Establishment of the Permeability Model for Soft Solid Sludge Conditioned with Flocculants

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ABSTRACT: Permeability plays a decisive role in the dewatering process and reflects the difficulty of filtration, especially for soft solid material such as sludge. In this paper, the physicochemical properties and dewatering performance of sludge conditioned with different kinds of flocculants were investigated. Results showed that the flocculant could change the sludge microstructure such as floc morphology, specific surface area, and fractal dimension. Compared with filtration pressure, flocculants had a greater influence on sludge permeability which was a significant negative correlation with filtration pressure and was a positive correlation with flocculant dosage. In order to describe the fact that fluid flows through the porous voids for soft solid sludge, the improved Kozeny constant was corrected. Research showed that permeability was more significant in the dewatering process for the sludge conditioned with inorganic flocculants than that with organic flocculants. The Kozeny constant was not only relevant with suspension nature but also with filtration pressure. The range of the improved Kozeny constant was reasonably determined based on flocculant type, concentration, and filtration pressure, which was of great help to project applications. For raw sludge, the improved Kozeny constant was 958 times than that of the original value, and it decreased significantly for conditioned sludge.

1. INTRODUCTION

Sludge is the product of sewage treatment. In recent years, with development of cities and industries, the output of sewage sludge increased sharply, causing serious environment pollution.1,2 It is estimated that the production of sludge (moisture content of about 80%) in China increased by 20 million tons from 2010 to 2017. Sewage sludge is a kind of soft solid material with high water content (up to 99%), high organic content, fine particles, and small specific gravity. Extracellular polymeric substance in sludge passes through hydrophilic functional groups, such as COOH and OH, to provide a strong cohesive force, making inorganic particles and microbial cells organically linked and attached to the surface of microbial cells. As a result, a gel-like bio-colloid structure similar to the sodium alginate was formed.3–5 These sludge characteristics determine that sludge is difficult to be dewatered and cannot even dewater without pretreatment.6,7 Sludge dewatering is an essential process in sludge disposal, which has close relation with the final processing cost.

In order to improve sludge dewaterability, many methods are employed, including chemical, physical, biological, and hybrid conditioning.8–21 Chemical conditioning aims to change sludge physicochemical or electrochemistry properties by adding acids, coagulants/flocculants, and it is a relatively mature, cost-effective, and user-friendly sludge dewatering technology.12–21 Physical conditioning mainly refers to thermal hydrolysis, ultrasonic conditioning, freeze–thaw treatment, and biological conditioning adopts enzymes and bioflocculants.22–25 In many pretreatments, coagulants/flocculants are the most common agents for sludge conditioning in practice because of lower cost and higher efficiency.10,16–22 Cationic polyacrylamide (CPAM) with a linear high molecular weight is a typical organic flocculant and has a variety of active groups (such as CONH2). Physical and chemical acting force (main including hydrogen bonding forces and van der Waals forces) facilitates adsorbing and bridging between sludge particles and CPAM, which could contribute to forming strong flocculation and good dewaterability at low dosage.13,17,20 Polyaluminum chloride (PAC) and ferric chloride (FC) are inorganic flocculants and could hydrolyze into hydroxyl compound in aqueous solution. Because of bridging of hydroxyl ions and polymerization of polyvalent anions, the molecular weight of PAC is larger than FC. However, PAC and FC could both facilitate conglomeration of sludge particles into flocs through charge neutralization and enmeshment. Besides, inorganic flocculants have been reported to be served as skeleton builders and improve the porosity of the sludge cake.8,10,18,29,30

