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Mathematical modeling of water flocculation process with high turbidity: studies and comparative analysis between methods and models

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ABSTRACT

The quality of water used by a population is directly proportional to the efficiency of its treatment. Mathematical modeling appears in this context as a tool for optimizing processes in order to make them more efficient, economical and sustainable. This work analyzed and compared the effectiveness of phenomenological mathematical models used in flocculation kinetics in water treatment. The results were obtained by comparing the Argaman and Kaufman Model with the Bratby Method; the Aggregation and Rupture Equation Method - MEAR; and the Method of the First Partial Derivative in Relation to the Velocity Gradient in Flocculation - MPDPG and a model that includes a new term (K_C) that contemplates a supposed process of irreversibility of floc. The mathematical modeling was validated and compared with experimental data. The coefficients of the models and methods were obtained using the Excel® solver® tool using spreadsheets from the same application. It was possible to identify that the Bratby Method, which obtained an average deviation of the order of 30%, was the least efficient, while the MEAR and MPDPG Methods, which obtained about 18% of average deviation and the Kc Model with a deviation of the order of 19%, proved to be efficient in describing the experimental data used.

Keywords: flocculation, mathematical modeling, water treatments.

Modelagem matemática do processo de floculação de águas com turbidez elevada: estudos e análise comparativa entre métodos e modelos

RESUMO

A qualidade da água utilizada por uma população é diretamente proporcional à eficiência de seu tratamento. A modelagem matemática surge nesse contexto como uma ferramenta para otimização dos processos com o intuito de torná-los mais eficientes, econômicos e sustentáveis. Este trabalho teve por objetivo analisar e comparar a eficácia dos modelos matemáticos fenomenológicos utilizados na cinética de floculação no tratamento de água. Os resultados foram obtidos por meio de uma comparação do modelo de Argaman e Kaufman com os métodos de Bratby; o Método da Equação de Agregação e Ruptura - MEAR; e o Método da

Primeira Derivada Parcial em Relação ao Gradiente de Velocidade na Floculação - MPDPG e um modelo que inclui um novo termo que contempla um suposto processo de irreversibilidade dos flocos (K_C). A modelação matemática foi validada e comparada com dados experimentais. Os coeficientes dos modelos e métodos foram obtidos utilizando a ferramenta solver® do Excel® com o uso de planilhas eletrônicas do mesmo aplicativo. Foi possível identificar que o método de Bratby, que obteve um desvio médio da ordem de 30%, mostrou-se o de menor eficácia, ao passo os métodos MEAR e MPDPG, que obtiveram cerca de 18% de desvio médio e o modelo K_C com um desvio da ordem de 19%, mostraram-se eficientes em descrever os dados experimentais utilizados.

Palavras-chave: floculação, modelagem matemática, tratamento de água.

1. INTRODUCTION

Mathematical modeling transforms real-world problems into mathematical processes in order to seek concrete solutions (Bertone *et al.*, 2014; De Lima *et. al.*, 2022). It can be used in flocculation systems for water treatment.

According to Di Bernardo *et al.*, (2017), mathematical modeling in the flocculation process aims to evaluate the performance of flocculation, through the phenomena of aggregation and rupture.

In Water Treatment Plants (WTP), flocculation corresponds to a step in which conditions are met to provide contact and aggregation of previously coagulated particles, facilitating their removal by sedimentation, flotation or rapid filtration (Bratby 2016; Lopes *et al.*, 2020; Li *et al.*, 2021; Moruzzi and Oliveira, 2020; Moruzzi *et al.*, 2022). The efficiency of the flocculation unit depends on the performance of the rapid mixing unit, and is affected by the following factors: coagulant type, coagulant pH, water temperature, concentration and age of the coagulant solution, rapid mixing time and rate of change of color, types and form of coagulant and quality of raw water (Di Bernardo *et al.*, 2017; Russo *et al.*, 2020).

Argaman and Kaufman in 1970 analyzed a mathematical model that explains the kinetics of collisions between particles during flocculation. It combines aggregation and rupture coefficients (K_A and K_B, respectively) whose values are determined through tests in pilot-scale continuous flow reactors. The experiments require costly equipment, so in 1981 Bratby, in the search for a low-cost experiment, adapted the method in order to improve the values of the average flocculation velocity gradient in units with continuous flow based on tests carried out in static reactors with long settling time.

Di Bernardo *et al.* (2005), emphasize studies carried out by Brito (1998), of methods to determine K_A and K_B, based on turbidity data and the number of remaining primary particles, considering the sedimentation velocity. The proposed methods were the aggregation and rupture equation – MEAR (Modification of the Bratby method of 1981) and First Partial Derivative in Relation to the Flocculation Velocity Gradient (MPDPG).

Proposing an alternative method of flocculation kinetics, Argaman and Kaufman (1970), and Marques and Ferreira Filho (2016; 2022), included a third component in the modeling resulting in three kinetic constants that are named, K_A (Aggregation Constant), K_B (Rupture Constant) and Kc (Permanent Rupture Constant); these new terms would determine what they called the “irreversible floc breaking process”.

In the flocculation process, aggregation and rupture occur simultaneously, and these effects are promoted by agitation. Thus, an increase in agitation with an increase in the average velocity gradient with the flocs already formed occurs in a few seconds, with an increase in shear forces and their partial or total decrease; but if you return to the initial stage of agitation, there will be the possibility of regrowth of the flakes or re-flocculation (Voltan, 2007; Ali and Chassagne, 2022). According to Santos *et al.* (2012), the size of the regrown flakes is limited. On the other

hand, Marques and Ferreira Filho (2017) say that after the flake breaks there is an irreversibility. This work therefore aims to compare the methods and mathematical models used by these authors.

The main contribution of this work lies in the simulation of results in mathematical models and methods for flocculation in water treatment and their comparison. The models used in this research were the mechanistic models in the area of flocculation in water treatment; obviously there are other methods and models (Oliveira and Donadel, 2019; Moruzzi *et al.*, 2022; Garcia-Gil *et al.*,2022). Most are statistical/probabilistic models and not based on phenomenology (Al-Saati *et al.*, 2019; Hernandez-Crespo *et al.*, 2022; Ezemagu *et al.*, 2020; Okey-Onyesolu *et al.*, 2022). Phenomenological models can be useful tools in the design of water treatment plants.

1.1.Mathematical modeling

The use of mathematical modeling related to the kinetics of flocculation aims to estimate its performance considering the phenomena of aggregation and rupture. The correct understanding of flocculation mechanisms depends on the study of flocculation kinetics, whose efficiency is linked to several parameters, such as floc sedimentation speed, coagulant dosage, velocity gradient and concentration of primary particles, among others (Hespanhol and Ferreira Filho, 2016; Castamann *et al*., 2022).

