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Characteristics of the dynamic distribution of suspended particles in the flocculation process*

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Abstract: Polyaluminium chloride (PAC) synthetic water was selected as a coagulant and kaolin suspension particles as objects to be removed. Online instruments such as the turbidimeter and particle counter were employed to monitor the flocculation process online and collect test data. The aim of the experiments was to study the dynamic distribution characteristics of suspension particles in the flocculation process. The 3D flow field in the reacting vessel was also simulated at different slow stirring speeds. The experiments showed that particle collision and aggregation in the flocculation process is in compliance with the Sutherland cluster aggregation model. This study further indicated that under appropriate hydrodynamic conditions, the distribution of turbulent flow in the reactor could be improved to increase the odds of effective particle collision and restrain the breakup of formed flocs by vortex shearing force. A good flocculation effect could therefore be produced.

Key words: Suspension particle, Particle counter, Flocculation process, Dynamic distribution
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INTRODUCTION

As suspension particles in water lower the quality of drinking water and damage delivery and distribution channels, they can have a serious impact on the water supply system and human well-being. The content of suspension particles in water is usually used as an important parameter to evaluate the results of water treatment, monitor the optimization of hydrodynamic conditions and measure the water quality of outflow (Luo *et al.*, 2000; Yang *et al.*, 2003). During water treatment, it is critical to remove the suspension particles from water at large.

In the water treatment industry, flocculation techniques are generally adopted to turn a large number of small particles into a smaller number of larger ones. However, the flocculation of particles depends on collisions, and the reactor should provide appropriate hydrodynamic conditions to stimulate such collisions. Once a coagulant is added, the flocculation process can be divided into two stages: rapid mixture and flocculating reaction. Rapid mixture is needed to ensure that the coagulant disperses rapidly and evenly into raw water so that it is evenly hydrolyzed and polymerized to cause the colloid particles to destabilize and aggregate. It requires swift and intense stirring by applying hydraulic or mechanical power and should be completed in a short time. The flocculating reaction requires an appropriate stirring intensity and flow velocity to prevent the breaking of formed particles and needs some time to allow the flocs to agglomerate progressively (Wang and Tambo, 2000).

The removal result in subsequent processes and the ultimate quality of outflow are directly related to the quality of floc particles, among which only those of the right size and viscosity can withstand the shear force of subsequent vortex flows and produce good

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treatment results. One key aspect of water treatment is to study the distribution characteristics of the suspension particles with a particle counter. This has proven to be an effective instrument due to its ability to provide an immediate estimate of the particle population and floc size as well as displaying their microscopic characteristics (Cui *et al.*, 2004; Bushell and Amal, 2000).

Flocculation is a chaotic and unstable process affected by physical and chemical factors in which particles collide and aggregate randomly showing non-linear characteristics. The development of non-linear sciences brought about a number of models for studying the floc collision-aggregation process including the vold ballistic aggregation model (VBAM) and the Sutherland cluster aggregation model (SCAM). VBAM considers floc formation to be a result of random superposition of primary particles along a linear path. The particles adhere to the enlarging floc at their original contact points and merge into the floc without internal reassembly. Through further aggregation, the flocs establish a third level floc structure and come to form palpable particles that can settle rapidly. Unlike VBAM, SCAM considers the growth of flocs to be a result of collision and aggregation among flocs that contain different numbers of particles rather than that of the adherence of single particles. Disregarding floc breakup, a regular flocculation process is to grow ever larger clusters out of small ones that are formed by primary particles (Sutherland, 1967). Such models have provided a scientific foundation for studying the flocculation process microscopically.

The hydraulic conditions in the reacting vessel will directly impact on the formation of flocs and the quality of outflow. The result of water flow and energy dissipation under different hydrodynamic conditions can be visualized with simulated 3D flow field in the reactor using FLUENT, the most widely used commercial Computational Fluid Dynamics (CFD) software. It is an intuitive and reliable method for studying the impact of different hydrodynamic conditions on the flow structures and flocculation process as well as describing and explaining such sophisticated processes (Park *et al.*, 2001).

