



Advances in the modelling of concentration-dependent relative viscosity data for nanofluids by introducing the Dispersion Factor



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ABSTRACT

The viscosity ratio (relative viscosity) of a nanofluid to its base liquid is related to the nanoparticle volume fraction by various developed theoretical and empirical equations. However, the theoretical framework used up to now is often inadequate for modelling experimental data. Now, a new parameter denoted as the Dispersion Factor (DF) is proposed to advance the accurate modelling of relative viscosities of nanofluids dependent on nanoparticle volume fraction. Literature data of TiO₂, γ-Al₂O₃ and SiO₂ nanofluids have been selected and subjected to our new theoretical treatment using the Chen equation adapted with the Dispersion Factor to model the relative viscosity in relation to the nanoparticle volume fractions. A much better agreement with the experimental data has been obtained. The value of DF that is identified by the mathematical modelling of relative viscosity data reflects the comprehensive effect of particle size, shape and chemical composition on the interactions between the nanoparticle and the base liquid on the one hand, and solvated nanoparticle and nanoparticle interactions on the other hand. The DF parameter may present a possible tool that can be used to tailor and tune the nanofluid design to meet specific application requirements.

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1. Introduction

Nanoparticles dispersed in a base liquid form a nanofluid. Due to the small size and large specific surface area of nanoparticles, a small amount of functionalised nanoparticles has demonstrated a significant modification of the physiochemical properties of the base liquid, e. g. gas/vapour absorption, wettability alteration of the carbonate rock surface, interfacial tension, and viscosity [1–3]. Such physiochemical property modifications are of high interest in processing intensification methods/technologies, such as CO₂ capture [4], dehumidifying in air-conditioning[5], and more interestingly, enhanced oil recovery [6–10]. Enhanced oil recovery is the oil production stage after water flooding where water cannot effectively replace the oil existing in the porous reservoir anymore. Using more viscous fluids to flood is one of the strategies to recover more oil. Metal oxide nanoparticles dispersed in water as nanofluid have been extensively studied for their viscosity enhancement, of which silica (SiO₂) [8,6], alumina (Al₂O₃) [11–13] and titanium dioxide (TiO₂) [11,12,14,15] are among the most studied.

The viscosity enhancement is evaluated by relative viscosity, i.e. the viscosity ratio of the nanofluid to the base liquid. It increases with the particle volume fraction, as described by the equations reported by e.g. Einstein [16], Batchelor [17], Krieger and Dougherty [18] and Pak and Cho [19]. These equations incorporate molecule-to-molecule interactions mediated by the viscosity of the base fluid, but do not include interactions of solvated molecules to nanoparticles, nor nanoparticle-to-nanoparticle interactions. The nanoparticle volume fraction expressed in the equations is described for spherical particles without the inclusion of any parameters that distinguish particle size, shape and composition of the particles (e.g. surface chemistry). However, the effect of particle properties on the viscosity enhancement can be very significant, as has been demonstrated by the studies of particle size [20–23], shape, i.e. morphology [24,25], material type, i.e. chemical composition [26] in relation to nanoparticle concentration [20,21,26,3,27,28]. Unexplained phenomena such as the relative viscosity decreasing with particle size reduction towards the lower nanometre range [29,30] and inconsistency in experimental data from different laboratories [31–34] have been repeatedly observed. For example, various groups studied TiO₂ aqueous nanofluids with a nanoparticle volume fraction of 0.02. For TiO₂ nanoparticles of 21 nm Duangthongsuk and Wongwises [31] determined a relative

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viscosity of 1.13, whereas Turgut et al. [32] reported a value of 1.60 at the same temperature of 25 °C. The origin of this discrepancy is unclear. It is even more puzzling that at the volume fraction of 0.02 for an aqueous nanofluid of TiO₂ nanoparticles of 90–200 nm, Jarahnejad et al. [29] reported a relative viscosity of 1.88. Similarly enigmatic results have been reported for Al₂O₃ aqueous nanofluids, in that at a volume fraction of 0.03, Duan et al. [30] reported a relative viscosity of 1.5 for 25 nm Al₂O₃ nanoparticles, whereas Chandrasekar et al. [33] reported a relative viscosity of 1.45 for 43 nm Al₂O₃ nanoparticles, and Moldoveanu et al. [34] reported a relative viscosity of 3.0 for 45 nm Al₂O₃ nanoparticles. There is a definite need to explain these as yet inexplicable experimental results to get insight into other factors, beyond the nanoparticle volume ratio and nanoparticle size, that affect the magnitude of viscosity enhancement.

