calculate the viscosity (the proof beeing you have the same initial viscosity in blue as the one listed in your parameters in fig. 1) but in this way you are considering two different physical systems.

This problem on the viscosity evaluation and on the equations you used make the subsequent physical discussion completely irrelevant.

I suggest you evaluate the correct viscosity expression starting from eq. 7 and use it to generate a new plot for fig. 2. In order to assess if the contributions from the triplets are relevant I suggest you compare the results from your model with triplets with the one with only doublets and singlets.

The same considerations given for fig. 1 and fig. 2 holds true also for fig.3 and fig. 4 respectively, being fig. 4 wrong for the same reasons of fig. 2.