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van Netten, K.; Zhou, J.; Galvin, K. P.; Moreno-Atanasio, R. "Influence of magnetic and hydrodynamic forces on chain-aggregation and motion of magnetisable particles and composites" Chemical Engineering Science Vol. 93, p. 229-237 (2013)

Available from: <http://dx.doi.org/10.1016/j.ces.2013.01.028>

Accessed from: <http://hdl.handle.net/1959.13/1049108>

Influence of Magnetic and Hydrodynamic Forces on Chain-Aggregation and Motion of Magnetisable Particles and Composites

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ABSTRACT

The motion of ferromagnetic single particles and linear aggregates of spheres subjected to a gradient magnetic field in the presence of a non-magnetic fluid was investigated through the experimental validation of a new theoretical model. The theoretical model combines the correction of the mutual magnetisation between particles as well as the interaction with the external magnetic gradient and the hydrodynamic drag corrections for linear aggregates for the first time. The drag for linear aggregates of spheres was corrected using a semiempirical equation which was developed from published work on the drag correction for linear clusters of particles.

The experimental results show that the ratio of the velocity of aggregates to the velocity of a single particle with the same diameter can be described by the aspect ratio of the aggregate. The solution for the motion equation obtained by numerical analysis showed an excellent agreement with the experimental data for linear aggregates of spheres, while very similar results were obtained for non-spherical particles.

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This study also examined the influence of the thickness of a non-magnetic coating around the ferromagnetic particles. Core-shell composites were created by dip-coating ferromagnetic particles in a non-magnetic material. When a pair of the core-shell composite particles (doublet) was formed, the velocity of the doublet was found to be lower than for the aggregates of the non-coated particles. This effect was examined as a function of the film thickness. When the thickness of the shell was approximately 50% of the particle radius, the velocity of the doublet was similar to that of a single uncoated particle.

This work establishes an understanding of the fundamental effects of thin coatings on magnetic aggregation. Particle coatings provide a novel and simple means for controlling the separation between the magnetic cores and in turn the kinetics of the aggregation and subsequent magnetisation of the final aggregates.

Keywords: composites; suspensions; magnetisation; mathematical modelling; particulate processes; drag corrections.

1. INTRODUCTION

Minerals and composite particles with magnetic properties are commonly found and used in a wide range of industrial and scientific processes such as mineral beneficiation (Lopez-Lopez, 2005, Svoboda 2004, Meshani et al. 2004, Garcia-Martinez et al. 2004), drug delivery (Nawwab et al. 2011, Yoshida et al. 2007, Weyh et al. 2004), magnetorheological fluids (Han et al. 2010) and the design of new materials with specific properties (Sacanna et al. 2012, Sacanna and Pine, 2011, Li et al. 2009; Lee and Lidell, 2009; Papaefthymiou, 2009; Ge et al. 2007).

Minerals and many other types of solids containing magnetic materials can aggregate, forming linear chains due to dipole-dipole interactions (Phatak et al. 2011; Lee et al. 2009; Zerrouki et al. 2008; Ge et al. 2007; Claesson et al. 2007; Veyret et al. 2005; Bizdoaca et al. 2002; Svoboda, 1982). These particulate solids can be separated from other existing components through aggregation and sedimentation or by means of an external magnetic field gradient. Some examples include separation of minerals (Nakai et al. 2011; Li et al. 2007; Murariu et al. 2005; Arol and Aydogan, 2004; Svoboda and Fujita 2003; Svoboda 1982), catalysts (Claesson et al. 2007) and particles used in the capture of viruses (Veyret et al., 2005).

However, in cases in which the particles need to fulfil a given role, magnetic aggregation (flocculation) may produce premature sedimentation of the particles before they have fulfilled their function (Martinez-Pedrero, 2008). Therefore, the hindrance and control of aggregation is highly desirable. This control is usually achieved by modification of the particle surfaces through the coating of particles with a monolayer of

molecules that exhibit specific physicochemical properties (Martinez-Pedrero 2007; Cousin et al. 2003). This coating can have a double functionality, to hinder the magnetic flocculation and to give to the composite particles a specific physicochemical functionality (Arsianti et al. 2011). Magnetic aggregation can also be controlled by the use of a bimodal particle size distribution which has proved to be an effective way of avoiding aggregation due to the creation of a steric barrier whilst keeping the required response to an external magnetic field (Lopez-Lopez et al. 2005, Viota et al. 2007).

The kinetics of magnetic aggregation is a fast process since the magnetic force of attraction between dipoles depends on the inverse of the 4th power of their separation distance (Zhang and Widom, 1995). Hence the force becomes increasingly significant just prior to aggregation and, thus, it is not possible to achieve sufficient resolution to study the particle motion just before particles collide against each other. Nevertheless, our previous studies have shown that the velocity of two iron spheres, located within a gradient magnetic field in the presence of a non-magnetic fluid (Zhou et al. 2008; van Netten *et al.* 2008) doubles due to the increase in magnetisation when the particles finally aggregate. In addition, as particles aggregate the magnetisation of each individual particle, and thus the interaction force with the external field, changes due to the proximity of other magnetic particles. This force is responsible for driving the motion of the aggregates in a direction perpendicular to their poles (Zhou et al. 2008; van Netten *et al.* 2008).

