AIChE WILEY InterScience® DISCOVER SOMETHING GREAT

Flocculation Analysis and Control System by Laser Diffractometry at Industrial Scale

Mohamed Bizi and Hervé Gaboriau
BRGM Environment and Process Div., 3, Avenue C. Guillemin, 45060 Orleans Cedex 2, France

DOI 10.1002/aic.11352
Published online November 13, 2007 in Wiley InterScience (www.interscience.wiley.com).

The flocculant injection control system efficiency was evaluated on-site in an aggregate quarry, by means of laser diffraction and analysis of the size and texture of flocs. The configuration of the feed tank and the laser particle size measurement cell installed at the facility (280,000 t/year of aggregates) allowed characterization of flocs with particle size between 4.2 and 3473 μm under hydrodynamic conditions that were highly favorable for the examination of large and very fragile flocs. Two days of analysis of the floc formation process along the path followed by the slurry showed that flocculation was optimal during standard operation of the facility when the flow rate of waste fines and concentrations of solids were close to those used to calibrate the flocculant injection control system. Conversely, when the concentration of solids in the flocculator feed slurry dropped by 57.3%, the flocculant dosing fluctuated during stabilization of the mechanism, the kinetics of flocculation slowed, the mean size of flocs arriving at the settling tank dropped by 69%, and the mode of smaller flocs shifted from 77.8 to 10.4 μm in relation to normal operation. On-site analyses confirmed that the measurements made with laser diffraction (using a methodology developed in the laboratory) allow determination of the effects of conditioning on the characteristics of flocs in terms of particle size, porosity, density, and volume fraction in the slurry. Evolution of these characteristics according to the local parameters of conditioning (mean detention time, mean slurry velocity, and mean velocity gradient) provides a significant part of basic information necessary to a diagnosis of the operation of an industrial circuit of flocculation. *AIChE J,* 54: 132–137, 2008
Keywords: flocculation, density, laser diffractometry, porosity, quarry, regulation

Introduction

Separation of solids and liquids by flocculation is a widely used process in industry. For example, in the aggregates sector (400 Mt/year in France), the main purpose of flocculation is to ensure rapid recirculation of process water and thickening and consolidation of solid waste (60% of production), within the space limits imposed by the need for facilities to encroach as little as possible on their surrounding environments. The regulation of flocculation in aggregate quarries is controlled via measurement of the settling velocity of flocs. The flocculant injection control system regulates the dosing to maintain the required settling velocity permanently, with only slight variations.

The objective of this work is to demonstrate the advantages of a system for in-situ analysis and monitoring in industrial sectors that, for economic and environmental reasons, recycle process water and consolidate mineral slurries by flocculation. Current management of conditioning circuits generally includes control systems located far upstream of the settling tank inlet well. In quarries producing aggregates,

Correspondence concerning this article should be addressed to M. Bizi at m.bizi@brgm.fr.

for example, the regulation of flocculation is based on floc settling velocity measured by means of transparent-tube optical sensors requiring sampling of the flocculated slurry, with samples often being taken too far upstream of the thickener feed. This study presents the analysis of flocculation and the effect of sensor response on the hydrodynamics and texture of flocs in two situations that are typical of operation of an aggregate quarry. The combination of a hydrodynamic model and a methodology for particle size analysis by laser diffraction developed in the laboratory was used to study the effects of the control of system's capacity on the flocs reacting on the flocculator feed slurry concentrations.

