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Review

Could Plant-Based Flocculants Substitute the Conventional Synthetic Chemicals in the Sludge Dewatering Process?

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Abstract: Due its high water content, sewage sludge dewatering is not just a simple operation; rather, it is a challenging process and a costly management task. Its final handling is usually preceded by several dewatering steps, and among them is the conditioning process known as the flocculation stage, which is carried out using synthetic chemical reagents. Despite the abilities of these additives to reduce sludge volume and extract its bound waters, they are suspected to cause serious environmental and health threats. Their substitution by natural and efficient additives originating from plant extracts could thus be a safe and an eco-friendly alternative, overcoming ecosystem damages. It is within this context that the present review paper critically investigates the efficacy and feasibility of plant-based flocculants, aiming to enhance sludge dewatering and dispense with environmental burdens. To do so, the types of the conventional chemical flocculants, their drawbacks, and their impacts on the ecosystem and human health were addressed. In parallel, the potential dewatering efficiency of plant extracts toward sludge treatment was compiled, and their mechanistic dewatering paths performances were thoroughly discussed. The challenges associated with dewatered sludge and its potential exploitation were also highlighted to motivate scientific communities to further explore green resources for sludge processing. It is suggested that green resources such as Moringa, Cactus, Aloe, and Okra could be used as green flocculants instead of chemical ones, which would provide a promising and eco-sustainable approach to sludge dewatering and might represent a path towards an environmentally friendly and clean technology.

Keywords: sludge; dewatering; conditioning; chemical flocculants; plant-based flocculants

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1. Introduction

With the aim of protecting ecosystems and ensuring the continuity of human life, great efforts have been devoted to wastewater remediation and its related technologies. Yet, despite the well-established paths therein and the effectiveness of conventionally applied wastewater treatment processes, it is believed that their associated sludge is one of the detrimental causes behind environmental and human health degradation, primarily if they are disposed of without efficient treatment means [1]. Furthermore, in addition to its unpleasant odor and high quantity of suspended solids (SS) content, the presence of large amounts of water therein renders its management more challenging, and its transport to the final drying beds is very costly. Its dewatering is hence a crucial step towards ensuring successful solid–liquid separation, reducing its volume, and alleviating its handling charge. To do so, the sludge is usually conditioned in advance to promote its associated water removal and extend the life of the mechanical devices used for dewatering (plate and membrane filter presses), which are precious and energy-consuming [2].

This latter task is usually performed via the supply of synthetic flocculants, also called synthetic conditioners, such as aluminum sulphate, poly-aluminum chloride, ferrous chloride, and polyacrylamide [3,4]. The key role of these chemicals is to agglomerate the suspended matter into large and settleable flocs and free them from the water to which they are bound. Consequently, the sludge volume will be reduced upon the release of the trapped water into solid particles, thus leading to an increase in its settling velocity and compactness [5]. However, in spite of their efficiency in facilitating solid–liquid separation, the effective application of these chemicals is doubtful, as it is reported to cause serious environmental burdens; thus, it is a growing human health concern [6].

In fact, due to their nonbiodegradability, metal salts and organic polymer residues may accumulate in the treated sludge and thus hinder its usage. Further, if valorized in agriculture, those residues may leach out into soil and groundwater, endangering fauna, flora, and human health [7]. Likewise, it is accepted that severe diseases such as Alzheimer's disease and neurotoxic and carcinogenic illnesses have been ascribed to the extensive application of these chemicals in wastewater treatment and sludge dewatering [8,9]. Additionally, the high cost and non-availability of such chemicals, especially in developing countries, could represent the main hurdles to their supply. Taking these drawbacks and limitations into consideration, in sludge conditioning, replacing chemical flocculants with natural ones derived from plant extracts is both worth considering and deemed to be a safe and environmentally friendly option. In this vein, although some prior studies have highlighted the uses of some plant-based flocculants (such as Moringa [10], Cactus [11], and Okra [12]) as surrogates for their chemical equivalents, and although they have been evaluated for their sludge-dewatering performance, the application of green conditioners in this field, compared to the conventional synthetic flocculants that are extensively exploited in wastewater treatment plants, is still in its infancy, [13–16]. In order to persuade environmentalists and stakeholders to invest in the replacement of chemical flocculants with safe and green ones, additional works exploring natural flocculants in the dewatering process are required.

