UNSTEADY SETTLEMENT TEST AND NUMERICAL CALCULATION OF ACTIVATED CARBON PARTICLES IN STATIC WATER

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ABSTRACT

Powdered activated carbon (PAC) has strong adsorption capacity because of its large specific surface area and developed pore structure. It is currently used in advanced treatment of industrial sewage. In the sewage treatment technology using activated carbon, the settling velocity of activated carbon in static water is an important parameter. Based on the process and results of the static water settlement test of powdered activated carbon and combining the physical properties of powdered activated carbon, it was found that the powdered activated carbon does not settle in the static water simply as a single particle, but settles in the form of flocs due to effect of flocculation. Activated carbon flocs are accompanied with flocculation and diffusion when settling, and the change of the intensity of flocculation and diffusion results in the change of the equivalent size of flocs with time, thus affecting the settling characteristics of flocs. Therefore, the hypothesis of "flocculation-diffusion" is put forward when activated carbon particles settle, and the fitting formulas of activated carbon flocs equivalent particle size changed with time are given according to experimental data. Based on BBO equation of unsteady particle movement, the settling process of activated carbon particles in static water is numerically simulated. The calculated results are in good agreement with the experimental values.

Keywords: activated carbon particles; unsteady settlement; flocculation and diffusion; numerical calculation

1. INTRODUCTION

Powdered activated carbon is widely used in the advanced treatment of industrial wastewater because of its large surface area and well-developed pore structure. And the settling velocity of activated carbon in static water is an important parameter in many technical treatments of industrial wastewater using activated carbon. The particle size of powdered activated carbon is very small, so when settling in static water, it is influenced by the turbulent action of flow and the movement of Brownian (Liu et al, 2019), which makes activated carbon particles settle not in the form of simple single particles but flocs. Therefore, the parameters such as equivalent particle size and equivalent density of flocs have become important factors affecting their settlement in static water. There are abundant researches on the settlement characteristics of fine cohesive sediment. Jin et al (2002) confirmed the influence of salinity, sediment concentration, mineral characteristics of sediment and other factors on the flocculation of fine cohesive sediment by experiments and analysis. Chen (2013) studied the relationship between the settling velocity in static water of fine cohesive sediment in Changjiang Estuary and water temperature, salinity and sediment concentration by using new type of settling tube and measuring instrument. Guo et al (2019) studied the effect of sediment concentration and turbulent intensity of flow on particle size and effective density of fine cohesive sediment flocs in annular flume by using high-power camera, and furthermore studied their influence on the settling velocity. Based on the similarity of settlement characteristics between fine cohesive sediment and activated carbon, the static water settling characteristics of activated carbon particle group were studied in this paper according to the different physical properties of activated carbon and sediment particles. From the results of experimental observation, it is found that the equivalent particle size of activated carbon particles varies from time to time because of the flocculation and diffusion, and the fitting formula of "flocculation-diffusion" of the equivalent particle size of activated carbon particles was proposed. Therefore, the characteristics of unsteady settlement of the two kinds of powdered activated carbon with and without adsorbing pollutants were numerically studied.

2. MEASUREMENT OF PHYSICAL PROPERTIES OF ACTIVATED CARBON AND THEORETICAL CALCULATION OF THE SETTLING VELOCITY

Activated carbon and fine grained sediment have certain similarity to each other, so the parameters of fine grained sediment are equally important for activated carbon particles, such as median particle size, density and so on. However, because activated carbon particles are porous media, they have fully developed and complex pore structure. After absorbing pollutants and putting them into water, some pollutants and water will be filled in the pore, which makes the equivalent density of activated carbon flocs when settling in water is the weighted mixture density of carbon, water and pollutants. Therefore, for powdered activated carbon, it is necessary to measure its pore volume, tapped density (i.e. mass per unit volume after vibration by instrument) and true density (i.e. mass per unit volume of solid after removal of internal pore volume).

