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Review

Advances in Chemical Conditioning of Residual Activated Sludge in China

Yankai Liang, Rui Wang, Wenquan Sun and Yongjun Sun *

College of Urban Construction, Nanjing Tech University, Nanjing 211800, China
* Correspondence: sunyongjun@njtech.edu.cn

Abstract: Municipal sludge is characterized by high organic matter content, high viscosity, and fine particles, resulting in poor dewatering performance. This article analyzes the composition and properties of municipal sludge, examines the factors affecting the dewatering performance of sludge and the mechanisms corresponding to each influencing factor, and introduces chemical conditioning in detail. Chemical conditioning includes flocculation conditioning, oxidation conditioning, acid-base conditioning, and aggregate conditioning. The principles and applications of existing sludge conditioning technologies are systematically analyzed. By comparing the advantages and disadvantages of different technologies, it is pointed out that the key to developing sludge conditioning technology lies in developing a more appropriate combination of the sludge conditioning and dewatering process according to the sludge quality of different municipal wastewater treatment plants, taking into account their local environment, input costs, subsequent sludge disposal methods, and other factors, and further optimizing the sludge dewatering process by developing new efficient and environmentally friendly sludge conditioning agents.

Keywords: sludge conditioning; sludge dewatering; dewatering performance; sludge water content

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1. Introduction

With rapid economic development and urbanization, the volume of wastewater treatment in urban and rural sewage treatment plants is also increasing [1]. According to the 2020 Urban and Rural Construction Statistical Yearbook released by China, as shown in Figure 1, the total annual wastewater treatment in Chinese cities reached 5.7 billion cubic meters by the end of 2020, and the wastewater treatment rate of urban sewage treatment plants reached 97.53%. The primary sediment produced in the primary settlement tank and the secondary sediment produced in the secondary settlement tank in the wastewater treatment are collectively referred to as sludge, which is the main by-product of wastewater treatment [2]. The type of sludge produced by municipal sewage treatment plants is municipal sewage sludge. Since the annual volume of municipal wastewater treatment increases year after year, the sludge production also increases significantly [3]. According to the empirical formula of 1.5 tons of sludge dry matter from treating 10,000 tons of wastewater, China will produce about 4,285,200 tons of sludge with 80% water content in 2020. The first step in the process of promoting reduction and ingenuity in sludge treatment and disposal is sludge dewatering. Therefore, the promotion of sludge treatment and disposal, stabilization, harmless and gentle treatment, and disposal of sludge from sewage treatment plants is of positive importance for the stable operation of sewage treatment plants and energy saving and CO_2 reduction.

Sludge dewatering is an important part of sludge treatment and disposal. Due to the high organic matter content, good hydrophilicity, and fine floc particles of municipal sludge, it has been difficult to reduce the water content of the sludge below 60% [4]. As shown in Table 1, China has introduced a series of regulations that impose specific requirements on the water content of sludge under various treatment and disposal conditions. In practical engineering, sludge dewatering without conditioning is very poor [5]. The more difficult moisture to remove in mechanical sludge dewatering is bound to water that cannot be removed by simple mechanical pressure [6]. In China's municipal sewage treatment plants, inorganic polymeric coagulants such as polymeric aluminum chloride (PAC) and organic polymeric flocculants such as polyacrylamide (PAM) are generally added to thickened sludge for sludge conditioning prior to dewatering. Due to the high organic content and viscosity of municipal sewage sludge, the conditioned sludge is usually dewatered in a plate and frame filter press. After dewatering by the above process, the water content of the sludge can be reduced to 70–80%, which is much higher than the national requirement of 60% water content [7]. In addition, sludge conditioning agents such as PAM and lime are currently expensive, and subsequent transportation of the sludge cake and disposal costs such as landfill and incineration are high due to the large volume of dewatered sludge cake [8]. For this reason, sludge conditioning before dewatering is a very important part of the process to achieve the desired dewatering effect, stabilize the sludge, make it harmless and resource-saving, reduce the cost of sludge treatment, and increase the safety of the subsequent disposal of the sludge mud. The development of efficient and environmentally friendly sludge conditioning agents and the rational use of various sludge conditioning methods in combination with conditioning is an urgent task.

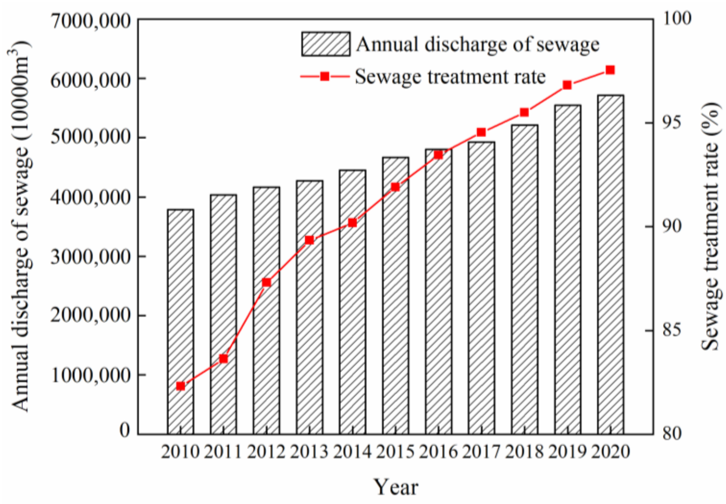

Figure 1. Annual sewage discharge and sewage treatment rate in China, 2010–2020.

Sludge conditioning is the use of physical, chemical, and biological methods to reduce the water content of sludge by applying energy or chemical reagents to the sludge, destroying the floc structure, changing the surface structure and charge of the floc mud, and weakening the interaction between the mud particles to create sufficient conditions for to create the subsequent sludge dewatering while releasing part of the intracellular water and pore water from the sludge [9]. The sludge conditioning process can be divided into physical and pore water processes. Sludge conditioning processes can be divided into physical, chemical, and biological conditioning [10]. Chemical conditioning includes floc conditioning, oxidation conditioning, acid-base conditioning, aggregate conditioning, etc. The principles of sludge conditioning are generally based on the following three principles [11]. 1. Improving the settling properties of sludge through adsorption bridges and electro-neutralization reactions by adding coagulants/flocculants [12]. 2. Deep drainage through oxidation reactions that destroy extracellular polymers (EPS) and release bound water [13]. 3. Building a skeleton structure reduces the compressibility of the sludge, builds the water filtration channel of the sludge cake, and reduces the viscosity of the sludge, thereby improving the dewatering performance of the sludge. The chemical method has the advantages of rapid reaction, simple addition of equipment, and convenient operation. At the same time, it also has the limitations of corroding equipment, causing secondary pollution, and difficult to determine the dosage.

Table 1. Requirements for sludge moisture content for different treatment and disposal methods in China (partial).

