Freeze–thaw combined with activated carbon improves electrochemical dewaterability of sludge: analysis of sludge floc structure and dewatering mechanism

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Received: 23 February 2021 / Accepted: 27 September 2021 / Published online: 3 November 2021
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Abstract
Freeze–thaw (F/T) and electrochemistry both are environment-friendly and efficient sludge treatment technologies. In this study, the sludge samples were frozen at −15 °C, and 20% g/gTss activated carbon (AC) was added to the dissolved sludge. Finally, the uniformly mixed sludge was treated at a voltage of 15 V for 25 min. During the experiment, the effect of F/T on the floc structure was analyzed by a laser particle analyzer and scanning electron microscope. F/T treatment improved the dewatering performance of the sludge and promoted the aggregation of sludge flocs into larger particles either. At the same time, the median diameter (D50) increased from 45.27 to 128.94 μm. AC was added to the thawed sludge solution before electrochemical treatment (EP). The conductivity of AC enhanced the effect of EP, thereby cracking the sludge flocs. Therefore, the three-dimensional excitation–emission matrix (3D-EEM) intensity of tightly bound extracellular polymeric substances (TB-EPS) decreased significantly. The protein in TB-EPS decreased from 54 to 33%, and the D50 was also reduced to 105.3 μm. The final specific resistance of filtration and water content were reduced by 96.39% and 32.17%, respectively. The dehydrated cake elemental analysis showed that increased AC improved the sludge cake’s combustion efficiency significantly. Moreover, the preliminary economic analysis indicated that the cost of this research was low, which implied the potential application value of combined treatment.

Graphical abstract

Keywords 
Freezing treatment · Electrochemical pretreatment · Carbon-based material · Waste activated sludge · Extracellular polymeric substances · Economic analysis

Introduction
In recent years, more attention has been paid in the treatment of sludge, and sludge dewatering has been an effective method for saving cost and reducing the risk of environmental pollution (Sha et al. 2021). Therefore, dewatering has become a crucial step before the disposal and utilization of sludge (Guo et al. 2019; Neyens et al. 2004; Ren et al. 2020). Among the dewatering technologies, green environmental treatment methods are fashionable among researchers, such as freeze–thaw (F/T) and electrochemical pretreatment (EP) (Guo et al. 2021).

F/T is an efficient sludge dewatering technology. The primary reason for dewatering is the formation of ice crystals during freezing (Halde 1980; Mowla et al. 2013; Tuan and Sillanpää 2010; Xu et al. 2021). In general, the freezing temperature is the primary factor affecting the
efficiency of sludge F/T dewatering, whereas the thawing conditions have little effect on the sludge dewatering performance (Wu et al. 2020b). According to the research of Vesilind and Martel (1990), slow freezing rate results in desirable dewatering performance of the sludge during freezing; the freezing rate is determined by the freezing temperature. Hu et al. (2011) also confirmed this finding in the study of sludge F/T dewatering. Sludge that has been slowly frozen at $-20^\circ\text{C}$ has a better dewatering effect than that quickly frozen at $-80^\circ\text{C}$. F/T treatment not only improves the dewatering performance of sludge but also reduces the activity of harmful microorganism cells and eliminates microorganisms either. According to Diak and Örmeci (2018), in the study of ferrate pre-treatment combined with F/T dewatering, ice crystals formed by freezing would puncture part of the microbial cells to reduce the microbial activity and the content of harmful cells in the sludge. Liu et al. (2018) studied the effects of pH and F/T on the solubility and dewatering performance of sludge and found that the solubility of sludge treated with F/T under alkaline conditions is higher and the dewatering performance of sludge treated with freeze–thaw under acidic conditions is better. The method of freezing activated nitrite pretreatment is also used in sludge conditioning by Liu et al. (2020a). It is found that it can improve sludge dewatering performance and methane production. Therefore, F/T is applied in cold areas to enhance the sludge dewatering performance, avoiding the difficulty of biological treatment and the freezing of equipment caused by low temperature (Martel 1993). Although F/T has numerous advantages in sludge treatment, the large particles of sludge formed by the extrusion of ice crystals during freezing leads to tight combination of the material and water inside the floc (Sun et al. 2018). If this tightly coupled structure is destroyed, the sludge dewatering performance will be further improved.

