

## Characterization and utilization of water treatment sludge for coagulation of raw water

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### ABSTRACT

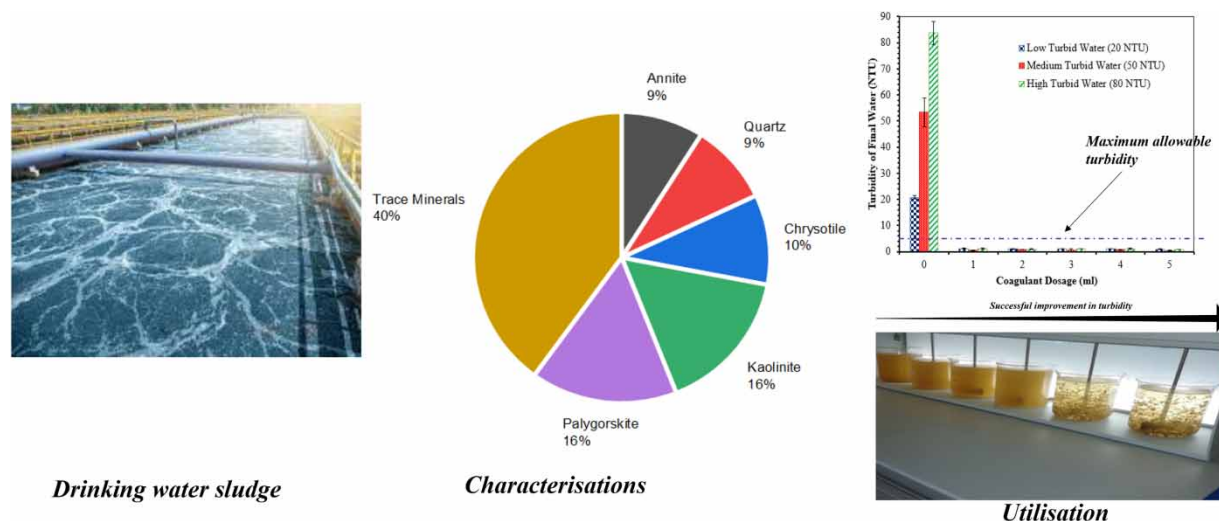
One of the critical steps in the treatment of raw water for human consumption is clarification. It encompasses coagulation and flocculation to reduce the concentration of suspended solids in turn producing a semi-solid slurry called water treatment sludge (WTS). It is necessary to characterize and find economic and sustainable ways to utilize the WTS. The WTS used in this work was characterized and contained aluminum (Al) (20.3%  $\text{Al}_2\text{O}_3$ ) and iron (Fe) (5.28%  $\text{Fe}_2\text{O}_3$ ) making it suitable for the development of a coagulant. A less energy-intensive way, through atmospheric acid leaching of WTS, was developed to extract Al and Fe for use as a coagulant. The sulphuric acid concentration and solid loading that gave the highest Al concentration were determined to be 3N and 5 g/100 mL, respectively. Turbidity reduction achieved with the developed coagulant ranged from 91 to 99% and was higher than that obtained in related studies due to the higher concentrations of Al and Fe in the WTS used in this study. Since the coagulant was very acidic, it was co-dosed with calcium oxide at 10 g/L of water to adjust the pH of the final water to allowable pH values for potable water (6.5–9.5).

**Key words:** acid treatment, alumina coagulant, coagulation, leaching, water treatment sludge

### HIGHLIGHTS

- Water-treated sludge (WTS) can be leached to produce a coagulant.
- WTS is rich in aluminum which is essential for coagulation.
- Acid leaching effectively extracted 35.7 mg/g Al and 4.5 mg/g Fe from WTS.
- > 90% turbidity reduction was achieved for low, medium and high turbid water.
- WTS can be used in other applications based on its Al, Fe and Si content.

### GRAPHICAL ABSTRACT



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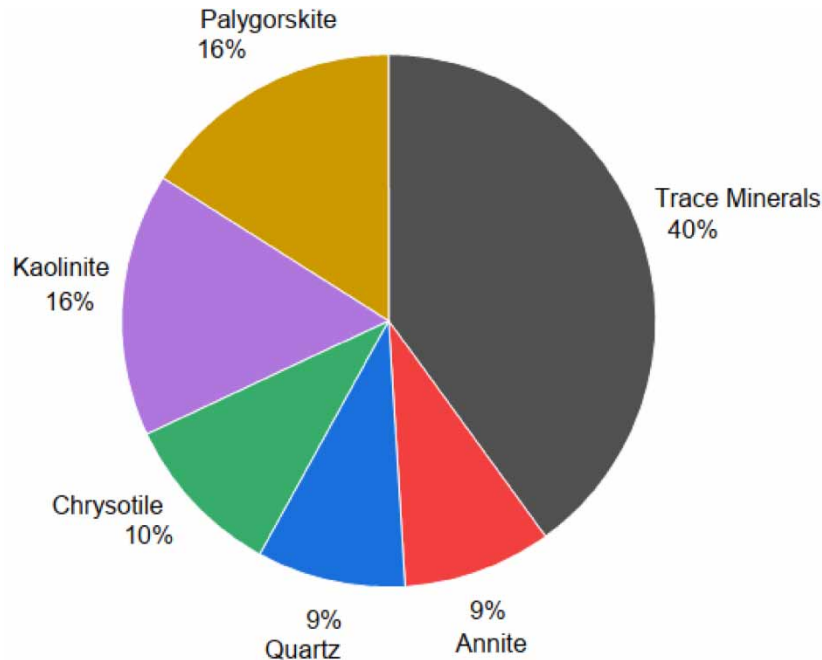
## 1. INTRODUCTION

Coagulation is a chemical process in the treatment of raw water to make it safe for human and animal consumption and it involves the use of alumina and ferrous oxides, sulphates, chlorides and polymers like poly-silicate iron and poly-aluminum chloride (Aktas *et al.* 2013). The rate at which these chemicals are being used is proportional to the demand for clean water and since the world's population has been increasing over the past years, there has been a tremendous increase in demand for quality drinking water. As a result, it is predicted that by the year 2050, about six billion people could lack access to clean potable water (Gosling & Arnell 2016). To counteract this pressing need, water treatment plants have opted for the large-scale usage of coagulants to meet the increasing demand. Coagulants are used during the clarification step of the water treatment process for the removal of suspended particles from raw water, so as to reduce the turbidity. The desired clear supernatant water is accompanied by water treatment sludge (WTS), an inevitable by-product precipitated during the clarification process, which settles as solids at the bottom of clarifiers. WTS is mainly made up of metallic oxides, minerals and humic matter as well as clay/sand with coagulant residuals (Babatunde & Zhao 2007).

