



## Article

# Simultaneous Environmental Waste Management through Deep Dewatering of Alum Sludge Using Waste-Derived Cellulose

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**Abstract:** To simultaneously solve problems in an eco-friendly manner, introducing a waste residual as a sustainable conditioner to aid alum sludge dewatering is suggested as a cradle-to-cradle form of waste management. In this regard, the superiority of deep dewatering alum sludge with a powdered wood chip composite residual as a novel conditioner was explored, whereby traditional conventional conditioners, i.e., polyelectrolytes and lime, were substituted with powdered wood chips. Initially,  $\text{Fe}_3\text{O}_4$  was prepared at the nanoscale using a simple co-precipitation route. Next, wooden waste was chemically and thermally treated to attain cellulosic fine powder. Subsequently, the resultant wood powder and  $\text{Fe}_3\text{O}_4$  nanoparticles were mixed at 50 wt % to attain a wood powder augmented with iron, and this conditioner was labeled nano-iron-cellulose (nIC-Conditioner). This material (nIC-Conditioner) was mixed with hydrogen peroxide to represent a dual oxidation and skeleton builder conditioning substance. Characterization of the resultant conditioner was carried out using transmission electron microscopy (TEM) and Fourier transform infrared (FT-IR) transmittance spectrum analysis. The feasibility of the experimental results revealed that the moisture content in the sludge cake was lower after conditioning, and the capillary suction time (CST) was reduced to 78% compared to that of raw alum sludge after 5 min of dewatering time. Moreover, the optimal system parameters, including nIC-Conditioner and  $\text{H}_2\text{O}_2$  concentrations, as well as the working pH, were optimized, and optimal values were recorded at 1 g/L and 200 mg/L for nIC-Conditioner and  $\text{H}_2\text{O}_2$ , respectively, with a pH of 6.5. Additionally, scanning electron microscope (SEM) analyses of the sludge prior to and after conditioning were conducted to verify the change in sludge molecules due to this conditioning technique. The results of this study confirm the sustainability of an alum sludge and waste management facility.



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## 1. Introduction

Alum sludge is an inevitable aluminum-based waste product of waterworks plants [1,2]. Due to urbanization and industrialization, the amount of aluminum-based sludge waste is continuously increasing, as water treatment is inevitable and unpreventable [3–5]. Aluminum-based sludge has a high water content, special physical texture, and specific handling and transportation requirements, which continue to cause issues for managing such waste [6]. Moreover, alum sludge is difficult to dewater, and thus, its water reduction is essential to controlling its disposal in an economic way [7]. In this regard, the conditioning of alum sludge in waterworks plants is essential to facilitate dewatering techniques [8,9]. The alum sludge conditioning scheme's objective is to bind the physically and/or chemically bound water onto a solid surface and then take water into the sludge flocs [10]. Therefore, the conditioning

policy is to transform the floc strength together with the quantity of bound water [11]. Significant enhancement in filterability has been achieved, as alterations in solid/liquid phase hydrodynamics have been shown to play a vital role in sludge's water content [3]. The result is a sludge that possesses a high-comparability nature [3,4]. Thus, the search for a special conditioner that possesses environmentally benign characteristics with cost-efficient criteria is attracting scientists' attention.

Many potential sludge-conditioning approaches have been introduced, especially for alum sludge, to facilitate its dewatering performance. For instance, polyelectrolytes have shown a pronounced effect in alum sludge conditioning, although their toxicity and danger to the environment are concerning [12]. Additionally, freezing/thawing, thermal hydrolysis, and sonication combination [13] have been introduced as conditioning methods; however, these are uneconomic techniques due to their high energy consumption. Chemical conditioners such as lime, gypsum, and oxidants have also previously been applied [14,15], but the extensive use of extra reagents as non-benign chemicals still causes severe environmental damage. The advanced oxidation conditioning method has been proposed as being superior due to its environmental friendliness [16]. Hence, exploring and taking into account decreased chemical use, but attaining superior sludge treatment, is a target for researchers [17–19]. Among the conditioning methods used, advanced oxidation processes (AOPs), especially the Fenton-like reaction, are the superior proposed dewatering methods [20].

The utilization of a transition metal source, especially an iron source augmented with a hydrogen peroxide reagent, could generate the hydroxyl radical ( $\cdot\text{OH}$ ) that might attack sludge molecules and enhance their floc formation, which improves dewatering performance. Previous studies [13,20–22] demonstrated the use of such an oxidative reaction test in sludge conditioning. Moreover, numerous substances have been applied to skeleton builder sludge conditioning, such as gypsum [23], carbonaceous-based materials [24], squeezed sugar cane residuals [25], and zeolite-type materials [26]. However, to the best of the authors' knowledge, the augmentation of Fenton-type materials, especially nanoparticle structure materials and wood chips as waste skeleton builder material, has not yet been carried out. Such augmentation might possess the advantages of both the oxidation-type reaction and skeleton builder materials, which provide high porosity throughout filtration [27]. Scientists' and researchers' mission is to provide a superior conditioner from the perspective of environmental benignity and efficiency [28]. Hence, augmentations of the above-mentioned methodologies are significant, as multilevel features of advantages support the concept of eliminating serious environmental destruction.

The sustainable endpoint solution of dewatered alum sludge is also gaining researchers' interest. Dewatered sludge management is being studied due to its potential to offer a green sustainable waste disposal opportunity that conserves the ecosystem. Thus, alternative alum sludge disposal methods that facilitate its conversion to value-added material are valuable options that satisfy environmental restrictions and laws. In this regard, crucial research has been conducted, and numerous sludge cake applications have been suggested. Such applications include using dried cake in soil enhancements and amendments, in water treatments as a coagulant or adsorbent material, in construction materials, in sludge conditioning to enhance the dewaterability of various sludge types, and in wetland applications [18]. Although these dewatered sludge cake applications have been proposed, unexplored concerns require study before widening limited real-world applications. Such concerns include anti-clogging techniques used in constructed wetland applications and problems linked to the medium's pH. Careful evaluation is essential in soil applications. Thus, further investigation is required before undertaking specific applications.

In this study, the dual-conditioning concept of chemical oxidation and physical skeleton builder materials was applied to waterworks residual alum sludge. The benignity of such work supported the use of this suggested technique for real-world applications. The physical wood chips conditioner and co-chemical Fenton oxidation conditioning were conducted as an advanced oxidation process conditioning, and results were compared with

commercial polyelectrolytes conditioners. This study suggested using alum sludge from a waterworks plant in dewaterability conditioning. The operating parameters were studied and optimized for better performance.

## 2. Experimental Section

### 2.1. Materials

#### 2.1.1. Alum Sludge

Alum sludge is a waste by-product from waterworks plants, formed as a result of using aluminum sulphate as a primary coagulant. The alum sludge used in this study was collected from the primary channel after the secondary clarifier. Subsequently, the sludge was transferred to the laboratory in a washed plastic container for analysis. The sludge had a high moisture content (MC) of 99.59%, suspended solid (SS) content of 12,043 mg/L, and capillary suction time (CST) of 46 s. The water supernatant's turbidity was 243 NTU, and it had a pH of 8.5.