Chinese Standards	Standard Number	Requirements for Sludge Moisture Content
Pollution control standards for domestic waste landfills	GB 16889-2008	The water content of the sludge to be disposed of in a domestic landfill must be less than 60%
Pollutant Discharge Standards for Urban Sewage Treatment Plants	GB 18918-2002	The water content of sludge for aerobic composting should be less than 65%; urban wastewater treatment plants should dewater the sludge and the water content should be less than 80%.
Municipal wastewater treatment plant sludge disposal—mixed landfill sludge	CJ/T249-2007	The moisture content of the sludge should be less than or equal to 60% when used in mixed landfill applications and less than 45% when used as landfill cover
Municipal sewage treatment plant sludge disposal—sludge for land improvement	CJ/T291-2008	When sludge is used as a land improvement sludge the moisture content should be less than 65%.
Municipal sewage treatment plant sludge disposal—brick making sludge	CJ/T289-2008	When sludge is used for brick making the moisture content should be less than 40%

This work provides an overview of floc and water composition and properties in municipal sludge and analyzes the factors affecting sludge dewatering and their respective mechanisms of action in relation to five aspects: sludge zeta potential, sludge particle size, rheological properties, floc properties, and EPS properties, and examines the chemical method of sludge conditioning. Chemical processes include coagulation and flocculation, oxidation treatment, acid-base processes, and aggregate conditioning. The technical parameters and treatment efficiencies of the different sludge conditioning methods are summarized, the mechanisms of each conditioning method are studied, and their advantages and disadvantages and adjustment conditions are analyzed. Geographical conditions, inherent sludge properties, intended use, level of economic development, and climatic conditions should be considered before selecting a sludge conditioning process for any municipal sewage treatment plant. This report can provide industry researchers with a summary of their experiences and new ideas for using a combination of sludge conditioning processes for municipal sludge with a view to developing a more efficient, economical, and environmentally friendly sludge conditioning process for dewatering. At present, the goal of chemical conditioning is to further optimize the treatment process of sludge dewatering by developing a new type of highly efficient and environmentally friendly sludge conditioner to achieve high efficiency and low energy consumption.

2. Composition of Municipal Sludge

The composition and properties of sludge largely depend on the quality of the water treated in the treatment plant and can generally be divided into two types: municipal sludge and industrial sludge. Municipal sludge is the sludge produced by domestic sewage treatment plants, which has a higher organic content and a relatively low content of toxic and harmful substances such as heavy metals and persistent toxic and harmful organic substances compared to industrial sludge [14]. The classification of municipal sludge is shown in Table 2, which is divided into primary sludge (96–98% water content), residual sludge (99% water content), thickened sludge (95–97% water content), and digested sludge (95% water content), depending on the mud source. The sludge consists of sludge flakes and water; the composition of the sludge flakes is shown in Figure 2. The sludge flocs are mainly composed of microorganisms, including protozoa, filamentous bacteria, microcolonies, organic fibers, inorganic particles, and extracellular polymeric substances (EPS). The water in the mud is distributed in four forms, as shown in Figure 3: bound water, interstitial water, surface adsorbed water, and chemically bound water. Research manuscripts reporting large data sets deposited in a publicly accessible database should indicate where the data is deposited and provide the appropriate accession numbers. If accession numbers are not available at the time of submission, please indicate that they will be provided during review. They must be provided prior to publication [15].

Table 2. Classification of municipal sludge.

Sludge Type	Sludge Sources	Moisture Content
Primary sludge	Sedimentation tanks and primary settling tanks	96–98%
Residual sludge	Bioreactors and secondary sedimentation tanks	99%
Thickened sludge	Sludge thickening tank	95–97%
Digestion of sludge	Sludge after digestion	95%

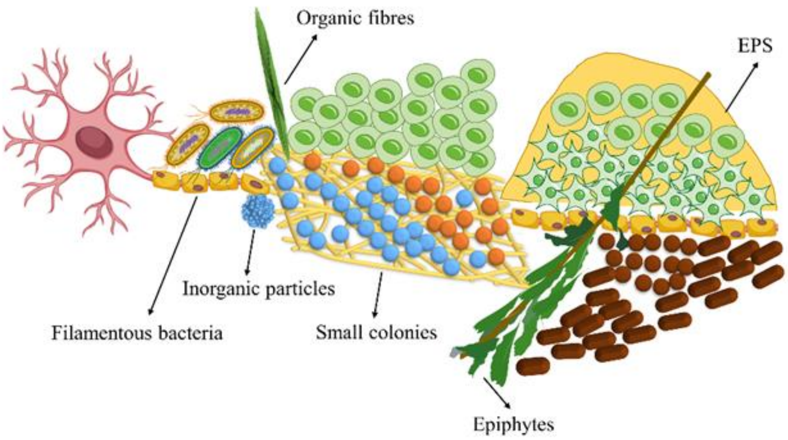

Figure 2. Schematic diagram of municipal sludge flocs.

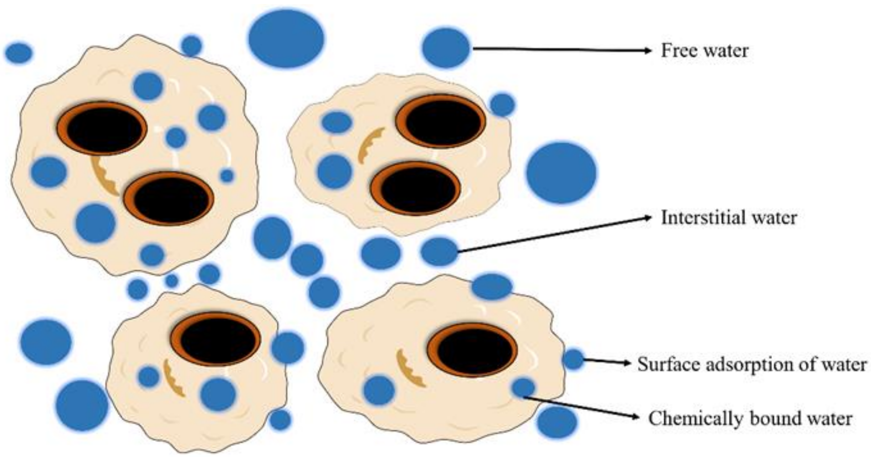

Figure 3. Form of water present in sludge.

Municipal sludge consists primarily of organic fibers, inorganic particles, cells or microcolonies, and colloidal EPS material. The mud flocs have a high surface area and porosity, and the floc structure is relatively loose and held together by van der Waals forces, electrostatic attraction, adsorption bridge forces, and hydrophobic physical bonding. Where EPS is the product of cell division and macromolecular hydrolysis, it also contains adsorbed material from the wastewater treatment process. It mainly includes substances such as proteins with macromolecular composition, polysaccharides with long-chain structure, nucleic acids, and lipids, which are organic colloids with a high degree of hydrophilicity, high specific surface area, poor drainage performance, and the mass of which accounts for 40–60% of the dry weight of the sludge. The main components of inorganic particles in residual sludge include some soil particles, soil colloids, inorganic salts (N, P, K, etc.), inorganic nutrients, heavy metals, etc. It features a variety of sources, diversity, complex morphology, and easy migration and accumulation.

Water is the largest component of sludge and the water content of all sludges in wastewater treatment plants is typically between 90 and 99%. The water in the mud can be easily divided into two categories, free water and bound water, depending on the strength of the water's interaction with the mud floc. Free water is the water in the mud that is unaffected by solid particles and whose properties have not changed; bound water is the water that is physically or chemically bound to the floc and whose chemical potential energy is different from that of free water. As shown in Figure 3, there are three types of bound water depending on how it is bound to the mud flocs: 1. interstitial water: water bound between the mud flocs and between the pores; 2. surface adsorbed water: water bound to the surface of sludge flocs by physical action; 3. chemically bound water: water chemically bound to the solid particles of the mud floc.