A typical water treatment plant like the Palapye Water Treatment Works (PWTW) (Botswana) which treats about 4,200 m<sup>3</sup>/day discards about 100,000 tons per year of sludge, that is, 0.065 tons of sludge/m<sup>3</sup> of raw water, while on a global scale, more than 10,000 tons of sludge is produced daily (Lukasiewicz 2016). The amount of sludge generated at the PWTW is comparable to the 100,000 tons/year of sludge generated in a typical wastewater treatment plant (WWTP) (Ahmad *et al.* 2016). Generally, the sludge produced is made up of about 60% fine sand/silt particles along aluminum (Al) oxides, iron (Fe) oxides, and silica, as well as some heavy metals and trace elements (Ahmad *et al.* 2016). The management of sludge is a serious concern as most water treatment plants across the world dispose of sludge into water bodies and drainage channels whereas in Botswana, untreated sludge is mainly disposed of in dug structures called sludge lagoons. This WTS can leach hazardous components into rivers, streams, ponds, lakes and drains and lead to eutrophication in water bodies and contamination of water sources (Ahmad *et al.* 2016). With the rise in the need for environmentally friendly economic practices as well as laws governing residual effluents, it is, therefore, necessary to come up with sustainable sludge management strategies.

The current drive to achieve sustainable development requires that product processing plants implement the concepts of the circular economy in their processes and this approach has gained traction within the water treatment industry. It is, therefore, necessary to study the characteristics of the waste sludge and find good utilization alternatives to fulfill the desire to turn water treatment plants into water resource recovery facilities (WRRFs). This will enable such facilities to deliver clean water, renewable energy, agricultural nutrients like phosphorus and other valuable bio-based materials from raw water. Given that WTS is one of the major waste products from water treatment plants, developing waste valorization routes for it will ensure the sustainable operation of water treatment plants. Based on past research, for instance, Tarique *et al.* (2016) concluded that a 1% concentration of WTS treated with 0.05 mL/mL sludge of 2.5 N (normality) of sulphuric acid behaved as a good coagulant at pH range 6–8 with a dosage of 8 mL/L. However, Keeley *et al.* (2014) studied the use of Donnan dialysis membrane (without using chemical substances like acids) for the selective recovery of 70% Al from WTS for use as a coagulant. There are other several coagulant recovery techniques like alkaline treatment which is carried out at pH range of 11.4–11.8 for NaOH and 11.2–11.6 for Ca(OH)<sub>2</sub> (Masschelein *et al.* 1985). Keeley *et al.* (2014) stated that WTS with high Fe content can be used directly as a coagulant in vegetable refinery wastewater with the ability to remove oil and grease, with high yields achieved when coupled with Al-rich sludge while Ishikawa *et al.* (2007) concluded coagulant recovered from sludge using sulphuric acid can be used for the treatment of domestic wastewater, as well as wastewater from food industries. Acid leaching was chosen over alkaline leaching because previous studies showed that acids are more effective in the extraction of Al compared to bases (Kyriakogona *et al.* 2017). Chigondo *et al.* (2015) also stated that sulphuric acid can be used for the large-scale extraction of alum from kaolin clay. Therefore, considering that the WTS used in this study is rich in Kaolinite (Figure 1), a principal mineral in kaolin clay, it was best to leach with sulphuric acid. In addition to the limited information on the extraction of Al from kaolinite-rich WTS, the key novelty is developing a less energy-intensive hydrometallurgical process that is easily adaptable to WWTP's to recover and re-use coagulants from WTS.

An atmospheric hydrometallurgical process using agitation leaching is less energy intensive, easier to operate and requires low capital investment compared to both pressurized hydrometallurgical processes and pyro metallurgical processes. This makes it more suitable for small-scale operations. In addition, the WTS used in this study contains remnants of a polymer blend of Zetafloc 553 L. This type of WTS has not been extensively studied. In summation, this study was aimed at



**Figure 1** | Quantitative results of dry WTS XRD analysis.

characterizing and utilizing a Zetafloc 553 L containing WTS from a local treatment plant, the PWTW in the Central District of Botswana as a coagulant. The study goes on to determine the physical and chemical characteristics of the sludge. Moreover, the identification of a more suitable less energy-intensive technique to treat the sludge and to use the treated sludge for coagulation of raw water was the key motivation for the study.

## 2. MATERIALS AND METHODS

### 2.1. Collection and preparation of WTS samples

Samples of the WTS were collected from open sludge lagoons at the PWTW, Botswana. The dry samples were collected by grab sampling and were put in tight plastic bags and the wet WTS was collected from both the sludge lagoons and recovery ponds and put in a closed polyvinylchloride (PVC) bucket. Both samples were dried at about 105–110 °C to remove excess water. The samples were then pulverized at 300 rpm for 3 min to ready them for characterization and the leaching process.

### 2.2. WTS characterization

A Zetium X-ray Fluorescence (XRF) PW5400 3 kw machine was used for chemical analysis, specifically elemental oxides while PANalytical X'Pert Pro ([XRD5]) X-Ray Diffraction (XRD) was used to study the mineralogy of WTS. Vertex 70 v vacuum Fourier transform infrared (FTIR)-Spectrometer gave the identity of functional groups which can be related to various compounds in the WTS, within the wavenumber ranges of 4,000 to 400  $\text{cm}^{-1}$ . The amount of Al and Fe in the leachate (coagulant) was measured using flame ContrAA-700 Atomic Absorption Spectroscopy (AAS).

### 2.3. Leaching process (production of coagulant)

From Table 1 and Figure 1, dry WTS proved to have high Al and Fe content and was therefore used for the leaching process. 98% analytical grade sulphuric acid ( $\text{H}_2\text{SO}_4$ ) supplied by Rochelle Chemicals was diluted with distilled water to produce the desired normalities (1.0, 2.0, 2.5 and 3.0 N). Solid loading was varied from 3 g WTS/100 mL of leaching agent, 5 g WTS/100 mL of leaching agent to 10 g WTS/100 mL of leaching agent.