The present study extends our previous work to cover the behaviour of linear aggregates, non-spherical particles and composite spheres subjected to a magnetic field gradient. The variation in velocity with the aspect ratio of the particles and aggregates is

determined. The results are presented for different magnetic field gradients, pole geometries and vessel sizes.

We also focus on the behaviour of core-shell composite particles thus providing a way to control the kinetics of aggregation. A novel experimental method was developed using thin coatings on the particles to prevent mechanical contact between the magnetic cores. Therefore, the strength of the magnetic interaction, and in turn the aggregate motion, was influenced by the presence of the coating which acted as a steric barrier.

Therefore, the objective of this work is to help to understand the motion of particles and linear aggregates in magnetic field gradients. In order to achieve this objective we have developed a model of the motion of individual particle chains as a function of the number of particles in the chains. The new model will be of strong benefit in areas as diverse as mineral processing and drug delivery where magnetic gradients are used as a method to separate magnetic components and chain formation is often observed (Li et al. 2007, Yoshida et al. 2007; Weigh et al. 2004; Svoboda 2004).

2. THEORY

2.1 Motion in a gradient magnetic field

This study aims to provide a description of the motion of chains of spheres and cylindrical particles along the direction of a magnetic field gradient.

The net force acting on a particle can be written as the sum of the magnetic, F_{mag} , and drag, F_{drag} , forces acting on the particles. That is,

$$\vec{F} = \vec{F}_{mag} + \vec{F}_{drag} \quad (1)$$

The magnetic force acting on a particle located in a non-magnetic fluid in the direction of a magnetic field gradient is given in a scalar form (Zhenghua et al. 2010; Svoboda 2004):

$$F_{mag} = MV \frac{dB}{dx} \quad (2)$$

where M is the total magnetisation of the particles and db/dx is the magnetic field gradient. The total magnetisation depends on the external field and the mutual magnetisation of the particles. In this way the final particle velocity and acceleration are influenced by the mutual magnetisation of the particles, the magnetisation due to the external field and the magnetic field gradient.

At low particle Reynolds numbers and infinite dilution the hydrodynamic drag force varies linearly with the velocity of the sphere as given by Stokes Law as:

$$F_{drag} = 6\pi\eta aU \quad (3)$$

where η is the viscosity of the fluid, a the radius of the particle and U the velocity of the sphere relative to the fluid in the direction of interest.

Thus, the equation of motion in the direction perpendicular to the poles of the magnet can be written as a function of the particle density, ρ , and volume, V as:

$$\rho V \frac{dU}{dt} = MV \frac{dB}{dx} - 6\pi\eta a U \quad (4)$$

where the magnetisation of a single particle can be written as a function of the external magnetic field induction, B , the effective magnetic susceptibility of the particle material, χ_{eff} , and μ_0 , the vacuum permeability as:

$$M = \chi_{eff} \frac{B}{\mu_0} \quad (5)$$

The effective magnetisation of the particles, χ_{eff} , depends on the material properties and the demagnetisation factor, K , which introduces a correction for specific particle shapes.

$$\chi_{eff} = \frac{\chi}{1 + K\chi} \quad (6)$$

The constant K has the value of 1/3 for spheres. Please note that the effective magnetic susceptibility trends to 3 when the material susceptibility trends to infinity, as occurs for iron.

We can generalise Equation (4) for linear aggregates made of N monodisperse particles by incorporating the hydrodynamic resistance function, $f(N)$, dependent on the number of particles that form the linear aggregate and on the aggregate shape. In addition, if we consider that the velocity of all particles in the chain (rigid chain) is approximately the same, we can obtain a generalised result as:

$$\rho V N \frac{dU_N}{dt} = V \frac{dB}{dx} M_N - 6\pi\eta a U f(N) \quad (7)$$

where M_N is the total magnetisation of the chain, or the non-spherical particles.

Now, we can define the aspect ratio of linear chains, κ , as the ratio between the length of the chain, L , and the particle diameter, $2a$:

$$\kappa = \frac{L}{2a} \quad (8)$$

It is clear that for linear aggregates the aspect ratio, κ , coincides with the number of particles in the linear chain, N . Therefore, Equation 7 can be rewritten as a function of the aspect ratio.

$$\rho V \kappa \frac{dU_\kappa}{dt} = V \frac{dB}{dx} M_\kappa - 6\pi\eta a U f(\kappa) \quad (9)$$

where the total magnetisation M_T has now been written as an explicit function of the aspect ratio.

