

# Physicochemical interaction and its influence on deep bed filtration process

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**Abstract:** The capillary model was used to analyze the hydraulic conditions in the deep bed filtration process. The physicochemical interaction forces between the filter media and suspended particles and their influence on deep bed filtration process were also studied theoretically. Through the comparison of the hydraulic and physicochemical forces, the key influencing factors on the filtration process were proposed and investigated. Pilot study of the micro-flocculation deep bed filtration was carried out in the No. 9 Potable Water Treatment Plant of Beijing, and the experimental results of hydraulic head loss, particle distribution and entrapment were presented. The theoretical prediction was reasonably consistent with the experimental results under different conditions, which indicated that the regulation and control of micro-flocculation and deep bed filtration could be realized by the evaluation of the physicochemical interactions. Further theoretical and experimental research should be carried out to investigate the interaction mechanism and its application in the deep bed filtration and other cases.

**Keywords:** capillary; micro-flocculation; filtration; physicochemical; interaction

## Introduction

Filtration process in the production of potable water is primarily aimed to remove suspended particles from water, and it is the last unit process that will directly affect the tap water quality (Pontius, 1990). Deep bed filtration, which is an efficient and cost effective method for the treatment of low turbidity and low temperature water, has been widely studied and used (Ives, 1980; Shandalov, 1997; Yao, 1971; Bai, 2000).

Deep bed filtration involves a variety of complex mechanisms in the particle removal process, and the surface interactions between suspended particles and filter grain have been thoroughly investigated by many researchers (Payatakes, 1974; Huang, 1999; Jegatheesan, 2000; O' Melia, 1997; Rajagopalan, 1976; Srinivasa, 1998). The particles to be removed are generally much smaller than the pores in the packed bed, so the physicochemical interaction taken place in the filter will predominantly influence the particle removal. The shear force caused by water flow will also affect the particle adsorption and desorption to the filter grain (Warszynski, 2000; Guo, 2002). Because of its influence on the surface potential and size distribution of the suspended particles, the chemical pretreatment is essential to deep bed filtration. The influence of flocculent on particle aggregation and its attachment to the filter was also investigated extensively to improve the filtration efficiency (Berre, 1998; Habibian, 1975; Leprince, 1984; O' Melia, 1967; 1985; Tang, 1987). Filtration is a promising unit process deserve further study, and there are still plenty of room for improvement in filtration and its relative areas.

On the basis of uniform medial filtration, the turbidity removal and head loss in filtration process was analyzed by capillary model theoretically (Jing, 2000). And in this paper, the capillary model is combined with the physicochemical forces between the suspended particles and filter grain to analyze the micro-flocculation deep bed filtration. The shear force distribution and hydraulic slope in the capillary is studied firstly. Then, the physicochemical forces including London—van der Waals attraction force, electric double-layer force, Born repulsion force and

structural (or hydration) force between the particle and the filter grain and their influence on the filtration process are analyzed theoretically. The theoretical results are used to explain the experimental data, and good prediction could be obtained.

## 1. Theoretical analysis

### 1.1 Capillary model and hydraulic slope

When water transfers through the porous media as laminar flow in the packed bed filtration process, the suspended particles in it will interact with the filter grain. And the particle will be removed if the total interaction force is strong enough to bond the particle to the grain surface. At the beginning of filtration operation, the particles will adhere to the high-attraction positions in the packed bed. The forces on the filter pore surface will be about equal when stable operation stage is arrived, then the pores in the filter bed can be considered as circular shaped capillaries (Guo, 2002). The following equations can be formulated through comparison of capillary hypothesis with actual filtration system (Jing, 2000):

$$\delta = n \cdot d_c^2 \cdot \pi / 4, \quad (1)$$

$$f = n \cdot \pi d_c, \quad (2)$$

where  $\delta$  is the filter bed porosity,  $f$  is the specific surface area of the filter bed,  $m^{-1}$ ,  $n$  is the number of capillaries per unit surface area of the filter bed,  $m^{-2}$ ,  $d_c$  is the capillary diameter, m. For non-spherical filter grain, the specific surface area can be expressed as (Jing, 2000):

$$f = 6\alpha(1 - \delta) / d_c, \quad (3)$$

where  $\alpha$  is the surface shape factor and  $d_c$  is the equivalent diameter of a filter grain, m.