Materials and Methods

Study site and material

The methodology developed in the laboratory by using laser diffraction to characterization the flocs was used to analyze flocculation and control system response in an industrial flocculation plant. The analysis was carried out in an operating aggregates quarry producing 14,000 tonnes (dry weight) of waste fines of less than 80 μm from the aggregate washing. The flow rate of washing water consumed was between 250 and 300 m^3/h. The facility is equipped with a water recycling system that includes a flocculant preparation and dosing unit and an 11-meter diameter clarifier. For normal working of the deposit, 46% of the waste material constitutes a large silicate fraction, the other 54% constituting a clayey (-2 μm) fraction that includes kaolinite, illite, and clayey minerals of the montmorillonite group. The specific surface areas of this material and its less than 2 μm component—measured by BET multipoint nitrogen adsorption with a "Micromeritics TriStar 3000" analyzer, for a relative pressure range of 0.05–0.3—are 34.8 and 62.2 m^2/g, respectively. In addition to the influence of particle size, the clay phase could lead to stability of these fines in the washing water. Clarification of the water is therefore highly compromised. At the quarry in question, clarification of this water with its load of fines is obtained by a system comprising a continuous conditioning circuit. The conditioning energy is provided by gravity flow of the suspension through a four-compartment flocculation tank and a tank-thickener transfer pipe, with controlled head loss. The configuration of the circuit is shown in Figure 1; its main operating characteristics are summarized below:

- Standard concentration of solids in feed slurry is around 17 g/l;
- Nominal feed slurry flow rate is 380 m^3/h;
- Mean velocity in the flocculation tank is 0.3–0.4 m/s, 1 m/s in the transfer pipe; mean detention time is 23 s;
- Specific power of 425 W/m^3, corresponding to a mean velocity gradient of 516 s^{-1};
- Flocculation is obtained with an anionic sodium acrylamide–acrylate copolymer supplied by SNF Floerger. The anionicity of this flocculant is 30 mole % and the average molecular weight is about 12×10^{-21} kg. The mean dose is around 180 g/t, based on the solids content; the flow rate for addition of the diluted solution of flocculant measured in stable circuit operating conditions is 1696 l/h.
- Regulation of flocculation is controlled via measurement of floc settling velocity. The flocculant injection control system regulates dosing to maintain the required settling velocity permanently, with only slight variations;
- Flocculant is added in two incremental steps: 20% is injected in the flocculation tank compartment consisting of channels 2 and 3 (Figure 1); the remaining 80% is distributed at the end of the last compartment, at the head of the transfer pipe, so as to improve conditioning of the slurry over the mean duration of transport.

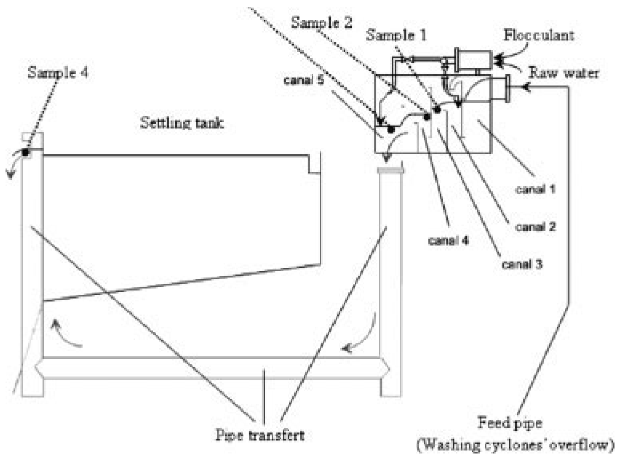

Figure 1. Configuration of flocculation circuit for -80 μm washing fines slurry.

These operating characteristics differ considerably from those encountered most frequently in potable water and wastewater treatment systems, where the values for $\overline{G}$ rarely exceed 100 s^{-1} and detention times are rarely less than 10 min. These differences illustrate that the objectives of treatment are not comparable: the aggregate producer is looking for rapid recycling of process water favored by high settling velocities and sludge consolidation. The priority for treatment of potable water is to minimize turbidity and lower concentrations of residual solids, which requires long conditioning times to capture fine particles and moderate velocity gradients to limit adverse effects on floc size. The velocity gradients in the different compartments are greatly influenced by the viscosity of the slurry, η_p (0.0157 poise). This latter value corresponds to a flocculated slurry with mean floc porosity, ε_F, estimated to be 0.93. Viscosity is considered to be independent of the velocity gradient; it is estimated from the Thomas equation[1]:

$$\frac{\eta_P}{\eta_0} = 1 + 2.5\Phi_F + 10.05\Phi_F^2 + 0.062\exp\left(\frac{1.875\Phi_F}{1 - 1.595\Phi_F}\right) \quad (1)$$

where Φ_F is the volume concentration of the floc in the slurry, as a decimal fraction, and η_0 is the viscosity of water at ambient temperature.

