Evaluation of sludge handling using acidification and sequential aluminum coagulant recovery: case study of El-Sheikh Zayed WTP

Mahmoud M. Fouad, Ahmed S. El-Gendy and Taha M. A. Razek

ABSTRACT

Huge amounts of water treatment sludge are wasted annually in Egypt. One common practice of sludge handling is to discharge into waterways or by disposal after drying into landfills. Currently, there is a critical need for sustainable solutions that provide environmental protection as well as benefits for water producers. Coagulant recovery is not new but is still a good solution for sludge handling and reduction of operational costs. This work aims at completing the picture on coagulant recovery from alum sludge in El-Sheikh Zayed water treatment plant (ESZ-WTP) by acidification using hydrochloric acid. This study also investigates the effect of sequential recovery on the efficiency of the coagulant and treated water quality. Sludge characterization was done through determination of metals' concentration and composition, water content and organic matter concentration. The influence of acid concentration, sludge mass, acid/sludge mass ratio, mixing speed, temperature, and mixing time on aluminum leaching was studied. At optimum values of these parameters, 82.4% aluminum recovery was obtained. In addition, repeated recoveries were carried out at laboratory scale using raw water from ESZ-WTP. Our research revealed that recovered coagulant can be used for three times' recoveries as the trihalomethanes remain within acceptable limits.

Key words | acidification method, aluminum recovery, alum sludge, sequential recovery, water purification

Mahmoud M. Fouad (corresponding author)
Quality and Environmental Affairs Department,
Holding Company for Water and Wastewater,
Cairo,
Egypt
E-mail: *mahmoudfouad82@gmail.com*

Ahmed S. El-Gendy
Department of Construction Engineering, School of Sciences and Engineering,
The American University in Cairo,
New Cairo,
Egypt

Taha M. A. Razek
Environmental Basic Sciences, Institute for Environmental Studies and Research,
Ain Shams University,
Cairo,
Egypt

INTRODUCTION

Coagulation is one of many steps used in surface water treatment to remove suspended particles, colloids, organics, color, and microorganisms from water before drinking (Keeley *et al.* 2014a; Fouad *et al.* 2017a, 2017b). Coagulation is carried out by mixing a coagulant with raw water. Common coagulants used worldwide include aluminum sulfate (alum), ferric chloride, and poly-aluminum chloride. Huge quantities of sludge from water treatment plants are produced when coagulants are added to raw water. In Egypt, alum is the common coagulant used in water clarification. Egyptian treatment plants annually consume about 365,000 tons of aluminum sulfate in the water clarification process. More than a million tons of wet alum sludge is produced daily by water treatment in Egypt.

In many developing countries such as Egypt, a common practice for the disposal of water treatment sludge is by dumping it into water bodies or sanitary sewers (Nair & Ahammed 2015). Disposal of water treatment sludge into waterways is reported to be toxic to aquatic life (Xu *et al.* 2017). In Egypt, due to regulatory changes, water treatment sludge has to be disposed into landfills. Since the levels of pollutants in the water treatment sludge are relatively low, as the best quality raw water sources are generally selected for drinking water production, recycling of alum sludge may be a feasible option instead of disposal. A number of research efforts has been made, particularly in recent years, to use the water treatment sludge in many beneficial ways. These include its use in building and construction materials (Fouad *et al.* 2017a, 2017b), in

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wastewater treatment (Nair & Ahammed 2015), and for soil conditioning (Keeley *et al.* 2014a). Two different approaches have been tested for the use of water treatment sludge in water and wastewater treatment. In the first approach, a coagulant is recovered from the sludge and is used for water and wastewater treatment. In the second approach, wet or dry sludge itself is used as a coagulant or adsorbent for removal of different contaminants. Recovery of coagulant metal from the sludge is an attractive option and has been reported in studies (Keeley *et al.* 2014a, 2016). Generally, four coagulant recovery methods can be employed on the chemical sludge generated from a water treatment plant. These methods include acidification, basification, ion exchange, and membrane processes (Keeley *et al.* 2014b). Most of the studies reported recently used acidification for coagulant recovery due to its high efficiency and low cost compared to other methods (Chen *et al.* 2012; Fouad *et al.* 2017a). Several factors are known to affect coagulant metal recovery using acidification (Keeley *et al.* 2014b; Fouad *et al.* 2017a, 2017b). Coagulant recovery from water treatment sludge was found to minimize the operational cost of water purification, preserve natural materials, and follow the new laws and regulations regarding disposal of sludge by reusing it after recovery instead of disposal in waterways. In addition, this will also improve the environmental quality from bioaccumulation of aluminum in the environment if released without treatment, which will protect aquatic life and, hence, human health and life.

In this study, the authors have tried to explore the feasibility of aluminum recovery from sludge using acidification to produce efficient coagulant and determine the limitations of the sequential recovery process to produce water with acceptable quality as drinking water. Especially trihalomethane (THM) concentration as the organic matter accumulation through the recovery process is the major concern for water production using recovered coagulant solutions.

MATERIALS AND METHODS

This research work tested different parameters that affect aluminum leaching from El-Sheikh Zayed water treatment plant (ESZ-WTP) sludge using hydrochloric acid. Moreover, optimum conditions detected were used to prepare an aluminum coagulant solution. The efficiency of the coagulant was compared to the commercial one and the effect of the two coagulants on the produced water quality was tested. Three sequential aluminum recoveries were studied for the recovered coagulant and the quality of the produced water was monitored.