The velocity gradient is considered of paramount importance in the design of flocculation units and is related to the variation of the velocity profile in space, including turbulence mechanisms for the transport of destabilized particles. The models proposed for the study of the kinetics of the flocculation process, are mostly based on experiments carried out in batch mode. After these tests, the results obtained are often extrapolated to the projects of continuous systems with quantities of one or more flocculation chambers in series (Moruzzi and Oliveira, 2010).

1.2. Aggregation and disruption

During flocculation, the kinetics of the encounters between the particles promotes two effects simultaneously: aggregation and rupture (Moruzzi and Silva, 2018). Aggregation is the result of the encounter of chemically destabilized particles, by the action of the coagulant, where its agitation promotes conjoining with each other, forming the flakes (Rau *et al.*, 2018; Pawignya *et al.*, 2019). Breakage is the breakage of flocs by shearing forces, which can occur over a long time of flocculation or by intense agitation (Voltan, 2007; Seneda *et al.*, 2021).

According to Oliveira and Teixeira (2014), high velocities can generate the formation of velocity gradients, which, before their removal, allow the breakage and fragmentation of the flocs. Thus, according to the authors (Santos *et al.*, 2014; Voltan, 2007), when returning to the initial conditions of agitation the flakes grow again; this effect is called “refloculation”. In the aforementioned studies, the effects of rupture and reflocculation on floc size in static reactors were verified, and that reflocculation depends on the stirring time and the rupture velocity gradient (Oliveira and Teixeira, 2014; Santos, *et al.*, 2012).

Two mechanisms are responsible for the breakdown: surface erosion of primary particles present in the flocs and floc fragmentation. The first is caused by the drag of water acting through the shear forces on the surface of the flakes, with turbulent flow, while the second occurs as a function of dynamic pressure differences on opposite sides of the flakes, deforming and fragmenting them (Di Bernardo *et al.*, 2005).

Aggregation and disruption (disaggregation) during flocculation results in the formation of a stable floc size defined by Equation 1, (Di Bernardo, 2002).

$$d_{fes} = K_{fes}(G_{med})^{kfes} \quad (1)$$

Where, d_{fes} represents the size of the stable flake (cm), k_{fes} the coefficient related to the

strength of the stable flake (cm.s^{kfes}) and k_{fes} (dimensionless coefficient), the coefficient that depends on the way in which the flake rupture occurs and the size of the eddies causing this rupture, while G_{med} is the mean velocity gradient (s^{-1}). When there is erosion of flocs larger than η (Kolmogorov turbulence microscale – [cm]), k_{fes} = 2 is obtained and, for flocs smaller than h, k_{fes} = 1 results. When the predominant action is fragmentation, it has k_{fes} = 0.5 for the two floc size conditions with respect to h. Results of some experiments with k_{fes} = 1 indicate that the maximum floc size is inversely proportional to G_{med} (Di Bernardo, 2017).

According to Di Bernardo *et al.* (2005), previous research on flocculation suggested the following relationships between d_{fes} and Gmed to obtain a stable floc size, with the k_{fes} coefficient encompassing erosion and fragmentation actions, which can be observed in Equations 2 and 3:

$$d_{fes}\alpha\ (G_{med})^{-(0{,}65\ a\ 0{,}76)} to\ d_{fes} <<< n \tag{2}$$

$$d_{fes}\ \alpha\ (G_{med})^{-(0{,}8\ \ a\ 1{,}0)} \quad to\ d_{fes} > n \tag{3}$$

Still on the mathematical modeling of flocculation, aggregation and rupture, Di Bernardo *et al.* (2005), present and detail Equations 4, 5, 6 and 7:

Equation 4 expresses the primary particle production rate due to floc rupture, dn_1/dt.

$$\frac{dn^1}{dt} = K_B n^0 (G_{med})^{kes} \tag{4}$$

Where: K_B is the breakage (rupture) coefficient (s), n^0 is the number of particles per unit volume at time t = 0 (m^{-3}), and the coefficient kes, is equal to 4 for flocs with d > n, and equal to 2, for flakes with d < n.

Considering the phenomenon of aggregation:

$$\frac{dn}{dt} = -\frac{4\alpha}{n}\ \Phi_f n G_{med} = -K_{ag}\Phi_f n G_{med} \tag{5}$$

In Equation 5, K_{ag} is an empirical coefficient that depends on the chemical characteristics of the system and the physical characteristics of the mixture; Φ is the volumetric fraction of the flocs and n is the number of particles per unit volume (m^{-3}).

That is, $K_A = K_{ag}\Phi_f$:

$$\frac{dn^1}{dt} = -K_A n^1 G_{med} \tag{6}$$

Where n_1 is the number of particles per unit volume at time t (m^{-3}). Combining Equations 4 and 6, the general flocculation Equation 7 results:

$$\frac{dn^1}{dt} = K_B n^0 (G_{med})^{kes} - K_A n^1 G_{med} \tag{7}$$

Argaman and Kaufman (1970), setting kes = 2, applied Equation 7 to a flocculation unit consisting of "m" completely mixed (constant G_f) chambers (reactors), in series, resulting in Equation 8:

$$\frac{n_1{}^m}{n_1^0} = \frac{1+K_B G_f^2 \frac{T_d}{m} \sum_{1=0}^{m-1}\left(1+K_A G_f \frac{T_d}{m}\right)^i}{\left(1+K_A G_f \frac{T_d}{m}\right)^m} \quad (8)$$

Where: n_1^0: Number of primary particles per volume unit present at the beginning of flocculation (m^{-3}); n_1^m: Number of primary particles per unit volume present at the exit of the m-th (m^{-3}); G_f: Mean flocculation velocity gradient (s^{-1}); m: Number of Chambers; Td: Total flocculation time (s); K_A: Aggregation coefficient; K_B: Breakage coefficient (s).

Argaman *et al.* (1970), exposed a model that contemplated the variation of velocity gradients in different flocculation chambers in series, according to Equation 9:

$$\frac{n_1^{i-1}}{n_1^i} = \frac{1+K_A G_f \frac{T_d}{m}}{1+\frac{n_1^0}{n_1^{i-1}} K_B G_f^2 \frac{Td}{m}} \quad (9)$$

In Equation 9, $\frac{n^{i-1}}{n_1^i}$ is the ratio between the number of primary particles (or turbidity) effluent and influent from flocculation chambers in sequence.

