

Research Article

Research on Dewatering Ability of Municipal Sludge under the Treatment of Coupled Acid and Microwave

Xudong Zhang¹, Peng Ye¹, Yajun Wu¹, Zhen Guo^{2,3}, Yuan Huang⁴, Xingtao Zhang¹, Yuncong Sun¹, and Haiqiang Zhang¹

¹Department of Civil Engineering, Shanghai University, 200444, China

²College of Civil Engineering, Tongji University, Shanghai 200092, China

³Key Laboratory of Geotechnical and Underground Engineering of the Ministry of Education, Tongji University, Shanghai 200092, China

⁴Institute of Environmental Pollution and Health, School of Environmental and Chemical Engineering, Shanghai University, 200444, China

Correspondence should be addressed to Yajun Wu; wjlddz@shu.edu.cn and Zhen Guo; zhenguo@tongji.edu.cn

Received 24 May 2021; Accepted 9 July 2021; Published 6 August 2021

Academic Editor: Dayang Xuan

Copyright © 2021 Xudong Zhang et al. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

Municipal sludge is a by-product of urban sewage treatment; it has high water content and contains many hydrophilic extracellular polymeric substances (EPS). It is difficult to achieve deep dewatering of sludge. Therefore, it is necessary to find an efficient method to treat sludge. In this study, an acid-microwave treatment method was proposed to improve the dewatering ability of sludge. A vacuum filtration test and particle size analysis test were used to explore the improvement effect of dehydration performance. After centrifugation, the concentrations of soluble proteins and polysaccharides in the supernatant were measured to explore the disintegration of acid-microwave treatment on EPS. Finally, the microstructure of sludge treated by acid-microwave was studied via the SEM test. The results showed that short-term microwave radiation was conducive to sludge dewatering. However, the sludge dewatering performance deteriorated significantly when the treatment time was too long. The optimal microwave treatment condition was 700 W for 180 s, and the SRF of sludge was the lowest, which was 10.11×10^{12} m/kg. After acid-microwave treatment, sludge particle size increased significantly. The concentration of protein and polysaccharide in supernatant increased with the increase of microwave treatment time. Acid-microwave treatment could significantly improve the release efficiency of the polysaccharide. Through the SEM test, it can be verified that the bulk floc structure of sludge is broken after microwave treatment, and acid-microwave treatment is beneficial to the aggregation of floc fragments to form large particles, which can provide larger pores for drainage.

1. Introduction

Municipal sludge is the product of municipal sewage and industrial wastewater treatment by wastewater treatment plants (WWTPs). Many suspended substances are usually trapped in the wastewater treatment process, and sludge is composed of these suspended substances after settling. In recent years, with the rapid development of urbanization, the treatment rate of urban sewage and industrial wastewater is increasing, resulting in a significant increase in sludge production. As a result, the cost of municipal sludge treatment is

significantly increased, reaching about 50% of the operating cost of WWTPs [1, 2]. Sludge is formed by the precipitation of many pollutants, which results in a very complex composition of the sludge. Sludge mainly includes organic substances such as proteins and carbohydrates, and it also contains a large number of pathogens and heavy metals [3–5]. If the sludge is not correctly treated, it will pollute the environment and be harmful to health. Therefore, the treatment of municipal sludge has also attracted more and more attention. Municipal sludge treatment methods usually include agricultural utilization, fuel utilization, sanitary landfill, construction material

utilization, etc. [1]. In China, there are many sludge disposal methods. The ultimate goal is to achieve sludge reduction, stabilization, harmless, and resource utilization. At present, sludge is often first treated by landfill, then secondary treatment to achieve the purpose of resource utilization of sludge. However, due to the high water content and fluid state of sludge, municipal sludge is usually transported by pipeline, and the cost is expensive. If the sludge is dehydrated before transportation, the cost will be significantly reduced. In 2008, the water content standard of landfill sludge in China dropped from 80% to 60% [6], indicating that the sludge volume will be reduced by 50% before landfill. Therefore, reducing the water content of sludge to achieve volume reduction has become an urgent problem to be solved.

Sludge dewatering ability is an important factor affecting the efficiency of sludge volume reduction. Therefore, many scholars have carried out research on sludge conditioning. In previous studies, sludge treatment methods mainly include chemical conditioning, physical conditioning, and biological conditioning.

Chemical conditioning, i.e., adding oxidant, flocculant, acid, or alkali to the sludge, changes the dewatering ability of sludge through chemical reactions. The interaction between chemical reagents and sludge has been proved by experiments [7, 8]. For example, Zhang et al. [9] compared the effects of Fenton and FeCl_3 treatment on the consolidation characteristics of landfill sludge and explained the conditioning mechanism of Fenton and FeCl_3 . In the study of Zhen et al. [10], Fe^{2+} -activated sodium persulfate was used to regulate the sludge, and the sludge was oxidized by sodium persulfate to improve the dehydration ability. Li et al. [11] compared the effects of acid and alkali on the capillary suction time and the specific filtration resistance of sludge and concluded that only acidification treatment could improve the dewatering ability of sludge. Niu et al. [12] studied the effect of inorganic coagulants (FeCl_3 , PAC, and HPAC) on sludge dewatering ability and explained the mechanism of inorganic coagulation. The results showed that the effect of iron salt was slightly better than that of aluminium salt. At present, due to the low cost of chemical conditioning, it is mostly used in practical applications. However, chemical conditioning also has many problems. The use of chemicals will cause environmental pollution, and a large number of metal elements remain in sludge, which is not conducive to resource utilization.

Microbial conditioning using the bioflocculant as the conditioner makes the colloidal suspended substances in sludge condense and precipitate each other to enhance the dewatering ability. Zhang et al. [13] used microbial flocculants (TJ-F1) produced by *Proteus mirabilis* TJ-1 as a novel conditioner for improving the dewatering ability of sludge, and the results demonstrated that the microbial flocculant TJ-F1 is an effective conditioner in improving the dehydration ability of sludge. Guo et al. [14] used a composite of bioflocculant MBFGA1 and P(AM-DMC) to enhance the dewaterability of sludge and pointed that the dry solids and SRF appeared as 29.9% and 2.2×10^{12} m/kg at the optimal condition. Microbial flocculants are pollution-free and can be degraded. However, owing to the high cost, it has not been applied in practice.

Physical conditioning is the use of physical methods to change the characteristics of the sludge. For instances, Feng et al. [15] proposed using ultrasonic treatment of waste activated sludge; low specific energy dosage slightly improved sludge dewatering capacity, while large specific energy dosage significantly deteriorated sludge dewatering capacity. In addition, the dewatering ability of sludge can be greatly improved by heat treatment. Bougrier et al. [16] studied the treatment of sludge by ultrasonic, heat, and ozone, and the results showed that heat treatment could significantly reduce the apparent viscosity of sludge and improve the filtration capacity, which was the most effective method to improve the sludge dewatering ability.