Description of ESZ-WTP

ESZ-WTP consists of four main treatment phases: coagulation/flocculation; clarification using tube settler design; filtration using rapid sand filters; and pre- and post-chlorination for water disinfection. Aluminum sulfate (alum) was used in the coagulation of water at ESZ-WTP. Chlorine and coagulant doses were 5.5 and 30 mg/L, respectively, at the time of sludge sampling. Drying lagoons were used for drying the alum sludge produced and collected from the tube settler clarifiers. Three drying lagoons were operated in the following modes: one lagoon in the filling mode, one lagoon in the drying mode, and the last lagoon in the standby mode.

Experimental setup

The current study was carried out through three main phases. In Phase I, sampling, preparation, and characterization of sludge were done. In Phase II, the recovery of aluminum from sludge was applied using hydrochloric acid. Six different parameters that affect the recovery process were studied. The optimum values of the recovery parameters were applied to obtain a solution that had the best recovery of aluminum, then characterized and tested in water clarification and compared with the virgin coagulant (aluminum sulfate). In Phase III, sequential coagulant recovery from sludge using hydrochloric acid was evaluated. To sum up the experimental work phases and experiments, Table 1 was designed to provide a summary of the experimental steps carried out throughout the research.

Phase I: sludge sampling, preparation, and characterization

Experiment 1: sludge sampling and preparation

The sludge used in the current study was collected from different points in the lagoon operated in the drying mode, as

Table 1 | Summary of experimental phases and steps

Experiment	Description
Phase I: Sludge sampling, preparation, and characterization	
Experiment 1: Sludge sampling and preparation	Includes the methods used in sampling and preparation of sludge from ESZ-WTP before recovery
Experiment 2: Sludge characterization	This section exhibits a detailed illustration of the characterization of sludge showing different analytical techniques used
Phase II: Aluminum recovery, coagulant preparation, and characterization	
Phase II, Part I: Aluminum recovery using hydrochloric acid	
Experiment 1: Effect of hydrochloric acid concentrations	Various concentrations of hydrochloric acid were used to estimate the effect of acid concentration on the recovery efficiency of aluminum with fixing other conditions to constant values
Experiment 2: Aluminum recovery at different masses of sludge	Applying fixed conditions from temperature, rotational speed, acid concentration, and time, different sludge weights were used to evaluate its impact on aluminum recovery
Experiment 3: Effect of mixing time	By using constant values for other conditions, various mixing times were used to test its effect on the aluminum recovery
Experiment 4: Effect of temperature	Three different temperatures were used with constant values for other conditions to evaluate the effect of temperature on the recovery rate of aluminum coagulant
Experiment 5: Effect of mixing speed	Different mixing speeds were used to test its effect on aluminum leaching by stabilizing other conditions
Phase II, Part II: Testing the recovered coagulant solution	
Experiment 1: Preparation of a coagulant solution from alum sludge using hydrochloric acid	In this experiment, optimum conditions obtained from the above experiments were used to prepare coagulant solution
Experiment 2: Evaluation of the recovered coagulant solution in water clarification	In this step, the recovered coagulant was applied to the raw water of ESZ-WTP to evaluate its efficiency in clarification of water versus a commercial one
Phase III: Evaluation of sequential recovery using hydrochloric acid	
Experiment 1: Evaluation of virgin coagulant and proceeding first recovery	In this experiment, fresh coagulant was applied to raw water in ESZ-WTP and the resulting water analyzed to evaluate the coagulant. Afterwards, the sludge produced underwent recovery using the optimum conditions obtained through the experiments in Phase II, Part I
Experiment 2: Evaluation of first recovered coagulants and proceeding second recovery	The same as Experiment 1, Phase III but starting with applying the first recovered coagulant to the raw water and the produced water was analyzed then the sludge used to produce the second recovered coagulant
Experiment 3: Evaluation of second recovered coagulants and proceeding third recovery	The same as Experiments 1 and 2, Phase III but starting with applying the second recovered coagulant to the raw water and the produced water was analyzed then the sludge used to produce the third recovered coagulant
Experiment 4: Evaluation of third recovered coagulants	The same as Experiments 1, 2 and 3, Phase III but applying the third recovered coagulant to the raw water and the produced water analyzed to evaluate its efficiency in clarification of water

shown in Figure 1, then transferred to the laboratory in polyethylene bags. The collected sludge was dried in a laboratory oven (Venti-cell model: C100923) at 105 °C for 24 hours. Then, the dried sludge was ground and allowed to pass through a 2 mm sieve then mixed together to ensure the homogeneity before being used in the following experiments.

Experiment 2: sludge characterization

The sludge was characterized by measuring: (1) total volatile solids by weight loss on ignition at 550 °C (Santisteban *et al.* 2004); (2) water content through drying 10 g of wet sludge at 105 °C for 2 hours and reweighing it (USEPA 2001); (3)