Method

The floc size distribution by volume (FSD) and the textural characteristics of the flocs (density ρ_F or porosity ε_F) were determined by laser diffraction ($\lambda = 0.632$ μm) using a long-bench Malvern MasterSizer S particle size analyzer and a 10-mm wide measurement cell with gravity feed for the flocculated suspension diluted to a measurement concentration whose attenuation effect on the central laser does not exceed 16% obscuration. The suspension is homogenized in a 4-liter supply reactor with a slowly revolving “perforated sheet” stirrer ($G \leq 20\ s^{-1}$). For each analysis, the floc sample taken from the flocculation circuit (Figure 1) is introduced gently into the water in the reactor using a “ladle” of known volume. This configuration is able to determine FSDs between 4.2 and 3 473 μm. It avoids the need for the pumped recirculation of the suspension during measurement that is used in conventional analyzer configurations, and which alters floc FSD and texture significantly. Analysis of the optical data is obtained with a Fraunhofer optical model, a special case of Lorentz–Mie theory.[2,3] This model gives a good approximation of the Lorentz–Mie theory for particles with diameters greater than five times the wavelength of the incident beam, i.e. 3.2 μm. Laser diffraction measurements also provide access to mean porosity, volume concentration Φ_F, and mean relative density of flocs,[4] using the following equations:

$$\varepsilon_F = 1 - \frac{C_S}{10^{-2} C_V \rho_S} \tag{2}$$

$$\rho_F = \rho_S(1 - \varepsilon_F) + \rho_f \varepsilon_F \tag{3}$$

ε_F is the mean floc porosity, where C_s is expressed in kg/m^3; ρ_s is the mean density of solids, in kg/m^3, and C_v is the volume concentration of floc measured by the analyzer as a percentage. The porosity complement ($1 - \varepsilon_F$) corresponds to the solid fraction (V_S/V_F). The ratio between the floc volume “V_F” and the volume of slurry “V_P” therefore represents the volume concentration “Φ_F” of the flocs in the flocculated suspension with initial concentration of solids C_s. This concentration is determined from the following equation:

$$\Phi_F = \frac{\frac{C_s}{\rho_s}}{1 - \varepsilon_F} \tag{4}$$

The FSD obtained are normalized in frequency according to relation (6). They are then broken up into four Log-normales populations (Table 3), large, medium, small, and fine flocs, to attempt to clarify the changes in FSD after addition of flocculant and conditioning characterized by $\overline{t_r}$ and $\overline{G}$, along the path of the particles and flocs in the flocculation tank and transfer pipe. The FSD, represented by the function $F(D)$, is modeled by a linear combination of four populations F_1, F_2, F_3 and F_4, and then normalized as follows:

$$F(D) = a_1F_1(D) + a_2F_2(D) + a_3F_3(D) + a_4F_4(D) \tag{5}$$

$$\int_0^D F(D)dD = \sum_{i=1}^{4} a_i \int_0^D F_i(D)dD = 1 \quad \text{where} \quad \sum a_i = 1 \tag{6}$$

where the integrated terms represent the cumulative relative volumes at dimension D for the FSD and the component populations. The coefficients a_1, a_2, a_3, and a_4 appear as volume weighting coefficients in the floc distributions.

The presentation of the grading curves is given at the same time in linear scale and logarithmic scale. The floc size frequency curves with a linear scale provide a better representation of the relative proportions of large flocs with sizes greater than around 800 μm; those with log scales show more clearly the differences between distributions in the central and fine areas.

Table 1. Hydrodynamic and Textural Characteristics for the First Series

Parameter	F.	S.1.1	S.1.2	S.1.3	S.1.4
P (kW)		0.07	0.16	0.21	0.56
V_P (m^3)		0.35	0.14	0.38	1.51
$\overline{t_r}$ cumulative (s)		3.3	4.7	8.3	22.6
$\overline{G}$ (s^{-1})		367	545	564	516
DmF (μm)	14.5	279	327	273	428
d90/d10	20.9	6.9	11.0	11.1	7.4
DSI	2.23	0.95	1.37	1.26	1.15
SSA BET (m^2/g)	35				
ε_F		0.935	0.934	0.935	0.939
ρ_F (g/cm^3)		1.110	1.113	1.111	1.104
Φ_F		0.098	0.095	0.097	0.104

F: feed sample; S.1.b (b: sampling zone).