EP is an advanced oxidation technology, which generates strong oxidizing free radicals when electrolysis water. In recent years, it has gradually become a popular research direction in sludge dewatering (Arenas et al. 2021; Patermarakis and Fountoukidis 1990; Rajeshwar et al. 1994). During EP, electrophoresis, electromigration, and chemical reactions on the electrode will occur when an electric field is applied. These processes directly or indirectly affect the sludge flocs, prompting the extracellular polymeric substances (EPS) to release the contained substances and moisture and improve the sludge dewatering performance (Ding et al. 2021; Mahmoud et al. 2010). For example, Yuan et al. (2010) studied the effects of voltage, time, and electrode spacing on the electrochemical dewatering of sludge. When the voltage is 21 V, the electrode distance is 5 cm; the electrolysis time is 12 min; the CST reduction efficiency is 18.8% ± 3.1%. According to the research of Zeng et al. (2019), EP can not only reduce the moisture content of the sludge but also stabilize the sludge and reduce the number of microbial cells in the sludge either. When the voltage is 15 V, the cell wall is destroyed, resulting in a reduction of particle size up to 50%. The concentration of Escherichia coli and indicator pathogens dropped by 5 log10, reaching the US Environmental Protection Agency’s health standards. Compared with the traditional methods, EP has a negligible impact on the environment with a better sludge pretreatment effect; thus, it is considered as an environment-friendly sludge decomposition technology (Li et al. 2021; Yuan et al. 2010). It can be seen from the studies of these researchers that EP can not only enhance the sludge dewatering performance but also kill harmful microbial cells to stabilize the sludge. However, after a single EP, the dewatering performance of sludge still needs further amelioration (Lv et al. 2019), and reducing the energy consumption of electric dewatering is also a problem that scholars are concerned about (Anh and Sillanpää 2020). It is well known that a lot of energy is consumed in the EP process, but a higher conductivity can improve the EP treatment effect and reduce the power-on time to achieve the purpose of reducing energy consumption. Therefore, improving the conductivity in the EP process plays an important role in reducing energy consumption. Activated carbon (AC) has excellent electrical conductivity, which can improve electrochemical performance and reduce energy consumption (Hadi et al. 2015; Sheng et al. 2010). Among the electro-osmotic dewatering of sludge, Cao et al. (2019) added AC to increase the electrophoretic mobility of the current and conductivity and to improve the efficiency of electro-osmosis in the sludge solution and reduce energy consumption either. In addition, AC, as a carbon-based material, has a higher calorific value and can be used as an additive material to increase the calorific value of the final product (Liu et al. 2021; Wu et al. 2020a).

Therefore, in this study, a method of combining F/T and EP was proposed, and AC was added to improve both of the electrochemical efficiency and the sludge dewatering performance and reduce energy consumption. The operating parameters such as freezing temperature, electrochemical voltage time, and AC dosage were set on the basis of the changes in specific resistance of filtration (SRF) and water content (Wc) during the treatment. The electrical properties and structural changes of floc in the sludge were tested after F/T and EP treatments. After the treatment, the three-layer EPS in the sludge flocs were extracted to analyze the changes in the internal organic matter of the flocs and explore the dewatering mechanism. After dewatering, the calorific value of the sludge cake...
was analyzed to determine the benefits of AC. Finally, an economic analysis of the energy consumption in the research process was carried out to evaluate the potential of this research in practical applications.

**Materials and methods**

**Waste activated sludge**

The activated sludge sample used in this study was collected from a sludge thickening tank in a sewage plant in Hohhot, China. The wastewater treatment capacity of this water plant is 50,000 m$^3$/day. The retrieved samples were filtered through a 4-mm screen to remove large particles of impurities and then stored at 4 °C for 2 days to ensure that no major change in the composition of the sludge samples occurred during the experiment. The primary characteristics of the sludge are shown in Table 1.

**Experimental procedures**

**F/T treatment**

The F/T treatment was designed to simulate the effect of the cold environment on the sludge in winter, and the lowest temperature in Hohhot in winter was −25 °C. Two liters of sludge was collected and frozen at −5 °C, −10 °C, −15 °C, −20 °C, and −25 °C, respectively. Then, the sludge sample was placed in a plastic bottle and wrapped with a foam barrier to simulate an environment where the temperature gradually decreased under natural conditions. Preliminary experiments showed that the sludge was completely frozen within 18 h; therefore, 18 h was selected for all experiments. In addition, considering that long-term thawing would cause anaerobic reaction of the sludge, which would change the properties of the sludge (Örmeci and Vesilind 2001), the frozen sludge was thawed at room temperature for 12 h.