Compared with traditional treatment methods of sludge, microwave conditioning does not pollute the environment and can transfer energy directly into the sludge to avoid wasting energy [17]. Therefore, the selective heating by microwave irradiation is an effective method. Microwave treatment usually includes "thermal effect" and "nonthermal effect." Wojciechowska compared the effects of microwave and conventional sludge heating on dewaterability and pointed out that the change of dewatering ability was not only caused by the sludge sample temperature; the nonthermal effect was also an important factor effecting the dewatering ability [18]. In the process of microwave irradiation, the orientation of polar molecules and dipoles is changed, which is the nonthermal effect, leading to hydrogen bonding fracture and denaturation of complex biological molecules. The thermal effect of microwave treatment is reflected in the heating effect on sludge, and the change of temperature field will affect the migration of the water field [19]. Therefore, heat treatment has the potential to improve the sludge dewatering ability. As an efficient clean energy, the microwave is also used for sludge conditioning. The study of Gil et al. [20] showed that microwave treatment had a positive effect on the biodegradability of sludge. Sheng et al. [21] found that the efficiency of microwave dehydration was generally better than that of thermal heating.

As the main component of the flocculent structure in sludge, EPS contains a large number of hydrophilic groups [22], which have a significant impact on the flocculation ability, settling ability, dehydration ability, and adsorption ability of sludge [21]. The free water content decreases because the EPS capture water in the sludge, resulting in the reduction in free water content of the sludge. In addition, sludge flocs have a large amount of negative charge, which repel each other and are difficult to aggregate. These are the critical reasons for the deterioration of sludge dewatering capacity. Therefore, the destruction of floc structure EPS is the key to improve sludge dewatering performance.

At present, most of the studies on sludge dewatering capacity only focus on a single method treatment, and it is difficult to avoid secondary pollution in the treatment process. Therefore, it is essential to find an effective sludge treatment method, which is environment-friendly and beneficial to the resource utilization of sludge. In this study, acid-microwave treatment was proposed as a new composite conditioning method to improve sludge dewatering ability. A vacuum filtration test was carried out to explore the optimal

microwave radiation time and pH value. In addition, to explore the mechanism of acid-microwave treatment improving sludge dewatering capacity, the particle distributions of sludge and soluble protein and polysaccharide in the supernatant were analyzed. The floc structure changes of treated sludge can be directly observed through the SEM test, which is important to investigate the mechanism of acid-microwave conditioning. Therefore, after the vacuum filtration test, the SEM test was carried out on the sludge cake to observe the microstructure of the sludge, and the effect of the microstructure on the sludge dewatering ability was analyzed. The results obtained from this study can provide guidance for the actual engineering.

2. Material and Methods

2.1. Sampling Area and Sludge Samples. In this study, municipal sludge was obtained from the Wusong Wastewater Treatment Plant in Shanghai. The sampling point is located at Haijiang Road, Baoshan District, Shanghai, and at the entrance of the Yangtze River. The annual average temperature of Shanghai is 15°C–22°C, which belongs to the subtropical monsoon climate. The sewage treatment plant mainly treats municipal sewage and industrial wastewater from the Baoshan District of Shanghai. The sludge samples in this study are sediments from secondary sedimentation tanks. The basic properties of municipal sludge are shown in Table 1. The water content of the municipal sludge is 87%. The organic content is 49.3%, and the permeability coefficient is 7.13×10^{-7} cm/s. The void ratio is 9.55, which indicates that the sludge has many voids, and these voids are filled with organic matter and water, which is the main reason for the large volume of the sludge. In addition, due to the complex composition of municipal sludge and high organic matter content, it is not suitable to dry at 105°C in the geotechnical test specification when determining the water content of the sludge. This is because the organic matter of sludge will be burned under high temperature, resulting in a change of the sludge engineering characteristics [23]. Therefore, in order to prevent organic particles from burning sludge at high temperature, 70°C for 48 h was used in the water content measurement test [24, 25]. Sludge samples were burned in a muffle furnace at 550°C for 2 hours in an organic matter measurement test [16].

2.2. Test Apparatus and Devices. The test equipment and instruments mainly include a microwave oven (Galanz P70F20EN3P-ZSB, 2450 MHz), centrifuge, air compressor, Brinell funnel, vacuum conversion valve, jars, catheter, vacuum gauge, valve, particle size analyzer (Malvern HYDRO), scanning electron microscope, spectrophotometer, plastic box, and polyethylene film. The vacuum filtration test device is shown in Figure 1(c).

2.3. Test Procedure

2.3.1. Municipal Sludge Treated by Acid-Microwave. The experimental procedure of this study is shown in Figure 1; the microwave oven was used to treat sludge. 500 g sludge was placed in a plastic box (205 × 140 × 70 mm), and the exposed area of the sludge was 287 cm². Since water molecules are heated under microwave radiation, the water content of

the sludge has a significant impact on the microwave treatment efficiency. For the purpose of avoiding the influence of water evaporation on sludge samples during microwave heating, the plastic box was sealed by polyethylene membrane (in Figure 1(b)). To explore the dewatering ability influenced by the microwave treatment time, the first group of sludge samples was irradiated by microwave at 700 W for 60 s, 180 s, 300 s, 420 s, and 600 s. According to the results of the first group of experiments, the sludge in the second group was adjusted to different pH values (pH 5, 4, and 3) by adding an appropriate amount of HCl standard solution (1 mol/L). After acid treatment, the sludge was radiated by microwave at 700 W for 180 s. All the treated sludge samples were cooled to indoor temperature (25°C), and then, the following tests was carried out.

2.3.2. Vacuum Filtration Test. The specific filtration resistance (SRF) is the resistance of the unit dry weight filter cake, and it is also an important indicator to evaluate the sludge dewatering ability. A high SRF value means that the sludge is difficult to filter, and the dewatering capacity is poor [26]. The mechanism diagram of the test device for measuring SRF is shown in Figure 2. During the test, 200 g sludge was poured into the Brinell funnel filled after setting a filter paper, and 75 kPa vacuum pressure was applied. The volume of the filtrate collected at different times was recorded until no additional water was discharged from the sludge [26]. The SRF can be calculated as follows:

$$r = \frac{2bpA^2}{\mu c}, \quad (1)$$

where A is the filter area, m²; c is the dry weight of filter cake obtained from per unit volume of filtrate, kg/m³; μ is the viscosity of the filtrate, N(s)/m²; p is the filtration pressure, N/m²; and b is the slope of the line between the filtration time and the filtrate volume.

2.3.3. Particle Size Analysis Test. The particle composition of sludge is one of the important characteristics. Since sludge is generated from urban sewage, it usually contains many small particles, fibrous substances, and impurities. Therefore, the traditional screening test is difficult to evaluate the particle size distribution. In this study, particle analysis was performed on the original sludge and the sludge after acid-microwave treatment. The particle size distribution of sludge samples was measured by a particle size analyzer (Malvern HYDRO) (in Figure 1(d)). The measurement results will be analyzed to explore the effect of acid-microwave treatment on municipal sludge particle size.

2.3.4. Measurement of Polysaccharide and Soluble Protein. Dewaterability of sludge is related to EPS in sludge cells. Polysaccharide and soluble protein are the main components of EPS. To explore the breaking effect of microwave radiation on EPS, the treated sludge was centrifuged at 4000 rpm for 30 min. The polysaccharide content in the supernatant was measured by the phenol-sulfuric acid method, and glucose was used as the standard substance [27]. The soluble protein

TABLE 1: Characteristic of sludge sample.

