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ORIGINAL PAPER

LATENT COAGULATION OF SUSPENDED SOLIDS CONTAINING P-PO$_4$ – COMPUTER- SIMULATION

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Abstract

Coagulation is an important process in water and wastewater treatment, including the removal of P-PO$_4$. This process has not been fully examined and explained. It results from not only the complexity of this process, the quantity of factors influencing its process, but from the number of restrictions in the field of laboratory experiments. This paper presents a new method of researching complex processes of coagulation through a computer simulation. This computer program is based on a semi-empirical concept, inspired by observations of coagulation-flocculation-sedimentation of sewage containing, among others, P-PO$_4$, carried out in both natural and model systems. In this study, a computer simulation was performed to examine the kinetics of latent coagulation, that can be the main determinant of coagulation efficiency. Several factors that influence the rate of latent coagulation were analyzed, including coagulant overdosing and underdosing, type of coagulant, initial concentration (load) of wastewater containing P-PO$_4$, wastewater dispersion and initial velocity of sol and coagulant particles. The rate of latent coagulation decreased in response to both coagulant underdosing and overdosing. The time of latent coagulation was highly correlated with the degree of system dispersion. An increase in the size of sol and coagulant particles accelerated the rate of latent coagulation. The results of the study indicate that the costs associated with coagulant preheating are not compensated by the resulting increase in the rate of latent coagulation. A better response was observed by preheating or stirring sewage, in particular wastewater with a low load.

Keywords: computer simulation, latent coagulation, sol, wastewater, P-PO$_4$.

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INTRODUCTION

Coagulation is one of the most important processes in chemical wastewater treatment (Lee et al. 2014, Loloei et al. 2014, The et al. 2016). Coagulation plays an important role in removing P-PO_4 from water and wastewater (Langer et al. 2017). Phosphorus compounds occurring in colloidal systems due to their dimensions create a stable suspension. One of the most effective methods of removing this type of contamination is the coagulation process. The main aim of this process is to coagulate dispersed colloidal substances with the use of electrolytes. Colloidal particles are destabilized to form larger clusters, flocs, capable of sedimentation. Every coagulation/flocculation process begins with particle aggregation, which decreases the degree of dispersion. This stage does not produce visible signs of coagulation, such as changes in turbidity or color, and it is often referred to as latent coagulation. Successive processes induce changes which are easy to observe in the system, and this stage is known as active coagulation. In lyophobic colloids, latent coagulation is a very short process, which is why most research has long focused on active coagulation. Meanwhile, latent coagulation can be the main determinant of coagulation efficiency (Duan, Gregory 2003, Starow 2010, Smoczyński et al. 2014).

Coagulation can be influenced by various chemical and physical factors, in particular the type and dose of the coagulant, concentration and type of water impurities, pH and chemical composition of water. Coagulation is also influenced by the conditions of the coagulation process, including temperature, time and stirring rate. For these reasons, coagulation is a complex process that continues to attract researchers' interest (Grant et al. 2001, Sillanpää et al. 2018, Xu et al. 2018). Coagulation is studied in natural systems, such as wastewater (Al-Shannag et al. 2012, Zhao et al. 2014), as well as in modeled systems, such as silica suspensions (Shi et al. 2011, Skvarla 2013, Habasaki, Ishikawa 2014). Researchers increasingly often rely on computer simulations which create practically unlimited possibilities for experimentation and support analyses of factors that influence the kinetics and efficiency of coagulation. The results of computer simulations expand fundamental databases, they have practical applications and could create an interesting alternative to applied research (Smoczyński et al. 2009*a*, Yu 2014, Zangooei et al. 2016).

This article discusses the results obtained in the ZB2 computer program which simulates the coagulation-flocculation of suspended solids containing P-PO_4. In this study, a computer simulation was performed to examine the kinetics of latent coagulation, a process that does not produce visible symptoms and is very difficult to observe in a laboratory. Several factors that influence the rate of latent coagulation were analyzed, including coagulant overdosing and underdosing, type of coagulant, concentration (load) of wastewater containing P-PO_4, wastewater dispersion and the initial velocity of sol and coagulant particles.