To test the efficiency of the leaching agent and acid concentration in recovering Al and Fe from the WTS, the leaching process was carried out at a constant solid loading for all three different normalities. The leaching process was carried out in 1,000 mL beakers using a VELP Scientifica JLT6 Flocculation tester for agitation purposes. The mixing speed was set at 50 rpm to achieve homogeneity during the leaching process and the extraction was carried out for 60 min. Starting from

**Table 1** | Elemental oxides compositions for wet and dry WTS

Sample type	Na <sub>2</sub> O (%)	K <sub>2</sub> O (%)	MgO (%)	CaO (%)	TiO <sub>2</sub> (%)	MnO (%)	Fe <sub>2</sub> O <sub>3</sub> (%)	Al <sub>2</sub> O <sub>3</sub> (%)	SiO <sub>2</sub> (%)	P <sub>2</sub> O <sub>5</sub> (%)
Dry WTS	0.153	0.817	0.743	0.849	0.460	1.06	5.28	20.3	31.9	0.439
Wet WTS	0.171	0.604	0.592	1.69	0.338	1.34	3.65	16.5	31.3	0.503

the initial stage of extraction, the pH of the mixture was measured every 10 min until the end of the experiment. The process was carried out at room temperature and pressure and after the specified hour of extraction, the unreacted solids were allowed to settle before the filtration stage was conducted. The experiments were repeated twice.

The classical approach was used in this instance because the numbers of factors considered for leaching were mainly two that is acid concentration (studied at four levels) and solid loading (studied at three levels). The other factors like temperature and agitation were kept constant. Atmospheric leaching at room temperature was preferred in order to make the process less energy intensive and easier to implement at WWTP's. A minimum agitation speed of just enough to suspend the sludge particles was also used to conserve energy and reduce costs. It is known for leaching operations that solid loadings beyond 10% present operational challenges in pumping and mixing as the solution becomes saturated (Shiba & Ntuli 2017), thus there was no need to go beyond this value. Based on this and the fact that the classical approach is most suitable for three factors or less it was chosen over the design of experiments (DOE) approach. Using the classical approach, the number of trials required to get reliable data is  $N = k(p - 1) + 1$  for  $k$  factors and  $p$  levels is 7. Thus, a reasonable number of experiments could be conducted without compromising the quality of the data. However, it is also important to highlight that the classical approach has its own drawbacks, e.g. inability to reveal interaction effects.

#### 2.4. Al and Fe content analysis

The filtered extract was analyzed for the composition of Al and Fe before it was used for coagulation using AAS. The samples were further filtered with a 0.45 µm sterile syringe filter to avoid clogging of AAS liquid transfer tubes. Since the samples were too acidic for the AAS, the samples were diluted by a factor of 10 either with distilled water for Fe or nitric acid for Al depending on the element tested and the type of flame used.

For Fe analysis, five Fe concentrations were prepared: 0.5, 1.0, 2.0, 5.0 and 10 ppm along with three blanks of 2.0, 2.5 and 3.0 N of the leaching agent and distilled water. These standards were used for calibration purposes and leachate samples were tested with acetylene flame in the AAS and results were recorded in mg/L along with the absorbance.

Al analysis also required five Al concentrations; 5.0, 10, 15, 20 and 25 ppm. The standards were of higher concentration compared to those used for Fe because of the expected high Al concentrations in leachate samples since the coagulant used in the treatment plant was alumina based. For Al analysis, the leachate samples were diluted by a factor of 10 using 2.0% nitric acid since nitrous oxide flame was used.

#### 2.5. Coagulation: jar tests

##### 2.5.1. Preparation of synthetic turbid water

The turbid water used for coagulation was prepared from Bentonite clay supplied by Nature's Choice Wholefood Specialists. 0.01 g/mL stock solution was prepared and stirred at a speed of 200 rpm for 60 min, the solution was then left to settle for about 24 h. This was also meant to make sure that the Bentonite particles are completely hydrated before using the solution for the jar tests.

##### 2.5.2. Jar test experiments

The jar test experiment was used to analyze the efficiency and percentage removal of turbidity achieved by the leachate (coagulant). Synthetic turbid water samples were classified into low (20 NTU), medium (50 NTU) and high (80 NTU) to study the effectiveness of the coagulant at different levels of turbidity. Six 1,000 mL beakers were filled up to the mark with the 20 NTU turbidity water prepared by adding 10 mL of the 0.01 g/mL stock solution in 1,000 mL of water and assembled on the bench-top VELP Scientifica JLT6 Flocculation tester. The first beaker acted as a reference point and therefore was not dozed with the leachate coagulant while the remaining five were dozed starting with 1.0 mL to 5.0 mL with increments of 1.0 mL.

The agitation started with fast mixing at 230 rpm for 2 min followed by slow mixing at 40 rpm for 15 min to facilitate the formation of flocs. Stirrers were finally switched off to allow the settling of flocs at a settling time of 30 min. This was also done for 50 NTU (25 mL of stock solution in 1,000 mL of water) and 80 NTU turbid (40 mL of stock solution in 1,000 mL of water). The same procedure was carried out in natural raw water collected from the Lotsane river in Palapye, Botswana, with an initial turbidity of 381 NTU. The sample was collected during the peak rainy season when water turbidity levels are normally extremely elevated. This was to enable the developed WTS coagulant to be tested under extreme conditions.

### 2.5.3. Turbidity and pH analysis

The initial turbidity of the synthetic turbid water was measured using Hanna HI-93703 portable microprocessor turbidity meter while the pH was measured using pH 700 Bench-top meter from OAKTON Instruments. After the settling time (30 min), the turbidity of the supernatant water was measured and the pH of the supernatant water was also measured to check if it falls within the World Health Organization (WHO) standard value of 6.5–9.5 and also to analyze how the leachate coagulant affects pH of raw water.

### 2.6. Supernatant water pH adjustment

The coagulant used was very acidic because sulphuric acid was used as the leaching agent therefore the pH of the supernatant was below the recommended 6.5–9.5. To adjust this pH, lime (calcium oxide) solution was used to increase the pH to the recommended standard. The pH of lime was 12 at a concentration of 10 g/L of water.

## 3. RESULTS AND DISCUSSION

### 3.1. Characterization of WTS

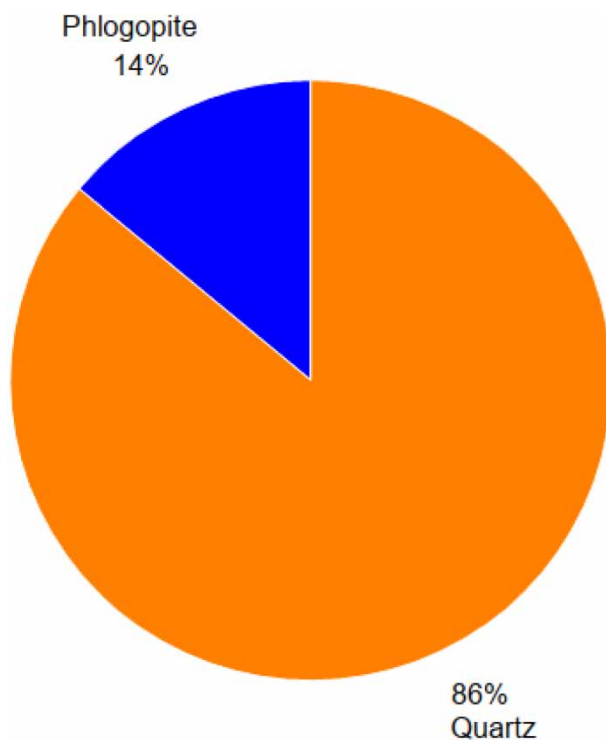
#### 3.1.1. Mineralogy of WTS (XRD)