This study adopted the particle counting technique to monitor online variation of the dynamic distribution of suspension particles in the flocculation process, to study how different doses and hydrodynamic conditions would affect the distribution of suspension particles and to understand their innate principles. The study may provide valuable guidance on how to develop the mechanism and process of flocculation in water treatment and may have significance in scientific research and the development of engineering industry.

MATERIALS AND METHODS

Instruments and reagents

Instruments used in this study included a PCX2200 particle counter (HACH, USA), a MICRO TOL online turbidimeter (HF, USA), an RW20 N-type multiple velocity agitator (IKA, Germany) (7 cm diameter impeller at a distance of 6 cm to the bottom surface of the reacting vessel), a rectangular reacting vessel (18 L effective volume, 28 cm long, 28 cm wide and 23 cm effective height).

A conventional flocculation experiment is to sample water at selected locations and measure the water quality offline with analytical instruments. However, such a method may reproduce the results poorly since the sampling locations and instrument readings may be manly intervened. The uniqueness of this study is that the amount of suspension particles in water and the particle distribution were monitored with online instruments. The water samples, collected at a fixed sampling mouth, flowed continuously over the instruments and the parameter values of the water quality were recorded online. Contrary to the conventional experiment, the results of such an experiment are rarely affected by subjectivity and their immediacy and reproducibility can be guaranteed.

All experimental results were obtained by reading the online instruments and their accuracy was determined by the accuracy of the instruments. The precision of the PCX2200 particle counter and the MICRO TOL online turbidimeter can reach 4-bit and 5-bit respectively (Wang and Chen, 2003).

Kaolin suspension water was used in experiments (kaolin: SiO_2: 46%, w/w; Al_2O_3: 39%, w/w; calcination loss: 20%, w/w) and polyaluminium chloride (PAC: analytical reagent, content>99.0%, w/w; pH=4.9; basicity: 82.4%, w/w; water insolubility: 0.95%, w/w) was selected as the coagulant.

Preparation of coagulant and water samples

First, PAC was made into 1% synthetic water and kaolin clay was soaked for some time before the experiment. Next, the clay was mixed with tap water in a rectangular vessel to make 18 L of suspension water with a turbidity of 100 NTU. The turbidimeter and the particle counter were then turned on to collect samples at a speed of 100 ml/min. When the turbidity reading of the samples stabilized, the experiments started in the order of coagulant addition, rapid stirring, slow stirring and sedimentation.

Method

The flocculation experiment used the stirring method to study the dynamic distribution characteristics of suspension particles in the flocculation process. The experiment was performed in the following steps:

(1) In the flocculation process, the dose is important in determining the flocculation effect. Different doses will bring about different flocculation effects and particle distributions. Studying the dynamic distribution of suspension particles in the flocculation process under different doses helps us to explore the properties of the flocculation reaction from a microscopic perspective. To identify the optimal hydrodynamic conditions in water flows, it is essential to first decide at what coagulant dose (or optimal dose) the best water quality can be produced. A flocculation experiment is generally adopted by the water treatment sector (Yukselen and Gregory, 2004) to determine the optimal dose. In this experiment, we used the residual turbidity at 20 min after sedimentation to evaluate the results and the infinite approximation method to determine the optimal dose. The doses ranged from 5 to 100 mg/L. The experiment first identified the lowest value of residual turbidity when the PAC dose was 10 mg/L and then approximated to the optimal dose by progressively increasing the dose. The experiment indicated that the lowest residual turbidity after sedimentation and the best coagulation effect was found at a PAC dose of 10 mg/L.