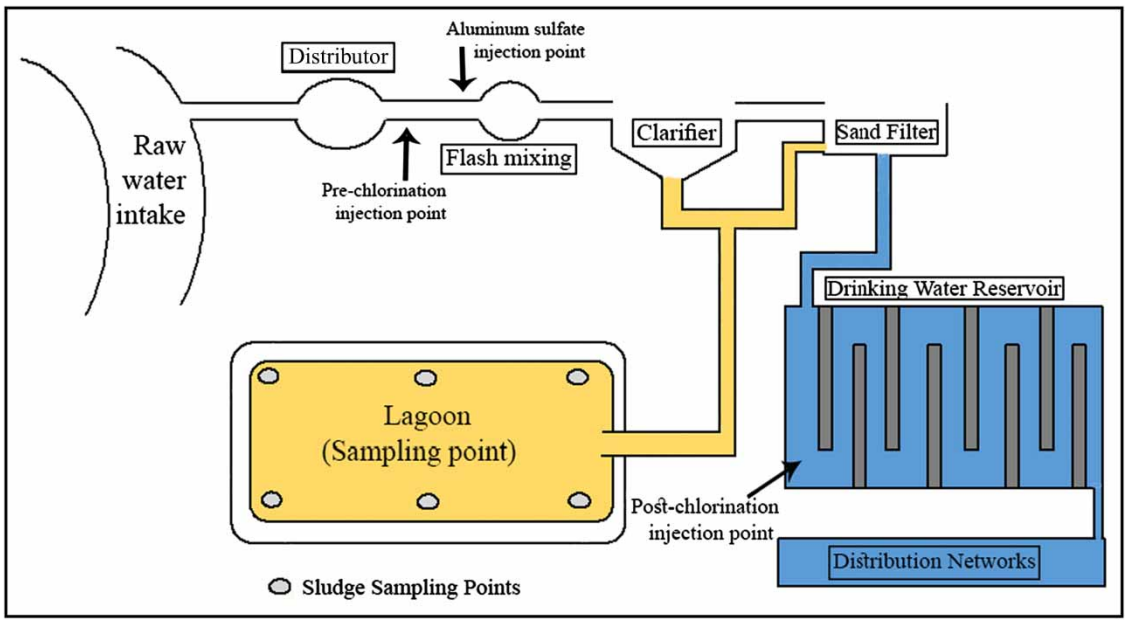

Figure 1 | Schematic diagram for ESZ-WTP showing the sludge collection points.

total metals' content was determined through acid digestion according to the US-EPA Method 3050 B (USEPA 1996). It was applied through mixing 1 g of sludge with 10 mL of 1:1 HNO_3 and covering with a watch glass. The sample was heated to 95 °C ± 5 °C and refluxed for 10–15 min without boiling, then it was allowed to cool for 5 min. Afterwards, 5 mL of concentrated HNO_3 was added and the sample heated up to 95 °C ± 5 °C and refluxed. This process was carried out repeatedly until the entire sample was digested.

After digestion, aluminum, iron, magnesium, cobalt, manganese, vanadium, strontium, chromium, nickel, lead, sodium, potassium, calcium, and zinc were analyzed using inductively coupled plasma optical emission spectroscopy (Optima 8300 dv ICP-OES, manufactured by PerkinElmer, USA) according to the US-EPA Method 200.7 (USEPA 1994).

Phase II: aluminum recovery, coagulant preparation, and characterization

This phase is composed of two parts. The first part is aluminum recovery, which includes five different experiments carried out to study the parameters and their optimum values that affect the aluminum extraction from sludge. These parameters include acid concentration, sludge mass, acid/sludge mass ratio, mixing speed, temperature, and mixing time. The second part is coagulant preparation using optimum conditions obtained from the first part and coagulant characterization through metals' analysis. All experiments in Part I were carried out using a jar test apparatus. In each experiment, leaching of aluminum from sludge was performed by adding 500 mL of hydrochloric acid solution to a known mass of sludge collected and prepared as explained in Phase I. The efficiency of the aluminum leaching process was investigated using the different parameters mentioned above. After acid/sludge mixing, the solution was allowed to settle for 10 min then filtered through a 0.45 μm filter paper. Then, the filtrate was analyzed for aluminum using Optima 8300 dv ICP-OES, according to the US-EPA method no. 200.7 (USEPA 1994). Each sample was measured three times and the average value is presented. All experiments were carried out in duplicate. The percentage of aluminum recovery was calculated using Equation (1):

$$\text{Al recovery } \% = \frac{\text{Al measured in the filtrate after recovery (mg Al/g dry sludge)}}{\text{Al measured in the filtrate after recovery (mg Al/g dry sludge)}} \times 100 \quad (1)$$

Phase II, Part I: aluminum recovery using hydrochloric acid

Five experiments were carried out to get the optimum conditions for coagulant preparation from sludge using hydrochloric acid, and were as follows.

Experiment 1: effect of hydrochloric acid concentration

In this experiment, the effect of hydrochloric acid concentration expressed as acid normality on aluminum recovery was studied. In this test, 500 mL of different hydrochloric acid solutions with different normality values was added to 5 g of alum sludge. The hydrochloric acid solutions were prepared at different normality values of 0.1, 0.5, 1.0, 1.5, and 2.0 N. Leaching with the different acid normality values was performed at a constant temperature of 25 °C. The acid solution and sludge were mixed at a constant rotational speed of 80 rpm for 20 min. After mixing, solutions were allowed to settle, then filtered. The filtrates were sampled to determine the concentration of recovered aluminum, hence, leaching efficiency.

Experiment 2: aluminum recovery at different masses of sludge

Here, aluminum recovery was investigated using 1 N hydrochloric acid solution and 5, 10, 15, 20, and 25 g of alum sludge prepared as discussed before. The leaching of aluminum was performed at 25 °C. The acid solutions were mixed with the different masses of sludge at a constant paddle speed of 80 rpm for 20 min. The produced solutions were allowed to settle, then filtered, and aluminum was analyzed in filtrates and recovery efficiency was calculated.

Experiment 3: effect of mixing time

In this trial, five mixing times were examined in five different jars. In each jar, 500 mL of 1.0 N hydrochloric acid solution was added to 5 g of sludge. Then, the solution was mixed at a constant speed of 80 rpm. The mixing was performed at a fixed temperature of 25 °C for 10, 20, 30, 40, and 50 min. At the end of the mixing, solutions were allowed to settle then filtered. The filtrates were sampled to determine the amount of recovered aluminum and the leaching efficiency.