Equation 9 is the motion equation of aggregates and/or non-spherical particles with an aspect ratio of κ , under the assumption that the aggregate structure aligns perpendicular to the direction of motion. Thus it is assumed this alignment is not affected by the hydrodynamic resistance.

Since the magnetisation depends on the external magnetic field induction, as the particle moves towards stronger magnetic fields, the magnetisation increases and therefore the particle accelerates until reaching a constant velocity. Nevertheless, if the fluid is very viscous we can ensure a high rate of dissipation of the energy due the drag force and therefore the inertial force is negligible. The particle will accelerate rapidly towards the following quasi-steady state velocity,

$$U_{\kappa} = \frac{V}{6\pi\eta a f(\kappa)} \frac{dB}{dx} M_{\kappa} \quad (10)$$

The ratio of the velocity of the aggregate of $N=\kappa$ particles, U_{κ} , to the velocity of a single independent particle, U_1 , is given as:

$$\frac{U_{\kappa}}{U_1} = \frac{M_{\kappa}/M_1}{f(\kappa)/f(1)} \quad (11)$$

Therefore, the relative velocity of the aggregate with respect to the individual particles, U_1 , is directly proportional to the ratio of the magnetisation of the chain to the magnetisation of the individual particles, M_1 , and the hydrodynamic resistance function of a chain.

2.2 Hydrodynamic resistance function

Different theoretical and experimental studies in the literature provide ways to calculate the drag resistance function for the motion of linear and non-linear aggregates (Happel and Brenner, 1965). However, at low Reynolds numbers, an exact calculation of the drag resistance function only exists for the case of two particles moving either parallel or perpendicular to the flow for different interparticle distances due to the complexity of the calculations (Happel and Brenner, 1965; Ganatos et al. 1978).

Ganatos et al. (1978) have modelled non-axisymmetric Stokes flow for linear aggregates. They provided a plot of the drag correction factors of the individual particles forming part of a seven particle chain. Specifically, these authors plotted the case corresponding to an interparticle distance value of 1.0025 times the particle diameter. We took this case as a good approximation of the circumstance which involves particles that are touching, with a distance of 1.0000 times the particle diameter.

In addition, Filippov (2000) obtained the dynamic shape factor (aspect ratio) for linear chains using the multipole expansion method (MEM). The dynamic shape factor expresses the ratio of the drag force of the actual cluster to the drag force of an isolated sphere having a volume equal to the volume of the cluster.

Table 1 shows the drag correction coefficients, λ , for different particle chains as well as for the case of a single particle obtained from the plots and data given by Ganatos et al. (1978) and Filippov (2000), respectively.

Table 1

Figure 1 shows the values given in Table 1 plotted as a function of the number of particles in the chain (Figure 1 left axis) and the hydrodynamic resistance function (Figure 1 right axis). Further, we provide an empirical fit of the data in Figure 1a, obtained using a least squares fit of the functional form applied to finite cylinders (Happel and Brenner, 1965). The best fit result obtained is,

$$\lambda(\kappa) = \frac{0.3075}{0.1884 \ln(\kappa) + 0.3075} \quad (12)$$

The drag resistance function is thus the drag correction factor, λ_κ , multiplied by the aspect ratio, κ , which coincides with the number of particles in the chain. Hence,

$$f(\kappa) = \kappa \lambda(\kappa) = \frac{0.3075 \kappa}{0.1884 \ln(\kappa) + 0.3075} \quad (13)$$

Figure 1

The drag correction factor, $\lambda(\kappa)$, trends to zero due to the fact that the average drag per particle decreases with aspect ratio. However, this fact does not imply that the chain stops feeling the fluid drag since the actual drag resistance function grows with aspect ratio, κ (or equivalent number of particles in the system) divided by the $\ln \kappa$. Therefore, the longer the chain the larger the hydrodynamic resistance function and thus the total drag force.

It is important to note that Equation 13 has the value of 1 for a single particle whose aspect ratio is clearly 1. Therefore, Equation 13 can also be interpreted as the ratio of the hydrodynamic resistance function of a chain to that of a single particle. In addition, Equation 13 can be interpreted as the product of a linear function, as predicted by Stokes law, and an exponential decay function that corrects the drag for the presence of other particles. This equation becomes approximately linear for large values of aspect ratio, κ , since the exponential decay will trend to zero.

2.3 Magnetization of a finite chain of particles

When particles are close to one another the magnetisation changes according to the geometrical arrangements and the distance from the other particles. Therefore, the effect of the mutual particle magnetisation is observed in the final aggregate velocity. The magnetisation of a specific particle located in a chain containing an infinite number of particles, was given by Zhang and Widom (1995). In our work we have adopted the same equation but considered explicitly the finite sum for a chain made of N particles. This expression (Eq. 14) is valid if all the particles feel the same magnetic induction and therefore there are no boundary effects. In addition, if we make N trend to infinity in Eq. 14 we will recover the original expression given by Zhang and Widom (1995).

$$M_i = \frac{1}{\mu_0} \frac{\chi_{eff}^{VB}}{1 - \frac{2}{3} \chi_{eff} \sum_{n=1, n \neq i}^N \left(\frac{a}{nd} \right)^3} \quad (14)$$

where d is the distance between two particles in contact (equivalent to the sum of their radii), nd is the distance between two given particles, χ_{eff} is the magnetic susceptibility of particle i and μ_0 the magnetic permeability of the particles. It should be noted that when the magnetic interaction with other particles is not considered the sum of the terms in Eq. 14 becomes identically null reducing to the expression of the magnetisation of a single particle. Therefore, the total magnetisation of a linear chain with aspect ratio κ is given by the sum of the individual magnetisations of the particles.

