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# Viscosity of multimodal concentrated suspensions in a Newtonian fluid.

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ABSTRACT. A new theoretical formulation about the viscosity of a multimodal concentrated suspension of non-colloidal spherical rigid particles in a Newtonian fluid is presented: the relative viscosity can be linked both to the volume fraction of the suspended particles in a total volume unity and to the solid fraction of the dry mixture. Therefore, it takes into account hydrodynamic interactions and geometrical interactions between particles. Concerning the first ones, it resorts to an iterative approach by change of scale method according to the concept of Farris. Concerning the second ones, when the volume fraction of the solid skeleton which is predicted by the Compressible Packing Model (CPM). This one takes into account both the loosening effect on big particles by interstitial small ones and the wall effect within assemblies of small particles near a big one. Our predicted viscosities are in good agreement with experimental data of relative viscosities obtained on binary mixtures with 3 different size ratios, for glass beads in silicone oil.

RÉSUMÉ. Une nouvelle formulation théorique concernant une suspension concentrée multimodale de particules sphériques rigides non-colloïdales dans un fluide Newtonien est présentée: la viscosité relative peut être reliée à la fois à la fraction volumique des particules suspendues dans un volume total unité et à la compacité du mélange sec. Elle tient compte des interactions hydrodynamiques et des interactions géométriques entre particules. Concernant les premières, elle fait appel à une approche itérative de type changement d'échelle conformément au concept de Farris. Concernant les secondes, lorsque la fraction volumique des particules suspendues atteint sa valeur critique, la suspension devient empilement et le mélange atteint la compacité du squelette solide prédite par le Modèle d'Empilement Compressible (MEC). Celui-ci tient compte à la fois de l'effet de desserrement provoqué par les fines particules sur les grosses et de l'effet de paroi provoqué par ces dernières sur les plus petites. Nos viscosités calculées sont en bonne adéquation avec les viscosités relatives expérimentales obtenues sur des mélanges binaires avec 3 rapports de tailles différents, pour des billes de verre dans une huile de silicone.

KEY WORDS: Relative viscosity, concentrated suspension, change of scale method, solid fraction, Compressible Packing Model (CPM), spheres.

MOTS-CLÉS : Viscosité relative, suspension concentrée, méthode de changement d'échelle, compacité, Modèle d'Empilement Compressible (MEC), sphères.

#### 1. Introduction.

Knowledge of the viscosity of concentrated suspensions involves a significant number of sectors like biology and medicine (human red cells), manufacturing and materials science (nanopowder systems), mineral processing (drilling fluids), soil science (flow of sediments). In civil engineering, a multitude of heterogeneous materials, like concrete, bitumen binder, are easy to flow if their viscosity is low. At the same time, a dense mixture must be sought, not only to reach a good strength, but also with the aim of decreasing the permeability of the hardened material. The study of the concentrated suspensions viscosity allows to optimize the composition of the granular skeleton while assuring a good workability necessary to be placed easily. The influence of the particle size ratio and of the volume fraction of the fine class on the relative viscosity of a bimodal suspension has been recently investigated by Qi and Tanner [QI 11]. Their approach presents two inconveniences. The first one is that the curve representing the maximum volume fraction of the bidisperse system as a function of the volume fraction of the fine particles is too smooth when compared with numerical data (see for example [ROQ 16] for 0.2 and 0.4 size ratios). The second one is that their study is limited to a bimodal case. The main objective of this study is to highlight a new relation between the relative viscosity  $\eta_r$  of a multimodal concentrated suspension and both the

volume fraction of the suspended particles in a total volume unity and the solid fraction of the dry mixture. We shall focus on an ideal suspension compound of spherical, inert and rigid particles in a Newtonian fluid. The predictions of the new model are compared with available experimental data from [STO 87] for three different size ratios of glass beads in silicone oil.