#### 2.1.2. Conditioner Preparation

Ferrous and ferric sulfate precursors supplied by Qualikems Fine Chem Pvt. Ltd. (Vadodara, India) were used to prepare magnetite nanoparticles. Next, 2 mol of  $\text{Fe}_2(\text{SO}_4)_3$  and 1 mol of  $\text{Fe}(\text{SO}_4)$  were mixed using 50 mL of water; then, NaOH solution was added in a drop-wise manner to raise the pH to the alkaline range (it reached a pH of 11.0). Thereby, magnetite nanoparticles began to precipitate in the solution mixture. Subsequently, the solution was heated at 80 °C and stirred for one hour prior to being repeatedly washed to reduce the pH to 7.0. The attained  $\text{Fe}_3\text{O}_4$  was dried in an electric oven at 80 °C until its weight was reduced.

Wood waste chips were collected, sequentially washed in distilled water, and then dried at 105 °C for 12 h to attain a cellulosic fiber. The cellulosic fiber was hydrolyzed through HCl treatment by heating at 90 °C for 15 min using an HCl concentration of 25 N. Hydrolysis was conducted using 25 g of sawdust exposed to 200 mL of HCl. The resultant mixture was filtered and successively washed to attain a neutral pH prior drying at 60 °C to reach a constant weight. Next, the material was bleached using  $\text{H}_2\text{O}_2$  (3%) and heated for one hour at 90 °C. It was then washed until its pH was neutral and finally dried at 105 °C; the resultant material was isolated cellulose.

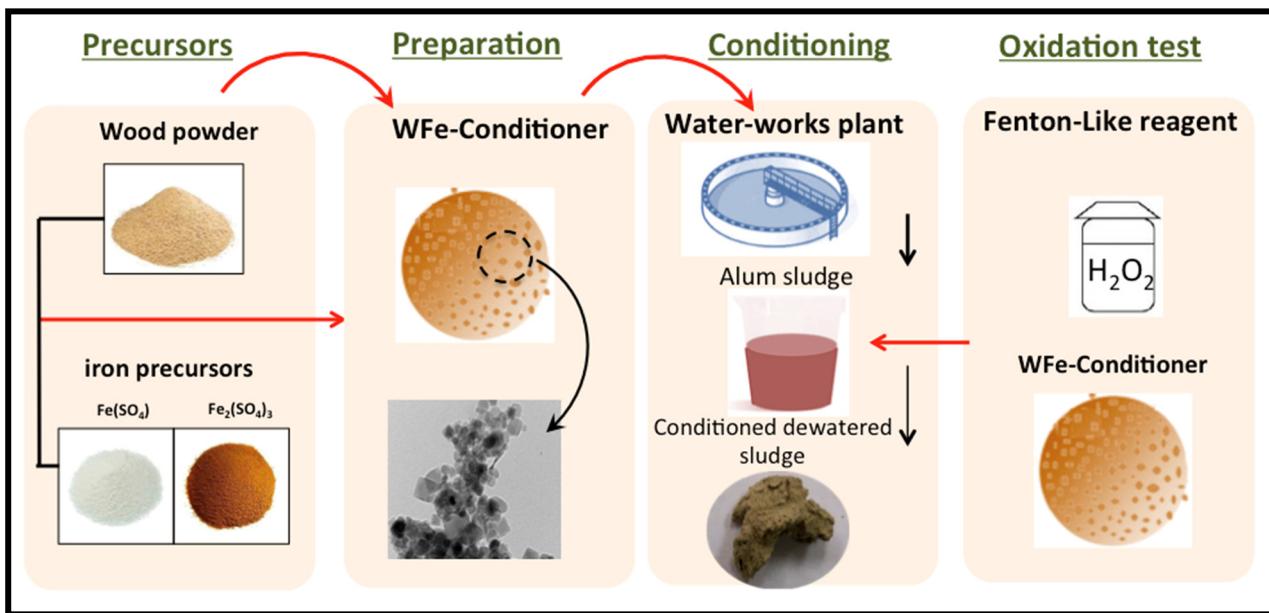
To prepare a dual catalyst/skeleton builder conditioner, a specific amount of the as-synthesized  $\text{Fe}_3\text{O}_4$  nanoparticles was mixed with the attained cellulose material at a mixture weight percent of 50%. The mixture was ground until it was homogenous, then heated in a microwave oven for 5 min at 200 watts. The result was a brownish composite identified as nIC-Conditioner (nano-iron-cellulose) that was ready for use as a conditioner.

Furthermore, the suggested conditioner was compared with commercially available conditioners. Both anionic and cationic polyelectrolytes are applied as conditioners. LT-25 supplied by CIBA Specialty Chemicals was used as the anionic polyelectrolyte, and Magnafloc FO-4140 supplied by Snf Sas Zac De Milieux (Andrezieux, France) was applied as the cationic polyelectrolyte; both were used as received, with no further purification or treatment.

### 2.2. Methodology

First, 100 mL of the sludge sample was poured into a beaker and subjected to a jar test experiment. The solution's pH was adjusted, when required, using drops of diluted sulfuric acid (1:9) and/or 1 M sodium hydroxide. Next, the nIC-Conditioner (nano-iron-cellulose) was added to the beakers in specific amounts ranging from 0.2 to 1.0 g/L. To initiate the oxidation reaction, the addition of the catalyst (nIC-Conditioner) was followed by the addition of analytical-grade hydrogen peroxide (40%, *w/w*). The hydrogen peroxide's role was to initiate the oxidation reaction through radical formation. Next, the jar test was applied to produce the conditioning reaction, and magnetic stirring was applied (initial rapid stirring for the first 30 s followed by slow stirring according to the required conditioning time) and was checked and investigated through the 1 to 6 min range. After conditioning, the sludge

was analyzed using a capillary suction time (CST) test. Figure 1 demonstrates a schematic graphical illustration of the experimental steps.



**Figure 1.** Schematic graphical representation of the experimental steps.

### 2.3. Analytical Determinations

The capillary suction time (CST) test was applied to assess the sludge's dewatering performance as an indication of better conditioning performance. Capillary suction time testing is a rapid method for characterizing the filterability and dewaterability of a given sludge. The test is conducted by pouring slurry into a small open tube resting on a piece of filter paper. Capillary suction is applied to extract the filtrate, and the cake is formed at the bottom of the tube. The filtrate's extraction rate depends on the cake's resistance. Filterability is measured by calculating the time it takes to wet the filter paper. This method relies on the varying resistance applied by water moving through the filter paper. Each test can be completed in a few minutes, and the rate of water passing through the filter paper varies depending on the condition of the sludge and the filterability of the cake formed on the filter paper [20]. A Triton-WPRL type 304M CST (Triton Electronics Ltd., Essex, UK) was used to measure the CST capability. The pH was adjusted to desired values using a digital pH meter (Model AD1030, Adwa instrument, Szeged, Hungary). The supernatant's turbidity was investigated using an ICM turbidimeter (Columbus, OH, USA), and its suspended solids (SS) content was inspected using standard methods [29].

