Effect of Particle Size on Breakup of Flocs in a Turbulent Flow

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In our recent paper (*Langmuir* Vol. 15, pp. 4351-4356, 1999), we proposed a simple model of floc strength expressed as the product of the cohesive force between primary particles (f) and the number of contacts (N_c) between clusters (small flocs) in the floc. This model was verified by the experiment of floc breakup, where N_c was controlled. In the present study, to confirm the effectiveness of the model including the former effect, i.e., floc strength is proportional to f, we performed a series of floc breakup experiment in the turbulent flow generated with a mixing vessel. In the experiment, the magnitude of the cohesive force was controlled by changing diameters of primary particles composing flocs. The obtained relationships, maximum diameter of floc versus turbulent shear stress as well as the mean number of particles composing a floc versus turbulent shear stress, showed good agreement with the prediction based on the model.

Key Words: *Colloidal particle, floc breakup, locally isotropic turbulence, particle size, cohesive sediment*

1. INTRODUCTION

To understand the physical properties (size, structure and strength) of flocs (aggregates of colloidal particles) is very important for the prediction and/or control of transport of cohesive sediment in water environment[1-3].

Thus far, the Smoluchowski's equation[4] and the fractal geometry[5] have given us the framework to treat the growth kinetics of flocs and the structure of formed flocs. However, the backbone of floc breakup, which reduces floc size, is insufficient because the theoretical expression of floc strength against fluid force is unclear.

It can be interpreted that floc strength depends on both the cohesive force between primary particles composing a floc and the structure of a floc. With regard to the former, Sonntag & Russel[6] broke up flocs which were formed with monodisperse polystyrene latex colloidal spheres in various concentrations of electrolyte solutions in a simple shear flow and obtained relationships between the average number of primary particles composing a broken floc and the shear stress at each salt concentration. They demonstrated that differences in relationships between the average number of primary particles composing a floc and the shear stress can be normalized by the cohesive forces between two colloidal spheres predicted by the so-called Derjaguin-Landau-Verwey-Overbeek theory[6, 17]. This finding implies that the cohesive force between primary particles directly controls the floc strength. On the other hand, Tambo, Yamada & Hozumi[7] pointed out the significance of the floc structure and derived floc strength as a function of floc density, which decreases with increasing floc size. They concluded that floc strength is proportional to the floc diameter. Recently, however, the Tambo's model was opposed by Yeung & Pelton[8], who directly measured floc strength from the distortion of two micropipettes rupturing a floc. From the measurement, they found that floc strength does not change with floc size but change with fractal dimension of flocs.

It is well known that flocs are fractal objects[5, 9, 10] reflecting the sequence of binary collision between clusters (small flocs). Fractal dimension (D) of a floc increases with increasing the number of contacts between the two colliding clusters (N_c) when the floc is formed[9, 10]. Thus, it can be expected that the increase of fractal dimension increases floc strength through the increase of the number of contacts between clusters (N_c).

Very recently, on the basis of these findings[6, 8, 10], we proposed a simple model of floc strength (F_{floc}) expressed as the product of the cohesive force between primary particles (f) and the number of contacts between clusters (N_c) [11]:

$$F_{floc} = f \cdot N_c \qquad (1)$$

The latter effect of the model, F_{floc} is proportional to N_c, was validated by the experiment where we broke up three types of flocs with different values of N_c in a turbulent flow[11]. In the experiment, flocs were formed by the coagulation of monodisperse polystyrene latex (PSL) model spheres in KCl solution. However, the effect of the former is not examined. Thus, to complete the expression of eq. (1), it is also necessary to obtain the experimental proof that F_{floc} is proportional to f. For this purpose, we planed the experiment to break up flocs formed with different sizes of PSL spheres because f is proportional to the diameter of a primary particle.

