Solving the inverse problem for aggregation in activated sludge flocculation using a population balance framework

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Abstract Many systems contain populations of individuals. Often, they are regarded as a lumped phase, which might, for some applications, lead to inadequate model predictive power. An alternative framework, Population Balance Models, has been used here to describe such a system, activated sludge flocculation in which particle size is the property one wants to model. An important problem to solve in population balance modelling is to determine the model structure that adequately describes experimentally obtained data on for instance, the time evolution of the floc size distribution. In this contribution, an alternative method based on solving the inverse problem is used to recover the model structure from the data. In this respect, the presence of similarity in the data simplifies the problem significantly. Similarity was found and the inverse problem could be solved. A forward simulation then confirmed the quality of the model structure to describe the experimental data.
Keywords Flocculation; inverse problem; population balances; particle sizing

Introduction

Mathematical modelling of wastewater treatment plants has reached a level of maturity that allows these models to be applied to solve a wide range of problems (design, operation, control,…). However, looking at the complexity of the models for the different process units, it is still apparent that the models used for secondary clarification are considerably simpler than the ones used for the biodegradation processes. The latter is not really an issue when the clarifier is built sufficiently large and does not cause any problem for the overall plant performance. However, when one wants to exploit the full capacity of the clarifier (i.e. pushing it to its limits) these simple clarifier models become useless as they are too far off from reality and will lead to too optimistic predictions of performance.

In the last decade, a lot of knowledge about the hydrodynamics has been gained by means of Computational Fluid Dynamics (CFD) (McCorquodale *et al.*, 1991; Krebs *et al.*, 1996; Lakehal *et al.*, 1999; De Clercq *et al.*, 2007). This has already led to better design of clarifiers (e.g. modified inlet structures, baffles,…). However, for an even better performance of clarifier models, the flocculation of the activated sludge flocs and the possible handles to interact with that process should be better understood.

Many environments contain populations of individuals. These individuals can be of all kind, e.g. sludge flocs in wastewater treatment systems, rivers and estuaries, or particles in sewer systems, to name but a few. These populations are mostly treated as lumped entities exhibiting mean properties (e.g. sludge concentration in ASM models). However, for some applications this approach is too rough leading to inaccurate model predictions.

Hence, a special modelling framework is needed for this type of applications: Population Balance Models (PBM). The so-called segregated models that are used in this PBM framework have been around for quite some time (Hulburt and Katz, 1964), mainly in chemical engineering. However, due to their intensive calculation requirements they have not been used extensively until roughly 2 decades ago.

Recently, however, the PBM-framework has found its way to biological systems as well (Biggs and Lant, 2002; Nopens *et al.*, 2002). The main challenge for their successful application has been to retrieve the correct model structure within the PBM framework because the functions that have been used so far in other fields to describe the discrete events in flocculation such as aggregation and breakage seem not to be valid (Nopens *et al.*, 2005). In this contribution, an alternative procedure based on solving the inverse problem is applied in order to recover the model structure from experimental data. The procedure is applied to an aggregating activated sludge. Prior to this, typical flocculation experiments and the general PBM framework are introduced.

Flocculation experiments

Several attempts have been made in literature to study activated sludge flocculation. Biggs and Lant (2002) was the first to obtain *on-line* experimental data of activated sludge flocculation. They used a 1L mixed vessel filled with diluted activated sludge and pumped this through a laser diffraction device (Malvern Mastersizer, Malvern, UK) allowing retrieval of quantitative information about the flocculation process. The online nature of these experiments allowed Floc Size Distributions (FSD) to be collected with a frequency of approximately 1 per minute. Experiments were conducted at different levels of shear intensity and calcium additions. A mass mean diameter ($D[4,3]$ calculated from the 4th and 3rd integral moments of the FSD) can be used to illustrate the dynamics of the FSD (Biggs and Lant, 2002; Govoreanu, 2004). The results of a typical (de)flocculation experiment are shown in Figure 1. It is clear that the average floc size changes drastically under different regimes of shear. The frequency of the data allows for mathematical modelling of the activated sludge process. The experimental period between 2000 s and 4000 s is used in this contribution. It describes the flocculation from a deflocculated sludge (developed under a high shear intensity of $G = 113 \mathrm{s}^{-1}$) at a decreased shear
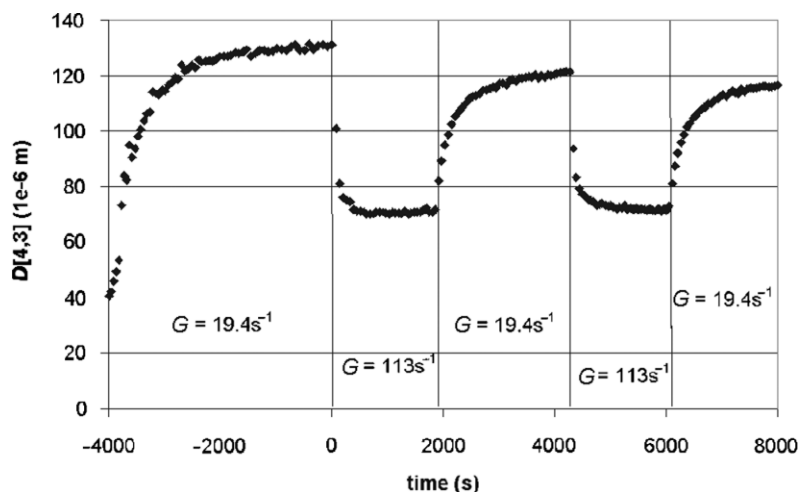