Items	Specific gravity	Water content* (%)	Bulk density (g/cm ³)	Organic content	Permeability coefficient (10 ⁻⁷ cm/s)	Void ratio
Sludge sample	1.92	87%	1.01	49.3%	7.13	9.55

*Represents the water mass divided by the sludge mass.

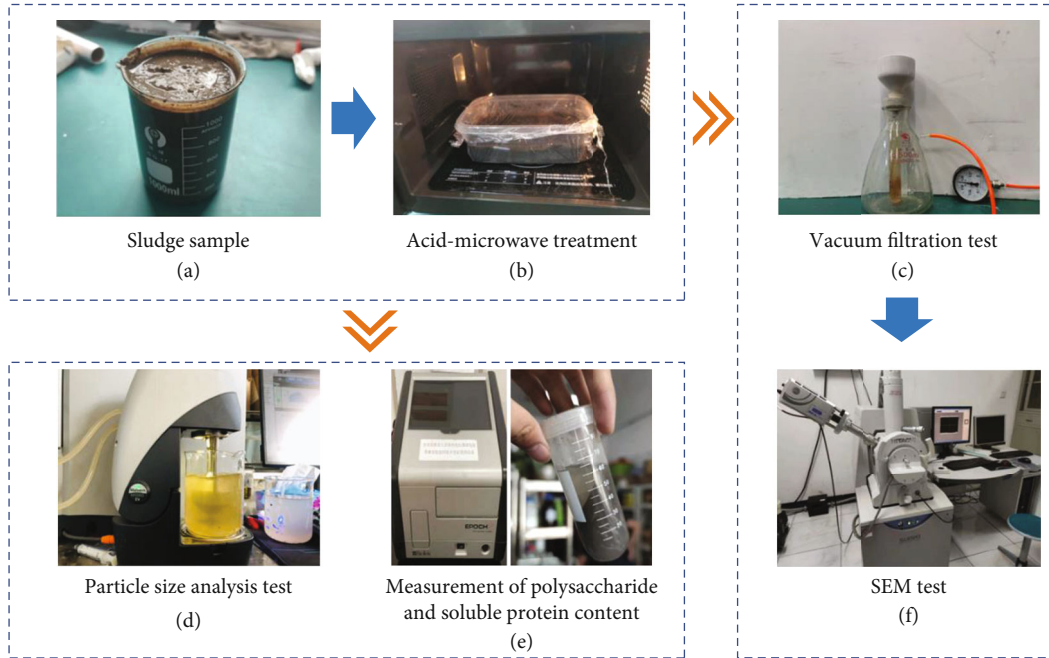


FIGURE 1: Flow diagram of this study.

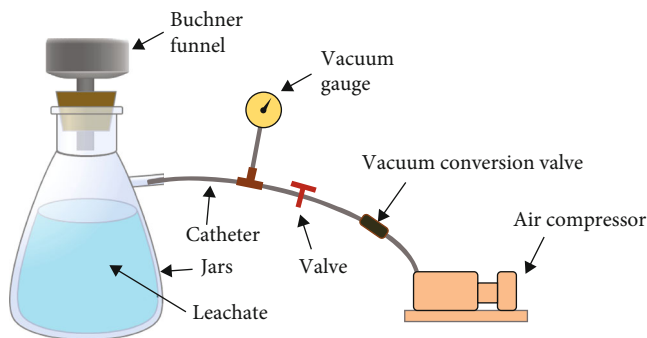


FIGURE 2: Schematic diagram of vacuum filtration test.

in the supernatant was measured by Coomassie brilliant blue staining, and bovine serum protein was used as the standard substance [28].

2.3.5. SEM Test. In order to further explore the influence mechanism of microwave treatment on sludge, it is necessary to observe the microstructure of sludge samples after vacuum filtration. After the vacuum filtration test, the sludge cake was cut into small pieces and frozen in liquid nitrogen. Then, the sludge sample was put into a freeze dryer for drying. The ice in the sludge will directly sublimate into the gas phase in a

vacuum, so the structure of the sludge will be preserved. After drying for two hours, the sludge sample was fixed on the aluminium plate for the conductive coating, and the sludge sample was observed under SEM at 2000x after the conductive coating was finished (Figure 1(f)).

3. Results and Discussion

3.1. Vacuum Filtration Test Results. The measurement results of SRF are shown in Figure 3. It can be seen from Figure 3(a) that microwave treatment significantly improves the SRF of sludge. The SRF of the original sludge is 16.36×10^{12} m/kg. After 700 W microwave treatment for 180 s, the SRF of municipal sludge is 10.11×10^{12} m/kg, which decreases by 38.2%. However, with the further increase of microwave irradiation time, the SRF of the sludge increases again, and the sludge dewatering ability deteriorates. When the microwave treatment time is 600 s, the SRF of the sludge is 14.27×10^{12} m/kg, which is only 12.78% lower than that of the original sludge.

Figure 4 is used to explain the mechanism of microwave treatment of municipal sludge. As shown in Figure 4(a), the water in sludge is mainly composed of free water, interstitial water, and bound water [29–32]. The sludge contains many hydrophilic colloid particles. Thus, interstitial water and

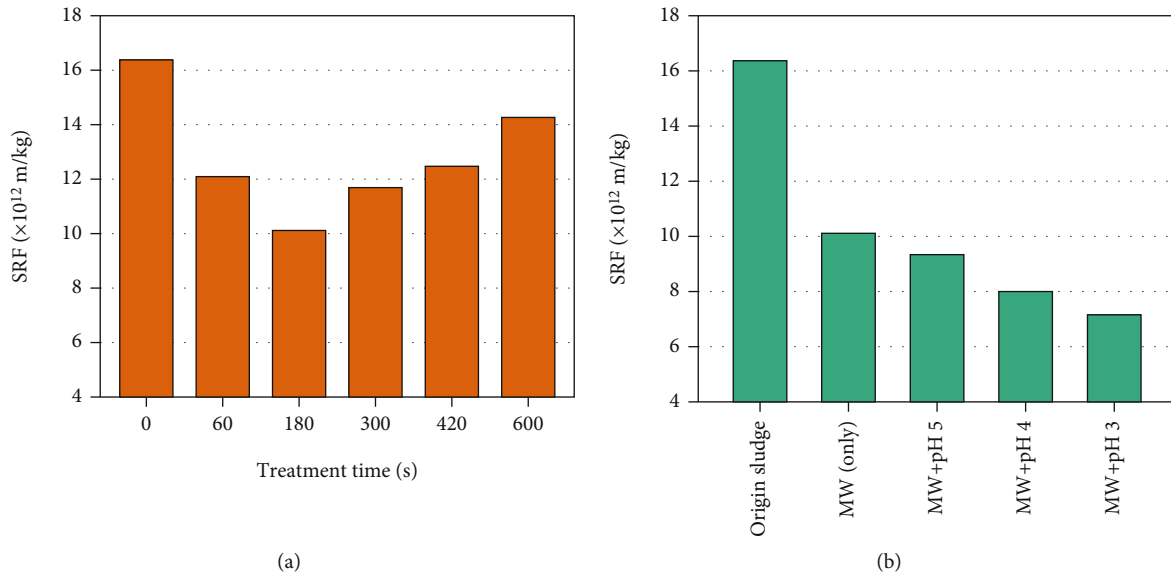


FIGURE 3: SRF measurement results: (a) SRF of sludge treated by 700 W microwave for different times; (b) SRF of sludge with different pHs treated by microwave (MW) at 700 W for 180 s.