Among the main minerals characterized in the dry WTS was Palygorskite at 16%, a mineral made up of Al, Mg, O and Si while Kaolinite (Al, O, Si) also made up to 16% of the dry WTS. Chrysotile (Mg, O, Si) and Annite (Al, Fe, K, O, Si) contributed 10 and 9%, respectively, to the mineralogy of dry WTS whereas quartz (Si, O) was at 9% (Figure 1). The remaining percentage was made up of trace minerals of Fe, Al, calcium, magnesium and silica. It is this high Al and Fe content (Ahmad *et al.* 2016) that makes coagulation a possible alternative utilization route for WTS. Mineralogy of wet WTS (Figure 2) composed of high quartz content (86%) and Phlogopite (Al, Ba, F, Fe, Mg, Na, Si) at 14% which, when compared to dry WTS, shows very few mineral contents. This might be because dry WTS had well-detectable minerals among its amorphous structure (Nayeri & Mousavi 2020) than wet WTS as shown in Figure 3.

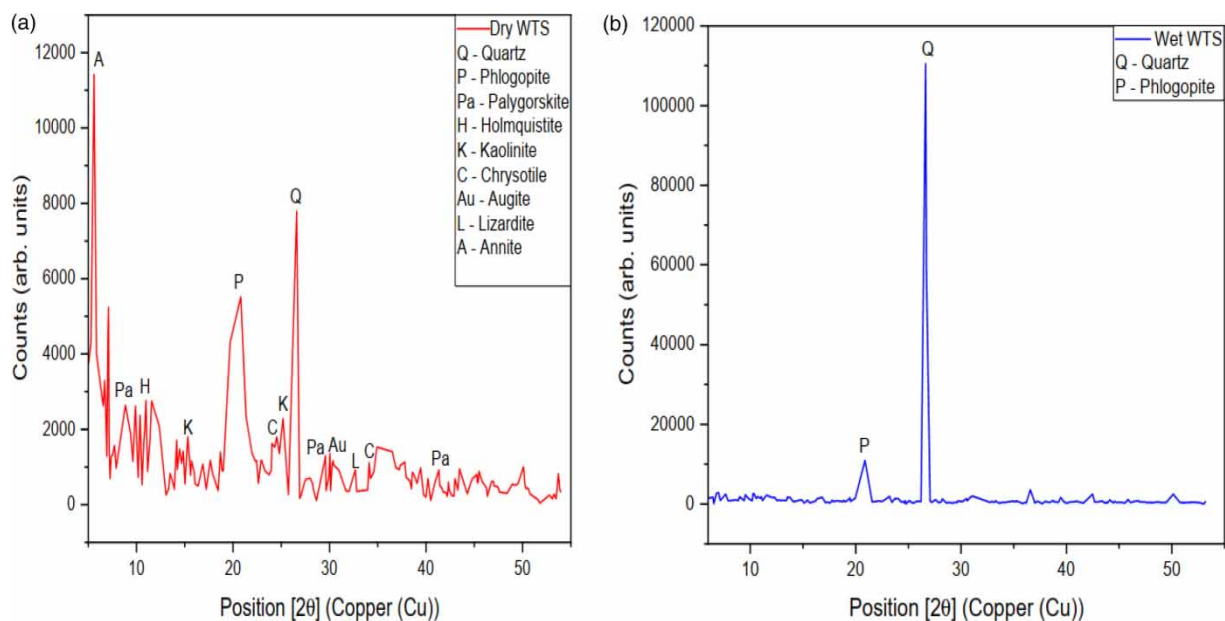
#### 3.1.2. Oxides composition analysis (XRF)

The two sludge samples have the same content of oxides with varying percentage compositions (Table 1). The main metallic oxides, as also observed elsewhere by Tarique *et al.* (2016), include  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$  which are essential for coagulation as most coagulants are either Al or Fe based. The dry WTS contains up to 5.28%  $\text{Fe}_2\text{O}_3$  and 20.3%  $\text{Al}_2\text{O}_3$  while wet WTS has 3.65 and 16.5%  $\text{Fe}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3$ , respectively. Al oxide composition is higher than iron oxide from additional Al in alumina-based coagulant used at the water treatment plant where the sludge was collected. Dry WTS has a higher Al and Fe content than wet WTS while the silica content ( $\text{SiO}_2$ ) content is similar with 31.9% for dry and 31.3% for wet WTS. The remaining percentage is made up of trace elements, almost equal in both dry and wet WTS. These XRF results are in consonance with the analysis made by Ramirez *et al.* (2018) in river water sludge that the WTS had higher concentrations of aluminum oxide ( $\text{Al}_2\text{O}_3$ ), silicon dioxide ( $\text{SiO}_2$ ) and iron oxide ( $\text{Fe}_2\text{O}_3$ ). These oxides account for about 69.9% of the total chemical composition of wet WTS and 92.6% of calcined WTS. Wołowiec *et al.* (2019) also undertook XRF analysis on groundwater treatment sludge (GWTS) and found that iron oxides are the predominant chemical components (over 43%) with Silica and Manganese oxides at (7.2%) and (3.11%), respectively. The sludge had calcium oxide (3.93%) and phosphorus oxide (4.02%). The difference between this groundwater sludge and river water sludge, apart from the fact that it was treated with iron (III) sulfate (IV) coagulant is that groundwater tends to have high concentrations of health-giving minerals such as calcium, magnesium and Fe because of interactions with several underground rocks and minerals from which this water is transmitted or contained (Nwankwo *et al.* 2020).