### 2.4. Structural Characterization and Morphology

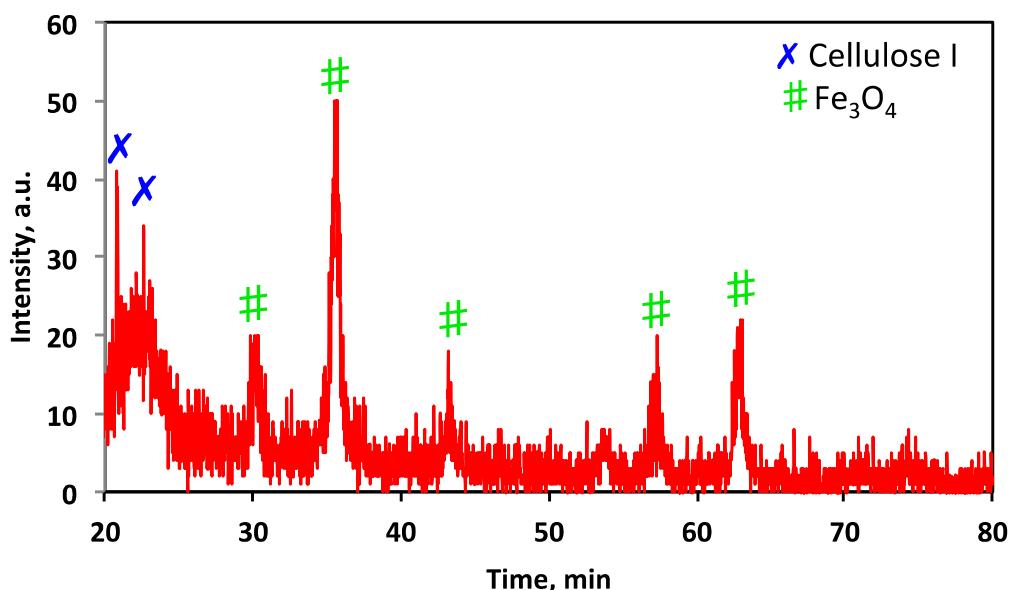
An XRPhilips X'pert diffractometer model MPD3040 (Malvern, UK) was used to explore the crystal structure of the prepared nIC-Conditioner sample, using a Cu K $\alpha$  radiation source ( $\lambda = 1.5406$ ) run at 40 kV and 40 mA at a step-scan mode of 0.02°. However, the nIC-Conditioner sample's morphology was investigated via scanning electron microscopy (SEM) accompanied by energy dispersive X-ray spectroscopy (EDX) and transmission electron microscopy (TEM). SEM, using model Quanta FEG 250 (Facultad de Químicas, Madrid, Spain), was used to obtain SEM images, and TEM, using model Tecnai G20 (Beijing, China), facilitated the use of FEI (Beijing, China) to investigate the sample's morphology through TEM image analysis, with applied typical magnifications of  $\times 8000$  and  $\times 60,000$ .

### 3. Results and Discussion

#### 3.1. Characterization of nIC-Conditioner

##### 3.1.1. X-ray Diffraction Characterization

Figure 2 shows the X-ray diffraction (XRD) patterns of the prepared conditioner (nIC-Conditioner) (Beijing, China). The attained pattern had almost identical diffraction line positions and verified the formation of cellulose and the  $\text{Fe}_3\text{O}_4$  composite. The data presented in the figure reveal that the recorded lines were in accordance with standard crystallographic data in the reference pattern (JCPDS 36-1451) [30]. According to the data displayed in Figure 2, the pure cubic spine crystal structure of magnetite peaks was present in the sample. The appearance of hkl at  $30.0^\circ$  [220],  $35.3^\circ$  [311],  $43.0^\circ$  [400],  $56.9^\circ$  [511], and  $62.8^\circ$  [440] verified the  $\text{Fe}_3\text{O}_4$  diffraction peaks in the composite. Moreover, the structurally sharp peaks of cellulose I displayed hkl of an  $2\theta$  at  $22.4^\circ$  [101] and  $2\theta$  at  $15.6^\circ$  [002], confirming the presence of cellulose from wood residuals.



**Figure 2.** X-ray diffraction patterns of the nIC-Conditioner catalyst composite.

##### 3.1.2. TEM Images

The isolated cellulose from wood chip residuals and the composite of cellulose coated with magnetite (nano-iron-cellulose, nIC) were exposed under a transmission micro-scope, as seen in Figure 3A,B. The images in Figure 3 illustrate the sheet-like morphology of a pristine cellulose substance (Figure 3A). However, Figure 3B, showing a TEM micrograph of the synthesized composite nanoparticles of cellulose augmented with magnetite, displays uniformly shaped dense spheres of magnetite nanoparticles aggregated over the sheet cellulose's surface.