Therefore, the total magnetisation of a linear chain with aspect ratio κ is given by the sum of the individual magnetisations of the particles

$$M_{\kappa} = \sum_{i=1}^{\kappa} \frac{1}{\mu_0} \frac{\chi_{eff} VB}{1 - \frac{2}{3} \chi_{eff} \sum_{n=1, n \neq i}^N \left(\frac{a}{nd} \right)^3} \quad (15)$$

Equation 15 can be rewritten as a function of the magnetisation of one particle, M_1 , in the absence of particle-particle interaction.

$$M_{\kappa} = \sum_{i=1}^{\kappa} \frac{M_1}{1 - \frac{2}{3} \chi_{eff} \sum_{n=1, n \neq i}^N \left(\frac{a}{nd} \right)^3} \quad (16)$$

Equation 16 can be rewritten for a linear chain of N touching spheres ($d=2a$) as:

$$M_{\kappa} = \sum_{i=1}^{\kappa} \frac{M_1}{1 - \frac{1}{12} \chi_{eff} \sum_{n=1, n \neq i}^N \left(\frac{1}{n} \right)^3} \quad (17)$$

Figure 2 shows a double y axis plot of the total magnetisation of a chain of aspect ratio, κ , and the average magnetisation per particle in the chain, M_{κ}/M_I . The total magnetisation has been calculated by using Equation 17 which considers the sum of the individual particle magnetisations which, in turn, depends on the presence of all the other particles. For instance, if a chain is made of 30 particles it was necessary to evaluate 870 terms (since for each particle it was necessary to evaluate the denominator of Equation 17 which includes the interaction with the other 29 particles in the system). These calculations have been performed by using a computer code in Fortran 77.

Figure 2

Figure 2 clearly shows that the total magnetisation of the chains increases with the number of particles in the aggregate. However, the average magnetisation per particle reaches a plateau. The main reason for this plateau is found in the fact that the mutual magnetisation decays as $(1/n)^3$ (eq. 17) and therefore the sum of the series in the denominator converges to the value of 1.202 (Weigh et al. 2004, Zhang and Widom 1995). This is due to the fast decay of the magnetic interactions with interparticle separation distance. In addition, given that the effective magnetic susceptibility of a spherical particle cannot be larger than 3, the denominator in Equation 17 is always positive.

It is also important to note that Equation 17 does not depend on particle size explicitly (with the exception of M_I) but on the ratio between interparticle distance and particle diameter. Therefore, the relative aggregate velocity as given by the combination of Equation 11 and Equation 17 does not depend on the individual particle size either.

Once established, the expression for the relative magnetisation and relative drag for linear chains is of interest in order to understand how these terms affect the behaviour of the system. Therefore, the ratio of the relative magnetisation as given by Equation 17 (Figure 2) to the relative drag given by Equation 13 (Figure 1) is plotted in Figure 3. Since the relative magnetisation is close to a linear relationship for large values of κ (Figure 2), the tendency observed in Figure 3 is clearly logarithmic due to the influence of the hydrodynamic resistance function (Figure1).

Figure 3

Therefore, according to Equation 11, the relative velocity will follow a logarithmic shape associated with the influence of the drag corrections.

2.4 Magnetization of two coated particles

The magnetization of a doublet made of two equal sized spheres of radius a can be expressed according to Equation 16 as:

$$M_2 = \frac{2M_1}{1 - \frac{2}{3}\chi_{eff} \sum_{n=1, n \neq i}^N \left(\frac{a}{d}\right)^3} \quad (18)$$

If two magnetic particles of radius a are coated with a layer of a weak magnetic material it would be expected that the total susceptibility of the material would not be drastically modified by the presence of a weak magnetic shell. This would not be the case if the

external layers were made of a ferro- or ferri- magnetic material which would need to be explicitly taken into account in Equation 18.

Given that in this case the magnetic cores would not be touching, the interparticle distance no longer coincides with the diameter of the core ($2a$). In this case d can be expressed as the sum of twice the radius of the core plus the thickness of the coating.

Equation 18 is then rewritten as:

$$M_2 = \frac{2M_1}{1 - \frac{2}{3}\chi_{eff} \sum_{n=1, n \neq i}^N \left(\frac{a}{2(a + \delta)} \right)^3} \quad (19)$$

Equation 19 shows that the total magnetisation of a core-shell doublet depends on the magnetic properties, size of the core, and the ratio of the core to the composite size.