Thus, the present review paper sheds light on the feasibility of using plant-based flocculants for sludge dewatering and offers a comparison of their performance with those of their chemical equivalents. Although previous reviews and studies mention other bioflocculants derived from animals, plants, or microorganisms, the majority of these works focused on sludge composition, conditioning, and dewatering methods [17–19]. It is also worth noting that flocculants originating from animals or microorganisms are difficult to acquire and require sophisticated technology and substantial precautions. Furthermore, most of the recent review articles presented a holistic summary of the coagulation/flocculation process in sludge dewatering [20], i.e., they reviewed the commonly used chemical and natural flocculants without comparing their dewatering capabilities. Few reviews have discussed and assessed the potential use of plant-based flocculants instead of chemical ones for sludge treatment [21]. As a result, and as has already been indicated, this paper offers a thorough analysis of the viability of replacing the notorious chemical flocculants with those derived from plants. An overview of the sludge's physicochemical properties, its conditioning techniques, and, in particular, the flocculation process is presented. In the present review, the use of chemical flocculants as conditioners and their impact on the environment and human health are highlighted with the aim of drawing the attention of environmentalists and wastewater stakeholders to the necessity of considering green surrogates. Thus, also in this paper, the performances of the plant-based flocculants that are most widely used in sludge dewatering are compared to those of their chemical equivalents. Challenges to and the future the prospects of their application are addressed in the hopes that it will help to establish an effective, green, and environmentally friendly dewatering process.

2. Sludge Properties and the Necessity for Dewatering

Sludge is a suspension of thick semi-solid deposits (slurry, organic matters, trace metals and nutrients, etc.) and microorganisms, which form more than 96% of the sludge's weight and volume [22]. This high water content can be classified into free and bound water. Free water is easily separated from the sludge by simple filtration or sedimentation. However, bound water, which consists of vicinal, interstitial, and internal water, is difficult to separate. According to its position in the sludge matrix, the vicinal water is held on the solids' surface, the interstitial one is captured in the interstices, and the internal water is chemically bound to the solids [23]. This kind of water is deemed to be the main barrier for dewatering because it is tightly attached to the sludge's constituents (microorganisms and organic and inorganic matter) and binds them together. As a result, sludge porosity decreases, thus preventing water withdrawal.

Otherwise, microorganisms and organic polymers, namely the extracellular polymeric substances (EPS), are considered to be the prime factors to maintain the sludge's moisture due to their ability to trap water. Thus, based on the water distribution within, the EPS can be classified into three types, namely the soluble (S-EPS) and the loosely bound (LB-EPS) and tightly bound extracellular polymeric substances (TB-EPS) [24]. Despite its high bound water content, the EPS forms stable and viscous gel networks due to the prevalence of proteins and polysaccharides in their composition [25]. The ionization of these biopolymers imparts a negative surface charge to the sludge particles and leads to the aggregation of organic matter and microbial cells [26]. Consequently, important water volume is retained, and flexible flocs with low settleability are produced, resulting in dewatering difficulties. Considering the complex structure of sludge, dewatering is necessary to promote the further treatment, transportation, and disposal of sludge and to elucidate the causes of its exploitation.

Generally, sludge dewatering is carried out by conventional mechanical means using different pieces of equipment and methods, such as centrifuging, belt filter presses, plate and frame filters, etc. [27]. Although, the efficacy of these methods in eliminating free water, as well as a small portion of bound water and the dry solids produced after dewatering, does not exceed 40% [28]. Therefore, the sludge is typically subjected to a pretreatment stage by different conditioning technologies involving biological (enzyme treatment), physical (ultrasonic, thermal, and microwave treatments, filter aid applications), and chemical (acid or alkali, oxidation, and flocculation treatment) methods [29–31]. To elucidate their efficiency in aiding sludge dewatering, the principles behind each conditioning method, as well as their limitations, are briefly described in Table 1.

Table 1. Conditioning methods for improving sludge dewatering.

Conditioning Methods	Principle	Limitations
Biological methods	Lysis of EPS and microbial cells to release the bound water in the sludge through the addition of enzyme or microbial leachate [32].	- Long processing time. - Difficult microorganism cultivation.
Physical methods	Modification of the structural properties of the sludge to increase its filterability and settle ability via the addition of solid and porous material (skeleton builder) or through thermal treatment (freeze-thawing, thermal, and microwave treatment) [33].	- Energy-consuming. - Voluminous sludge cake.
Chemical methods	Modification of the physicochemical properties of the sludge via the addition of chemical reagents conducive to either solids degradation to ease bound water release as a result of pH alteration (alkali or acid treatment) or by the consolidation of solids to increase its permeability and promote solid/liquid separation (coagulation/flocculation) [34,35].	- Careful pH monitoring. - Questionable safety of the used chemicals.