Figure 1 Results from a typical (de)flocculation experiment in terms of $D[4.3]$ versus time (after Biggs and Lant, 2002)

intensity of $G = 19.4\text{s}^{-1}$. Such flocculation may actually occur when sludge moves between different shear zones in a wastewater treatment plant.

Population balance framework

The first literature report of the general framework of population balances was by Hulburt and Katz (1964). An excellent review of the state-of-the-art in population balances can be found in Ramkrishna (2000). PBMs have been widely applied to describe processes involving dynamical behaviour of population properties. Applications can be found in different scientific areas such as crystallisation (Randolph and Larson, 1971), flocculation of inorganic dispersed systems (Spicer and Pratsinis, 1996), polymerisation (Blatz and Tobolsky, 1945), precipitation (Manjunath *et al.*, 1994), flotation (Haarhof and Edzwald, 2001), granulation (Iveson, 2002), cell culture dynamics (Mantzaris *et al.*, 1999) and aerosol dynamics (Hidy and Brock, 1970), to name but a few. Depending on the number of properties being described by the model, a PBM can be categorized as either one- or multidimensional. The general format of a one-dimensional number-based PBM looks like (Hulburt and Katz, 1964; Ramkrishna, 2000):

$$\frac{\partial f(x,t)}{\partial t} + \frac{\partial}{\partial x}(\dot{X}(x,t)f(x,t)) = h(x,t) \tag{1}$$

where x is a property of the individuals, $f(x,t)$ is the number-based property distribution function ($\#.\text{cm}^{-3}$), $\dot{X}(x,t)$ is the time derivative of the property x (s^{-1}) and $h(x,t)$ the net aggregation rate ($\#.\text{cm}^{-3}.\text{s}^{-1}$). The right-hand-side of Equation 1 often contains integral functions of $f(x,t)$ describing aggregation and/or breakage processes, turning it into an integro-differential equation which is hard to solve analytically. Alternative techniques to solve this type of equations are summarised in Ramkrishna (2000).

In the particular application of the aggregation part of activated sludge flocculation, x was chosen to be the particle size and $h(x,t)$ consists of 2 processes: aggregation birth (AB) and aggregation death (AD). Indeed a particle of size x can both be formed and disappear through aggregation. Aggregation models are often based on the Smoluchowski model (Thomas *et al.*, 1999; Ramkrishna, 2000):

$$h(x,t)_{\text{agg}} = \frac{1}{2}\int_0^x \alpha(x-x',x')f(x-x',t)f(x',t)\mathrm{d}x' - f(x,t)\int_0^\infty \alpha(x,x')f(x',t)\mathrm{d}x' \tag{2}$$

where $\alpha(x,x')(\text{cm}^3.\#^{-1}\text{s}^{-1})$ is the collision frequency for particles of volume x (cm^3) and x' (cm^3). However, Nopens *et al.* (2005) showed that the expressions found in the literature to describe these phenomena (also called *kernels*) did not perform well when confronting the model predictions with experimental data.

The inverse problem for aggregation

An alternative way of retrieving the kernels based on experimental data is to solve the inverse problem (Wright and Ramkrishna, 1992; Sathyagal *et al.*, 1995). The different steps of this process are depicted in Figure 2.