#### 2. The concept of Farris.

Farris (1968) [FAR 68] proposed a theoretical treatment for the prediction of the viscosity of multimodal suspensions if each class size is completely independent of the others:  $d_1 >> d_2 \dots >> d_n$ . In this case, all smaller spheres added to the suspending fluid behave towards the larger ones as a viscous continuous phase. Farris extends the theory developed by Einstein (1906) [EIN 06] by considering that the relative viscosity depends only on the solid volume concentration in the liquid:

$$\eta_{\rm r} = H(\phi)$$
[1]

Let us carry out our reasoning (Stovall, Buil, Such [STO 87]) on a suspension constituted by two spherical classes with diameters  $d_1 >> d_2$  occupying volumes  $V_1$  and  $V_2$ .  $V_0$  represents the volume of the suspending fluid. The finest class is first introduced. Its relative viscosity  $\eta_{r2}$  can be expressed as follows:

$$\eta_{r_2} = H(\psi_2, \psi_2^{MAX}) \text{ where } \psi_2 = \frac{V_2}{(V_0 + V_2)}$$
 [2]

In this precise case,  $\psi_2^{MAX}$  corresponds to the maximal volume fraction of the class 2 of the packing constituted by uniform-sized spherical particles. Let us now add the component with size  $d_1$ . As the condition of separation of scales is checked according to the Farris' assumption, the model of unimodal suspension can be used to predict the viscosity variation linked to the introduction of this new class. The final suspension has thus a relative viscosity corresponding to the product of the relative viscosities associated with each class. Assuming that the mathematical form of the law H is always valid, the viscosity of the suspension becomes:

$$\eta_{r1} = \eta_{r2} H(\psi_1, \psi_1^{MAX}) = H(\psi_1, \psi_1^{MAX}) \times H(\psi_2, \psi_2^{MAX}) \text{ where } \psi_1 = \frac{V_1}{(V_0 + V_1 + V_2)}$$
 [3]

 $\psi_1^{MAX}$  is the maximal volume fraction of the class 1 that can be added in the suspension before causing its jamming, taking account of the presence of the finest class 2:  $\psi_1^{MAX} = \text{function}(\psi_2)$ . Let us generalize this reasoning for the n-components.

$$\eta_{r} = \prod_{i=1}^{n} H\left(\psi_{i}, \psi_{i}^{MAX}\right)$$

$$[4]$$

by pointing out that  $\psi_i^{MAX}$  depends on the presence of the finest classes:  $\psi_i^{MAX} = \text{function}(\psi_{i+1}, \dots, \psi_n)$ . As the viscosity must diverge when  $\psi_i \rightarrow \psi_i^{MAX}$ , it is better to write the relative viscosity as follows:

$$\eta_{r} = \prod_{i=1}^{n} H\left(\frac{\psi_{i}}{\psi_{i}^{MAX}}\right)$$
[5]

#### 3. Binary mixture without geometrical interactions: apollonian model.

We consider  $d_1 >> d_2$ . Let us call  $\beta$  the dry solid fraction of each monosize class, supposed to be constant. We start by identifying the "dominant coarse particles" field and the "dominant fine particles" field where each class respectively constitutes the matrix of the dry mixture.

In the first case, coarse particles fill the mold without being disturbed by the finest ones. Their volume fraction in a total volume unity is  $\phi_1$ , their critical value being  $\phi_1^*$ . We have then to verify that:

$$\phi_1 \le \phi_1^* = \beta \tag{6}$$

In the second case, fine particles fill the porosity of coarse particles. Concerning  $\phi_2$  and  $\phi_2^*$ , we have to respect the following condition:

$$\phi_2 \le \phi_2^* = \beta \left( 1 - \phi_1 \right)$$

$$[7]$$

We can then write that:

$$\psi_{1}^{MAX} = \phi_{1}^{*} = \beta, \quad \psi_{2}^{MAX} = \frac{\phi_{2}^{*}}{\left(1 - \phi_{1}\right)} = \beta \quad [8]$$

As no geometrical interaction occurs, maximal volume fractions of classes 1 and 2 are equal to the solid fraction of each monosize class. Let us set  $\frac{V_j}{\sum_{k=0}^{n-2} V_k} = \phi y_j$  where  $y_{j\neq 0}$  is the volume fraction of the class j by  $\sum_{k=0}^{n-2} V_k$ 

reference of the total solid volume and  $\phi$  the volume fraction of the suspended particles in a total volume unity. By expressing  $\psi_1$  and  $\psi_2$  as functions of  $\phi$ ,  $y_1$  and  $y_2$ , the relative viscosity can be calculated as follows:

$$\eta_{r} = H\left(\frac{\psi_{1}}{\psi_{1}^{MAX}}\right) \times H\left(\frac{\psi_{2}}{\psi_{2}^{MAX}}\right) \text{ with } \psi_{1} = \phi y_{1}, \ \psi_{2} = \frac{y_{2}}{\left(\frac{1}{\phi} - y_{1}\right)}, \ \psi_{1}^{MAX} = \beta, \ \psi_{2}^{MAX} = \beta$$
[9]