##### 3.1.3. EDX Analysis

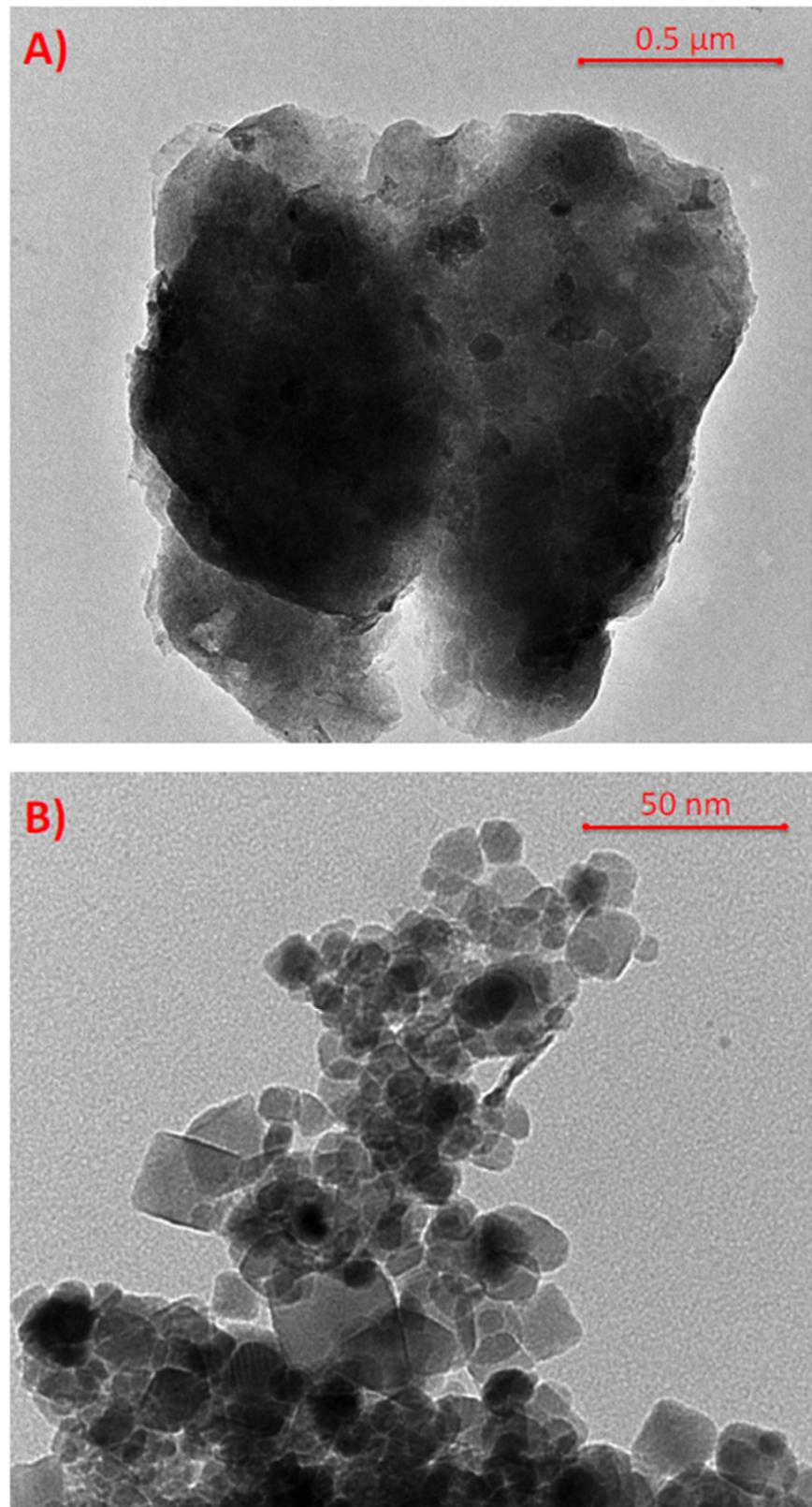
An elemental analysis of alum sludge was undertaken using EDX. The alum sludge's main component was C (33%), along with Si (8.7%), Al (9.5%), and small amounts of S, Ca, Na, and Fe. The addition of aluminum sulfate as a primary coagulant in the waterworks treatment plant was represented and reflected in the sludge's composition. Moreover, the loss on ignition was 39%.

### 3.2. Dual Chemical Conditioner/Skeleton Builder Dewatering

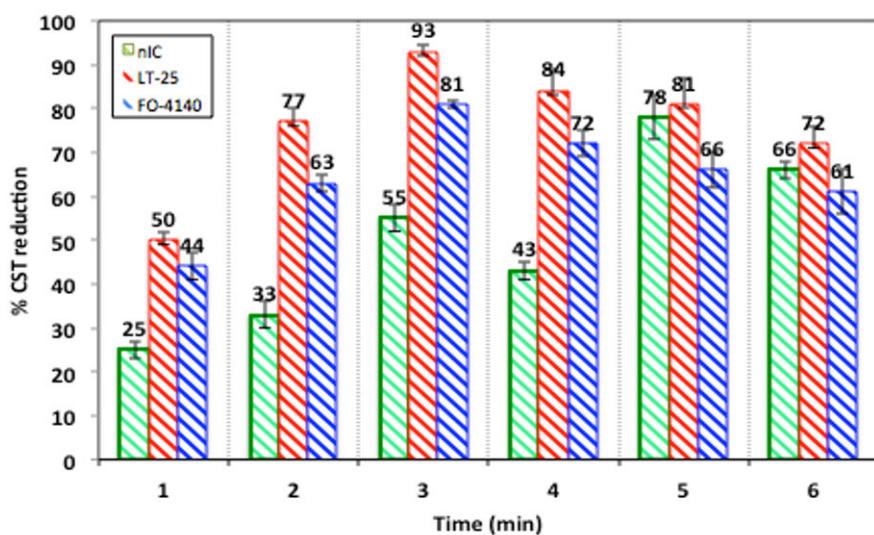
#### 3.2.1. Conditioning Time

The effect of the hybrid nIC-Conditioner/ $\text{H}_2\text{O}_2$  and its action as a skeleton builder were investigated in accordance with the conditioning time, as shown in Figure 4. Furthermore, a comparison of this environmentally benign conditioner with commercial

polyelectrolytes is presented in Figure 4. This conjugated comparison was based on the reduction in the CST value. Generally, the minimum value of the capillary suction time (CST) is preferred, as it corresponds to the maximum dewatering performance. The reason for the observed values remains unclear in the current work without further exploration.



**Figure 3.** TEM images of “(A)” pristine cellulose and “(B)” nIC-Conditioner catalyst composite.



**Figure 4.** CST reduction efficiency of various conditioners at various conditioning times.

The conditioning test results for the various conditioner types were evaluated according to various flocculation times in the range of one to six minutes. According to the preliminary results, LT-25 (Johannesburg, South Africa) and Magnafloc FO-4140 (Johannesburg, South Africa) were added according to their optimal dose of 10 mg/L. As shown in the data displayed in Figure 4, composite nIC-Conditioner/H<sub>2</sub>O<sub>2</sub>, as the source of the Fenton conditioner, was obtained and compared with LT-25 and Magnafloc FO-4140, which are anionic and cationic polyelectrolytes, respectively. When nIC-Conditioner/H<sub>2</sub>O<sub>2</sub> was applied, CST reduction was enhanced compared to that with both polyelectrolytes. Using the nIC-Conditioner/H<sub>2</sub>O<sub>2</sub>, the CST reduction reached 78% within a 5 min oxidation time. However, the conditioning rate reached 93% and 81% using polyelectrolyte conditioners LT-25 and Magnafloc FO-4140, respectively, with a 3 min conditioning time. Thus, the results revealed that the highest conditioning performance was noted for the anionic polyelectrolyte's CST reduction. This can be explained by the presence of the anionic polymer promoting the dry solid content in the sludge for floc formation [12].

In comparison, the nIC-Conditioner/H<sub>2</sub>O<sub>2</sub>-based Fenton system could not bridge the high floc size of alum sludge molecules. Consequently, the produced sludge flocs for polyelectrolytes were larger than those based on nIC-Conditioner/H<sub>2</sub>O<sub>2</sub> Fenton conditioning were. Thus, the water discharge from the sludge declined when using the nIC-Conditioner compared to that when using the polyelectrolyte conditioner, which was signified by a decline in its CST percentage [20].

