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Experimental study on reversible formation of 2D flocs from plate-like particles dispersed in Newtonian fluid under torsional flow

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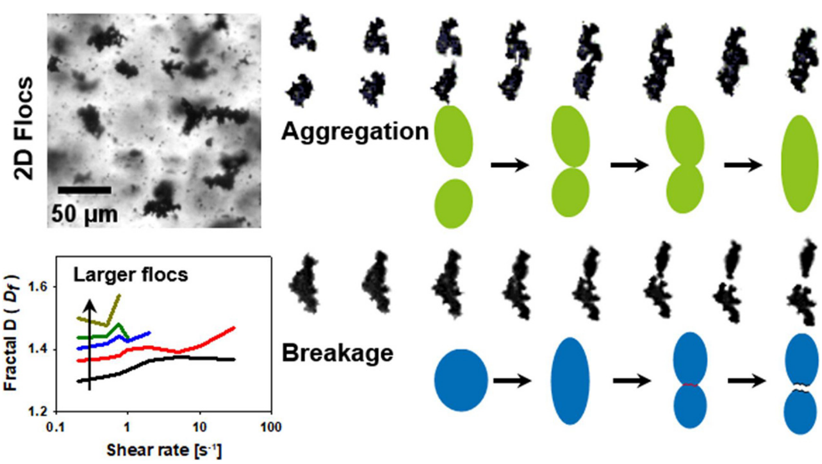
GRAPHICAL ABSTRACT



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ABSTRACT

In this study the reversible aggregation and breakage of two dimensional (2D) flocs in the bulk phase are investigated experimentally in a suspension of platelike particles under a torsional flow between two parallel plates. The suspension was prepared by dispersing platelike particles in a Newtonian fluid. The Newtonian fluid has a viscosity of 1.27 Pa s and the average dimension of the particle face is approximately 1 μm with a thickness of 250 nm. Peclet number of the particle is over 100 and the aggregation is orthokinetic. The microstructure was examined under an optical microscope without disturbing the flow or particle motions. The result shows that 2D flocs of fractal types with the aspect ratio of over 20 between the average face diameter and thickness are formed in the bulk phase by the aggregation of platelike particles and the face of the planar floc is oriented in the same direction of the velocity gradient. The flocs have non-circular faces with size dependent fractal dimensions. The flocs tend to be oriented along the vorticity or the flow direction depending on shear rate and the orientation strongly affects floc breakage and floc size. The breakup of flocs occurs mostly at the middle of the floc after being aligned along the flow direction.

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1. Introduction

Flocculation of particles in suspensions plays an important role in many industrial and environmental processes. For example, in wastewater treatment, dyes or metal ions are removed by flocculation [1–3]; in the tire industries the flocculation of carbon black or silica particles in a very viscous rubber during the tire forming process is an important issue in the performance of tires [4]. Flocculation is also important in food processing [5], paper making [6,7], dust collection [8] and mining [9,10]. Therefore in these industries the deep understanding of flocculation is essential.

Flocculation of particles in fluids can be divided into two major categories of orthokinetic flocculation and perikinetic flocculation [11]. In the orthokinetic flocculation particles are aggregated by the flow while in perikinetic flocculation particles are aggregated by Brownian motions. The theory of flocculation of spherical particles was originated from the historic paper by Smoluchowski [12]. Smoluchowski set up a second order kinetic model describing the coagulation of two spherical particles of different sizes as follows:

$$\frac{dn_i}{dt} = -\sum_{k=1}^{M-i} K_{ki}^{A} n_k n_i + \frac{1}{2}\sum_{k=1}^{i-1} K_{k,i-1}^{A} n_k n_{i-k} \tag{1}$$

In the above equation n_i is the number density of flocs composed of i particles, K_{ki}^{A} is the aggregation kernel (the rate constant of aggregation) between aggregates of size k and i. Also M is the maximum size of the aggregates. The first term at the right hand side of the equation represents the decrease rate of aggregate of size i by the collision with flocs of other sizes to form larger aggregates and the second term represents the formation rate of aggregate of size i by the collisions between sizes $i - k$ and k and between k and $i - k$. Smoluchowski derived the aggregation kernel for a shear flow by assuming that particles collide without being disturbed by the presence of the other particles and once they collide they become one spherical body so that

$$K_{ki}^{A} = \frac{4}{3}\dot{\gamma}(a_i + a_k)^3 \tag{2}$$

where $\dot{\gamma}$ is shear rate and a_i and a_k are the radii of the particles of size n_i. and n_k, respectively. When coagulation is controlled by Brownian diffusion the kernel becomes

$$K_{ki}^{A} = \frac{kT}{3\mu}(a_i + a_k)\left(\frac{1}{a_i} + \frac{1}{a_k}\right) \tag{3}$$

where k is the Boltzmann constant, T is temperature and μ is the viscosity. Following the pioneering work of Smoluchowski many studies have been reported and reviewed as recently done by Ramkrishna [13] and hence an extensive review will not be repeated here.

