Modelling the activated sludge flocculation process combining laser light diffraction particle sizing and population balance modelling (PBM)

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Abstract A technique based on laser light diffraction is shown to be successful in collecting on-line experimental data. Time series of floc size distributions (FSD) under different shear rates (G) and calcium additions were collected. The steady state mass mean diameter decreased with increasing shear rate G and increased when calcium additions exceeded 8 mg/l. A so-called population balance model (PBM) was used to describe the experimental data. This kind of model describes both aggregation and breakage through birth and death terms. A discretised PBM was used since analytical solutions of the integro-partial differential equations are non-existing. Despite the complexity of the model, only 2 parameters need to be estimated: the aggregation rate and the breakage rate. The model seems, however, to lack flexibility. Also, the description of the floc size distribution (FSD) in time is not accurate.

Keywords Activated sludge flocculation; floc size distribution; laser light diffraction; population balance modelling

Introduction

In the final step of the activated sludge process (clarification) the sludge needs to be separated from the treated water. Since the sludge is present in flocs, this separation can be done gravitationally. However, the floc size and structure are of great importance in this settling process. In order to improve floc formation, flocculation units are often used prior to the final clarification step. To date, considerable research in the area of activated sludge flocculation has been performed, however, this has not really resulted in sufficient knowledge of the process. Due to this poor understanding, this step in the treatment process is often a bottleneck in the overall system. Indeed, bad flocculation may lead to poor settling of the sludge resulting in bad effluent quality and/or a washout of the sludge from the system.

To improve insight in the activated sludge flocculation process it is necessary to gather quantitative information about floc size distribution and floc structure. Research has shown that this is a difficult task. Some of the techniques applied in the past for this purpose are microscopy, image analysis and Coulter counting. Using these techniques, different studies have resulted in some insight in the flocculation process (Barbusinski and Koscielniak, 1995; Li and Ganczarczyk, 1991; Spicer and Pratsinis, 1996; Serra and Casamitjana, 1998; Grijspeerdt and Verstraete, 1997). Some practical drawbacks encountered by these methods are that they: (1) cannot be used on-line in a flocculation experiment that proceeds, (2) are very time consuming, (3) do not cover the needed floc size range or (4) need dilution with electrolyte.

Alternatively, other variables such as SVI, turbidity and settling velocity have been used frequently to describe flocculation and settling. Wahlberg *et al.* (1994) used standardised flocculation tests to follow turbidity of the clarifier supernatant in order to monitor and compare the sludge settling properties of 21 different WWTP. Wilén (1999) monitored SVI

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and turbidity to monitor influences of dissolved oxygen concentration on the flocculation of the sludge. However, these variables are unable to describe the detailed interactions of activated sludge flocculation, which are needed to understand the underlying mechanisms and eventually model the process.

Other techniques that are able to quickly provide quantitative information about the process under study are light scattering techniques such as laser rotating and laser light diffraction. The first technique was successfully applied to monitor particle aggregation and break-up in a shear flow (Serra *et al*., 1996). The latter method has been applied to characterise the floc structure (Guan *et al*., 1998; Waite, 1999). It can also be applied to monitor changes in floc size distribution (Wilén, 1999; Biggs and Lant, 2000).

Recently, Biggs and Lant (2000) customised an on-line technique developed by Spicer *et al*. (1998) to follow dynamic changes in the floc size distribution (FSD) of activated sludge using on-line laser light diffraction. This technique allows the dynamics of the activated sludge flocculation process to be followed in a quantitative way.

To improve the understanding of several biological and physico-chemical processes and to be able to control them, mathematical modelling has already been shown to be a valuable tool. In the area of wastewater treatment this has resulted in Activated Sludge Models 1–3 (ASM), in which the biological processes taking place in the treatment process (e.g. COD-removal, (de)nitrification and phosphorous removal) are described. However, in the complete treatment plant models built around these ASMs, the model of the final clarification step tends to be the weakest part.

Efforts have been performed to model clarifiers by using Computational Fluid Dynamics (CFD) (Stamou and Rodi, 1989; Krebs, 1991; Zhou and McCorquodale, 1992; Lyn *et al*., 1992; Armbruster *et al*., 2000). CFD primarily aims at modelling the fluid dynamics of the clarifier by using continuity, momentum and turbulence equations. This allows us to predict a velocity and sludge concentration pattern for the complete clarifier both in space and time.