The comparison of these various reported experimental results with the calculated values resulting from the different theoretical treatments shows that the theoretical equations of Einstein [16], Brinkman [35], Batchelor [17] and Krieger-Dougherty [18] all give values for the relative viscosity of nanofluids that are significantly lower than the experimental ones, while the calculated values resulting from the empirical equation by Pak and Cho [19] represent the upper limit in the nanoparticle concentration range for which their empirical equation is derived. The values of the literature data that are located between the values calculated using the treatments by Einstein or Pak and Cho cannot be explained. Chen and co-authors [36] modified the Krieger-Dougherty equation by introducing an agglomeration ratio parameter to describe the particles' agglomeration in the dispersion, but the modelling result leads to the explanation that the viscosity enhancement increases with the agglomeration ratio, which does not obey the general understanding of dispersion systems and to guide the relevant practice work.

In this paper, at first an overview of the theoretical treatments developed so far for describing the relative viscosity of particle fluids is presented. The particular focus is on the merits and limits of each of these various theoretical frameworks. Our research involves the detailed analysis of selected relative viscosity literature data for nanofluids of differently sized TiO₂, γ-Al₂O₃ and SiO₂ nanoparticles. The data is subjected to a variety of theoretical models, after which the agreement between the experimental relative viscosity data and the theoretical values is analysed and discussed, revealing significant discrepancies showing that the present theoretical framework is inadequate for modelling the experimental data. Therefore, it has been proposed to introduce and use a new parameter, namely the Dispersion Factor (DF), to comprehend and mathematically describe the effect of particle size, shape and type (i.e. chemical composition) on the interactions between the nanoparticle and the base liquid on the one hand, and solvated nanoparticle and nanoparticle interactions to each other on the other hand, on the viscosity change of the base liquid. We shall emphasise the importance of the DF parameter as a tool for the future design and testing of nanofluids allowing them to be tailored and tuned to suit the application requirement. The DF parameter also allows for the quantification of differences in viscosity data for nanofluids (such as those due to nanoparticle aggregation and/or agglomeration) that could up to now not be captured, described, examined and analysed by a numerical parameter.

2. Overview of theoretical treatments

Various theories have been developed to explain and predict the influence of particle addition on fluid viscosity. The first attempt has been reported by Einstein as early as 1906 [16]. Equation (1) shows the Einstein equation, which expresses the ratio of the effective

dynamic viscosity of the nanofluid (η_{eff}) and the dynamic viscosity of the base liquid (also termed base fluid, abbreviated as bf) (η_{bf}) at a particular temperature as being equal to $1 + 2.5 \varphi$, in which φ is the volume fraction of the spherical nanoparticles suspended in the base liquid.

$$\frac{\eta_{\text{eff}}}{\eta_{\text{bf}}} = (1 + 2.5\varphi) \quad (1)$$

The Einstein equation is valid for dilute suspensions with a nanoparticle volume fraction less than 0.01, hence its limitation is that it does not consider the interaction between hard-spherical particles in the suspension.