The mean hydrodynamic conditions were deduced from a hydrodynamic model of the flocculation tank, assuming piston type flow and a parabolic profile of the outer slurry layers at overflows. Head losses and levels were calculated using published data from the literature.[5–8] The mean velocity gradient is derived from the power absorbed per unit volume of the suspension[9]:

$$P = Qg\rho_P\Delta H \tag{7}$$

$$\overline{G} = \sqrt{\frac{P}{V_P\eta_P}} \tag{8}$$

Results and Discussion

The analysis of flocculation at the above-mentioned industrial facility was carried out over 2 days during which time two series of analyses were made. The general layout of the circuit and sampling points are shown in Figure 1.

Analysis of the first sample series

The mean concentration of solids in the clarifier feed slurry measured on the first day of analysis was 17.1 g/l. This value is as expected, based on the circuit’s design and operating data. The analysis of flocculation is represented below by the behavior of four representative samples. The mean hydrodynamic conditions (Table 1) characterizing each of the samples were estimated on the basis of a slurry with a concentration of 17.1 g/l flowing at 380 m^3/h, of circuit configuration, and the slurry levels measured in the flocculation tank as the distance between the upper edge of the side wall and slurry–air interfaces.

The cumulative distributions and breakdowns into frequency distribution populations (Tables 1 and 3) show:

- marked aggregation dynamics between samples 1 and 2 for the first addition of flocculant (Figure 2). This is evident from the increases in the modes of the three populations of large, medium, and small flocs, in the absence of a significant increase in fine flocs (7%). The existence of a small erosion component is revealed by the reduc-

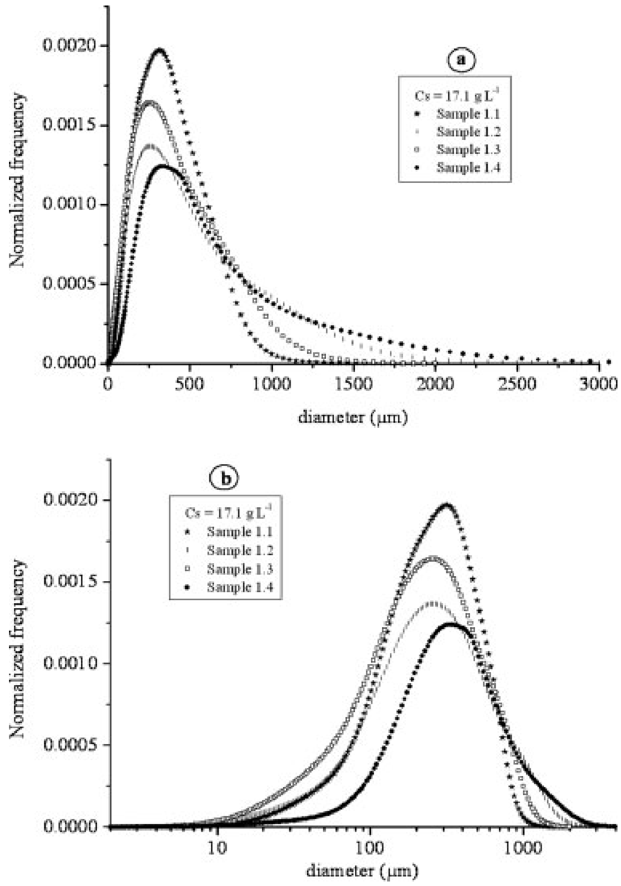

Figure 2. Particle size distribution for the first series.

(a) Normalized frequencies with a linear scale and (b) normalized frequencies with a log scale.

tion in mode for the fine floc population, accompanied by an increase of DSI in the range of 0.826–1.028. The coexistence of aggregation and erosion causes an increase in range for the entire floc population. The predominant aggregation leads to an increase in the mean size of flocs, DmF, which goes from 279 to 327 μm.