2.1 Physical parameter testing

The particle size gradation of activated carbon particles adsorbing and without adsorbing pollutants was measured by Microtrac S3500 particle size analyzer. The total pore volume of the two activated carbons was measured by gas adsorption using JW-BK122W surface and pore size distribution analyzer. The tapped density of two kinds of activated carbon was measured by BT-301 tapped density tester. True density of the two activated carbons was measured by gravity bottle method. Relevant studies have shown that when activated carbon settles in water, its pores are almost filled with water and the moisture content is close to 100%. Based on this, the equivalent density of the two activated carbon particles in water can be obtained. The results are shown in Table 1.

Table 1. Physical parameters of the two particles

	ACTIVATED CARBON PARTICLES ADSORBING POLLUTANTS	ACTIVATED CARBON PARTICLES WITHOUT ADSORBING POLLUTANTS
AVERAGE PARTICLE SIZE d50/μm	22.41	33.67
TOTAL PORE VOLUME/(cm^3/g)	0.0722	0.1032
TAPPED DENSITY/(g/cm^3)	0.735	0.766
TRUE DENSITY/(g/cm^3)	2.283	1.386
EQUIVALENT DENSITY OF STATIC WATER SETTLEMENT/(g/cm^3)	2.215	1.355

2.2 Theoretical calculation of final settling velocity

Through theoretical and experimental studies, some scholars have put forward various theoretical and empirical formulas for calculating sediment settling velocity (Zhang, 2008), such as

Stokes formula

$$\omega = \frac{1}{18}\frac{\rho_s - \rho}{\rho} g \frac{d^2}{\nu} \tag{1}$$

Zhang R.J.-Hasen-William-Clay formula

$$\omega = \sqrt{\left(13.95\frac{\nu}{d}\right)^2 + 1.09\frac{\rho - \rho_s}{\rho} gd} - 13.95\frac{\nu}{d} \tag{2}$$

and Sha Y.Q. formula, which is for laminar flow (d<0.1mm)

$$\omega = \frac{1}{24}\frac{\rho_s - \rho}{\rho} g \frac{d^2}{\nu} \tag{3}$$

According to the data in table 1, the theoretical final settling velocity of single particle of activated carbon was separately calculated using above formulas respectively, where the particle size takes the median particle size d50 and the density takes the equivalent density. The results are shown in Table 2.

Table 2. The theoretical settling velocity of the two kind of activated carbon particles (unit: cm/s)

	STOKES	ZHANG R.J.- HASEN-WILLIAM-CLAY	SHA Y.Q.
ADSORBING POLLUTANTS	0.019	0.014	0.014
WITHOUT ADSORBING POLLUTANTS	0.029	0.021	0.022

After inspection, the results in Table 2 are in accordance with the conditions for each formula.

3. ACTIVATED CARBON SETTLEMENT TEST

This experiment adopts time-displacement method. The diagram of experimental device is shown in Figure 1.

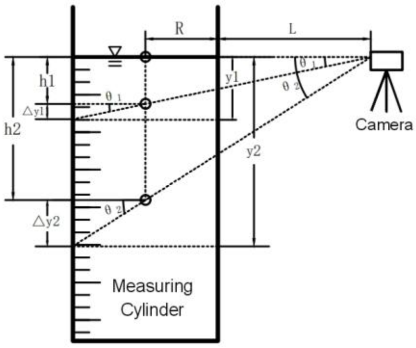


Figure 1. Schematic diagram of experimental device.

Put the activated carbon into the water at the vertical direction with zero initial velocity. At the initial moment t_0, the camera lens is horizontal with the water surface, so the reading of the position coordinate of the activated carbon is consistent with the actual position coordinate and recorded as y_0. However, with the settlement of activated carbon, it will have a height difference with the camera lens, which makes the reading of the position coordinate of the activated carbon have an error with the actual position coordinate. To eliminate this error, consider the following amendments:

$$\tan\theta_a = \frac{y_a - y_0}{L + 2R} \tag{4}$$

where L is the minimum distance between the camera lens and the wall of the measuring cylinder.