**Dosing of AC**

The AC used in this experiment was purchased from Fuchen Chemical Reagent Co., Ltd., with an average particle size of 75 μm. Before the experiment, AC was dried in an oven, sealed, and stored. One liter of thawed sludge was collected and poured into the reaction vessel. On the basis of the total suspended solid (Tss) content in the sludge, the tested dosage of AC was 20%g/gTSS (0.0028 g/L sludge) and magnetically stirred at 200 r/min for 20 min to evenly mix the AC and sludge solution without breaking the sludge flocs.

**EPS analysis**

A three-dimensional fluorescence spectrometer (F-7100, Hitachi, Japan) was used to detect the 3D-EEM spectrum of
the EPS extraction, and the EPS was extracted according to the method provided by Li et al. (2019). The primary components of sludge EPS were protein and polysaccharides, and the changes of protein and polysaccharides had a great relationship with the dewatering performance (Yan et al., 2020). Therefore, in this study, the Folin phenol method was used to determine the protein content in EPS, and the anthrone colorimetric method was used to determine the polysaccharide content in EPS, and the change mechanism of substances in EPS was analyzed.

Other methods

The multimeter was connected to the EP device to measure the current change in the electrochemical process. The treated sludge was sampled in a 100-mL graduated cylinder, and the sedimentation effect was observed after standing for 30 min. The mass percentages of [C], [H], [N], [O], and [S] were measured by an elemental analyzer (Vario EL cube, Elementar, Germany), and the calorific value of sludge was calculated by elemental analysis.

Economic analysis

Economic analysis is a necessary condition to determine whether a new technology could be applied. The cost of this study primarily included the energy consumption of the electrochemical device and the cost of adding AC. The electricity price adopted the general industrial and commercial use standard, which was $0.0854/kWh. Therefore, energy consumption was calculated according to formula (1) (Brillas et al. 2009):

Energy consumption \( \frac{\text{kWh}}{\text{L}} \) = \( \sum_{t=0}^{n} \frac{E_{\text{cell}} \cdot I \cdot t}{1000 V_{S}} \)

where \( E_{\text{cell}} \) is the applied voltage (V); \( I \) is the current (A); \( t \) is the time (h), and \( V_{S} \) is the sample volume (L).

The cost of buying AC is $16.9686/kg. Therefore, the total cost is calculated according to formula (2) (Olvera-Vargas et al. 2019):

Total Cost $/\text{L sludge} = \frac{\text{Powercost} + \text{Materialscos}}{\text{Volume of sample (L)}}

Results and discussion

Effect of EP, AC, and F/T on sludge dewaterability

Figure 1 showed that without F/T treatment, 15 V was used to control the EP during EP and EP + AC; the AC dosage was 20% g/gTss, and SRF and Wc changed with EP time. SRF and Wc decreased with the treatment time. Wc dropped to the lowest point at 20 min, from 88.93 to 85.56%. At this time, the SRF decreased from 0.535 \( \times \) 10^{13} m/kg to 0.271 \( \times \) 10^{13} m/kg. As shown in Fig. 1, EP and AC were combined after F/T treatment in advance; the EP time and voltage were controlled to 15 V and 20 min; the AC dosage was 20% g/gTss; and the SRF and Wc changed with freezing time. As shown in Fig. 1, remarkable dewatering performance was achieved after F/T + AC + EP treatment at −15 °C; SRF decreased from 0.107 \( \times \) 10^{13} m/kg to 0.066 \( \times \) 10^{13} m/kg; and Wc decreased from 77.85 to 67.42%. The reduction rate of sludge SRF was 96.39%, and the reduction rate of Wc was 32.17%, which was better than F/T and F/T + EP alone. As shown in Fig. 1, the dewatering performance of F/T + AC + EP combined treatment was significantly better than that of single treatment, and it had evident advantages compared with other studies (Gao et al. 2020; Hu et al. 2020; Yang et al. 2020; Zeng et al. 2020).

As shown in Fig. 1, the addition of AC can improve the effect of sludge EP treatment. This is because the conductivity of AC can enhance the ability of EP treatment to crack the sludge flocs, releasing more internal substances and water in the flocs (Mowla et al. 2013). After combining F/T and EP + AC, the sludge dewatering performance is further improved, but its dewatering mechanism needs to be studied. Therefore, this study will discuss the effect of F/T and F/T combined with EP + AC on the structure of sludge flocs and explore its dewatering mechanism.