- predominant fragmentation and erosion of flocs between sample points 2 and 3, in the absence of new addition of flocculant. Dimensional degradation of flocs between 2 and 3 is indicated by reduction in the modes of small, medium, and large floc populations. The marked increase in the proportion of the population of fine flocs—from 7 to 12%, with almost constant mode and slightly smaller range—indicates active erosion of flocs between points 2 and 3. Despite a greater proportion of the population of large flocs at point 3, in comparison to point 1, the severe erosion between 2 and 3 leads to a comparable mean size: 273 μm against 279 μm, with a greater range for the entire floc distribution.
- strong aggregation between points 3 and 4, as a consequence of the second addition of flocculant and conditioning of the suspension during transportation in the transfer pipe. This is clearly identified by:
  - – increase in the mode of the small floc population;
  - – increase in the sum of proportions of small, medium, and large flocs in floc distribution at point 4. The new addition of flocculant tends to even out the sizes of small and medium flocs, a tendency indicated by the fact that population 3 (medium flocs) has almost disappeared;
  - – the low proportion of fine flocs (population 1), correlating with the increase in the sum of populations 2–4. There was relatively little floc erosion, in spite of a longer transport time with a still high mean gradient. This observation indicates high resistance of the flocs to shearing stresses, which can be explained by the elevated addition of polymer (80% of total dosage).

The cumulative distribution data confirm that the new addition of flocculant leads to high aggregation between points 3 and 4, marked by a considerable increase in the mean floc size, which goes from 273 to 428 μm, and a reduction in the FSD range.

Analysis of the second sample series

On the second day of quarry working, the mean concentration of solids in the flocculator feed slurry did not exceed 7.3 g/l. This level of production reflects operating conditions that are far from normal, based on the nominal capacity of the washing circuit (17 g/l). The 380 m^3/h flow rate of -80 μm slurry is a reference parameter subject to slight variations, given the somewhat inflexible hydraulic capacities of the cyclone and flocculator. The reduction in concentration indicates either a flow rate of solid raw material lower than nominal capacity or a proportion of -80 μm fines in the raw material markedly below the mean. A concentration of solids very much lower than the standard mean concentration implies:

- – a higher initial settling velocity V_o, for a flocculant dose, a duration and mean velocity gradient comparable to those for conventional operation;
- – greater sensitivity of V_o to variations in discharge rate from the flocculant injection pump if the concentration of primary flocculant solution is unchanged. In this case, the control system will tend to greatly reduce pump discharge to attempt to keep V_o within tolerances;
- – reduction in the kinetics of flocculation and floc erosion, related to the reduction in volume concentration Φ_F of the flocs in the suspension, especially if flocculant input is reduced to stabilize V_o.

Particle size distribution data (Tables 2 and 3) show:

Table 2. Hydrodynamic and Textural Characteristics for the Second Series

Parameter	F.	S.2.1	S.2.2	S.2.3	S.2.4
P (kW)		0.07	0.16	0.21	0.56
V_P (m^3)		0.35	0.14	0.38	1.51
$\overline{t_r}$ cumulative (s)		3.3	4.7	8.3	22.6
$\overline{G}$ (s^{-1})		367	545	564	516
DmF (μm)	17.6	75	147	634	133
d90/d10	34.0	6.5	5.2	7.0	9.5
DSI	3.64	0.93	0.83	1.05	1.21
SSA BET (m^2/g)	44.7				
ε_F		0.937	0.951	0.985	0.893
ρ_F (g/cm^3)		1.106	1.083	1.026	1.183
Φ_F		0.043	0.055	0.176	0.025

F: feed sample; S.2.b (b: sampling zone).