MATERIALS AND METHOD

The simulation model

The ZB2 program simulating the process of coagulation-flocculation of suspended solids was based on long-term studies of coagulation and electrocoagulation processes carried out on natural sewage i.e. agricultural, municipal, cellulose and paper waste as well as a silica solution model or a cellulose-paper model. The program has been positively verified based on the principles of the classical kinetic theory and dispersoid diffusion, and it complies with the logic and the general principles of colloidal system destabilization (Smoczyński 2009*b*, 2013, Wardzyńska, Załęska-Chróst 2016, 2018). ZB2 is a stochastic dynamic model that relies on random variables, and the system's state changes over simulation time. The program's core is a module solving the equation of motion for a given number of particles in a closed vessel. The ZB2 program simulates the process of destabilizing the polydisperse sol; there are spherical particles of the I and II type that can differ in size, mass, and initial velocity. These particles can simulate, respectively, coagulant particles and sol particles. The program simulates rapid perikinetic coagulation where every collision of a coagulant particle and a sol particle produces an unbreakable bond and where particles of the same type do not aggregate. The sedimentation of clusters/flocs begins when the coagulation threshold (E) is exceeded. The coagulation threshold denotes the number of sol particles which initiate the sedimentation of a cluster/floc, where a given number of sol particles corresponds to one coagulant particle. The sedimentation rate increases with an increase in the number of sol particles in a cluster. The initial particle location values are randomly generated based on homogeneous distribution in the vessel. The direction of particle movement and their initial location are randomly drawn using the RANDOM command. The angle at which a particle bounces off the vessel wall is always equal to the angle of incidence. To simulate the friction between the particle and the fluid, floc speed in the direction of the surface is decreased by 0.1% per unit of displacement.

Variable parameters of the simulation model

The following input data are used in simulation tests:

N_c – number of coagulant particles, 20 - 200;
N_s – number of sol particles P-PO_4, 200, 400, 600, 800, 1000, 1200;
V_c – initial velocity of a coagulant particle, 50, 100, 150, 200, 250, 300;
V_s – initial velocity of a sol particle P-PO_4, 50, 100, 150, 200, 250, 300;
E – coagulation threshold, 10 = const;
F – sedimentation coefficient, 0.2 = const;
r_c – radius of a coagulant particle, 1 - 10;
r_s – radius of a sol particle, 1 - 10;
m_c/m_s – ratio of the coagulant particle mass to the sol particle mass, 1 = const.

The parameters of the simulation model used in this research are based on observations of coagulation-flocculation-sedimentation of wastewaters containing P-PO_4, carried out in both natural and model systems. All the units are the simulated values, for instance a time unit is the simulated second. Time (*t*) was measured in seven replications. One maximum value and one minimum value were discarded, and the arithmetic mean of the remaining five values was computed. Standard deviation values are shown in the respective diagrams.

RESULTS AND DISCUSSION

The effect of coagulant underdosing and overdosing on the latent coagulation

Unlike experimental methods, computer simulations support analyses of the initial stage of coagulation, also known as latent coagulation, which can determine the effectiveness of the entire process. In the presented simulations, the time of latent coagulation was defined as the time elapsed between the addition of the coagulant and the formation of the first sedimenting floc.

The choice of a coagulant and its dose are the procedural problems in wastewater treatment by chemical coagulation (Feo et al. 2008, Manamperuma et al. 2017). Coagulant overdosing is undesirable due to high cost and possible contamination with secondary impurities.

The effect of coagulant underdosing and overdosing on the time required for the formation of the first sedimenting floc in wastewater with different initial concentrations, N_s = 600, 800 and 1000, is presented in Figure 1. The initial velocity and radius of sol and coagulant particles were constant $V_s = V_c = 50$, $r_s = r_c = 1$. The optimal coagulant dose (100% of coagulant) was calculated stoichiometrically, i.e. the applied dose led to the complete removal of sol particles from the system at the coagulation threshold of E=10 (for example, for the initial sol concentration of Ns = 1000, the optimal coagulant dose was 100). Significant underdosing (20% of the optimal dose) prolonged the formation of the first floc by 38-46% relative to the optimal dose, subject to the initial sewage load. The data presented in Figure 1 indicate that both underdosing (≤ 40% of the optimal dose) and overdosing did not significantly influence the rate at which the first floc was formed, in particular in wastewater with a low initial concentration (low load). Overloading could be expected to significantly shorten the time required for the formation of the first floc; however, it delivered the opposite effect in some cases. The above could be attributed to the fact that coagulation in the presented simulation model was driven by the interactions between two colloidal systems formed by sol and coagulant particles, and coagulant overdosing