The data also revealed the optimal time associated with the maximum reduction in CST. However, more than 3 min of polyelectrolyte conditioning was unfavorable, as was more than 5 min of Fenton conditioning. This could be linked to the size of the floc attained after the flocculation, with the Fenton-based nIC-Conditioner substance being principally accountable for improving the dewaterability [31]. Moreover, the sawdust functioned as a skeleton builder; therefore, the Fenton-based sawdust was a dual conditioner, as it was a skeleton builder and oxidized the alum sludge.

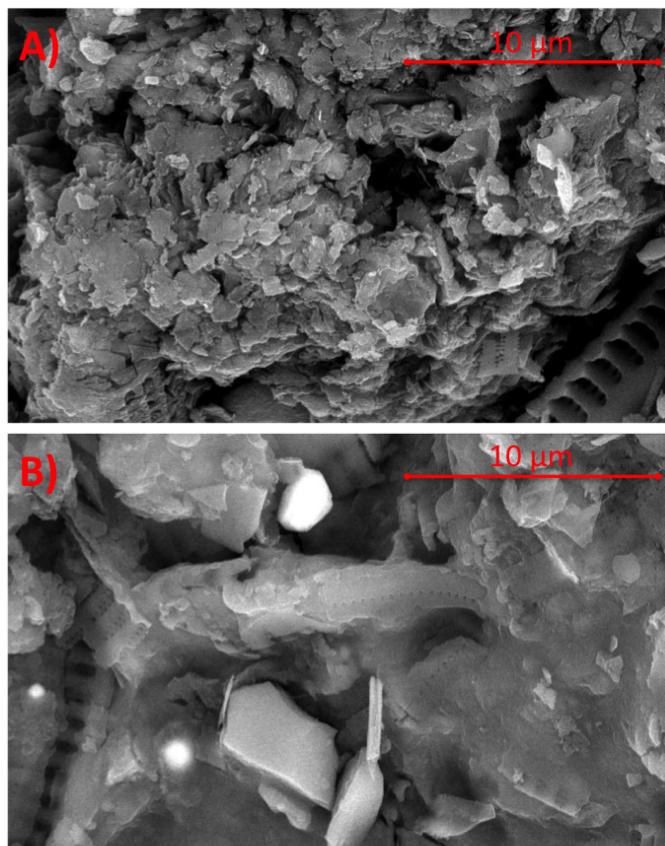
In addition, the presence of polyelectrolytes promoted the dry solid content in the sludge for floc development. Although using polyelectrolytes improved the dewatering performance, with a higher yield compared to the Fenton system, it is noteworthy that the nIC-Conditioner is a waste valorization material and associated with a circular economy [32], whereas polyelectrolytes are toxic substances [23].

The zeta potentials of the alum sludge before and after conditioning were investigated and compared using a Zetasizer Ver. 6.32 (Worcestershire, UK) at 25 °C. The zeta potential of the raw sludge was -12.1 mV, and that of the conditioned sludge was 0.23 mV. Hence, the raw sludge's zeta potential changed to a more positive value, and the sludge flocs were

bigger than those in the raw sludge. Furthermore, when the particle sizes were investigated, alum sludge particles had an average size of  $1.87\text{ }\mu\text{m}$ , and conditioned sludge particles, on average, were  $2.43\text{ }\mu\text{m}$ . Thus, the floc size increased as expected.

#### SEM Images

Figure 5A,B illustrates the SEM analyses of the surface micromorphologies of raw sludge and sludge conditioned using the Fenton-based nIC-Conditioner. These images illustrate that the raw and conditioned sludge cakes possessed various textures that signified floc formation in the conditioned sludge, which had a better dewaterability. Figure 5A shows that the sludge molecules and particles settled in a lamellar structure. The raw sludge's surface was smooth, and its structure was non-porous. However, Figure 5B shows a particle size increase, which signifies an increase in the floc size due to conditioning [32]. This was due to the presence of hydrogen peroxide augmented with the sawdust/magnetite conditioner, which acted as a skeleton builder and oxidizing agent, and led particles to agglomerate and increase in size [23]. Moreover, conditioned sludge cake structures with flocculants bridged the adjacent colloids. This was also a factor in the change in the conditioned sludge's surface in comparison with the raw alum sludge.