#### 4. Binary mixture with geometrical interactions.

The objective now is to combine the concept of Farris and the Compressible Packing Model (CPM) from de Larrard [DEL 00] which takes into account the geometrical interactions. To this end, we are going to follow the scientific process used by Bournonville, Coussot, Chateau [BOU 05]. The approach adopted by Stovall, Buil, Such [STO 87] is indeed incomplete in the sense that volume fractions of each class don't change when seeking to quantify the maximal partial volume of each ones when causing the jamming. However, during this operation, a certain amount of fluid will be deleted, the solid phase remaining unchanged. The developed reasoning is thus inappropriate with the concept of Farris which proceeds by successive incorporations. The flow can then be jammed when a supplementary class is introduced if a sufficient number of particles is added. For the concept of Farris to be valid, volumes of granular classes already present in the suspension need to be maintained constant when a volume  $\delta V_i$  of a class i, supposed to be the dominant one, is progressively added. As long as the jamming does not occur, the solid phase can not be considered as a packing. According to the CPM from de Larrard, when the class 1 (coarse particles) is dominant, we have to verify that:

$$\phi_1 \le \phi_1^* = \beta - a_{12} \phi_2$$
 [10]

where  $a_{12}$  is the loosening effect coefficient (Figure 1).

In this case:

$$\phi_{1} = \frac{\left(V_{1} + \delta V_{1}\right)}{\left(V_{0} + V_{1} + V_{2} + \delta V_{1}\right)} \text{ and } \phi_{2} = \frac{V_{2}}{\left(V_{0} + V_{1} + V_{2} + \delta V_{1}\right)} \text{ (dominant class 1)}$$
[11]

When the class 2 (fine particles) is dominant, the relationship can be written:

$$\phi_2 \le \phi_2^* = \beta (1 - \phi_1) - (1 - \beta) b_{21} \phi_1$$
 [12]

where  $b_{21}$  is the wall effect coefficient (Figure 1).

In this case:

$$\phi_{1} = \frac{V_{1}}{\left(V_{0} + V_{1} + V_{2} + \delta V_{2}\right)} \text{ and } \phi_{2} = \frac{\left(V_{2} + \delta V_{2}\right)}{\left(V_{0} + V_{1} + V_{2} + \delta V_{2}\right)} \text{ (dominant class 2)}$$
[13]

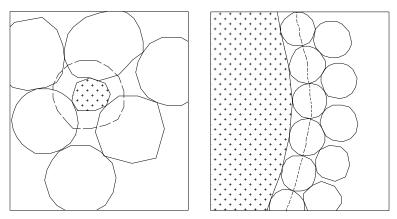


Figure 1. Illustrations of the loosening effect (left) and the wall effect (right).

From the precedent relationships, we can deduce the maximal volume  $\Delta V_i$  of the class i that can be added to jam the suspension:

$$\Delta \mathbf{V}_{1} = \frac{1}{\left(1-\beta\right)} \left(\beta \sum_{j=0}^{n-2} \mathbf{V}_{j} - \mathbf{V}_{2} \mathbf{a}_{12} - \mathbf{V}_{1}\right)$$
 if the class 1 is dominant [14]

$$\Delta V_2 = \frac{1}{(1-\beta)} \left( \beta \sum_{j=0}^{n-2} V_j - V_1 \left( \beta + (1-\beta) b_{21} \right) - V_2 \right)$$
 if the class 2 is dominant [15]

The value  $\Delta V_1$  takes into account the loosening effect from fine particles of the class 2 on coarse particles of the class 1. The value  $\Delta V_2$  takes into account the wall effect from coarse particles on finer ones. When the jamming occurs, we can deduce that:

$$\phi_1^* = \frac{(V_1 + \Delta V_1)}{(V_0 + V_1 + V_2 + \Delta V_1)} \text{ and } \phi_2 = \frac{V_2}{(V_0 + V_1 + V_2 + \Delta V_1)} \text{ (dominant class 1)}$$
[16]

$$\phi_{1} = \frac{V_{1}}{(V_{0} + V_{1} + V_{2} + \Delta V_{2})} \text{ and } \phi_{2}^{*} = \frac{(V_{2} + \Delta V_{2})}{(V_{0} + V_{1} + V_{2} + \Delta V_{2})} \text{ (dominant class 2)}$$
[17]