2. MODEL OF FLOC BREAKUP

(1) Brief Description of Model

From the analogy of breakup of droplets in a locally isotropic turbulent flow[12], it is considered that a floc is broken up when the hydrodynamic force acting on the floc exceeds the floc strength (F_{floc}) given by eq. (1). On the basis of this criteria, we derived the following equations for the maximum value of floc diameter ($d_{f,max}$) in a certain turbulent flow field[11].

$$d_{f,\max} \propto \left(\frac{\mu G}{f \cdot N_c}\right)^{-1/2} \quad \text{for } d_f < \eta, \qquad (2)$$

$$d_{f,\max} \propto \left(\frac{\rho \varepsilon^{2/3}}{f \cdot N_c}\right)^{-3/8} \quad \text{for } d_f >> \eta. \qquad (3)$$

where $G=(\varepsilon/\nu)^{1/2}$ and $\eta=(\varepsilon/\nu^3)^{-1/4}$ are local shear rate and Kolmogorov microscale of turbulence, respectively. Other symbols, μ, ε, ν and ρ denote viscosity of fluid, rate of energy dissipation per unit mass, kinematic viscosity of fluid and density of fluid, respectively. These equations show that the size of flocs decreases as ε increases. Eq. (2) was validated in the previous paper[11].

(2) Effect of Particle Size

If coagulation is induced under the condition of sufficiently high ionic strength, the electrical repulsive force between particles is negligibly small. In this case, f is written as the following equation based on the van der Waals attractive forces between two spheres with an equal diameter of d_0.[6, 17]:

$$f = \frac{A d_0}{24 h^2}. \qquad (4)$$

where A is the Hamaker constant and h is the minimum distance of separation between particle surfaces. Supposing A and h are constants, one can derive the following equations from eqs. (2-4).

$$d_{f,\max} \propto \left(\frac{\mu G}{d_0 \cdot N_c}\right)^{-1/2} \quad \text{for } d_f < \eta, \qquad (5)$$

$$d_{f,\max} \propto \left(\frac{\rho \varepsilon^{2/3}}{d_0 \cdot N_c}\right)^{-3/8} \quad \text{for } d_f >> \eta. \qquad (6)$$

When a floc is a fractal object, the relationship between the number of primary particles composing the floc (i) and the diameter of the floc (d_f) is written as the following equation[13]:

$$i = \left(\frac{d_f}{d_0}\right)^D \qquad (7)$$

where D is so-called fractal dimension and is in the range of $1 \le D \le 3$. Substituting eq. (7) into eqs. (5, 6), one can derive the following equations for the relationship between i and ε.

$$i_{\max} \propto \left(\frac{\mu G d_0}{N_c}\right)^{-D/2} \quad \text{for } d_f < \eta, \qquad (8)$$

$$i_{\max} \propto \left(\frac{\rho \varepsilon^{2/3} d_0^{5/3}}{N_c}\right)^{-3D/8} \quad \text{for } d_f >> \eta. (9)$$

To verify these equations taking into account d_0, we carried out the following experiment, where flocs formed with different d_0 of PSL spheres were broken up in a turbulent flow.

3. EXPERIMENT

(1) Materials

In the present experiment, flocs were formed by the coagulation of monodisperse PSL spheres in the solution of KCl. To form flocs with different f, we adapted PSL spheres with three different diameters (480, 1356 and 1956 nm). All these spheres were synthesized by aqueous polymerization without surfactant. From now on, we denote the flocs using the value of particle diameters; "480 floc" is the floc that consists of particles with d_0 = 480 nm. Before experimental use, all distilled waters and

KCl solutions were filtered by using the Millipore filter with a 0.22 μm mesh size.

(2) Floc Formation

The formation of flocs was induced by mixing 1 mL of 2.236 mol/L KCl solution to 1 mL of the PSL suspension, whose volume fractions (ϕ_0) were 0.00162, 0.0182, 0.00182, 0.0891 and 0.00101 for 480 floc, 1356 A floc, 1356 B floc, 1356 C floc and 1956 floc, respectively. After mixing, the mixed suspension was sucked up into a sampler (a syringe with a glass tube) and left over three days to be fully coagulated. The KCl concentration of the mixed suspension is adjusted to have the equivalent density of PSL spheres and is higher than critical coagulation concentration[14]. Thus, the van der Waals attractive force is dominant for the force between particles.