Substituting Equation (1) and (2) into (3), the following results can be obtained:

$$d_c = \frac{2\delta}{3\alpha(1 - \delta)} d_c, \quad (4)$$

$$n = \frac{9\alpha^2(1 - \delta)^2}{\pi\delta \cdot d_c^2}. \quad (5)$$

The average flow rate in each capillary must be equal to that of the filter pores, i. e.,  $\bar{u} = Q/A_s$ , where  $\bar{u}$  is the average flow rate in capillaries, m/s,  $Q$  is the apparent

filtration velocity, m/s,  $A_\delta$  is the pore area per unit area of filter medium  $A_\delta = \pi (d_c/2)^2 n = \delta$ . As mentioned above, the flow in the capillary is considered as laminar flow, and the velocity distribution of laminar flow in a circular shaped capillary can be expressed as  $u = \gamma J (r_0^2 - r^2) / 4\mu$ , where  $r_0$  is the capillary radius. The average velocity is (Wen, 1992):

$$\bar{u} = \gamma J d_c^2 / 32\mu, \quad (6)$$

where  $r$  is the distance from an arbitrary point to the capillary center, m,  $\gamma$  is the fluid density, kN/m<sup>3</sup>, and  $\mu$  is the dynamic viscosity of the fluid.  $J$  is the hydraulic slope:  $J = h_f/l$ , where  $h_f$  is the hydraulic head loss, m,  $l$  is the capillary length, m.

Through the analysis conducted above, the theoretical hydraulic slope during uniform media filtration could be achieved (Wen, 1992)

$$J = 32 \cdot \bar{u} \mu / \gamma d_c^2. \quad (7)$$

For the uniform flow in pressurized circular capillary, the relation between shear force  $\tau$  and hydraulic loss can be given as:  $\tau = \gamma \cdot r \cdot J/2$ , and the average hydraulic head loss of laminar flow in the capillary can be written as:  $\bar{h}_f = J \times l$ . Thus the shear force distribution in the circular shaped capillary can be obtained from Equation (7):

$$\tau = 16 \cdot \mu \cdot \bar{u} \cdot r / d_c^2. \quad (8)$$

It can be concluded from Equation (8) that the shear force in the capillary center equals zero, and it is in direct proportion to the distance between the capillary center and calculation position. The maximum value  $8\mu\bar{u}/d_c$  is reached on the inner wall of capillary. Supposing that the separation distance between the capillary wall and particle is  $z$ , then  $r = d_c/2 - z$ , and the fluid shear force in position  $z$  can be calculated by the following equation:

$$\tau = 16\mu\bar{u}(d_c/2 - z)/d_c^2. \quad (9)$$

## 1.2 Physicochemical forces between the particle and capillary wall

During filtration process, the influencing physicochemical forces on the particle attachment onto the wall of filter bed pores can be divided into two groups according to their function scope (Ives, 1980; Jegatheesan, 2000). The Born repulsion force ( $F_B$ ) and structural (or hydration) force ( $F_h$ ) are termed as short-range forces due to their influence on particles being dominant only if the particles are less than 5 nm away from the interaction surface. However, the influence of London-van der Waals attraction force ( $F_L$ ) and electric double-layer force ( $F_e$ ) (either attractive or repulsive) exist even when the distance between the particle and interaction surface is about 100 nm, so they are called long-range forces. During filtration process, these forces will affect the particle adsorption and desorption on the filter pore surface. The equations governing these forces are given below:

Retarded London-van der Waals force (Gregory, 1981; Jegatheesan, 2000):

$$F_L = (A_{cwp} a_p / 6) [1 + 28(z/\lambda_c)] / \{z^2 [1 + 14(z/\lambda_c)]^2\}. \quad (10)$$

Electrical double-layer force (Gregory, 1975; Jegatheesan, 2000):

$$F_e = -64\pi a_p \epsilon K [k_B T / Ze]^2 \tanh[Ze\psi_1 / 4k_B T] \tanh[Ze\psi_2 / 4k_B T] \exp(-kz). \quad (11)$$

