Towards mechanistic models for activated sludge flocculation under different conditions based on inverse problems

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ABSTRACT

Experimental data of Ca-induced activated sludge flocculation under different conditions of temperature and dissolved oxygen are investigated in order to model the influence of changing physical and chemical factors. However, current kernel structures for collision frequency and efficiency are unable to describe activated sludge flocculation data. Therefore, an earlier developed methodology based on an inverse problem is applied, yielding empirical models, to find out how flocculation is affected by these different environmental conditions. This contribution shows the useful application of inverse problems to improve the understanding of complex aggregation mechanisms.

Key words | calcium, flocculation, inverse problem, population balance

INTRODUCTION

Secondary settling in conventional activated sludge (CAS) systems and membrane filtration in membrane bioreactor (MBR) applications suffer respectively from poor settling and fouling which are highly related to the flocculation state of the sludge. Small and low density flocs will settle poorly. Similarly, they will lead to less permeable cake layers due to a higher packing density compared with large sludge flocs and, hence, lower void fractions and lower permeability. Therefore, research to improve the knowledge of the flocculation process still deserves ample attention as understanding the process is the key to optimization and control.

The activated sludge flocculation process can be described by means of a population balance model (PBM). This modelling framework allows the investigation of the entire distribution of floc sizes during flocculation. The dynamics of the number concentration of the flocs ($N_i$) in different size classes (i) can be written as (Hounslow et al. 1988):

$$\frac{dN_i}{dt} = \text{Birth}_{\text{aggregation}} - \text{Death}_{\text{aggregation}}$$

$$+ \text{Birth}_{\text{breakage}} - \text{Death}_{\text{breakage}}$$

The flocculation rate in the PBM depends on the collision frequency ($\beta$) (governed by transport) and the efficiency ($\alpha$) of collisions (i.e. the probability that a collision is successful). The latter rate expressions are called kernels and are embedded in the PBM. Several model structures for $\alpha$ and $\beta$ have been presented in literature based on physical phenomena and conceptual models. However, the application of these models to experimental data showed that these structures are not able to describe the flocculation dynamics of activated sludge (Nopens 2005). The reason for this finding is probably that important processes are missing from the model. Determining the exact cause using a forward simulation (i.e. numerically solving the equation above to obtain $N_i(t)$) is difficult.

An alternative approach is to solve the inverse problem based on dynamic experimental data, yielding empirical models (Wright & Ramkrishna 1992; Nopens et al. 2007). These models have a drawback in that they do not have a physical meaning and, therefore, cannot be directly used for system analysis or process optimization. They can, however, provide valuable insights into the true dynamics of the flocculation mechanism and allow evaluating where conventional kernel structures are failing.

The knowledge related to the flocculation process can be improved only by a careful analysis of the physico-chemical factors affecting it. Both environmental conditions and process parameters will influence the floc properties.
A deeper, thorough investigation of these effects will eventually allow understanding the complex processes related to the floc formation. This could improve the activated sludge performance with regard to separation. In this context, the present research will deal with a model-based analysis of short-term effects of temperature and dissolved oxygen (DO) concentration on the structure of the aggregation process. Evidence is provided in literature that these factors have a significant influence on the flocculation process of activated sludge (Sürüçü & Çetin 1989; Wilén 1999; Liao et al. 2001).

Temperature has a complex influence on the performance of the activated sludge flocculation process. The temperature effect is related to both transport and attachment of the flocs. When temperature increases, the mixed liquor viscosity decreases, improving its mixing behaviour and enhancing Brownian motion of small particles. The latter is, however, only important for relatively small particles (<1 μm) and is, therefore, often not considered to be important in activated sludge flocculation compared with the magnitude of other transport mechanisms. Next to the viscosity effect, the separation properties of the activated sludge are also influenced by temperature due to a change in the function and charge of the EPS (Sürüçü & Çetin 1989; Wilén 1999). However, very little is known about the direct impact of the short-term temperature effect on floc size dynamics.

Li & Ganczarczyk (1993) found that the availability of DO is one of the most significant factors which influence the size distribution of activated sludge flocs. Wilén & Balmér (1999) performed an intensive research study regarding the effect of the DO concentration on flocculation. Long-term effects were mostly governed by growth of filamentous bacteria. From a short-term perspective, oxygen limitation had a pronounced effect on the amount of small flocs in the supernatant after settling and introduced an increase in effluent turbidity, although the size and structure of the larger flocs was not significantly affected.

**MATERIAL AND METHODS**

**Experimental data**

Govoreanu (2004) conducted several experiments under different conditions in which flocculation dynamics were measured by on-line monitoring of particle size distributions (PSD). This was done by recording the number of particles in each of the 44 particle size classes every 30 s (MastersizerS). The workflow of each experiment is shown in Figure 1.