In such models the transport of water can be included quite easily. To be able to predict the sludge concentration profile in the settler, however, more detailed information about the activated sludge settling process is needed. This sludge settling depends on floc settling properties like floc size and floc structure. These are influenced by local flocculation conditions including shear forces (induced by the flow regime), dissolved oxygen concentration, loading rate, sludge concentration, ionic strength, etc., which vary throughout the settler. Detailed information about the activated sludge flocculation process and the interactions between the fluid and solid phase should, therefore, be included in the model.

To model the flocculation process to a reasonable degree of detail for inclusion in a CFD model, a more sophisticated "segregated" model, often referred to as Population Balance Model (PBM), is needed (Ramkrishna, 1979; Frederickson, 1991).

In this contribution a PBM and the laser light diffraction technique will be combined to collect flocculation-relevant experimental data to support these models and show how this combination of new approaches can further improve our knowledge about the activated sludge flocculation process and the influences it is subject to.

Experimental data collection using laser light diffraction

Quantitative information about floc structure, floc size distributions (FSD) and their derived equivalent diameters can be obtained by using a Malvern Mastersizer (Malvern Instruments, Malvern, UK) (Guan *et al*., 1998; Waite, 1999; Wilén, 1999; Biggs and Lant, 2000). The measurement technique of this apparatus is based on laser light diffraction. Particles (i.e. flocs) are brought in a laser beam where they cause forward-scattering of the incoming light. Particles of different sizes will scatter the light differently. Hence, a heterogeneous particle population will result in a diffraction pattern. This pattern is detected and the Mie diffraction model (van de Hulst, 1981) or an approximation to it can be used to translate it into a FSD. Another drawback of the diffraction model is the assumption of sphericity of particles, which makes the interpretation of the FSD difficult. A drawback of the technique is the need for considerable sample dilution (< 0.2 g/l) to avoid multiple scattering since this is not taken into account in the diffraction model that derives the FSD from the diffraction pattern. Measurements were therefore performed using samples diluted with filtered effluent (0.45 µm). The flow rate was determined in a way to minimise shear effects and pump pulsation influences (to approximate isokinetic sampling). A flow rate of 3ml/s was selected (Biggs and Lant, 2000).

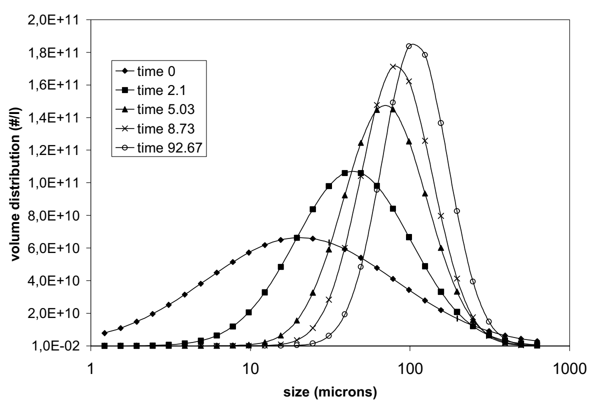
However, the main advantage of laser light diffraction over the previously mentioned techniques is that it can easily be used on-line in an experimental set-up, which makes it a useful and fast tool to follow dynamic changes in FSD's as shown in Figure 1 (left).

A characteristic derived from the FSD that is often used to describe the floc size is the volume weighted equivalent diameter or the mass mean, D[4,3]. This variable is calculated as:

$$D[4,3]=\frac{\sum_{i=1}^{n}\Delta F_N(x)_i\, x_i^4}{\sum_{i=1}^{n}\Delta F_N(x)_i\, x_i^3} \tag{1}$$