As presented in Table 1, it is worth mentioning that long processing times and high energy requirements impede the real application of biological and physical conditioning methods on an industrial scale. So, for economic reasons, among all of the chemical conditioning approaches, coagulation/flocculation is the most applied one for sludge dewatering due to its rapidity, cost-effectiveness, and efficacy in separating water from solid particles [36]. Coagulation/flocculation is based on the addition of chemicals to aggregate the (SS) into large and compacted flocs, enabling the removal of the bound water of sludge and improving its settleability. Likewise, regarding wastewater treatment, the efficiency of the coagulation/flocculation process to assist in sludge dewatering depends on the nature and physicochemical properties of the used chemical coagulants/flocculants, which are generally aluminum salts, iron salts, and acrylamide-based synthetic polymers [37,38].

3. Chemical Flocculants for Improving Sludge Dewatering: Efficiency and Effects on the Environment and Human Health

The conventional synthetic flocculants, such as aluminum sulphate ($Al_2(SO_4)_3$, aluminum chloride ($AlCl_3$), polyaluminium chloride (PACl), ferric sulphate ($Fe_2(SO_4)_3$, and ferric chloride ($FeCl_3$), as well as cationic and anionic polyacrylamides (PAM), have been applied in sludge dewatering for a long time [14,39,40]. The efficacy of these chemicals has been assessed in several research works. For instance, in a study conducted by Masihi and Gholikandi [41], the dewatering performances of aluminum ($Al_2(SO_4)_3$ and $AlCl_3$) and iron ($Fe_2(SO_4)_3$ and $FeCl_3$) salts were evaluated with respect to their specific resistance to filtration (SRF), capillary suction time (CST), time to filter (TTF), and moisture content (MC) in treating an anaerobically digested sludge. As listed in Table 2, significant SRF reductions of 95% and 93% were found using 150 mg/g of $AlCl_3$ and $FeCl_3$, respectively, while the higher dosages of 300 mg/g for $Al_2(SO_4)_3$ and $Fe_2(SO_4)_3$ lessened the SRF by more than 80%. Even the addition of these metal salts to the sludge also minimized the CST, TTF, and MC, thus improving its dewatering. Moreover, a notable SRF decrease from 11.30×10^{12} m/kg to 3.90×10^{12} m/kg was achieved using PACl as a flocculant to treat a biological sludge [42]. Interestingly, comparing the dewatering efficacy of PACl with that of $FeCl_3$, Niu et al. [43] revealed that the best decrease in SRF (from 1.72×10^{12} m/kg to 0.07×10^{12} m/kg) was gained using $FeCl_3$. In this study, the investigators demonstrated that inorganic salts enhance sludge dewatering by neutralizing the charge of particles or through compression in the double layers of the EPS, which is carried out by trivalent cations (Al^{3+} or Fe^{3+}) [43]. As a result, the aggregation of sludge colloids and the seepage of bound water renders the sludge amenable to dewatering.

Otherwise, synthetic polymers, namely PAMs, have been commonly used in sludge conditioning to enhance its dewatering [44,45]. On account of their long chain and high charge density, polyacrylamide-based flocculants bridge the small solids together to form large and settleable flocs. Consequently, the separation of solids from water content in the sludge may occur, enabling efficient dewatering. In fact, as displayed in Table 2, the application of PAM led to important decreases in the dewatering properties (SRF, CST, and MC) of sewage sludge [46]. Likewise, in treating a waste-activated sludge, Wu et al. [47] found that prominent reductions in SRF (from 14.20×10^{12} m/kg to 0.40×10^{12} m/kg) and CST (from 225 s to 19.70 s) were achieved by a cationic PAM.

Table 2. Efficiency of chemical flocculants for sludge dewatering.

Raw Sludge	Chemical Flocculant	Dewatered Sludge Parameters					References
		SRF × 10^{12} (m/kg)	CST (s)	TTF (s)	DS (%)	MC (%)	
Anaerobically digested sludge SRF = 265 × 10^{12} m/kg CST = 283 s TTF = 600 s MC = 97.80%	$Al_2(SO_4)_3$ Optimal dose = 300 mg/g	51.94	57	140	–	89	[41]
	$Fe_2(SO_4)_3$ Optimal dose = 300 mg/g	43.99	51	137	–	88	
	$AlCl_3$ Optimal dose = 150 mg/g	18.29	35	118	–	87	
	$FeCl_3$ Optimal dose = 150 mg/g	13.25	32	129	–	86	
Biological sludge SRF = 11.30 × 10^{12} m/kg DS = 12% MC = 98.50% CST = 132 s	PACl Optimal dose = 10%	3.90	55	–	22.50	80.80	[42]
Sewage sludge SRF = 1.72 × 10^{12} m/kg	$FeCl_3$ Optimal dose = 10%	0.07	–	–	–	–	[43]
	PACl Optimal dose = 10%	0.50	–	–	–	–	
Sewage sludge MC = 98% SRF = 2.19 × 10^{12} m/kg CST = 150 s	Cationic PAM Optimal dose = 2 g/kg	1.07	9.77	–	–	74.70	[46]
Waste-activated sludge SRF = 14.20 × 10^{12} m/kg CST = 225 s	PAM Optimal dose = 3 mg/g	0.40	19.70	–	–	–	[47]