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Fluid flowing in porous media (including filter media and filter cake) is the filtration essence. As the filtration process proceeds, solid particles in suspension continuously deposit on the surface of medium to form a filter cake or block inside pores of medium. Then, flow resistance increases, and filtration rate decreases rapidly.\(^{31,32}\) Permeability is a key parameter for studying the filtration process, and it is the most basic problem in studying seepage of porous cake materials.\(^{33}\) However, the permeability in both the Ruth equation and Kozeny–Carmen (K–C) equation is treated as a constant. At the same time, particles in the K–C equation are treated as uniform, hard spheres, and the contact form between particles is point. Furthermore, the Kozeny constant \((K_c)\) is usually fixed in a constant of 5, in fact, which is related to material properties and has larger fluctuation. Many research studies have been done and several empirical models have been constructed to study the permeability of deformable porous media, including index models, power models, and logarithmic models.\(^{34−36}\)
while most of them focus on rocks and are not applicable to soft solid gel materials such as sludge. Therefore, an appropriate permeability model in sludge filtration should be studied.

In this paper, in order to accurately describe the mechanism of filtration dehydration for soft solid gel materials, CPAM, FC, and PAC were selected as sludge conditioners to obtain different kinds of suspension with various physicochemical characteristics. On the basis of Darcy’s law, the Kozeny constant in the K–C equation was improved and established.

2. RESULTS AND DISCUSSION

2.1. Effect of Flocculant on Sludge Physicochemical Properties. Figure 1 shows the effect of the flocculant on the floc size, pH, one-fractal dimension, and specific surface area. It can be seen that as the addition amount of inorganic flocculant PAC and FC increased to 120–140 mg/g, the pH of sludge suspension decreased from 6.88 of original sludge to 6.09 and 5.54, respectively. Hydrolysis of PAC and FC could cause higher concentration of the hydrogen ion in the solution, and thus, the pH of the solution decreased.8,29 Furthermore, stronger hydrolysis performance of FC led to lower pH than that of PAC. However, CPAM could inhibit sludge dissolution, hydrolysis, and acid production. As a result, when the amount of CPAM was 3 mg/g, the pH increased from 6.88 to 7.50. The variation trend of the sludge particle size and specific surface area with the dosage of PAC and FC was not a monotone decrease, and the minimum (maximum) occurred at 40 and 60 mg/g for PAC and FC, respectively. While for all sludge samples conditioned with PAC and FC, the smaller particle size and larger specific surface area were observed, compared with original ones (original sludge particle size: 71.89 μm, specific surface area: 946.7 cm²/g). It is possibly due to their high charge density, which could effectively compress the double electricity layer. In addition, the CPAM dosage was negatively correlated with the specific surface area and positively correlated with the particle size, which was on account of the effect of electrical neutralization on particle agglomeration and flocculation. One-fractal dimension characterizes the regularity of the aggregate structure, and it is negatively correlated with flocculant dosage for all the treatments, which is consistent with the results of Zhao.37 It is possible that more compact and more peripheral regular flocs are formed under charge neutralization between electro-negative particles and cations within flocculants, leading to a smaller one-fractal dimension. As a result, improved porosity of the filter cake and better dewaterability were obtained.38,46

2.2. Effect of the Flocculant and Filtration Pressure on Filter Cake Permeability. 2.2.1. Effect of the Flocculant on Filter Cake Permeability. Figure 2 shows the effect of dosage of three kinds of flocculants (PAC, FC, and CPAM) on permeability at a constant pressure gradient of 3.4 × 10⁵ Pa, respectively. With the increase of flocculant dosage, permeability increased first and then decreased. As the dosage of PAC was at 100 mg/g, it increased by 50 times compared to that of original sludge (from 5.89 × 10⁻¹¹ cm² for original sludge to 2.88 × 10⁻⁹ cm²). In the filtration process, the charge neutralization effect could reduce the thickness of the hydrated shell of sludge particles and enhance free water content by compressing the electric double layer and weakening the surface tension of sludge water, leading to better dewaterability.39 Meanwhile, excessive dosage might cause sludge particles to be covered with PAC, resulting in the regeneration of the suspension stability, that is, the “restabilization effect”. Therefore, the optimal dosage of PAC was 100 mg/g in this work, which was consistent with the previous work.14 However, as the sludge treated with organic flocculant CPAM (at the dosage of 3 mg/g), the permeability was only 3.86 × 10⁻¹⁰ cm², which increased by less than 7 times, indicating that filtration velocity is faster for sludge treated with an inorganic flocculant at this condition.