with: $x_i =$ diameter of size class i

$\Delta F_N(x)_i =$ number fraction in size class i $= N_i \big/ \sum N_i$



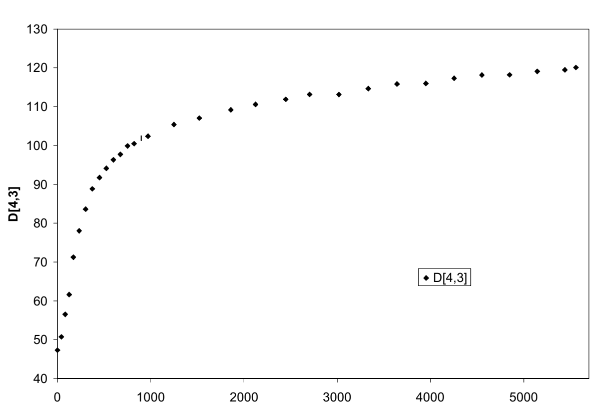


Figure 1 Example of an on-line measurement of the dynamical change in D[4,3] due to applying a constant average velocity gradient of 37 s^{-1}

In the experimental results shown in this paper the sludge was sonicated prior to each experiment to bring it into a standard condition. It was observed that at time zero, a large number of particles have a diameter around 10 μm. These particles are often referred to as microcolonies. They consist of a few micro-organisms bridged together with polyvalent cations that bind to the extracellular polymers present around the organisms (Higgins and Novak, 1997).

By using the on-line technique elaborated by Biggs and Lant (2000), the evolution of both the FSD and the mass mean from this standard condition can be easily followed. An example of the dynamic change in FSD caused by applying a constant average velocity gradient, G, of 37 s^{-1} to a sonicated sludge is shown in Figure 1. The evolution of the FSD (Figure 1 left) reveals a shift towards larger sizes, revealing that flocculation is occurring. This shift almost reaches a steady state at the end of the experiment. The shift to larger sizes can also be clearly observed in the evolution over time of the mass mean (Figure 1 right).

Population balance modelling

An early effort to model the activated sludge flocculation process was performed by Parker *et al.* (1972) in order to be able to describe changes in settling characteristics. The proposed model describes the changes in supernatant primary particles (turbidity measurements) after settling and was also validated by Wahlberg *et al.* (1994). Floc aggregation and break-up appeared to be key processes that occur simultaneously. However, the model does not allow overall modelling of the settler, since it only provides information concerning primary particles in the supernatant, determining effluent suspended solids.

Instead of approaching the biomass as a “lumped biophase”, sludge can be viewed as a segregated population of individual flocs. This individuality approach implies that all floc properties are no longer average values, but are given by number distributions (distributed model). These segregated models, also called Population Balance Models (PBMs), allow the description of dynamic changes in these property distributions when the conversion terms are known. Considering floc size as the floc property, conversion terms can be interpreted as aggregation and break-up of flocs and the number distribution based on floc size (N) becomes:

$$\frac{dN}{dt} = aggregation + breakage \tag{2}$$

PBMs have been successfully applied in a number of different disciplines dealing with particle or droplet populations (Hounslow *et al.*, 1988; Kusters, 1991; White and Ilievski, 1996; Spicer and Pratsinis, 1996; Ramkrishna, 2000). The main difference of a PBM compared to the model proposed by Parker *et al.* (1972) is that it describes changes in the complete particle size distribution instead of only the fraction of primary particles.

Biggs (2000) showed that a PBM based on the aggregation model introduced by Hounslow *et al.* (1988) and the breakage model described by White and Ilievski (1996) could be used to describe the activated sludge flocculation process. Both aggregation and breakage can lead to “birth” and “death” of flocs of a certain size. The evolution of the volume-based number distribution n(v) is a result of these four mechanisms and is given by:

$$\frac{\mathrm{d\ n(v)}}{\mathrm{dt}} = \mathrm{B(v)_{agg}} - \mathrm{D(v)_{agg}} + \mathrm{B(v)_{break}} - \mathrm{D(v)_{break}} \tag{3}$$

Volume-based integral expressions of aggregation birth ($B(v)_{agg}$) and aggregation death ($D(v)_{agg}$) can be found in Hounslow *et al.* (1988), those for breakage birth ($B(v)_{break}$) and breakage death ($D(v)_{break}$) in White and Ilievski (1996). Solving this “integro-partial