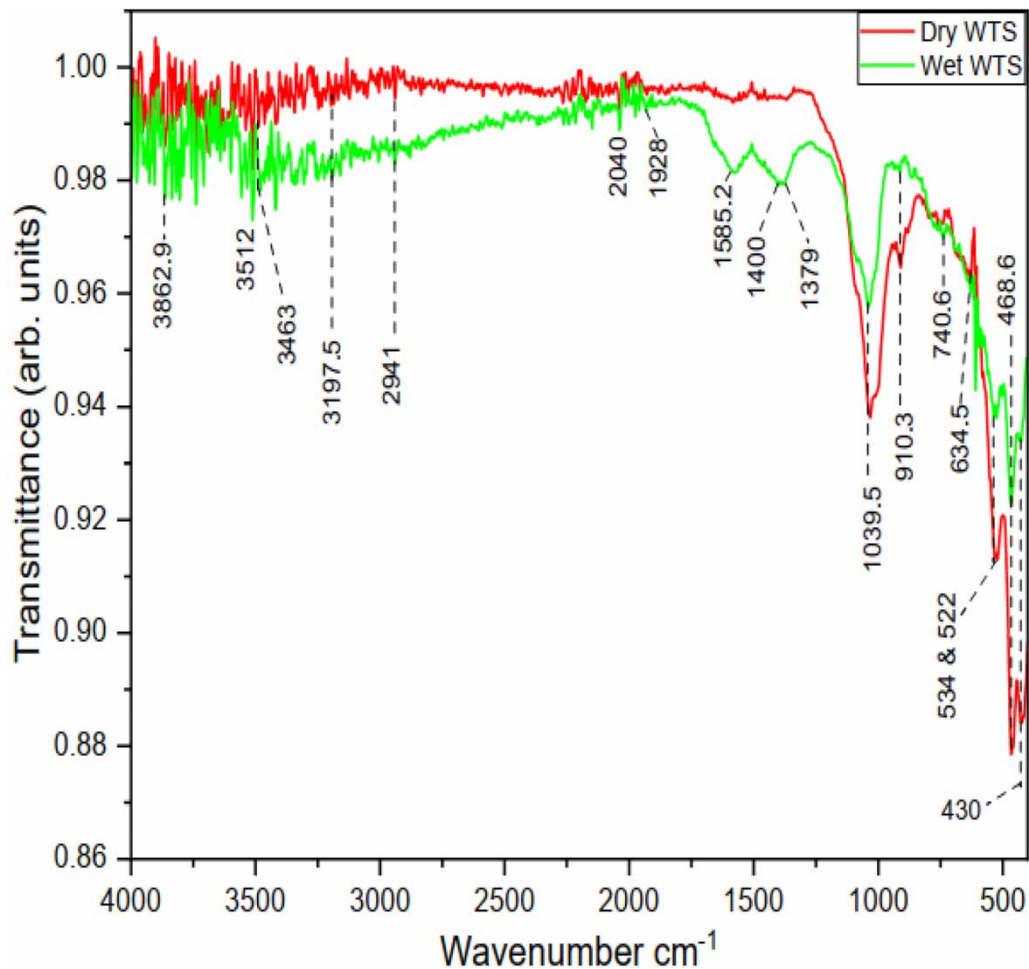
**Figure 2** | Quantitative results of wet WTS XRD analysis.



**Figure 3** | XRD Patterns for (a) dry and (b) wet WTS.

### 3.1.3. Functional groups of WTS (FTIR)

The FTIR spectrum shows that on the wavenumber region of  $4,000$  to  $400\text{ cm}^{-1}$ , both dry and wet WTS have almost similar peaks hence similar functional groups (Figure 4). The bands at  $3,862.9$ ,  $3,512$  and  $3,197.5\text{ cm}^{-1}$  are in the region of the anti-symmetric and symmetric O–H stretch mode (Tarique *et al.* 2016) while tensile O–H vibrations can be found at  $3,463\text{ cm}^{-1}$



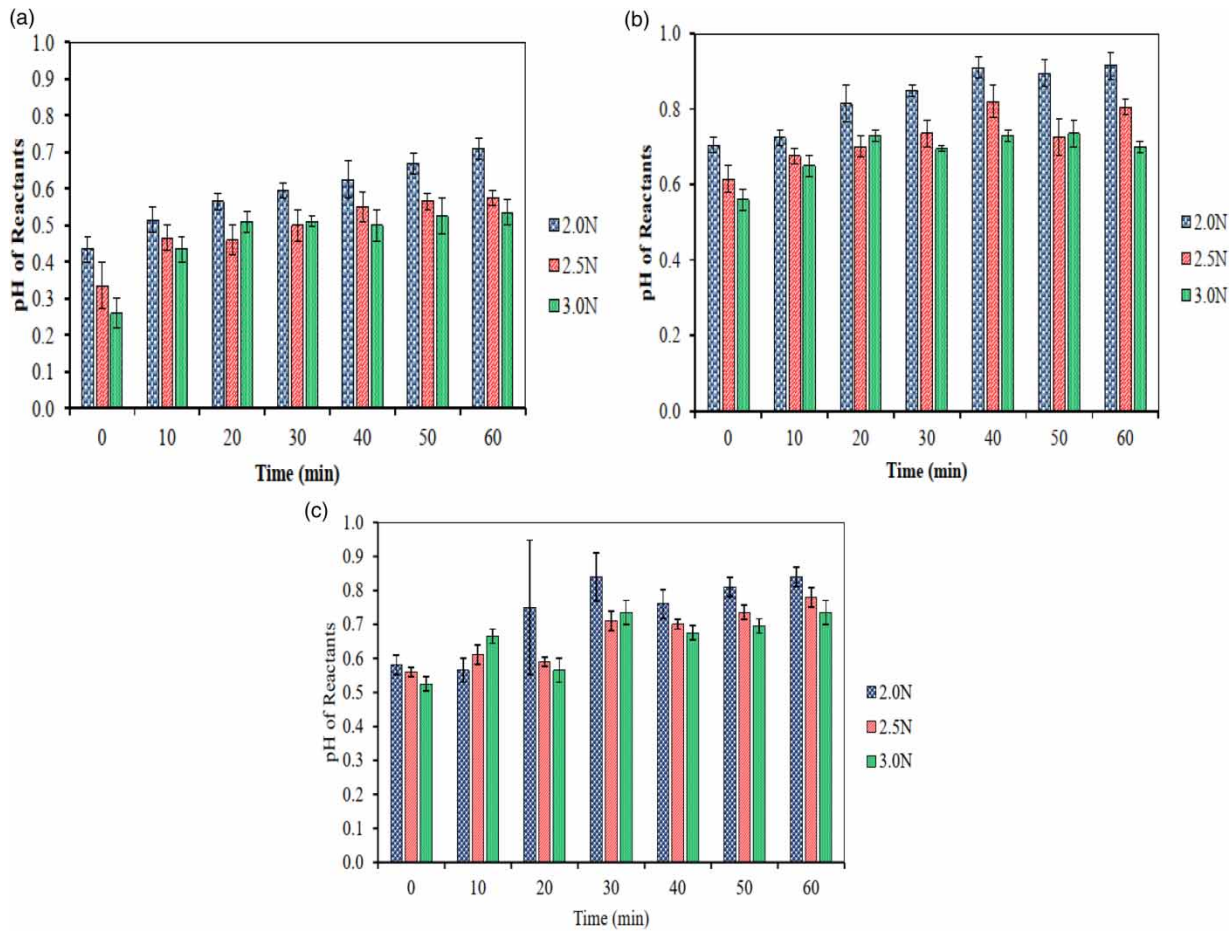
**Figure 4** | FTIR spectrum for dry and wet WTS.