3. Factors Influencing Sludge Dewatering Performance

3.1. Sludge Zeta Potential

The zeta potential is a measure of the forces of attraction or repulsion that exist between particles and represents the stability of the colloidal system to be measured. The zeta potential of mud is generally in the range of −30 to −10 mV. The higher the absolute value of the zeta potential, the more negatively charged the sludge, the more stable the sludge system, and the poorer its dewatering performance. The larger the extracellular protein/extracellular polysaccharide, the higher the sludge zeta potential, indicating more ionized polymers on the sludge flake surface, which increases the hydrophilicity of the sludge, resulting in more bound water in the sludge floc, which is detrimental to the sludge dewatering affects. Acidification of the sludge generates H^+, which neutralizes the negatively charged functional groups on the sludge surface, while the extracellular polymers of the sludge also detach from the cells under highly acidic conditions, resulting in an increase in the zeta potential of the sludge. Studies have shown that flocculants condition sludge primarily by reducing electrostatic repulsion between sludge particles through electro-neutralization and compressive double layer effects that destabilize sludge colloids, reduce the sludge's zeta potential value, improve sludge settling properties, and facilitate dewatering treatment. Sun et al. [16] conditioned the sludge with various cationic levels of polyacrylamide and the results showed that the zeta potential was positive and continued to increase with increasing dose. The higher the cationic degree of the flocculant, the better and stronger the charge neutralization ability. The zeta potential of the starting sludge without flocculant was −9.04 mV. When the flocculant was added, the negative charge in the sludge was quickly neutralized and the zeta potential changed from negative to positive, resulting in a significant improvement in the dewatering performance of the sludge.

3.2. Sludge Particle Size

The sludge particle size is a direct indication of the size of the sludge floc particles and to some extent reflects the aggregation or dispersion of the sludge. In general, the greater the fines content in the sludge, the greater the sludge's viscosity per solids concentration, and the greater its water-binding ability, the poorer the dewatering of the sludge. At the same time, these fines can clog the sludge dewatering channels and degrade the dewatering performance of the sludge. The filtration and densification properties of sludges are usually improved by adding flocculants and coagulants to increase the proportion of large colloidal particles in the sludge. If the particle size of the sludge floc is too large, not only will the filter sludge increase in volume after dewatering, but also chemicals will be wasted, and the sludge will not be dewatered. This suggests that both large and small particle sizes are detrimental to sludge dewatering. Studies have shown that the larger the D50 (the particle size corresponding to a sample with a cumulative particle size distribution of 50%), the better for sludge dewatering. Xia et al. [17] used fly ash in conjunction with polymeric iron sulfate and polymeric aluminum chloride to treat residual sludge and showed that the flocs of the original sludge were relatively fluffy. The addition of complex conditioning agents can neutralize the negative charge on the surface of the flakes and reduce the repulsion between colloidal ions, making it easier for the flakes to collide and aggregate, thereby forming flocculated particles with large particle size and dense structure. The best results are obtained with fly ash coupled with polymeric iron sulphate where D50 reaches 2.92 µm in sludge particle size.

3.3. Rheological Properties

The rheological properties of sludge are a composite indicator of the interaction between sludge particles, the colloidal stability of microbial agglomerates, and the strength of the floc network, which can be influenced by its physicochemical properties. The parameters that describe the rheological properties are generally: viscosity, elastic modulus, viscous modulus, yield stress, etc. Temperature has been shown to be one of the key factors affecting the flow properties of mud and can greatly affect the rheological properties, by changing the size, shape, and dispersion of the sludge floc particles, thereby affecting the dewatering performance of the sludge. Hill et al. [18] showed that the sludge after thermal hydrolysis pretreatment still had viscoelastic properties similar to the original sludge and that the energy storage and loss moduli decreased with increasing treatment temperature. The pH also had a significant impact on the flow properties of the sludge, as the network strength and surface charge of the sludge particles changed with decreasing pH. Wang et al. [19] studied the effect of floc structure and surface properties on sludge dewatering and rheological properties in low pH environments by acidifying sludge and found that microbial cells in sludge from acidic environment undergo hydrolysis, thereby reducing the content of bound water and significantly changing the rheological properties of the mud.

3.4. Floc Characteristics

The floc structure of mud is closely related to drainage performance, and the mud fractal dimension is typically used to characterize the structural shape of mud. The fractal dimension D ranges from 1 to 3, with larger values indicating higher sludge density and better sludge dewatering performance. The fractal dimension of the mud flakes is mainly related to the physical and chemical properties of the mud itself and the reaction formation process. The addition of coagulants and flocculants can change the properties of the flocs, so that the originally loose and broken flocs of mud can be aggregated into large flocs of mud through adsorption bridging and charge adsorption to improve the settling and compressibility of the mud flocs. The dewatering performance of sludge can be improved by adding fly ash, red mud, diatomaceous earth, etc., to form a skeletal structure in the sludge floc and form hydrophobic channels [20]. A sludge dehydrating agent, carboxymethyl chitosan-grafted polyacrylamide-methacryloxyethyl trimethyl ammonium chloride (CCPAD), was prepared by UV-induced graft polymerization using carboxymethyl chitosan (CMCS), acrylamide, and methacryloxyethyl trimethyl ammonium chloride as the copolymer monomers. The best sludge dewatering performance was obtained when CCPAD was used as a sludge conditioner with pH between 5.0 and 6.0, a characteristic viscosity of 1267 mg/L and a concentration of 20 mg/L. The turbidity of the supernatant was 6.54 NTU, the water content was 76.26% and the filtration specific resistance was 1.09×10^{13} m/kg. CCPAD can increase the sedimentation behavior and compressibility of sludge through adsorption bridges and electrical neutralization. CCPAD can increase sludge settling and compressibility through adsorption bridging and electro-neutralization, bringing the originally loose and broken sludge flocs together to form larger, more compact sludge flocs. This improves the dewatering performance of the sludge.

3.5. EPS Characteristics

EPS is a product of cell division and macromolecular hydrolysis that contains adsorbed material from the wastewater treatment process. EPS is generally distributed outside the cells as well as wrapped around the microbial matrix of the sludge. It accounts for about 80% of the total sludge and 50–60% of the organic matter content in the sludge. The mud EPS components include proteins, polysaccharides, humic acids, and nucleic acids. Based on the spatial distribution properties and compactness of EPS, as shown in Figure 4, EPS can be divided into dissolved EPS (S-EPS), loosely bound EPS (LB-EPS), and tightly bound EPS (TB-EPS) from the outside in. The amount of sludge EPS is negatively correlated with sludge dewatering performance, and the amount of protein and humic matter affects the amount of negative charge carried on the surface of the sludge floc, which in turn affects the dewatering performance of the sludge. Ge et al. [21] developed the PFS/O_3 process as a novel environmentally friendly method to break down sludge flocs and EPS to improve dewatering performance. Deep dewatering of the sludge is easily accomplished when the optimal dosage levels of PFA and O_3 are 40 mg/g TS and 60 mg/g TS, respectively. The results of the PFS/O_3 sludge treatment showed that the percentage of dead cells in the sludge was 47.3%, indicating that the microbial cells suffered greatly and that the proteins and polysaccharides in EPS were released through hydrophobic channels, to improve the dewatering performance of the sludge. The interaction between the EPS and the sludge cells had a significant impact on the coagulability of the microorganisms. The coagulation mechanism of cationic coagulants is based on adsorption bridges and two-electron layer compression, and ionic bridges between multivalent cations (including Ca^{2+} and Mg^{2+}) and EPS are also considered as a key factor in sludge flocculation. Chen et al. [22] studied a hydrothermal process in combination with $FeSO_4$-$7H_2O$/$Ca(ClO)_2$ for sludge conditioning to improve sludge dewatering performance. The results showed that the surface water content of the mud decreased and the free water content increased significantly under the hydrothermal method and $FeSO_4$/$Ca(ClO)_2$, indicating that the EPS in the mud was degraded and part of the water was absorbed in the EPS was published.