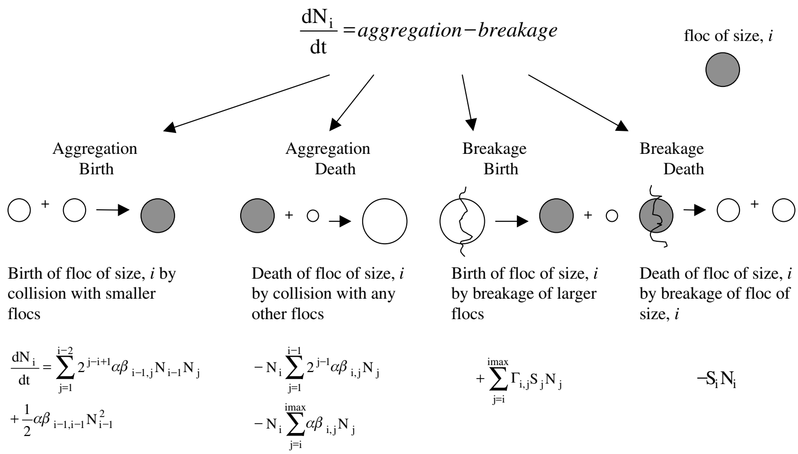


Figure 2 Aggregation and breakage dynamics of the discretised population balance (Biggs, 2000)

differential equation" is, however, no trivial task. Since an analytical solution is not possible, a discretisation allowing numerical integration is needed. This discretisation divides the particle size range into a number of classes, each represented by a floc size and volume. The discretised PBM and the expressions used to describe the different processes were obtained by Hounslow *et al.* (1988) and are summarised in Figure 2.

In this model N_i (#.l^{-1}) is the number concentration of flocs of size i, α (–) is the collision efficiency, and β_{ij} ($m^3.s^{-1}$) is the collision frequency for particles of volume v_i (m^3) and v_j (m^3). S_i (s^{-1}) is the breakage rate of flocs of size i and $\Gamma_{i,j}$ (–) is the breakage distribution function which defines the volume fraction of the fragments of size i produced from j sized flocs. The parameter α (collision efficiency) was introduced in the discretised model in the aggregation expressions. This parameter (value between 0 and 1) represents the number of successful collisions since not every collision will result in aggregation. It can be interpreted as a correction factor that is introduced to correct the number of collisions (β), which is obtained only on the basis of the floc size as floc property. Details on the discretisation-grid and the mass conservation can be found in Hounslow *et al.* (1988). Discretisation resulted in 28 floc size classes and therefore in 28 differential equations that need to be solved simultaneously.

In order to make the described model operational, functional relationships of the collision frequency $\beta_{i,j}$, the breakage rate S_i and the breakage distribution function $\Gamma_{i,j}$ must be available. Spicer and Pratsinis (1996) described the collision frequency, $\beta_{i,j}$, in terms of the volume of the particles (v_i,v_j) that collide and the average velocity gradient, G:

$$\beta_{i,j} = 0.31\,G\left(v_i^{1/3} + v_j^{1/3}\right)^3 \tag{4}$$

in which: $$G = \left(\frac{\varepsilon}{\upsilon}\right)^{\frac{1}{2}} \tag{5}$$

with: υ= the kinematic viscosity ($m^2.s^{-1}$)
ε = the average turbulent energy dissipation rate ($m^2.s^{-3}$)

Through Eqs (4) and (5), the PBM becomes directly linked to the CFD results, since the value of ε is produced by the CFD-model.

The breakage rate (S_i) can be described as function of the particle volume (Spicer and Pratsinis, 1996; Serra and Casamitjana, 1998):

$$S_i = A\, v_i^a \tag{6}$$

with: a = constant (=1/3)
A = the breakage rate coefficient ($cm^{-3a}s^{-1}$)

It is assumed that only binary breakage occurs. This means that flocs break into 2 smaller flocs with equal volume.

