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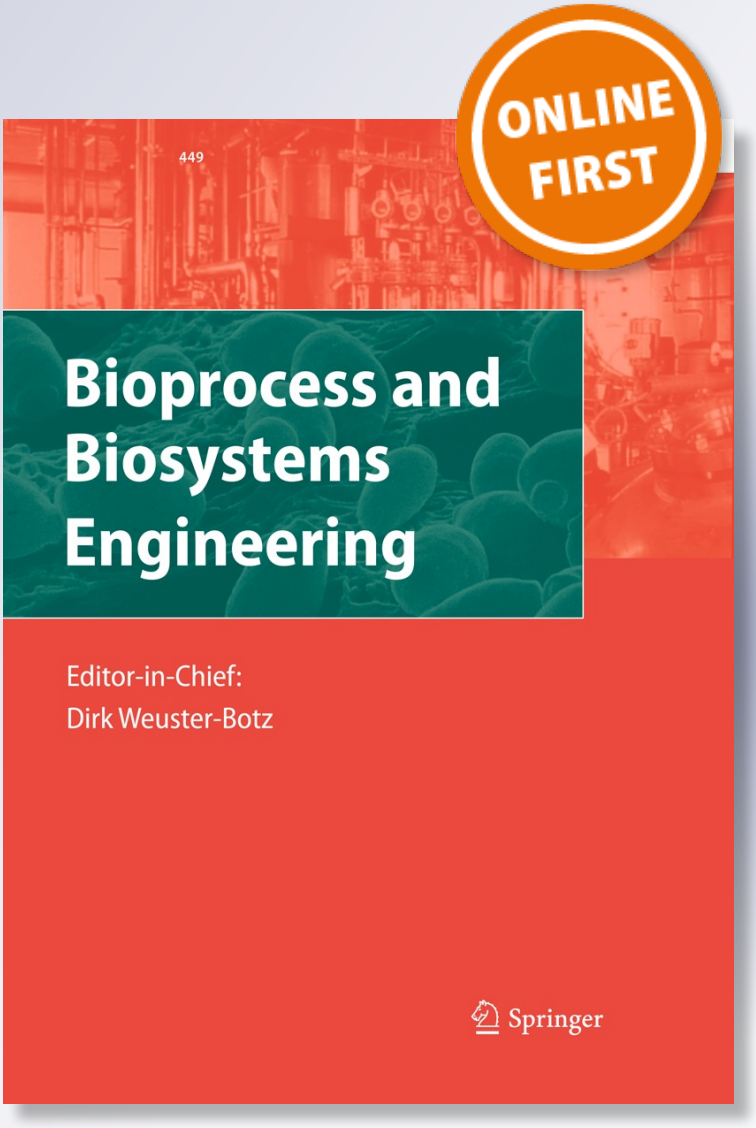

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ORIGINAL PAPER

Mathematical modeling and analysis of the flocculation process in chambers in series

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Abstract In this study, the flocculation process in continuous systems with chambers in series was analyzed using the classical kinetic model of aggregation and break-up proposed by Argaman and Kaufman, which incorporates two main parameters: K_a and K_b. Typical values for these parameters were used, i. e., $K_a = 3.68 \times 10^{-5}$–$1.83 \times 10^{-4}$ and $K_b = 1.83 \times 10^{-7}$–$2.30 \times 10^{-7}$ s^{-1}. The analysis consisted of performing simulations of system behavior under different operating conditions, including variations in the number of chambers used and the utilization of fixed or scaled velocity gradients in the units. The response variable analyzed in all simulations was the total retention time necessary to achieve a given flocculation efficiency, which was determined by means of conventional solution methods of nonlinear algebraic equations, corresponding to the material balances on the system. Values for the number of chambers ranging from 1 to 5, velocity gradients of 20–60 s^{-1} and flocculation efficiencies of 50–90 % were adopted.