It is thus possible to give a mathematical definition to the partial volumes used in the calculation of the relative viscosity:

$$\psi_{1} = \frac{V_{1}}{(V_{0} + V_{1} + V_{2})}, \quad \psi_{2} = \frac{V_{2}}{(V_{0} + V_{2})}, \quad \psi_{1}^{MAX} = \frac{(V_{1} + \Delta V_{1})}{(V_{0} + V_{1} + V_{2} + \Delta V_{1})}, \quad \psi_{2}^{MAX} = \frac{(V_{2} + \Delta V_{2})}{(V_{0} + V_{2} + \Delta V_{2})}$$
[18]

The expressions of  $\Delta V_1$  and  $\Delta V_2$  provided above lead to:

$$\psi_{1}^{MAX} = \frac{\left(\beta V_{0} + V_{2} \left(\beta - a_{12}\right)\right)}{\left(V_{0} + V_{2} \left(1 - a_{12}\right)\right)}, \quad \psi_{2}^{MAX} = \frac{\left(\beta V_{0} - V_{1} b_{21} \left(1 - \beta\right)\right)}{\left(V_{0} - V_{1} b_{21} \left(1 - \beta\right)\right)}$$
[19]

By dividing each volume by  $\sum_{k=0}^{n=2} V_k$ , by setting  $\frac{V_j}{\sum_{k=0}^{n=2} V_k} = \phi y_j$  and by taking into account that  $\sum_{j=1}^{n=2} y_j = 1$ , we can

express  $\psi_1\,,\,\psi_2\,,\,\psi_1^{MAX}$  and  $\psi_2^{MAX}$  as functions of  $\varphi$  :

$$\psi_{1} = \phi \ y_{1}, \ \psi_{2} = \frac{y_{2}}{\left(\frac{1}{\phi} - y_{1}\right)}, \ \psi_{1}^{MAX} = \frac{\left(\beta\left(\frac{1}{\phi} - 1\right) + y_{2}\left(\beta - a_{12}\right)\right)}{\left(\frac{1}{\phi} - 1 + y_{2}\left(1 - a_{12}\right)\right)}, \ \psi_{2}^{MAX} = \frac{\left(\beta\left(\frac{1}{\phi} - 1\right) - y_{1} \ b_{21}\left(1 - \beta\right)\right)}{\left(\frac{1}{\phi} - 1 - y_{1} \ b_{21}\left(1 - \beta\right)\right)}$$
[20]

According to the CPM, the solid fraction  $\gamma$  of a dry binary mixture is calculated by using the expression of  $\gamma_1$  in the "coarse dominant" field and  $\gamma_2$  in the "fine dominant" field:

$$\gamma = \min(\gamma_1, \gamma_2), \ \gamma_1 = \frac{\beta}{(1 - (1 - a_{12})y_2)}, \ \gamma_2 = \frac{\beta}{(1 - (1 - \beta)(1 - b_{21})y_1)}$$
[21]

By combining precedent relationships, it is possible to obtain  $\psi_1$ ,  $\psi_2$ ,  $\psi_1^{MAX}$  and  $\psi_2^{MAX}$  as functions of  $\beta$ ,  $\phi$   $y_1$ ,  $y_2$  and of solid fractions  $\gamma_1$  and  $\gamma_2$ :

$$\eta_{r} = H\left(\frac{\psi_{1}}{\psi_{1}^{MAX}}\right) \times H\left(\frac{\psi_{2}}{\psi_{2}^{MAX}}\right)$$

$$\psi_{1} = \phi y_{1}, \quad \psi_{2} = \frac{y_{2}}{\left(\frac{1}{\phi} - y_{1}\right)}, \quad \psi_{1}^{MAX} = \frac{\left(\beta\left(\frac{1}{\phi} - \frac{1}{\gamma_{1}}\right) + y_{1}\left(1 - \beta\right)\right)}{\left(\frac{1}{\phi} - \frac{\beta}{\gamma_{1}}\right)}, \quad \psi_{2}^{MAX} = \frac{\left(\beta\left(\frac{1}{\phi} - \frac{1}{\gamma_{2}}\right) + y_{2}\left(1 - \beta\right)\right)}{\left(\frac{1}{\phi} - \frac{\beta}{\gamma_{1}} - y_{1}\left(1 - \beta\right)\right)}$$