(2) An orthogonal experiment was employed to achieve the optimization of stirring conditions. With experiments properly designed, high quality and highly reliable data can be obtained to help us understand the inherent correlations among experimental variables and identify the optimal parameters and techniques. In this experiment, we used the 10 mg/L PAC dose and residual turbidity at 20 min after sedimentation as parameters to evaluate the results. We divided the stirring speed into rapid and slow levels and selected four segments, i.e., rapid stirring speed, rapid stirring time, slow stirring speed and slow stirring time, which were sub-divided into three levels (Table 1), to identify the optimal hydrodynamic conditions. Table 2 shows the schemes and experiment results.

It is known that ratio (*R*), is an important factor for evaluating the degree of data fluctuation. The largest value of *R* corresponds to the most important factor. The impact of stirring conditions on the flocculation effect was evaluated in the order of slow stirring speed, rapid stirring speed, rapid stirring time and slow stirring time (Table 2). Slow stirring speed had the greatest impact. Comparing the values of k_{mean} corresponding to each segment at each level, we

Table 1 Factors and levels of the orthogonal experiment

Level	RSS (r/min)	RST (s)	SSS (r/min)	SST (min)
1	200	10	60	10
2	250	20	90	15
3	300	30	120	20

RSS: rapid stirring speed; RST: rapid stirring time; SSS: slow stirring speed; SST: slow stirring time

Table 2 Schemes and results of the orthogonal experiment

Experiment	RSS (r/min)	RST (s)	SSS (r/min)	SST (min)	RT (NTU)
1	200	10	60	10	17.31
2	200	20	90	15	21.37
3	200	30	120	20	31.70
4	250	10	90	20	16.55
5	250	20	120	10	31.07
6	250	30	60	15	16.78
7	300	10	120	15	31.59
8	300	20	60	20	17.98
9	300	30	90	10	21.52
k_1	70.38	65.45	52.07	69.90	
k_2	64.40	70.42	59.44	69.74	
k_3	71.09	70.00	94.36	66.23	
$k_{1\text{mean}}$	23.46	21.82	17.36	23.30	
$k_{2\text{mean}}$	21.47	23.47	19.81	23.25	
$k_{3\text{mean}}$	23.70	23.33	31.45	22.08	
R	2.23	1.65	14.09	1.22	

RSS: rapid stirring speed; RST: rapid stirring time; SSS: slow stirring speed; SST: slow stirring time; RT: residual turbidity found the optimal stirring conditions of 250 r/min rapid stirring speed, 10 s rapid stirring time, 60 r/min slow stirring speed and 20 min slow stirring time.

(3) Under the optimal hydrodynamic conditions, PAC dose was varied from 3 to 40 mg/L to study the variation in the dynamic distribution of suspension particles.

(4) The results showed that the flocculation process was most affected by the slow stirring speed. The experiment simulated the variation of hydrodynamic conditions by changing the slow stirring speed. Under 10 mg/L PAC dose, the slow stirring speed was varied from 60 to 200 r/min to study the variation in the dynamic distribution of suspension particles.

Experimental results

A sample of clear liquid was collected at 10 cm below the free surface of the reacting vessel. The flocculation effect was evaluated using the residual turbidity at 20 min after sedimentation.

The particle counter was used to monitor the variation in the dynamic distribution of floc particles over time and to microscopically observe the process of aggregation and growth of flocs in various size ranges. Table 3 shows the channels (i.e., particle size ranges) in the particle counter.

Since kaolin suspension water shows properties similar to many rivers, it is usually used as raw water in conventional water treatment experiments (Jarvis *et al.*, 2005a). Table 4 shows the particle size distribution of raw water and settled water under PAC doses of 3, 10, 20 and 40 mg/L, respectively. After flocculation, the particle size ranges were distributed mainly in CH1~CH5 due to the small sizes of kaolin clay particles and the breakup of the particles by constant sampling. Repeated monitoring of different stages of the flocculation process revealed similar changes (Table 4). Thus, only variation of particle populations in CH1~CH5 were studied in subsequent experiments.