(Nayeri & Mousavi 2020). This band can be related to O–H stretching of  $\text{Al}(\text{OH})_3$  (kaolinite and illite) while the small peak at  $3,197.5 \text{ cm}^{-1}$  is a representation of intermolecular hydrogen bonding associated with  $\text{Al}(\text{OH})_3$  (Fathima *et al.* 2013). Based on analysis from Fathima *et al.* (2013), the band at  $2,941 \text{ cm}^{-1}$  is due to the aromatic C–H stretching. The bands  $2,040$  and  $1,928 \text{ cm}^{-1}$  can be attributed to H–O–H (Liu *et al.* 2012).  $1,029.5 \text{ cm}^{-1}$  band is in the region of Al–O bands (Lekgoba *et al.* 2021) while the range from  $1,092$  to  $782 \text{ cm}^{-1}$  is related to stretching vibrations of Si–O–Si associated with quartz and from the FTIR analysis of WTS, that stretching vibration is shown at  $910.3 \text{ cm}^{-1}$ . The band at  $740.6 \text{ cm}^{-1}$  can be associated with Al–O of  $\gamma\text{-AlOOH}$  of boehmite mineral (Liu *et al.* 2012). Fe–O band is represented by  $634.5 \text{ cm}^{-1}$  which is associated with  $\gamma\text{-Fe}_2\text{O}_3$  while the bands at  $468.6$  and  $430 \text{ cm}^{-1}$  represent the bending vibrations of O–Si–O (Tarique *et al.* 2016).  $468.6 \text{ cm}^{-1}$  relates to the stretch of Si–O–Fe. Fe–O of  $\text{Fe}_2\text{O}_3$  can also be found at  $534$  and  $522 \text{ cm}^{-1}$  and the last peak at  $430 \text{ cm}^{-1}$  may be attributed to a mixed vibration of Si–O in quartz (Vijayaragavan *et al.* 2013).

## 3.2. Leaching process

### 3.2.1. Changes in pH during leaching

The changes in pH were monitored throughout the leaching process for all the solid loading ratios as shown in Figure 5(a)–5(c). Before adding  $\text{H}_2\text{SO}_4$ , the pH of water and WTS was between 6.9 for a lower solid loading (3 g WTS/100 mL leaching agent) and 7.02 for a higher solid loading (10 g WTS/100 mL leaching agent) from which it can be said that the WTS was neutral. Upon addition of the acidic leaching agent, the pH dropped instantly to a range of 0.27–0.68 as the acid leaching treatment process decreases the pH of samples due to the acidic strength of sulphuric acid. Reactants with lower WTS loading



**Figure 5** | pH of reactants against time for various solid loading; (a) 3 g/100 mL leaching agent, (b) 5 g/100 mL leaching agent and (c) 10 g/100 mL leaching agent.

had a lower pH compared to those with high WTS loading since an increase in WTS concentration in water due to higher contents of alkaline constituents dissolved namely, CaO and MgO, which slightly increases the pH. The pH increased after the leaching process started as the acidic ions reacted with the metal and non-metal ions present in WTS. Since the pH during acid leaching should increase until it reaches stability based on leaching duration (Chu & Ko 2018), pH of reactants stabilized around 0.64–0.91 after 60 min of leaching. As a result, the pH of the resultant leachate was acidic.

### 3.2.2. Al and Fe concentration analysis of leachate

Elemental composition of the extracted leachate was analyzed to deduce the concentration of Al and Fe which are necessary for the coagulation process. Table 2 shows concentrations of Al and Fe of filtered leachate in mg/L measured by AAS. Al

**Table 2** | Al and Fe concentrations in leachate

Solid loading (g/100 mL H <sub>2</sub> SO <sub>4</sub> )	Al Conc. (mg/g of WTS)			Fe Conc. (mg/g of WTS)		
	3	5	10	3	5	10
H <sub>2</sub> SO <sub>4</sub> Conc. (N)						
<b>2</b>	34.0 (1,019)	26.8 (1,339)	14.0 (1,402)	5.0 (148.7)	3.9 (194.4)	3.1 (312.6)
<b>2.5</b>	35.5 (1,066)	28.1 (1,404)	9.7 (971)	5.4 (161.0)	3.8 (189.1)	3.2 (316.9)
<b>3</b>	35.4 (1,063)	<b>35.7 (1,785)</b>	12.2 (1,215)	4.7 (142.4)	<b>4.5 (224.8)</b>	3.1 (314.3)

Note: Actual solution concentrations (mg/L) are indicated in brackets. Figures in bold represent the aluminum and iron concentrations of the chosen coagulant.



concentration was higher in the range of 9.7–35.7 mg/g of WTS compared to 3.1–5.4 mg/g of WTS for Fe. As aforementioned, this is because the sludge used was collected from a processing plant that uses an Al-based coagulant which will inherently increase the Al content of the sludge. Since a high concentration of reactants increases the rate of a chemical reaction (Sodiqovna 2020), high Al concentration in WTS resulted in its high rate of extraction compared to Fe which was at a lower concentration.

The highest value of extracted Al was 35.7 mg/g of WTS at a solid loading of 5 g WTS/100 mL of H<sub>2</sub>SO<sub>4</sub> and 3.0 N. 3.0 N gave an extraction pH range of 0.56–0.77 for the 5 g/100 mL solid loading. This is, therefore, the optimum extraction condition for Al because the concentration of leached Al as a function of pH follows an amphoteric leaching pattern where high leachate concentrations can be achieved at lower pH ranges (Cui *et al.* 2019). The pH levels of 5 g/100 mL leaching agent were similar to those of 10 g/100 mL leaching agent but the leachate concentration of the latter is lower because even though high concentration increases extraction efficiency, high solid loading can result in the production of diffusion-resistant sulphate precipitates on the surface and pores which results in a self-inhibition effect on mass transfer at the leaching sites (Seidel & Zimmels 1998). Fe leaching closely follows the cationic leaching pattern where the concentration of leached Fe decreases with an increase in pH (Cui *et al.* 2019) and therefore high concentrations of Fe were obtained at lower pH too, in the range of 0.58–0.59 at 2.5 N of sulphuric acid. The best combination of concentrations was picked based on the idea that the coagulant to be produced is alumina based. Therefore, a combination of high Al concentration of 35.7 mg/g of WTS and 4.5 mg/g of WTS Fe was chosen which is obtained at 3.0 N acid concentration and a solid loading of 5 g WTS/100 mL leaching agent. This condition also gave the highest solution concentrations of Al and Fe, 1,785 and 224.8 mg/L, respectively, compared to 1,063 mg/L Al and 142.4 mg/L Fe obtained by using 3 N acid concentration and solid loading of 3 g WTS (Table 2). Thus, the leachate coagulant obtained using 3.0 N acid concentration and a solid loading of 5 g WTS/100 mL was used for coagulation tests. The removal of the dominant heavy metals (Al and Fe) also generates a sludge with very low concentrations of heavy metals making it suitable for other applications from an environmental point of view.