Experiment 4: effect of temperature

In this experiment, the effect of solution temperature on aluminum leaching from alum sludge was investigated. Three different temperatures of 25, 40, and 60 °C were tested. The temperature was adjusted using a hot plate and a mercury thermometer. The temperature was kept constant during the mixing time. At each temperature, 500 mL of 1.0 N hydrochloric acid solution was added to 5 g of sludge mass. Then, the solution was mixed at a constant speed of 80 rpm for 20 min. At the end of mixing, solutions were allowed to settle then filtered. The filtrates were sampled to determine the recovered aluminum.

Experiment 5: effect of mixing speed

In this experiment, five mixing speeds were tested in five different jars. In each jar, 500 mL of 1.0 N hydrochloric acid solution was added to 5 g of sludge mass. Then, the solution was mixed at stirring speeds of 20, 40, 60, 80, and 100 rpm for 20 min. The effect of mixing speeds was performed at a constant temperature of 25 °C. After mixing the sludge with the acid solution, the suspension was allowed to settle then filtered as indicated before. The filtrates were sampled to determine the amount of dissolved aluminum.

Phase II, Part II: testing the recovered coagulant solution

This part of Phase II, consists of two experiments. In the first one, the optimum conditions obtained from the five experiments above were used to recover a coagulant in hydrochloric acid solution. In the second experiment, the recovered coagulant was tested against the commercial aluminum sulfate (alum) in the clarification of raw water sampled from ESZ-WTP using jar test.

Experiment 1: preparation of a coagulant solution from alum sludge using hydrochloric acid

After completing the five experiments in the first part of Phase II, the optimum values of the recovery parameters obtained from Part I of Phase II were applied to prepare a solution that had the optimum recovery of aluminum, hence used as

a coagulant in the treatment process. The percent of aluminum recovered in the coagulant solution was measured. In addition, the recovered solution was evaluated by comparing its composition with the commercial alum that was used in ESZ-WTP at the time of the study.

Experiment 2: evaluation of the recovered coagulant solution in water clarification

In this experiment, raw water samples were collected from the intake of ESZ-WTP to perform the testing. Jar tester was used to evaluate the performance of recovered coagulant solution versus commercial alum in clarification of raw water. The commercial alum and recovered coagulant were added to 1 L of raw water at the same doses, which were 20, 25, 30, 35, 40, and 45 mg/L. Jar test was performed by filling six rounded jars of 1 L capacity for each by raw water, then coagulant was added and mixed with raw water at a mixing speed of 120 rpm for 2 min. Then, the water was mixed slowly at a mixing speed of 40 rpm for 18 min, which simulates the operating conditions in ESZ-WTP. The water was allowed to settle for 10 min before samples were collected from the supernatant. The collected samples of raw and clarified water were analyzed for pH, turbidity, temperature, conductivity, total dissolved solids, total alkalinity, chlorides, aluminum, iron, manganese, total THMs, algal count, total plate count, and total coliform bacterial count. All analyses of samples were performed according to *Standard Methods for the Examination of Water and Wastewater* (APHA, AWWA, WEF 2012).

Phase III: evaluation of sequential recovery using hydrochloric acid

In this phase, sequential coagulant recovery was studied in order to evaluate the accumulation of different contaminants in the recovered coagulants and to determine how long the sludge would be recyclable without affecting the quality of the treated water. The experiments in this phase were carried out using jar test (Reid & Edison 2004). Water samples were collected and analyzed according to *Standard Methods for the Examination of Water and Wastewater* (APHA, AWWA, WEF 2012). Four different experiments were the building blocks for this phase and they were as follows.

Experiments 1–4: evaluation of virgin and recovered coagulants and process of the sequential recoveries

Jar test was carried out by applying optimum doses of virgin aluminum sulfate and chlorine on raw water and the produced sludge was subjected to recovery using hydrochloric acid as explained before to produce the first aluminum coagulant. This process was sequentially repeated twice to produce and evaluate the second and third recovered coagulants. The treated water each time was collected and evaluated through analyzing pH, turbidity, temperature, conductivity, total dissolved solids, total alkalinity, chlorides, aluminum, iron, manganese, total THMs, algae count, total plate count at 35 °C, and total coliform bacteria.

RESULTS AND DISCUSSION

Sludge characteristics

Alum sludge was characterized to explore the feasibility of coagulant recovery using hydrochloric acid and its application as an alternative for the commercial coagulant. The composition of alum sludge from ESZ-WTP is a reflection of the structure of the virgin coagulant used in the treatment process and the contaminants in raw water. The analysis of sludge collected from ESZ-WTP showed that the average values of the water and organic contents were 86% and 25%, respectively. In addition, Table 2 shows the concentrations of metals in the sludge collected from ESZ-WTP. As shown, the major metal constituents of the sludge were aluminum 13.3%, iron 2.2%, and calcium 1.2%. These are nearly the same constituents of the commercial alum used in the treatment process. This indicates that the main source of metals in the sludge is the coagulant used in the treatment and not from raw water.

Effect of different leaching parameters on aluminum recovery

The following part shows the results obtained from the different experiments (Experiments 1–5) carried out in Phase II, Part I to determine the optimum conditions for aluminum leaching from sludge.