Table 3 Particle size ranges in particle counter

	Channel					
	CH1	CH2	CH3	CH4	CH5	CH6
Min size (μm)	2	5	10	18	25	40
Max size (μm)	5	10	18	25	40	750

Table 4 Particle size distribution of water sample (counts/ml)

Experiment	Channel					
	CH1	CH2	CH3	CH4	CH5	CH6
Raw water	8921	9673	4852	98	11	5
Settled water at 3 mg/L PAC dose	5153	4843	4023	354	47.2	4
Settled water at 10 mg/L PAC dose	2167	1752	1134	127	35	7
Settled water at 20 mg/L PAC dose	3654	2957	2108	243	41	6
Settled water at 40 mg/L PAC dose	5083	4013	2463	95.4	5.6	2

RESULTS

Variation of suspended particle distribution in the flocculation process

In the stirring experiment, PAC dose was changed under optimal hydrodynamic conditions. Fig.1 shows the variation in suspended particle distribution in the flocculation process.

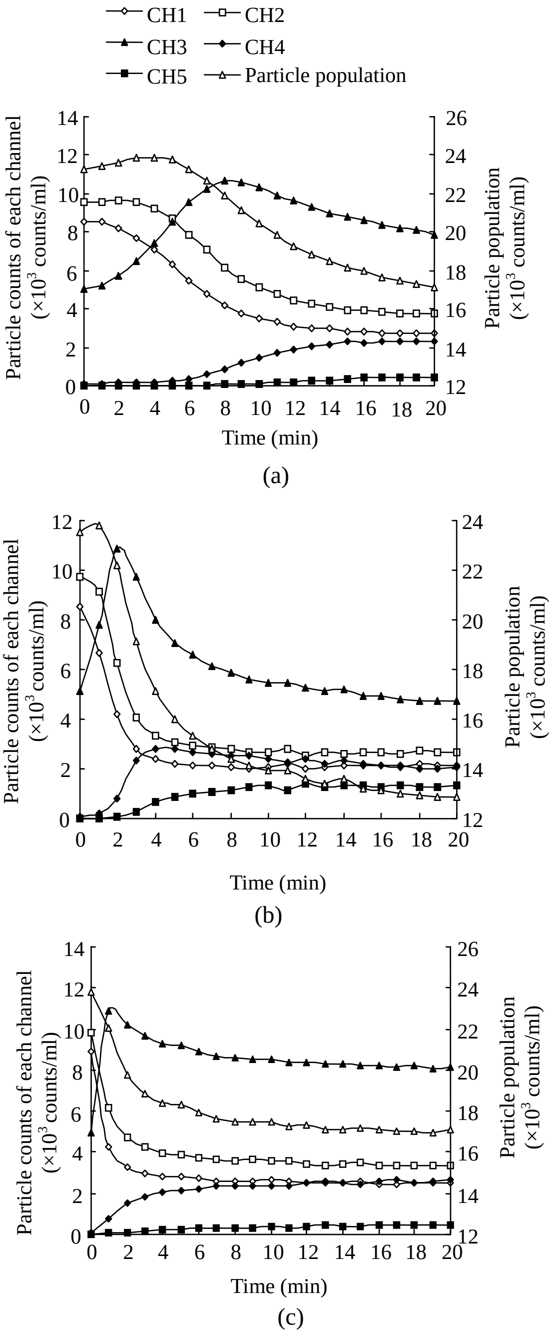
Suspension particles aggregated rather rapidly in the early stages of flocculation (Fig.1) such that small particles decreased sharply and large ones increased dramatically. However, the aggregation slowed down over the reacting time. By studying the variation in particle populations in CH1~CH5, we concluded that large flocs did not aggregate directly from primary particles. Instead, they were formed over time through binding of small flocs and primary particles.

Such a process is in compliance with SCAM. The curve of particle population in Figs.1a~1c also demonstrated that the particle population was decreasing throughout the flocculation process as many small suspension particles were gradually turning into large particles.

Impact of dose on residual particle distribution

Dose was varied under optimal hydrodynamic conditions. Fig.2 shows the variation in residual particle distribution and residual turbidity at 20 min after sedimentation.