First, a sludge sample was taken from a SBR breeding reactor and transferred into a flocculation vessel (separate setup). Initial conditions in this vessel were set by diluting the sludge to the desired concentration (step 1) and controlling the experimental parameters (e.g. DO, T,...) at reference values (step 2), to which the sludge was given time to acclimatize (t₁). This approach allowed each experiment to start with a similar sludge, acclimatized under identical conditions. PSD were recorded at these initial conditions during t₂ = 10 min (step 3). Subsequently, one or more of the experimental parameters was changed to correspond to the design of the specific experiment (step 4). Once again, the sludge was given time to acclimatize (t₃) after which PSD were measured at the new design point during t₄ = 10 min (step 5). Finally, calcium (Ca) was added to promote flocculation (step 6) in order to study the Ca-induced flocculation dynamics for the new experimental conditions (step 7).

Next to these dynamic experiments, a second set of experiments was performed which consisted of consecutive variations of each studied parameter in the reactor vessel without addition of Ca. These experiments allow comparing the steady state floc size for different experimental conditions. We will refer to these experiments as the steady state experiments.

In this contribution, the effect of two parameters (temperature and DO) on the flocculation dynamics is studied based on the experimental data collected by Govoreanu (2004). The DO concentration limits were set between anaerobic conditions (DO = 0 g/l) and an aerobic level at...
DO = 4 g/l. DO = 2 g/l was chosen as an intermediate reference value. To raise the oxygen in the vessel O₂-gas was supplied through a permeable silicon tube, from where the gas diffuses into the liquid in a bubble-less manner (avoiding additional shear). To decrease the oxygen level, N₂-gas was used in a similar way. Temperature experiments were conducted at resp. 5, 15 and 25 °C.

Inverse problem solution and kernel recovery methodology

The methodology for solving the inverse problem is adopted from Wright & Ramkrishna (1992). Figure 2 provides a schematic representation of the different steps that comprise the solution of an inverse problem and the recovery of a valid kernel. A brief explanation of the different steps is provided below. For a more detailed explanation the reader is referred to Wright & Ramkrishna (1992) and Torfs et al. (2012).

In the first step, a similarity analysis is performed which is essentially a data transformation. The idea is to find a scaling function that can transform the data in such a way to make all the separate PSD collapse into one single distribution (the similarity distribution). Through this process, the time component is eliminated from the system which allows rewriting the PBM in a simplified form. The population balance equation (PBE) as a function of the similarity variable η is given by:

\[ \eta \Phi'(\eta) = \int_0^\infty d\eta' \Phi'(\eta') \int_\eta^{\infty} \frac{d\eta''(\Phi'(\eta'')) (a(\eta', \eta''))}{\eta''} <\alpha> \]

Here, \( \Phi'(\eta) \) is the first derivative of the similarity distribution and \( a(\eta', \eta'')/\langle\alpha\rangle \) is the unknown aggregation frequency function that needs to be recovered. The similarity distribution \( \Phi(\eta) \) can be approximated by an expansion of \( \gamma \)-functions (step 2). The unknown aggregation frequency is the product of collision frequency (\( \beta \)) and collision efficiency (\( \alpha \)) and can be represented as a linear combination of basis functions (Laguerre polynomials).

The inverse problem can then be solved by fitting the right hand side of Equation 1 (which can be computed) to the left hand side (that can be extracted from the experimental data), resulting in an optimal set of coefficients for the linear combination of Laguerre polynomials (step 3). The optimization method is a constrained one where symmetry is enforced. Identifiability was not investigated as such, however it is expected that this factor would not pose problems. After all, the recovered optimal coefficients have no actual physical meaning and the goal of this research is to study the structure of the recovered kernel, which will not be influenced by identifiability in the coefficients. Finally, the recovered kernel can be reconstructed from the linear combination of Laguerre coefficients, using the optimal set of coefficients (step 4). The quality of the recovered kernel can be checked by implementing this kernel in the PBM and performing a forward simulation (step 5).

The inverse problem has already been successfully applied to shear induced (Nopens et al. 2007) and Ca-induced (Torfs et al. 2012) flocculation data. The latter proposes a generic methodology to compare kernels recovered through the inverse problem \( \alpha_{inv}\beta_{inv} \) with kernel expressions from literature (step 6). A schematic overview of this methodology is given in Figure 2.