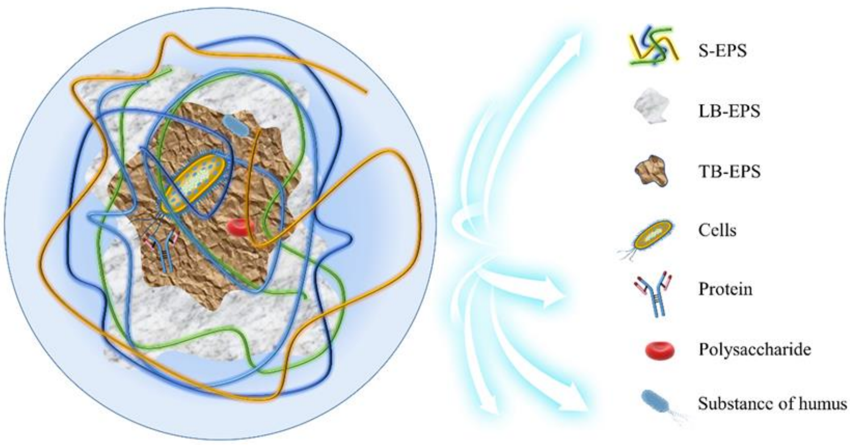

Figure 4. Schematic diagram of the EPS structure.

4. Chemical Conditioning

4.1. Flocculation Conditioning

Coagulation and flocculation is one of the most common methods of sludge conditioning in sewage treatment plants today. During coagulation and flocculation, small colloidal particles from large and dense flocs, which helps accelerate floc settlement, increase drainage rate, and increase the solid content of the sludge cake. In the coagulation process, the charge on the surface of the fine suspended colloids is weakened by the neutralization of the oppositely charged coagulant, which destabilizes the aggregation and precipitation. To improve the bridging effect, flocculants with longer molecular chains can be added. Through the functional groups with a special effect on the molecular chain, and the colloidal particles through adsorption bridge action, the colloidal particles form flakes and quickly settle.

As shown in Figure 5, electro-neutralization and adsorption bridging are two important mechanisms in the coagulation and flocculation process. Electro-neutralization effectively destabilizes the charged particles in the sludge, causing them to aggregate and precipitate, followed by mechanical dewatering. In addition, electro-neutralization reduces the surface tension of the water by compressing the two-electron layer of the colloidal mud particles, reducing the amount of bound water. Adsorption bridging can be explained by two mechanisms: adsorption and bridging. Electrostatic gravitational forces and hydrogen bonding are usually the forces responsible for adsorption. Electrostatic gravity refers to the strong adsorption of polymeric flocculants with oppositely charged colloidal mud particles under the influence of electrostatic gravity. The polymer flocculant can be adsorbed onto the surface of the colloidal particles through hydrogen bonding. The principle of bridging means that the polymeric flocculant adsorbs to the surface of the colloidal sludge particles in lower concentrations and is simultaneously adsorbed to other colloidal sludge particles, thereby binding multiple colloidal sludge particles together, similar to bridging, and forming a flocculation. There is a wide range of coagulants/flocculants that can be classified according to structural properties, charge properties, ionic strength, functional groups, and molecular weight, and different coagulants/flocculants are used for different treatment goals.

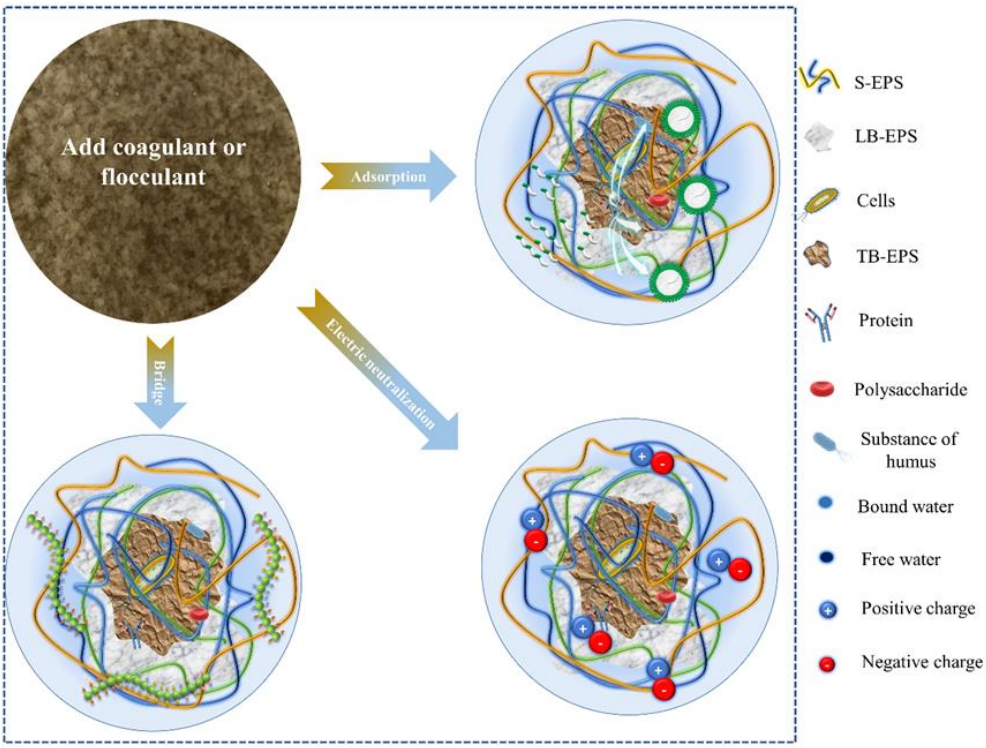

Figure 5. Mechanism of coagulation/flocculation conditioning of sludge.

4.1.1. Inorganic Flocculants

The most commonly used inorganic flocculants are aluminum and iron salts; they can be divided into low molecular weight inorganic coagulants, such as $AlCl_3$, $Al_2(SO_4)_3$, $FeCl_3$, and $Fe_2(SO_4)_3$, and inorganic polymeric coagulants, such as polymeric aluminum chloride

(PAC) and polymeric ferrous sulfate (PFS). Inorganic polymeric coagulants generally have a higher charge density and higher molecular weight than lower molecular weight inorganic flocculants. In addition to the classic aluminum and iron salts, Na_2SiO_3, which is commonly used as a coagulation aid, is used for deep conditioning of sludge because of its good bridging effect but is not suitable for use on its own. Zhang et al. [23] evaluated the effect of chemical conditioning of various titanium-based coagulants (TSCs) in combination with magnetic nano-Fe_2O_3 on dewatering performance. The results showed that different TSC treatments all improved the dewatering performance of activated sludge, with TSCs with an alkalinity of 0.5 (TSC0.5) treatment being more effective. Sludge flocs formed by TSC0.5 treatment had larger floc size and higher floc strength than those formed by other TSCs treatments. The compressibility of the sludge floc decreased from 1.24 to 0.91. SRF decreased to 5×10^{12} m/kg; and the water content of the sludge cake decreased to 77.4%.