Despite the complexity of the above model, only 2 parameters need to be estimated from the FSD data: α, the collision efficiency, and A, the breakage rate coefficient. More important, however, in view of the future use of such a model for better understanding and prediction of flocculation (and thus settling) is that one may try to derive relationships between these two key model parameters and (1) applied environmental conditions such as shear rate, ionic strength, temperature, … and (2) floc properties (structure, strength, density, settling velocity and SVI). These relationships should then be built into the PBM. Indeed, it can be expected that both α and A will not be constant throughout the complete flocculation process and that they could be time-varying (through a changing environmental factor or floc property). To model this time-dependency, however, more detailed knowledge about the flocculation process is needed with regard to relationships between floc growth and floc density on the one hand and the rate of both aggregation (α) and breakage (A) on the other hand. Some work on the functionality of α has already been reported in the literature (Han and Lawler, 1992; Thomas *et al.*, 1998).

Illustrative results and discussion

This PBM-framework was used to check (1) the influence of shear rate (G), (2) the effect of addition of calcium ions on the flocculation dynamics and (3) the capability of the model to describe activated sludge flocculation.

Shear rate

To investigate the influence of shear rate on the flocculation process, different levels of shear (G) were applied. At most applied G-values (except $346s^{-1}$), an initial steep increase in mass mean diameter is observed when starting from standardised (sonicated) sludge. A steady state is, however, not reached and a slow increase continues. At G = $346s^{-1}$, no significant increase was observed. As expected, it was found that at higher average velocity gradients the final mass mean floc size decreased (Figure 3, left).

A power law relationship was found between the average velocity gradient and the final mass mean floc size.

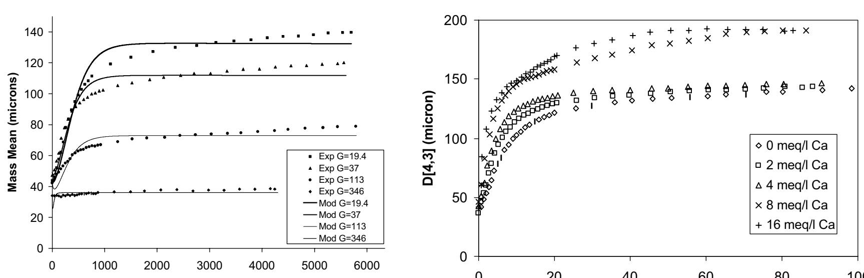


Figure 3 (Left) Change in floc size with time for different average velocity gradients (Biggs, 2000); (right) effect of different calcium concentrations on the flocculation dynamics

By increasing the average velocity gradient for a short time (1 min) and immediately returning it to its original value, the regrowth behaviour of the activated sludge flocs was demonstrated. A slightly lower final mass mean floc size was reached compared to the one observed before the increase in average velocity gradient (Figure 4a).

Biggs (2000) also investigated the effect of shear history by increasing the average velocity gradient for a longer period of time (35 min). Again, regrowth was observed. However, a smaller final mass mean floc size was reached (Figure 4b). This phenomenon was already observed by Spicer *et al*. (1998), who found smaller and more dense flocs after a cycled shear regime. Lou and Ghosh (1988) proposed that the extent of damage to the extracellular polymers could be larger for an extended period of increased shear.

Calcium addition

The effect of the calcium concentration on the floc size was investigated by adding different concentrations of $CaCl_2$ to sonicated sludge mixed at 19.4 s^{-1}. Similar flocculation dynamics are obtained compared to the experiments with different shear. It appeared that an addition of more than 8 meq/l of calcium was needed to observe a significant increase of the final mass mean floc size (Figure 3, right).

By measuring calcium, sodium and magnesium ion concentrations before and after the experiments, it was also found that ion exchange took place. Calcium was consumed, while both sodium and magnesium were released.

Model capability

The PBM was applied to fit the evolution of the mass mean of activated sludge flocculation experiments at different shear levels. Although it is clear that the model is able to approximate the data from Figure 3 (left), it can be seen that the model predicts a steady state mass mean floc size which is not observed in reality. The model as described in this paper is thus not flexible enough to accurately describe the experimental data. A possible explanation for this lack of flexibility can be that the model structure is not correct. This can be caused (1) by the fact that processes are described incorrectly or processes are missing or, (2) as stated before, dependencies of α and A on environmental conditions and floc properties have to be investigated and need to be implemented.