Based on all of this information, it can be concluded that synthetic flocculants certainly improve sludge dewatering. However, due to their chemical nature, the application of these flocculants requires many precautions as it can harm both the environment and human health [48,49]. In fact, as reported in the literature, the enhancement of sludge dewatering using metal salts or PAM as flocculants generally requires carefully controlled pH and a specific dosage to ensure the aggregation of solids and rule out the re-stabilization of sludge flocs [43]. Furthermore, admittedly, sludge treated by such chemicals is voluminous and acidic, thus impeding its transport and disposal [24]. Additionally, dewatering devices may be damaged as a result of the intensive use of corrosive ferric salts [50]. Moreover, due to their non-biodegradability, chemical residuals ascribed from synthetic flocculants may remain embedded in the dewatered sludge matrix, subsequently leading to secondary pollution and limiting the use of this sludge [51]. Consequently, sludge handling becomes much more costly as further treatment is required. Generally, serious environmental pollution is associated with with the use of these synthetic polymers. For example, the resulting alum sludge is a source of toxic aluminum that is harmful to both human and marine life. Aluminium may be associated with Alzheimer's disease, and its toxicity towards fish has previously been reported [7,52]. However, obtaining an exact estimation of the toxic sludge produced in a region is difficult as various factors may control its production (the polymer type, water characteristics, sludge characteristics, the process used, etc.) [53].

Generally, the mechanisms of coagulation/flocculation include charge neutralization and the bridging effect, which are appropriate for sludge dewatering. The used polymers can efficiently destroy the relative stability of the charged particles and allow them to agglomerate into large flocs prior to sedimentation and mechanical dewatering. In the dewatering process, the resulting charge neutralization reduces the thickness of the hydrated shell of sludge particles and allows for the enhancement of free water content by compressing the electric double layer and weakening the sludge water surface tension. In deep dewatering, chemicals weaken the water-trapping capability of sludge by destroying the network structure of sludge. Moreover, the bridging effect is also essential in sludge dewatering [54–56].

Considering all of these adverse effects and limitations associated with the application of chemical flocculants as conditioners for sludge dewatering, looking for natural, efficient, and harmless surrogates has become the primary focus of environmentalists, not only to comply with the standard regulations but also to provide safe and costless sludge treatment, transport, and disposal. Thus, natural flocculants derived mainly from plant extracts have been evaluated as possible alternatives to enhance the dewatering process.

4. Plant-Based Flocculants for Sludge Dewatering: Efficiency and Comparison with Chemical Flocculants

Over the last few years, more than 57 plants have been identified as natural coagulants/flocculants for the treatment of various wastewaters [57]. Despite efforts to prove their efficiency in removing various water pollutants, the exploration of plant-based flocculants for sludge dewatering is still limited to only a few plants, such as Moringa, Cactus, Aloe, and Okra, which are believed to be the most effective sludge conditioners due to their active agent content [11,12,58,59]. Indeed, the application of these popular plants as green flocculants could be attributed to their reliable ability to aggregate sludge's colloidal particles into big flocs, allowing for easy sedimentation and separation from the treated water [13,60,61]. Thus, this flocculating activity has piqued the interest of scholars and has spurred on many in the scientific community to exploit them in sludge dewatering and apply them as surrogates for chemical conditioners. The following section sheds light on the most extensively studied plants in the context of sludge dewatering. Later on, a comparison will be made between the plant-based flocculants and synthetic chemical flocculants.

4.1. Moringa

Moringa, and chiefly the *Moringa oleifera* (MO) species, is the most investigated plant in the field of sludge dewatering. The MO seeds allow for the congregation of sludge's solid particles into dense and settleable flocs. In other words, this natural flocculant enhances sludge dewatering due to its ability to strengthen solids by reducing its compressibility and improving its permeability to facilitate bound water release. To gain more insight into Moringa's capacity to aid sludge dewatering, see Table 3, which compiles the previous research works that have assessed the variations in dewatering properties (such as the SRF and the CST) that occur when using Moringa as a flocculant. In fact, Rabea et al. [62] found that the powder of MO seeds decreased the SRF by up to 70%. This result denotes that filterability becomes greater via powder supplementation, hence promoting sludge water removal.