(3) Agitation Apparatus to Generate Turbulent flow

Turbulent flows to break up flocs were generated with an agitation apparatus composed of a Rushton type stirrer equipped in the cylindrical vessel with equally placed four baffles. To evaluate hydrodynamic force and Kolmogorov microscale (η), we evaluated ε from the measurement of the rate of coagulation[15] in the vessel. Evaluated values of ε were consistent with the rate of energy input per unit mass measured with a torque meter[15]. The schematic illustration of the vessel and the values of ε were described elsewhere[11, 15].

(4) Procedure

The coagulated suspension in the sampler was carefully poured into the vessel filled with 1.118 mol/L KCl solution. In this handling, volume fractions of PSL in the vessel (ϕ_b) were adjusted to 5.84×10^{-7}, 5.25×10^{-7}, 5.25×10^{-7}, 5.14×10^{-7} and 4.67×10^{-7} for 480 floc, 1356 A floc, 1356 B floc, 1356 C floc and 1956 floc, respectively. These values are so low that the effect of the regrowth of broken flocs can be regarded negligibly small[11]. After agitation for 90 minutes, which is sufficient time for the size distribution of broken flocs to reach steady-state[11], a small amount of suspension containing broken flocs was carefully extracted by using a microslide (glass tube with rectangular cross section). Suspended flocs in the slide were photographed through a microscope and the maximum lengths of the projected flocs (D_m) were measured as an index of d_f one by one. To determine the value of N_c, we evaluated the value of the fractal dimension of flocs (D) by solving the mass conservation equation of primary particles[11, 13)

$$N(0)\cdot V_s = \sum_{n=1}^{N_f} i_n = \sum_{n=1}^{N_f}\left(\frac{D_{m,n}}{d_0}\right)^D \qquad (10)$$

on the assumption that all monitored flocs in the prescribed volume (V_s) have the same D, where $N(0)$ $(=6\phi_b/(\pi d_0^3))$ and N_f are the number concentration of primary particles and the number of flocs in V_s, respectively.

From the experiment described above, D_m, $\langle i \rangle$, and D were obtained. The value of N_c is deduced from D.

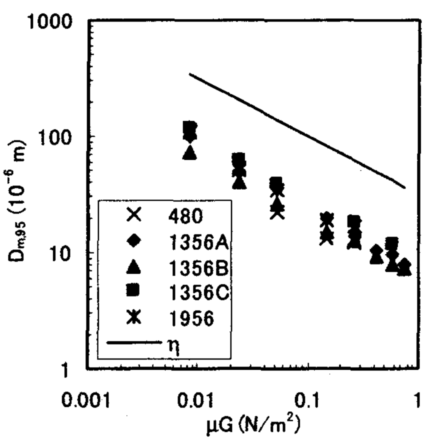

Fig. 1 The 95 % D_m, $D_{m,95}$ (used as the maximum diameter of flocs) vs. turbulent shear stress, μG. Solid line denotes Kolmogorov microscale, η.

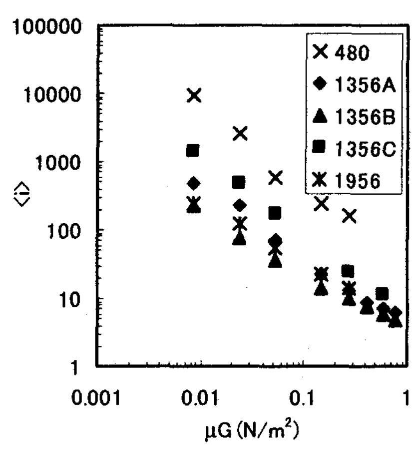

Fig. 2 The mean number of particles composing a floc, $\langle i \rangle$ vs. turbulent shear stress, μG.