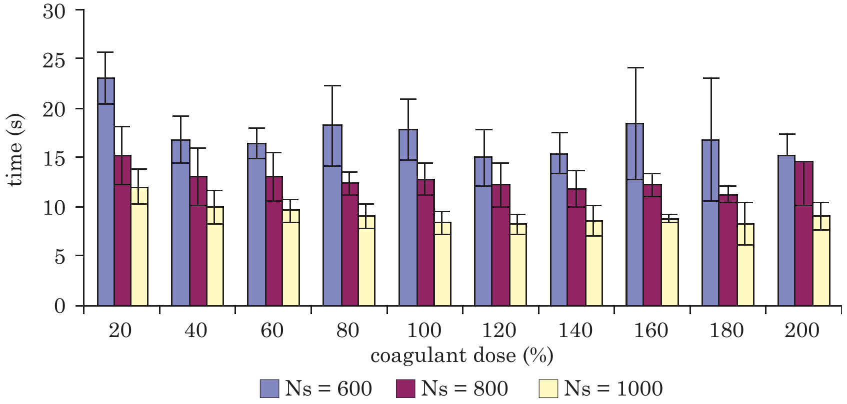

Fig. 1. Time required for the formation of the first sedimenting floc containing P-PO_4, subject to coagulant dose and wastewater concentration (number of sol particles P-PO_4 -N_s)

(relative to the coagulation threshold) contributed to the predominance of differently charged particles and the formation of a new colloidal system.

The influence of coagulant particle size on the rate of latent coagulation

Selection of a coagulant is a critical step, and several tests need to be performed to choose the optimal dose and type of a coagulant. Despite chemical similarities, various types of the same coagulant with minor differences in structure and composition are used in practice. However, these minor differences can significantly influence coagulation efficiency.

The influence of coagulant particle size on the rate of latent coagulation, expressed by the time required for the formation of the first floc, is presented in Figure 2. The initial velocity of sol and coagulant particles and the radius of sol were constant $V_s = V_c = 50$, $r_s = 1$. An increase in the coagulant particle

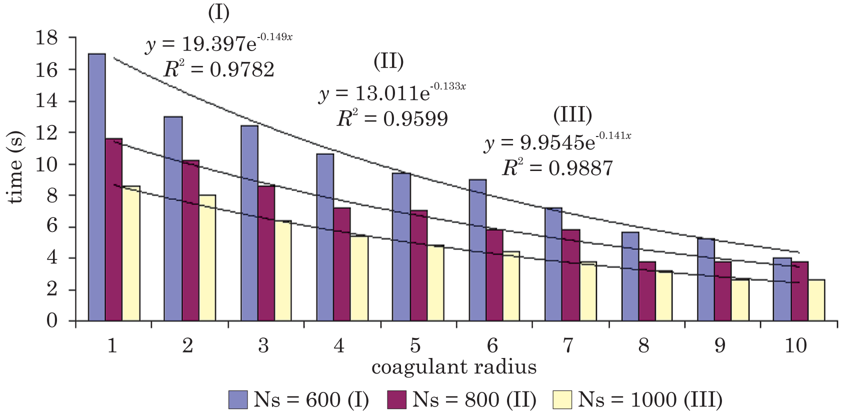

Fig. 2. The influence of coagulant particle size on the rate of latent coagulation, subject to initial wastewater load (N_s – P-PO_4)

size led to a decrease in coagulant density because the ratio of the mass of a coagulant particle to the mass of a sol particle remained constant ($m_c/m_s=1$). The size of sol particles was constant at $r_s=1=\text{const}$. The influence of coagulant particle size on the rate of latent coagulation was compared in sewage with different initial concentrations, $N_s=600$, 800 and 1000. An increase in coagulant particle size shortened the time required for the formation of the first floc. A 5-fold increase in coagulant particle size led to a 1.8-fold increase in the rate of latent coagulation regardless of the initial wastewater load. Variations in the rate of latent coagulation in wastewater with different concentrations were observed when coagulant particles with a radius of $r_c \geq 8$ were used. A 10-fold increase in the coagulant particle size induced a 4.15-fold increase in the rate of latent coagulation in wastewater with initial load of $N_s=300$, a 3-fold increase in wastewater with initial load of $N_s=800$, and 3.3-fold increase in wastewater with initial load of $N_s=1000$. Therefore, larger coagulant particles had a greater influence on the rate of latent coagulation in wastewater with a lower load than in sewage with a higher load.