Compared to the parameter values found for inorganic flocculating systems (Spicer and Pratsinis, 1996), the value found for α in activated sludge flocculation was significantly smaller (10^{-4}). Despite this low collision efficiency value, aggregation does take place (Figure 3, left).

In terms of functional relationships between PBM-parameters and shear rate G, a power law relationship (Spicer and Pratsinis, 1996) between the breakage rate coefficient A and the average velocity gradient G was adopted:

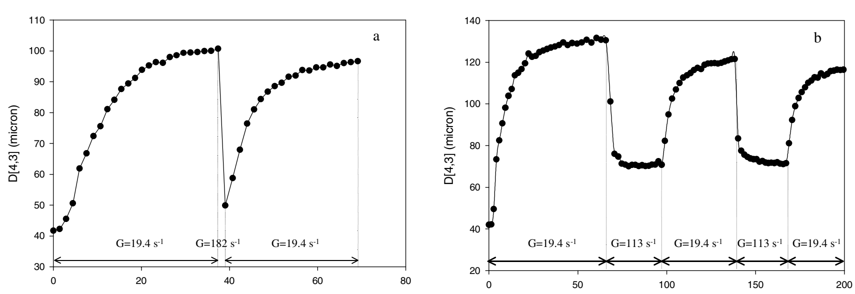


Figure 4 Illustration of the effect of shear history on the activated sludge flocculation (Biggs, 2000): (a) short (1 min) increase of G and (b) longer increase (35 min) of G

$$A = A' G^y \tag{8}$$

where A' and y are constants. Values of A' = 0.62 and y = 0.45 were found, which were quite different from those found in inorganic systems (A' = 0.0047 and y = 1.6) (Spicer and Pratsinis, 1996). Both parameters A' and y can be related to floc strength and floc structure. The collision efficiency α decreased at higher shear rates.

The PBM was also successfully used to model the flocculation dynamics at different calcium concentrations (results not shown). The collision efficiency α increased with increasing calcium concentration. This is logical since calcium is thought to provide cationic bridging between bacteria. From this it can be concluded that α indeed is dependent on environmental conditions, as stated before. However, no clear relationship could be found.

Conclusions

The combination of the experimental technique developed by Biggs and Lant (2000) and the PBM-framework allows us to gather more insight into the activated sludge flocculation process. However, the model appears to lack flexibility and the model structure should be reviewed by means of (1) changing current process descriptions or adding processes or (2) adding some parameter dependencies on other factors. The latter requires additional relationships between key model parameters and (1) environmental conditions and (2) floc properties need to be identified and implemented in the model. This will lead to an improved understanding of the two mechanisms of major importance in flocculation, aggregation and breakage respectively, represented in the model by a and A.

Since the PBM-framework also takes into account the fluid characteristics such as viscosity and shear stress (Eqs (4) and (5)), it is already linked to the CFD-model and can therefore be included in it. Local flocculation rates could therefore be predicted and settling rates could be connected to it. This would allow for a complete modelling of the final clarifier in both space and time, allowing it to predict its performance.

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References

Armbruster, M., Krebs, P. and Rodi, W. (2000). Numerical modelling of dynamic sludge blanket behaviour in secondary clarifiers. *Wat. Sci. Tech.*, **43**(11), 173–180.

Barbusinski, K. and Koscielniak, H. (1995). Influence of substrate loading intensity on floc size in activated sludge process. *Wat. Res.*, **29**, 1703–1710.

Biggs, C.A. (2000). *Activated sludge flocculation: Investigating the effect of shear rate and cation concentration on flocculation dynamics*. Ph.D thesis, University of Queensland, Australia.

Biggs, C.A. and Lant, P.A. (2000). Activated sludge flocculation: On-line determination of floc size and the effect of shear. *Wat. Res.*, **34**, 2542–2550.

Frederickson, A.G. (1991). Segregated, structured, distributed models and their role in microbial ecology: a case study based on work done on the filter-feeding ciliate *Tetrahymwena pyriformis*. *Microb. Ecol.*, **22**, 139–159.

Grijspeerdt, K. and Verstraete, W. (1997). Image analysis to estimate the settleability and concentration of activated sludge. *Wat. Res.*, **31**, 1126–1134.