Table 2 | Concentration of different metals (expressed in gram of metal per 100 g sludge on dry mass basis) in sludge samples collected from the drying lagoons of the ESZ-WTP

Metal	**Al**	**Ba**	**Co**	**Cr**	**Cu**	**Fe**	**Li**
Conc.	13.34	0.01	0.002	0.013	0.001	2.21	0.001
Metal	**Mn**	**Ni**	**Pb**	**Sr**	**V**	**Zn**	
Conc.	0.27	0.004	0.087	0.017	0.012	0.011	
Metal	**K**	**Na**	**Ca**	**Mg**			
Conc.	0.216	0.164	1.203	0.902			

Effect of hydrochloric acid concentration on the leaching of aluminum

Figure 2 shows the effect of acid concentration on the leaching efficiency of aluminum. As shown, the leaching efficacy increases with hydrochloric acid concentration. That is due to the aluminum dissociation from aluminum hydroxide in the sludge being enhanced much more by the increase in hydrogen ion concentration. In other words, decreasing the solution pH promotes the aluminum hydroxide transformation to aluminum chloride, hence increasing the leaching efficiency. Equation (2) shows the formation of aluminum chloride as a response to sludge (aluminum hydroxide) acidification.

$$Al(OH)_3 + 3HCl \leftrightarrow AlCl_3 + 3H_2O \quad (2)$$

As presented in Equation (2), more aluminum chloride was produced by increasing the added acid. Based on the tested range of acid concentration, the maximum normality that can produce the highest recovery of aluminum from an economic viewpoint is 2.0 N as more leaching efficiency is expected to be achieved by higher normality according to the literature (Fouad *et al.* 2017a, 2017b). 2.0 N acid solution was capable of achieving aluminum recovery of 67.9% from sludge. The results of the current work agree with the findings in the literature that acid extraction led to higher solubility rates for iron and manganese (Boaventura *et al.* 2000; Fouad *et al.* 2017b) and aluminum leaching increases with lowering the solution pH (Chen *et al.* 2012; Fouad *et al.* 2017a). It was also reported that aluminum leaching from alum sludge could reach 61–100% when using acidification at a pH range of 1–3 (Xu *et al.* 2009). Fan *et al.* (2013) also recorded that the leaching rates of Ni and Co obviously increased relatively slowly by increasing the concentration of sulfuric acid to 40% (mass basis) and the recovery rate was above 96%.

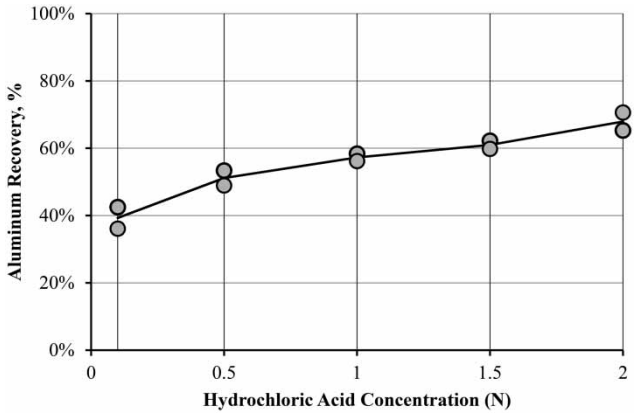

Figure 2 | Effect of acid concentration on aluminum recovery from alum sludge (sludge mass = 5 g; temperature = 25 °C; mixing speed = 80 rpm; mixing time = 20 minutes; each point is the average value of analyzing a sample three times).

Effect of alum sludge mass on the leaching of aluminum

Figure 3 shows the change in aluminum dissolution from the sludge at different sludge masses using 1.0 N hydrochloric acid. As shown, higher aluminum recovery is obtained with a lower mass of sludge. This is due to the lower sludge weight being applied for acidification; higher contact between acid and sludge particles is conducted, hence, more aluminum recovery is achieved. This agrees with the literature that the leaching rates of aluminum, iron, and manganese increased with the decrease in sludge mass (Boaventura *et al.* 2000; Fan *et al.* 2013; Fouad *et al.* 2017a, 2017b). Under the mentioned conditions, Figure 3 shows that the best recovery was 59.5% at a sludge mass of 5 g.

Effect of acid/sludge mass ratio on aluminum recovery

As shown previously in Figures 2 and 3, the concentration of acid and the mass of sludge are two major controlling factors that affect directly aluminum dissolution from sludge. Therefore, to normalize their effect, the impact of acid to sludge mass ratio on the aluminum recovery has been investigated. Figure 4 shows the effect of acid/sludge mass ratio (g/g) on aluminum recovery. The figure shows that by increasing acid/sludge ratio aluminum recovery increases. Aluminum recovery increased significantly until an optimum ratio of 1.83 (g/g), which gives aluminum an average recovery of 52.7%. Beyond the optimum ratio, a high value is needed to give only a little increase in aluminum recovery. Almost three times the optimum value is required to show an increase in the recovery of about 8%. Therefore, an acid/sludge mass ratio of 1.83 is recommended for aluminum recovery from sludge. This agrees with the literature, that the recovery of metals from alum sludge or waste sludge increases by decreasing pH of the solution and the sludge mass (Boaventura *et al.* 2000; Xu *et al.* 2009; Chen *et al.* 2012; Fan *et al.* 2013; Fouad *et al.* 2017a, 2017b).