When the dose was changed from 3 to 10 mg/L, the residual turbidity decreased as coagulant increased. The particle population of each channel declined noticeably and the flocculation effect improved. However, when PAC dose was increased above 10 mg/L, the flocculation effect became poor due to excessive coagulant addition and both the residual



(a)

(b)

(c)

Fig.1 Variation in suspended particle distribution over time. (a) 3 mg/L PAC dose; (b) 10 mg/L PAC dose; (c) 35 mg/L PAC dose

50 r/min rapid stirring speed, 10 s rapid stirring time, 60 r/min slow stirring speed, 20 min low stirring time

turbidity and particle population of each channel increased. Under the dose of 10 mg/L, both the residual turbidity and the particle population reached their lowest values and the flocculation effect was maximized.

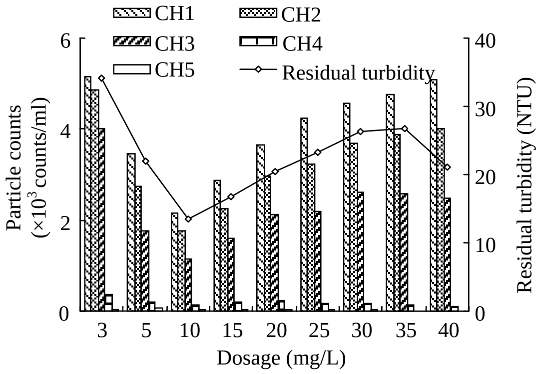


Fig.2 Variation in residual particle distribution and residual turbidity with different PAC doses under optimal hydrodynamics

250 r/min rapid stirring speed, 10 s rapid stirring time, 60 r/min slow stirring speed, 20 min low stirring time

The PAC dose affects not only the function of charge neutralization and the absorption bridging mechanism but also the sufficiency of flocculating cores in treated water that supposedly work to stimulate effective flocculation. In other words, with the same water quality and under the same hydrodynamics, a little PAC could instantly distribute evenly in water. Due to the small number of flocculating cores in water, there would be few free particles to be caught, making it harder for small particles to bump into each other and clump together into larger ones. Consequently, the flocs would show poor settling properties and high residual turbidity.

Under the optimum dose, the relative number of flocculating cores increases, causing an increase in aggregation points and the formation of large-size, high-density floc particles with good settling properties (Tang *et al.*, 2005). When the dose is further increased, PAC, as an inorganic polymer coagulant, forms a protective coating over the exterior surface of the colloid particles, making them disperse rather than coagulate. However, the size to which a floc can grow is limited under certain hydrodynamic conditions. When a floc grows excessively large, its ability to withstand the shear force becomes weaker and the floc breaks apart. Large flocs with low intensity are broken into small ones, causing small particles to increase and large ones to decrease. As a result, the removal of sediment becomes more difficult and both the residual turbidity and the particle population of each channel increases.

Although residual turbidity was almost the same at 5 and 40 mg/L (21.89 and 21.13 NTU, respectively), the particle population of the respective channel was remarkably different (Fig.2). Turbidity is an optical measurement that is used to evaluate the impact of suspension particles on light due to various densities, sizes, shapes, colors and surface characteristics, whereas a particle counter is used to calculate the number of suspension particles in water and measure their sizes. Research suggests that particles in different size ranges produce different values of turbidity (Ceronio and Haarhoff, 2005). When the numbers of particles with low production values are too high or too low, they will not substantially affect the turbidity. This explains why residual turbidity remained the same when residual particle distribution was remarkably different.

Fig.3 shows the relationship between the residual particle population and residual turbidity under different doses and the same optimal hydrodynamics.

The residual particle population was related to residual turbidity under selected doses (correlation coefficient, R^2=0.8671).