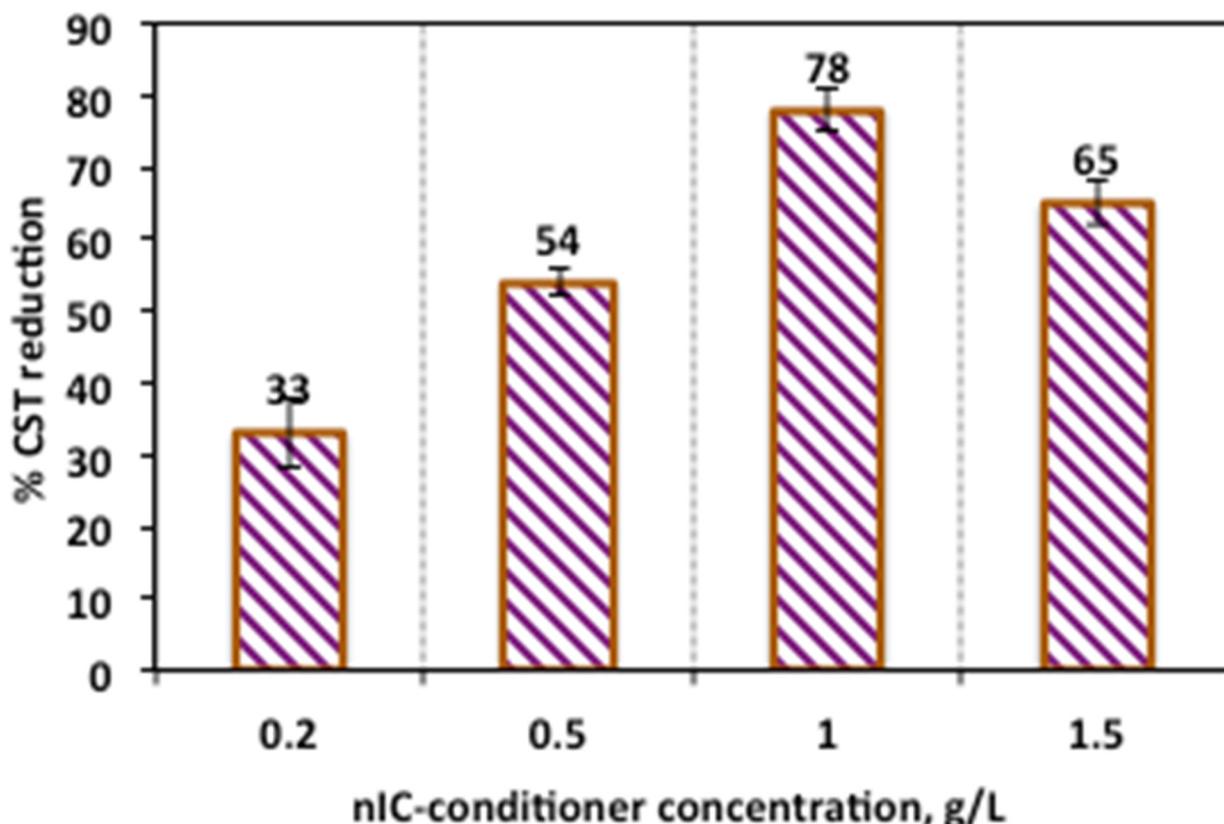


**Figure 5.** SEM images of “(A)” raw sludge and “(B)” sludge conditioned using the nIC-Conditioner catalyst composite.

#### 3.2.2. Effect of nIC-Conditioner Dose

The effect of the nIC-Conditioner dose on the Fenton oxidation and skeleton builder conditioner is an important parameter, as the dose plays a vital function in the conditioning system's efficiency. Experiments were conducted using  $200\text{ mg/L}$  of hydrogen peroxide at a pH of 6.5 and a nIC-Conditioner catalyst in an amount that varied from 0.2 to  $1.5\text{ g/L}$ ; the resulting CST reduction efficiencies were examined. The data exhibited in Figure 6 show that the nIC-Conditioner catalyst dose affected the CST reduction associated with the filterability regime. These data illustrate that the rate-limiting stage in the conditioning

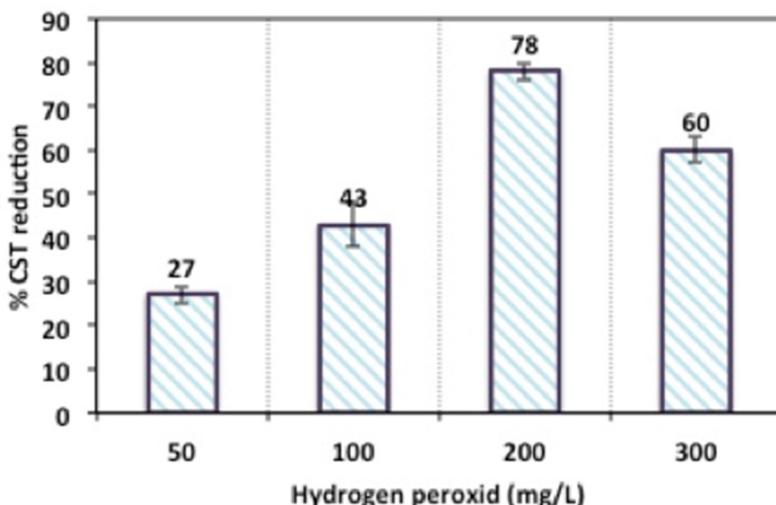
system is the conditioner dose. An increase in the conditioner dose resulted in an increment in CST reduction efficiency. Optimal performance was linked to a dose of 1 g/L. However, a conditioner dose greater than 1 g/L resulted in a decline in dewatering efficacy. This might be credited to the excess magnetite incorporated within the cellulose sample, which resulted in extra hydroxyl radical production. The OH radicals' maximum yield was associated with the optimal nIC-Conditioner dose. The difference in doses that resulted in a reduction in CST efficiency was linked to the amount of OH radicals, which changed as the catalyst dose changed [33,34]. Consequently, the extra catalyst dose acted as an oxidation inhibitor, as it reduced the amount of OH radicals, rather than acting as a generator [35].



**Figure 6.** CST reduction efficiency at various nIC-Conditioning doses.

### 3.2.3. Effect of Hydrogen Peroxide Dose

The oxidizing agent, hydrogen peroxide, played a significant role in the oxidizing test. Alum sludge dewatering experiments using the Fenton-based nIC-Conditioner were conducted while changing the hydrogen peroxide reagent concentration from 50 to 400 mg/L, with all other operating parameters kept constant. As shown in Figure 7, the experimental data revealed that elevating the H<sub>2</sub>O<sub>2</sub> concentration from 50 to 200 mg/L resulted in a CST increment that reached 78%. However, when the amount of reagent was increased to 300 mg/L, the result was a decline in CST efficiency, which indicated a reduction in dewaterability.



**Figure 7.** CST reduction efficiency at various hydrogen peroxide doses.

Hydrogen peroxide was applied as an initiator for the nIC-Conditioner catalyst to generate the oxidation reaction and hydroxyl radicals. These radicals were mainly responsible for oxidizing and conditioning the sludge. However, when the  $\text{H}_2\text{O}_2$  reagent dose was more than 200 mg/L, the result was a reduction in CST reduction efficiency. This might be attributed to triggering hydroxyl (OH) radical generation via extra hydrogen peroxide radicals with hydroxyl radicals rather than producing them. This explanation and these results are in accordance with a previous work [36].

### 3.2.4. Effect of pH Value

As the Fenton reaction is highly influenced by the pH of the medium, the oxidation reaction for alum sludge conditioning based on the modified Fenton skeleton builder/conditioner (nIC-Conditioner) was conducted under varying pH to assess its influence. The pH was varied from the sludge's original pH (6.5) into acidic and alkaline ranges. The alkaline pH range was unfavorable, whereas the acidic pH was better than the alkaline one, and the natural pH (6.5) corresponded with the highest CST reduction. The results displayed in Figure 8 indicate that the highest CST (78%) was reached when the pH was 6.5; however, when the pH was reduced to an acidic level, the CST was 72%. This might have been related to a release of metal ions, especially aluminum and iron, from the alum sludge to promote flocculation. In contrast, the basic medium exhibited a negative effect on the sludge dewatering performance, as deduced from the capillary suction time. The high pH may have reduced the amount of hydroxyl radicals generated, which are the driving force of the oxidation system. However, when the pH was increased to the alkaline range, the CST percentage declined. This might be attributed to the alteration of the sludge particles' surface and characteristics due to the pH's impact on the extracellular substance (nIC-Conditioner) in the alum sludge. Alum sludge molecules might have adsorbed both  $\text{H}^+$  and/or OH ions present in the solution in acidic and alkaline pH ranges. Nevertheless, in both acidic and alkaline circumstances, the alum sludge's colloidal particles might have hydrolyzed and flocs might have formed. However, using the alum sludge without a pH adjustment was more reliable and economical, which facilitated the process [37–39]. This investigation of the sludge's original pH showed that the optimal performance was in accordance with previous data reported in the literature [40].

### 3.2.5. Dual Thermal/Chemical Conditioning

A temperature increase might affect the conditioning process and enhance its de-waterability. Therefore, chemical conditioning based on the nIC-Conditioner/ $\text{H}_2\text{O}_2$  was conducted under changes in temperature, while all other parameters were kept constant (pH 6.5, 200 mg/L  $\text{H}_2\text{O}_2$  and 1 g/L nIC-Conditioner). The temperature was elevated from room temperature to 40, 50, and 60 °C; the experimental data are displayed in Figure 9.