Table 3. Sludge dewatering performance when using Moringa as an alternative to chemical flocculants.

Raw Sludge.	Flocculant	Sludge Dewatering Performance	References
Sewage sludge SRF = 1.22 × 10^{16} m/kg	MO powder Optimal dose = 6 g/L	SRF = 3.43 × 10^{15} m/kg	[62]
Waste-activated sludge SRF = 4.50 × 10^{12} m/kg	MO powder Optimal dose = 4 g/L	SRF = 2.50 × 10^{12} m/kg	[63]
Sewage sludge SRF = 8.00 × 10^{10} m/kg CST = 6.8 s	MO powder Optimal dose = 2 g/L (for SRF) Optimal dose = 3 g/L (for CST)	SRF = 3.30 × 10^{10} m/kg CST = 5.60 s	[64]
	Chemical polymer (Zetag 7653) Optimal dose = 0.05 g/L	SRF = 3.30 × 10^{10} m/kg CST = 3.60 s	
Sewage sludge SRF = 4.45 × 10^{11} m/kg CST = 6.90 s	MO water extract Optimal dose = 4.69 g/L	SRF = 1.22 × 10^{11} m/kg CST = 4.50 s	[65]

Table 3. *Cont.*

Raw Sludge.	Flocculant	Sludge Dewatering Performance	References
Sewage sludge SRF = 0.90 × 10^{12} m/kg CST = 9 s	MO water extract Optimal dose = 3 g/L (for SRF) Optimal dose = 5 g/L (for CST)	SRF = 3.64 × 10^{11} m/kg CST = 7.10 s	[66]
	Chemical polymer (Zetag 8140) Optimal dose = 13 mg/L	SRF = 1.48 × 10^{11} m/kg CST = 5.50 s	
Synthetic Kaolin sludge SRF = 3.90 × 10^{11} m/kg	MO salt extract Optimal dose = 235.58 mg/L	SRF = 1.10 × 10^{11} m/kg	[67]
	Alum Optimal dose = 212.02 mg/L	SRF = 0.80 × 10^{11} m/kg	
Water treatment sludge SRF = 35.10 × 10^{12} m/kg CST = 175.4 s	MO salt extract Optimal dose = 40 mL/L	SRF = 12.10 × 10^{12} m/kg CST = 59.70 s	[68]
	Alum Optimal dose = 40 mL/L	SRF = 6.64 × 10^{12} m/kg CST = 42.20 s	
Water treatment sludge SRF = 1.61 × 10^{13} m/kg CST = 44 s	*Moringa Pergerina* Optimal dose = 100 mL/L	SRF = 1.21 × 10^{13} m/kg CST = 19 s	[69]
	Ferric chloride Optimal dose = 100 mL/L	SRF = 0.97 × 10^{13} m/kg CST = 9 s	

Furthermore, the effects of the extraction methods of MO-based flocculants on sludge dewaterability have been within the scope of interest of many different investigations. In this respect, Muyibi et al. [63] appraised the flocculating efficacy of MO dry powder used directly and dissolved in water or salt solution as conditioning agents to treat waste-activated sludge. Both the dry powder and its water extract exhibited better dewatering performance than the salted MO seed solution. As shown in Table 3, the MO powder decreased the SRF from 4.50 × 10^{12} m/kg to 2.50 × 10^{12} m/kg. Additionally, the treated sludge showed the smallest volume and the most notable settling rate of 66.70% compared to 48% using the salted solution. The results of this study are in accordance with those reported by Wai et al. [64]. The three MO forms—powder, water, and NaCl-aqueous extracts—disclosed comparable CST variations of around 5 s, while the lowest SRF value was registered using the powdered MO form. The latter allowed for a decline in the SRF from 8.00 × 10^{10} m/kg to 3.30 × 10^{10} m/kg (Table 3). In comparison to a cationic polyacrylamide (Zetag 7653, a commercial flocculant) and in the context of sewage sludge dewatering, Wai et al. [64] reported that the efficiency of MO powder to reduce the SRF was as good as that of Zetag 7653. As depicted in Table 3, similar SRF decreases from 8.00 × 10^{10} m/kg to 3.30 × 10^{10} m/kg were achieved using 2 g/L of MO and 0.05 g/L of Zetag 7653. Even though a high natural flocculant dosage was required, the availability, biodegradability, and safety features of MO as a conditioning agent make it an alternative eco-sustainable solution to mitigate all of the threats associated with the use of chemical flocculants.