Born force (Ruckenstein, 1976; Jegatheesan, 2000):

$$F_B = -A_{cwp} a_p \sigma_1^6 (180z^8). \quad (12)$$

Hydration force (Israelachvili, 1992; Jegatheesan, 2000):

$$F_h = -2\pi a_p K_1 h \exp(-z/h), \quad (13)$$

where  $F_B$ ,  $F_e$ ,  $F_h$ ,  $F_L$  are the Born force, electrical double-layer force, hydration force and London-van der Waals force respectively (Nm/s<sup>2</sup>),  $a_c$ ,  $a_p$  are the radii of the filter grain

and suspended particles (m),  $z$  is the distance between a particle and the capillary wall (m),  $\lambda_c$  is the characteristic wavelength of the interaction (m), defined as  $2\pi c/\omega_c$ ,  $c$  is the light velocity,  $2.9979 \times 10^8$  m/s,  $\omega_c$  is the dispersion frequency, for most materials  $\lambda_c$  takes a value of 100 nm (Jegatheesan, 2000),  $k$  is the inverse Debye length (m<sup>-1</sup>), which can be approximated as (Stumm, 1981):  $\kappa^{-1} \approx 2.8/\sqrt{I}$  nm,  $I$  is the ionic strength, mole/L.  $k_B$  is the Boltzmann's constant,  $1.3807 \times 10^{-23}$  J/K,  $T$  is the suspension temperature, K,  $Z$  is the charge number of the electrolyte used,  $e$  is the charge of an electron,  $1.6022 \times 10^{-19}$  C,  $\psi_1$  and  $\psi_2$  are the surface potential of particles and filter grain respectively,  $\sigma_1$  is the collision diameter (m),  $K_1$  and  $h$  are empirical constants,  $\epsilon$  is the suspension permittivity ( $\epsilon = \epsilon_r \cdot \epsilon_0$ ),  $\epsilon_0$  is the permittivity of the vacuum and  $\epsilon_r$  is the dielectric constant of the suspension.  $A_{cwp}$  is the Hamaker constant when a particle (p) and a filter grain (c) are separated by water (w) (Nm<sup>2</sup>/s<sup>2</sup>). Assuming the suspended particles are mainly composed of silica, then the Hamaker constant between the particle and the capillary wall can be approximated as:  $A_{cwp} = (A_{Si}^{1/2} - A_{H_2O}^{1/2})^2$ . The Hamaker constant of the water and quartz are  $5 \times 10^{-20}$  and  $1 \times 10^{-19}$  J, respectively (Stumm, 1981; Zhou, 1996), so  $A_{cwp} = (A_{Si}^{1/2} - A_{H_2O}^{1/2})^2 = 8.58 \times 10^{-21}$  (J).

Under the condition of 20° centigrade, the interaction forces between a particle and the capillary wall in the univalent electrolyte solution can be simplified as follows:

$$F_L = 1.43 \times 10^{-2} a_p (1 + 0.28z) \cdot (z + 1.4 \times 10^{-3} z^2)^{-2}, \quad (14)$$

$$F_e = -0.334 I^{1/2} a_p \times 0.74 I^{0.5z} \tanh(9.756\psi_1) \cdot \tanh(9.756\psi_2), \quad (15)$$

$$F_B = -4.77 \times 10^{23} a_p \sigma_1^6 z^8, \quad (16)$$

$$F_h = -6.28 a_p K_1 h \times \exp(-z/h). \quad (17)$$

## 2 Experimental study

### 2.1 Experimental methods

Pilot study of micro-flocculation deep bed filtration was carried out in the No. 9 Potable Water Treatment Plant of Beijing, the raw water of which was from Miyun Reservoir. The schematic flow chart for the micro-flocculation deep bed filtration is shown in Fig. 1

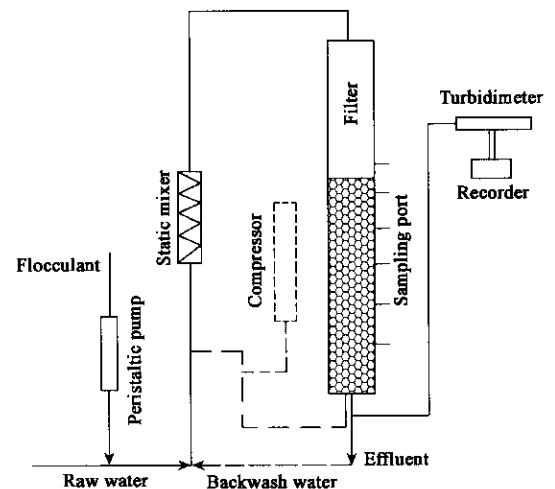


Fig. 1 Schematic flow chart of the micro-flocculation deep bed filtration

Injected by peristaltic pump (Model 7523-37, Cole-Parmer Instrument Cor.), the coagulant was mixed with raw water in the static in-line mixer (Model 1-40C-4-12-2, Koflo Cor.), and the coagulated water flowed into the packed filter

column through the pipelines. The effluent turbidity was monitored by online turbidimeter (Model 8220, Great Lake Co. Ltd.), and the results were sent to the recorder (Model 056-3002, Hitachi Cor.) for analysis and printing.