As the kernel structure recovered from the inverse problem is the product of \( \alpha_{inv} \) and \( \beta_{inv} \), comparison with literature kernels becomes quite challenging because numerous combinations of existing \( \alpha \) and \( \beta \) kernels are possible. The methodology represented in Figure 3 overcomes this problem by assuming a fixed expression for \( \beta \) from literature. The methodology is generic and can thus be applied for any choice of \( \beta \) from literature. However, the validity of the chosen \( \beta \) will determine how well the dynamics of collision frequency and efficiency are separated. If the chosen function for \( \beta \) is not valid, a portion of the transport effect will be incorporated in the collision efficiency resulting in an expression for \( \alpha \) which is not satisfactory (e.g. values larger than 1, shape not in correspondence to experimental findings from literature, etc.). If this is the case, the...
chosen $\beta$ kernel needs to be re-evaluated. The transport kernel used here (Lee et al. 2000), accounts for shear (via $G$) and incorporates fractal dimension of the colliding flocs ($D_f$).

Moreover, for the experiments at different DO concentrations, care was taken to supply O$_2$/N$_2$ gas in a bubbleless manner so as not to cause additional mixing and shear. The effect of temperature on the transport kernels is limited to very small particles ($<1 \mu m$) by influencing their Brownian motion. However, the sludge that was used here had already undergone some degree of flocculation prior to the initialization of the experiments, as a result of which the smallest size classes consisted of particles of approximately 4 $\mu m$. Hence, the effect of temperature and DO concentration on the transport of particles is eliminated.

Determining whether the resulting $\alpha$ is satisfactory can be accomplished by comparing the results with literature findings. First, as $\alpha$ represents the probability of a successful collision, its values should lie between 0 and 1 for all size classes. Second, Ding et al. (2006) found that the collision efficiency decreases with particle size.

### RESULTS AND DISCUSSION

#### Dissolved oxygen

To study the effect of DO on the aggregation dynamics of activated sludge, data were used from experiments at three different DO concentrations, resp. 0, 2 and 4 mg/l. Figure 4 shows the evolution of the mean floc diameter for both the dynamic and steady state experiments. From both experiments it can be observed that altering the DO concentrations results in different mean floc diameters. The latter indicates that the floc size distributions have been influenced by the DO concentration.

To investigate the aggregation dynamics for the entire distribution instead of merely for the mean floc diameter, the inverse problem was solved for each dataset. The performance of the inverse problem solution relies entirely on the information content of the measurements. This finding means that the intervals at which measurements are taken should be sufficiently small in order to capture the dynamics and allow the inverse problem to accurately recover the corresponding kernels. For the first few minutes after Ca addition, the inverse problem could not be solved as.
the aggregation dynamics are too fast in comparison with the measurement interval. The inverse problems were solved for the time interval as indicated by the black box in Figure 3. In this interval the aggregation dynamics have returned to shear induced aggregation allowing investigation of the DO effect without interaction of Ca.

The quality of the recovered aggregation frequency can be verified by performing a forward simulation. This entails the implementation of the recovered kernel structure into a PBM model. The model should be able to follow the experimental data from which it was extracted. The goodness of fit was evaluated by calculating the Sum of Squared Errors (SSE). This evaluation was done by calculating and summing the squared errors between experimental and predicted volume percentage data for each of the 44 size classes at every measurement time within the indicated black box (Figure 4). Simulation of the data for DO concentrations of 0, 2 and 4 mg/l resulted in SSE values of resp. 92, 23 and 68. For reasons of comparison, the data for 0 mg/l DO were also simulated using the frequency kernel by Lee et al. (2000) and a constant collision efficiency. The value of the collision efficiency was determined through an optimization experiment. The simulation resulted in an SSE of 2,443. These results show that the inverse problem was able to extract a valid kernel structure from the experimental data.

Figure 5 represents the collision efficiencies obtained by dividing the recovered kernels by the literature expression for the collision frequency as shown in Figure 3. To determine whether these structures for \( \alpha \) are satisfactory, both shape and size are evaluated. The recovered structures show high collision efficiencies for small particles and a decreasing efficiency for larger particles indicating that collision between two large particles is less likely to form a new floc than collision between smaller particles. This is consistent with findings from the literature (Ding et al. 2006). Secondly the values of the collision efficiency should lie between 0 and 1. However, for very small particles \( \alpha \) values larger than 1 were obtained suggesting that for these particles transport is underestimated by the collision frequency kernel from literature (Lee et al. 2000).