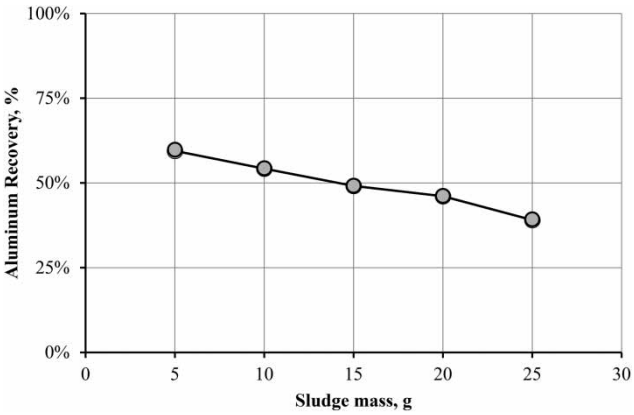

Figure 3 | Change in aluminum recovery with the change in sludge dry mass (acid normality = 1.0 N; temperature = 25 °C; mixing speed = 80 rpm, mixing time = 20 minutes; each point is the average value of analyzing a sample three times).

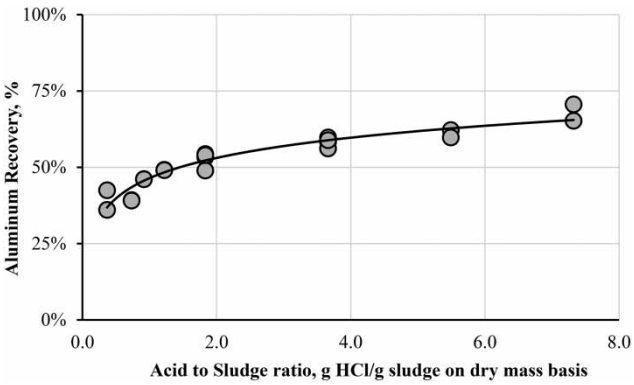

Figure 4 | Efficiency of aluminum recovery from alum sludge at different acid/sludge ratio on dry mass basis (temperature = 25 °C; mixing speed = 80 rpm; mixing time = 20 minutes).

Effect of temperature on aluminum recovery

Figure 5 shows the effect of temperature on aluminum leaching from sludge. The leaching of aluminum from sludge increases with temperature until 40 °C, after which, only a small increase in aluminum recovery is noted by elevating the temperature. The highest recovery of aluminum (75.1%) is achieved at 60 °C. This is compared to an aluminum recovery of 73.1% at 40 °C. Therefore, the optimum temperature recommended for the aluminum recovery from sludge is 40 °C. The effect of temperature on aluminum leaching is explained through higher temperatures allowing the acid solution to accept more aluminum from the sludge and raising the degree of the solution saturation. In addition, increased temperature pushes the aluminum ions to become easily liberated from the sludge and react with the chloride ions in hydrochloric acid solution to show more leaching efficiency. This agrees with the findings in the literature, that by applying different temperatures and at a particular reaction time, leaching rate of aluminum in acid solution increases with temperature (Cheng *et al.*2012; Fouad *et al.* 2017a). In addition, it has been proved that the leaching

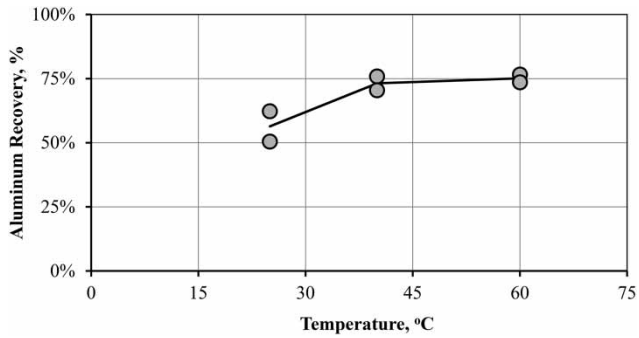

Figure 5 | Effect of temperature (°C) on aluminum recovery from alum sludge (acid normality = 1.0 N; sludge mass = 5 g; mixing speed = 80 rpm; mixing time = 20 minutes; each point is the average value of analyzing a sample three times).

rate of metals such as Ni and Co in acid solution from waste super-alloys is more influenced by increasing temperature. A leaching rate of 96.68% (weight basis) was reported for Ni and Co when the temperature was elevated to 85 °C (Fan *et al.* 2013). Our results also comply with Fouad *et al.* (2017b), who reported increased iron, manganese, and chromium leaching from alum sludge using sulfuric acid with increasing the temperature.

Effect of mixing speed on aluminum recovery

The results in Figure 6 show that the leaching efficiency of aluminum increases with the increase in mixing speed until it reaches 80 rpm. Beyond 80 rpm, leaching efficiency does not show a significant increase. This reveals that, when mixing speed reaches a threshold value, the thickness of the diffusion layer can no longer be effectively reduced causing any more aluminum dissolution. This diffusion layer controls the dissolution of aluminum from the dry sludge into the acid solution. Therefore, mixing intensity beyond a threshold value does not affect aluminum leaching rate (Cheng *et al.* 2012). Figure 6 shows that at the optimum mixing speed of 80 rpm, the average aluminum recovery can reach 49.3%. This shows that the more rotational speed is applied, higher agitation to the sludge with the acid is achieved, which helps more aluminum to react with hydrochloric acid and thus more recovery occurs. The dissipated power through rotational speed allows aluminum leaching until a threshold is reached, which is 80 rpm in this study. That could be attributed to the rotational speed being unable to provide more aluminum leaching within the applied conditions from acid concentration, temperature, time, and sludge weight. Similar results were obtained in the literature by applying mixing speeds in the range of 20–120 rpm. It was found that the leaching ratio of aluminum increases with the increase in the mixing speed until 80 rpm, beyond which the mixing speed had no effect on the leaching of aluminum (Cheng *et al.* 2012). In addition, Gürmen (2005) reported that metals' dissolution, namely cobalt, iron, and nickel from cemented carbide scrap in acidic solutions, increased by increasing stirring speeds between 700 and 900 rpm.