The idea behind a similarity analysis is to look for a collapsing solution in the dynamic part of the experimental data, i.e. one looks for a function that allows to transform the set of experimental distributions collected at different time instants into a set of distributions that collapse onto a single distribution. This collapsing solution allows to rewrite the PBM in a simpler form, which considerably simplifies the solution of the inverse problem. In an aggregating context, Wright and Ramkrishna (1992) showed that this practically means that a scaling function $h(t)$, given by the ratio of two successive
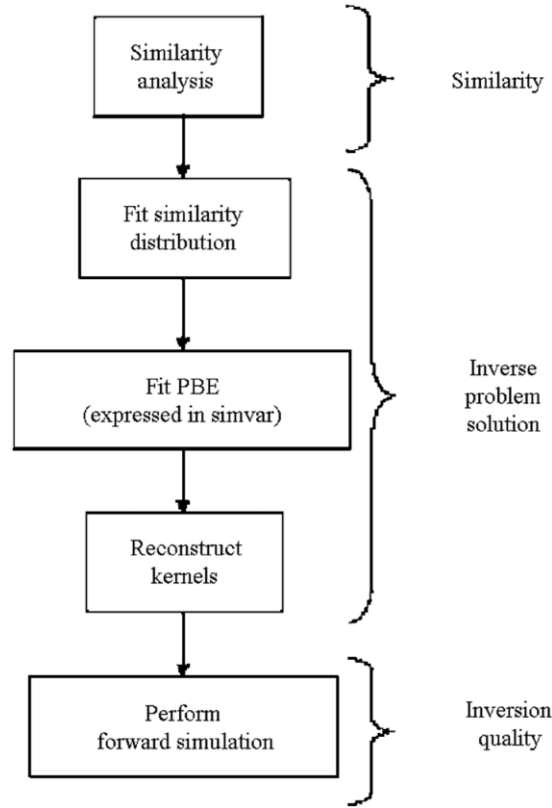
Figure 2 Schematic overview of the steps related to the inverse problem solution

integral moments μ of the number density function $f_1(x, t)$,

$$h(t) = \frac{\mu_{k+1}(t)}{\mu_k(t)} = \frac{\int_0^\infty x^{k+1} f_1(x, t) dx}{\int_0^\infty x^k f_1(x, t) dx} \tag{3}$$

is computed and is used to rescale the particle volume distribution into the similarity variable

$$\eta \equiv \frac{x}{h(t)} \tag{4}$$

Since the cumulative distribution $F(x,t)$ is itself a candidate for the self-similar solution, this distribution is rescaled and a k-value is looked for that makes all distributions collapse onto one single distribution, the self-similar distribution. The process of determining k proceeds through trial and error.

The next step to solve the inverse problem (Figure 2) is to redefine the PBM in terms of the similarity variable as shown by Wright and Ramkrishna (1992):

$$\eta \Phi(\eta) = \int_0^\eta d\eta' \, \Phi(\eta') \int_{\eta-\eta'}^\infty d\eta'' \, \frac{\Phi(\eta'')}{\eta''} \, \frac{\alpha(\eta', \eta'')}{\langle \alpha \rangle} \tag{5}$$

Here, $\alpha(\eta',\eta'')/\langle\alpha\rangle$ is the unknown function that needs to be reconstructed. It represents the scaled aggregation frequency divided by the mean scaled aggregation frequency.

At this moment, a constrained quadratic minimisation problem can be formulated. The similarity coordinate is discretised into a set of $\{\eta_i\}$ and $\alpha(\eta', \eta'')$ is expanded in terms

of basis functions

$$\frac{\alpha(\eta', \eta'')}{\langle \alpha \rangle} = \sum_{j=1}^{n_b} a_j A_j(\eta', \eta'') \tag{6}$$

A matrix $X_{i,j}$ is formulated

$$X_{i,j} = \int_0^{\eta i} d\eta' \, \Phi'(\eta') \int_{\eta i - \eta'}^{\infty} d\eta'' \frac{\Phi'(\eta'')}{\eta''} A_j(\eta', \eta'') \tag{7}$$

which results in the following quadratic minimisation problem:

$$\min(a^T (X^T X + \lambda_{\mathrm{reg}} D) a - 2a^T X^T g \tag{8}$$

that allows to obtain the unknown vector a. To cure for the ill-posedness of the problem Tikhonov regularisation (λ_{reg}) is included in the minimisation (Wright and Ramkrishna 1992).

The constraints that are used are physical constraints derived from the definition of the aggregation frequency: (1) the aggregation frequency is a symmetric function of its arguments, and (2) the aggregation frequency is positive everywhere.