The influence of wastewater dispersion and load on the rate of latent coagulation

The coagulation process was also influenced by the type, composition and dispersion of wastewater.

The influence of wastewater dispersion on the rate of latent coagulation in wastewater with initial load of $N_s=600$, 800 and 1000, assuming the constant initial velocity of sol and coagulant particles $V_s=V_c=50$, is presented in Figure 3. Coagulation was carried out with the use of coagulant particles of the same type and density, with radius $r_c=1$. The radius of sol particles (r_s) increased from $r_s = 1$ to $r_s = 10$. The degree of sol dispersion considerably influenced the rate of latent coagulation. A 5-fold increase in dispersion led

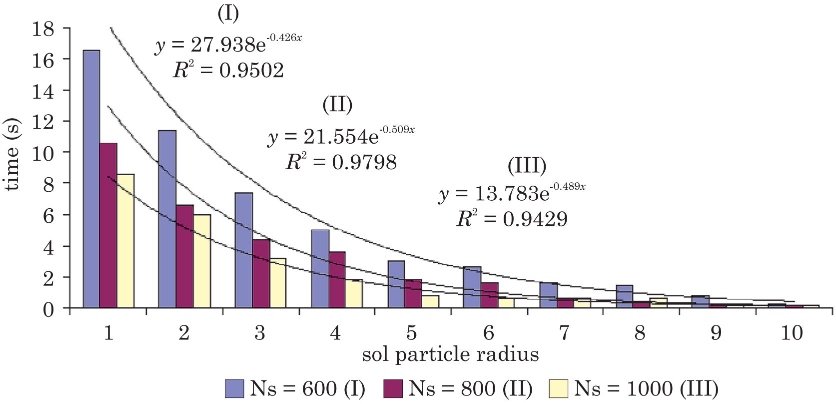

Fig. 3. The influence of wastewater dispersion and load (N_s - P-PO_4) on the rate of latent coagulation

to a 10.8-fold increase in the rate of latent coagulation in wastewater with the initial load of N_s=1000, a 5.9-fold increase in wastewater with the initial load of N_s = 800, and a 5.5-fold increase in wastewater with the initial load of N_s = 600.

The theory of rapid coagulation kinetics developed by Marian Smoluchowski applies only to monodisperse systems with spherical particles of identical size. According to SMOLUCHOWSKI'S equation (1917), the probability that two particles with the same radius will collide is expressed by coefficient *k*:

$$k = 4\pi AD \tag{1}$$

where A is the radius of the attraction zone, and D is the diffusion coefficient. The radius of the attraction zone is roughly equal to particle diameter.

In practice, however, most systems are polydisperse, and they contain differently sized particles. The simplest polydisperse sol is composed of two types of particles with radii r_1 and r_2. The radius of attraction zone A equals:

$$A = r_1 + r_2 \tag{2}$$

whereas the probability that a larger particle and a smaller particle will collide equals:

$$k_1 = 2\pi A(D_1+D_2) \tag{3}$$

where D_1 and D_2 are diffusion coefficients for sol particles with radii r_1 and r_2.

In our simulation, an increase in the radius of sol or coagulant particles increased the radius of the attraction zone and, consequently, the probability of particle collision and aggregation, which increased the rate of latent coagulation. The course of latent coagulation in monodisperse and polydisperse systems clearly differed when the ratio of particle radii was high.

The results of the performed simulation also indicate that the initial wastewater load significantly influences the rate of latent coagulation. According to Fick's second law (Sonntag, Strenge 1987), the value of D is determined by the system's concentration, and the relationship between diffusive flux δc/δt and concentration gradient δc/δx can be expressed with the use of the following equation:

$$\delta c/\delta t = \delta/\delta x[D\ (\delta c/\delta x)] \tag{4}$$

where x is a generalized coordinate.

Equation (4) indicates that an increase in diffusive flux δc/δt is directly proportional to D. Theoretically, an increase in concentration should lead to an increase in the value of D and the probability that two particles will collide. The above is validated by the results of our simulation where an increase in the initial wastewater load always increased the rate of latent coagulation.

The influence of the initial velocity of sol and coagulant particles on the rate of latent coagulation

Coagulation is also influenced by physical factors, such as temperature, time and stirring intensity.