2.2.2. Effect of Pressure on Filter Cake Permeability. Figure 3 illustrates the effect of filtration pressure on sludge filter cake permeability, where the sludge was treated with three kinds of flocculants (PAC, FC, and CPAM) at the concentration of 100, 120, 3 mg/g, respectively. It can be seen that with the increase of filtration pressure, the filter cake permeability first dropped sharply and then tended to be gentle, which may be due to the decreased effective gap in the filter cake at higher filtration pressure. As filtration pressure

Figure 2. Permeability at different flocculant concentrations.

![Figure 2](image-url)
was greater than $3.4 \times 10^5$ Pa, its influence on permeability became apparent, maybe due to the formation of a highly compact skin at the interface between the cake and filter medium. In addition, the permeability for the sludge conditioned with CPAM reduced by 91.7% as filtration pressure increased from $1 \times 10^5$ to $3.4 \times 10^5$ Pa, which was obviously above those of sludge treated with PAC and FC, maybe attributing to the higher compressibility of sludge conditioned with CPAM.

Figure 4 demonstrated the relationship between the filtration rate and filtration pressure. Obviously, a weak correlation was observed. In theory, higher filtration pressure could contribute to the outflow of the filtrate, but concurrently, the partial effective pores of the filter cake are gradually blocked, resulting in the reduction of effective porosity. Consequently, the filtration rate could not be proportional to pressure. Also, at the filtration pressure of $3.4 \times 10^5$ Pa, the filtration rate of original sludge was 0.0086 cm$^3$/s, while it increased by 44.2, 30.2, and 5.6 times for sludge conditioned by PAC, FC, and CPAM, respectively. Inorganic flocculants (PAC and FC) can destroy the sludge network structure and weaken its water-trapping capability. As a result, flocs became more peripheral, regular, and compact with a smaller specific area, smaller settlement drag coefficient, and the effective porosity in the filter cake became larger, leading to better dewaterability performance. Furthermore, the improvement of the filtration rate for CPAM-conditioned sludge is not significant compared with inorganic flocculants PAC and FC, maybe due to its limited dosage.

2.3. Establishment of K–C for Sludge. Common expression for the permeability is expressed as a product of a preset constant. The best-known formula is perhaps the K–C relation, which is shown as eq 1.

$$K = \frac{\varepsilon^3}{K_c (1 - \varepsilon)^2 S_0^2}$$  \hspace{1cm} (1)

where $\varepsilon$ refers to local porosity, $%$; $S_0$ refers to specific surface area of particles, m$^2$/m$^3$; $K_c$ refers to Kozeny constant, which is related to suspension nature, including shape and compressibility of particles, dimensionless. For grain-like particles, $K_c$ is usually defined as $S_0^{−1}$ and it can be deduced and calculated by eq 1 in this paper.

Permeability prediction is an important aspect of the filtration process design. The K–C equation (Formula 1) belongs to a class of relationships relating the pressure gradient and flow rate of fluid flowing through porous media, and it is applicable to randomly packed media composed of spherical or nearly spherical particles. However, the filtration process of sludge with soft solid gel properties and complex physicochemical properties is difficult to be described by K–C, where the Kozeny constant is determined as about 5.