4.1.2. Organic Polymer Flocculants

Organic polymer flocculants are now widely used in municipal sewage treatment plants for sludge conditioning; the most commonly used of these are polyacrylamide type flocculants. Polyacrylamide flocculants are divided into anionic (APAM), cationic (CPAM), amphoteric (AmPAM), and nonionic (NPAM). Compared to inorganic flocculants, organic flocculants are more adaptable to a wider pH range and can form larger flocs and create drainage pore channels. Natural polymeric flocculants and their modified polymeric flocculants, represented by starch, chitosan, and cellulose, have also become research hot spots in recent years due to their low cost and no secondary pollution. Feng et al. [24] prepared hydrophobic conjugated cationic polyacrylamide (HACPAM) by micellar polymerization under UV irradiation using V-50 (azodiisobutylamine hydrochloride) as the initiator and acrylamide, acryloyloxyethyl trimethyl ammonium chloride, and butyl methacrylate as substrates. The sludge was conditioned with HACPAM to improve sludge dewatering performance. The results showed that sludge dewatering performance improved significantly with increasing HACPAM dosage, with the best dosage being 3.532 kg/t dry matter of HACPAM. The water content of the sludge cake was reduced to 68.8%

4.1.3. Bioflocculants

Bioflocculants are of interest because of their high degradability, non-toxicity, and high efficiency compared to traditional coagulants/flocculants. There are two major types of bioflocculants; the first common bioflocculant, which is similar to traditional conditioning agents, and the second complex bioflocculant, which requires a longer inoculation time and requires isoenergetic substances to enhance flocculant production. Liu et al. [25] studied the optimized production process of a new Klebsiellabi flocculant, M-C11, and its application in sludge dewatering. The results showed that the optimal bioflocculation activity of M-C11 2.56 mL and $CaCl_2$ 0.37 g/L was 92.37%. SRF decreased from 11.6×10^{12} m/kg to 4.7×10^{12} m/kg, indicating that bioflocculant treatment significantly improved sludge dewatering performance (Table 3).

Table 3. Efficiency of different types of flocculants/coagulants for sludge conditioning.

Types of Flocculants/Coagulants	Flocculation/Coagulation Methods	Conditioning Conditions	Conditioning Efficiency	References
Inorganic coagulants	Titanium-based coagulants (TSCs)	Dosing rate: 0.009 g/g TSS	SRF down to 5×10^{12} m/kg Mud cake moisture content reduced to 77.4%	[23]
	PFS	PFS dosing rate: 300 mg/gDS	SRF down to 9×10^{11} m/kg Mud cake moisture content reduced to 82%	[26]
	Poly aluminum chloride (PACl)	PACl dosing rate: 3.8 gAl/L	SRF down to 2×10^{8} m/kg	[27]

Table 3. *Cont.*

Types of Flocculants/Coagulants	Flocculation/Coagulation Methods	Conditioning Conditions	Conditioning Efficiency	References
Organic flocculants	Organic polymer flocculant hydrophobically-conjugated cationic polyacrylamide (HACPAM)	HACPAM dosing rate: 3.532 kg/t	Mud cake moisture content 68.8%	[24]
	CPAM modified diatomaceous earth	pH 3.5 CPAM modified diatomaceous earth dosage: 0.4% wt	SRF reduced to 0.92×10^{12} m/kg, remaining cake moisture content reduced to 68.1%	[28]
	PAM	PAM dosage: 3.0 mg/g	SRF down to $(0.4 \pm 0.1) \times 10^{12}$ m/kg CST drops to 19.7 ± 0.8 s.	[29]
Complex Conditioning	PAM-FeCl3	PAM-FeCl3 Optimal dosage: 20 mg·L−1	SRF down to 5.4×10^{10} $m \cdot kg^{-1}$	[30]
	Starch-3-chloro-2-hydroxypropyltrimethylammonium chloride (St-WH)	St-WH optimum dosing rate: 10 mg/gTSS	SRF down to 2.5×10^{11} $m \cdot kg^{-1}$	[31]
Bioflocculants	A new type of Klebsiella bioflocculant M-C11	M-C11 dosing volume: 2.56 mL	SRF down to 4.7×10^{12} m/kg	[25]
	Complex iron bioflocculant produced by acidified ferrous oxide thiobacillus	Processing time: 1 h	92% and 91% reduction in CST and SRF, respectively	[32]

4.2. Oxidative Conditioning

Oxidative conditioning refers to improving the dewatering performance of sludge by adding a proportion of oxidizer to the sludge to break down the cell walls of the EPS and sludge cells, break down large organic matter molecules, and release the bound water in the sludge. The oxidation methods generally include Fenton reagent oxidation, ozone oxidation, Fe^{2+} activated persulfate, potassium permanganate oxidation, hypochlorite oxidation, etc.

4.2.1. Fenton Conditioning

The Fenton reaction can generate strong oxidizing and non-selective –OH (its redox potential is 2.8 V), which can effectively promote the decomposition of mud flake EPS and release the water bound in it; eliminating the water that cannot be removed mechanically, which is converted into clear water; and to realize a deep dewatering of the sludge process. As shown in Figure 6, the mechanism of sludge conditioning by Fenton's reagents is generally divided into two steps. First, the strong oxidizing radicals such as –OH generated during the Fenton reaction destroy the sludge floc and release the internally bound water. Then, Fe^{3+} generated during the Fenton reaction agglomerates the fine flakes of sludge destroyed by –OH into large particles, further releasing the bound water content in the sludge flakes, and finally achieving a significant improvement in sludge dewatering performance. Chen et al. [33] used fast and efficient treatment of activated sludge by electro-Fenton oxidation. The electrolyte was used with a final concentration of 50 mmol/L Na_2SO_4. In addition, Fe^{2+} was concentrated to 50 mmol/L and bubbled with an air pump at a rate of 3 L/min to continuously generate H_2O_2. The results showed that SRF decreased to $5.28 \pm 0.01 \times 10^{11}$ m/kg (30.17% decrease). The zeta potential converges to 0.

4.2.2. Ozone Conditioning

Ozone as an oxidant for sludge conditioning can be divided into two cases, an ozone oxidation conditioning technology alone and a second catalyst-catalyzed ozone oxidation conditioning technology. Ozone is a strong oxidizing agent with a redox potential of 2.07 V. The oxygen atoms in the ozone molecule are strongly electrophilic or protonophilic, and the new ecological oxygen atoms produced by decomposition have high oxidation activity. The direct action of the ozone molecule on the pollutants is selective, and most of the oxidation process is very slow and not complete enough. This is also influenced by ozone's low solubility and easy decomposition and stability. A number of studies have shown that ozone alone can increase the amount of protein and polysaccharides in sludge EPS and has some effect on COD removal, but the effect of ozone on sludge conditioning is very high due to its selective and less stable oxidation limits for pollutants and insufficient and complete oxidation. Therefore, in general, the ozone oxidation technology catalyzed by the second catalyst is widely used in sludge conditioning. As shown in Figure 7, the ozone molecule can be indirectly oxidized under the catalytic action of the catalyst. Under the influence of the catalyst, the ozone undergoes a decomposition reaction to generate strongly oxidizing hydroxyl radicals (–OH). The catalytic ozone is non-selective in its action on the pollutant, with a faster reaction speed and faster decomposition of the pollutant. Among the many catalysts for catalyzing ozone, iron-based catalysts are an excellent catalyst with the advantages of easy production, plentiful nature, and high catalytic efficiency. Ge et al. [21] developed the PFS/O_3 process as a novel environmentally friendly method to decompose sludge populations and EPS to improve dewatering performance. The optimal dosing rates for PFS and O_3 were 40 mg/gTS and 60 mg/gTS, respectively. The moisture content of the dewatered sludge cake was at least 59.79%; the bound water content was limited to 2.32 g/gTS. PFS/O_3 treatment increased the mud zeta potential to −4.23 mV.