The correlation between the rate of latent coagulation and the initial velocity of sol and coagulant particles is presented in Figure 4*a*. Sol and coagulant particles had the same size $r_c = r_s = 1 = \text{const.}$, density $m_c/m_s=1=\text{const.}$ and initial velocity $V_c=V_s$. The absence of differences in the initial velocity of sol and coagulant particles implies that coagulation took place in the presence of a coagulant, in a system that was heated or intensively stirred, where the initial velocity of sol and coagulant particles increased from $V_c = V_s = 50$ to $V_c = V_s = 300$. Every increase in the initial velocity of sol and coagulant particles led to an increase in the rate of latent

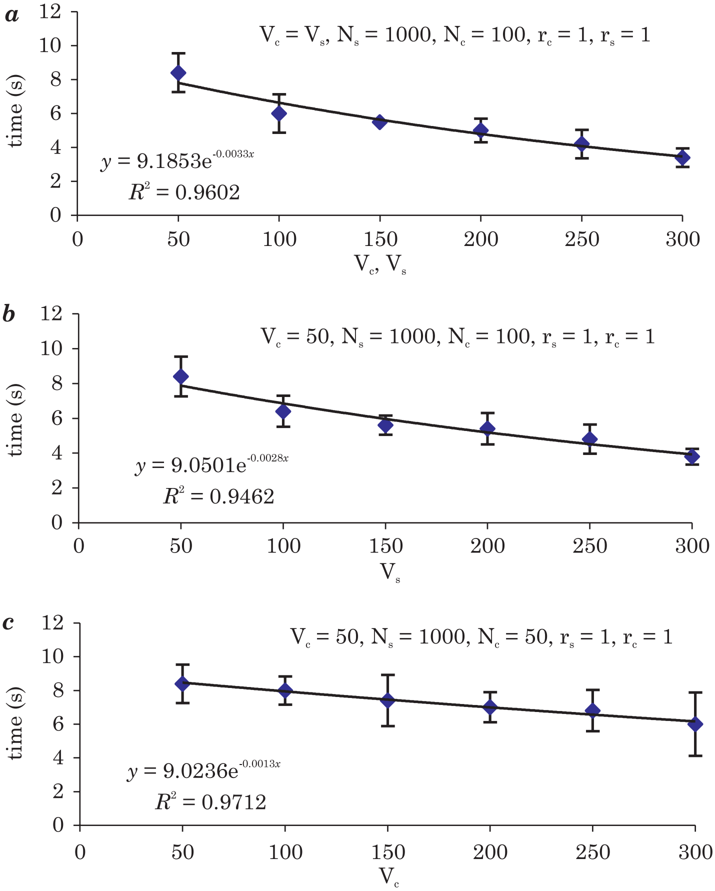

Fig. 4. The influence of the initial velocity of: *a* – sol P-PO_4 and coagulant particles, *b* – sol P-PO_4 particles, *c* – coagulant particles, on the rate of latent coagulation

coagulation. A 6-fold increase in the initial velocity of sol and coagulant particles shortened floc formation time 2.5-fold.

The influence of the initial velocity of sol particles on coagulation rate, expressed by the time required for the formation of the first sedimenting floc, is presented in Figure 4*b*. The initial velocity of coagulant particles remained constant at $V_c = 50$, whereas the initial velocity of sol particles increased from $V_s = 50$ to $V_s = 300$. Therefore, differences in the initial speed of sol and coagulant particles can simulate a system where the coagulant is added to preliminarily heated (before coagulant addition) or intensively stirred wastewater to increase the initial velocity of sol and coagulant particles from $V_s = 50$ to $V_s = 300$. The increase in the initial velocity of sol particles shortened the time required for the formation of the first floc and increased the rate of latent flocculation. A 6-fold increase in the initial velocity of sol particles led to a 2.2-fold increase in the rate of latent coagulation, which corresponds to the data presented in Figure 4*a*.

The influence of the increase in the initial velocity of coagulant particles on the rate of latent coagulation is presented in Figure 4*c*. The initial velocity of sol particles remained constant at $V_s = 50$ = const. The presented system can simulate a process where the coagulant was preheated before addition, which increased the initial velocity of coagulant particles from $V_c = 50$ to $V_c = 300$. The plotted curve indicates that the increase in the initial velocity of coagulant particles had a minor influence on the formation of the first sedimenting floc. A 6-fold increase in the velocity of coagulant particles increased the rate of latent coagulation only 1.4-fold. Therefore, an increase in the initial speed of coagulant particles (heating, stirring) before addition had no significant effect on the rate of latent coagulation.