Guan, J. , Waite, T.D. and Amal, R. (1998). Rapid structure characterization of bacterial aggregates. *Environ. Sci. Technol.*, **32**, 3735–3742.

Han, M.Y. and Lawler, D.F. (1992). The (relative) insignificance of G in flocculation. *J. Am. Water Works Assoc.*, **84**, 79–91.

Higgins, M.J. and Novak, J.T. (1997). The effect of cations on the settling and dewatering of activated sludges: Laboratory results. *Water Environ. Res.*, **69**, 215–224.

Hounslow, M.J., Ryall, R.L. and Marshall, V.R. (1988). A discretized population balance for nucleation, growth and aggregation. *AIChE Journal*, **34**(11), 1821–1832.

Krebs, P. (1991). The hydraulics of final settling tanks. *Wat. Sci. Tech.*, **23**(4–6), 1037–1046.

Kusters, K.A. (1991). *The influence of turbulence on aggregation of small particles in agitated vessels*. Ph.D Thesis, Eindhoven University of Technology, Eindhoven, The Netherlands.

Lou, R.J. and Ghosh, M.M. (1988). Polyelectrolyte characteristics and flocculation. *J. Am. Wat. Works Association (AWWA)*, **80**, 159–167.

Li, D.H. and Ganczarczyk, J.J. (1991). Size distribution of activated sludge flocs. *Res. J. Wat. Pollut. Control Fed.*, **63**, 806–814.

Lyn, D.A., Stamou, A.I. and Rodi, W. (1992). Density currents and shear-induced flocculation in sedimentation tanks. *J. Hydr. Eng.*, **118**, 849–867.

Malcherek, A. (1994). Numerical modelling of cohesive settling velocities. *Int. J. of Sediment Research*, **9**, 97–106.

Ramkrishna, D. (1979). Statistical models of cell populations. *Adv. Biochem. Eng.*, **11**, 1–47.

Ramkrishna, D. (2000). *Population Balances: Theory and applications to particulate systems in engineering*. Academic Press, London, UK, 355p.

Serra, T. and Casamitjana, X. (1998). Effect of the shear and volume fraction on the aggregation and breakup of particles. *AIChE Journal*, **44**, 1724–1730.

Serra, T., Colomer, J. and Casamitjana, X. (1996). Aggregation and breakup of particles in a shear flow. *J. Colloid Interface Sci.*, **187**, 466–473.

Spicer, P.T. and Pratsinis, S.E. (1996). Coagulation and fragmentation: universal steady state particle size distribution. *AIChE Journal*, **42**, 1616–1620.

Spicer, P.T., Pratsinis, S.E., Raper, J., Amal, R., Bushell, G. and Meesters, G. (1998). Effect of shear schedule on particle size, density, and structure during flocculation in stirred tanks. *Powd. Techn.*, **97**, 26–34.

Stamou, A.I. and Rodi, W. (1989). Numerical modeling of flow and settling in primary rectangular clarifiers. *J. Hydr. Res.*, **27**, 665–682.

Thomas, D.N., Judd, S.J. and Fawcett, N. (1998). Flocculation modelling: a review. *Wat. Res.*, **33**, 1579–1592.

Van de Hulst, H.C. (1981). *Light scattering by small particles*. Dover Publications, NY, 470p.

Waite, T.D. (1999). Measurement and implications of floc structure in water and wastewater treatment. *Coll. and Surf.: A: Physicochemical and Engineering Aspects*, **151**, 27–41.

Wahlberg, E.J., Keinath, T.M. and Parker, D.S. (1994). Influence of activated sludge flocculation time on secondary clarification. *Water Environ. Res.*, **66**, 779–786.

Wilén, B-M. (1999). Properties of activated sludge flocs. Ph.D Thesis, Department of Sanitary Engineering, Chalmers University of Technology, Goteborg, Sweden.

White, E.T. and Ilievski, D. (1996). The use of the population balance for modelling metallurgical systems. *Emerging Separation Technologies for Metals* II. R.G. Bautista (Ed.), The Minerals, Metals and Materials Society: 91–103.

Zhou, S. and McCorquodale, J.A. (1992). Modeling of rectangular settling tanks. *J. Hydr. Engrg.*, **118**, 1391–1405.