In 1952, Brinkman [35] expanded on Einstein's treatment by estimating the concentration-dependence of any particle quantity at higher concentrations if the result for infinite dilution is known, which resulted in Equation (2) that is valid for a particle volume fraction of up to 0.04.

$$\frac{\eta_{\text{eff}}}{\eta_{\text{bf}}} = \frac{1}{(1 - \varphi)^{2.5}} \quad (2)$$

When the volume fraction of suspended solid particles increases further, the interaction among the particles increases due to Brownian motion. In 1977, Batchelor [17] modified Einstein's equation by including the effect of Brownian motion of spherical hard particles for an isotropic suspension, thus resulting in Equation (3), which is suitable for predicting the suspension viscosity for particle volume fractions of up to 0.1.

$$\frac{\eta_{\text{eff}}}{\eta_{\text{bf}}} = 1 + 2.5\varphi + 6.2\varphi^2 \quad (3)$$

In 1998, Pak and Cho [19] specified Equation (4), resulting from experimental data of aqueous suspensions of γ-Al₂O₃ and TiO₂ nanoparticles, which has a similar form to that of Batchelor but the constants involved are much larger.

$$\frac{\eta_{\text{eff}}}{\eta_{\text{bf}}} = 1 + 39.11\varphi + 533.9\varphi^2 \quad (4)$$

Building on the equations related to Einstein's model, Krieger and Dougherty [18] introduced their power law-based equation in 1959, Eq. (5), which predicts the effect of the volume fraction of micro/millimetre-sized particles in suspensions on the relative viscosity. It predicts the viscosity of non-diluted suspensions with randomly monodispersed hard spherical particles by introducing the maximum particle volumetric fraction (φ_m), at which the particles are just able to flow.

$$\frac{\eta_{\text{eff}}}{\eta_{\text{bf}}} = \left(1 - \frac{\varphi}{\varphi_m}\right)^{-[\eta]\varphi_m} \quad (5)$$

$$\text{where } [\eta] = \lim_{\varphi \rightarrow 0} \frac{\eta - \eta_0}{\eta_0 \varphi}$$

The maximum particle volume fraction φ_m varies from 0.50 to 0.54 under quiescent conditions, while at high shear rates the value is approximately 0.61. $[\eta]$ is the intrinsic viscosity (the measure of a solute's contribution to the viscosity of a solution) which is 2.5 according to Einstein [16], for well-dispersed hard spherical particles [37].

In 2007, Chen et al. [36] modified the Krieger-Dougherty equation by introducing φ_a , the volume fraction of aggregation, to address the particles' aggregation as shown in Equation (6).

$$\frac{\eta_{\text{eff}}}{\eta_{\text{bf}}} = \left(1 - \frac{\varphi_a}{\varphi_m}\right)^{-[\eta]\varphi_m} \quad (6)$$

Where $\varphi_a = \varphi \left(\frac{a_a}{a}\right)^{3-D}$, in which a_a and a are the radius of aggregate and the radius of the primary particle, respectively, and D is the fractal index indicating the extent of change in the packing fraction from the centre to the edge of the aggregate. It is typically 1.6–2.5 for diffusion-limited aggregation and 2.0–2.2 for rate-limited aggregation for spherical particles [38–40].

For spherical particles with $D = 1.8$ [41], $[\eta] = 2.5$ and $\varphi_m = 0.605$ [18], Equation (6) can be simplified as Eq. (7):

$$\frac{\eta_{\text{eff}}}{\eta_{\text{bf}}} = \left(1 - \frac{\varphi}{0.605} \left(\frac{a_a}{a}\right)^{1.2}\right)^{-1.5125} \quad (7)$$

Much progress has been made in the development of the theoretical framework describing the relationship between the viscosity changes with the nanoparticle volume fraction, but further progress is needed for the description of the viscosity change with respect to the properties of the added solid nanoparticles and their interaction to the base liquid and each other in the base liquid, especially when the nanoparticle population becomes large (e.g. for small particle size and large volume fraction) and the interaction of the nanoparticle to the base liquid and other particles is strong.

To make further progress, we propose to analyse literature relative viscosity data of nanofluids using various reported theoretical models, and we shall show that a better interpretation of the literature data is afforded by our adaptation of Chen's equation via introducing a new parameter denoted as the Dispersion Factor (DF) to describe relative viscosities of nanofluids dependent on nanoparticle volume fraction.