Keywords Mathematical modeling · Kinetics · Flocculation · Water and effluent treatment

List of symbols

ε	Total energy dissipated per unit mass of fluid ($L^2\ T^{-2}$)
E	Flocculation efficiency (−)
G	Average velocity gradient (T^{-1})
K_a	Aggregation coefficient (−)
K_b	Break-up coefficient (T^{-1})
β	Coefficient related to floc resistance (−)
m	Number of chambers in series
n	Number of primary particles per unit volume at time t for batch system or at the inlet stream for continuous system (L^{-3})
n_0 and n_m	Number of primary or destabilized particles in the water before flocculation and in the effluent of the mth chamber, respectively (L^{-3})
n_0	Number of primary particles at time $t = 0$ (L^{-3})
n_i and n_{i-1}	Concentrations of primary particles in the output of the ith and $(i - 1)$th flocculation chambers, respectively (L^{-3})
Q	Flow of water ($L^3\ T^{-1}$)
R	Flocculation efficiency expressed by the ratio n_0/n_m (−)
T	Total hydraulic retention time in the set of m chambers in series (T)
V	Volume of the flocculation chamber (equal for all chambers) (L^3)
v	Kinematic viscosity of the fluid ($L^2\ T^{-1}$)

Introduction

The treatment of water and effluents (domestic or industrial) can be performed using physical–chemical processes alone or in combination with biological processes. Flocculation is one of the stages in the physical–chemical treatment process. The goal of this step is to promote the meeting of destabilized particles to promote aggregation under the appropriate conditions for solid/liquid separation.

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The energy supplied to the liquid, either hydraulically by energy dissipation devices or by electromechanical equipment, is the main driving force responsible for the transport of particles. However, interactions resulting from Brownian motion and differential sedimentation occur concurrently. The relative importance of each transport mechanism depends mainly on the sizes of the particles. The break-up of previously formed agglomerates (called flocs), resulting from shear forces, occurs simultaneously with aggregation.

The use of mathematical modeling to describe the kinetics of flocculation aims to estimate the performance of the process considering the phenomena of aggregation and of floc break-up [1]. These phenomena are resultant of several combined mechanisms of transport and attachment that can be generalized by means of apparent coefficients. All models proposed for the kinetic study of the flocculation process are based on batch experiments, and the parameters estimated from the treatment of data obtained in these tests are commonly extrapolated to design continuous systems with one or more flocculation chambers in series. It is known that arrangement in series results in hydrodynamic behavior approaching ideal plug-flow as the number of chambers making up the series increases.

Di Bernardo et al. [1] presented a method for determining the apparent coefficients of aggregation (K_a) and of floc break-up (K_b) using data obtained in tests with batch reactors (jar-tests), which were treated by means of the model proposed by Argaman and Kaufman [2].

One of the most important parameters in the design of flocculation units is the average velocity gradient (G), which is related to variations in the velocity profile in space, incorporating turbulence mechanisms for destabilized particle transport. Camp and Stein [3] proposed the velocity gradient as a significant parameter in turbulent flocculation (Eq. 1).

$$G = \sqrt{\frac{\varepsilon}{v}} \tag{1}$$

where G is the average velocity gradient; ε is the total energy dissipated per unit of fluid mass; and v is the kinematic viscosity of the fluid.

Although there are limitations to the use of the parameter G related to the scale of turbulence (η) and the size of the particles (d_p), Saffman and Turner [4] and Argaman and *K*aufman [2] justified the practical application of G in sanitation when $d_p < \eta$. This criterion has been widely employed and is an important tool for the design and operation of treatment units.

In this study, mathematical modeling was performed along with analysis of the flocculation process in chambers in series to better understand the influences of certain design and operating parameters on the total retention time required for a desired flocculation efficiency.

Mathematical modeling

Coagulation and flocculation units are designed to promote changes in particle size distributions. Therefore, the optimization of their operating parameters should preferably be performed with the objective of obtaining particle size distributions suitable for the separation techniques used downstream (sedimentation, flotation, filtration, and others). The first attempt to mathematically describe the kinetics of particle size changes in aqueous suspensions was reported by Smoluchowski [5], who proposed a model based on six simplifying hypotheses: (i) the efficiency of collisions is 100 %; (ii) the flow of the liquid medium is laminar; (iii) the particles are monodisperse; (iv) there is no break-up of formed flocs; (v) all particles are spherical and remain so after collisions; and (vi) collisions occur only between two particles.

Nearly three decades later, Camp and Stein [3] incorporated the concept of the average velocity gradient (G) and the effects of differential sedimentation into the Smoluchowski equation [5], but the authors still assumed the six basic hypotheses presented earlier as valid.

In more recent studies, other phenomena, such as hydrodynamic interactions, heterodispersion, particle shape and others have been investigated, and more elaborate models have been proposed [6–9].