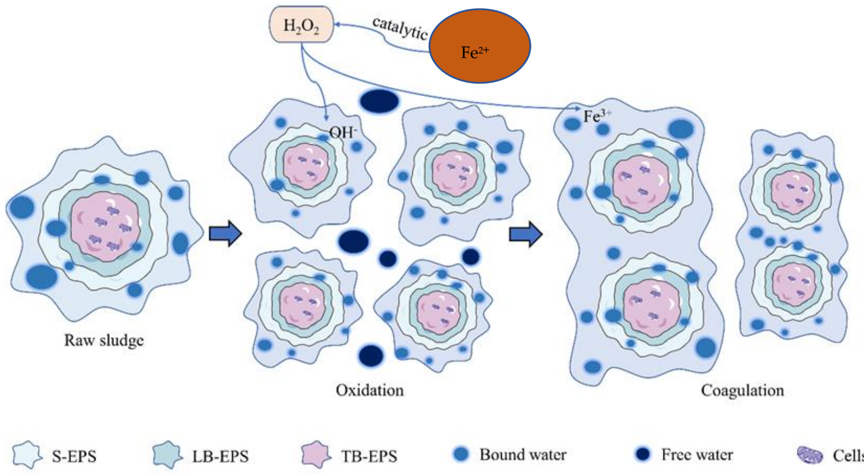

Figure 6. Two-step conditioning mechanism of oxidation and coagulation in Fenton reagent-conditioned sludge.

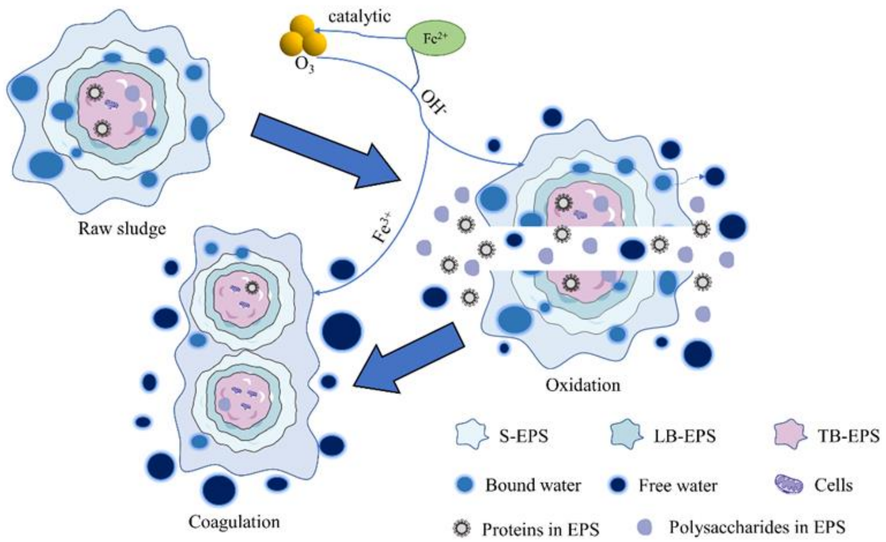

Figure 7. O_3 Two-step conditioning mechanism for oxidation and coagulation in conditioned sludge.

4.2.3. Activated Persulfate Conditioning

The advanced oxidation technology of activated persulfate uses SO^{4-} as the main reactive group and is divided into two types, peroxomonosulfates and peroxodisulfates, which are generally referred to as peroxodisulfates. As shown in Figure 8, activated persulfates are activated by light, heat, and transition metal ions, and the persulfate ion $S_2O_8{}^{2-}$ is activated and decomposed into SO^{4-}, destroying the specific functional groups of the fluorescent material, resulting in polymer chain breakage and damage the sludge cells, leading to the release of bound water, intracellular material, and intracellular water from the EPS, enhancing the dewatering performance of the sludge. The most commonly used activation method is Fe^{2+} activation, with a standard redox potential of 3.1 V for SO^{4-}, which exceeds that of the strongly oxidizing –OH and is more stable to oxidizing pollutants, making it more suitable for a wider range of applications. Guo et al. [34] uses Fe^{2+} activated potassium persulfate (PPS) for sludge treatment. The results showed that the Fe^{2+}-activated PPS improved the dewatering performance of the sludge with a lower PPS dosage and a shorter treatment time. When PPS and Fe^{2+} were dosed at 0.5 mmol/g and 1.0 mmol/g, respectively, the CST decreased sharply within 15 min and finally fell to only 25.1 s; SRF decreased to 1.75×10^8 m/kg.

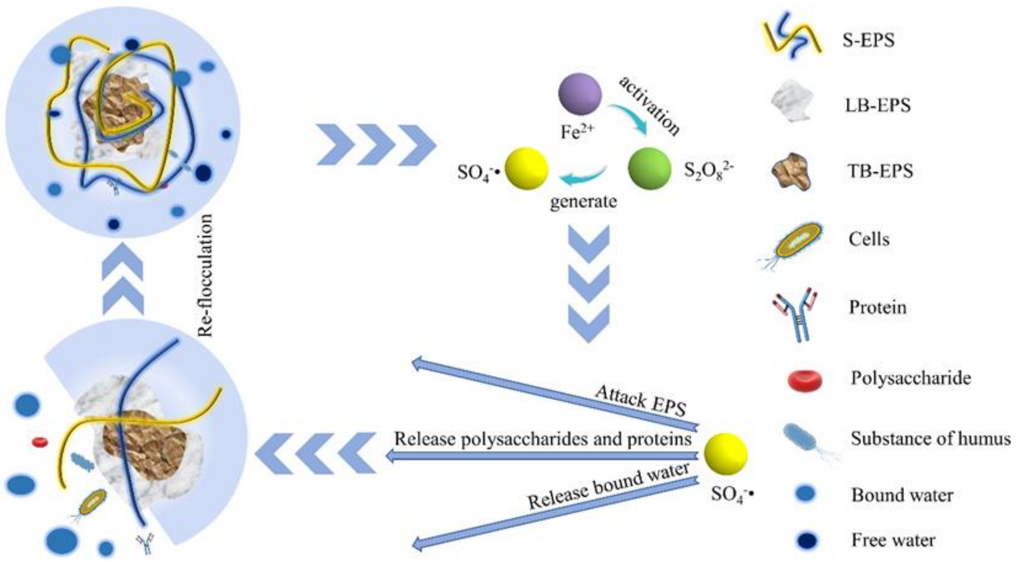

Figure 8. Fe^{2+} Activated persulfate sludge conditioning mechanism.

4.2.4. Hypochlorite Conditioning

Hypochlorite is a powerful oxidizer that has been used in sludge treatment in recent years. Typically, hypochlorite is used in combination with coagulants, where hypochlorite (ClO^-) reacts with water to produce hypochlorous acid (HClO), which destroys the organic matter attached to the outer layer of colloidal particles and thereby increases the electronegativity of the particles themselves and the effectiveness of coagulation. As shown in Figure 9, similar to other oxidants, hypochlorite decomposes ClO_2, destroys EPS, and releases organic matter such as bound water, polysaccharides, and proteins from EPS to achieve deep dewatering of sludge. Colloidal particles are better adsorbed by aluminum and calcium ions, which promotes the precipitation of formed humates, etc. The additionally added calcium ions can further improve the coagulation effect. Chen et al. [22] used hydrothermal treatment in combination with $FeSO_4$-$7H_2O/Ca(ClO)_2$ to treat sludge. The results showed that the optimal $Ca(ClO)_2$ dosage was 0.04 g/g TS and the optimal molar ratio of $Ca(ClO)_2$ to $FeSO_4$-$7H_2O$ was 1.25. The bound water in the sludge was converted into free water, indicating that the EPS in the sludge was degraded by ClO^- and part of the water absorbed in the EPS was released. This leads to deep drainage of the sludge. Table 4 summarizes the conditioning efficiencies of the different types of oxidatively conditioned sludge.