The significant dewatering performance of MO powder compared to MO water and salt extracts could be ascribed to the capacity of this powder to, once dissolved in the sludge matrix, release the cationic proteins that are responsible for its flocculating ability. Due to its low molecular weight and positive charge density, the cationic MO proteins neutralize the negatively charged solids to generate dense and compacted flocs that are readily settleable via gravitational decantation. On the other hand, the un-dissolved MO powder serves as an adsorbent to collect sludge solids and assist in its dewatering. In fact, it may produce rigid and porous solids of low compressibility and good permeability to enhance water removal.

Regarding MO water and salt extracts, researchers assume that their effectiveness is due to the total solubility of proteins as active agents. Along this line, Tat et al. [65] revealed that MO water extract was better than dry powder and salt (NaCl) extracts with respect to reducing SRF and CST. Also, in dewatering a sewage sludge using this natural flocculant, Mohammad et al. [66] found significant decreases in SRF (from 0.90 × 10^{12} m/kg to 3.64 × 10^{11} m/kg) and CST (from 9 s to 7.10 s). Nonetheless slightly better SRF (1.48 × 10^{11} m/kg) and CST (5.50 s) declines were observed using a synthetic cationic polyacrylamide (Zetag 8140).

Likewise, the MO seeds extracted by NaCl salt exhibited a comparable SRF reduction (72%) to that found using alum (79%) in a sludge dewatering process based on a drinking water treatment method [67]. These results are in agreement with those reported by Ghebremichael and Hultman [68].

Interestingly, with the increase in awareness about the necessity to look for natural resources as alternatives to chemical flocculants for sludge dewatering, another species of Moringa, *Moringa peregrina* (MP), was explored for the first time by Mazaheri et al. [69] to dewater a sewage sludge. Similarly, to MO, the MP was prepared by dissolving its seeds powder in NaCl solution. A significant decline in CST from 44 s to 19 s was achieved using 100 mL/L of MP. Similarly, the SRF value dropped from 1.61 × 10^{13} m/kgto 1.21 × 10^{13} m/kg, whereas larger reductions in CST (9 s) and SRF (0.97 × 10^{13} m/kg) were found using 100 mL/L of $FeCl_3$. From an environmental point of view, due to its efficiency, safety, biodegradability, and sustainability, the application of MP as a bioflocculant instead of $FeCl_3$ as a synthetic flocculant is favorable.

4.2. Cactus and Aloe

In contrast to Moringa, a few studies have addressed sludge dewatering using Cactus and Aloe. To the best of our knowledge, only two research works have investigated the efficiency of these plants in assisting in the sludge dewatering process. For instance, in a study related to the application of Cactus (*Opuntia ficus indica*), Betatache et al. [11] reported that replacing a chemical flocculant with cactus juice for sewage sludge dewatering is feasible. In comparison with a few tested synthetic organic cationic (Chimfloc C4346), anionic (Sedipur AF 400), and non-ionic (Sedipur NF 102) polymers, as well as metal salts—namely alum (Al_2SO_4) and iron chloride ($FeCl_3$)—the cactus juice showed the best dewatering performance. In fact, as presented in Table 4, the SRF value obtained using the cactus juice (SRF = 0.17 × 10^{12} m/kg) was slightly less than those obtained using the cationic polymer (SRF = 0.3 × 10^{12} m/kg), the non-ionic polymer (SRF = 4 × 10^{12} m/kg), and alum (SRF = 1.3 × 10^{12} m/kg). This result implies that cactus-derived flocculants have the capacity to enhance sludge dewatering.

Table 4. Application of Cactus and *Aloe vera* as natural flocculants in sludge dewatering.

Raw Sludge	Flocculant	Sludge Dewatering Performance	References
Sewage sludge SRF = 1.03 × 10^{13} m/kg	Cactus juice Optimal dose = 0.4 g/kg	SFR = 0.17 × 10^{12} m/kg	[11]
	Cationic polyacrylamide (Chimfloc C4346) Optimal dose = 6 g/kg	SFR = 0.3 × 10^{12} m/kg	
	Non-ionic polyacrylamide (Sedipur NF 102) Optimal dose = 6 g/kg	SFR = 4 × 10^{12} m/kg	
	Alum Optimal dose = 40 g/kg	SFR = 1.3 × 10^{12} m/kg	
	$FeCl_3$ Optimal dose = 80 g/kg	SFR = 1.3 × 10^{12} m/kg	
Municipal wastewater sludge Settling rate = 55%	*Aloe vera* gel Optimal dose = 3 mL/L	Settling rate = 67.50%	[59]

On the other hand, regarding the use of *Aloe vera*, Jaouadi et al. [59] appraised the efficiency of aloe gel as a flocculant to treat sewage sludge. They noticed that the addition of this natural flocculant as a conditioner promotes the raw sludge's settleability, and an enhanced settling rate of 67.50% was achieved using 3 mL/L as a result of the aggregation of flocs. Moreover, it can be deduced that this improvement in particle strength allowed for the removal of the trapped bound water, hence facilitating solid–liquid separation.