However, with respect to the kinetic study of the flocculation process, the model most widely employed is that proposed by Argaman and Kaufman [2], in which aggregation and break-up are the main phenomena considered. The aggregation of destabilized particles as a result of various associated phenomena is represented in this model by the parameter K_a, and the break-up of previously formed flocs is represented by the parameter K_b. Equation 2 depicts the rate of change over time for the number of primary particles in a discontinuous flocculator:

$$\frac{dn}{dt} = K_b n_0 (G)^{\beta} - K_a n G \tag{2}$$

where n is the number of primary particles per unit volume at time t; K_b is the break-up constant; n_0 is the number of primary particles at time $t = 0$; β is the coefficient related to the resistance of the flocs; and K_a is the aggregation coefficient. For $d_p < \eta$ (microscale of turbulence), $\beta = 2$ [1].

Elaborating a mass balance and considering the kinetics of aggregation and break-up for units composed of m chambers operated in series with equal average velocity gradients (G), Argaman and Kaufman [2] obtained the following equation:

$$\frac{n_0}{n_m} = \frac{\left(1 + K_a G \frac{T}{m}\right)^m}{1 + K_b G^2 \frac{T}{m} \sum_{i=0}^{m-1} \left(1 + K_a G \frac{T}{m}\right)^i} \tag{3}$$

where T is the total hydraulic retention time in the set of m complete mixing chambers in series and n_0 and n_m corresponding to the concentrations of primary or destabilized particles in untreated water (before flocculation) and in the effluent of the mth chamber, respectively.

In the case of flocculation chambers operated in series with different G values, the mass balance at steady state in the ith flocculation chamber takes the form:

$$Qn_{i-1} - Qn_i - K_{a_i} n_i G_i V + K_{b_i} n_0 G_i^2 V = 0 \quad (4)$$

where Q is the flow of water; n_{i-1} and n_i are the concentrations of primary particles in the $(i-1)$th and ith chamber, respectively; and V is the volume of the flocculation chamber (equal for all chambers). The notations K_{a_i} and K_{b_i} indicate that these parameters depend on the value of the average velocity gradient (G_i) in each chamber.

The average hydraulic retention time (t) in each unit, assuming ideal hydrodynamic behavior (complete mixture), can be obtained through the ratio V/Q and the relation $t{\cdot}m$, which expresses the value corresponding to the total hydraulic retention time (T) in m chambers in series. Thus

$$n_{i-1} - n_i - K_{a_i} n_i G_i t + K_{b_i} n_0 G_i^2 t = 0 \quad (5)$$

$$\frac{n_{i-1}}{n_i} - 1 - K_{a_i} G_i t + K_{b_i} \frac{n_0}{n_i} G_i^2 t = 0 \quad (6)$$

$$\frac{n_{i-1}}{n_i} - 1 - K_{a_i} G_i t + K_{b_i} \frac{n_0}{n_i} \frac{n_{i-1}}{n_{i-1}} G_i^2 t = 0 \quad (7)$$

$$\left(1 + \frac{n_0}{n_{i-1}} K_{b_i} G_i^2 t\right) \frac{n_{i-1}}{n_i} = 1 + K_{a_i} G_i t \quad (8)$$

Therefore, Eq. (9) can be written for the values of G varying in the m chambers in series, as proposed by Argaman and Kaufman [2]:

$$\frac{n_{i-1}}{n_i} = \frac{1 + K_{a_i} G_i \frac{T}{m}}{1 + \frac{n_0}{n_{i-1}} K_{b_i} G_i^2 \frac{T}{m}} \quad (9)$$

Di Bernardo et al. [1] reported that the kinetic coefficients K_a and K_b should be obtained through tests in continuous reactors, but on the other hand this procedure could be indeed limited by the cost and time required. According to the same author, citing Bratby et al. [10], these coefficients may assume constant values when the number of chambers in series approaches infinity ($m \to \infty$), under plug-flow or in a static (batch) reactor.

To obtain the apparent kinetic coefficients (K_a and K_b), the proposal of Bratby et al. [10] uses settling times of such length that the remaining particles were present only as primary particles. In this context, Pádua [11] and Di Bernardo [1] challenged the validity of the extrapolation suggested by Bratby et al. [10] and recommended that sedimentation times practiced in water treatment plants be employed in obtaining the aggregation and break-up coefficients, K_a and K_b, respectively.