When a floc is placed in a shear flow it cannot grow in size indefinitely because the floc breaks up when it is subjected to a shear or extensional force that is greater than the strength of the floc. For flocs in a turbulent flow, the floc size is known to be determined by Kolmogorov length scale of the turbulence and floc strength [14–16]. For flocs in a laminar flow, no such length scale exists and hence a different approach is required. For a spherical floc in a simple shear flow, the breakage can occur by the separation of particles from the extensional direction [11]. Adler and Mills [17] considered the rupture of a uniformly porous sphere in a linear shear field and obtained the critical shear for an aggregate. Sonntag and Russel [18] developed a model for the breakup for flocs with fractal geometry to show that the breakage pattern of a spherical floc with a position-dependent internal structure can be varied depending on fractal dimension, Poisson's ratio and the power-law parameter representing the structure. They also showed that the possible breakage patterns are ruptures at the center, equatorial plane or polar axis. It has been experimentally shown that flocs are elongated and then fractured or they are eroded under an extensional flow [19]. Through experiments and population balance modeling Pandya and Spielman [19] and Lu and Spielman [20] deduced that breakage occurs not only by splitting of the parent aggregate into smaller fragments but also by the erosion of constituent particles from the parent floc under turbulent flow conditions. Most of the later studies [21–25] on breakage use the population balance modeling because direct observations of the aggregation and breakage are practically impossible due to the experimental difficulties. Especially, even though a considerable number of theoretical and experimental studies on shear-induced aggregation and breakup exists, direct observations are absent except for those studies for an isolated floc under well-defined flow conditions [11,19,26].

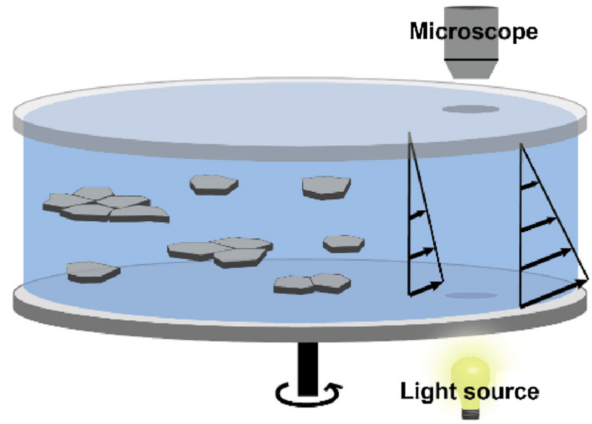

Fig. 1. Schematic diagram of the experimental system. Platelike particles suspended in the liquid become aggregated into 2D flocs in the bulk phase and the flocs are oriented as shown.

In the present study we carried out an experimental study on aggregation and breakup of platelike particles in a Newtonian fluid under shear flows and directly observed the formation of two dimensional (2D) flocs of platelike particles in the bulk phase in situ without disturbing the flow or particle motions using the experimental system shown schematically in Fig. 1. The formation of 2D flocs in the bulk phase is a new finding as far as the authors are aware of. Goodarz-Nia and Sutherland considered the floc formation of non-spherical particles with a size distribution, but the flocs tend to have a prolate-spheroid form with an aspect ratio of 2 [27]. 2D flocs observed by Reynaert et al. [28] were formed at the oil-water interface, not in the bulk phase. Hence the physicochemical origin of the floc formation is basically different. It is also different from the liquid-crystalline structure formation of platelike particles at a high particle loading at a static state by the geometric exclusion potential [29–33]. We also found that flocs of different sizes have different fractal dimensions. This new finding was possible by examining the individual flocs directly under the optical microscope. It is noted that, if a scattering technique was used, only the average value over the whole flocs was obtained.

The Newtonian fluid used here has a viscosity of 1.27 Pa s and the largest dimension of a particle is approximately 1 μm with a thickness of 250 nm. For this particle in the shear flow, Pe is found to have an order of 100, and hence the Brownian motion hardly contributes to the aggregation. In addition to the formation of 2D flocs in the bulk phase, some differences are observed between 2D flocs here and the 3D flocs studied in the literature. A noticeable difference is that the floc is broken into two parts almost at the middle of the floc in the case of 2D flocs in contrast to the case of 3D (three dimensional) floc breakage in which a part of the surface elements or a single particle is taken off from the floc [11,18]. Hence the underlying mechanics appears fundamentally different.

Not all platelike particles aggregate to form 2D flocs. For example, in the case of red blood cells, rouleaux are formed within blood [34] while spherical aggregates are formed in the case of platelets [22]. The shape should depend on hydrodynamic interaction as well as van der