In order to account for the fact that fluid flows through the voids of a porous media for soft solid sludge, the improved Kozeny constant is proposed, which reflects the effect of sludge properties on dewatering and represents dewatering performance. Consequently, Formula 1 was corrected by (Formula 2).

$$K = \frac{\varepsilon^3}{K'_c (1 - \varepsilon)^2 S_0^2}$$  \hspace{1cm} (2)

where $K'_c$ is the improved Kozeny constant, suggesting that the medium morphology should be taken into account. In order to clearly explain the effect of three kinds of flocculants on dewatering performance, the ratio of the improved Kozeny constant to the original one ($K'_c/K_c$) was introduced ($K'_c/K_c$). The bigger value of $K'_c/K_c$ suggested higher dewatering resistance.

2.3.1. Effect of the Flocculant on the Kozeny Constant. Figure 5 exhibits the values of the improved Kozeny constant ($K'_c$) for raw and conditioned sludge at the filtration pressure of $3.4 \times 10^5$ Pa. Obviously, for the original sludge, $K'_c$ was about 958 times contrasting to that given in literature studies ($K_c$ was about $S_0^{−1}$). While for conditioned sludge, the value decreased prominently, from 4787 to several hundreds. Actually, the Kozeny constant has great relation to flocs, reflecting the physical nature of sludge. Original sludge contains various interlaced and disordered substances. On the contrary, conditioned sludge with more regular floc particles was a more regular arrangement, resulting in significant reduction of the Kozeny constant.

Interestingly, with the increase of dosage, the value of $K'_c$ reduced. As the PAC dosage achieved 100 mg/g, the minimum value of $K'_c$ (36) was observed, suggesting that the optimum dosage was determined, which is consistent with the above discussion. Furthermore, the minimum value of $K'_c$ was closed to $K_c$ (the value was 5), indicating that the floc characteristics for sludge conditioned by PAC was compact and less compressible, which can contribute to dewatering. While for the sludge conditioned with CPAM, the value of $K'_c$ was larger, maybe due to larger floc and higher compressibility.
2.3.2. Effect of Filtration Pressure on the Kozeny Constant. Figure 6 presents the relationship between filtration pressure and the Kozeny constant $K_c'$ at the following conditions (PAC: 100 mg/g, FC: 120 mg/g and CPAM: 3 mg/g). As above mentioned, the Kozeny constant is a material property, which refers to floc morphology, particle size, and compressibility. Therefore, the pressure gradient should not be taken into account during filtration. However, it is unexpected that $K_c'$ was obviously positively correlated with filtration pressure, which increased significantly from 9.6 to 48 for the sludge conditioned with PAC (filtration pressure: from $1.0 \times 10^5$ to $4.2 \times 10^5$ Pa). Perhaps, because of the particular characteristics for soft solid sludge (high compressibility and soft particles), extremely high variation in floc morphology and fractal structure appeared in higher pressure, resulting in an increment of the Kozeny constant. Therefore, for organic flocculant CPAM, $K_c'$ was larger than those of inorganic.

2.4. Prediction of the Improved Kozeny Constant. Sludge is a typical soft solid material with difficult dewaterability because of high content of the organic matter. The Kozeny constant as a comprehensive parameter is relevant to a material characteristic in the filtration process, reflecting dewatering performance indirectly.

As shown in Figure 5, $K_c$ decreased first with increase of flocculant dosage and then increased at one point. The minimum values of $K_c'$ were about 36 and 162 for sludge conditioned with PAC and FC, respectively, which decreased significantly compared to unconditioned sludge ($K_c$ is 4787). The variety trend of $K_c'/K_c$ with flocculant dosage at the filtration pressure of $3.4 \times 10^5$ Pa is shown in Figure 7, from which it can be seen that the ratio was at the range of 7.2 to 457. As the dosage of flocculant was appropriate, $K_c'$ would not exceed 50 and 160 for PAC and FC, respectively. Compared to $K_c$, the improved Kozeny constant increased by 7.2 times for conditioned sludge with PAC and increased by 32.4 times to 125.6 times, for conditioned sludge with FC, respectively. While for CPAM conditioned sludge, the maximum value of $K_c'$ is around 2000, and it could not be less than 590 usually.