3. Our analysis of literature experimental data vs. calculated results

Nanoparticles of metal oxides are a group of widely studied viscosity enhancers for their applications in processing engineering. We have selected some experimentally measured relative viscosity data from representative studies of different-sized TiO₂ [29,20,42,43,31,32], γ -Al₂O₃ [29,34,30,33,44,45,43], and SiO₂ [34,44] nanoparticles dispersed in water, and have compared the experimentally measured (data points) and calculated (lines) relative viscosity vs. nanoparticle volume content for each type of metal oxide as shown in Figs. 1–3, respectively.

To allow a strict comparison, all the data selected were measured at a temperature between 20 and 25 °C. Figs. 1–3 comprise a double triad of data, i.e. three different groups of literature data points for TiO₂, γ -Al₂O₃ and SiO₂ nanofluids, with lines resulting from the data being subjected to the various reported theoretical

and empirical equations (Fig. 1a, 2a, 3a) and to Chen's equation (Fig. 1b, 2b, 3b).

If we examine the data points, then it becomes clear that in general, the relative viscosity increases with the nanoparticle content, but the increment does not vary with particle size consistently. The relative viscosity ratio also varies with particle composition, which in turn affects the particle surface properties and its interaction with the base liquid. The calculated lines derived from the Einstein and Pak and Cho equations, included in Fig. 1a, 2a and 3a, represent the low and high boundaries of the experimental data in the studied concentration range, respectively. The lines calculated by the equations of Brinkman and Batchelor are very close to that of the Einstein equation; in fact, they virtually overlap at the scale used, thus can be considered as similar in the discussion (*vide infra*), hence only the Einstein line has been included in the figures. Fig. 1a, 2a and 3a show that when the nanoparticle volume content is less than 1 vol%, the calculated relative viscosity given by the Einstein equation is close to the experimental data, which is consistent with the Einstein equation being valid for spherical particles at a content less than 1 vol%. Consequently, when the nanoparticle volume content is above 1 vol%, all data points, even the low boundary of experimental values, lie significantly above the Einstein line.

The relative viscosity varying with nanoparticle volume content calculated using the Pak and Cho equation shows a line at the high boundary of the experimental data (Fig. 1a, 2a and 3a). The Pak and Cho equation has a similar form to that of the Batchelor equation, but the constants were empirically derived from experimental data of γ -Al₂O₃ and TiO₂ nanoparticle aqueous suspensions in the concentration range of 1–3 vol%. The Pak and Cho line fits better with the high boundary of the experimental data of Al₂O₃ (Fig. 2a) and SiO₂, (Fig. 3a), while it lies slightly higher than the data points for TiO₂ (Fig. 1a). While this supports the experimental findings that γ -Al₂O₃ nanoparticles provide better viscosity enhancement than TiO₂ [34,44,32,29], the theory is unable to satisfactorily model the difference in data for these different metal oxides. It is particularly noteworthy that the data points located between the Einstein and Pak and Cho lines in Fig. 1a, 2a and 3a could not be described or predicted by any of these reported equations.

The present analysis and discussion of literature data show the limitation of the relative viscosity equations of Einstein [16], Brinkman [35], Batchelor [17] and Pak and Cho [19] in modelling the experimental data. The origin of this discrepancy lies in the fact that these equations relate the relative viscosity only to nanoparticle content, but do not reflect particle size, shape and/or chemical composition nor the interaction between particles or particles to the base liquid. The important implication is therefore, that these