4. RESULTS AND DISCUSSION

In Fig. 1, the 95 percentile D_m of the size distribution ($D_{m,95}$) of broken flocs used as $d_{f,max}$, is plotted against turbulent shear stress (μG). This figure indicates that floc size decreases as μG increases and floc breakup occurs in the region smaller than η represented by a solid line. Similar tendency is shown in the relationships between the mean number of particles composing a floc ($\langle i\rangle$) and μG (Fig. 2). Fractal dimension (D) is plotted against $\langle i\rangle$ in Fig. 3. This figure indicated that D decreases steeply with the decrease of $\langle i\rangle$ in $\langle i\rangle \leq 70$ as previously reported[11,16]. This is due to the lower cut-off limit of scaling relation[16]. That is, flocs composed with smaller i have the smaller values of D even if flocs have the same N_c value[16].

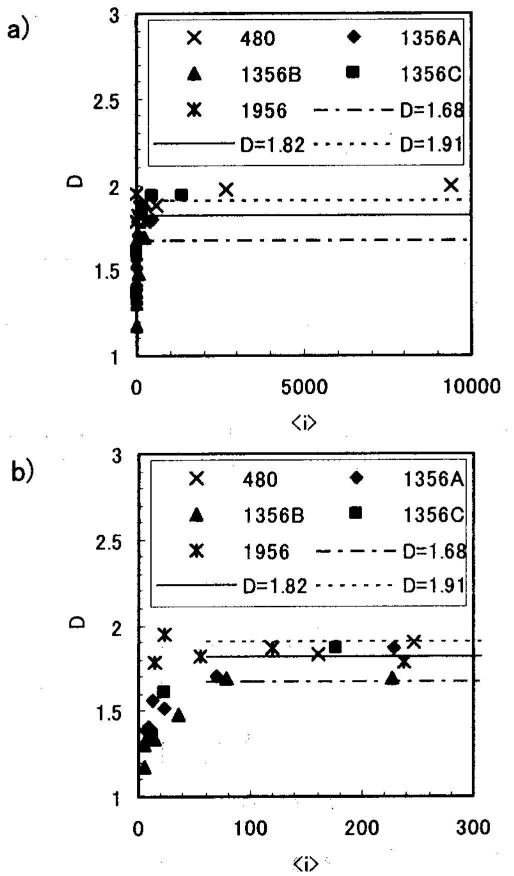

Fig. 3 Fractal dimension, D vs. the mean number of particles composing a floc, $\langle i\rangle$. a) all data. b) magnification.

Mean values of D in $\langle i\rangle \geq 70$ are 1.91 (480 floc), 1.82 (1956 floc), 1.79 (1356 A floc), 1.68 (1356 B floc) and 1.91 (1356 C floc). Comparing these values to those of simulated flocs[9, 10] as done in the previous report[11], we determine that $N_c = 3$ (480 floc, 1356 C floc), 2 (1356 A floc, 1956 floc), 1 (1356 B floc)[11].

According to the model (eqs. (5, 8)) with N_c values determined above, we replotted $D_{m,95}$ and $\langle i\rangle$ against $\mu G/(N_c d_0)$ and $\mu G d_0/N_c$ in Figs. 4 and 5, respectively. As indicated in these figures, differences shown in Figs. 1 and 2 are normalized. Further, the power relation, $D_{m,95} \propto (\mu G/(N_c d_0))^{-0.511}$ agrees well with the model.