Thus, it is evident that the ratio n_0/n_m depends on the solid/liquid separation technology employed after flocculation, where sedimentation is commonly employed as a separation operation. In the specific case of this study, sedimentation was considered using the kinetic parameters presented by Di Bernardo et al. [1], which estimated K_a and K_b from experimental data obtained in batch tests with a sedimentation rate of 2.5 cm min^{-1}.

It should be noted that other solid/liquid separation technologies can be employed, such as dissolved air flotation (DAF) and different forms of direct filtration, and that in such cases, it is essential to assess the method in which the apparent kinetic coefficients are obtained.

Methods

For the simulations performed, the values of G and kinetic coefficients reported by Di Bernardo et al. [1], presented in Table 1, were adopted. The coefficients estimation was made using experimental data obtained in batch flocculation tests followed by sedimentation at velocities of 2.5 and 5.0 cm min^{-1}. The coefficients concerning to sedimentation velocity of 2.5 cm min^{-1} were chosen based on typical reported values for full-scale sedimentation units. According to Di Bernardo et al. [1], the studied water from which the K_a and K_b coefficients were estimated, had the following characteristics: pH = 7.7 ± 0.2; turbidity = 27 ± 1 NTU; apparent color = 210 ± 10 Hu; true color = 20 ± 5 Hu; alkalinity = 23 ± 1 mg $CaCO_3$ L^{-1}; conductivity = 46.0 ± 0.5 μS cm^{-1}; hardness = 13 ± 1 mg $CaCO_3$ L^{-1}.

Systems were simulated with the number of chambers in series (m) ranging from 1 to 5, i.e., $m = 1, \ldots, 5$, with fixed or variable velocity gradients in each unit.

When the units were operated with the same velocity gradients, for each value of G, T values were obtained for the different flocculation efficiency (E) values expressed indirectly by the ratio $R = n_0/n_m$, where $E = 1 - (1/R)$. The values of T were obtained using the Newton–

Table 1 Values for K_a and K_b obtained by Di Bernardo et al. [1] for a sedimentation velocity of 2.5 cm min^{-1}

G (s^{-1})	K_a (−)	K_b (s^{-1})
20	1.83×10^{-4}	1.83×10^{-7}
30	9.40×10^{-5}	1.91×10^{-7}
40	6.97×10^{-5}	2.33×10^{-7}
50	7.93×10^{-5}	2.42×10^{-7}
60	3.68×10^{-5}	2.30×10^{-7}

Raphson's method [12] to determine the root of Eq. (10) obtained from Eq. (3):

$$f(T) = R\left[1 + K_b G^2 \frac{T}{m}\sum_{i=0}^{m-1}\left(1 + K_a G\frac{T}{m}\right)^i\right] - \left(1 + K_a G\frac{T}{m}\right)^m = 0 \quad (10)$$

When the flocculation chambers were operated with distinct velocity gradients, the calculation procedure consisted of the following steps:

1. Enter the values m, $R_{specified.}$, G_i, K_{a_i} and K_{b_i};
2. Make an initial estimate for T;
3. Calculate the n_{i-1}/n_i ratio in each chamber using Eq. (9);
4. Calculate the productory $\prod_{i=1}^{m}\left(\frac{n_{i-1}}{n_i}\right)$, denoting the result as $R_{calculated}$;
5. Determine the root of the equation $f(T) = R_{specified} - R_{calculated} = 0$ using the Newton–Raphson's method.

In both cases, programs were written in the FORTRAN language for determining the values of T corresponding to the various simulated conditions. Values of G from 20 to 60 s^{-1} and of R from 2 to 10, corresponding to E values from 0.5 to 0.9, were used.

To confirm the results provided by the Newton–Raphson's method, the Wegstein's method [12], another method of solving nonlinear algebraic equations, was used. Unlike the Newton–Raphson's method, the Wegstein's method does not require the calculation of the function derivative over the iterations.

Results and discussions

Both methods of solving the nonlinear algebraic equations yielded the same results for the total time of flocculation (T).

Tables 2, 3 and 4 show the calculated values of total flocculation (T) for different values of the operating parameter G (G = 20, 30, 40, 50, 60 s^{-1}) and the parameter R (R = 2, 3, 10) as examples.