### 3.3. Coagulation (jar tests)

#### 3.3.1. Synthetic raw water

The extracted leachate hereby termed coagulant was filtered and used in jar tests to analyze its effectiveness in turbidity reduction. Three different synthetic water samples were used; low turbid water at 20 NTU, medium turbid water at 50 NTU and high turbid water at 80 NTU as shown in Figure 6. For low turbid water, it takes a coagulant dosage of 2 mL/1,000 mL of turbid water to get the highest turbidity removal (97.9%). The results show that percentage removal increases with dosage until an optimum point is reached and then the percentage removal will start to decrease. This is because lower coagulant concentration results in reduced collision between suspended particles, making it difficult to form flocs while the opposite happens with high coagulant concentration with flocs being not properly formed which can even increase turbidity (Febrina *et al.* 2020).

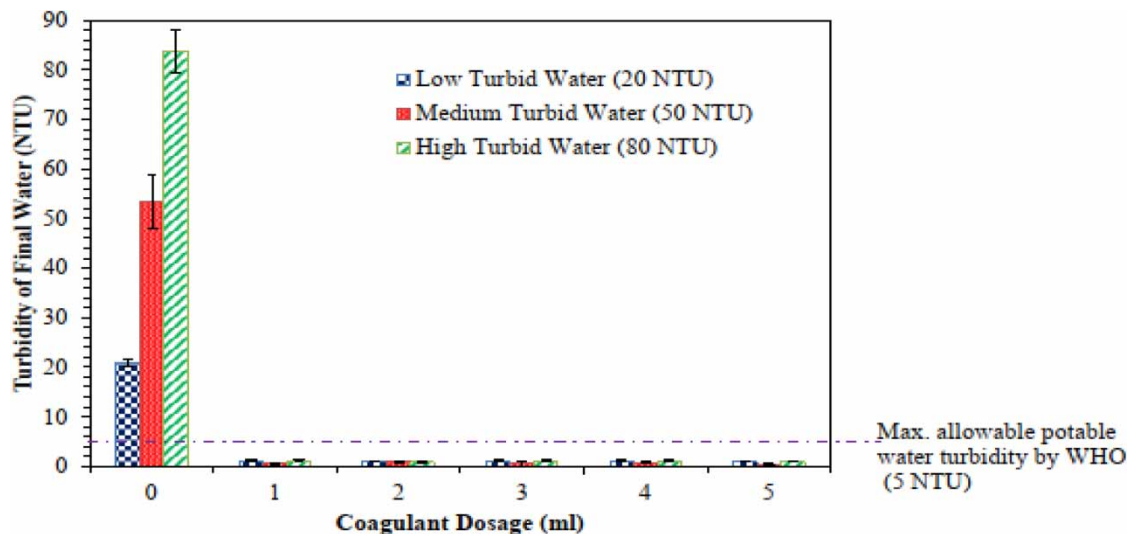


Figure 6 | Final turbidity against coagulant dosage for synthetic raw water.

Medium turbid water (50 NTU) showed the same trend of coagulant dosage being directly proportional to percentage removal as in low turbid water. The highest percentage of removal was recorded with a dose of 3 mL/1,000 mL giving 99.9% turbidity removal. Just as it is with low turbid water, the percentage removal increases with the dosage until an optimum is reached and it starts dropping.

Results for high turbid water (80 NTU) indicated that high turbid water required a bit higher coagulant dose of 5 mL/1,000 mL compared to low and medium turbidity in order to achieve an optimum percentage removal of 99.5%. This is because it requires the application of high coagulant dosage in order to improve turbidity removal efficiency for high turbidity water (Baghvand *et al.* 2010).

For all the coagulant dosages and levels of initial turbidity, the final water turbidity was within the acceptable WHO standard of at most 5.0 NTU for final water. Medium and high turbidity waters have higher percentage of turbidity removal compared to low turbidity water for all coagulant dosages since it is difficult to get particle collisions in low turbid water making it hard to facilitate the formation of flocs. This high-efficiency turbidity removal of the extracted coagulant is due to its high Al content. Al produces trivalent Al ions,  $Al^{3+}$  which are very effective in neutralizing the charge of colloidal particles since the charge of cations is directly proportional to the effectiveness of charge neutralization (Malik 2018). Alumina and ferrous compounds in the coagulant hydrolyses in water to form cationic species which can be absorbed by negatively charged particles that cause turbidity and neutralize their charge. This will destabilize particles that cause turbidity and induce flocculation (Baghvand *et al.* 2010).

### 3.3.2. Natural raw water

Raw water from Lotsane river, Palapye Botswana, was used to analyze the performance of the extracted coagulant on natural raw water. The initial turbidity was 381 NTU and it decreased with an increase in coagulant dosage as shown in Figure 7. The final water turbidity decreased until it reached the optimum of 11.35 NTU, giving a percentage turbidity reduction of 97.0% at a dosage of 5 mL/1,000 mL of raw water. Beyond the 5 mL/1,000 mL dosage, the turbidity started to increase. This is because high coagulant concentrations beyond the optimum point result in a more positive net charge of particles and therefore disturb the formation of flocs (Karnena *et al.* 2022).

During the jar tests for all the different levels of turbidity and dosages, it was observed that flocs were formed faster which facilitated a high settling rate. Large flocs were formed which also increased the settling rate as shown in Figure 8.