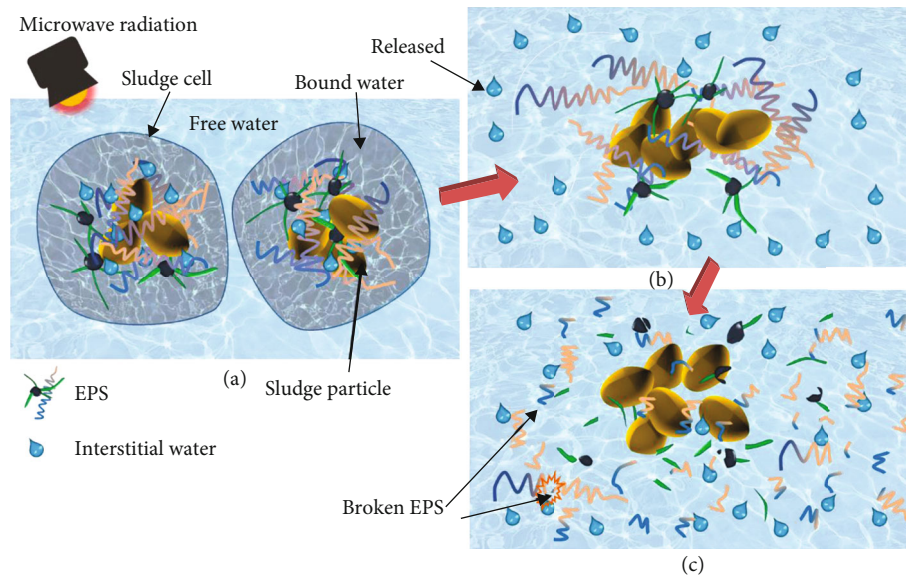


FIGURE 4: Mechanism diagram of sludge disintegration by microwave radiation.

bound water are trapped in sludge cells and are difficult to remove, which is also the main reason for poor sludge dewaterability. Therefore, breaking EPS and the floc structure, releasing interstitial water and bound water, and increasing the free water content are the most effective ways to improve sludge dewatering efficiency [33]. The reduction of the SRF is mainly because the sludge flocculation is destructed by microwave radiation [34, 35]. As shown in Figure 4(b), under microwave irradiation, the hydrophilic EPS in sludge cells are destroyed, and free water and interstitial water are released. Therefore, the free water content in sludge increases significantly, which is conducive to improving the sludge dewatering ability. However, the dewaterability deteriorates with the increase of microwave irradiation time, and it suggests that

the floc structure is excessively destroyed by microwave irradiation, resulting in the overflow of a large number of intracellular substances in the sludge. Therefore, the viscosity of the sludge also increases (in Figure 4(c)), which is also why the increase of SRF in the sludge is treated by microwave for more than 180 s [36, 37].

To investigate the effect of acid-microwave treatment on sludge dewatering capacity, a HCl standard solution was added to the original sludge (pH 6.57) to adjust the pH value of the sludge. The pH value of the sludge was adjusted to 5, 4, and 3, and then, a 700 W, 180 s microwave treatment was carried out. After microwave treatment, vacuum filtration test results are shown in Figure 3(b). With the decrease of the sludge pH value, the SRF of the sludge further decreases.

The lowest SRF is 7.15×10^{12} m/kg, which is 29.28% lower than that of the sludge without acid treatment. Compared with the original sludge, it decreases by 56.32%. It can be concluded that the acid-microwave treatment significantly improves the dewatering ability of the sludge. Li and Yang [38] pointed out that the surface of solid particles in sludge cells has many negative charges, so sludge cells are mutually exclusive and difficult to aggregate, which is not conducive to sludge filtration and dewatering. After the acid-microwave treatment, the concentration of cations in the sludge increased, and pH neutralization was achieved on the surface of the particles. As a result, the double electric layer of sludge particles becomes thinner, which weakens the repulsion in the sludge and facilitates the aggregation of sludge particles. The water molecules in the sludge cells were heated by high-speed rotation under the action of the microwave, and it will break the sludge cells and lead to the destruction of the sludge stability [39, 40]. The increase in sludge particle size provides a larger channel for the seepage of water in the sludge. The brokenness of sludge cells reduces the resistance of hydrophilic flocs to water in the seepage process. Therefore, acid-microwave treatment of the sludge has excellent potential to improve the dewatering ability of the sludge.

3.2. Particle Size Analysis. Previous studies have shown that the particle size of sludge significantly impacts the dewatering ability [41, 42]. As shown in Figure 5, the particle distribution curves of municipal sludge show a “double peak” shape; the first “peak” in the particle diameter is 10-50 μm and the second “peak” in the particle diameter is 200-500 μm . The particle size of the sludge is greatly affected by the different treatment methods. The original sludge has lots of small particle sizes. After the microwave radiation at 700 W for 180 s, the first “peak” of the sludge decreases, the second “peak” began to grow, and the particle size increases. This is because the sludge is composed of hydrophilic groups with many negative charges on the surface [43]. In the natural state, the sludge remains in the stable state because of electrostatic repulsion. Under the microwave irradiation treatment, the polar molecules in the sludge rotate rapidly, resulting in the destruction of the floc structure and the breaking of the “stable state” in the sludge; it is the reason that the particle size of the sludge will increase, and the dewatering ability will be enhanced [44]. It can be seen from Figure 4 that the particle distribution results show the same trend as the vacuum filtration test results. When the sludge was treated by acid-microwave, with the decrease of pH value, the particle size of the sludge shows an increasing trend. The second “peak” of the particle distribution curve increases, and the dewatering ability of the sludge is further improved. When pH 3, the content of large particles in the sludge is the highest, which also corresponds to the results in Section 3.1. Municipal sludge particles have negative charges owing to carboxyl and amino functional groups in EPS, which is also a reason for the stability of sludge flocs. The charge is greatly affected by the pH value. Acidification treatment leads to the decrease of negative charges in the sludge, the decrease of double-layer thickness liquid, and the decrease of zeta potential of the sludge [43, 45]. There-

fore, after the acid-microwave treatment decomposes the sludge, the sludge floc is destroyed; the sludge particles will gather into large particles because the repulsive force between the sludge particles is reduced.

In addition, the sludge particles can be divided into four particle groups: sand group (2000-75 μm), silty group (75-5 μm), clay group (5-2 μm), and colloidal group (<2 μm) [46]. The composition of the sludge particle group is shown in Figure 6. After the original sludge treated by the microwave, the sand group's content decreases from 25.6% to 24.4%, while the silty group's content increases from 66.6% to 68.3%. Due to the fact that there are many floc structures between large particles in the sludge, the floc structure is disrupted under the microwave treatment, so the content of the silty group increases [47]. In addition, the stable state of the sludge is broken by microwave treatment, so the small particles show a trend of aggregation, and the content of the colloidal group decreases from 2.53% to 1.83%, which is 27.67% lower than that of the original sludge. Karr and Keinath [41] reported that the number of colloidal particles was an essential factor affecting the filtration capacity of the activated sludge. The dewatering ability is better on account of the small proportion of colloidal particles. Because the presence of colloidal particles would increase the viscosity of the sludge, the effect of the microwave on colloidal particles is also a fundamental reason for the improvement of sludge dewatering capacity [48].