Ignore the thickness of the cylinder wall and according to triangle similarity, we can see that the modified value of activated carbon in this position coordinates is

$$\Delta y_a = R\tan\theta_a \tag{5}$$

where R is the radius of the water surface in the measuring cylinder.

Therefore, the actual settling height of activated carbon at ta moment is

$$h_a = y_a - \Delta y_a \tag{6}$$

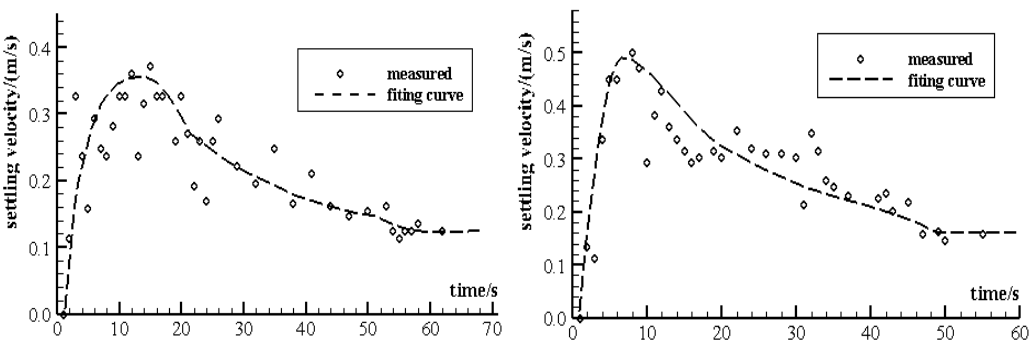
Similarly, the actual settling height h_b of activated carbon at t_b moment is obtained.

Therefore, the average settling velocity ω_{ab} of activated carbon during the t_a and t_b is

$$\omega_{ba} = \frac{h_b - h_a}{t_b - t_a} \tag{7}$$

When the time between t_a and t_b is short enough, the average settling velocity can be approximated as instantaneous settling velocity.

During the experiment, the two kinds of activated carbon powders were respectively prepared into aqueous suspension of certain concentration, and drop it into the water at the initial vertical velocity of 0. Record the settling process by camera, and intercept the image of the activated carbon liquid cluster at different moment. Using WinDIG software to read the position coordinates in the vertical direction of the activated carbon liquid cluster, and the average velocity in the time interval is approximated as its instantaneous settling velocity at different moments. Based on this, the settling velocity - time curve can be made to analyze the characteristics of the settling velocity of powdered activated carbon with time in static water. The curve is shown as Figure 2.



(a)Activated carbon without adsorbing pollutants (b)Activated carbon adsorbing pollutants

Figure 2. Settling velocity-time curves of two kinds of activated carbon.

Relevant studies (Yin et al, 2016; Huang and Wei, 2003) have shown that the settlement of particles in static water is variable accelerated motion, and the settling velocity of particles increases with time, but the amplitude of the increase becomes smaller and smaller, and finally reaches the state of near uniform settlement, which is different from the results of this experiment. As is shown in Figure 2, the settling velocity increases with time firstly, reaching a large value and then slowing down, while finally reaches a state close to uniform motion. In addition, the final uniform settling velocities of the activated carbon that adsorbing and without adsorbing pollutants obtained from the test are about 0.12 cm/s, 0.16 cm/s respectively, which are also quite different from the theoretical values in Table 2.

According to the studies on flocculation of fine sediment particles (Guo et al, 2019), it is shown that the settling speed of the flocs is about 6-28 times of that of the single particle, which is close to the multiple of the experimental value and the theoretical value. And it can be seen from the Stokes formula that the equivalent particle size of the flocs has a great influence on its settling velocity. At the same time, it is found that the diffusion phenomenon of activated carbon aqueous suspension settling in water is accompanied by the change of local concentration, which will cause the change of floc particle size (Cheng et al, 2007). Therefore, it can be considered that the powdered activated carbon is not a simple single particle settlement when settling in water, but in the form of flocs, and the equivalent particle size of flocs is not constant during the settling process, but changes with time.