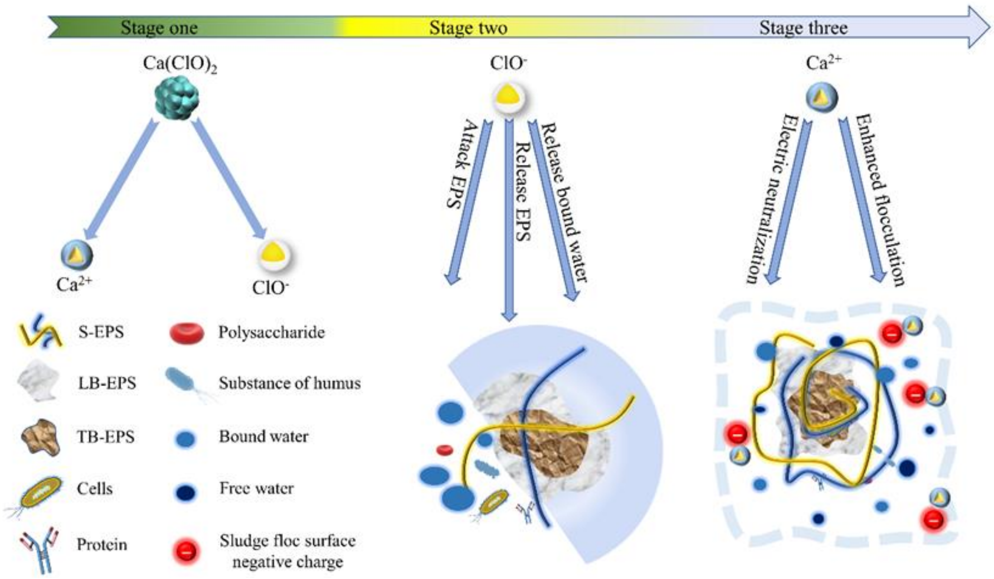

Figure 9. $Ca(ClO)_2$ sludge conditioning mechanism.

Table 4. Efficiency of different types of oxidation techniques for conditioning sludge.

Oxidation Methods	Conditioning Processes	Conditioning Conditions	Conditioning Efficiency	References
Fenton	Fenton—Electrodialysis	Fe^{2+} at a concentration of 50 mmol/L was bubbled with an air pump at a rate of 3 L/min to continuously produce H_2O_2	SRF reduced to $5.28 \pm 0.01 \times 10^{11}$ m/kg (30.17% reduction)	[33]
	Fenton-Cetyltrimethylammonium Bromide (CTAB)	The water content response doses for Fe^{2+}, $H_2 O_2$ and CTAB were 89, 276, and 233 mg/gDS, respectively	Mud cake moisture content reduced to 66.8%	[35]
Ozone	PFS-O_3	The optimum dosing rates for PFS and O_3 are 40 mg/gTS and 60 mg/gTS, respectively	The moisture content of the de-cemented cake is limited to a minimum of 59.79% bound water content of 2.32 g/gTS	[21]
	Peroxymonosulfate—Ozone	pH 11.0 PMS/O_3 = 0.06; O_3 = 12.5 mmol/L	70% reduction in sludge filtration time (TTF) relative to virgin sludge	[36]
Activated persulfates	PPS-Fe^{2+}	The optimum dosing rates for PPS and Fe^{2+} are 0.5 mmol/g and 1.0 mmol/g, respectively	CS down to 25.1 s SRF down to 1.75×10^8 m/kg	[34]
	Fe^{2+}—perthiolate—tannic acid (TA)	Fe^{2+} (0.3 mmol/gTS (total solid phase))/Persulfate (0.6 mmol/gTS) process with an effective TA/Fe^{2+} (molar ratio) of 0.25	The capillary adsorption time, filtration ratio, and water content of the dewatered sludge cake were reduced by 61.5%, 35.3%, and 6.4%, respectively, compared to the Fe^{2+}/persulfate treatment	[37]
Hypochlorite	$Ca(ClO)_2$-$FeSO_4$-$7H_2$ O	The optimum dosage of $Ca(ClO)_2$ is 0.04 g/g DS and the optimum molar ratio of $Ca(ClO)_2$ to $FeSO_4$-$7H_2O$ is 1.25	The conversion of bound water to free water in the sludge indicates that the EPS in the sludge is degraded by ClO^- and some of the water absorbed in the EPS is released	[22]
	NaClO-cationic starch-based flocculant (St-WH)	Optimal dosing of NaClO: 11 mg/gTSS	SRF down to 1.2×10^{11}	[38]

4.3. Acid–Base Conditioning

Acid conditioning protonates negatively charged functional groups (carboxyl and amino groups, etc.) in the sludge floc, increasing the sludge's zeta potential from negative to nearly zero, reducing electrostatic repulsion, and improving flocculation, aggregation, and dewatering of the sludge. Acidification increases the dewatering capacity of the sludge through the combined action of sludge floc fragmentation and protonation. The former degrades EPS, promotes conversion of some of the inner EPS into the outer layer, and induces the release of bound water. The latter effectively compacts the released EPS and improves drainage performance. Wei et al. [39] adjusted the pH of the sludge between 7.0 and 3.0 to study the effect of acidification on the dewatering performance of sewage sludge. The results showed that the SRF and sludge cake water content of the sludge decreased to a minimum of 2.5×10^{12} m/kg and 82% at pH 3.0. Combined conditioning by acid digestion and various other physical/chemical conditioning methods is also a new fashionable topic. Fan et al. [40] studied the conditioning of sludge by acidification combined with multi-stage elution (AME). The results showed that the AME pre-treatment reduced the water content of the sludge cake to 67.3%, increased the sludge dewatering rate by 84.5%, and increased the organic sludge content by 10.3%. Zhang et al. [41] investigated the respective roles of acidification and oxidation in the Fenton process in improving the dewatering performance of anaerobically digested sludge. The results showed that acidification and Fenton oxidation had a synergistic effect, with CST decreasing from 200 s to 100 s when pH was lowered from 7.0 to 3.0.

The alkaline pre-treatment is used to destroy the cell structure of the sludge and to dissolve the EPS in the sludge through solubilization of membrane proteins and saponification of membrane lipids, thereby converting the TB-EPS in the sludge floc into a liquid in the form of organic matter. Alkaline hydrolysis in combination with oxidation has been a hot topic in sludge pretreatment. Liu et al. [42] prepared alkaline ferrate containing Fe(VI) and KOH (NPAF) that effectively destroyed sludge structure while improving dewatering performance. The results showed that when NPAF was dosed at 500 mg/L, sludge settling capacity and dewatering capacity increased by 55.1% and 7%, respectively. The addition of alkali increases the oxidation stability. In addition, the formed hydroxide flocculates microbial debris, organic matter, and inorganic particles; flocculates the sludge; and reduces sludge volume and the cost of sludge treatment and disposal. Budych-Gorzna et al. [43] investigated low-temperature alkaline pretreatment (WAS) as a method to improve the economics of water treatment plants. Results showed that methane production increased from 210.4×10.9 mL CH_4/gVS to 248.8×13.1 mL CH_4/gVS at 16 g NaOH/kg TS WAS, 60 °C, 60 min, methane production was increased by 27% and the dewatered sludge volume was reduced by 8.7%. Table 5 summarizes the sludge conditioning efficiency for different types of acid–base conditioning.