It can be concluded that the improvement of sludge dewatering ability by microwave radiation is reflected in the disruption of large particle flocs and the aggregation of colloidal particles. However, after acid-microwave treatment, the content changes of the sand group and clay group are more significant. It can be seen from Figure 6 that with the decrease of the sludge pH value, the content of the clay group decreases, and the content of the sand group increases significantly. Acid pretreatment is conducive to the aggregation of sludge particles after microwave treatment, and the treatment efficiency is significantly better than that of microwave treatment only.

3.3. Polysaccharide and Soluble Protein Content Test Results. As the most abundant substances in EPS, protein and polysaccharide are closely related to sludge dewatering characteristics and account for 70-80% of the total organic matter in EPS [49].

Under different treatment conditions, the concentrations of soluble protein and polysaccharide in the supernatant are shown in Figure 7. It can be seen from Figure 7(a) that the concentrations of soluble protein and polysaccharide in the original sludge are 13.62 mg/L and 82.68 mg/L, respectively. Under the 700 W microwave treatment, with the increase of treatment time, the soluble protein and polysaccharide content in the supernatant increases significantly. After microwave treatment for 600 s, soluble protein and polysaccharide concentrations are 35.1 mg/L and 245.14 mg/L, respectively, which increase by 157.7% and 196.5%, respectively, compared with the original sludge. It can be concluded that the EPS in sludge will be significantly cracked by microwave irradiation, and the release of polysaccharide in EPS is more obvious.

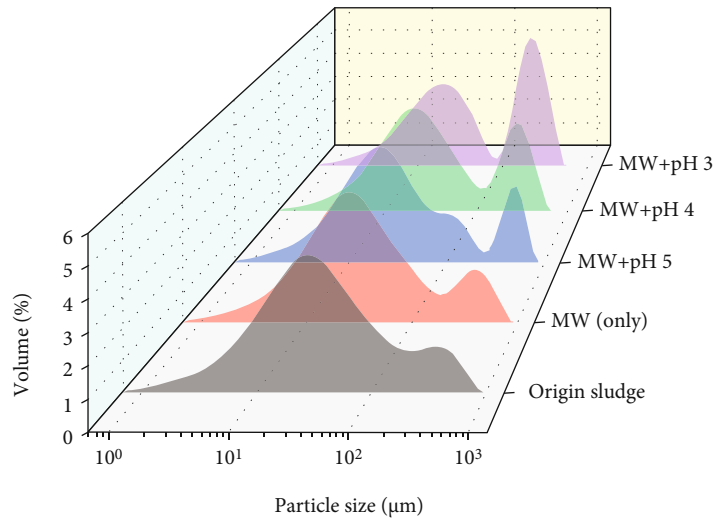


FIGURE 5: The particle size distribution curves of sludge with different pHs treated by microwave at 700 W for 180 s.

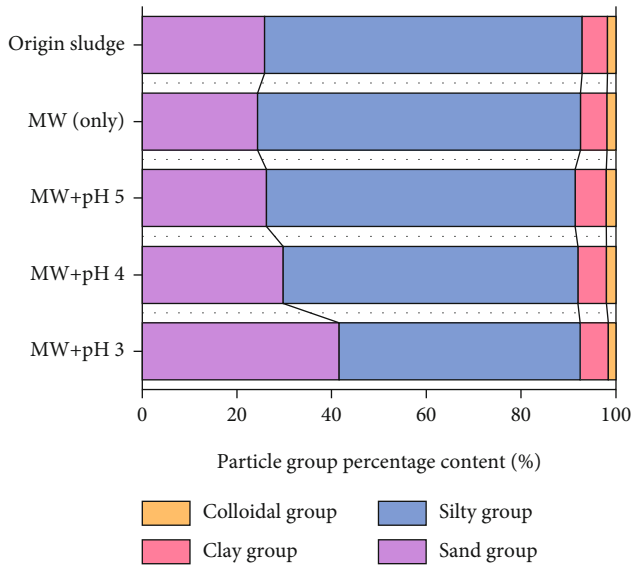


FIGURE 6: Percentage content of particle groups in sludge treated by acid-microwave.

However, when the treatment time is more than 420 s, the protein concentration does not change significantly. When the treatment time is about 180 s, the content of polysaccharide and soluble protein in EPS is moderate. The appropriate amount of polysaccharides is conducive to the dewatering performance of the sludge. Excessive radiation results in high content of protein and polysaccharide in the sludge, which will lead to the increase of sludge viscosity and the deterioration of dewatering performance. The conclusion is similar to that of SRF.

After the sludge is treated by acid-microwave, polysaccharide and protein content is shown in Figure 7(b). Compared with proteins, polysaccharides are more significantly affected by acid-microwave treatment. Under the condition of the 700 W microwave treatment for 180 s, with the decrease of the pH value in the sludge, the polysaccharide

content in the supernatant increases significantly. The polysaccharide content is 285.19 mg/L at pH 3. Compared with only microwave treatment, it is increased by 155.7%, compared with the original sludge increased by 245%. In an acidic environment, excessive fragmentation of EPS is conducive to the release of interstitial water and the reaggregation of floc fragments. The increase of the large particle content provides a larger channel for sludge drainage, which positively impacts the improvement of the sludge dewatering performance. When the sludge is treated by acid-microwave, the sludge becomes diluted substantially; it appears that the acid destroys the colloids, and the interstitial water is released. Thus, the free water content of the sludge increases, and the proteins are diluted. Under the microwave irradiation, the water of the sludge colloid is released rapidly. This also led to the dilution of protein and the decrease of the concentration. It can be seen from Figure 8 that the suspension after the acid-microwave treatment is clearer and has more volume. Qin and Liu [50] observed a similar phenomenon when using HCl to extract protein from sludge. Acid-microwave treatment of sludge can effectively improve the disintegration efficiency of EPS and improve the sludge dewatering performance.

3.4. SEM Test Results. According to the results of the vacuum filtration test, the original sludge, the sludge after microwave treatment (700 W for 180 s), and the sludge after acid-microwave treatment (pH 3, 700 W for 180 s) were selected for the SEM test.