The results demonstrated that temperature elevation resulted in enhanced CST reduction efficiency, which reached 94% when the temperature was elevated to 60 °C. As expected, the thermal effect might have had a high impact on CST reduction due to its pronounced effect on water release and removal. Moreover, the temperature elevation enhanced OH radical production, as it facilitated good contact between the catalyst and the hydrogen peroxide reagent. Additionally, the temperature might have affected the cellulose's performance as a skeleton builder. Hence, the overall conditioning was improved. However, an extreme temperature increase might have the opposite effect, as above a certain limit, the temperature increase could not lead to an increase in the hydroxyl radical yield. In contrast, the result may have been the disappearance of OH radicals. Dewaterability enhancement using the thermal effect was previously reported by researchers treating various types of sludges [33–35].

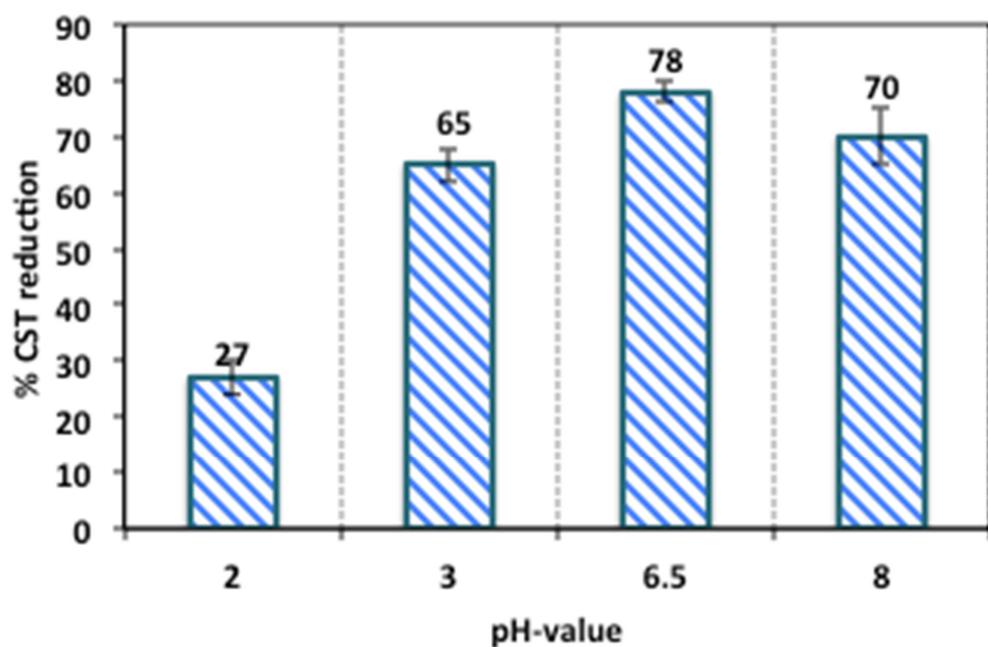


Figure 8. CST reduction efficiency linked to sludge pH using nIC-Conditioner/H<sub>2</sub>O<sub>2</sub>.

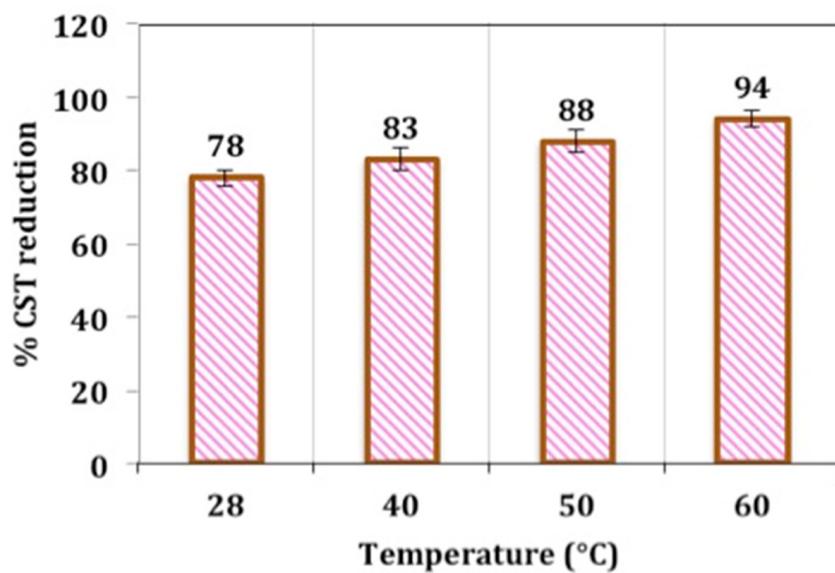


Figure 9. CST reduction efficiency linked to temperature effect using nIC-Conditioner/H<sub>2</sub>O<sub>2</sub>.

### 3.2.6. Comparative Data Analysis

The current study's dewatering results were compared with previously investigated dewatering sequences cited in the literature that used commercially available conditioners. The data in Table 1 summarize previous research regarding the dewatering performances of various alum sludges collected from water treatment plants in different regions. Experiments on alum sludge conditioning with the aim to enhance dewatering capabilities revealed that polymer dewatering displayed higher treatment efficiencies than the use of other available conditioners. However, it is essential to note that the toxicity related to polymers is problematic. Thus, several current research projects are focusing on environmentally benign alternatives.