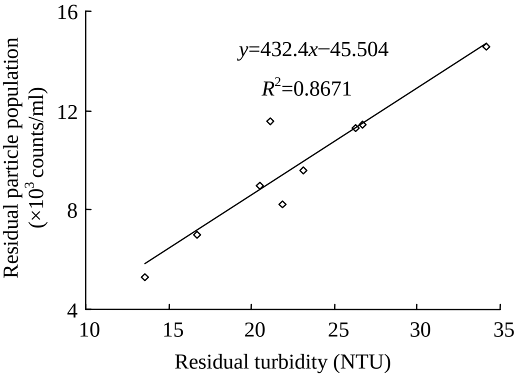


Fig.3 Relationship between residual particle population and residual turbidity under different doses

250 r/min rapid stirring speed, 10 s rapid stirring time, 60 r/min slow stirring speed, 20 min low stirring time

Impact of hydrodynamics on residual particle distribution

Experiments were also carried out to study the effect of varying the slow stirring speed under the optimal dose. Fig.4 shows the variation in residual particle distribution and residual turbidity 20 min after sedimentation.

The flocculation effect is largely determined by particle characteristics and hydrodynamic conditions.

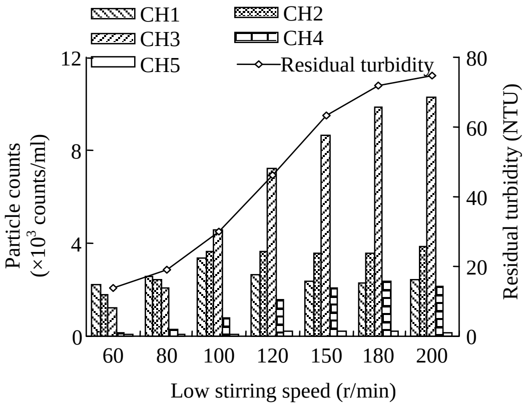


Fig.4 Variation in residual particle distribution and residual turbidity at slow stirring speed under optimal PAC dose

10 mg/L PAC dose, 250 r/min rapid stirring speed, 10 s rapid stirring time, 20 min low stirring time

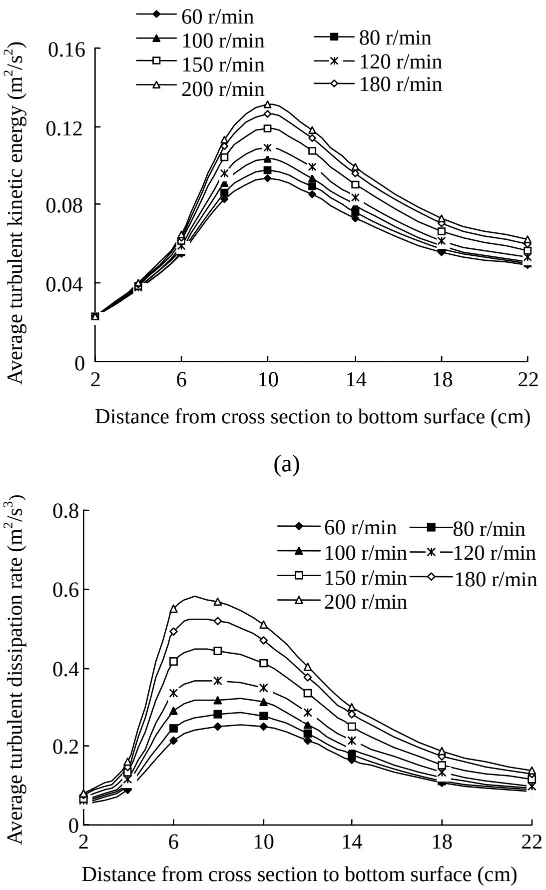
Fig.4 shows that variation in hydrodynamics has a major impact on the residual turbidity. When the slow stirring speed changed from 60 to 200 r/min, the residual turbidity started increasing and the particle population of each channel increased, particularly in CH3. The addition of PAC would cause flocs of various size ranges to form in water. At an appropriate slow stirring speed, the chances for particle collision would increase. Small particles would be stimulated to aggregate into large ones so that large formed flocs could be protected from excessive damage. When particle aggregation and breakup reaches a dynamic balance, a small number of large-size, high-density particles that could gradate and settle down well would remain in water and better withstand the destructive force of subsequent processes, facilitating the removal of suspension particles at large during subsequent solid-liquid separation and improving the quality of outflow (Jarvis *et al.*, 2005b).