An interesting set of basis functions that can be used for this kind of inversion problem are so-called *Laguerre polynomials*, given by:

$$L_n(x) = \sum_{m=0}^{n} (-1)^m \frac{n!}{(n-m)! m!^2} x^m \tag{9}$$

In order to simplify the matrix equations for the determination of a, as introduced above, a straightforward method is needed for the representation of the similarity distribution $\Phi'(\eta')$ (note that this is the derivative of the cumulative similarity distribution $\Phi(\eta')$). Other advantages of the fitting of the similarity curve lie in the limitation of experimental error and the guarantee that the similarity distribution is continuous. The mathematical expression that is fitted to the distribution should exhibit analogous qualitative and quantitative behaviour of the self-similar distributions. A linear combination of distributions exhibits that behaviour, since γ-distributions have the ability to be singular at the origin, but also can show small η-behaviour falling back toward the origin (two different types of behaviour that have been observed in similarity distributions of known aggregation kernels in practice). The expansion of the similarity distribution in terms of γ-distributions is:

$$\Phi'(\eta) = \sum_{k=1}^{n_{\mathrm{term}}} A_k \eta^{\alpha_k - 1} \exp(-\beta_k \eta) \tag{10}$$

where n_{term} is the number of γ-distributions used in the expansion and A_k, α_k and β_k are parameters of the kth γ-distribution.

Since an analytical form is available for $\Phi'(\eta')$, certain simplifications of the matrix equations can be accomplished. Equation 7 can be rewritten as

$$\bar{X}_{ij}(m) = \int_0^{\eta_m} d\eta' \, \Phi'(\eta') L_i(\eta') \bar{Y}_j(\eta_m - \eta') \tag{11}$$

where η_m is a point of discretisation and is $\bar{Y}_j(\eta_m - \eta')$ defined by

$$\bar{Y}_j(\eta_m - \eta') = \int_{\eta i - \eta'}^{\infty} d\eta'' \frac{\Phi'(\eta'')}{\eta''} L_j(\eta'') \tag{12}$$

Results and discussion

The data used in this exercise were obtained from dynamic experiments in a 10 L flocculator. The floc size distributions were measured using a Malvern Mastersizer (Malvern, UK), a technique based on forward laser scattering. A thorough description of the experiments can be found in Govoreanu *et al.* (2004).

First, a similarity analysis was performed on a dataset of aggregating activated sludge that was sonicated prior to aggregation. However, no collapsing distribution could be obtained for any value of k that was tested. However, a value of k and, hence, similarity could be found for an aggregating experiment where the flocs were broken through an increased shear intensity. The initial and collapsed cumulative distributions are given in Figure 3 and Figure 4 respectively. The optimal k-value was found to be -0.35. Another experiment conducted under the same conditions confirmed this similarity, albeit for a slightly different k-value (-0.3). The fact that this similarity did not exist for the case where sonication was applied prior to flocculation suggests that a different kind of aggregation is taking place, i.e. a different expression will be needed to describe this type of reflocculation after sonication. Since sonication does not occur in the real activated sludge process, it can be concluded that lab experiments studying flocculation starting from a sonicated sludge will not lead to the extraction of the correct flocculation behaviour (i.e. aggregation kernel), and the results obtained cannot be extrapolated to the full-scale case.

In a next step an analytical description of the similarity distribution is needed so as to simplify the inverse problem solution. In this case 2 γ-distributions were found appropriate to describe the similarity distribution (not shown).

Finally, the inverse problem was solved by computing the left-hand side of Equation 5 at each discrete value of the similarity variable. The goal of inversion is to reproduce this left-hand side with a linear combination of double integrals present at the right-hand side of the PBE for different combinations of Laguerre polynomials. This is done at all discretised values of the similarity variable. The number of Laguerre polynomials (*nb*) was
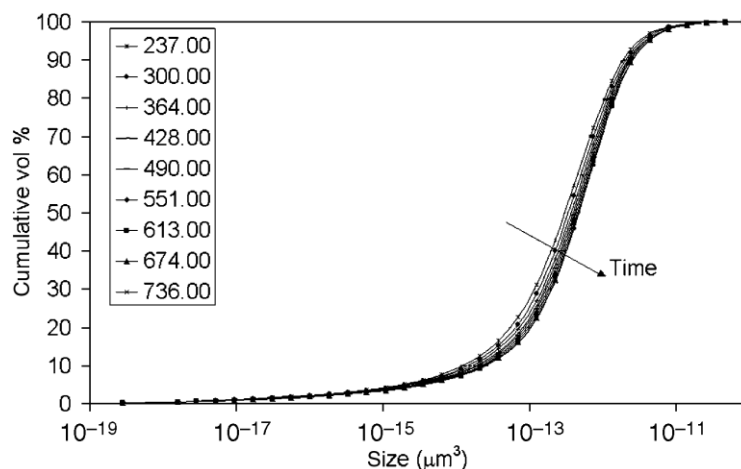