Changes in sludge electrical properties and sludge morphology

Changes in current and conductivity

Sludge treatment is accompanied by changes in its properties. For example, EP will break the flocs, and the internal
substances will be released, leading to changes in current and conductivity in the electrochemical process (Lv et al. 2019; Zeng et al. 2019). Figure 2 shows the current change with time during sludge treatment, showing a state of increasing first and then becoming stable. The current detected in the EP + AC process is the largest, and the current in the F/T + EP process is the smallest. The currents obtained by EP and F/T + AC + EP are found between EP + AC and F/T + EP. This change may be due to the increased EP cracking efficiency of sludge flocs, releasing more conductive substances and increasing the efficiency of charge conduction; thus, the current is the largest in the process of EP + AC treatment. For the sludge that has undergone F/T treatment first, the conductive substances in the sludge solution are tightly compressed, resulting in a smaller current in the F/T + EP process. In the F/T + EP + AC process, with the enhancement of AC, the current will only increase slightly, but based on the SRF and Wc, the dewatering effect is best at this time. Figure 2 shows that the change trend of conductivity is basically the same as the current change. Therefore, EP and F/T affect the structure of sludge flocs and increase the dewatering performance, accompanied by changes in the properties of the sludge.

Changes in zeta potential

In addition to changes in current and conductivity during the treatment process, the release of negatively charged organic matter in each layer of EPS will also cause changes in the zeta potential in the sludge solution (Hu et al. 2020). Figure 2 shows the change trend of zeta potential with processing time and freezing temperature. The zeta potential of raw sludge is 8.85 mV, and the zeta potential when the best dewatering performance is obtained after F/T + AC + EP treatment is −15.8 mV. After treatment, the zeta potential of the
sludge increases, indicating that F/T and AC can enhance the electrophoretic mobility of the sludge during EP, and the increased electrophoretic mobility will weaken the polarity between the substances in the sludge, resulting in the reduction of hydrophilicity, thereby enhancing the dewatering performance, which is consistent with the study of Cao et al. (2019) in electro-osmotic dewatering of sludge. Moreover, according to the DLVO theory, the negative increase of the zeta potential in the sludge solution can enhance the electrostatic repulsion between the flocs and prevent the sludge from forming tighter flocs and difficult dewatering (Guo et al. 2017).

**Analysis of the sludge floc structure**

**Changes in sludge particle size**

The effect of sludge particle size on sludge dewatering performance was analyzed by measuring sludge particle size distribution using wet laser particle size...
analyzer. Figure 3 shows the cumulative percentage of sludge particles with a particle size of 0–200 μm. The cumulative percentage curve of sludge particle size after F/T treatment moves to the right, indicating that the percentage of large particles in the treated sludge is greater than that in the original sludge. The percentage curve after F/T + EP + AC treatment shifts to the left, indicating that large-particle sludge is broken into relatively small particles. Zeng et al. (2020) used Pearson's correlation analysis to determine the correlation between the median diameter D50 and the dewaterability of sludge. Therefore, by analyzing the change of the median diameter D50 during the treatment process, the relationship between the change in the size of the sludge particle size and the dewatering performance can be more intuitively reflected. As shown in Fig. 3, the D50 of the original sludge is 45.27 μm, and the D50 after F/T treatment is 128.94 μm. The D50 of the sludge treated by F/T + EP + AC is 105.3 μm, and the F/T + EP + AC treatment has the best dewatering performance. Thus, not the larger or smaller the particle size, the better the dewatering performance. Feng et al. (2009) found that CST and SRF decreased with the increase of sludge particles in the process of polyelectrolyte combined with ultrasonic treatment of sludge. This finding is due to the bridging effect of polyelectrolyte between the sludge flocs, thereby forming large sludge particles (Lee and Liu 2000). Zeng et al. (2020) found that CST and SRF decreased with the decrease of sludge particles during the electrochemical treatment of sludge, and the irreversible disintegration of sludge caused by electrochemistry was the primary reason for dewatering. In this research, the sludge after F/T treatment will form larger particles because of extrusion, and the larger sludge particles will be broken apart after EP treatment to form smaller sludge particles. Therefore, it can be hypothesized that the dewatering performance of sludge is not determined by the size of sludge particles, but the process of F/T and EP affects the structure of sludge flocs, which leads to changes in sludge particle size. In this process, internal moisture is released.