To further evaluate the impact of hydrodynamic conditions on the flocculation effect, FLUENT was employed to simulate the 3D flow field in the reacting vessel under different slow stirring speeds. The experiment used a rectangular vessel that could effectively control the flow co-rotation along the impeller with flow resistance. Some part of the flow would bump into the walls of the vessel and form reverse counterflow, increasing the chances of collision and adherence among floc particles. Meanwhile, at a certain slow stirring speed, turbulent fluctuation would moderate facilitating floc growth in the vessel. Since flocs in dead zones are unable to be involved in major flow circulation, they would eventually aggregate in small sizes with poor settling properties and the flocculation effect would be affected. However, as the slow stirring speed increases, such phenomena would effectively improve.

Based on the micro-vortex theory, the flocculation effect is determined by how much energy can be turned into effective energy at each flocculation stage rather than how much energy the main flow can possess in the reactor. The smaller the eddy size of the turbulent region, the greater the viscous force and the pulsating deformation, as well as the deformation work and coefficient of effective energy dissipation. Thus, the turbulent kinetic energy and the turbulent dissipation rate were used to indicate the turbulence in the flow field and the degree of impact of the flow field on the flocculation effect. Turbulent kinetic energy refers to kinetic energy per unit mass, a measurement of the intensity of speed fluctuation. The turbulent dissipation rate is the dissipation rate of kinetic energy per unit mass, which is used to indicate the speed when small scale vortices in homogeneous isotropic turbulence transfer mechanical energy into thermal energy. Fig.5a and Fig.5b show the distributions of mean turbulent kinetic energy and mean turbulent dissipation rate, respectively, at different distances from the cross section to the bottom surface of the vessel. In this study, the distance between the impeller and the bottom surface was 6 cm.

Variation in the slow stirring speed had a major impact on the water flow around the impeller (Fig.5a and Fig.5b). As the speed increased, the differences in the distribution of mean turbulent kinetic energy and mean turbulent dissipation rate within different distance ranges became more pronounced. Mean turbulent kinetic energy and mean turbulent dissipation rate in the vicinity of the impeller (at distances of from 6 to 14 cm) increased noticeably while those in distant zones remained low and changed only moderately. Fig.6 shows clearly the impact of slow stirring speed on the process of floc growth.

At a particular slow stirring speed, the residual particle population tended to stabilize at a certain level in the later flocculation stage and increased gradually as the slow stirring speed increased (Fig.6). Comparing the variation of the impact of slow stirring speed on the flow turbulence around the impeller with FLUENT results showed that, at a certain slow stirring speed, the energy input in the water remained



(a)

(b)

Fig.5 Distribution of (a) mean turbulent kinetic energy and (b) mean turbulent dissipation rate at different slow stirring speeds in the reactor

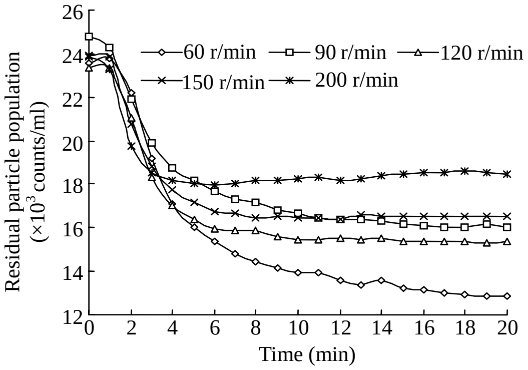


Fig.6 Variation in the residual particle population size at different slow stirring speeds over time