**Table 1.** Summarized comparison of the current alum sludge conditioning with relevant sludge studies.

Type of Conditioner	Conditioner Amount	Suspended Solids (mg/L)	Sludge pH	Conditioning Time (min)	Dewaterability Reduction (%)	Refs.
Nano-iron-cellulose/H <sub>2</sub> O <sub>2</sub> Fenton	1 g/L	12,043	6.5	5	78%	Current investigation
LT-25 anionic polyelectrolyte	20 mg/L	9300	7.0	1.5	20%	[41]
Dried alum sludge	63 kg/t-dry solids	33,900	6.4	30	76%	[42]
Cationic polymer PC-320	20 mg/L	-	-	1	85%	[43]
Chitosan	-	60,000	7.0	1	88%	[44]
Cationic polyelectrolyte Praestol 650 TR	1.8 kg/t	33,900	6.2	30	96%	[42]
Anionic polyelectrolyte Praestol 2540 TR	1.8 kg/t	33,900	6.2	30	93%	[42]
Cationic polymer PC-325	-	60,000	7.0	1	96%	[44]
Anionic polymer Flohmiser 50	3.85 mg/L	1500	6.3	-	70%	[45]
Anionic surfactant SDS (sodium dodecyl sulphate)	1 mg/L	-	-	60	65%	[46]
Moringa oleifera seed extract	125 kg/t-dry solids	33,900	6.2	30	66%	[42]
P FO-4140 cationic polyelectrolyte	50 mg/L	2985	6.3	1.16	89%	[47]
LT-25 anionic polyelectrolyte	30 mg/L	2985	6.3	1.16	90%	[47]
Fenton oxidation	Fe <sup>2+</sup> 21 mg/g-DS+ H <sub>2</sub> O <sub>2</sub> 105 mg g-dry solids	2850	6.0	1	48%	[20]
LT-25 anionic polyelectrolyte	3.5 mg/g-dry solids	2850	6.0	1	67%	[40]
FO-4140 cationic polyelectrolyte	7.0 mg/g-dry solids	2850	6.0	1	82%	[40]
Solar/Fenton (Fe <sup>2+</sup> /H <sub>2</sub> O <sub>2</sub> )	50 mg Fe/L + 800 mg H <sub>2</sub> O <sub>2</sub> /L	2364	8.5	7	78%	[48]
Dual solar/LT-25 anionic polyelectrolyte	10 mg/L	2364	8.5	3	97%	[48]
Cationic polyacrylamide CPAM1	8 mg/L	-	7.0	2.5	95%	[49]
Cationic polyacrylamide CPAM2	16 mg/L	-	7.0	2.5	97%	[49]
Fenton-like oxidation (Cu <sup>2+</sup> /H <sub>2</sub> O <sub>2</sub> )	Cu <sup>2+</sup> 20 mg/g-dry solids + 125 H <sub>2</sub> O <sub>2</sub> mg/g-dry solids	2850	6.0	1	7%	[32]
Zetag-89 polymer	3 kg/t-dry solids	-	7.0	-	20%	[50]
Praestol 650 TR polymer	1.8 kg/t-dry solids	33,900	6–10	30	96%	[42]
Gypsum/PW 85 polymer	20 mg/L	8453	7.0	60	16%	[23]

Filterability performance is compared via the filtration capability and water release. The comparative results in Table 1 illustrate the significance of different materials, including a variety of conditioners, such as the Fenton oxidation reaction, skeleton builders, and polyelectrolytes, which are widely applied as commercial conditioners. Conditioning behaviors are associated with time periods. Using different conditioners changed the dewatering time. As there are strict limits on toxic material use in drinking water treatment

plants to avoid environmental damage, using greener technologies, even if they are less efficient, is a must. Moreover, the environmental impacts and potential cost of polymers stand against their use. Furthermore, the use of sawdust/magnetite technology improves the overall sustainability of the sludge treatment system, as it reduces the chemicals used and thereby reduces the associated costs and safety hazards. Consequently, the proposed method presents a cost-effective and environmentally friendly opportunity for sludge conditioning and dewatering. Notably, the monomers used in polymer production are more toxic than the polymer itself; nonetheless, extreme levels of toxicity are sustained, especially with regard to acrylamide. In this regard, rigorous regulations guiding polymer use and polymer toxicity would not seem to be significant problems associated with the proposed method.

Furthermore, it is well known that pronounced dewatering is achieved when anionic and cationic polyelectrolytes are applied; however, the cationic polyelectrolyte showed a pronounced dewaterability that reached 97%. Moreover, dual conditioning improved the performance. However, dewaterability reached only 78% in the current study. Moringa oleifera seed extract achieved 65% dewatering.

Notably, the material used in the current study is environmentally benign, as it comprises magnetite as a conditioner augmented with wood waste powder, which is a green alternative conditioner. The Fenton system is an eco-friendly option compared to the toxic polymer-based conditioners used in polyelectrolyte conditioning.

Compared with polymer conditioning, the cost of the modified Fenton's oxidation system based on waste sawdust is low. Notably, there might be a cost increase associated with Fenton's reagent due to its dual-reagent composition. Such cost increases may be associated with equipment installation, operation, and control processes. However, even with increased costs, Fenton's reagent has the potential advantage of eliminating the perceived long-term risk associated with residual polymers in the environment.

Magnetite nanoparticles' superior characteristics make them a suitable candidate for numerous applications, including environmental and medical applications. For example, they are used in wastewater treatment, electronics, drug delivery, cosmetics, and magnetic resonance imaging. Furthermore, the toxicity of magnetic nanoparticles is not well studied. In some applications, they are considered nontoxic. Notably, a limited amount of magnetite nanoparticles is used in the proposed composite. Numerous studies report that the toxicity potential of magnetite nanoparticles is low or non-existent until high exposure levels less than 100 mg/mL [44].

Notably, after dewatering, the sludge was suitable for numerous applications, as mentioned in previous sections. Therefore, using sawdust to dewater alum sludge could decrease the sludge's compressibility, release and reduce the bound water content, and improve the sludge cake's porosity. Thus, using a cellulosic sawdust substance for dewatering might enhance the sludge's calorific value; no energy input is required when sludge dewatered using this technique is disposed of via incineration. Moreover, although higher efficiency might be attained using other techniques, the current conditioning system improves the sludge's calorific value [51].

Sludge treatment and disposal in real-world applications and scalability suggestions for any particular waterworks plant location may initially require a reduction in the volume of generated alum sludge in order to compact this sludge so that it has a higher solids content [23]. Subsequently, chemical conditioning, such as the suggested nIC-Conditioner, would be applied using an oxidation reaction. In such a dewatering and drying step, the water would be removed and the sludge's volume reduced. Finally, depending on the sludge's treatment rate, its impact on the environment and legal disposal would be legitimate concerns. Thus, providing an appropriate reuse facility is essential. Priority disposal is required after chemical conditioning, which might be conducted through a range of processing options (including gravity, flotation, centrifuge, or elutriation via heat treatment) and requires consideration. The principal design criterion is solids loading expressed as the number of kilograms of solids applied per square meter of bottom area

per day ( $\text{kg}/\text{m}^2 \cdot \text{d}$ ). Thus, the chemical addition is associated with the type of sludge in a particular plant and with the sludge's dry solid content.

#### 4. Conclusions

A dual Fenton's reagent/skeleton builder conditioner based on wood chip powder augmented with magnetite as the catalyst source, initiated with a hydrogen peroxide reagent, was applied as a dual chemical conditioner and skeleton builder to alum sludge to improve its dewaterability. The optimal operating parameters were 1 g/L nIC-Conditioner, 200 mg/L  $\text{H}_2\text{O}_2$ , and a pH of 6.5, which corresponded to the maximum 78% dewaterability. Additionally, the demonstrated results were verified in the morphology change after conditioning, which was enhanced by an increase in the flocculation affinity. Moreover, the Fenton/skeleton builder conditioner and commercial conditioners, such as anionic LT-25 and cationic FO-4140 polyelectrolytes, were compared for verification purposes. The wood chip powder augmented with magnetite was found to be an ideal and environmentally benign alternative to the toxic commercial conditioners. Notably, further research is essential to explore and expand the real-world applications for dewatered sludge cakes. Dewatered alum sludge could be introduced as an adsorbent material, fuel source, or building material, and used in wetland applications. However, extra data are essential prior to these applications due to the anxiety associated with public beliefs regarding releasing alum sludge into the environment, as concerns associated with its release are still under consideration.

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