Table 3. Results of Breakdown of Particle Size Distributions of Flocs for Both Series, as Four Log-Normal Populations

		First Series			Second Series		
Sample	Population	Volume Fraction (Weighting)	Mode (μm)	Range (ln σ)	Volume Fraction (Weighting)	Mode (μm)	Range (ln σ)
First sample	P1	0.07	84	0.826	0.04	14	0.783
	P2	0.45	207	0.526	0.42	52	0.533
	P3	0.37	393	0.334	0.44	100	0.364
	P4	0.11	628	0.193	0.11	169	0.214
Second sample	P1	0.07	78	1.028	0.03	35	0.921
	P2	0.52	223	0.671	0.43	103	0.515
	P3	0.26	479	0.483	0.44	181	0.360
	P4	0.15	1030	0.281	0.10	319	0.204
Third sample	P1	0.12	78	0.932	0.05	206	0.765
	P2	0.43	206	0.596	0.47	435	0.568
	P3	0.30	405	0.413	0.35	899	0.375
	P4	0.15	731	0.250	0.13	1579	0.201
Fourth sample	P1	0.02	78	1.122	0.003	10	0.593
	P2	0.88	332	0.687	0.45	74	0.755
	P3	0.006	451	0.106	0.49	168	0.530
	P4	0.09	1434	0.304	0.06	534	0.208

1 → fine; 2 → small; 3 → medium; 4 → large.

- predominant aggregation for flocs and particles from sampling points 1–3 in the feed-flocculation tank (Figure 3). This results in a large increase in the modes of the four populations and in the mean floc sizes. Comparison with distributions observed on the previous day indicates slower growth of flocs between the point of the first addition of flocculant and point 2. The slower aggregation kinetics are indicative of a lower concentration of solids, implying lower floc volume concentrations. For points 1–3, the predominant dynamics of aggregation are continuous, without visible erosion due to constant increase in the fine floc population mode;
- an abnormally high contrast between the characteristics of samples 2.3 and 2.4. Sample 2.3 has a large mean floc size at 634 μm, and an FSD with moderate range, implying a high initial settling velocity V_o, given the low floc volume concentration. In the interval between samples 2.3 and 2.4 (around 10 min), the settling velocity V_o of the slurry—monitored at the flocculator outlet by the flocculant control system—probably went well beyond the setting, corresponding to an excess of flocculant, causing a large decrease in flocculant distribution pump discharge. When sample 2.4 was taken, the control system, dependent on the time between two V_o measurements, had not restored sufficiently high pump discharge to provide satisfactory flocculation. Regulation was probably disrupted by the large decrease in concentration of solids relative to that for normal circuit operation. The search by the control system for a pump discharge rate that leads to the set interval for V_o is laborious and causes significant temporary fluctuations in pump discharge and settling velocity.

Comparison of size distributions by volume and specific surface area for two feed influents shows variability in the characteristics of washing fines on the site that can affect flocculation performance. The highest specific surface area of the second series feed sample contributes to an increase in the sensitivity of V_o to changes in flocculant pump discharge for a constant concentration of flocculant. Examination of data from the flocculation circuit operating station indicates that instantaneous dosage of flocculant corresponding to second day sample 2.3 is higher than the standard dose of 180 g/t. This leads to a total instantaneous dosage of flocculant that is even higher in the industrial circuit at the time sample 2.3 was taken.