Figure 8 presents the relationship between filtration pressure and $K_c'/K_c$. For an inorganic flocculant, the ratio value was small, and especially for PAC, the minimum was only 1.9 at the pressure gradient of $1.0 \times 10^5$ Pa, which was close proximity to the original Kozeny constant. Interestingly, at lower pressure, it could be concluded that physical characteristics of the sludge

![Figure 5](https://example.com/figure5.png)

(a) The value $K_c$ at various dosages of PAC and FC

Figure 5. Value of $K_c$ at various dosages of (a) PAC and FC; (b) CPAM.

![Figure 6](https://example.com/figure6.png)

(b) The value of $K_c'$ at various dosages of CPAM

Figure 6. Relationship between $K_c'$ and filtration pressure.

![Figure 7](https://example.com/figure7.png)

(a) The value of $K_c'/K_c$ at various dosages for PAC and FC

Figure 7. Value of $K_c'/K_c$ at various dosages for (a) PAC and FC; (b) CPAM.

![Figure 8](https://example.com/figure8.png)

(b) The value of $K_c'/K_c$ at various dosages for CPAM

Figure 8. Relationship between $K_c'/K_c$ at different filtration pressure.
conditioned by PAC was similar with less compressible materials, maybe due to its skeleton effect. While, for sludge conditioned with CPAM, it could be inferred that the minimum value of the Kozeny constant is about 100, and the maximum value should not exceed 600 as the added dosage of CPAM is reasonable (at the range of filtration pressure of \(1.0 \times 10^5\) to \(4.2 \times 10^5\) Pa). Obviously, the Kozeny constant for organic flocculant-conditioned sludge is in a larger range than that of inorganic conditioned sludge. The main reasons may be the significantly changed viscosity and strong cohesive force for organic conditioned sludge, which deforms sludge floc particles and in turn affects the range of the Kozeny constant.

Although the Kozeny constant is influenced by many factors, it fundamentally reflects the nature of suspension. Based on the above discussion, it can be concluded that the parameter of the filtration rate has a strong correlation with the Kozeny constant, which provides a more reliable basis for inference of the Kozeny constant. No matter what kind of flocculant is added and how much pressure is applied, the goal is to improve dewatering performance of sludge, and the filtration rate just reflects it. Figure 9 shows the relationship between the Kozeny constant and filtration rate for three kinds of flocculants with different dosage at filtration pressure of \(3.4 \times 10^3\) Pa. Interestingly, linear correlation with a high correlation coefficient was observed between the Kozeny constant and filtration rate. Also, the fitted formula could be only appropriate as the dosage of the flocculant was under the optimum value. However, as the addition was excess, it could be not applicable, because of irregular changes in physicochemical properties of sludge.

3. CONCLUSIONS

The sludge was conditioned by organic and inorganic flocculants, and the Kozeny constant in the permeability model for soft solid gel materials (such as sludge) was corrected and established. The specific conclusions were as follows.

1. Flocculants could change physicochemical properties of sewage sludge. The organic flocculant makes the floc particle size larger, the specific surface area smaller, and the fractal dimension smaller. While with the increase of the inorganic flocculant dosage, the fractal dimension decreased gradually. Furthermore, in the particle size and specific surface area, an inflection point was observed at the dosage of 40 and 60 mg/g for PAC and FC, respectively. The particle size decreased first and then increased. Correspondingly, the specific surface area increased first and then decreased.

2. Compared with filtration pressure, the flocculant has a greater influence on sludge filtration. In addition, permeability has a significantly negative correlation with filtration pressure and a positive correlation with flocculant dosage.