10 mg/L PAC dose, 250 r/min rapid stirring speed, 10 s rapid stirring time, 20 min low stirring time

unchanged and a flow structure was established in the vessel. The particles continued to collide and aggregate under the flow turbulence. When the particles aggregated into larger flocs, small scale vortices would shear them powerfully. In particular, the turbulent force around the impeller broke the formed particles and prevented the aggregation of new flocs until the particle aggregation and breakup reached a dynamic balance and the particle population stabilized at a certain level. As the slow stirring speed increased, turbulent kinetic energy and turbulent dissipation rate in the reactor (especially around the impeller) increased accordingly. Since the eddy size is determined by effective energy dissipation, the smaller the eddy size, the more powerful the vortex and the greater the shear force, causing more damage to the flocs and more floc particles to break into small ones. As a result, the particle population increased when the stirring stopped.

Fig.7 shows the relationship between residual particle population and residual turbidity under different hydrodynamics. At the optimal dose and selected slow stirring speeds, the residual particle population was closely related to the residual turbidity (R^2=0.9399).

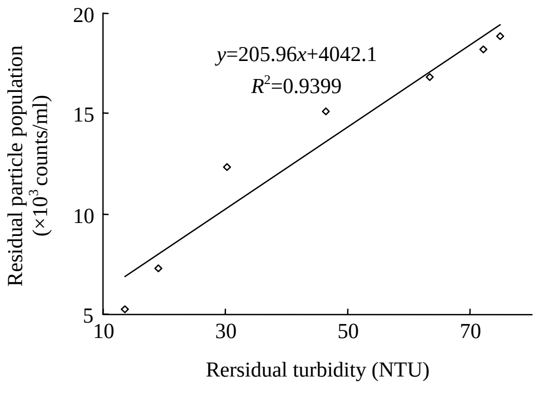


Fig.7 Relationship between residual particle population size and residual turbidity at different slow stirring speeds
10 mg/L PAC dose, 250 r/min rapid stirring speed, 10 s rapid stirring time, 20 min low stirring time

Fig.3 and Fig.7 both show the relationship between the residual turbidity and the particle population. As stated above, since the two parameters have different bases, they will not be absolutely equivalent when describing the characteristics of water quality. Since the two parameters have different bases, they will not be absolutely equivalent when describing the characteristics of water quality.

In summary, flocculation is a complicated physical-chemical process in which floc particles constantly collide with each other, aggregate and become sheared and dispersed in water. A comprehensive consideration of particle aggregation and breakup will be essential for the optimization of hydrodynamic parameters to stimulate the growth of flocs strongly bound together with excellent settling properties and to improve the flocculation effect and quality of outflow.

CONCLUSION

(1) By monitoring the flocculation process online with a particle counter, this study found that large flocs did not aggregate directly from primary particles. Instead, they were progressively formed by the binding of small flocs and primary particles. These findings are in compliance with SCAM.

(2) By changing doses and hydrodynamic conditions, this study showed that the quality of settled water was closely related to the dynamic distribution of suspension particles. Large-size and high density flocs with good settling properties could be formed only under appropriate dose and hydrodynamic conditions to remove the suspension particles at large and guarantee the quality of outflow.

(3) This study used FLUENT software to simulate the 3D flow field in the reacting vessel at different slow stirring speeds and successfully explained the impact of hydrodynamics on the distribution of suspension particles based on experimental results.

(4) There was a good relationship between the residual particle population and residual turbidity. The particle counter could be used as a useful supplement to the turbidimeter. Combined use of the turbidimeter, characterized by economic benefits and practical application, and the particle counter, characterized by high sensitivity and comprehensive uses, efficiently reveals the results of water treatment and guarantees water quality. Nowadays, as the requirements for water quality are becoming more stringent, it is expected that the particle counter will be more widely used and that the particle population will become one of the key parameters for evaluating the

quality of drinking water (Wang *et al.*, 2006).

(5) How the particles of various size ranges affect turbidity values will be studied further. A quantitative description will be developed that may be widely applicable in scientific research and in the engineering industry.

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