The change of sludge floc structure will inevitably lead to the change of sludge sedimentation performance. Sludge sedimentation performance is a restrictive factor of sludge dewatering performance, any attempt to improve sludge sedimentation performance is beneficial to sludge dewatering. Sludge settling performance is shown in supplementary material (Text S2).

Changes in the sludge cake structure

The analysis of the sludge cake of the original sludge and the treated sludge can enhance the understanding of the structural changes of sludge (Ramachandra and Devatha 2020). As shown in Fig. 4, the SEM image of the treated sludge cake was analyzed through two different magnifications. Figure 4 are the morphology of 500-μm sludge cake after treatment with raw sludge, F/T, and F/T + EP + AC. The whole raw sludge is finely broken and dense, with uniform particle size distribution (Fig. 4). As shown in Fig. 4, the sludge treated by F/T is frozen and squeezed to form large massive particles, and many cracks are generated for the water supply to escape. As shown in the sludge treated by F/T + EP + AC (Fig. 4), the particles of the sludge are reduced because of electrochemical action, and the particles are more loose. Figure 4 are the morphology of the sludge cake observed at 20 μm after treatment.

![Fig. 3 Changes in sludge particle size: a cumulative percentage and b change in median diameter D50](image_url)
Fig. 4 Structural change of sludge cake under different sizes: a–c are 500 μm; d–f are 20 μm

Fig. 5 Three-layer EPS three-dimensional excitation–emission matrix. (All samples were diluted by 50 times before EEM spectra measurement)
of raw sludge, F/T, and F/T + EP + AC. The surface of the original sludge is smooth without pores and cracks, which makes the removal of water difficult (Fig. 4). The sludge after F/T treatment (Fig. 4) has large irregularly shaped massive particles, voids, and cracks that can be used for water removal, which is similar to the sludge morphology under medium and low magnifications (Fig. 4). In sludge treated by F/T + EP + AC (Fig. 4), the fine pores and cracks disappeared, forming a porous and loose pore structure. The porous structure observed during the treatment is the same, which is conducive to the release of water.

**EPS analysis**

During sludge dewatering, the properties and structure of the sludge changed, which causes the sludge EPS flocs and microbial cells to crack, thereby releasing internal substances and moisture and improving the sludge dewatering performance (Huang et al. 2021; Sheng et al. 2010). Studies have shown that sludge EPS is composed of microorganisms and their metabolites, and the primary components of which are protein, polysaccharides, and other organic substances (Wei et al. 2019). 3D-EEM fluorescence spectrum analysis was performed on the extracted three-layer EPS to explore the changes of organic matter in EPS, and the results are shown in Fig. 5. Figure 5 shows four peaks (A, B, C, and D) in the original sludge TB-EPS. According to previous studies (Li et al. 2016; Lv et al. 2019), different peaks correspond to different types of protein substances. The wavelength Ex/Em of peak A is 230/350, which is in the range of aromatic proteins. The wavelength Ex/Em of peak B is 280/350, which is in the range of tryptophan-like proteins. The wavelength Ex/Em of peak C is 235/315, which is in the range of tyrosine proteins. The wavelength Ex/Em of peak D is 275/300, which is a by-product of soluble microorganisms. Based on the fluorescence spectrum in Fig. 5, the light intensity in the TB-EPS in the raw sludge is the largest, and the fluorescence intensity in the other two layers of EPS is weak. After treatment with different methods, the fluorescence intensity of each layer of sludge EPS will change differently. The fluorescence intensity in TB-EPS is all reduced, and the fluorescence intensity in S-EPS gradually increases. By contrast, in LB-EPS, the change of fluorescence intensity is small, which is similar to the change of EPS fluorescence intensity in the study by Zhen.
et al. (2012), when persulfate oxidation improves sludge dewatering. Combining the fluorescence intensities at different peaks (Table 2), the fluorescence intensities of the four substances in TB-EPS have greater changes during EP and EP + AC treatments, and more protein substances are released to S-EPS. When F/T + EP and F/T + EP + AC are processed, fewer substances are released from TB-EPS species. This phenomenon may indicate that the sludge flocs are compacted during F/T treatment, and the internal substances are more tightly bound and difficult to release. Based on the change in fluorescence intensity (Fig. 5), the internal substances of the flocs will transfer among different EPS layers during sludge treatment. According to the study of Zhang et al. (2020), proteins and polysaccharides in the internal substances of flocs are related to sludge dewatering performance.