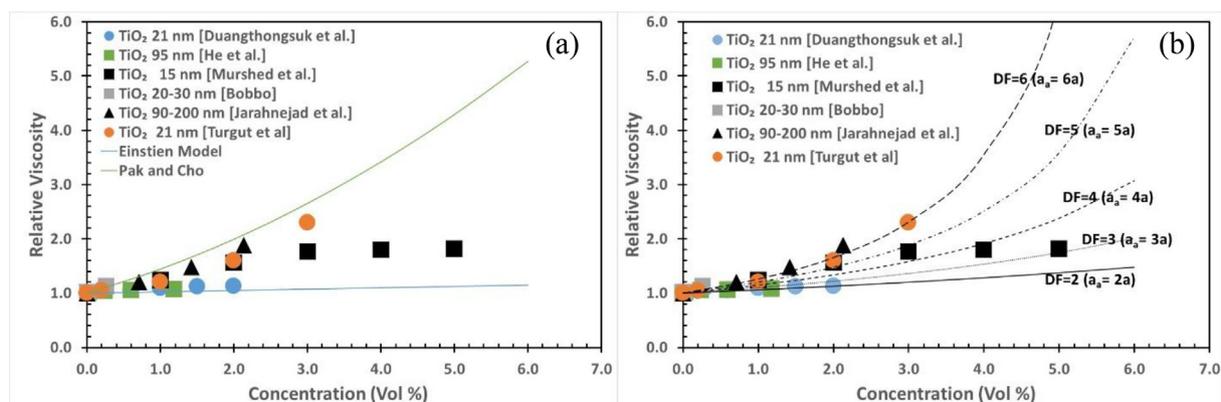


Fig. 1. The experimental relative viscosity data (data points) for different TiO₂ nanofluids vs. their concentration (in vol%) compared to the calculated lines based on (a) the Einstein model and the Pak and Cho equation, and (b) Chen equation that includes the Dispersion Factor (DF) parameter.

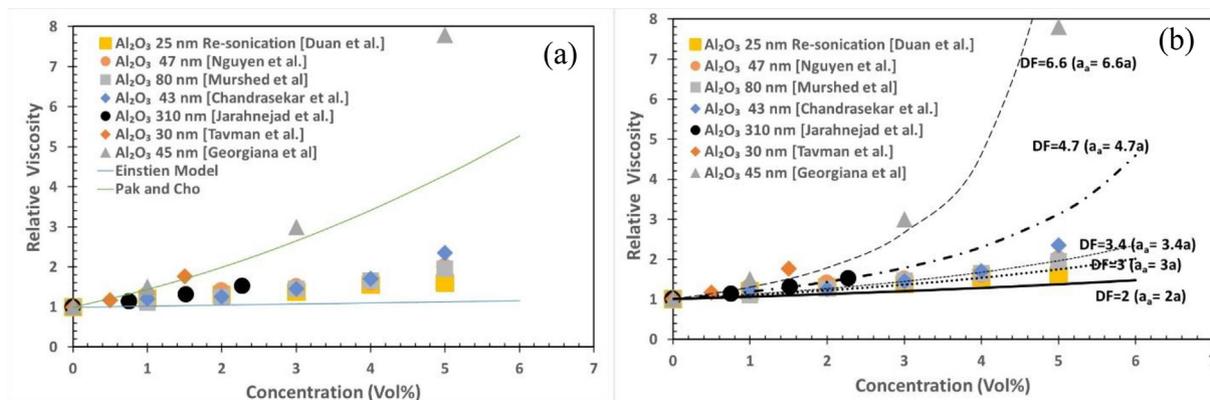


Fig. 2. The experimental relative viscosity data (data points) for different γ - Al_2O_3 nanofluids vs. their concentration (in vol%) compared to the calculated lines based on (a) the Einstein model and the Pak and Cho equation, and (b) the adapted Chen equation that includes the Dispersion Factor (DF) parameter.