For Di Bernardo *et al.* (2005), the determination of the values of the coefficients K_A and K_B can be obtained by carrying out tests in pilot installations with continuous flow. This fact made it difficult to use the model due to the cost involved and also the relatively long time required to carry out the tests. As K_A and K_B remain constant in Equation 8 for complete series mixing chambers, according to Bratby *et al.* (1977) such coefficients theoretically should not be changed if the number of chambers tends to infinity, that is, for piston-type or static-reactor flow.

The equation to describe the kinetics of flocculation in a static reactor is similar to Equation 7, presented by Equation 10, (Bratby *et al.*, 1977).

$$\frac{dn^1}{dt} = -K_A n_t^1 G_f + K_B n_0^1 \left(G_f\right)^2 \quad (10)$$

Where: n_0^1: Number of primary particles per unit volume at time t = 0 (m^{-3}); n_t^1: Number of primary particles per unit volume at time t (m^{-3}); $\frac{dn^1}{dt}$: Variation of particles per unit volume with respect to time ($s^{-1}.m^{-3}$).

Integrating Equation 10 and rearranging the terms, Equation 11 is obtained:

$$\frac{n_0^1}{n_{Tf}^1} = \left[\frac{K_B}{K_A} G_{f+} \left(1 - \frac{K_B}{K_A} G_f\right) e^{-K_A G f T f}\right]^{-1} \quad (11)$$

Where n_{Tf}^1 represents the number of primary particles after the flocculation time T_f. The coefficients K_A and K_B, determined by using Equations 10 and 11, can be used in a system of several complete mixing chambers in series, with G_f values smaller than 100 s^{-1}.

The settling time used in the assay should be relatively long, so that the supernatant would present only primary particles, and also so that the remaining turbidity values could be used to relate them to the number of primary particles in the supernatant (Bratby *et al.*,1977).

When questioning the validity of the data found, Pádua (1994), contested this methodology, because with a long settling time, there is a corresponding very low settling velocity, different from what actually happens in water treatment plants, in which there are sedimentation velocity values in the decanters of the order of 1 to 5 $cm.min^{-1}$.

Bratby *et al.* (1977), considering that the number of primary particles is equal to the remaining turbidity, when integrating Equation 10, resulted in Equation 12 below:

$$\frac{N_0}{N_1} = \left[\frac{K_B}{K_A}\, G_{f+} \left(1 - \frac{K_B}{K_A} G_f\right) e^{-K_A G f T f}\right]^{-1} \quad (12)$$

Where: N_0: Initial turbidity of the supernatant (uT) and N_1: Final turbidity of the supernatant after long sedimentation time (uT).

1.3. Aggregation Model and Rupture and Irreversible Rupture

Marques and Ferreira Filho (2016), presented a proposed amendment to the classic model of Argaman and Kaufman, including a new term that contemplates a supposed process of irreversibility of the flakes. This inclusion would result in the appearance of particles that cannot be removed by sedimentation, and that will not form a floc again. The mathematical model proposed by the authors is demonstrated by Equations 13, 14 and 15:

$$N(t) = \frac{K_B}{K_A}.G.N_0 + \left(N_0 - \frac{K_B}{K_A}.G.N_0\right).e^{-K_A G.t} \quad (13)$$

$$F(t) = \frac{\left(K_A.G.N_0 - K_B.G^2.N_0\right)}{\left(K_C.G - K_A.G\right)}.\left(e^{-K_A G.t} - e^{-K_C G.t}\right) \quad (14)$$

$$T(t) = \left[\frac{K_C.G^2.N_0.(K_A - K_B.G)}{(K_C.G - K_A.G)}\right].\left[\frac{e^{-K_C G.t}}{K_C.G} - \frac{1}{K_C.G} - \frac{e^{-K_A G.t}}{K_A.G} + \frac{1}{K_A.G}\right] \quad (15)$$

Where: N(t) = turbidity resulting from the presence of primary particles N at time t (uT); F(t) = turbidity resulting from the presence of F particles at time t (uT); T(t) = turbidity resulting from the presence of T particles at time t (uT); K_A = aggregation constant (s); K_B = Breakage constant (s); Kc = irreversible rupture constant (s); G = mean velocity gradient (s^{-1}); t = time (s); N_0 = initial turbidity resulting from the presence of primary particles (uT).

2. MATERIAL AND METHODS

The experimental data used for the validation of models and methods in the development of the methodology proposed in this work were obtained by Voltan (2007), who studied water with the following characteristics: pH from 7.50 to 7.55, turbidity between 99 and 103 uT, apparent color between 420 to 440 uC, true color 2 uC, alkalinity from 25.6 to 26.2 mg. CaCO3 L^{-1}; conductivity of 46.5 µS.cm^{-1} and hardness of 17 to 18 mg.L^{-1} of $CaCO_3$. For this validation, an average flocculation velocity gradient was used (G = 25, 30, 35, 40, and 60 s^{-1}), with sedimentation velocities (Vs = 1.0; 2.5 and 5.0 cm-min), and flocculation time (T_f = 300; 450; 600; 750; 900; 1050; 1200; 1350; 1500; 1500; 1500; 1800; 2100; 2400 s).

For the comparison of methods and models, experimental data from Dantas *et al.* (2000), Voltan (2007), Constantino (2008) and Brito *et al.* (2016) were used. Waters with the following characteristics were studied: Dantas, 2000, pH from 7.35 to 7.55, turbidity between 24 and 28 uT, apparent color between 175 and 215 uC, alkalinity from 23 to 27 mg.L of $CaCO_3$; conductivity of 45.9 µS.cm; Constantino (2008), pH from 5.8 to 6.2, turbidity between 4 and 6 uT, apparent color between 100 and 150 uH, alkalinity from 14 to 18 mg.L of $CaCO_3$; conductivity of 78.5 µS.cm; Brito *et al.*, 2016, pH from 6.9 to 7.5, turbidity between 458 and 633.67 uT, apparent color between 720 and 750 uH, alkalinity from 42 to 43 mg.L^{-1} of $CaCO_3$; conductivity of 106.67 µS.cm^{-1}.

The comparison of models and methods (Bratby, MEAR, MPDPG and Kc – See Figure 1) was performed using an electronic spreadsheet in the Microsoft Excel 201 program for each test performed. The values of K_A and K_B and K_C were determined in electronic tables and expressed in graphical form.