Figure 9 shows the microstructures of the original sludge and the acid-microwave treated sludge at 2000x magnification. As shown in Figure 9(a), the original sludge has few voids between the flocs, and the particles are very tightly bound in a large floc structure, which is difficult to provide a channel for water seepage. Therefore, the bound water and interstitial water in the original sludge are captured by hydrophilic EPS, showing poor dehydration performance. After microwave treatment at 700 W for 180 s, the morphology of sludge changes significantly. As shown in Figure 9(b), the floc structure of the sludge is loose and dispersed, and the

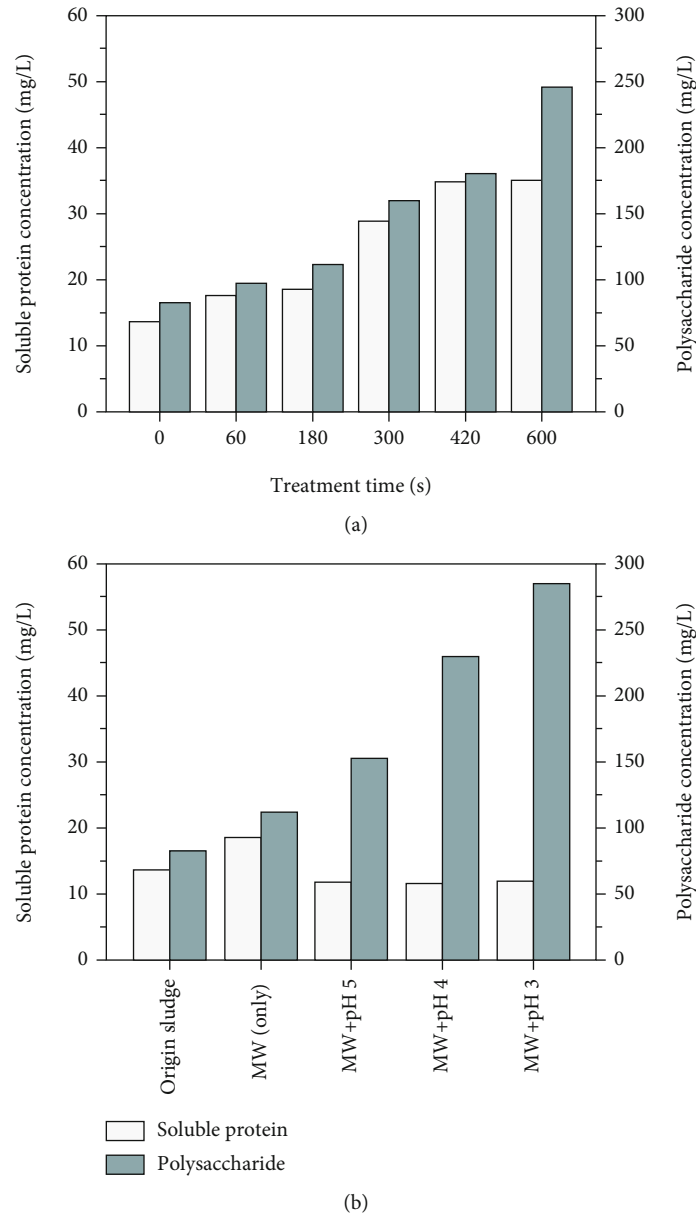


FIGURE 7: Polysaccharide and soluble protein concentration: (a) sludge treated by 700 W microwave for different times; (b) sludge with different pHs treated by microwave (MW) at 700 W for 180 s.

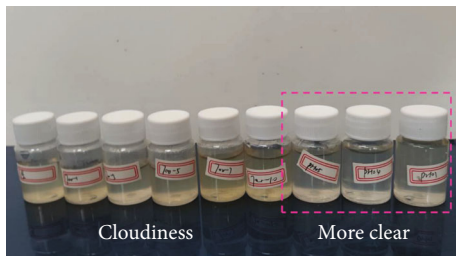


FIGURE 8: Sludge suspensions at different treatment conditions.

floc fragments are easy to find in the sludge sample. After microwave radiation, the sludge floc structure is destroyed by microwave radiation. The destruction of the floc structure will cause the release of interstitial water and bound water in

the sludge, so the dewatering performance of the sludge is improved to a certain extent. However, after acid-microwave treatment, the floc fragments of the sludge showed a trend of aggregation (in Figure 9(c)), and the number of large particles in the sludge increases significantly. Moreover, owing to the increase of large particles, the spacing and channel in the sludge become more obvious, which is more conducive to the discharge of water in the sludge; this is also the reason why the acid-microwave coupling treatment can significantly improve the dewatering performance of the sludge. The results from the SEM test are consistent with the observation from the particle size test; the sand group content decreases, and the silty group content increases when the sludge is treated by microwave (only). However, the particle size is increased after being treated by acid-microwave.

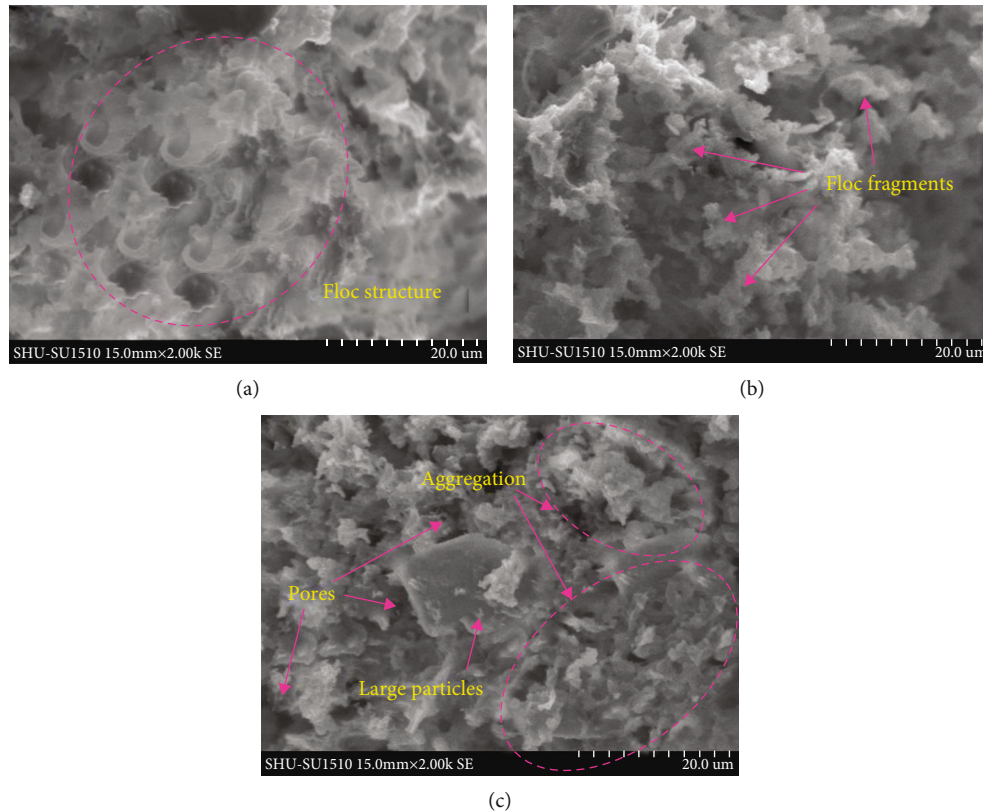


FIGURE 9: SEM photos of the samples at 2000x: (a) original sludge; (b) sludge treated by microwave (only); (c) sludge treated by acid-microwave.