3. EXPERIMENTAL METHODOLOGY

The top view of the experimental system used in this study is shown in Figure 4. The two poles were made of mild steel and formed a gap that tapered. The distances between the poles of the magnets were 50 mm and 90 mm for the shortest and longest distances respectively. Since the magnetic field induction, B , depends upon the distance between the poles, there is a magnetic field gradient in the x direction from left to right (Figure 4). The magnetic field lines have been schematically added to Figure 4. A detailed analysis of the magnetic field in the magnet has already been given by Zhou et al. (2008). The value of the magnetic field gradient in the x direction was 0.5 T/m. Two

cells constructed from 0.005 m thick Perspex were used. The smaller one had a height of $z=0.045\text{m}$, length $x=0.06\text{m}$ and width $y=0.03\text{m}$. The larger cell was 0.15m high, 0.09m long and 0.05m wide.

The fluid was located within a non-magnetic rectangular cell which was positioned between the two magnetic poles. With the magnet switched off, the individual particles were placed, with great care, into the fluid at approximately the same position along the x direction (Fig. 4). The high viscosity of the fluid aids in this task by keeping the particles at their initial positions. The magnet was then switched on and the particles could move together to form the chain.

Figure 4

A magnetisable particle will tend to move along the x direction in the arrangement depicted in Figure 4 (Zhou et al. 2008, Svoboda 2004). Due to symmetry, particles near the centre line between the poles will tend to maintain their alignment and not move towards one of the poles. The particle will also settle in the vertical direction of gravity, perpendicular to the page.

A non-magnetisable fluid consisting of UCON90 (Polyalkylene Glycol) was used because of its high viscosity, *ca* 34 Nsm^{-2} at $24\text{ }^{\circ}\text{C}$. The fluid was purchased from Union Carbide Corporation, US. Care was taken when transferring the liquid into the cell given the strong tendency for air bubbles to form and in turn remain in the system. The high fluid viscosity ensured exceedingly low particle Reynolds numbers of order 10^{-5} , and therefore a state of creeping flow. Hence, in principle, it was possible to resolve the

particle motion into the specific components of interest and relate that motion directly to the corresponding hydrodynamic forces, and hence the magnetic forces. So although the particles also settled under the influence of gravity, the vertical settling was of little consequence in this study.

In this system any aggregate of spheres was aligned along the direction of the external magnetic field induction as can be seen in Figure 5 for the cases of 5, 10 and 20 spheres. Similarly, cylindrical rods and shots (cylinders with aspect ratios less than 1) were aligned with their longest dimension along the direction of the magnetic field lines, i.e. perpendicular to the poles.

Figure 5

The motion of single particles and particle assemblies, having a given aspect ratio, was studied using a Phantom high speed digital camera (Vision research Inc.) and analysed by image processing software (Control vision 6.3). The velocities of the individual particles were obtained from the position-data extracted from the video images. The steady velocity of the assembly was then determined. The assembly velocities were then normalised by dividing the velocity of the aggregate, U_N , by the value, U_I , obtained for a particle having an aspect ratio of 1.0.

The first half of this study consisted of two series of experiments. In the first series of experiments the motion of individual aggregate chains was studied. The aggregates were formed using a number of ferromagnetic spheres of the same diameter, with separate experiments conducted using spheres of 0.6 mm and 1.0 mm. When magnetised, the spheres tended to automatically align along the direction of the magnetic field induction, perpendicular to the centre-line between the poles. Care was taken to promote this initial

alignment of the spheres, and to ensure a common distance from the axis of symmetry. In addition, we have used cylinders and shots of given aspect ratio for comparison with the predictions for linear aggregate chains. A second series of experiments involved the motion of rods made of the same material as the spherical particles, with a diameter of 0.8 mm and lengths of between 2.0 and 15.5 mm.

We were interested in exploring a broad range of circumstances in order to establish the generality of our findings. One set of data involved magnetic pole sizes of 100 mm x 100mm, while a second set involved a magnetic pole size of 160 mm x 160 mm. For the smaller poles, the applied current was 0.5 Amps, and the magnetic field gradient 0.5 T/m. For the larger poles, one series of experiments involved an applied current of 0.95 Amps with the magnetic field gradient again 0.5 T/m. Here, 1.0 mm diameter spheres were used. However, additional experiments conducted using the larger poles involved 1.5 Amps and a magnetic field gradient of 0.71 T/m. In this case, the experiments involved 1.0 mm and 0.6mm diameter spheres.

The second part of the study was concerned with the investigation of the magnetisation of aggregates made of core-shell particles. The objective of this set of experiments was to study the change in magnetisation, arising during aggregation, due to a change in the shell thickness. The shells were formed using commercially available enamel, consisting of nitrocellulose in a solvent containing colour pigments (nail polish). The core particles were dipped into the liquid. Care was needed to ensure the film was allowed to dry completely. The thickness of the films was varied by applying multiple coatings.