Argaman and Kaufman of 1970 gave rise to the Bratby methods of 1981; MEAR and

MPDPG from 1998, proposed by Brito (1998). In 2016, Marques and Ferreira Filho published a new Model of Aggregation and Rupture and Irreversible Rupture, which in this work was named the Kc Model. Despite the innovative model of Marques and Ferreira Filho, it is based on the model of Argaman and Kaufman from 1970.

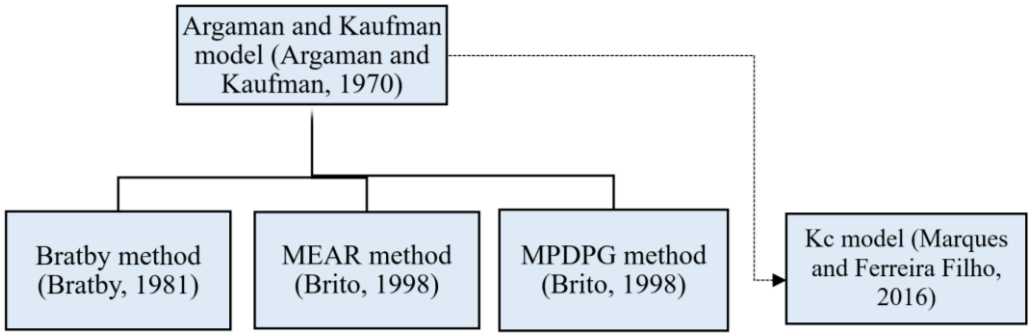


Figure 1. Models and mathematical methods for water treatment optimization.

2.1. Aggregation and disruption coefficients

2.1.1. Bratby method

For Bratby *et al.* (1977), the determination of the aggregation and breakage (rupture) coefficients in the jar test equipment, coagulation, flocculation and sedimentation tests are carried out (rest time greater than or equal to 2 h) for optimized conditions of rapid mixing, together with different times of agitation and of flocculation velocity gradient, and constructed N_0/N_1 figures as a function of flocculation time, for each velocity gradient studied. Rearranging Equation (16):

$$K_A = \frac{1}{G_f T_f} ln \left[\frac{\left(1-\frac{K_B}{K_A}G_f\right)}{\frac{1}{\frac{N_0}{N_1}}-\frac{K_B}{K_A}G_f} \right] \quad (16)$$

Assuming that no further aggregation or disaggregation of primary floc particles occurs, after a relatively long settling period in the jars of the jar test equipment, Equation 10 can be set to zero, resulting in Equation 17, (Bratby *et al.*,1977).

$$\frac{K_B}{K_A} = \frac{1}{G_f \frac{N_0}{N_1}} \quad (17)$$

According to Di Bernardo *et al.* (2005), from the horizontal portion of the best-fit curve of all experimental data (the tests must be conducted until a significant horizontal portion is produced), the value of N_0/N_1 is obtained for each value of G_f, obtaining the values of K_B / K_A. Using Equation 16 and with the values of K_B / K_A for each G_f (eq. 17), K_A and K_B are calculated. A curve is then constructed on which the values of K_B are plotted in ordinates and ln(G_f) in the abscissa axis. According to Bratby (1981), the value of K_B for any value of G_f is given by Equation 18:

$$K_B = k_{1b}\, ln\, G_f + k_{2b} \quad (18)$$

Where: k_{1b}, k_{2b} are dimensionless coefficients inherent to the water under study.

2.1.2. Aggregation and Rupture Equation Method - MEAR and First Partial Derivative Method with Relation to G_f - MPDPG

Brito (1998) studied two methods to determine K_A and K_B, from turbidity data and the

number of remaining primary particles, considering the effect of sedimentation velocity:

• The Aggregation and Rupture Equation – MEAR (modification of the method by Bratby, 1981), which considers the maximum ratio of N_0/N_1 for each value of G_f, equivalent to the shortest flocculation time in which that maximum efficiency is obtained. In this proposal, Equations 12, 16, 17 and 18 are used, considering variation in the values of the K_A and K_B coefficients for different sedimentation and G_f velocities.

• Method of the First Partial Derivative with respect to G_f - MPDPG, this method employs the first partial derivative of Equation 12 with respect to the parameter G_f (optimal value with maximum efficiency for T_f), equated to zero, giving Equation 19:

$$K_B = \frac{K_A^2 T_f e^{-K_A G_f T_f}}{\left(1+K_A G_f T_f e^{-K_A G_f T_f} - e^{-K_A G_f T_f}\right)} \quad (19)$$

To obtain Equation 19, the derivative was performed according to Equation 20, and the equation was multiplied by K_A and -1, with K_B in evidence:

$$\frac{dy}{dG_f} = \frac{KB\left(\frac{-1}{K_A} - \frac{K_A G_f T_f e^{-K_A G_f T_f}}{K_A} + \frac{e^{-K_A G_f T_f}}{K_A}\right) + \left(K_A T_f\right) e^{-K_A G_f T_f}}{\left(\frac{K_B}{K_A} G_f + e^{-K_A G_f T_f} - \frac{K_B}{K_A} G_f e^{-K_A G_f T_f}\right)^2} \quad (20)$$

2.2. Aggregation and Rupture Model and Irreversible Rupture - K_C Model

Marques and Ferreira Filho (2016), included a new term that determines the process of irreversible rupture of the flakes, resulting in the kinetic constants K_A, K_B and K_C. The method used to solve this model was based on Equation 14 and, when integrating this equation, Equation 21 was obtained:

$$\frac{N}{N_0} = \frac{(K_A.G - K_B.G^2}{K_C.G - K_A.G)} . \left(e^{-K_A.G.t} - e^{-K_C.G.t}\right) \quad (21)$$

Where: K_A and K_C are different from zero; and $K_A \neq K_C$.

To obtain the values of K_A, K_B and K_C, the "Solver" function was applied (convergence method "GRG Nonlinear"), selecting the option to minimize the value of the cell in question, varying the values initially arbitrated for the K_A constants, K_B and K_C.

3. RESULTS AND DISCUSSION

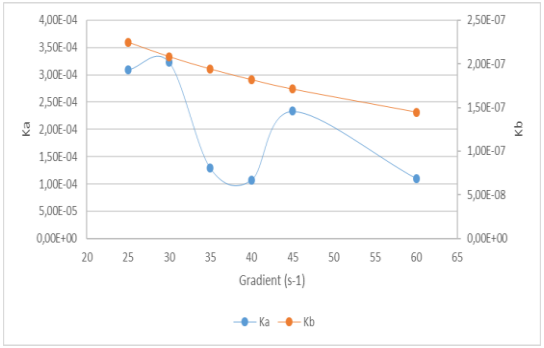
The results were obtained through simulation and compared the model of Argaman and Kaufman with the methods of Bratby; the Aggregation and Breakdown Equation method - MEAR; the Partial First Derivative method in relation to the Flocculation Velocity Gradient - MPDPG and the model K_C.