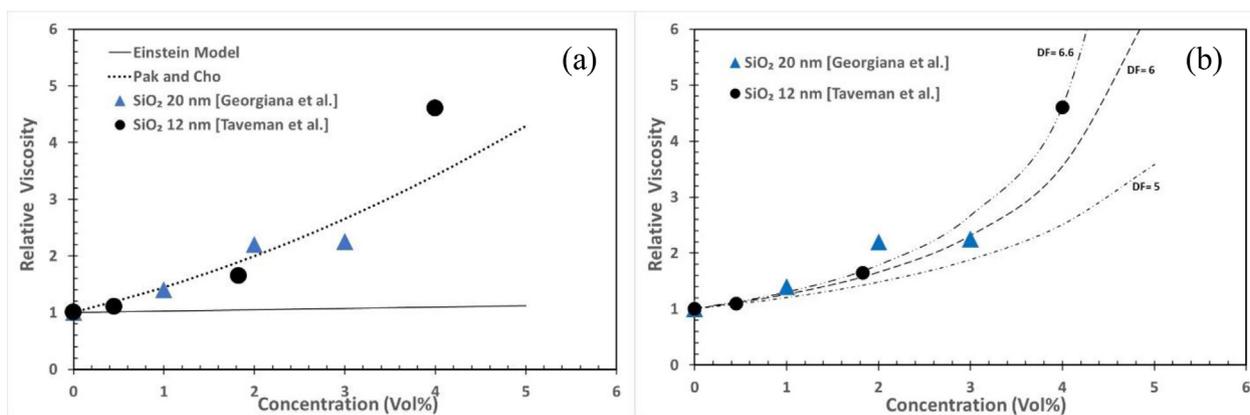


Fig. 3. The experimental relative viscosity data (data points) for different SiO_2 nanofluids vs. their concentration (in vol%) compared to the calculated lines based on (a) the Einstein model and the Pak and Cho equation, and (b) the adapted Chen equation that includes the Dispersion Factor (DF).

equations cannot realistically distinguish the viscosity enhancement resulting from a difference in particle size, shape and/or chemical composition nor can it take into account the interaction between nanoparticles or interactions between a nanoparticle to the base liquid. Further complications arise when nanoparticles aggregate, or agglomerate in the suspension; in such situations, the modelling becomes even more challenging.

Particle dispersion or aggregation/agglomeration is a major occurrence in suspension preparation. It is for this reason that Chen et. al. (2007) modified the Krieger-Dougherty equation (1959) by including a factor called agglomerate ratio (the ratio of agglomerate radius to that of the nanoparticle) to reflect the effect of particle agglomeration on the relative viscosity. We have used and expanded this approach by assigning different numerical values to the agglomerate ratio, which resulted in the various calculated lines showing the variation of the relative viscosity with nanoparticle volume content that is drawn in Fig. 1b, 2b and 3b. The experimental data can thus be described by a line with an agglomerate ratio in the range of 2 to 6.6.

It may be tedious or even premature to assign a particular Diffusion Factor (DF) value to a set of data originating from a particular study as the number of recorded data points is rather limited. To shed further light on this, more comprehensive experimental studies are required (and may become available in the future). However, at this stage, a focus on the general patterns can be offered.

In a general sense, the results could be interpreted by considering that the relative viscosity increases with the agglomerate ratio

at a given nanoparticle volume content; and the larger the aggregation, the larger the viscosity enhancement would be. Intriguingly, this conflicts with the general understanding that 1) the smaller particle size corresponds to a higher particle population for a given nanoparticle concentration thus resulting in a shorter distance between the nanoparticles and 2) the high nanoparticle surface area to its volume ratio boosts the interaction between the nanoparticle surface and the base liquid via the large interaction surface area. The consequence is that the van der Waals forces between the nanoparticles, as well as those between the nanoparticle and the base fluid, become larger when the nanoparticle is smaller, which leads to a large viscosity increase if the nanoparticles are dispersed well apart from one another in the base liquid.

Chen et.al. (2007) defined the agglomerate ratio as the quotient of agglomerate radius to nanoparticle radius, hence in the Chen equation the agglomerate ratio is strictly related to a radius or size ratio, rather than being correlated with the nanoparticle size itself or the chemical composition of the nanoparticle.