4. RESULTS AND DISCUSSION

4.1 Motion of aggregates and non-spherical particles in a magnetic field gradient

The motion of aggregates made of spherical particles as well as the motion of individual non-spherical particles (cylindrical particles and shots) was studied under different conditions of magnetic pole geometry and vessel size as described in section 3. In order to compare the data obtained using different diameter spheres, aggregates of spheres, and cylindrical particles of different length, the data were reported in terms of the dimensionless aggregate velocity. This dimensionless velocity was defined as the ratio of the aggregate velocity to the velocity of a single particle. The aggregates were always aligned in the direction of the magnetic field induction, i.e. perpendicular to the centre-line between the poles. Cylinders and shots were also aligned with their symmetry axis perpendicular to the direction of motion and therefore parallel to the line between the poles.

Figure 6 shows the normalised velocity of the aggregates made of N particles and with aspect ratio κ , relative to the velocity of a single particle, (U_κ/U_I) , as a function of the aspect ratio of the particles and aggregates. In the case of cylinders and shots their relative velocity was normalised using the corresponding particle with an aspect ratio of 1. The diameters of the spherical particles as well as the diameter of the base of the cylinders and shots are specified in the legend. The velocities were determined with a high speed camera (Phantom). In addition, the error associated with the absolute and relative velocities has been calculated and is presented in Figure 5. The error bars depicted are a consequence of the uncertainties in the measured values and the

subsequent propagation of these errors in the calculations carried out to find the velocity. It should be noted that the error bars correspond to twice the standard deviation.

Figure 6

Two important features can be observed in Figure 6. The first is the apparent collapse of the data for a broad range of particles onto a single curve, and the second, the apparent tendency of the relative velocity to reach an asymptotic value.

The tendency of all the data sets to show the same behaviour whether they belong to an aggregate, a single spherical, or a non-spherical particle, can be understood through Equation 11. Equation 11 shows that the relative velocity is an explicit function of the relative magnetisation and the hydrodynamic resistance function. However, the dependency on particle size is cancelled by the normalisation, while the dependency on exact shape (i.e. cylinders or spheres) is a secondary correction to the hydrodynamic resistance function and the magnetisation.

The asymptotic behaviour of the data shown in Figure 6 is due to the combination of two different factors. The first one is that the hydrodynamic resistance function becomes linear with the number of particles in the chain. The second factor is that the magnetisation also becomes linear with the number of particles in the chain. According to Equation 16 the denominator becomes constant due to the convergence of the series $1/N^3$ to the value of 1.202. Therefore, according to Equation 11 the velocity ratio as plotted in Figure 6 should trend towards a constant value. We are not aware of any previous study that has properly accounted for this result.

Based on the broad range of experimental conditions it was possible to conclude that the findings were largely independent of the system boundary conditions and particle geometry. In addition, it is possible to argue that given an adequate normalisation of the velocities the behaviour of non-spherical particles can be described in terms of the behaviour of aggregates with the same aspect ratio.

4.2 Motion of linear aggregates of spherical particles. Theoretical solution

Figure 7 shows the experimental results and the theoretical predictions of the aggregate velocities made of spheres of 1 mm in diameter as a function of the aspect ratio, κ ($\kappa=N$) of the aggregate. The theoretical predictions are given by Eq. 9 through the use of Equation 13. Three different cases are plotted in Figure 7: two of them correspond to a magnetic field gradient of 0.5 T/m and the third one corresponds to a magnetic field gradient of 0.71 T/m.

Figure 7

It is important to note that the theoretical model does not use any adjustable parameters to achieve the agreement, hence is based entirely on the conditions of the experiments and the well known mechanical and magnetic properties of iron. In order to compare with the experimental data, the initial position of the aggregates corresponded to an initial value of the magnetic field induction of 0.075 T and the final positions corresponded to a final value of 0.078 T.

According to Figure 7 the theoretical model shows excellent agreement for the three sets of data corresponding to two different values of the magnetic gradient. In fact the asymptotic behaviour of the velocity with aspect ratio is well predicted by the theory. Please note that aspect ratio was incorporated into the equations through the drag correction and the magnetisation correction by the presence of other particles in the linear chain.

This new theoretical model has been developed under the assumption that the chains of particles are fully rigid. This approximation may not hold true for extreme aspect ratios since the magnetisation of the boundary particles will be smaller than for the central particles and the drag would also be different. Future studies involving chains of hundreds of particles could be relevant in the study of magnetorheological brakes and other applications. The range of aspect ratios considered in this paper ($\kappa=1, 20$) is, however, large enough for this model to be validated for a broad range of conditions since no significant discrepancies between the experimental and theoretical results have been found.

Finally the range of conditions used in this work is relevant to the conditions governing aggregate motion in magnetic separators.