3. Kozeny constant in the permeability model for soft solid materials was established, which is not only relevant with suspension nature but also with filtration pressure. The range of the improved Kozeny constant was reasonably determined based on the flocculant type, concentration, and filtration pressure, which was of great help to project applications. For raw sludge, the Kozeny constant was 958 times than that of the original value. PAC and FC-conditioned sludge, the Kozeny constant decreased significantly, which was at the range of dozens to hundreds, while for CPAM-conditioned sludge, the Kozeny constant ranged from hundreds to thousands. The improved Kozeny constant was more significant in the dewatering process for the sludge conditioned with inorganic flocculants than that of organic flocculants, maybe due to differences of the action mechanism, and it could provide a certain experimental basis for filtration of soft solid materials.

Figure 9. Relationship between and filtration rate for sludge conditioned with (a) PAC; (b) FC; (c) CPAM.
4. MATERIALS AND METHODS

4.1. Sludge Samples. The municipal sludge was taken from efflux of the secondary sedimentation tank in the BeiJiao sewage treatment plant (Taiyuan, Shanxi province, China), and then was stored in a refrigerator at 4 °C to reduce properties change and ensure accuracy of following experiments. The sludge physicochemical properties were shown in Table 1.

Table 1. Physicochemical Properties of the Sludge

| Property                  | Value       |
|---------------------------|-------------|
| Total solids (%)          | 2.62        |
| d(0.5) (µm)               | 71.89       |
| pH                        | 6.88        |
| Zeta (mV)                 | −17.0       |
| Specific surface area (cm²/g) | 946.7     |

Before each experiment, the sludge was concentrated to a solid content of 7% and placed indoors for 30 min, which ensured that each set of experiments was carried out at room temperature. Furthermore, each experiment was performed 3 times, and the average value was taken for accuracy and repeatability.

4.2. Sludge Filtration Test. A batch filtration instrument was employed to complete the filtration experiment of sludge, which is mainly composed of a pressure filter with the cross-sectional area of 60.132 cm², an air compressor, and a gas storage tank.

The sludge was conditioned with different dosage of CPAM, FC, and PAC to improve physicochemical and dewatering properties, and the conditioning steps were as follows. First, a certain amount of the flocculant was added into sludge to prepare sludge suspensions. Second, the prepared sludge suspensions were stirred with a mechanical stirrer at 200 rpm for 5 min to accelerate mixing of the flocculant and sludge, and then stirred at 50 rpm for 15 min to promote sludge flocs growth, in order to obtain uniform sludge suspensions. Finally, batch filtration of conditioned sludge suspensions was carried out in the batch filtration instrument. The control variable method was used to investigate the effects of independent variables (including flocculant dosage and filtration pressure) on physicochemical properties and permeability during filtration. On the basis of previous studies, dosages of CPAM, FC, and PAC were determined as 0.12, 0.06, and 0.00 mg/g, respectively (relative to dry solid mass in sludge).\(^{14,41}\) The particle size and specific surface area of the sludge suspensions were measured by a laser particle size distribution analyzer (Betttersize 2000, China). Optical microscopy (Olympus, Japan) was used to observe and photograph sludge microscopic morphology was used to calculate sludge one-dimensional fractal dimension.\(^{44−49}\) Average cake porosity was obtained by the free water content in the filter cake.

4.3. Permeability Equation. For slow flows, fluid velocity through porous media is proportional to the pressure gradient, which obeys Darcy’s law as shown in eq 3.

\[
\frac{Δp}{L} = \frac{μ}{K} \times \frac{dV}{dr} \times \frac{1}{A}
\]

where \(Δp\) refers to pressure drop, Pa; \(L\) refers to dielectric layer thickness, cm, which is defined as the thickness of the filter cake in this work and can be obtained from experiment data; \(μ\) refers to liquid dynamic viscosity, Pa·s; \(dV\) refers to liquid volume flowing through the dielectric layer during time \(dt\), cm³; \(A\) refers to cross-sectional area of the filter cake, cm² (60.132 cm²); and \(K\) refers to permeability of the filter cake, cm², assumed constant. According to eq 3, \(K\) could be deduced.

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The authors declare no competing financial interest.

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