3.1. Bratby method

In the Bratby method, the values of K_B in ordinates and ln (G_f) in abscissa axis were plotted, using Equation 16 and the values of K_B / K_A for each G_f Equation 17, thus determining the values of K_A and K_B.

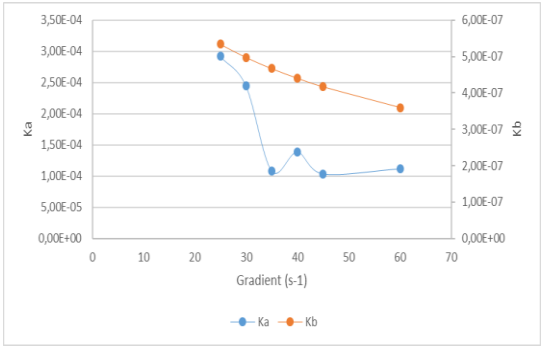
In obtaining the values of K_A and K_B as a function of G_f (s^{-1}), as seen in Figures 2-A, 2-B and 2-C, it is observed that there is a little fluctuation, and although it shows a slight tendency logarithmic for K_B values, the same is not true for K_A values.

(A)



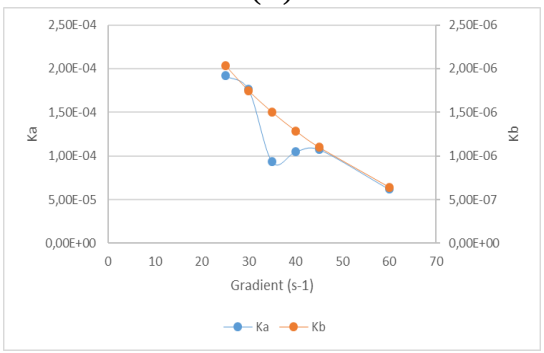
BRATBY (T_f = 2400 – Vs = 1,0 cm min^{-1})

(B)



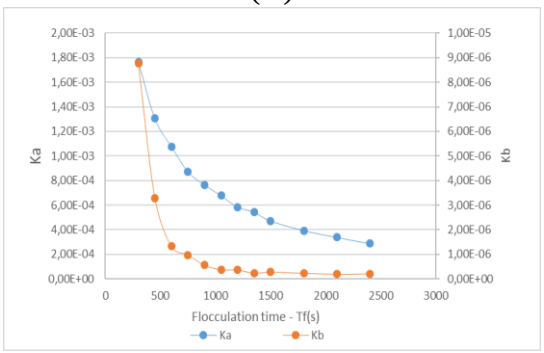
BRATBY (T_f = 2400 – Vs = 2,5 cm min^{-1})

(C)



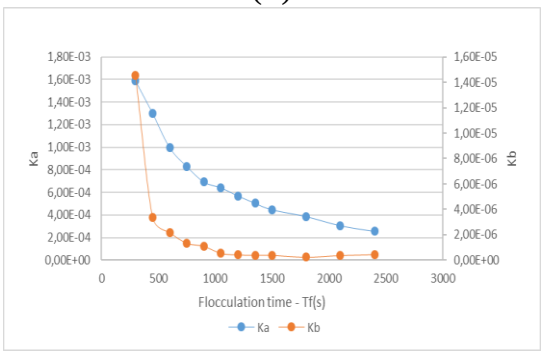
BRATBY (T_f = 2400 – Vs = 5,0 cm min^{-1})

(D)



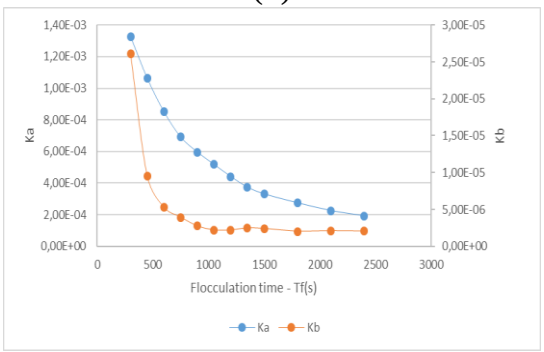
MEAR (G_f = 25 – Vs = 1,0 cm min^{-1})

(E)



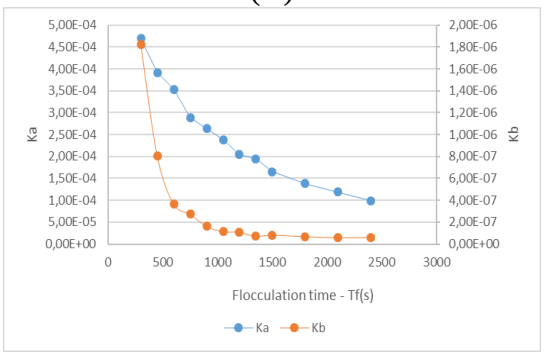
MEAR (G_f = 25 – Vs = 2,5 cm min^{-1})

(F)



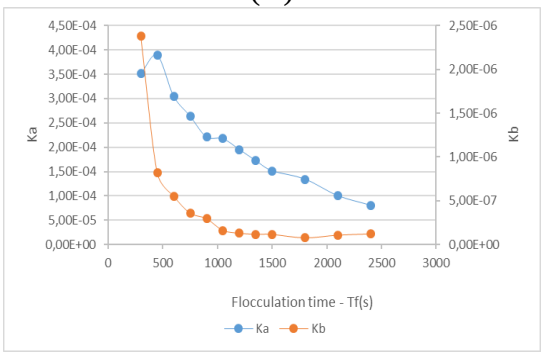
MEAR (G_f = 25 – Vs = 5,0 cm min^{-1})

(G)



MPDPG (G_f = 25 – Vs = 1,0 cm min^{-1})

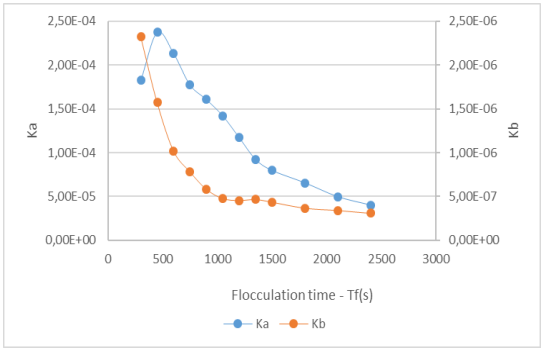
(H)