**2.2 Materials**

The filter column, which was made of plexiglass, was 5000 mm high with the inner diameter of 200 mm. The interval between the sample ports was 300 mm, and 200 mm height stone was used as support for the packed filter bed. The equivalent diameter of the filter media, which was made form anthracite, was 2.7 mm. The height of the filter bed was 2500 mm and the experiment was carried out under the filtration velocity of 6.7 mm/s. The particle size distribution in the raw water, coagulated water and effluent was measured (Multisizer II, Coulter Electronics) and analyzed. The size distribution of the suspended particles in the source and filtered water is given in Table 1.

**Table 1 Particle size distribution in the source and filtered water**

Particle diameter, $\mu\text{m}$	Filtered water		Source water, %
	PAC, % 0.5 mg/L	PFC, % 0.5 mg/L	
1-2	88.26	87.14	92.38
2-3	7.57	8.30	5.14
3-4	2.16	2.37	1.27
4-6	1.51	1.55	0.89
6-10	0.44	0.55	0.29
10-20	0.06	0.09	0.03

The coagulants used in the experiment were liquid polyaluminum chloride (Qingdao, China, hereinafter referred to as PAC) and polyferric chloride (made from liquid wastes of pickling industry, hereinafter referred to as PFC). The  $\text{Fe}^{3+}$  concentration in the PFC was 2.23 mol/L while the  $\text{Al}_2\text{O}_3$  content in the PAC was 15.6%. The alkalinities, i. e. the concentration ratio of  $\text{OH}^-$  to metal ion, were 1.4 and 0.31 for the PAC and PFC, respectively.

Since the raw water turbidity was low, the lower established limit for the head loss was set as 2000 mm, while that for the effluent turbidity was 0.3 NTU.

**3 Results and discussion**

**3.1 Experimental results**

It can be seen from Fig. 2 that the particle removal efficiency was in negative correlation with its diameter when no flocculent was dosed, i. e. the removal efficiency was decrease with the increase of particle size along the whole filter bed. However, different phenomena could be observed when flocculent was added. The influences of PAC and PFC on particle removal are shown in Fig. 3 and Fig. 4, from which it can be concluded that the particle removal efficiency increases with the increase of its size.

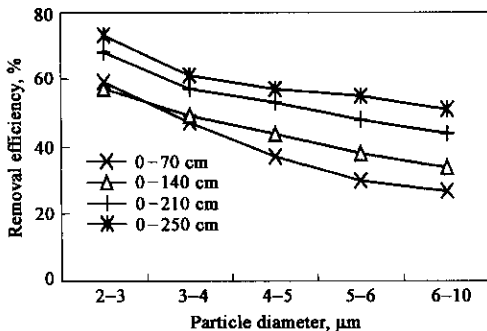


Fig. 2 Effect of particle diameter on its removal without flocculent dosage

Different influence of PFC and PAC on particle entrapment along the filter bed could be obtained through the

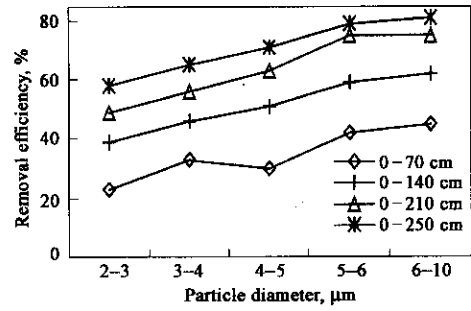


Fig. 3 Particle removal efficiency along the filter bed with 0.5 mg/L PAC dosage

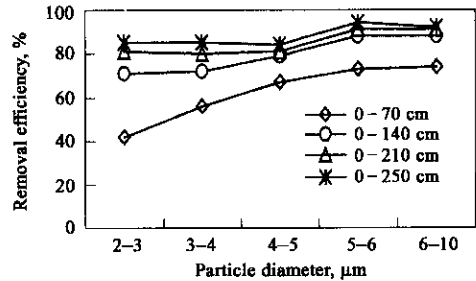


Fig. 4 Particle removal efficiency along the filter bed with 0.5 mg/L PFC dosage

comparison of Fig. 3 and Fig. 4. For PFC flocculent, the particle removal occurred mostly in the upper part of the filter column, while the entrapped particles rather evenly distributed along the whole filter bed for PAC flocculent. However, the head loss of PFC was lower than that of the PAC during the filtration operation, which may be resulted from the dehydration of the PFC flocs. The relationship between the head loss and filtration time is illustrated as Fig. 5.