It was also proved by Fan *et al.* (2013) that leaching rate increases greatly initially from waste super-alloys with increasing the stirring speed to 250 rpm. Nevertheless, when mixing speed is elevated more than 250 rpm, the leaching rate of Ni and Co decreases greatly because materials will be rotating with the solution and the effect of stirring decreased.

Effect of mixing time on the recovery of aluminum

As shown in Figure 7, the aluminum recovery increases with the increase in mixing time until it reaches 40 minutes when maximum recovery is achieved (80.4%). Beyond 40 minutes of mixing time, the aluminum recovery decreases. The reduction in recovery after 40 minutes may be due to the adsorption of aluminum on the rotating particulates from the remaining sediments from the sludge. This agrees with findings in the literature (Cheng *et al.* 2012), where it was

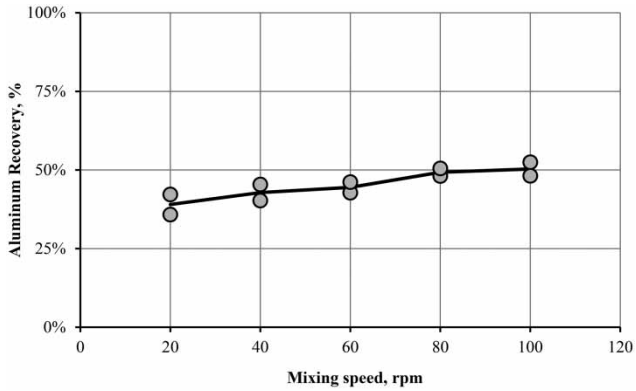

Figure 6 | Effect of mixing speed on aluminum recovery from alum sludge (acid normality = 1.0 N; sludge mass = 5 g; temperature = 25 °C; mixing time = 20 minutes; each point is the average value of analyzing a sample three times).

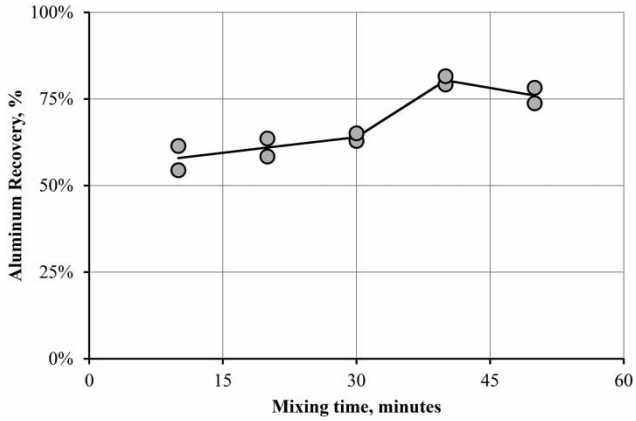

Figure 7 | Effect of mixing time on aluminum recovery from alum sludge (acid normality = 1.0 N; sludge mass = 5 g; temperature = 25 °C; mixing speed = 80 rpm; each point is the average value of analyzing a sample three times).

proved that aluminum leaching increased with reaction time up to 20 minutes, but after exceeding 20 minutes leaching reaction reached a steady state that is not influenced by acid concentration or leaching time. Our findings also comply with Fouad *et al.* (2017a, 2017b), who reported increased metals leached from alum sludge including aluminum by increasing mixing time.

Characteristics of prepared recovered coagulant solution

From the experiments of Phase II, Part I the optimum parameters of aluminum recovery from sludge were obtained. These parameters include a hydrochloric acid concentration of 2.0 N, an acid to sludge ratio of 1.83 on a mass basis, a mixing speed of 80 rpm, at a temperature of 40 °C and a mixing time of 40 minutes. The use of this combination of optimum values resulted in a higher recovery of aluminum from the sludge, which reached 82.4%. Table 3 shows the metals' content in the recovered coagulant solution compared with the commercial alum. It is obvious from Table 3 that the metals' concentrations in the commercial alum are higher than that in the recovered coagulant. This assures that the main source of metals in the recovered coagulant is the commercial one. In addition, the reduction in metals' concentrations of the recovered coagulant against the virgin solution is explained through two points: first, not all the metals are precipitated and removed in the sludge; and second, the leaching process did not perform 100% recovery for all the metals in the sludge.

Table 3 | Comparison between the metals' analysis of the recovered coagulant solution from the alum sludge and commercial alum (all metals are expressed in mg/L)

	Ba	Co	Cr	Cu	Fe	Li
C.A.S	0.06	0.01	0.08	0.03	13.31	0.01
R.C.S	0.06	0.01	0.06	0.02	10.70	0.00
	Mn	**Ni**	**Pb**	**Sr**	**V**	**Zn**
C.A.S	1.08	0.02	0.52	0.10	0.07	0.07
R.C.S	0.87	0.02	0.42	0.05	0.06	0.05
	K	**Na**	**Ca**	**Mg**		
C.A.S	0.782	0.882	14.52	4.78		
R.C.S	0.487	0.731	10.54	2.462		

C.A.S: commercial aluminum sulfate; R.C.S: recovered coagulant solution.