Table 5. Efficiency of different types of acid and alkaline conditioned sludge.

Conditioning Methods	Conditioning Conditions	Conditioning Efficiency	References
Acidification	pH 3.0	Sludge SRF and sludge cake moisture content reduced to a minimum of 2.5×10^{12} m/kg and 82%	[39]
Acidification combined with multistage elution (AME)	pH 2.2	Water content of sludge cake reduced to 67.3%, sludge dewatering rate increased by 84.5%, and organic content of sludge increased by 10.3%	[40]
Acidification—Fenton	pH 3.0	CST drops from 200 s to and 100 s	[41]
Acidification with sulphuric acid	pH 1.5	Mud cake moisture content reduced to 66.7%	[44]
Alkaline permethrate (NPAF)	NPAF dosed at 500 mg/L	Sludge settling capacity and dewatering capacity increased by 55.1% and 7%, respectively	[42]
Low temperature alkaline pre-treatment (WAS)	16 g NaOH/ kgTS WAS, 60 °C, 60 min	Dewatered sludge volume can be reduced by 8.7%	[43]
Alkaline thermal hydrolysis	160 °C/60 min/pH 10.0	Mud cake moisture content reduced to 40%	[45]
Alkaline thermal hydrolysis—mechanical dehydration (MDS)	Lime dosing rate: 20% DS	SRF down to 1.46×10^{10}	[46]

4.4. Aggregate Conditioning

Aggregates can reduce compressibility and improve the permeability of the mud cake. Commonly used aggregates include fly ash, lime, coal dust, red mud, chitosan, etc. Aggregate materials easily form a porous structure inside, and because of their small particle size and strong surface adsorption capacity, they can also separate water adsorbed on sludge mixed with sludge, resulting in charge neutralization and surface tension that is larger than the sludge adsorption power on adsorbed water. In addition, after mixing with sludge, the skeletal material can adsorb sludge flocs, forming a skeletal construct centered on its particles, thereby reducing the compressibility of the sludge, building the water filtration channel of the sludge cake, and greatly reducing the viscosity of the sludge, greatly improving the dewatering performance of the sludge. Wang et al. [47] compared the effects of rice husk powder (RHF), rice husk biochar (RHB), and rice husk mud cake biochar (RH—SCB) as physical conditioning agents on sludge dewatering performance. The results showed that the optimal dosage of RH—SCB was 90 g/g TS, SRF 7×10^{13} m/kg, and the moisture content of the sludge cake was 72%. Significant advances have been made in the conditioning of skeletal materials in combination with oxidation/flocculation processes or physical pretreatments. In Lee et al. [48], the municipal sewage sludge was conditioned by adding fly ash in combination with electrodialysis. The results showed that the optimal fly ash addition with a particle size of 25–75 μm was 20% by weight, which improved the dewatering efficiency by about 40% over the conventional mechanical dewatering method without the addition of fly ash. Tian et al. [49] used an aluminum ammonium booster and wood shavings to condition the mud. The results showed that the best conditioning effect was obtained when aluminum ammonium improver and wood chips were added at 2% and 20% of the sludge dry weight, respectively. Under these conditions, the specific filtration resistance and capillary wicking time of the sludge were reduced by 89.9% and 73.1%, respectively, compared to the original sludge. Table 6 summarizes the sludge conditioning efficiency for different types of aggregate conditioning. Fan et al. [50] prepared a new chitosan graft copolymer (C-Chitosan-g-PAM-AA) as a sludge conditioning agent. The results showed that the minimum water content of the filter cake was 61.41% at the optimal dosing rate of 37.5–45.0 mg·L^{-1}. Ding et al. [51] used potassium permalate and walnut shell to condition the mud. The results showed that the optimal dosages of potassium permalate and walnut shell were 60 mg/g TS and 0.8 g/g TS, respectively, resulting in a sludge cake moisture content of 70.2% and an SRF reduction to 1.107×10^{11}.

Table 6. Efficiency of sludge conditioning with different aggregate.

Skeleton Conditioner	Conditioning Conditions	Conditioning Efficiency	References
Rice husk sludge cake biochar (RH—SCB)	The optimum dosage of RH—SCB is 90 g/gDS	SRF of 7×10^{11} m/kg and mud cake moisture content of 72%	[47]
Fly ash—electrodialysis	The optimum amount of fly ash with a particle size of 25–75 um is 20% wt	Approximately 40% increase in dewatering efficiency	[48]
Ammonium aluminum enhancer—wood chips	Ammonium aluminum enhancer and wood chips added at 2% and 20% of the dry weight of the sludge, respectively	The specific filtration resistance and capillary suction time of the sludge were reduced by 89.9% and 73.1%, respectively, compared to the original sludge	[49]
Novel chitosan graft copolymer (C-chitosan-g-PAM-AA)	Optimal dosing level 37.5–45.0 mg L^{-1}	Mud cake moisture content reduced to 61.41%	[50]
Potassium permethrate- walnut shell	The optimum dosage of potassium permethrate and walnut hulls is 60 mg/gDS and 0.8 g/gDS, respectively	The sludge cake moisture content was 70.2% and the SRF was reduced to 1.107×10^{11}	[51]
Thermal hydrolysis—lignocellulose	Lignocellulose dosing: 0.2–0.5 g/g DS. 5 min assisted hydrolysis at 180 °C for medium temperature	Water content of sludge cake reduced by around 30%	[52]
Polyester textile fibers—FeCl3	The optimum dosage for polyester textile fibers is 20% DS	SRF down to 1.62×10^{12} m/kg	[53]

5. Conclusions

This paper provides an overview of the factors affecting sludge dewatering and sludge conditioning processes in relation to the composition and properties of municipal sludge. Factors affecting sludge dewatering performance include sludge zeta potential, sludge particle size, rheological properties, floc properties, and EPS properties. EPS is an important factor affecting sludge dewatering performance and is also an important factor limiting further reduction of sludge moisture content. Current methods of sludge conditioning are divided into physical, chemical, and biological conditioning, and this paper examines in detail the conditioning parameters and mechanisms of various chemical conditioning methods. Chemical conditioning includes coagulation and flocculation, oxidation, acid-base, and aggregate conditioning. Chemical conditioning is now widely used in municipal sewage treatment plants. Combined processes such as oxidation + coagulation and flocculation and acid–base + coagulation and flocculation are widely used for sludge conditioning in municipal sewage treatment plants at home and abroad. The mechanism of different chemical conditioners in sludge conditioning and dewatering process was clarified and the mechanism of deep dewatering of sludge was revealed. Through the comparison of the dewatering effect and cost analysis of these chemical techniques, the sludge conditioning method with controllable cost, high treatment efficiency, and low harm to the environment is found, which provides a certain theoretical basis and reference value for the deep dewatering of sludge and the selection of sludge disposal process. Sludge dewatering process research should focus on the development of more appropriate sludge conditioning and dewatering combination processes based on the sludge quality of different municipal sewage treatment plants in combination with their local environment, input costs, subsequent sludge disposal methods, and other factors. At the same time, the sludge dewatering process is to be further optimized through the development of new efficient and environmentally friendly sludge conditioning agents in order to achieve high efficiency and low energy consumption.

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