According to the Stokes formula, the equivalent particle size of the two kinds of activated carbon flocs can be inversely calculated from the test value of the final uniform settling velocity, where the density takes the equivalent density, and the kinematic viscosity coefficient takes $\nu=1.140\times10^{-6}m^2/s$ according to the water temperature at the time of the test. The results are shown in Table 3.

Table 3.Equivalent particle size for settlement of two activated carbon flocs

	EQUIVALENT PARTICLE SIZE/μm
WITHOUT ADSORBING POLLUTANTS	85.95
ADSORBING POLLUTANTS	51.85

They all meet the applicable conditions of Stokes formula after testing. The equivalent particle sizes of the two activated carbon flocs adsorbing and without adsorbing pollutants are 2.55 and 2.31 times of the median particle size of the single particle respectively. Considering the similar concentration of activated carbon aqueous suspension, the flocculation intensity of activated carbon particles is similar.

4. NUMERICAL SIMULATION OF UNSTEADY SETTLEMENT OFACTIVATED CARBONFLOCS

4.1 Introduction to the mathematical models

4.1.1 Equation of non-constant motion of particles under the condition of Stokes flow

Assuming that the particle moves transiently in the boundless flow field with a moving velocity of u(t), and the absolute velocity of the particle is equal to the relative velocity of the particle to flow field if fluid is static. When the flow field satisfies the Stokes flow condition, i.e. the particle Reynolds number Rep<<1, the equation of motion of the particle moving at any velocity $u_p(t)$ in the flow field can be expressed by Eq.(8), i.e. the generalized BBO equation (Huang et al, 2000).

$$\frac{\pi}{6}D^3\rho_s\frac{du_p}{dt}=3\pi\mu D\left(u-u_p\right)+\frac{\pi}{6}D^3\rho_f\frac{du}{dt}+\frac{\pi}{12}D^3\rho_f\frac{d\left(u-u_p\right)}{dt}+\frac{3}{2}D^2\sqrt{2\rho_f\pi\mu}\int_0^t\frac{d\left(u-u_p\right)/d\tau}{\sqrt{t-\tau}}d\tau+F_p \tag{8}$$

where u, u_p denote the velocity of the fluid and particles; ρ_f, ρ_s denote the density of the fluid and particles; D denotes the particle size; and μ denotes the dynamic viscosity coefficient of the fluid. The first four terms on the right side of the equal sign denote the quasi-constant Stokes viscous resistance F_s, buoyancy F_f, additional mass force F_m, Basset force F_B, respectively.

4.1.2 Non-constant motion equation of particles under the condition of high particle Reynolds number

Eq. (8) ignores the convection term in the N-S equation so that this equation holds only under the Stokes flow condition. Therefore, under the condition of high particle Reynolds number, the effect of convection term must be considered to correct this equation. If the gravity as well as Saffman force (shear lift caused by the non-uniformity of the flow field) are considered (Tsuji et al, 1991), the transient equation of the particles at the high Reynolds number is shown in Eq. (9).

$$\begin{aligned}\frac{\pi}{6}D^3\rho_s\frac{du_p}{dt}&=\frac{\pi}{6}D^3\left(\rho_s-\rho_f\right)f+c_1 3\pi\mu D\left(u-u_p\right)+\frac{\pi}{6}D^3\rho_f\frac{du}{dt}+\Delta_A\frac{\pi}{6}D^3\rho_f\frac{d\left(u-u_p\right)}{dt}\\&+\Delta_H\left(\frac{D}{2}\right)^2\sqrt{2\pi\rho_f\mu}\int_0^t\frac{d\left(u-u_p\right)/d\tau}{\sqrt{t-\tau}}d\tau+C_{LS}k_s\mu D^2\left(\frac{\xi}{\nu}\right)^{0.5}\left(\nu-\nu_p\right)\end{aligned} \tag{9}$$

