Summary

The separation of activated sludge and purified wastewater still remains a delicate issue in wastewater treatment. In both the traditional separation by means of gravitation in a secondary settling tank and the more recent membrane technology, it has been accepted that floc size and, hence, activated sludge flocculation plays an important role.

This dissertation deals with the investigation of the applicability of a population balance framework for modelling the activated sludge flocculation process. This analysis consists of several separate issues which should be treated in a logical order.

A first issue is related to the *numerical solution methods for population balance models*. To this end, three discretisation methods (Hounslow algorithm, fixed pivot and moving pivot) were taken from literature and subjected to a detailed simulation study in which several aspects like ease of implementation, accuracy, stability, ... were investigated for three distinct mechanisms (pure aggregation, pure breakage and the combined case) when using different initial conditions and grid densities. It was concluded that the moving pivot was superior in most cases. However, the choice is often a trade-off between accuracy and simulation speed.

In a second stage, a *comprehensive calibration methodology* was developed which deals with several issues related to the fitting of a population balance model to an experimental data set, i.e. the selection of the solution method, data transformation, determination of measurement errors and choice of fitting variable. The former was already investigated in the first part. Data transformations should be avoided, but are sometimes necessary, e.g. to determine the initial particle size distribution for the model. A new method based on the sludge concentration and the densities of water, flocs and solids was presented. Preliminary experiments for the measurement of the floc and solids densities were also conducted, where the former is based on pyknometry and the latter on a linear Percoll gradient. Initial results showed floc densities of 1.02-1.06 $\mathrm{g.ml^{-1}}$, whereas solids densities were found in the range of 1.60 $\mathrm{g.ml^{-1}}$. A method to determine measurement errors in the different size classes of a distribution was presented based on the variance of a multinomial distribution. Finally, the choice of fitting variable was investigated. Volume-based variables were able to capture the dynamics of the volume percentage distribution rather well. However, they completely failed at predicting the dynamics of the number distribution. Fitting on number-based variables resulted in poor predictions of the volume distribution dynamics, whereas a better prediction of the dynamic number distribution was found. The choice should be determined by the goal of the research. In this case, the number of small particles is of interest as it are the small particles that end up in the effluent and determine its quality.

In the previous steps, no attention was paid to finding an adequate *structure of the model*. Two different approaches were used to find one: evaluate existing kernels from literature and extracting the kernel structures from the experimental data.

In the first approach, different relevant kernels for aggregation and breakage were selected from literature. These were combined into four models that were subsequently fitted to the dynamic number distribution using a least squares method. The knowledge-based models incorporated mechanisms like shear-based orthokinetic aggregation, fractal dimension, hydrodynamic and Van der Waals interactions, turbulent breakage. However, none of the investigated models was able to describe the evolution of the number distributions. The best model was found to be composed of a constant collision efficiency, a shear-based orthokinetic aggregation frequency and power law breakage with an exponent of 1/9.

The second approach aims at solving the inverse problem to reconstruct the unknown aggregation and breakage kernels from the experimental data. Prior to applying this technique, a similarity analysis is performed since this can simplify the inversion problem.

In the pure aggregation case, the aggregation data obtained from a sonicated sludge did not exhibit self-similar behaviour. However, aggregation from a sludge that was previously exposed to a high shear environment did exhibit self-similar behaviour. This leads to the important conclusion that sonication is apparently not representative for the dynamic flocculation behaviour of an activated sludge in a full-scale plant. It should, therefore, be avoided in lab-scale studies on activated sludge flocculation.

Since flocculation behaviour was found to be different for the non-sonicated sludge, the previous approach using models from literature was repeated first. However, the optimisation algorithm was found to be sensitive to initial parameter values and also resulted in very high parameter variances. An investigation based on a scenario analysis showed that the objective function consisted of a large plateau and only exhibited a minumum around the origin. This would imply that the dynamic data could best be described by a static model (i.e. no aggregation or breakage) which is not consistent with the observations.

Continuing the second approach, the inverse problem was solved. In the scope of reconstructing the unscaled aggregation frequency, it was found that the kernel was not homogeneous and that an alternative method based on the derivative of the scaling function h(t) was to be used. The reconstructed unscaled aggregation kernels exhibited a different shape depending on the number of basis function that was used in the inversion. The regularisation parameter did not influence the results a lot, except in the case where three basis functions were used.

The quality of the inversion was checked by performing a so-called forward simulation. The case using four basis functions in the inversion yielded the best results and was able to capture the evolution of the complete volume distribution, except for the lower tail. However, in order to obtain this result, an additional factor had to be introduced. The origin of this factor is unclear. It causes the small size classes of the number distribution to be underestimated, whereas the remainder of the distribution is captured quite well.

The inversion was also investigated for the pure breakage case. Self-similar behaviour was found and resulted in a breakage rate described by a power law with exponent 3.7. The constant and the daughter distribution have to be recovered from the inverse problem. However, the basis functions had to be modified due to a singularity near the origin. This resulted in a solution of the inverse problem that was best for the lowest number of basis functions. The remaining constant of the breakage rate was determined to be 1.5. The daughter distribution could be extracted from the cumulative daughter distribution.

Unfortunately, the forward simulation was impossible to complete due to the high breakage rates that prohibited the solver to converge to a solution. Multiplying by a small factor allowed to perform the simulation, but resulted in poor predictions of the dynamic volume distribution.

Finally, existing *experimental data* and new experimental data from the newly built Flocunit were analysed with focus on the dynamic behaviour of the distributions instead of a summarising parameter such as the weighted average diameter. No universal relationship between the most abundant floc size and the Kolmogorov microscale was found for the different experiments. For the literature data, the Kolmogorov scale, calculated based on the assumption that the maximum energy dissipation rate near the impeller is tenfold of the average energy dissipation rate, was in good agreement with the most abundant floc size. The latter was not found for the Flocunit experiments. This is possibly due to differences in reactor configuration (larger volume, different impeller) and floc strength. On the other hand, the analysis revealed that in the dynamic part of aggregation/breakage experiments the assumption of neglecting the other mechanism (breakage/aggregation respectively) was valid. This assumption is an important requirement for conducting the similarity analysis.

The new experimental data investigated the influence of 5 possible influencing physico-chemical factors by means of a fractional factorial design, i.e. temperature, shear rate, dissolved oxygen, sludge concentration and Ca-addition. Shear has an important effect on the activated sludge flocculation. High shear destroys flocs and shifts distributions toward smaller sizes. Low shear allows larger flocs to be present in the system. Flocs are not disturbed when temperature is lowered. However, some immediate flocculation occurs when temperature is increased. Applying anaerobic conditions showed an immediate increase in floc size. At low sludge concentration, a slow deflocculation can be observed, caused by the smaller collision probability. Ca-addition results in flocculation.