4.3 Influence of thickness of the coating on the magnetisation of aggregates

The results described in this section are based on the motion of the particles in the x direction, parallel to the centre-line between the poles, as shown in Figure 4. The velocity of two iron spheres in the direction of the field gradient separated by the

viscous liquid was found to be constant prior to aggregation. Then, the velocity of each sphere quickly increased along the x direction during aggregation. The final stage of the aggregation was a rapid process due to the short range of the dipole-dipole interaction force (van Netten et al., 2008).

In order to better understand the aggregation process and the hydrodynamics, a novel series of experiments was conducted to determine the variation in the magnetisation with the distance between two magnetic spheres. Nine pairs of spheres with a core diameter of 1.5 mm were chosen for the experiments. Different core-shell composite particles were created by dip-coating the ferromagnetic core with a Nitro cellulose enamel, each with a different number of layers. Table 2 provides details on the number of thin films applied to the spheres, the film thickness, the system temperature, viscosity and measured velocity of each pair of spheres.

Table 2

The drag force depends directly on the viscosity, which in turn is influenced strongly by the temperature. Given that the experiments were conducted at slightly different temperatures it was necessary to correct the calculated drag force, and hence normalise the data to one temperature. The velocities were linearly adjusted to the result expected for a temperature of 24 °C based on the viscosity of the fluid. A second velocity correction was performed in order to address the effect of the film thickness on the particle diameter, and hence the drag force. An increase in the film thickness resulted in a greater hydrodynamic resistance which in turn led to a lower velocity. The velocities were adjusted to the result expected for a diameter of 1.5 mm, corresponding to the case

of no thin film. A dimensionless velocity was then produced for each experiment by dividing the corrected velocities for the doublet by the velocity obtained for a single uncoated sphere well before aggregation. A dimensionless thickness of the coating was obtained by dividing the thickness of the coating by the radius of the iron sphere.

Figure 8 shows the variation in the relative velocity as a function of the dimensionless film thickness around each of the spheres.

Figure 8

Clearly, the application of the hydrodynamic corrections to the experimental velocity measurements led to very good consistency in the data. A zero film thickness produced an aggregate velocity of approximately double the velocity of individual particles. When the thickness reached a distance of half the radius of the iron sphere, the velocity was approximately equal to the level obtained for an isolated sphere. In addition, the theoretical predictions predicted very well the behaviour of the doublets, especially since no fitted parameters have been used as described in section 4.2. When Equation 19 for the magnetisation of a doublet is combined with Equation 11 there is an explicit dependency on the thickness of the coating.

Equation 11 shows that the ratio of the velocity of the doublet to the velocity of the single particle is directly proportional to the magnetisation ratio. Therefore, Figure 8 can also be interpreted as the ratio of the magnetisation of the doublet. Thus, in the absence of a coating, the magnetisation of the doublet is approximately double the magnetisation

of a single individual particle. When the thickness is half the radius of the magnetic core, the magnetisation is approximately equal to the magnetisation of a single sphere.

Core-shell particles were clearly affected by the presence of the coating. Therefore, when controlling the speed of aggregation different coatings could be used to produce the desired speed without a corresponding loss of response of the particles to an external magnetic gradient.

5. CONCLUSION

A magnetic field gradient was used to drive the motion of ferromagnetic spheres along the direction of increasing field strength. The ratio of the velocity of the aggregates to the velocity of an individual particle as a function of the aspect ratio was found to reach a constant value independent of the aspect ratio value. In addition, the velocities of cylindrical particles and shots normalised to the velocity of single particles showed similar behaviour to those of the linear aggregates of spherical particles. This result opens up the possibility of using the theory of aggregates of spherical particles as a first approximation to explain the motion of non-spherical particles in a magnetic field gradient.

We have presented a new theoretical model of the motion of aggregates of spheres in a gradient field. This model included the corrections of the Stokes' hydrodynamics due to the presence of other particles. This correction has been performed by developing an expression for the hydrodynamic resistance function by using precise data from the literature. The theoretical model has also included the correction of the magnetisation

due to the presence of other particles. The theory has shown excellent agreement with the experimental data for aggregates of spheres for two different values of the gradient field and aspect ratio values between 1 and 20.

The study of aggregation of core-shell particles provided an understanding of the change in velocity, and also magnetisation, due to the presence of a coating shell. Particles separated by a distance equal to half the particle radius exhibited no increase in velocity or magnetisation. At closer separation distances the magnetisation increased almost linearly as the gap decreased. The new theoretical model has been extended to study the motion of core-shell particles by explicitly considering the thickness of the shell. The model has also provided very good agreement with the experimental results.

This work gives a new insight into the motion of aggregates and non-spherical particles in magnetic gradients, an area of great interest in chemical engineering and colloidal science. In particular, we have demonstrated that the motion of non-spherical particles can be described in terms of the motion of linear aggregates of particles and that the aggregation of magnetic particles can be controlled through the use of non magnetic coatings. In addition, this work provides a solid ground for further development of the theoretical model here presented to the case of multiple chains travelling simultaneously as typical in magnetorheological fluids.