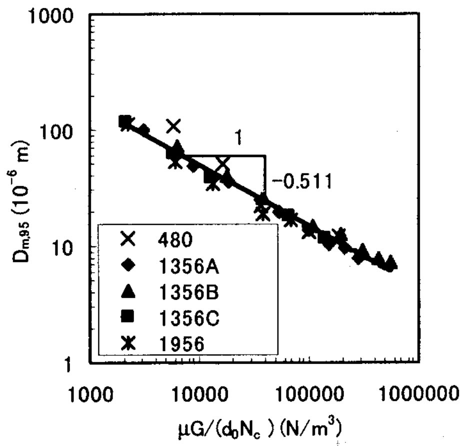

Fig. 4 The 95 % D_m, $D_{m,95}$ (used as the maximum diameter of flocs) vs. scaled turbulent shear stress, $\mu G/(d_0 N_c)$.

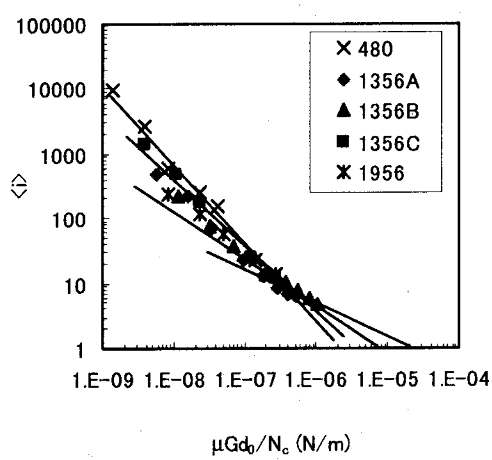

Fig. 5 The mean number of particles composing a floc, $\langle i\rangle$ vs. scaled turbulent shear stress, $\mu G d_0/N_c$. Four solid lines, whose slopes are -2.5/2, -2/-2, -1.5/2, and -1/2, from steep to gradual, are drawn for guide.

In Fig. 5, four solid lines are drawn for guide. These slopes, $d\log\langle i\rangle/d\log(\mu G d_0/N_c)$, are -2.5/2, -2/-2, -1.5/2, and -1/2, from steep to gradual. In the present model, the value of $d\log\langle i\rangle/d\log(\mu Gd_0/N_c)$ is given by $-D/2$; $d\log\langle i\rangle/d\log(\mu Gd_0/N_c) = -1.5/2$ for $D = 1.5$, for example. As shown in Fig. 5, experimental data obey one of lines whose slopes are $-1/2 > -D/2 > -2.5/2$. The range of D from $d\log\langle i\rangle/d\log(\mu Gd_0/N_c)$ in Fig. 5 shows reasonable agreement with that in Fig. 3. This confirms the validity of the present model.

From the extrapolation of $\langle i\rangle$ vs $\mu Gd_0/N_c$ for $N_c = 1$ (1356 B floc), the value of μGd_0 is expected to be between 6×10^{-6} (for $D = 1.5$) and 2×10^{-5} (for $D = 1$) at $\langle i\rangle = 1$. The meaning of $i = 1$ is that doublets even break at $\mu Gd_0 = 6\times 10^{-6}$ - 2×10^{-5}. From the criteria of doublet breakup in a laminar flow[17], the following relation is obtained for $i = 1$.

$$\mu Gd_0 = \frac{A}{36\pi h^2} \qquad (11)$$

The values of μGd_0 for i =1expected from the present experiment, 6×10^{-6} - 2×10^{-5}, can be obtained by substituting reasonable values of A and h, e.g. $A=1.0\times 10^{-21}$ J and h= 0.7-1.2 nm, into eq. (11). This agreement between experiment and calculation also confirms the validity of the present experiment and analysis.

5. CONCLUSIONS

To extend the applicability of the previously proposed model of floc strength, flocs formed with three different diameters of monodisperse PSL spheres were broken up in the turbulent flow generated in an agitation vessel. Experimental results show that floc strength is proportional to the cohesive force between primary particles. This result is in good agreement with the prediction based on the model. All findings including previously reported results indicate that floc strength is expressed as the product of the cohesive force between primary particles and the number of contacts between clusters when the floc is formed.

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