### 3.4. Adjustment of final water pH

The leachate coagulant (at 3.0 N and 5 g WTS/100 mL leaching agent) came out acidic and as a result, it altered the pH of the final water to acidic levels as shown in Figure 9. To counteract this effect, 10 g/L of water calcium oxide (CaO) solution was used. CaO is a base and will result in a neutralization reaction with acidic ions. At a pH of 12, CaO elevated the pH of the

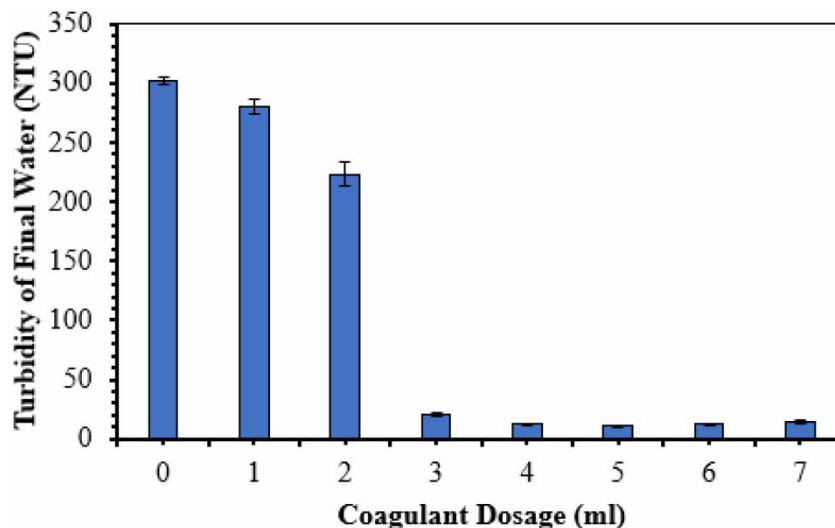
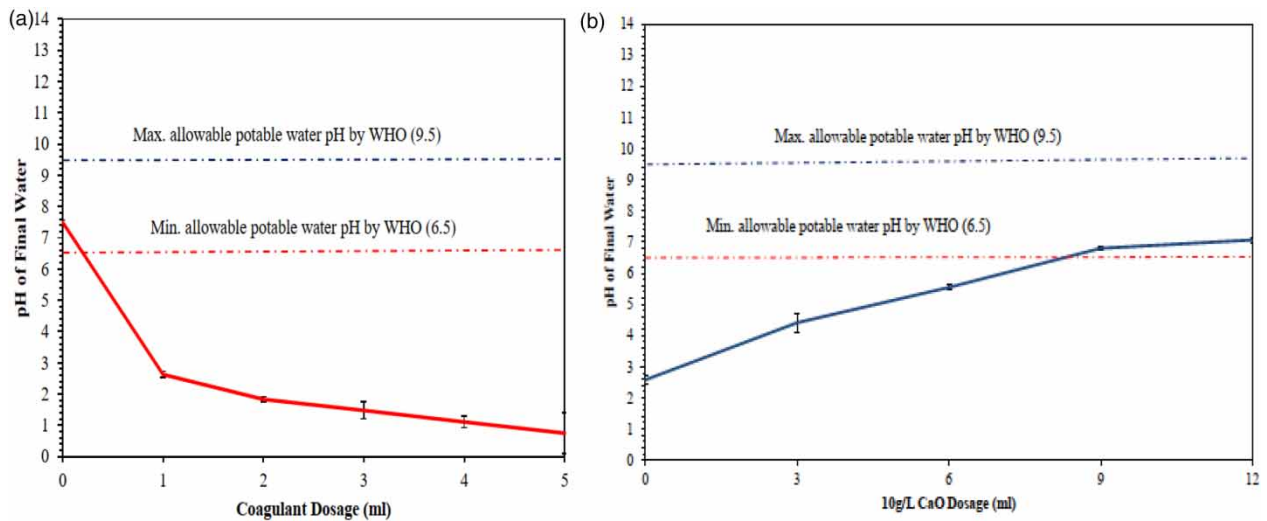


Figure 7 | Final turbidity against coagulant dosage for natural raw water.



**Figure 8** | Pictorial representation of floc formation during jar tests experiment.



**Figure 9** | (a) Change in pH of final water against coagulant dosage. (b) pH adjustment using 10 g/L CaO solution for final water treated with 1 mL of coagulant.

final water from 0.75 to 7.08 which is within the recommended WHO portable water pH range of 6.5–9.5. From [Figure 9](#), it requires 12 mL of 10 g/L of water CaO per 1 mL of coagulant to increase the pH to the recommended range of 6.5–9.5.

A comparison of results obtained in this study and previous studies is shown in [Table 3](#). The performance of the extracted coagulant was tested in the different levels of turbidity; low, medium and high while other studies tested only one level of turbidity. Coagulants from this study performed quite higher than those in comparison as shown in [Table 3](#). This is because the WTS used in this study has high Al and Fe content, which are very significant for coagulation compared to the WTS used by the other researchers. For instance, WTS used in this study had an Al content of 20.3% compared to the 9.58% used by [Tarique \*et al.\* \(2016\)](#). It also had 5.28% Fe content compared to 1.35% of Fe content in the WTS used by [Chigondo \*et al.\* \(2015\)](#). Further tests done with the real water sample, with elevated levels of turbidity, still indicated high levels of turbidity removal (97%). This shows the effectiveness of the coagulant obtained in this study and its potential for application in a

**Table 3** | Comparison of experimental results from this study to those of previous studies

	Experimental data from this study				Tarique <i>et al.</i> (2016)	Chigondo <i>et al.</i> (2015)	Sanga <i>et al.</i> (2018)
Raw water turbidity (NTU)	20	50	80	381	35	16.7	89.7
Treated water turbidity (NTU)	0.42	0.04	0.4	11.35	5	6.4	1.2
Extracted coagulant's dosage (mL/L)	2	3	5	5	8	0.04	–
% Reduction	97.9	99.9	99.5	97.0	85.7	61.7	98.7

typical WWTP. Thus, an effective environmentally benign method for preparing WTS coagulant has been achieved without compromising the water clarification levels.

#### 4. CONCLUSION

Characterization studies show that WTS mainly has high compositions of ferrous and alumina compounds as well as silica with the rest as trace elements. Functional groups present in the WTS are usually related to water, carbonates and oxides of Al, Fe and silicon. The utilization alternative studied in this project was the possibility of using WTS as a coagulant. Due to its high alumina and ferrous content, the WTS did quite well as a coagulant, giving turbidity removal percentages in the range of 91–99% with higher efficiency in the medium and high turbidity waters. The WTS coagulant, developed from a less energy-intensive method, also proved to be very effective for a real water sample with elevated levels of turbidity (381 NTU) giving turbidity reductions of 97%, fast forming and settling flocs. This demonstrated its potential to be used in typical water treatment plants adding a sustainability measure as waste is being utilized. The acidic effect of sulphuric acid used as a leaching agent reduced the pH of final water after coagulation and therefore it is necessary to co-dose the extracted coagulant with calcium hydroxide to reach the pH and turbidity recommended by the World Health Organization (WHO).

#### DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

#### CONFLICT OF INTEREST

The authors declare there is no conflict.

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First received 20 May 2022; accepted in revised form 13 March 2023. Available online 24 March 2023