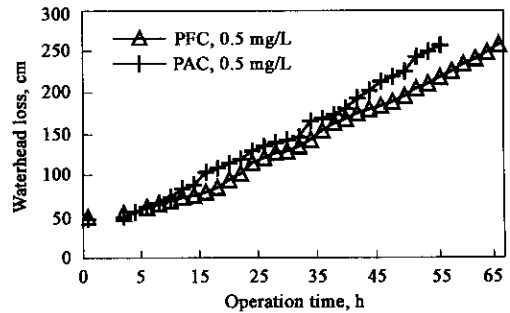


Fig. 5 Influence of operation time on the head loss of filter bed

**3.2 Theoretical results**

The ion strength in the theoretical calculation is 0.01 and the values for constant  $k_1$  and  $h$  are 0.00001 and 0.8 respectively. The temperature used in the calculation is 20°C. The shape factor of the filter grain is 2.08, while the other conditions are the same as those of the filter bed in the experimental part, i. e. 2500 mm packed anthracite grain whose equivalent diameter is 2.7 mm, and the filter bed porosity is 55%. The friction coefficient between the suspended particles and filter is assumed as 0.2. Other specific calculation conditions are given in the sections followed. Detailed calculation was presented in other researches (Guo, 2002).

**3.2.1 Influence of filter bed porosity on head loss**

According to the above-mentioned theoretical analysis, the influence of filter bed porosity on the hydraulic slope during filtration was calculated by Equation (7), and the theoretical results are shown in Fig. 6. It can be seen from

Fig. 6 that the hydraulic slope increases rapidly with the decrease of filter porosity, especially when the porosity is low, which means that great head loss change will occur if the porosity have small changes during the filtration process.

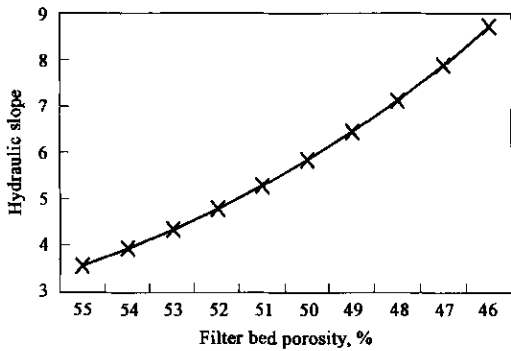


Fig. 6 Theoretical relation between the hydraulic slope and filter bed porosity

The PFC flocculent contains hydroxylic polymers, which can increase the collision of suspended particles and the collision between the particles and filter grain, thus accelerate the floc growth and enhance its entrapment by the filter. Dehydroxylation of the hydroxylic polymers in the PFC flocs will lead to the shrink of the flocs, thus slow down the porosity decrease (Leprince, 1984; O' Melia, 1967). As discussed in the theoretical analysis, small increase of the filter bed porosity will lead to rather great decrease of hydraulic slope. Therefore, the lower head loss of PFC compared with PAC during deep bed filtration may be resulted from the dehydroxylation of PFC flocs.

**3.2.2 Influence of particle diameter on the shear force**

Fig. 7 shows the influence of particle diameter on the shear force of suspended particles, which is calculated on the bases of Equation (9), and the separation distance  $z$  between the particle and capillary wall is assumed as 2 nm. The shear force on the suspended particle is the function of filter porosity, i. e. diameter of the capillary, which will determine the flow velocity in the capillary. Therefore, surface

characteristic of the particle and filter grain has slight effect on the shear force. According to the analysis of capillary model, the shear force has linear relationship with the particle diameter, which could be got from Fig. 7.

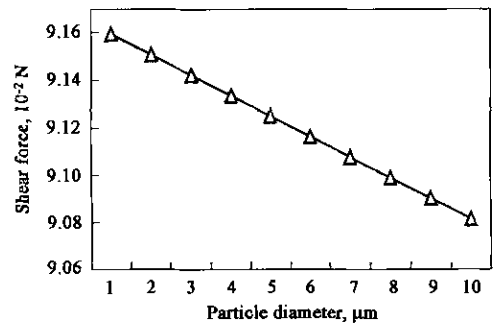


Fig. 7 Theoretical relation between the particle diameter and shear force

**3.2.3 Influence of surface potential on the particle removal**

The influence of surface potential on the particle removal shown in Fig. 8(a) and (b) are achieved when the surface potential of the suspended particles are -25 mV and -125 mV respectively.