Based on these promising findings, cactus and aloe vera have the potential to be bioflocculants for sludge dewatering, and their polysaccharide content and specifically their polygalacturonic acid content are regarded as the main agents responsible for their flocculating abilities [60]. Due to its long polymeric chains, polygalacturonic acid provides a bridge to adsorb the suspended sludge solids and binds them together in order to produce strong and dense aggregates. Consequently, the compressed solids enable the smooth withdrawal of bound water, facilitating sludge dewatering.

4.3. Okra

Like Cactus and Aloe, Okra (*Abelmoschus esculentus*) is a common polysaccharide-based flocculant widely applied for wastewater treatment [61,70]. However, its application in sludge treatment is still limited. To fill this gap, Lee's group has explored the dewatering efficiency and feasibility of using Okra in lieu of conventional synthetic flocculants for sludge dewatering [12,71,72]. Scholars have paid great attention to assessing how the methods used to extract bioflocculants (conventional hydrothermal and microwave-assisted extraction) affect their dewatering abilities. In a preliminary study conducted by Lee et al. [12], Okra water extract and its oven-dried powder (acquired after conventional hydrothermal extraction) were evaluated in terms of their efficiency in dewatering synthetic kaolin sludge. Both natural flocculants showed significant SS removal and water recovery rates exceeding 98% and 68%, respectively (Table 5). Further improvements in these dewatering properties were observed using microwave-extracted powder as salient SS removal (99%) and water recovery (75%) rates were attained [72]. The prominent dewatering capabilities of okra-based flocculants are merely attributed to the high solubility of their polysaccharides with the increase in extraction temperature when using microwave extraction. Likewise, compared to conventional synthetic flocculants (Table 5), Okra showed higher SS removal and water recovery rates than a cationic and anionic PAM. According to these interesting findings recorded by Lee et al. [12,71,72], the dewatering efficiency of Okra, and specifically its microwave-extracted powder, as a bioflocculant makes it a relevant candidate to replace the commonly used chemical flocculants.

Table 5. Sludge dewatering using Okra as natural flocculant.

Raw Sludge	Flocculant	Sludge Dewatering Performance	References
Synthetic Kaolin sludge	Okra water extract Optimal dose = 175 mg/L	SS removal $\geq$ 98% Water recovery $\geq$ 68%	[12,71,72]
	Okra oven-dried powder Optimal dose = 150 mg/L	SS removal $\geq$ 98% Water recovery $\geq$ 68%	
	Okra microwave-extracted powder Optimal dose = 30 mg/L	SS removal $\geq$ 99% Water recovery $\geq$ 75%	
	Cationic polyacrylamide (FO 4400 SH) Optimal dose = 70 mg/L	SS removal $\geq$ 98% Water recovery $\geq$ 65%	
	Anionic polyacrylamide (AN 934 SH) Optimal dose = 50 mg/L	SS removal $\geq$ 95% Water recovery $\geq$ 60%	

5. Potential Dewatering Flocculants: Synthetic or Plant-Based Ones

Overall, in spite of the paucity of research studies on the application of plant-based flocculants for sludge dewatering, the aforementioned results obtained using Moringa, Cactus, Aloe, and Okra have elucidated their ability to separate water from the solids of sludge, implying that their use in sludge dewatering is apt. For all of these plants, researchers have made efforts to optimize their extraction conditions (solvent concentration, extraction with water or salt, oven-drying, microwave-assisted extraction, temperature) to produce suitable bioflocculants with efficient dewatering activities. Considering the time and costs associated with their extraction procedures, the aforementioned plants are deemed to be effective biomaterials capable of enhancing sludge dewaterability.

Furthermore, as depicted in Tables 3–5, it can be seen that the SRF and CST are the most appraised dewatering parameters. Dry matter content, time to filter, sludge volume index, volume of filtrate, and water recovery rate were also assessed in some investigations [4,59,71]. Compared to the raw sludge, all of the discussed plant extracts exhibited outstanding SRF and CST decreases. The reduction in these two parameters reflects their potential to modify the physicochemical properties of sludge. As a matter of fact, the decrease in SRF values denotes an improvement in sludge filterability as a result of an increase in the porosity of its solids. Consequently, the sludge cake becomes more permeable to allow for the release of the bound water stuck in the EPS. In parallel, the compressibility of its solids decreases, allowing for water content to quickly decrease during filtration.