MPDPG (G_f = 25–Vs = 2,5 cm min^{-1})

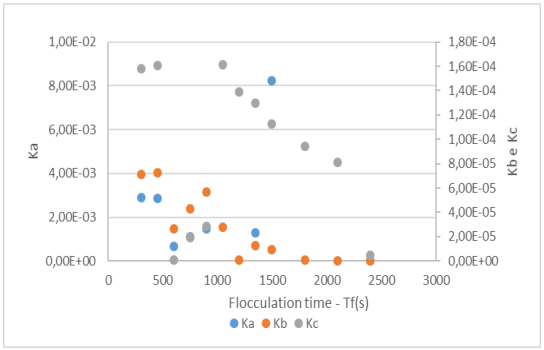
Figure 2. Graphs of validation of models and methods: obtaining of K_A and K_B values by the methods of BRATBY (A, B and C), MEAR (D, E and F), MPDPG (G, H and I) – Methods from the Model by Argaman and Kaufman (1970), and the values of K_A, K_B e Kc by the Model KC (J, K and L) - Model by Marques and Ferreira Filho (2016). Continue...

(I)



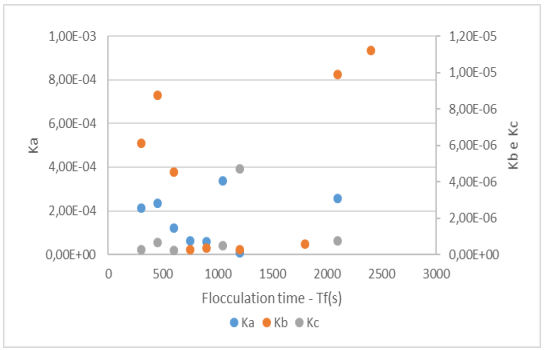
MPDPG (G_f =25 – Vs = 5,0 cm min^{-1})

(J)



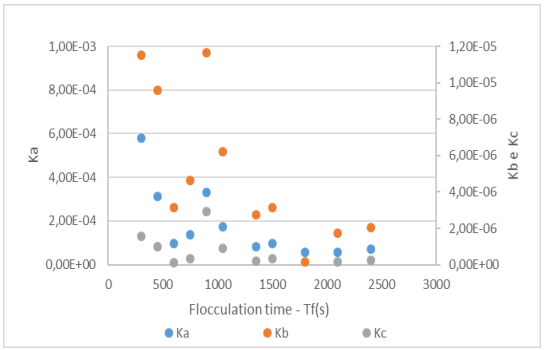
K_C model (G_f = 25 – Vs = 1,0 cm min^{-1})

(K)



K_C model (G_f = 25 – Vs = 2,5 cm min^{-1})

(L)



K_C model (G_f = 25 –Vs = 5,0 cm min^{-1})

Figure 2. Continued.

3.2. MEAR method

For the MEAR method, a routine was implemented in Excel, using Equations 12 and 17 through an iterative method, provided by the Solver tool of the Microsoft Excel program. The "Solver" function was then applied (convergence method "Nonlinear GRG") in the control cell (corresponding to the difference between the experimental and theoretical efficiency), selecting the option to minimize the value of the cell in question. In terms of restrictions, a condition of 10^{-4} was assigned, making the difference between experimental and theoretical efficiency greater than or equal to the condition in question, and the values of K_A and K_B restricted to positive values. Assigning an initial value of K_A between 10^{-1} e 10^{-20}, through the solver function, the values of K_A and K_B were obtained.

In determining the values of K_A and K_B as a function of flocculation time, it is possible to observe a logarithmic trend in almost all experiments, as shown in figures 2-D, 2-E and 2-F, with sedimentation velocity (V_s) of 1,0 cm.min^{-1}; 2,5 cm.min^{-1} and 5,0 cm.min^{-1}.

3.3. MPDPG method

In the MPDPG method, the "Solver" function (Non-Linear GRG convergence method) was applied to the control cell (corresponding to the difference between the experimental efficiency and the experimental efficiency), selecting the "value equal to zero" option of the cell in question, varying the values initially arbitrated for the constants K_A, K_B. In terms of restrictions, the values of K_A and K_B were restricted to necessarily positive values. Assigning an initial value of K_A between 10^{-1} and 10^{-20}, through the solver function, the values of K_A and K_B were obtained.

When analyzing the values obtained for K_A and K_B as a function of flocculation time, a logarithmic trend is observed with rare exceptions as shown in Figure 2-H and 2-I, where the K_A dropped below the K_B when the flocculation time was at approximately 400 T_f (s) and

ascending again when the flocculation time was close to 500 T_f (s).

3.4. Kc model

For the K_C model, the "Solver" function (convergence method "Nonlinear GRG") was applied to the control cell (corresponding to the difference between the experimental and theoretical efficiency), selecting the option to minimize the value of the cell in question, varying the values initially arbitrated for the constants K_A, K_B and K_C are used.

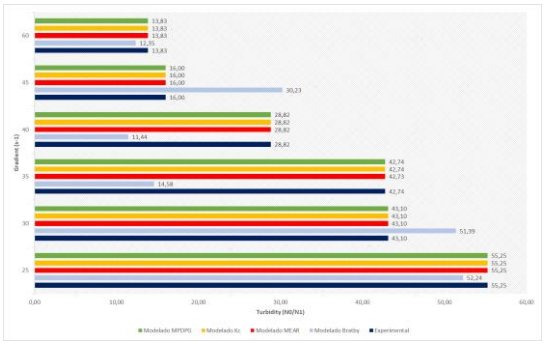
Differently from what was observed in this model, in obtaining the values of the constants K_A, K_B e K_C, at least visually there is no clear trend, as shown in Figures 2-J, 2-K and 2-L.

In terms of constraints, a condition of 10^{-10} was assigned, making the difference between experimental and theoretical efficiency greater than or equal to the condition in question, and the values of K_A, K_B and K_C restricted to positive values. Assigning an initial value of K_A, K_B and K_C between 10^{-1} and 10^{-20}, through the solver function, the values of K_A, K_B and K_C were obtained.