Comparing the \( \alpha \) kernel structures amongst the different DO concentrations shows that the highest efficiencies are found for the intermediate concentration. Both 0 mg/l DO and 4 mg/l DO exhibit less efficient aggregation dynamics compared with a concentration of 2 mg/l. Anaerobic conditions (0 mg/l) appear to be slightly worse than high DO concentrations (4 mg/l). These results clearly show the importance of the DO concentration with respect to the flocculation behaviour of the sludge. DO is strongly related to activity and production of soluble microbial products (SMP) which act as a glue for the flocs. However, at this stage of the research it is not possible to give an explanation of the observed behaviour, based on only three data sets. Unraveling the exact mechanism behind the influence of

![Figure 5](https://iwaponline.com/wst/article-pdf/65/11/1946/442116/1946.pdf)
DO is outside the scope of this work and needs further investigation involving more detailed data collection.

To investigate whether the influence of the DO concentration is related to the size of the floc, the ratios of the aggregation efficiencies were calculated as shown in Figure 6. Both 0 and 4 mg/l were compared with the optimal concentration of 2 mg/l. For the anaerobic conditions, the collision efficiency is reduced by a factor of about 5–7. The effect of higher DO concentrations is less severe with a collision efficiency that is about four times lower than for the optimal concentration. Furthermore, it can be observed that the impact of a change in DO concentration is more pronounced for the flocculation of larger particles than for smaller flocs. What exactly is the mechanism that drives this rather rapid phenomenon based only on a change in DO is unclear. Given the rapid nature, it is thought to be a physico-chemical mechanism.

**Temperature**

A similar analysis is performed for sludge flocculation at three different temperatures, resp. 5, 15 and 25 °C. The evolutions of the mean floc diameter for both the dynamic and steady state experiments are shown in Figure 7. An increase in floc size can be observed for a decrease in temperature. The observed differences are much more pronounced than was the case for the different DO concentrations.

The black box on Figure 7 shows the time interval for which the kernel was recovered. The aggregation dynamics right after Ca addition could not be captured by the inverse problem methodology because flocculation was occurring too fast.

The recovered kernels were implemented in a PBM in order to verify their quality. Forward simulations resulted in SSE values of 150, 55 and 81 for resp. 5, 15 and 25 °C confirming that the kernels extracted through the inverse problem are able to describe the experimental data.

Dividing the recovered kernels by the collision frequency kernel from literature, results in structures for the collision efficiency as shown in Figure 8. Evaluating the size and shape of the $\alpha$ kernels indicates that although the shapes correspond well to literature findings, values larger than 1 are found for collision of small particles. This finding confirms that the collision frequency equation from literature underestimates the transport in the small size classes.

Comparing the $\alpha$ kernel structures amongst the different temperatures shows a clear impact of temperature on the flocculation dynamics. Lowering the temperature to 5 °C induces a vast increase in collision efficiency whereas a decrease in $\alpha$ was observed at higher temperatures. These
results were investigated further by calculating the ratios of the collision efficiencies at different temperatures as shown in Figure 9. It becomes clear that the impact of temperature varies for different size classes. The aggregation of larger particles increases at lower temperature and, in contrast, smaller particles show an improved collision efficiency at higher temperatures. Also here, it is unclear what the exact mechanism is, but the analysis shows that there clearly is an impact of temperature on sludge flocculation.

However, these results should be interpreted with care. Although several studies revealed larger flocs and better settling at lower temperatures, the exact relation is difficult to quantify as different activated sludge samples respond differently to various temperatures depending on the temperature at which they were acclimatized (Wilén 1999).

**CONCLUSIONS AND PERSPECTIVES**

Flocculation data at different DO concentrations and temperatures were studied by solving their inverse problems. The aggregation kernels recovered through the inverse solution methodology allowed for accurate predictions of the particle size dynamics. The results showed that there was a clear influence of both DO concentration and temperature on the flocculation dynamics of activated sludge. For DO it was found that there exists a clear impact of the DO concentration on the flocculation dynamics. The most efficient aggregation was found at a concentration of 2 mg/l. Both an increase as well as a decrease in the DO concentration resulted in less efficient aggregation dynamics. The influence of the DO concentration was more pronounced for larger than for smaller particles. The data for temperature...
showed that aggregation of larger particles improves when temperature decreases while smaller particles exhibit faster aggregation dynamics at higher temperatures.

The kernels recovered through solving the inverse problem are able to accurately describe activated sludge flocculation dynamics under different conditions. This makes the inverse problem a powerful tool for system analysis and allows valuable insight into aggregation dynamics and the factors influencing flocculation. However, the resulting kernel structures are of a mere empirical nature and are therefore in their current form not applicable for predictive modelling purposes. Translation of the recovered kernels into actual physical expressions, should be the final step of the methodology. However, to develop this final step, more data and a thorough comparison between the recovered kernels and kernel expressions from literature is necessary. This comparison will be the scope of future work.

REFERENCES


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