4. Conclusions

- (1) Short-term microwave treatment is beneficial for sludge dewatering. However, the dewatering performance will deteriorate after sludge is overtreated by microwaves. The optimal treatment condition is 700 W microwave for 180 s. Under the optimal treatment condition, the SRF decreases by 38.2%. Acid-microwave treatment will further enhance dewatering ability. At pH 5, the SRF is 7.15×10^{12} m/kg, decreasing by 56.32% compared with the original sludge
- (2) The hydrophilic floc structure is broken after the sludge is treated by microwave. Therefore, the silty group content increases, and the sand group content decreases. Acid-microwave treatment can break the stable state of the sludge, which is conducive to the closer aggregation of sludge particles. Thus, the content of the silt group decreases, and the content of the sand group increases, which is beneficial to provide larger pores for sludge drainage
- (3) Microwave treatment can break the EPS in the sludge, and the interstitial water and bound water will be released. Suitable treatment is favorable to improve dewatering ability. However, if the microwave treatment time is too long, the EPS is excessively broken, and a large number of polysaccharides and proteins are also released, which is detrimental to sludge dewatering.

After acid-microwave treatment, the release of polysaccharides is more obvious than that of proteins. Moreover, under acidic conditions, excessive disintegration of EPS is conducive to the improvement of sludge dewatering capacity. Moreover, in an acidic environment, excessive disintegration of EPS is in favor of sludge dewatering

- (4) The results of the SEM test shows that the floc structure of the original sludge is large and loose, which is also a fundamental reason for the poor dewatering ability. After microwave treatment, the floc structure is broken, and it is easy to find floc fragments in the sludge. Under the acid-microwave treatment, the floc fragments gradually trend to aggregate and form large particles, providing more pores for sludge dewatering

Data Availability

The data support the results of this study and are reported in the table.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

The authors are grateful for the financial support for the study presented in this paper from the Shanghai Sailing Program (Grant No. 19YF1415500), National Natural Science Foundation of China (Grant No. 41772303), and National Key R&D Program of China (Grant No. 2019YFC1520500).

References

- [1] O. Nowak, "Optimizing the use of sludge treatment facilities at municipal WWTPs," *Journal of Environmental Science and Health Part a-Toxic/Hazardous Substances & Environmental Engineering*, vol. 41, no. 9, pp. 1807–1817, 2006.
- [2] A. G. Vlyssides and P. K. Karlis, "Thermal-alkaline solubilization of waste activated sludge as a pre-treatment stage for anaerobic digestion," *Bioresource Technology*, vol. 91, no. 2, pp. 201–206, 2004.
- [3] S. P. Mcgrath, "Metal concentrations in sludges and soil from a long-term field trial," *The Journal of Agricultural Science*, vol. 103, no. 1, pp. 25–35, 1984.
- [4] S. P. Mcgrath, A. C. Chang, A. L. Page, and E. Witter, "Land application of sewage sludge: scientific perspectives of heavy metal loading limits in Europe and the United States," *Environmental Reviews*, vol. 2, no. 1, pp. 108–118, 1994.
- [5] Y. Wu, X. Wang, X. Zhang et al., "Experimental study on the treatment of sludge discharged from an in situ soil washing plant by vacuum preloading," *Environmental Engineering Science*, 2021.
- [6] C/J/T249, *Disposal of Sludge From Municipal Wastewater Treatment Plant: Sludge Quality for Co-Landfilling*, Ministry of Building and Construction, P.R. China, 2007.
- [7] K. B. Thapa, Y. Qi, and A. F. A. Hoadley, "Interaction of poly-electrolyte with digested sewage sludge and lignite in sludge dewatering," *Colloids and Surfaces a-Physicochemical and Engineering Aspects*, vol. 334, no. 1-3, pp. 66–73, 2009.
- [8] Y. Q. Zhao and D. H. Bache, "Conditioning of alum sludge with polymer and gypsum," *Colloids and Surfaces a-Physicochemical and Engineering Aspects*, vol. 194, no. 1-3, pp. 213–220, 2001.
- [9] X. Zhang, Y. Lu, J. Yao, Y. Wu, Q. C. Tran, and Q. V. Vu, "Insight into conditioning landfill sludge with ferric chloride and a Fenton reagent: effects on the consolidation properties and advanced dewatering," *Chemosphere*, vol. 252, p. 126528, 2020.
- [10] G. Zhen, X. Lu, Y. Li et al., "Novel insights into enhanced dewaterability of waste activated sludge by Fe(II)-activated persulfate oxidation," *Bioresource Technology*, vol. 119, pp. 7–14, 2012.
- [11] C. W. Li, J. L. Lin, S. F. Kang, and C. L. Liang, "Acidification and alkalization of textile chemical sludge: volume/solid reduction, dewaterability, and Al(III) recovery," *Separation and Purification Technology*, vol. 42, no. 1, pp. 31–37, 2005.
- [12] M. Q. Niu, W. J. Zhang, D. S. Wang, Y. Chen, and R. L. Chen, "Correlation of physicochemical properties and sludge dewaterability under chemical conditioning using inorganic coagulants," *Bioresource Technology*, vol. 144, pp. 337–343, 2013.
- [13] Z. Q. Zhang, S. Q. Xia, and J. A. Zhang, "Enhanced dewatering of waste sludge with microbial flocculant TJ-F1 as a novel conditioner," *Water Research*, vol. 44, no. 10, pp. 3087–3092, 2010.
- [14] J. Y. Guo, L. C. Nengzi, J. Zhao, and Y. Z. Zhang, "Enhanced dewatering of sludge with the composite of bioflocculant MBFGA1 and P(AM-DMC) as a conditioner," *Applied Microbiology and Biotechnology*, vol. 99, no. 7, pp. 2989–2998, 2015.
- [15] X. Feng, J. Deng, H. Lei, T. Bai, Q. Fan, and Z. Li, "Dewaterability of waste activated sludge with ultrasound conditioning," *Bioresource Technology*, vol. 100, no. 3, pp. 1074–1081, 2009.
- [16] C. Bougrier, C. Albasi, J. P. Delgenès, and H. Carrère, "Effect of ultrasonic, thermal and ozone pre-treatments on waste activated sludge solubilisation and anaerobic biodegradability," *Chemical Engineering & Processing Process Intensification*, vol. 45, no. 8, pp. 711–718, 2006.
- [17] A. Zaker, Z. Chen, X. L. Wang, and Q. Zhang, "Microwave-assisted pyrolysis of sewage sludge: a review," *Fuel Processing Technology*, vol. 187, pp. 84–104, 2019.
- [18] E. Wojciechowska, "Application of microwaves for sewage sludge conditioning," *Water Research*, vol. 39, no. 19, pp. 4749–4754, 2005.
- [19] X. D. Zhang, E. C. Zhai, Y. J. Wu, D. A. Sun, and Y. T. Lu, "Theoretical and numerical analyses on hydro-thermal-salt-mechanical interaction of unsaturated salinized soil subjected to typical unidirectional freezing process," *International Journal of Geomechanics*, vol. 21, no. 7, 2021.
- [20] A. Gil, J. A. Siles, M. Toledo, and M. A. Martin, "Effect of microwave pretreatment on centrifuged and floated sewage sludge derived from wastewater treatment plants," *Process Safety and Environmental Protection*, vol. 128, pp. 251–258, 2019.
- [21] G. P. Sheng, H. Q. Yu, and X. Y. Li, "Extracellular polymeric substances (EPS) of microbial aggregates in biological wastewater treatment systems: a review," *Biotechnology Advances*, vol. 28, no. 6, pp. 882–894, 2010.
- [22] B. Frolund, R. Palmgren, K. Keiding, and P. H. Nielsen, "Extraction of extracellular polymers from activated sludge using a cation exchange resin," *Water Research*, vol. 30, no. 8, pp. 1749–1758, 1996.
- [23] A. Klein and R. W. Sarsby, "Problems in defining the geotechnical behaviour of wastewater sludges," *Geotechnics of High Water Content Materials*, pp. 74–87, 2000.
- [24] A. Arulrajah, M. M. Disfani, V. Suthagaran, and M. W. Bo, "Laboratory evaluation of the geotechnical characteristics of wastewater biosolids in road embankments," *Journal of Materials in Civil Engineering*, vol. 25, no. 11, pp. 1682–1691, 2013.
- [25] A. Arulrajah, M. M. Disfani, V. Suthagaran, and M. Imteaz, "Select chemical and engineering properties of wastewater biosolids," *Waste Management*, vol. 31, no. 12, pp. 2522–2526, 2011.
- [26] I. M. C. Lo, K. C. K. Lai, and G. H. Chen, "Salinity effect on mechanical dewatering of sludge with and without chemical conditioning," *Environmental Science & Technology*, vol. 35, no. 23, pp. 4691–4696, 2001.
- [27] M. Dubois, K. A. Gilles, J. K. Hamilton, P. A. Rebers, and F. Smith, "Colorimetric method for determination of sugars and related substances," *Analytical Chemistry*, vol. 28, no. 3, pp. 350–356, 1956.
- [28] Y. Q. Liu, Y. Liu, and J. H. Tay, "The effects of extracellular polymeric substances on the formation and stability of biogranules," *Applied Microbiology and Biotechnology*, vol. 65, no. 2, pp. 143–148, 2004.
- [29] J. Kopp and N. Dichtl, "Prediction of full-scale dewatering results by determining the water distribution of sewage