3.5. Comparison between methods and models

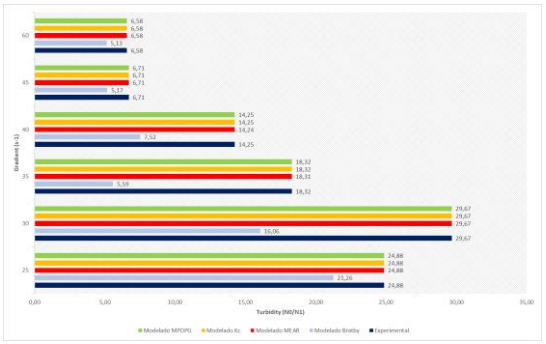
After validating the models and methods, the methods of Bratby, MEAR, MPDPG and KC model were modeled and compared in Figure 3, as well as for the authors listed in Tables 1 and 2. It can be identified in Figure 3-A, with Vs = 1.0 cm min^{-1}, in 3-B, with Vs = 2.5 cm min^{-1} and in figure 3-C, with Vs = 5.0 cm min^{-1} that only the Bratby Model oscillates in relation to the experimental data, emphasizing that in this experiment Voltan used aluminum sulfate as coagulant.

(A)



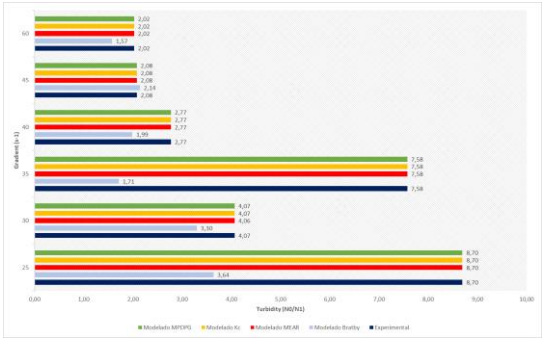
Voltan (2007) – Vs = 1,0 cm min^{-1}

(B)



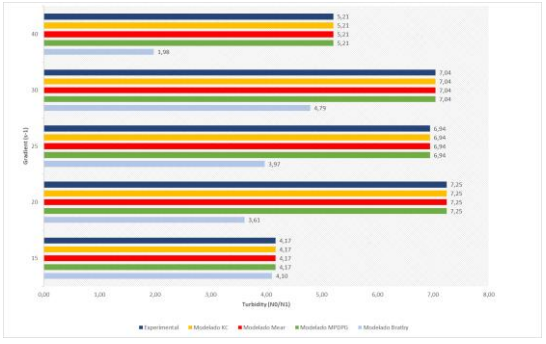
Voltan (2007) – Vs = 2,5 cm min^{-1}

(C)



Voltan (2007) – Vs = 5,0 cm min^{-1}

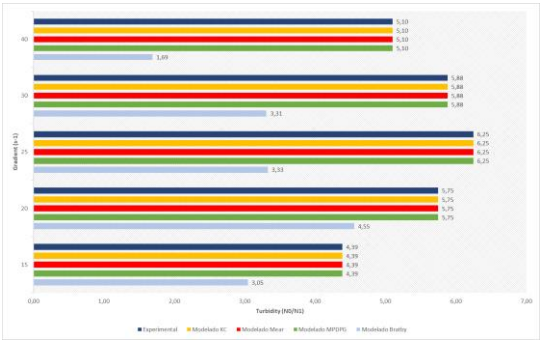
(D)



Constantino (2008) – Vs = 1,0 cm min^{-1}

Figure 3. Comparative analysis between the methods and models simulated for different sedimentation velocities in relation to the respective experimental data of the referenced authors. Continue...

(E) (F)



Constantino (2008) – Vs = 1,5 cm min^{-1} Constantino (2008) – Vs = 3,0 cm min^{-1}

Figure 3. Continued.

In Figure 3-D, with Vs = 1.0 cm min^{-1}, in 3-E, with Vs = 1.5 cm min^{-1} and in Figure 3-F, with Vs = 3.0 cm min^{-1}, which evidences the experimental data of Constantino (2008), although he used ferric chloride as a coagulant and a different sedimentation rate from the study by Voltan (2007), it was possible to observe similar results in relation to the compared models.

The experimental data from Dantas *et al.* (2000), Voltan (2007), Constantino (2008) and Brito *et al.* (2016) were simulated by the MEAR, MPDPG and K_C - Marques and Ferreira Filho Models. Table 1 presents the values with their respective deviations (errors), difference between experimental and theoretical efficiency. Table 2 presents the deviations obtained by the Bratby Method.

The optimal experimental values considered by the aforementioned authors for flocculation time (T_f) and velocity gradient (G_f) were used: Dantas *et al.* (2000): T_f = 1440 s and G_f = 25 s^{-1} for the Vs of (7.2 ; 3.95; 1.89 cm.min^{-1}), Voltan (2007): T_f = 1200 s and G_f = 25 s^{-1} for Vs of (7.0; 3.3; 2.07; 1.45; 0 .9 cm.min^{-1}), Constantino (2008): T_f = 1500 s and G_f = 10s^{-1} for Vs of (4.67; 2.2; 1.38; 0.97; 0.6; 0.42 cm .min^{-1}) for these data used the coagulant ferric chloride. For the aluminum sulfate coagulant, Constantino, 2008 considered a T_f = 900 s and G_f = 10 s^{-1} for the same Vs and Brito *et al.* (2016) considered a T_f = 900 s and G_f = 30 s^{-1} for Vs of (1 .0; 1.5; 2.0 cm.min^{-1}) for PAC coagulant and Aluminum Sulfate.

In Table 1, it is possible to verify that the MEAR and MPDPG Methods have an average of approximately 18% error deviation in relation to the experimental values of their respective references, while the Kc Model has 19% error deviation. However, when analyzing the different sedimentation velocities, it is possible to verify that there are also high error deviations, as in Constantino (2008) with a sedimentation velocity of 4.67 (using aluminum sulfate as coagulant) whose error deviation was approximately 127% for MEAR and MPDPG, while for the Kc Model it was 56%. In another situation, analyzing Brito *et al.* (2016) (which used the PAC coagulant and with Vs = 2.0 cm min^{-1}) the error deviation for the Kc Model was approximately 178%, while the other analyzed methods had a deviation of 64%; that is, there is times when the Kc Model has very high deviations in relation to the MEAR and MPDPG Methods and in other circumstances the opposite occurs.

Table 1. Performance of the modeling methods and models in relation to the experimental data of the cited references).