$$[22]$$

The advantage of these new expressions consists in highlighting a direct link between the relative viscosity  $\eta_r$ , even if the stiffening function is not completely defined, the volume fraction  $\phi$  of the suspended particles in a total volume unity, and the solid fraction of the dry binary mixture  $\gamma_1$  or  $\gamma_2$ . According to the approach adopted by Bournonville, Coussot, Chateau [BOU 05], it is now possible to create a suspension whose the solid phase is composed like a dry granular material with a solid fraction predicted by the CPM. When the latter is maximal by optimization of the granular skeleton, the critical solid fraction value is reached, the flow is jammed and the suspension becomes a packing. This "solid fraction - viscosity" association could also allow to improve the formulation of a liquefied concrete, for example, by following two steps: minimizing its porosity, i.e. the water volume in a volume unity, for a set viscosity but non-infinite.

#### 5. Ordinary mixture composed by n-classes.

Let us now study an ordinary mixture composed by n-classes of particles. Let us suppose the i<sup>th</sup> class be the dominant one by taking into account the wall effect from coarsest particles and the loosening effect from finest ones. Its volume fraction in a total volume unity  $\phi_i$  must be lower than its critical value  $\phi_i^*$ :

$$\phi_{i} \leq \phi_{i}^{*} = \beta - \sum_{j=1}^{i-1} \left( \beta + b_{ij} \left( 1 - \beta \right) \right) \phi_{j} - \sum_{j=i+1}^{n} a_{ij} \phi_{j}$$
[23]

where  $a_{ij}$  indicates the loosening effect coefficient and  $b_{ij}$  the wall effect coefficient.

 $\phi_i$  and  $\phi_{i\neq i}$  are calculated as follows when a volume  $\delta V_i$  of the class i is added:

$$\phi_{i} = \frac{\left(V_{i} + \delta V_{i}\right)}{\left(\sum_{k=0}^{n} V_{k} + \delta V_{i}\right)} \xrightarrow{\text{and}} \phi_{j\neq i} = \frac{V_{j}}{\left(\sum_{k=0}^{n} V_{k} + \delta V_{i}\right)} \text{ (dominant class i)}$$

$$[24]$$

From this system of equations, we can now deduce the maximal volume  $\Delta V_i$  of this class i to be added to jam the suspension.

$$\Delta \mathbf{V}_{i} = \frac{1}{\left(1-\beta\right)} \left(\beta \sum_{j=0}^{n} \mathbf{V}_{j} - \sum_{j=1}^{i-1} \mathbf{V}_{j} \left(\beta + \left(1-\beta\right) \mathbf{b}_{ij}\right) - \sum_{j=i+1}^{n} \mathbf{V}_{j} \mathbf{a}_{ij} - \mathbf{V}_{i}\right) \text{ if the class } i \text{ is dominant}$$
 [25]

The value  $\Delta V_i$  takes into account both the wall effect and the loosening effect. When the jamming occurs, we can write:

$$\phi_i^* = \frac{\left(V_i + \Delta V_i\right)}{\left(\sum_{k=0}^n V_k + \Delta V_i\right)} \text{ and } \phi_{j\neq i} = \frac{V_j}{\left(\sum_{k=0}^n V_k + \Delta V_i\right)} \text{ (dominant class i)}$$

$$[26]$$

It is now possible to define  $\psi_i$  and  $\psi_i^{MAX}$  of the class i in a mixture only constituted by finest classes, according to the concept of Farris:

$$\psi_{i} = \frac{V_{i}}{\left(V_{0} + \sum_{j=i}^{n} V_{j}\right)}, \quad \psi_{i}^{MAX} = \frac{\varphi_{i}^{*}}{\left(1 - \sum_{j=1}^{i-1} \varphi_{j}\right)} = \frac{\left(V_{i} + \Delta V_{i}\right)}{\left(V_{0} + \sum_{j=i}^{n} V_{j} + \Delta V_{i}\right)}$$
[27]

We can finally express  $\psi_i$  and  $\psi_i^{MAX}$  as follows:

$$\psi_{i} = \frac{y_{i}}{\left(\frac{1}{\varphi} - \sum_{j=1}^{i-1} y_{j}\right)}, \quad \psi_{i}^{MAX} = \frac{\left(\beta\left(\frac{1}{\varphi} - 1\right) - (1 - \beta)\sum_{j=1}^{i-1} y_{j} b_{ij} + \sum_{j=i+1}^{n} y_{j}(\beta - a_{ij})\right)}{\left(\frac{1}{\varphi} - 1 - (1 - \beta)\sum_{j=1}^{i-1} y_{j} b_{ij} + \sum_{j=i+1}^{n} y_{j}(1 - a_{ij})\right)}$$
[28]