- sludges,” *Water Science and Technology*, vol. 42, no. 9, pp. 141–149, 2000.
- [30] J. Kopp and N. Dichtl, “Influence of the free water content on the dewaterability of sewage sludges,” *Water Science & Technology*, vol. 44, no. 10, pp. 177–183, 2001.
- [31] L. Y. Jin, G. M. Zhang, and X. Zheng, “Effects of different sludge disintegration methods on sludge moisture distribution and dewatering performance,” *Journal of Environmental Sciences*, vol. 28, pp. 22–28, 2015.
- [32] B. D. Cao, T. Zhang, W. J. Zhang, and D. S. Wang, “Enhanced technology based for sewage sludge deep dewatering: a critical review,” *Water Research*, vol. 189, 2021.
- [33] W. W. Li and H. Q. Yu, “Insight into the roles of microbial extracellular polymer substances in metal biosorption,” *Biore-source Technology*, vol. 160, pp. 15–23, 2014.
- [34] C. Eskicioglu, K. J. Kennedy, and R. L. Droste, “Characterization of soluble organic matter of waste activated sludge before and after thermal pretreatment,” *Water Research*, vol. 40, no. 20, pp. 3725–3736, 2006.
- [35] S. A. Pino-Jelcic, S. M. Hong, and J. K. Park, “Enhanced anaerobic biodegradability and inactivation of fecal coliforms and Salmonella spp. in wastewater sludge by using microwaves,” *Water Environment Research*, vol. 78, no. 2, pp. 209–216, 2006.
- [36] Q. Yu, H. Lei, G. Yu, X. Feng, Z. Li, and Z. Wu, “Influence of microwave irradiation on sludge dewaterability,” *Chemical Engineering Journal*, vol. 155, no. 1–2, pp. 88–93, 2009.
- [37] Y. Tian, L. Fang, and J. L. Huang, “Influence of microwave pretreatment on activated sludge structure and dewaterability,” *China Environmental Science*, vol. 26, no. 4, pp. 459–463, 2006.
- [38] X. Y. Li and S. F. Yang, “Influence of loosely bound extracellular polymeric substances (EPS) on the flocculation, sedimentation and dewaterability of activated sludge,” *Water Research*, vol. 41, no. 5, pp. 1022–1030, 2007.
- [39] Q. Dai, N. Ren, L. Ma et al., “Research on dewaterability and properties of sewage sludge under modified phosphogypsum and acetic acid pretreatments,” *Biore-source Technology*, vol. 264, pp. 268–276, 2018.
- [40] Y. J. Wu, Z. Zhou, Y. T. Lu, and D. A. Sun, “Effects of pH value on stability, flocculation and consolidation characteristics of waste slurry,” *China Civil Engineering Journal*, vol. 51, no. 9, pp. 92–101, 2018.
- [41] P. R. Karr and T. M. Keinath, “Influence of particle size on sludge dewaterability,” *Water Pollution Control Federation*, vol. 50, no. 8, pp. 1911–1930, 1978.
- [42] P. B. Sorensen, J. R. Christensen, and J. H. Bruus, “Effect of small scale solids migration in filter cakes during filtration of wastewater solids suspensions,” *Water Environment Research*, vol. 67, no. 1, pp. 25–32, 1995.
- [43] Y. Liu and H. H. P. Fang, “Influences of extracellular polymeric substances (EPS) on flocculation, settling, and dewatering of activated sludge,” *Critical Reviews in Environmental Science and Technology*, vol. 33, no. 3, pp. 237–273, 2003.
- [44] D. A. Jones, T. P. Lelyveld, S. D. Mavrofidis, S. W. Kingman, and N. J. Miles, “Microwave heating applications in environmental engineering—a review,” *Resources, Conservation and Recycling*, vol. 34, no. 2, pp. 75–90, 2002.
- [45] W. J. Zhang, P. Yang, X. Y. Yang, Z. Chen, and D. S. Wang, “Insights into the respective role of acidification and oxidation for enhancing anaerobic digested sludge dewatering performance with Fenton process,” *Biore-source Technology*, vol. 181, pp. 247–253, 2015.
- [46] Y. Xu, Y. J. Wu, X. D. Zhang, and G. Chen, “Effects of freeze-thaw and chemical preconditioning on the consolidation properties and microstructure of landfill sludge,” *Water Research*, vol. 200, p. 117249, 2021.
- [47] C. Cai, H. L. Liu, and M. M. Wang, “Characterization of antibiotic mycelial residue (AMR) dewatering performance with microwave treatment,” *Chemosphere*, vol. 174, pp. 20–27, 2017.
- [48] G. H. Yu, P. J. He, and L. M. Shao, “Characteristics of extracellular polymeric substances (EPS) fractions from excess sludges and their effects on bioflocculability,” *Biore-source Technology*, vol. 100, no. 13, pp. 3193–3198, 2009.
- [49] M. F. Dignac, V. Urbain, D. Rybacki, A. Bruchet, D. Snidaro, and P. Scribe, “Chemical description of extracellular polymers: implication on activated sludge floc structure,” *Water Science and Technology*, vol. 38, no. 8–9, pp. 45–53, 1998.
- [50] X. Qin and Y. Liu, “Study on the processes of protein extraction from sludge by hydrochloric acid,” *Environmental Protection Science*, vol. 38, no. 2, pp. 48–52, 2012.