Table 2 Total flocculation time (T) as a function of both the number of chambers in series (m = 1, …, 5) and the velocity gradient for 50 % efficiency of flocculation (R = 2) under separation by sedimentation at a velocity of 2.5 cm min^{-1}

G (s^{-1})	R	T (min) $m = 1$	T (min) $m = 2$	T (min) $m = 3$	T (min) $m = 4$	T (min) $m = 5$
20	2	4.74	3.91	3.67	3.56	3.49
30	2	6.73	5.47	5.11	4.95	4.85
40	2	8.16	6.43	5.96	5.74	5.62
50	2	6.05	4.72	4.36	4.20	4.10
60	2	30.19	18.66	16.08	14.96	14.33

Table 3 Total flocculation time (T) as a function of both the number of chambers in series (m = 1, …, 5) and the velocity gradient for 67 % efficiency of flocculation (R = 3) under separation by sedimentation at a velocity of 2.5 cm min^{-1}

G (s^{-1})	R	T (min) $m = 1$	T (min) $m = 2$	T (min) $m = 3$	T (min) $m = 4$	T (min) $m = 5$
20	3	9.69	7.00	6.32	6.01	5.83
30	3	14.47	10.13	9.05	8.57	8.30
40	3	19.96	12.95	11.32	10.60	10.20
50	3	15.50	9.80	8.50	7.93	7.61
60	3	−120.77	nc	−78.49	nc	−102.61

nc no convergence

Table 4 Total flocculation time (T) as a function of both the number of chambers in series (m = 1, …, 5) and the velocity gradient for 90 % efficiency of flocculation (R = 10) under separation by sedimentation at a velocity of 2.5 cm min^{-1}

G (s^{-1})	R	T (min) $m = 1$	T (min) $m = 2$	T (min) $m = 3$	T (min) $m = 4$	T (min) $m = 5$
20	10	51.23	22.77	17.83	15.86	14.81
30	10	136.24	46.15	33.45	28.71	26.27
40	10	−159.57	nc	−70.85	nc	−87.10
50	10	−71.94	nc	−44.46	nc	nc
60	10	−24.70	nc	−52.42	nc	−82.22

nc no convergence

For a given value of G, the use of chambers in series reduced the total flocculation time (T). Conversely, increasing the value of G increased the value of T for all numbers of flocculation chambers in series (m = 1, …, 5).

Some of the sets of conditions shown in Table 3 resulted in negative values of T or no convergence of the method. The same behavior was observed for values of G above 30 s^{-1} and a flocculation efficiency of 90 % (R = 10) (Table 4). The negative T values may be explained by the nonlinearity of Eq. 10, which may exhibit multiple roots positive and/or negatives, or, otherwise, may not have real roots. In such particular conditions, several initial guesses inside a wide range of T values were tested and all of them converged to the same results. Although of no physical significance, such results can be interpreted as being due to the strong influence that the break-up phenomenon has on flocculation kinetics under relatively high fixed gradient values in all chambers, not being possible attain the desired flocculation efficiency. As an extrapolation, to achieve a high separation efficiency when operating with a relatively high fixed gradient, the residence time in the system should

be minimal, approaching zero, whereas the efficiency approaches to 100 % ($R \rightarrow \infty$).

Figures 1, 2, 3, 4 and 5 present the results of simulations performed for different values of the average velocity gradient (G), efficiencies expressed directly (as E) and indirectly (as R), chambers number (m) and total flocculation times (T) in minutes.

It is evident that the arrangement of chambers in series is beneficial to the process because shorter flocculation times were necessary to achieve the same efficiencies when increasing the number of units. This is advantageous because it results in smaller chamber volumes for the same flow of water treated. It can also be verified that increasing the value of G causes a decrease in the efficiency of the process, independent of the number of chambers in series. Operation with higher values of G requires impractical

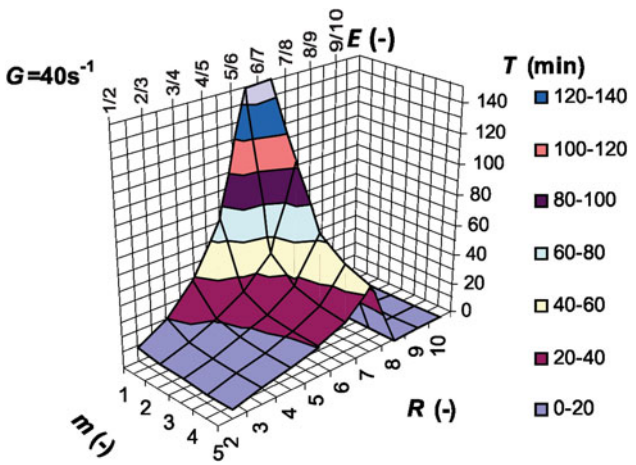

Fig. 3 Simulation of the total detention time (T) in chambers in series ($m = 1, \ldots, 5$) for several flocculation efficiencies (expressed as E or R) and for average velocity gradient (G) of 40 s^{-1}