According to the CPM, the solid fraction of a dry granular mixture composed by n-classes when the class i is dominant is:

$$\gamma = \gamma_{i} = \frac{\beta}{\left(1 - (1 - \beta)\sum_{j=1}^{i-1} (1 - b_{ij}) y_{j} - \sum_{j=i+1}^{n} (1 - a_{ij}) y_{j}\right)}$$
[29]

We can then determine the relative viscosity:

$$\eta_{r} = \prod_{i=1}^{n} H\left(\frac{\psi_{i}}{\psi_{i}^{MAX}}\right), \quad \psi_{i} = \frac{y_{i}}{\left(\frac{1}{\varphi} - \sum_{j=1}^{i-1} y_{j}\right)}, \quad \psi_{i}^{MAX} = \frac{\left(\beta\left(\frac{1}{\varphi} - \frac{1}{\gamma_{i}}\right) + y_{i}\left(1 - \beta\right)\right)}{\left(\frac{1}{\varphi} - \frac{\beta}{\gamma_{i}} - \left(1 - \beta\right)\sum_{j=1}^{i-1} y_{j}\right)} \quad [30]$$

The expression of the relative viscosity diverges to infinity as  $\phi$  goes to  $\gamma_i$  when the class i is dominant: the suspension becomes a packing. However, it is necessary to be conscious of the following paradox that may occur [BOU 05]: the concept of Farris assumes additions of successive classes by change of scale. The divergence can occur when the class i is introduced because this one reaches its critical volume fraction, the following classes having not yet been incorporated. However, the volume  $\Delta V_i$  is calculated by taking into account geometrical interactions with all the granular classes, including those not present yet in the suspension when the divergence occurs. The expression allowing to determine the relative viscosity must then be used carefully.

#### 6. Experimental verifications on binary mixtures.

Experimental data concerning binary mixtures are those presented by [STO 87]. They concern glass beads classified in rather narrow granular classes. The suspension liquid is a silicon oil with a viscosity equal to 17.9 Pa.s for a temperature of 20°C. It exhibits a Newtonian behaviour. Measurements are performed in coaxial cylinders.

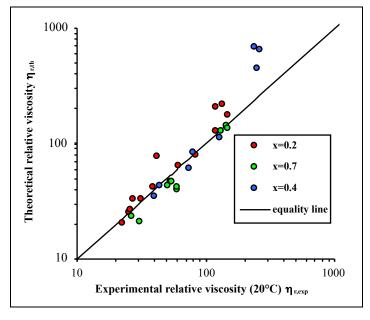
The relative viscosity is calculated by a power-law relation (Krieger-Dougherty type [STO 87]):

$$\eta_{r} = \prod_{i=1}^{n-2} \left( 1 - \frac{\psi_{i}}{\psi_{i}^{MAX}} \right)^{-[\eta_{i}]^{B}}$$
[31]

where  $[\eta]_1 = 2.5$  is the first-order intrinsic viscosity for spheres [EIN 06] and  $\beta$  the solid fraction of each monosize class. The wall effect coefficient  $b_{21}$  and the loosening effect coefficient  $a_{12}$  are determined from the expressions of de Larrard [DEL 88] which are specifically adapted to spheres:

$$b_{21}(x) = 1 - (1 - x)^{1.6}, a_{12}(x) = 1 - (1 - x)^{3.1} - 3.1 x (1 - x)^{2.9}$$
 [32]

where x is the size ratio fine/coarse particles diameters. Each mixture being composed by spherical particles, the solid fraction of each monosize class is taken equal to  $\beta = 0.608$ , corresponding to a random loose packing [ARI 09].



**Figure 2.** *Bimodal suspensions: comparison between relative predicted viscosities and relative experimental values from [STO 87] with 3 size ratios (x=0.2, x=0.4, x=0.7), for glass beads in silicone oil.* 

Predictions of the viscosity model are, for the studied suspensions, in good agreement with experimental data by taking into consideration that the solid fraction of each monosize class  $\beta$  is only estimated, and by considering high experimental uncertainties for the tested concentration ranges. For them, the exponent of the Krieger-Dougherty law equal to  $-2.5\beta$  seems to be appropriate.