Moreover, as shown in Tables 3–5, it can be said that the dewatering performance of bioflocculants is comparable (in case of Moringa) and even superior (in the cases of Cactus and Okra) to PAM. However, it is worth noting that, in spite of the fact that higher plant extract dosages were required to compete with their chemical counterparts, these natural products are beneficial for sludge dewatering as they may help the process become rid of the detrimental environmental damage and human health risks associated with it [73]. They are also highly biodegradable, abundant, locally available, and can be cultivated and derived from renewable resources [74]. Hence, the application of these natural flocculants to dewater sludge is regarded as economically feasible in terms of raw materials procurement.

Moreover, using these innocuous bioflocculants in dewatering processes would be easier compared to using chemicals because neither pH adjustments nor health precautions are required to reduce sludge volume. This volume was found to be three times smaller when Moringa was used instead of alum [75]. Consequently, this notable reduction could definitely minimize the cost of sludge handling, transport, and disposal.

Furthermore, due to the fact that plant-based flocculants are natural, the dewatered sludge treated using natural flocculants can be relatively free from chemicals depending on the type of the raw effluent. Taking into consideration their antimicrobial activity, the use of plants as flocculants also ensures the removal of pathogens and unpleasant odors from the sludge, further demonstrating their utility in protecting the environment and human health [59,76]. Moreover, the dewatered sludge is deemed to be a nontoxic byproduct suitable for applications in agriculture as a soil conditioner or fertilizer due to its high nutrient content. This sustainable strategy fits well with the concept of "waste to wealth". Thus, alongside their promising activity to aid the dewatering process, the application of plant-based flocculants instead of chemical ones opens up profitable opportunities to reuse the produced sludge, which may, in turn, alleviate the economic burdens related to the management, disposal, cost, and safety of sludge.

6. Challenges and Future Prospects in the Application of Plant-Based Flocculants for Sludge Dewatering

As evidenced by the studies highlighted in this review, the most frequently used plants to dewater sludge are polysaccharide-based flocculants such as Cactus, Aloe, and Okra. The use of these natural extracts as green conditioners will likely inspire researchers to look for other plants that contain polysaccharides to investigate their dewatering performance. Further studies should be also conducted to provide a greater understanding of the efficiency of bioflocculants. To fulfill this research objective, future studies should clarify the effects of sludge's physicochemical properties (water content distribution, nature of EPS, solids content) on its dewaterability. Investigations into its rheological properties are also highly recommended because sludge viscosity not only affects sludge's filterability (block of water release) but it may also deteriorate the equipment used for dewatering (e.g., the clogging of filters). Moreover, variations in EPS components and their interactions with plant-based flocculants during the conditioning stage are still lacking; nevertheless, most of the previous works in the literature have systematically correlated dewatering characteristics such as the SRF and CST with the capacity of sludge solids to release the bound water entrapped in the EPS matrix [12,64]. Hence, scientific evidence regarding this phenomenon is urgently required (for example, characterization through Scanning Electron Microscopy (SEM) or assessing a sludge cake via fluorescence spectroscopy). In addition, the mechanisms of dewatering when using plants as flocculants should be elucidated in future research studies. Furthermore, with the aim to scale up their application, techno-economic analyses conducted through the life cycle assessment (LCA) method are greatly needed to fully describe sludge dewatering processes involving raw sludge collection, treatment, bioflocculant preparation, and disposal, as well as the environmental impact associated with each stage of the process.

7. Conclusions

For the sake of environmental and human health preservation, substituting conventional synthetic flocculants in sludge dewatering with safe, clean, and natural ones derived from plants appears to be an ecofriendly approach. However, despite their biodegradability, availability, and cost-effectiveness, the potential utility of these natural extracts for use in the sludge dewatering process has still not been adequately addressed. Unfortunately, only four plants, namely Moringa, Cactus, Aloe, and Okra, have been investigated in the literature; however, all have shown efficient dewatering performances that are comparable and even superior to chemical flocculants. Prominent decreases in sludge SRF and CST transcending 90% were found for polysaccharide-based flocculants such as Cactus and Okra. Moreover, their application can be regarded as a profitable and an eco-sustainable strategy not only to enhance the dewatering process but also to promote environmentally friendly sludge disposal and reuse, especially in the field of agriculture as sludge with a high nutrient content can be deployed as a soil conditioner or fertilizer. Thus, on one hand, extensive studies are needed to fill the knowledge gaps in appraising the dewatering abilities of other natural resources to facilitate their large-scale application, and on the other hand, evaluating the reliability of the produced sludge for agriculture use is necessary to make sludge dewatering, management, and disposal more green and clean.

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