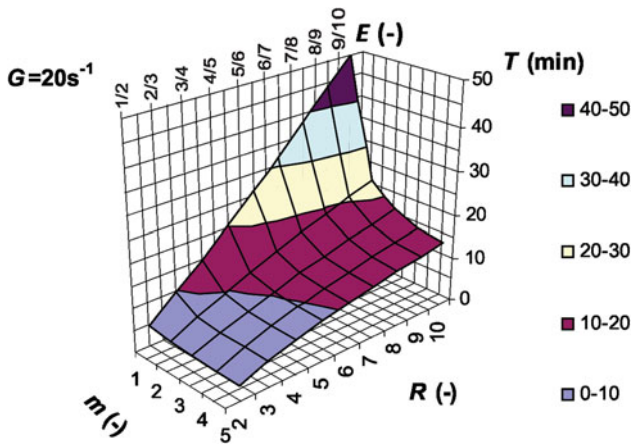

Fig. 1 Simulation of the total detention time (T) in chambers in series ($m = 1, \ldots, 5$) for several flocculation efficiencies (expressed as E or R) and for average velocity gradient (G) of 20 s^{-1}

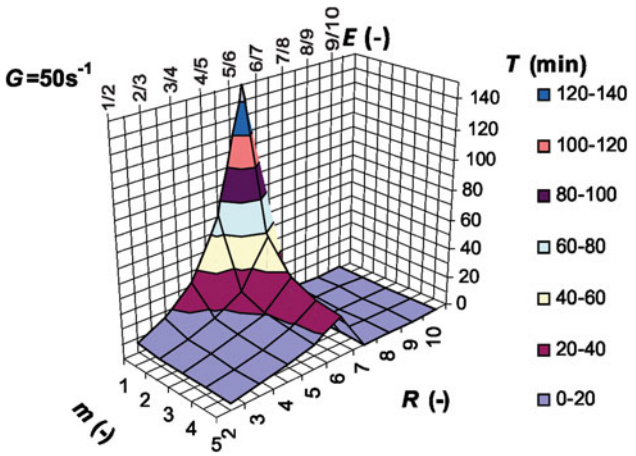

Fig. 4 Simulation of the total detention time (T) in chambers in series ($m = 1, \ldots, 5$) for several flocculation efficiencies (expressed as E or R) and for average velocity gradient (G) of 50 s^{-1}

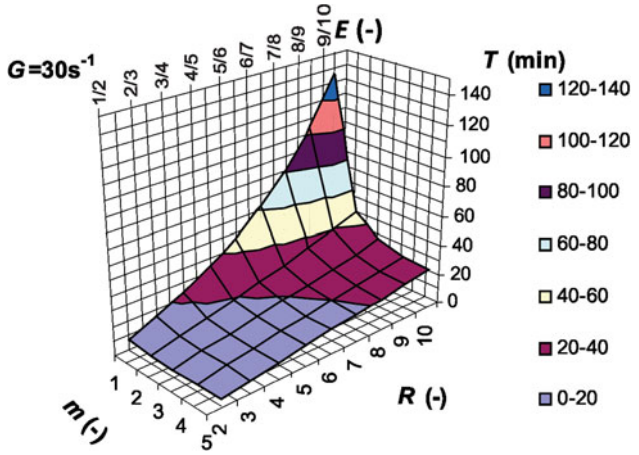

Fig. 2 Simulation of the total detention time (T) in chambers in series ($m = 1, \ldots, 5$) for several flocculation efficiencies (expressed as E or R) and for average velocity gradient (G) of 30 s^{-1}