In Fig. 6, the percentage changes of protein and polysaccharide in each layer of EPS in raw sludge and sludge treated by different methods were compared. The protein content in the original sludge TB-EPS accounts for more than 54%, and the polysaccharide content accounts for 45%. During EP and EP + AC treatment, the proportion of protein in TB-EPS has been significantly reduced to 33%, whereas the proportion of polysaccharides has not changed significantly. Murthy and Novak (1999) found that the binding effect of protein on bound water in TB-EPS is greater than that of polysaccharides; therefore, releasing more protein improves the dewatering performance of sludge. The reduced protein in TB-EPS is reflected in S-EPS, and its proportion has increased from 23 to 44%. In the treatment of F/T + EP and F/T + EP + AC, the proportion of protein and polysaccharide in TB-EPS has a small decrease, and the SRF and Wc effects of the sludge are

| Treatment | EPS fractions | Aromatic proteins | Tryptophan protein-like substances | Protein-containing tyrosine | Soluble microbial by-product |
|-----------|---------------|-------------------|----------------------------------|---------------------------|-----------------------------|
| Raw       | S-EPS         | 437.1             | 127                              | 502.4                     | 148                         |
|           | LB-EPS        | 809.8             | 126.7                            | 968                       | 297.7                       |
|           | TB-EPS        | 4321              | 4876                             | 9843                      | 2123                        |
| EP        | S-EPS         | 1357              | 251.1                            | 1618                      | 416.3                       |
|           | LB-EPS        | 799               | 292.3                            | 938.4                     | 303.6                       |
|           | TB-EPS        | 2944              | 5136                             | 8752                      | 1778                        |
| EP + AC   | S-EPS         | 3515              | 440.8                            | 5432                      | 966.3                       |
|           | LB-EPS        | 437.1             | 127                              | 502.4                     | 148                         |
|           | TB-EPS        | 1283              | 216.1                            | 1576                      | 332.1                       |
| F/T       | S-EPS         | 1804              | 3233                             | 6701                      | 1317                        |
|           | LB-EPS        | 1501              | 2895                             | 5957                      | 1220                        |
|           | TB-EPS        | 1483              | 3708                             | 7356                      | 1375                        |
| F/T + EP  | S-EPS         | 2815              | 3810                             | 8240                      | 1716                        |
|           | LB-EPS        | 2353              | 3024                             | 5650                      | 1633                        |
|           | TB-EPS        | 1842              | 3553                             | 6822                      | 1313                        |
the best. The EP + AC treatment will increase the release of substances in the aggregates formed by F/T to improve the dewatering performance of sludge. However, in LB-EPS, the proportion of protein and polysaccharides does not change significantly, which may hinder the further improvement of the dewatering performance; this result is consistent with the research of Liu et al. (2020b).

**Analysis of calorific value of sludge cake**

The calorific value is an important evaluation method for the sludge used for incineration treatment (Tan et al. 2015). The raw sludge and the sludge cake produced after treatment were collected, and the actual situation was simulated. Afterward, the sludge cake was placed in a ventilated place for 24 h for drying and then sampled. The moisture content was measured, and an elemental analyzer was used to determine the [C], [H], [N], [S], and [O] in the sludge cake. The percentage of element content was calculated using the formula provided by Coskun et al. (2020) to determine the HHV and LHV of the sludge cake, such as Formulas (3) and (4):

\[
HHV = 78.33C + 338.89(H - O/8) + 22.21S + 5.78N \quad (3)
\]

\[
LHV = HHV - (9H + H_2O) \cdot 5.8278 \quad (4)
\]

[C], [H], [N], [S], and [O] in the formula are the mass percentages of carbon, hydrogen, nitrogen, sulfur, and oxygen in the sludge, respectively, and H_2O represents the percentage of water in the sludge. HHV is the general term for the heat of combustion during fuel combustion and the heat of condensation generated by the moisture. LHV is the heat of combustion only generated by the fuel when the moisture is completely evaporated. A higher LHV represents a better fuel production effect. Table 3 shows the HHV and LHV of the raw sludge and treated sludge cake. The HHV and LHV of the original sludge are 1806.79 kcal/kg and 1045.04 kcal/kg, respectively, which are both smaller than the treated sludge (299.83 kcal/kg and 2285.40 kcal/kg), which increased by 66.04% and 118.79%, respectively. This result is due to the AC added during treatment, which is a carbon-based material with high calorific value (Wang et al. 2020). Using AC as an auxiliary additive can improve not only the EP treatment effect and dewatering performance of the sludge but also the final sludge. The combustion performance of the cake is conducive to the subsequent disposal and utilization of the dehydrated cake.