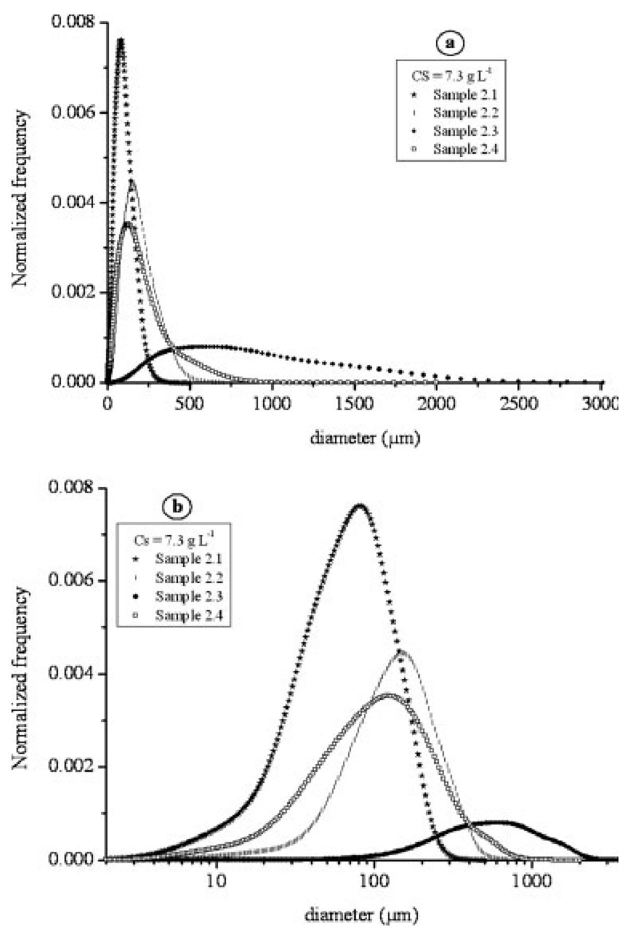

Figure 3. Particle size distribution for the second series.

(a) Normalized frequencies with a linear scale and (b) normalized frequencies with a log scale.

Conclusion

The particle size analysis carried out intermittently on flocculated slurries within the washing fines treatment system confirms that measurement by laser diffraction, using the method developed in the laboratory, is able to determine the effects of conditioning on the size and texture of flocs. The observations made show that the conventional control system located upstream of the settling tank inlet well is not, on its own, able to regulate operation of a flocculation system subjected to variations in production. Installation of a complementary analysis system providing information on floc texture in the settling tank inlet well could make a definite improvement to the level of stability of clarification of process water and to flocculant consumption. It would be possible, for example, to add an in-situ device for analysis and display of flocs moving within the settling tank. By determining a field of velocity and particle sizes for the flocs, such an instrument could ensure optimal control of flocculant injection and of water clarification.

Notation

C_s = solid concentration of the feed slurry (kg/m^3)
C_v = volume concentration measured by particle size analysis (%)
DmF = mean diameter by volume (m)
g = gravitational acceleration (m/s^2)
$\overline{G}$ = average velocity gradient (s^{-1})
ΔH = total head loss (m)
DSI = distribution spreading index [DSI = (d90 − d10)/(2 × d50)]
d_x = diameter corresponding to x% of cumulative undersize by volume (x = 10, 50, or 90)
P = dissipated power (W)
Q = slurry flow rate (m^3/s)
$\overline{t_r}$ = average residence (or conditioning) time (s)
V_F = floc volume (m^3)
V_P = slurry volume (m^3)

Greek letters

ε_F = mean floc porosity
ρ_F = mean floc density (kg/m^3)
ρ_P = slurry density (kg/m^3)
ρ_s = mean density of solids (kg/m^3)
η_0 = water viscosity (Pa s)
η_p = slurry viscosity (Pa s)
Φ_F = floc volume concentration

Literature Cited

1. Alessandrini A, Lapasin R. A class of rheological models for concentrated suspensions: their application to clay/kaolin aqueous systems. *Chem Eng Commun.* 1985;37:29–40.
2. Swithenbank J, Beer JM, Taylor DS, Abbot D, MacCreath GC. *A laser diagnostic technique for the measurement of droplet and particle size distribution, Presented at the AIAA 14th Aerospace Science Meeting*, Washington DC, 1976.
3. Van de Hulst HC. *Light Scattering by Small Particles*. New York: Dover, 1981, pp. 131–187.
4. Bizi M, Baudet G. Contribution of static light scattering to the textural characterization of large aggregates. *J Colloid Interface Sci.* 2006;300:200–209.
5. Idel'cik IE. *Memorandum of the Pressure Drop (in French)*. Paris: Editions Eyrolles, 1999.
6. Graf WH, Altinakar MS. *Hydrodynamics (in French)*. Paris: Eyrolles Editions, 1991.
7. Cauvin A, Guerrée H. *Elements of Hydraulics (in French)*. Paris: Eyrolles Editions, 1986.
8. Sinniger RO, Hager WH. *Constructions Hydrauliques, Vol. 15: Steady Flows. Treaty of Civil Engineering of the Polytechnic School of Lausanne (in French)*. Lausanne: Presses Polytechniques Romandes, 1989.
9. Akers R. *Flocculation*. Rugby, UK: IchemE, 1975.

Manuscript received Feb. 7, 2007, and revision received Sept. 5, 2007.