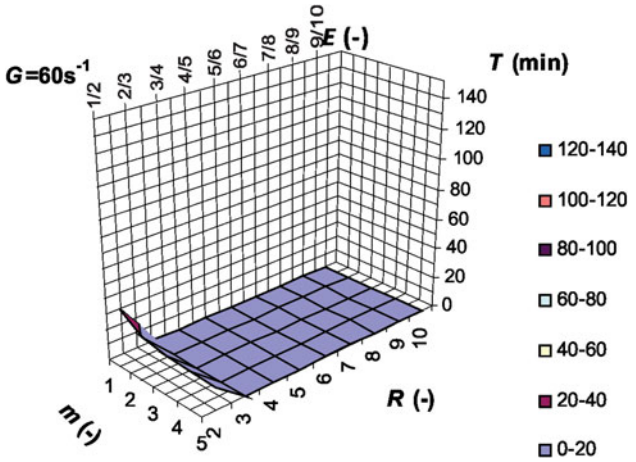

Fig. 5 Simulation of the flocculation process in chambers in series ($m = 1, \ldots, 5$), maintaining the average velocity gradient (G) at 60 s^{-1}

Table 5 Values required for the total flocculation time (T) for chambers in series with a scaled velocity gradient for a flocculation efficiency of 90 % ($R = 10$) by sedimentation at a velocity of 2.5 cm min^{-1}

m	G (s^{-1})	T (min)
1	30	136.24
2	40/30	56.21
3	60/30/20	30.15
4	60/30/30/20	25.51
5	50/40/20/20/20	16.96

total flocculation times ($T \rightarrow 0$), resulting from the intensification of the effect of the break-up of pre-formed flocs.

Thus, the system using chambers in series is both feasible and beneficial from the point of view of process efficiency. The process is, however, restricted to a limiting G value, above which, depending on the number of units used, it is not possible to achieve the desired efficiency due to the break-up of the flocs.

The scaled decrease in G is presented as an option for minimizing the effect of the break-up of pre-formed flocs. In this sense, different experimental studies have presented results suggesting the benefit of scaling (by decreasing successively the value of velocity gradient) on the quality of treated water, including that of Di Bernardo et al. [13]. However, these benefits depend on the quality of the raw water, the coagulant used, the hydrodynamics characteristics in the tanks, the relative effects of other transport mechanisms and the technology used for water treatment. However, in some cases the results of scaling can be minor.

Considering the operational flexibility that the scaling of G may provide for the flocculation process, this option was investigated by performing simulations with different values of G in the chambers. The simulations were performed for the highest value of R ($R = 10$), for which the operation with the fixed velocity gradient proved to be impractical for certain values of G. The results of these simulations are shown in Table 5, according to the scaling proposed by Di Bernardo et al. [1].

In fact, the use of scaled velocity gradients in the m chambers in series allows high efficiencies and permits operation with higher values of G.

Conclusions

The kinetic model of the aggregation and break-up of particles proposed by Argaman and Kaufman allowed the analysis of design and operating parameters for continuous flocculation systems using chambers in series. Application of the model assumes knowledge of the kinetic coefficients of aggregation and break-up, which may be obtained experimentally by trials in static reactors, as proposed in the literature. These parameters are apparent coefficients because they incorporate different transport and attachment phenomena. Thus, the values of the kinetic coefficients depend on the average velocity gradient in the flocculation chambers.

The flocculation efficiency in the continuous systems depends on the number of chambers in series and the velocity gradient applied in these chambers, which can be fixed or gradually decreased.

The use of a single flocculation chamber restricts the velocity gradient, or similarly the system efficiency, requiring longer retention periods and higher capacity chambers for the same flow rate.

The arrangement of chambers in series with fixed gradients permits the reduction of the total flocculation time up to a determined efficiency limit. However, for higher velocity gradients, it is not possible to operate the system with high efficiencies due to the increased break-up of the flocs. A scaled decrease of G permits the operation of the system with higher values of G and elevated flocculation efficiencies under separation by sedimentation at conventional velocities.

The combination of physico-chemical and biological processes for the treatment of wastewater is an increasingly common practice due to the benefits that it may provide. Despite the differing characteristics of the particulate material present in each wastewater, the process of solid–liquid separation requires coagulation and flocculation under conditions suitable for the subsequent separation technology. In this context, the present study presented data on the influence of operational and design parameters on the performance of the flocculation process in continuous systems with chambers in series, with the objective of separating the particulate impurities using a sedimentation technique. However, it is important to highlight that the main aim was to analyze the flocculation process on its fundamentals by seeking general conclusions. Therefore, the operational conditions, which lead to the best results, must be determined for each specific case.

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