The influence of the initial velocity of sol particles on the formation of the first sedimenting flock in wastewater with different loads is presented in Figure 5. The initial velocity of coagulant particles remained constant

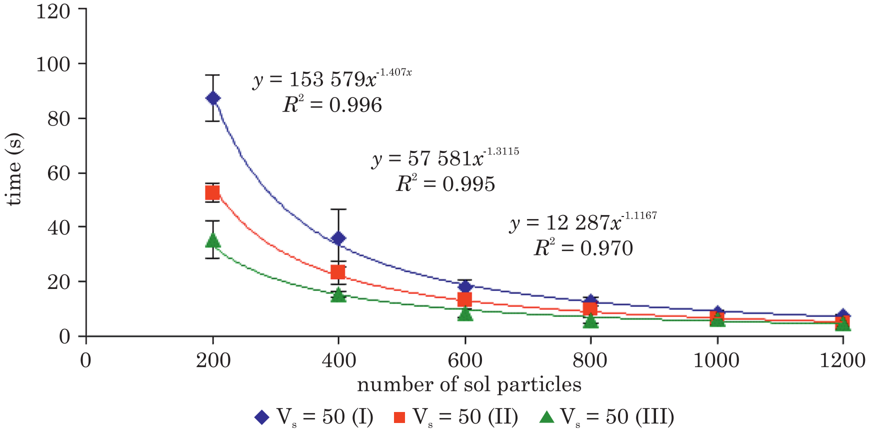

Fig. 5. The influence of the initial velocity (V_s) of sol particles P-PO_4 on the rate of latent coagulation in wastewater with different loads

at V_c=50=const. The initial velocity of sol particles had the greatest influence on the rate of latent coagulation in wastewater with low initial load. In wastewater with initial load of N_s=200, a 3-fold increase in the initial velocity of sol particles increased the rate of latent coagulation 2.5-fold. In wastewater with initial load of N_s=1200, a 3-fold increase in the initial velocity of sol particles shortened the formation of the first floc 1.5-fold. The above observations can have practical implications because higher initial velocity (achieved through heating or intensive stirring) can significantly shorten the time of latent coagulation, but only in wastewater with a low load.

In perikinetic coagulation, dispersion is decreased when destabilized colloidal particles collide due to Brownian motion. This type of coagulation takes place in preliminary sedimentation tanks and septic tanks in wastewater treatment plants. Colloidal particles suspended in sewage have low kinetic energy, which is why colloids are destabilized and precipitated at a slow rate. In practice, the initial velocity of any particle changes in response to variations in temperature or stirring intensity because both parameters always lead to changes in kinetic energy. According to Smoluchowski's theory, the probability that two particles will collide increases with an increase in displacement (in a unit of time) in Brownian motion. In practice, this displacement has to be directly correlated with the initial velocity of a particle in the ZB2 program, and it has to increase the probability of particle collision. The results of the performed simulation confirm the above observation. The probability that two particles will collide increases and the rate of latent coagulation decreases with a rise in the initial velocity of particles.

SUMMARY AND CONCLUSION

The presented simulation experiment revealed that both coagulant underdosing and overdosing lower the rate of latent coagulation, expressed by the time required for the formation of the first sedimenting floc. In the simulation model where coagulation was driven by interactions between two colloidal systems, coagulant underdosing and overdosing led to the predominance of differently charged particles and deterioration in coagulation conditions, which prolonged the time of latent coagulation.

An increase in the size of coagulant particles increased the rate of latent coagulation. The size of coagulant particles had a greater influence on the rate of latent coagulation in wastewater with a low load P-PO_4 than in wastewater with a high load. In polydisperse systems, the rate of latent coagulation was determined by the degree of dispersion. A 5-fold increase in dispersion induced a 10.8-fold increase in the rate of latent coagulation.

Preheating (or stirring) influenced the rate of latent coagulation. The temperature of coagulated sewage was raised to increase particle energy and mobility, which increases the probability of contact between particles and speeds up coagulation reactions. In the simulated system, an increase in the initial velocity of sol and coagulant particles increased the rate of latent coagulation, but this parameter increased significantly only in wastewater with a low load.

Higher initial velocity of coagulant particles does not exert a significant influence on the rate of latent coagulation, therefore, the costs associated with coagulant preheating are not compensated by the resulting increase in the rate of latent coagulation.

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