**Economic analysis**

A new technology required preliminary economic analysis before its application. The cost of sludge pretreatment was calculated to be $0.0382/m^3, which included the power consumption during the pretreatment and the cost of AC. Olvera-Vargas et al. (2019) spent $25.11/m^3 in the study of electrochemical peroxidation–electro-Fenton (ECP-EF) treatment of anaerobic sludge (excluding dry sludge disposal and anode consumption). Zhou et al. (2015) found that the traditional Fenton method adjusted the sludge cost to $0.0295/m^3. Assuming that the service population of the sewage treatment plant is 100,000, the total cost of the two methods is compared in Table 4. The Fenton method required reaction under acidic conditions. The experimental conditions were harsh and expensive. In this study, the reaction conditions were loose; no chemical agents were used; no pollution was generated, and costs were saved.

| Table 3 | Changes of element mass percentage and calorific value in sludge cake before and after treatment |
|---------|-------------------------------------------------|
| Samples | H_2O (%) | Element mass percentage (%) | Calorific value (kcal/kg) |
| RAW | 91.33 | 20.11 | 3.48 | 3.39 | 0.37 | 23.03 | 1806.79 | 1045.04 |
| F/T + AC + EP | 67.42 | 28.91 | 6.13 | 5.0 | 0.45 | 32.60 | 2999.83 | 2285.40 |

| Table 4 | Cost analysis of various dewatering methods |
|---------|---------------------------------------------|
| Treatment methods | Power cost ($/m^3 sludge) | Material cost ($/m^3 sludge) | Chemical cost ($/m^3 sludge) | Total cost ($/year) | References |
| EP | 0.48 | - | - | 8,760,000 | Zeng et al. (2020) |
| F/T + AC + EP | 0.0336 | 0.00464 | - | 697,883.18 | This study |
| ECP-EF | 7.83 | - | 17.28 | 458,257,500 | Olvera-Vargas et al. (2019) |
| Classical Fenton | - | - | 0.0295 | 538,000 | Zhou et al. (2015) |

**Conclusion**

This research combines F/T, EP, and AC and explores the potential application of combined treatment in sludge dewatering, aiming to improve the dewatering
performance of sludge and increase the utilization efficiency of dry sludge cake through green and energy-saving treatment technology. The analysis of the sludge floc structure during the research shows that the dewatering performance of the sludge is not always positively or negatively correlated with the size of the sludge, but the sludge is released when EP and F/T change the sludge floc structure. The internal moisture is removed, and the sludge particle size changes. Through EPS analysis, F/T will make the internal materials of EPS more closely combined, and EP has the opposite effect, cracking EPS and releasing internal materials. AC can further enhance the treatment effect of EP by enhancing the electrophoretic mobility during EP. Therefore, this study uses the squeezing effect of F/T and the cracking effect of EP, combined with AC to treat water-containing sludge and improve the dewatering performance of sludge. Under optimal conditions (−15 °C, 15 V, 25 min, and 20% g/gTss), the sludge was conditioned; the Wc of the sludge cake was reduced from 99.39 to 67.42%; and the SRF was reduced from 1.83 × 1013 m/kg to 0.066 × 1013 m/kg. The reduction rates were 32.17% and 96.39%, respectively, and good treatment effects were obtained. In addition, the calorific value of the sludge after the addition of AC has been improved, which promotes the utilization efficiency of the sludge cake, and the economic analysis also shows that this research consumes less energy and exhibits application value.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11356-021-16837-4.

Acknowledgements The authors wish to thank the Gongzhufu wastewater treatment plants for supply of samples for testing.

Author contribution Data analysis and the lead in writing the manuscript were performed by KH. LS contributed to the study conception and design. ZY contributed to writing. HS contributed to theory and helped shape manuscript structure. Material preparation was performed by ZW. Data collection was performed by WG. LX commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Funding This study was financially supported by the National Natural Science Foundation of China (No. 21107041) and the Natural Science Foundation of Inner Mongolia (No. 2020MS05028).

Data availability All data generated or analyzed during this study are included in this published article.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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