where

$$\Delta_A = 1.05 - \frac{0.066}{A_c^{\,2} + 0.12}, \quad \Delta_H = 2.88 + \frac{3.12}{(A_c + 1)^3}, \quad A_c = \frac{(u - u_p)^2}{D\left|d(u - u_p)/dt\right|} \tag{10}$$

where k_s denotes the coefficient of Saffman lift force; v, v_p denote the transverse velocity component of the fluid and the particles in the vertical direction of u, u_p; ζ denotes the transverse velocity gradient of the fluid; and the C_{LS} is the correction coefficient. Δ_A, Δ_H denote the non-constant resistance correction coefficient (Odar and Hamilton, 1964).

4.2 Flocculation-diffusion model of equivalent particle size

Assuming that the particle density in the settlement process is constant, based on the change of the settlement velocity shown by the experimental results, we proposes that there is also a process that the particle size grows from small to large and becomes small again, then eventually becomes constant.

The possible reasons for this process are that the activated carbon suspensions are more turbulent after full stirring, so the flocculation is weak, and the particle size of flocs is at a smaller value. Due to the decrease of the turbulent action and the large local concentration, the flocculation is enhanced, and the equivalent particle size of the flocs becomes larger. With the diffusion in the settlement process, the local concentration becomes smaller, the flocculation is weakened, and the equivalent particle size of flocs becomes smaller. When the flocculation and diffusion are in equilibrium, the particle size of the flocs will no longer change.

In this paper, the equivalent particle size of the flocs is assumed to be polynomial with time. According to the experimental data in last section, the functional relations of the equivalent particle size D_1 and D_2 with time T of the two activated carbon flocs adsorbed pollutants and without adsorbed pollutants are fitted as Eq. (11) and Eq.(9).

$$D_1(T) = \begin{cases} -(1.34898\times10^{-6})T^2 + (1.888572\times10^{-5})T + 8.50\times10^{-5}, & 0 < T \le 7 \\ (1.63885\times10^{-9})T^3 - (1.40122\times10^{-7})T^2 + (1.72079\times10^{-6})T & \\ +1.45358\times10^{-4}, & 7 < T \le 50 \\ 8.595\times10^{-5}, & T > 50 \end{cases} \tag{11}$$

$$D_2(T) = \begin{cases} -(1.13889\times10^{-6})T^2 + (1.36667\times10^{-5})T + 5.10\times10^{-5}, & 0 < T \le 6 \\ (2.04305\times10^{-9})T^3 - (1.4097\times10^{-7})T^2 + (1.471\times10^{-6})T & \\ +8.78077\times10^{-5}, & 6 < T \le 40 \\ 5.185\times10^{-5}, & T > 40 \end{cases} \tag{12}$$

4.3 Numerical result

In this paper, a kind of numerical calculation method (Huang and Wei, 2003; Huang et al, 2000) is used to simulate the unsteady settlement process of the above two activated carbon particle groups. Briefly, the Eq. (9) can be changed into first order differential equation group with initial values, that is,

$$\left.\begin{array}{ll} \dfrac{dx}{dt} = u_p, & \dfrac{du_p}{dt} = g(x, t, u_p, \dfrac{du_p}{dt}) \\ x(0) = 0, & u_p(0) = 0 \end{array}\right\} \tag{13}$$