Figure 3 Time evolution of the cumulative distribution for the flocculation experiment at a shear intensity of $G = 19.4$ s^{-1} starting from a deflocculated sludge through increased shear intensity
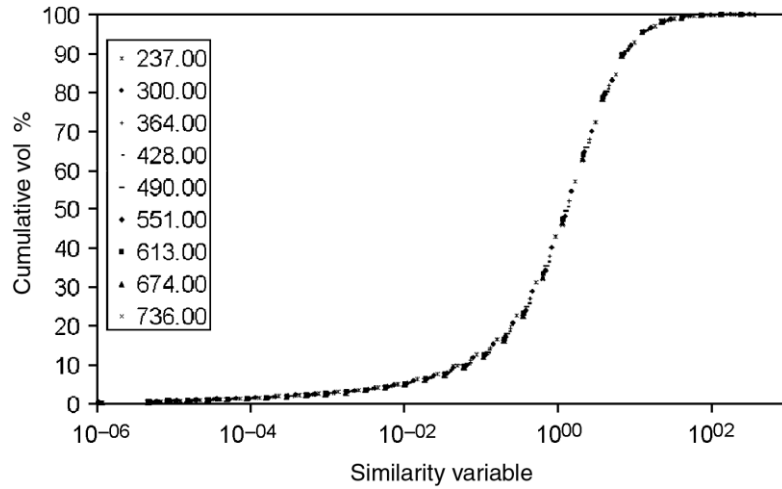
Figure 4 Cumulative distributions versus the similarity variable η for $k = -0.35$

varied from 2-4. It was concluded that $nb = 4$ was the best choice (results not shown). However, it might not be necessary to pursue such high accuracy of the approximation. The danger of using the sum of squared errors (SSE) as only criterion to base the choice on might be that one starts to fit experimental noise, which is not in our interest. Regularisation (Equation 8) did not affect coefficient values for $\lambda_{reg} < 1$. From the inverse solution, the vector a representing the weighting factors of the basis function expansion was obtained. It could then be used to reconstruct the aggregation kernel (results not shown).

Finally, the quality of the inversion is checked. This can be done by performing a forward simulation, i.e. simulating the model using the obtained kernel structure. In order to do so, the scaled aggregation frequency needs to be converted to the unscaled one. The results for the forward simulation were found to be in best agreement with the data for a regularisation factor of 1. Results are depicted in Figure 5 and show good description for the entire distribution dynamics, including the peak and tails.
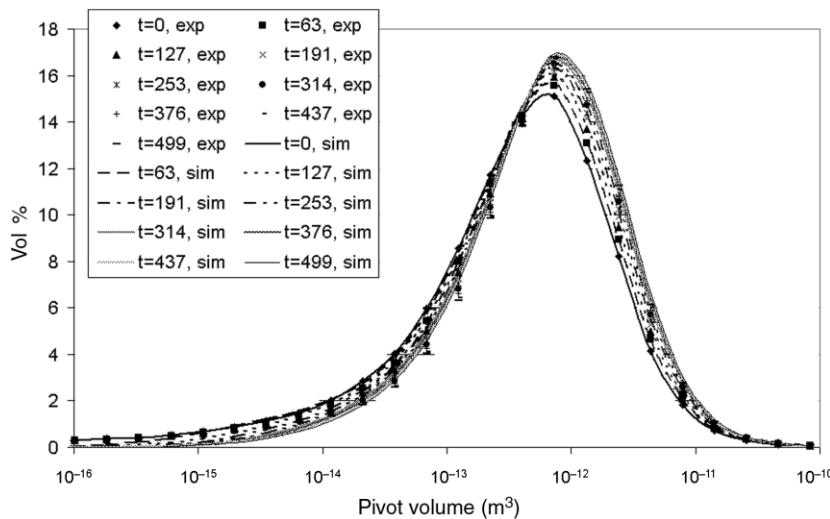

Figure 5 Forward simulation using the unscaled aggregation frequency recovered through inversion using 4 basis functions per axis, $\lambda_{reg} = 1$

Conclusions

A segregated modelling framework was adopted enabling to describe the evolution of distributed properties of populations. In search for an adequate model structure, an inverse solution methodology was successfully applied for the first time for a biological system. Similarity was found for aggregating sludge that was disrupted through excess shear. The latter was not the case for sonicated sludge, proving that the model and, hence, the flocculation process is different and might not be an adequate method to study activated sludge in a real treatment plant. The aggregation kernel was recovered and its quality proven by means of a forward simulation resulting in an adequate description of the distribution dynamics during activated sludge flocculation.

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