References – Coagulant Type	Sedimentation Velocities (Vs) ($cm.min^{-1}$)	Argaman & Kaufman Model		K_C Model - Marques and Ferreira Filho (2016)
		Deviations from methods: Mear and MPDPG		
		MEAR Deviation (% error)	MPDPG Deviation (% error)	Deviation (% error)
Dantas *et al.* (2000) - Aluminum Sulfate	7,20	8,34%	8,33%	9,09%
	3,95	17,72%	17,73%	15,06%
	1,89	7,39%	7,39%	6,88%
Voltan (2007) - Aluminum Sulfate	7,00	4,14%	4,14%	4,32%
	3,30	2,06%	2,06%	2,02%
	2,07	10,16%	10,16%	9,23%
	1,45	5,45%	5,45%	5,76%
	0,90	6,57%	6,57%	7,02%
Constantino (2008) - Ferric Chloride	4,67	5,68%	5,68%	5,38%
	2,20	15,28%	15,28%	18,03%
	1,38	7,94%	7,94%	7,35%
	0,97	0,00%	0,00%	0,00%
	0,60	11,29%	11,29%	12,73%
	0,42	0,00%	0,00%	0,00%
Constantino (2008) - Aluminum Sulfate	4,67	126,81%	126,83%	55,91%
	2,20	18,76%	18,77%	15,80%
	1,38	15,20%	15,19%	17,92%
	0,97	5,37%	5,38%	5,10%
	0,60	18,32%	18,32%	22,43%
	0,42	6,03%	6,02%	6,41%
Brito *et al.* (2016) – (PAC)* (DC–50)**	2,00	64,06%	64,06%	178,27%
	1,50	6,63%	6,63%	7,10%
	1,00	0,23%	0,23%	0,23%
Brito *et al.* (2016) – Aluminum Sulfate	2,00	59,02%	59%	37,11%
	1,50	32,83%	33%	24,72%
	1,00	3,97%	4%	26,83%
General average of deviation ± Standard Deviation of the Mean (%)		18 ± 27%	18 ± 27%	19 ± 35%
Coefficient of variation		1,55	1,55	1,81

* Aluminum Polychloride ** Coagulant Dosage

Table 2. Average Percentage of Error Deviation of the Bratby Method in relation to the experimental data of the cited references.

Reference-Coagulant Type	Sedimentation Velocities ($cm.min^{-1}$)	Velocity Gradient (Vs)	Average Deviation (% error)	Reference–Coagulant Type	Sedimentation Velocities ($cm.min^{-1}$)	Velocity Gradient (Vs)	Average Deviation (% error)	Reference – Coagulant Type	Sedimentation Velocities ($cm.min^{-1}$)	Velocity Gradient (Vs)	Average Deviation (% error)
Voltan (2007) (S.A)*	5,0	25	35,56%	Constantino (2008) (S.A)*	3,0	10	28,95%	Constantino (2008) (C. F)**	3,0	10	-
		30	18,73%			15	30,57%			15	25,08%
		35	37,80%			20	34,76%			20	24,76%
		40	23,86%			25	32,53%			25	27,78%
		45	32,90%			30	26,29%			30	26,10%
		60	23,55%			40	27,48%			40	41,75%
Voltan (2007) (S.A)*	2,5	25	36,19%	Constantino (2008) (S.A)*	1,5	10	28,78%	Constantino (2008) (C.F)**	1,5	10	-
		30	22,52%			15	24,36%			15	24,05%
		35	52,94%			20	27,55%			20	15,95%
		40	37,44%			25	41,64%			25	29,55%
		45	40,18%			30	40,98%			30	21,07%
		60	27,27%			40	43,61%			40	40,96%
Voltan (2007) (S.A)*	1,0	25	19,45%	Constantino (2008) (S.A)*	1,0	10	20,86%	Constantino (2008) (C. F)**	1,0	10	-
		30	48,75%			15	26,56%			15	4,92%
		35	49,55%			20	26,89%			20	27,46%
		40	50,82%			25	35,33%			25	28,83%
		45	43,62%			30	35,52%			30	23,19%
		60	28,87%			40	40,73%			40	42,22%
General average of deviation ± Standard Deviation of the Mean (%)											32 ± 10%
Coefficient of variation											0,31

*Coagulant used Aluminum Sulfate **Coagulant used Ferric Chloride.

The Bratby Method uses a very high flocculation time for the different G_f values, and it is not possible to apply it in the references used in Table 1. As a result, the analysis was performed separately, using other samples, with sufficiently high flocculation and very low velocity gradients for the application of this particular method, as shown in Table 2. It is worth noting that such gradients and sedimentation velocities are not commonly applied in ETA's or in academic works. It is observed that the average error deviation of the Bratby Method is approximately 32%, much higher than the MEAR and MPDPG methods and the Kc Model presented in Table 1.

Although the MEAR and MPDPG methods and the K_C Model present a lower mean deviation as shown in Table 1, there is a very high standard deviation of approximately 27%, 27% and 35%, respectively. Although Bratby's Method has a lower standard deviation, it has a high mean deviation compared to the others (Table 2).

4. CONCLUSION

The MEAR and MPDPG Methods and the Model Kc, compared with the references used in this study, and using as base the mathematical model of Argaman and Kaufman (1970), modeled well, while the method of Bratby did not obtain favorable results in the studied comparisons.

The Bratby Method presents greater consistency in relation to the other methods and models; however, it has a greater systematic error, since it is already part of an average of around 30% of error. However, the MEAR and MPDPG Methods and the K_C Model manage to reach an average of smaller deviations; however, they lead to deviations that can reach a high inconsistency, obtaining almost twice the average value, as identified in the K_C Model.

This is unlike the Bratby (1981) Method, which says that for each water sample there is a K_A and a K_B, and that they are evaluated for very large T_f values and for very low settling rates. On the other hand, the MEAR and MPDPG methods proved to be more feasible, with the reality of the experimental data used in present work. Brito (1998), when developing the MEAR method, said that there is a K_A and a K_B for each G_f and that it will change depending on sedimentation rate.

Thus, when analyzing the results of the mathematical methods of MEAR and Bratby, it was possible to identify that the MEAR Method follows a logarithmic trend for K_A and K_B, while Bratby did not obtain the same results. This trend is also identified in the MPDPG Method.

As for the Marques and Ferreira Filho (2016) Model, it may be an innovation, however, it did not represent a significant improvement in effectively describing the flocculation process in water treatment in terms of the mathematical model. It must be considered that the premise that the flocs can be broken in an “irreversible” way, as designated by the authors, can be confused with primary particles that were not even destabilized and presented efficiency similar to the MEAR and MPDPG Methods based on the Argaman and Kaufman Model (1970).

This work does not exhaust the options for studies on the models and methods discussed, but it advances understanding of their relevance and a comparison between them, considering the references used here. The results obtained in this work provide insights for the design of flocculation units in water treatment plant projects.

5. ACKNOWLEDGMENTS

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