### 7. Upper and lower limits for $\eta_{r,th}$ as a function of $\phi/\phi_m$ for a binary mixture.

If now, the theoretical relative viscosity corresponding to the three size ratios x = 0.2, x = 0.7, x = 0.4 is represented for  $y_2 = 0.30$  as a function of the fraction  $\phi/\phi_m$ , called the *normalized total solid volume fraction*, the results show that the curves are very close together. However, this is not a single master curve (Figure 3).

An *upper limit* of  $\eta_{r,th}$  can be found for the mixture with the highest solid fraction: the apollonian model. In this case:

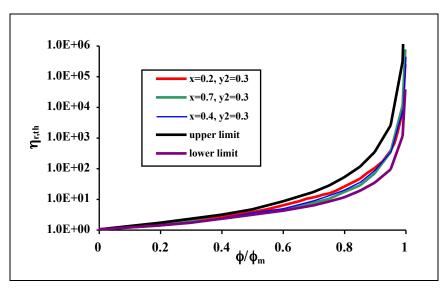
x = 0, b<sub>21</sub> = 0, a<sub>12</sub> = 0, 
$$\gamma_1 = \frac{\beta}{(1 - y_2)}, \gamma_2 = \frac{\beta}{(1 - (1 - \beta)y_1)}$$
 [33]

The highest solid fraction is reached when  $\gamma_1 = \gamma_2$  for which:

$$y_{1} = \frac{1}{(2-\beta)}, \quad y_{2} = \frac{(1-\beta)}{(2-\beta)}, \quad \phi_{m} = \beta \left(2-\beta\right), \quad \psi_{1} = \beta \frac{\phi}{\phi_{m}}, \quad \psi_{2} = \frac{(1-\beta)}{\left(\frac{1}{\beta}\left(\frac{\phi_{m}}{\phi}\right)-1\right)}, \quad \psi_{1}^{MAX} = \beta, \quad \psi_{2}^{MAX} = \beta$$
[34]

By using a power-law relation (Krieger-Dougherty type), we can deduce that:

$$\eta_{r,th}^{sup} = \left(1 - \phi / \phi_{m}\right)^{-[\eta]_{l}\beta} \times \left(1 - \frac{\left(1 - \beta\right)}{\left(\frac{1}{\phi / \phi_{m}} - \beta\right)}\right)^{-[\eta]_{l}\beta}$$
[35]



**Figure 3.** Evolution of the theoretical relative viscosity as a function of the normalized total solid volume fraction  $\phi / \phi_m$  for a volume fraction of the fine class  $y_2=0.30$  ( $\beta=0.608$ ).

A *lower limit* of  $\eta_{r,th}$  can also be found for the mixture with the lowest solid fraction when small particles are as fine as possible in infinitesimal quantity:

$$x = 0, b_{21} = 0, a_{12} = 0, y_1 \rightarrow 1, y_2 \rightarrow 0, \gamma_1 \rightarrow \beta, \gamma_2 \rightarrow 1, \phi_m \rightarrow \beta$$
 [36]

$$\psi_1 \rightarrow \phi, \ \psi_2 \rightarrow 0, \ \psi_1^{MAX} = \beta, \ \psi_2^{MAX} = \beta$$
 [37]

We can deduce that:

$$\eta_{r,th}^{inf} = \left(1 - \phi / \phi_m\right)^{-[\eta]_{f}\beta}$$
[38]

#### 8. Conclusion.

From a work of Bournonville, Coussot, Chateau, we have been able to develop a model of viscosity in a Newtonian fluid linking this latter to the volume fraction of the suspended particles in a total volume unity and to the solid fraction of the dry mixture. This theory takes into account both the hydrodynamic interactions, through the concept of Farris, and the geometrical interactions, through the CPM. However, to be compatible with these two approaches, the model initially proposed by Farris is modified to adopt the description of the wall effect and of the loosening effect used in the CPM. First results are positive because they give relatively good predictions about relative viscosities on suspensions of glass beads with three different size ratios. The model also allows to determine upper and lower limits for the theoretical relative viscosity as a function of  $\phi/\phi_m$  which

shows that a single master curve can not exist. Even if it requires improvements, this work could constitute an interesting comprehension tool to optimize granular skeletons for various kinds of materials where the densest mixture must be sought for a given viscosity.

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