It can be seen from Fig. 8(a) that the physicochemical forces between the particle and filter grain surface decrease rapidly with the increase of their separation distance. The influence of particle diameter on the physicochemical forces will be quite small when the separation interval is larger than 5 nm. However, in the range from 1 to 4 nm, the particle diameter has great effect on the physicochemical interaction force, and the attraction force increased sharply with the increase of particle diameter. The surface potential of the suspended particles (flocs) will be greatly increased when flocculent is dosed, and the filter surface potential will also greatly increased due to the floc attachment on the filter grain. Thus the physicochemical force between the suspended particles and filter grain will have the same variation trend as shown in Fig. 8(a).

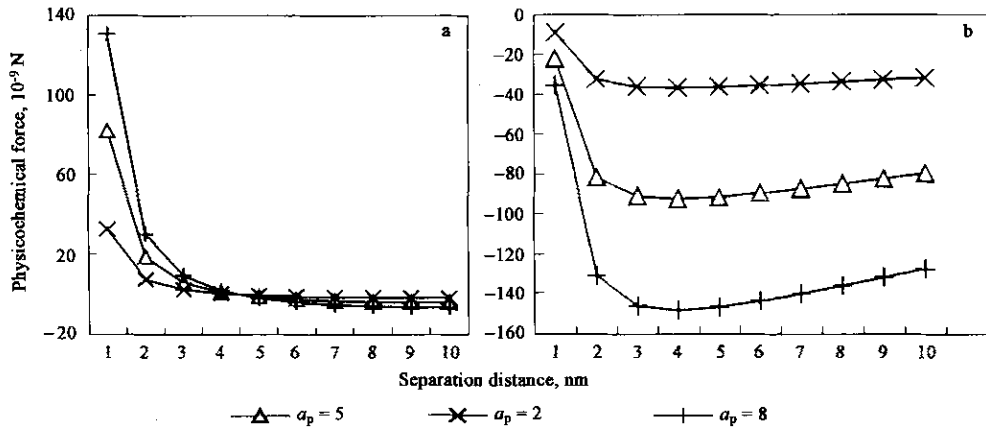


Fig. 8 Influence of separation distance on the physicochemical force between the particle and filter grain

As concluded from Fig. 7, the influence of particle diameter on the shear force is linear and quite small, which means that the obstacle to particle attachment has slightly relation with the particle diameter. Therefore, the changes of physicochemical force will greatly affect the particle attachment according to the theoretical analysis, and the flocculent dosage will result in higher removal efficiency for larger particles, which can be seen from Fig. 3 and Fig. 4.

It can be seen from Fig. 8(b) that the physicochemical force between the particle and capillary wall is minus when the surface potential is -125 mV, so the influencing factors

on particle removal under such conditions will be inertia, diffusion and other interactions.

The repulsion force between the particle and filter grain will increase with the increase of particle diameter and separation distance, which means that the attachment on the filter grain will be more difficult for larger particles. The influence of the separation distance will be quite small when the separation distance is larger than 4 nm, which is of the same trend observed in the high surface potential case. The surface potential of the particles and filter grain will be quite low without flocculent dosage, and as discussed above, the

removal efficiency will decrease significantly with the increase of particle size under such conditions, which can interpret the experimental results of Fig. 2 qualitatively.

#### 4 Conclusions

Using capillary model, the hydraulic shear force distribution in the pore of filter bed was simulated, and the physicochemical interaction forces between the suspended particle and filter grain and their influence during filtration process was investigated theoretically. The computation results were used to interpret the experimental data of pilot study for micro-flocculation deep bed filtration, which was carried out in the No. 9 Potable Water Treatment Plant of Beijing. The water head loss, particle size distribution and its removal efficiency along the packed filter bed were measured during the experiment. Theoretical calculation for the physicochemical forces between the suspended particle and filter grain, shear force on the particle caused by the flow in filter bed and the influence of the surface potential, particle size and separation distance were carried out. The computational results could explain the experimental phenomena qualitatively, and through the comparison of the theoretical and experimental results, it can be concluded that the physicochemical interaction and flow condition analysis could be used to study the regulation and control of the uniform media deep bed filtration. Further theoretical and experimental research should be carried out to study the interaction mechanism and its application in deep bed filtration and other cases, then the quantitative computation and analysis will be possible and the deep bed filtration process could be operated more easily and efficiently.

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