ACKNOWLEDGEMENT

The financial support of Australian Research Council for this study is much appreciated.

List of symbols

Latin symbols

a	Particle radius. Radius of the iron core.
B	Magnetic induction
d	Distance between two contacting particle centres
F_{drag}	Drag Force
F_{mag}	Force on a gradient magnetic field
K	Demagnetisation factor
L	Length of a linear aggregate
M	Magnetisation
N	Number of particles in a linear aggregate
U_N	Velocity of an aggregate of N particles
V	Volume of an individual particle
x	Spatial coordinate

Greek symbols

κ	Aspect ratio
μ_0	Vacuum permeability
χ	Material magnetic susceptibility
χ_{eff}	Effective magnetic susceptibility of a particle

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List of Tables

Table 1 Drag correction factor, $\lambda(\kappa)$, for individual particles.

No of particles	λ	Reference
1	1.0000	
2	0.7247	Ganatos et al. (1978)
3	0.6134	Filippov (2002)
4	0.5500	Filippov (2002)
5	0.5079	Filippov (2002)
6	0.4773	Filippov (2002)
7	0.4544	Ganatos et al. (1978)
10	0.4070	Filippov (2002)
15	0.3632	Filippov (2002)
20	0.3371	Filippov (2002)

Table 2. Analysis of aggregate velocity versus the thickness of the coating. The Table includes a velocity correction for viscosity based on a reference temperature of 24 °C, and a velocity correction to account for the different hydrodynamic diameters arising from the thin films. U_2 is the velocity of the doublet and U_1 (=0.228) is the velocity of a single uncoated particle.

No. of films	Coating thickness (mm)	Temp. (°C)	Fluid Visc. (Pa.s)	Vel. (mm/s)	Vel. correction factors		Corrected Vel. (mm/s)	Ratio of shell thickness to core radius	Vel. ratio U_2/U_1
					Visc.	Diam.			
0	0.000	20.5	41.1	0.376	1.202	1.000	0.452	0.000	1.987
1	0.016	24.0	34.2	0.421	1.000	1.021	0.430	0.021	1.890
3	0.048	24.0	34.2	0.390	1.000	1.064	0.415	0.064	1.823
7	0.112	24.0	34.2	0.326	1.000	1.150	0.375	0.149	1.646
10	0.160	27.0	29.2	0.330	0.855	1.212	0.342	0.213	1.504
15	0.240	27.0	29.2	0.259	0.853	1.321	0.292	0.320	1.283
20	0.320	27.0	29.2	0.204	0.853	1.431	0.249	0.427	1.094
24	0.384	19.0	44.5	0.114	1.298	1.514	0.224	0.512	0.984
28	0.448	27.0	29.2	0.166	0.855	1.599	0.227	0.597	0.997

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Figure 1. Double y axis plot of the dependency of the drag correction factor, $\lambda(\kappa)$, (left y axis) and the hydrodynamic resistance function, $f(\kappa)=\kappa \cdot \lambda(\kappa)$, (right y axis) on the aspect ratio of a linear chain orientated perpendicular to the flow.

Figure 2. Double y axis plot of the total magnetisation of a linear chain (right y axis) and the average contribution to the total magnetisation by individual particles (left y axis). Aspect ratio coincides with the number of particles in the chain.

Figure 3. Plot the ratio of the relative magnetisation, M_κ/M_1 to the relative hydrodynamic resistance function, $f(\kappa)/f(1)$ for chains of aspect ratio κ .

Figure 4. a) Top view of the experimental set up. b) Schematic representation of the experimental set-up showing a gradient magnetic field. Also shown is the motion of two spheres approaching each other in the normal direction, and the simultaneous motion in the tangential direction. Gravity is perpendicular to the plane of the figure.

Figure 5. Image of the linear chains made of 5, 10 and 20 particles travelling in the magnetic field gradient

Figure 6. Relative velocity of the aggregates and particles versus aspect ratio. The relative velocity was defined by the velocity of an aggregate of aspect ratio κ divided by the velocity of an aggregate of aspect ratio 1. For the cylindrical rods, the aspect ratio was varied simply by increasing the length of the rod rather than by forming aggregates

from smaller cylinders. Acronyms: LG: low gradient; HG: high gradient; SC: small container; BC: big container.

Figure 7. Velocity of magnetisation of a linear chain of spherical particles in the presence of a magnetic field gradient. Acronyms: LG: low gradient; HG: high gradient; SC: small container; BC: big container. Please note that the magnetisation model is exact, and the drag corrections correspond to the exact numerical solution provided in the literature, hence no adjustable parameters were involved.

Figure 8. The relative velocity is shown as a function of the dimensionless thickness of the coating. The velocity ratio was obtained by dividing the velocities of the two sphere aggregate by the velocity of a single uncoated sphere. The theoretical curve has not used any adjustable parameter