Our analysis suggests that the term agglomerate ratio can instead be represented by a parameter that has a pure numerical value. This treatment will enlarge the applicable scope of the model by including the effect of actual nanoparticle sizes and chemical compositions via the interaction between nanoparticles and the nanoparticles to the base liquid. To reflect the more comprehensive nature of this new ratio, we would like to suggest the new term Dispersion Factor (DF) that is to be included in Equation (7), thus yielding Equation (8).

$$\frac{\eta_{\text{eff}}}{\eta_{\text{bf}}} = \left(1 - \frac{\varphi}{0.605} (DF)^{1.2}\right)^{-1.5125} \quad (8)$$

In this equation, the DF is represented by a pure numerical value, which comprehensively reflects the effect of the nanoparticles in the base liquid and their interactions with the base liquid and themselves on the relative viscosity. The significance is that the value of DF can be determined from the experimental data, and its value captures crucial information in the following manner:

- If the modelling of experimental data gives a larger DF value, it means that there is 1) a smaller size and larger aspect ratio of nanoparticles; 2) a stronger interaction of particles to the base liquid and among themselves, or more complicatedly 3) the formation of secondary functional entities formed from the nanoparticles as, e.g. aggregates. The larger the aspect ratio of the secondary functional entities, the more significant their effect on the relative viscosity.
- When the nanoparticle sizes are comparable and the nanoparticles are suspended in the same base liquid, the DF value difference will reflect the relative interaction strength of the nanoparticles to the base liquid, as shown by the modelling of the data in Fig. 1b, 2b and 3b. The magnitude of the Dispersion Factor, therefore, can be used to evaluate the dispersion condition of nanoparticles in the base liquid or the interaction strength of the nanoparticle surface to the base fluid.

4. Conclusion

It has been known for a long time that adding a small number of nanoparticles to a base liquid causes a viscosity increase. This viscosity increase is explained via the relative viscosity by the equations of Einstein [16], Batchelor [17] and Krieger and Dougherty [18] and so on.

In our work, concentration-dependent relative viscosity data for TiO₂, γ-Al₂O₃ and SiO₂ nanofluids have been selected from the open literature, and the data has been subjected to the various reported theoretical and empirical equations. However, it appeared that the experimentally measured values of metal oxide nanofluids tend to be significantly higher than the calculated ones in the particle volume content range studied (less than 5%). Though the Pak and Cho equation [19], an empirical equation, reproduces the higher-end values of the experimental measurements to some extent, it is however rather unsuccessful in modelling the large number of experimental data whose values are lower than the higher-end values towards that estimated by, e.g. the Einstein equation. Our new theoretical treatment using the adaptation of Chen's equation [36] with the newly defined Dispersion Factor (DF) yielded a much better agreement with the wide range of experimental data. The DF value comprehensively includes the number and magnitude of the interactions between the nanoparticles themselves and with the base liquid and thus reflects the strength of these interactions. It enables to guide relevant practical work towards the directed obtaining of nanofluids with a desired, predetermined behaviour just by measuring viscosity enhancement, e.g. to examine the dispersity of nanoparticles in a working condition of the fluid rather than by the more elaborate method of using TEM in a dry state or DLS in an extensively diluted state.

By assigning the DF with a series of numerical values from 1 upwards, the literature data can be represented by the lines calculated with a DF value using the modified Chen equation. The DF values comprehend the effect of particle size/shape, and chemical composition via the interactions of particle surface (including pores) to the base liquid and particle to particle on the viscosity of the nanofluid. A larger DF value represents a stronger influence of the nanoparticles on the viscosity increment.

The DF is therefore a significant parameter describing the combined particle interactions and particle aggregations in nanofluids. Importantly, its value can be experimentally determined, which also means that it can be experimentally tailored and tuned to suit an application for which the nanofluid will be designed. Our ongoing work is testing the use and application of the DF parameter in the design of charcoal-based nanofluids to be used for enhanced oil recovery.

CRediT authorship contribution statement

Ifeoluwa Akande: Writing – original draft. **Tony Bridgwater:** Supervision, Validation. **Petra J. van Koningsbruggen:** Validation. **Qingchun Yuan:** Conceptualization, Methodology, Supervision.

Data availability

No data was used for the research described in the article.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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