where

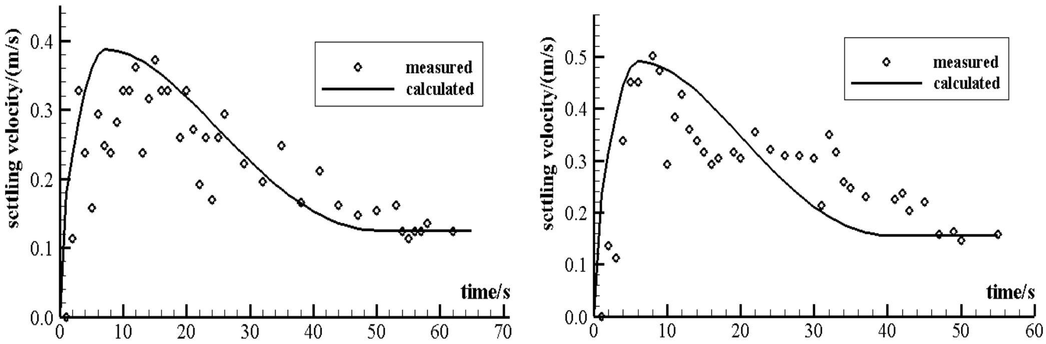
$$g(x,t,u_p,\frac{du_p}{dt}) = (1 - \rho_s/\rho_f)\ f + \frac{c_1 18\mu}{\rho_s}(u - u_p) + \frac{\rho_f}{\rho_s}\frac{du}{dt} + \Delta_A \frac{\rho_f}{\rho_s}\frac{d(u - u_p)}{dt}$$
$$+ \Delta_H \frac{3}{2\pi\rho_s D}\sqrt{2\pi\rho_f\mu}\int_0^t \frac{d(u - u_p)/d\tau}{\sqrt{t - \tau}}d\tau + \frac{6C_{LS}k_s\mu}{\pi\rho_s D}\left(\frac{\xi}{v}\right)^{0.5}(v - v_p)$$

At the first three time steps, four order Runge-Kutta formulations are used to calculate Eqs. (13) numerically. From the fourth time step on, Adams calculation scheme is employed to compute particle settling velocity step by step.

For activated carbon flocs without adsorbed pollutants, the equivalent particle size D_1(T)is taken as Eq. (11); from Table 1, the equivalent density of activated carbon without adsorbed pollutants is 1.355g/cm^3. The dynamic viscosity coefficient of water is taken as μ=1.140×10^{-3}Pa s according to the water temperature at the time of the test. The calculation step size is 2×10^{-4}s. The total time is 55s.

The equivalent particle size D_2(T) of activated carbon flocs adsorbed pollutants is taken as Eq. (12); from Table 1, the equivalent density of activated carbon adsorbed pollutants is 2.215g/cm^3. The dynamic viscosity coefficient of water is also taken as μ=1.140×10^{-3}Pa s according to the water temperature at the time of the test. The calculation step size is 1×10^{-4}s. The total time is 45s.

Numerically calculated simulation results compared with the test results are shown in Figure 3. It can be seen that the settlement process of the numerical simulation is consistent with the trend of the change of experimental values with time, and the data can be well qualitatively fitted. This indicates that the proposed "flocculation-diffusion" model and numerical calculation method can simulate the settling characteristics of activated carbon particle groups.



(a)Activated carbon without adsorbing pollutants (b)Activated carbon adsorbing pollutants

Figure 3. Variation curves of non-constant settlement of activated carbon particle groups

5. CONCLUSIONS

The process of static water settlement of powdered activated carbon and fine grained sediment is similar. The main difference between their physical properties is that the density of activated carbon in water is the weighted density of mixture of water, carbon and pollutants due to adsorption of water and pollutants in pore. In this paper, the fine static water settlement test was carried out for two kinds of activated carbon particle groups with and without adsorbing pollutants and obtained the experimental results of settling velocity of activated carbon particle group with time. By comparing with the theoretical calculation results of the settling velocity of single particle, it is found that the powdered activated carbon is not a simple single particle settling in static water, but settling in the form of flocs due to flocculation. And the particle size of the flocs can change with time due to diffusion. Accordingly, a flocculation-diffusion model with time-dependent equivalent particle size of activated carbon was proposed in this paper. Based on the experimental data, the empirical formula of the equivalent particle size of activated carbon particle flocs with time evolution is fitted. And based on the BBO equation of particle transient movement, the unsteady settlement process of activated carbon particle group in static water was numerically simulated. The calculated results are in good agreement with the experimental data, indicating that the proposed "flocculation-diffusion" model and numerical calculation method can well simulate the settlement characteristic of activated carbon particle groups in static water. It will have certain reference value for the industrial application of activated carbon.

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