Evaluation of using recovered coagulant solution versus commercial alum in clarification of raw water from ESZ-WTP

In Phase II, Part II of this study, the optimum doses of the recovered coagulant solution and commercial alum were applied to the raw water using jar test. The analyses of the treated water by the optimum doses of recovered coagulant solution and commercial alum are reported in Table 4. As shown, the quality of the treated water using recovered coagulant solution and commercial alum was almost similar. Turbidity was 1.4 and 1.9 NTU when using commercial alum and recovered aluminum solution, respectively. This reveals that the recovery process did not affect leached coagulant efficiency in turbidity removal. This agrees with the findings of Keeley *et al.* (2014a), who reported that the recovered coagulants were capable of treating water and removed 60–70% of raw water turbidity for river and upland reservoir waters.

Table 4 | Complete analyses of raw water from ESZ-WTP and treated water from jar testing using recovered coagulant solution and commercial alum

No.	Parameter	Unit	RW	TW – CAS	TW – RCS
1	pH	–	7.9	7.4	7.6
2	Turbidity	NTU	8.0	1.4	1.9
3	Temperature	°C	24.3	25.2	25.1
4	Conductivity	μS/cm	327	338	339
5	Total dissolved solids	mg/L	218	225	226
6	Total alkalinity	mg/L	147	126	134
7	Chlorides	mg/L	18	23	66
8	Aluminum	mg/L	0.05	0.18	0.17
9	Iron	mg/L	0.02	ND	ND
10	Manganese	mg/L	ND	ND	ND
11	Total THMs	mg/L	ND	0.07	0.09
12	Algae count	Organism/mL	5,130	207	217
13	Total plate count at 37 °C	Cell/mL	2,970	32	43
14	Total coliform bacteria	Cell/mL	1,485	160	190

RW: raw water of ESZ-WTP; TW – CAS: treated water from the jar test at the optimum dose of commercial aluminum sulfate; TW – RCS: treated water from the jar test at the optimum dose of recovered coagulant solution (three times recovery).

One of the main issues regarding alum sludge recycling using acidification is the accumulation of disinfection by-products, especially THMs, in sludge and their reintroduction into water being treated (Keeley *et al.* 2014b). As shown in Table 4, the concentration of the THMs in the treated water using the recovered solution is 0.09 mg/L, which is slightly higher than that when commercial alum is used (0.07 mg/L). Chloride concentration is also greater in the water treated using the recovered coagulant, which is attributed to the chloride ions introduced by the hydrochloric acid used in the recovery process. Another observation on the treatment using the recovered coagulant is the biological aspects of water including algal count; total plate count at 37 °C and total coliform bacteria were found to show a little difference in treated water using recovered coagulant if compared to the water produced using the commercial coagulant. Taking into account a fixed disinfectant dose and biological properties of raw water, the increase in algae and bacteria may reflect a small reduction in the efficiency of recovered coagulant in removing these organisms physically by coagulation and sedimentation.

Evaluation of sequential aluminum recovery for sludge produced from ESZ-WTP raw water treatment using jar test

This part discusses the results obtained from Experiments 1–4 of Phase III to evaluate sequential coagulant recovery and its effect on the resulting water quality.

As mentioned before, the main risk of using recovered coagulants is the accumulation of contaminants in sludge that are recycled in the recovery process of aluminum coagulants. To complete the picture regarding using recovered coagulants in water treatment safely without affecting the produced water quality, sequential (repeated) aluminum recovery was carried out by applying the optimum conditions for recovery using hydrochloric acid and then the produced waters were analyzed and are expressed in Table 5.

As shown in Table 5, although repeated recoveries do not show a significant change in the overall quality of treated waters, they show a remarkable increase in the total THMs. It can be seen that the highest concentration of the THMs appears using the third recovered coagulant;

Table 5 | Complete analysis for raw water and tap water in ESZ-WTP, jar test waters for commercial aluminum sulfate, first, second, and third recovery for aluminum coagulants using hydrochloric acids

No.	Parameter	Unit	Raw water	Tap water	J.T.W.C.A.S	C1	C2	C3
1	pH	–	8.1	7.3	7.4	7.6	7.7	7.6
2	Turbidity	NTU	8.6	0.35	1.4	1.9	1.8	1.9
3	Temperature	°C	24.3	25.2	25.2	22.6	23.8	22.4
4	Conductivity	μS/cm	329	335	338	339	341	340
5	Total dissolved solids	mg/L	218	224	225	226	227	226
6	Total alkalinity	mg/L	147	126	126	134	134	134
7	Chlorides	mg/L	18	22	23	66	66	67
8	Aluminum	mg/L	0.05	0.15	0.18	0.17	0.21	0.19
9	Iron	mg/L	0.01	ND	ND	ND	ND	ND
10	Manganese	mg/L	ND	ND	ND	ND	ND	ND
11	Total THMs	mg/L	ND	0.09	0.07	0.08	0.09	0.1
12	Algae count	Organism/mL	5,035	17	207	217	215	230
13	Total plate count at 35 °C	Cell/mL	2,830	5	32	43	37	45
14	Total coliform bacteria	Cell/mL	1,630	ND	150	190	210	180

ND: not detected; J.T.W.C.A.S: jar test water produced using virgin aluminum sulfate; C1: jar test water produced using recovered aluminum coagulant using HCl (first time); C2: jar test water produced using recovered aluminum coagulant using HCl (second time); C3: Jar test water produced using recovered aluminum coagulant using HCl (third time).

however, the water is still within acceptable limits for THM value for drinking water which is 0.1 mg/L. These findings are compatible with Keeley *et al.* (2014a), who reported that organic matter removed from surface water by a coagulation process is concentrated in the sludge. Acidification of alum sludge solubilizes organic matter as well as aluminum from sludge (Keeley *et al.* 2014b). In addition, performing the coagulant recovery process sequentially allows organic matter to be solubilized and reintroduced to water, then precipitated during coagulation and precipitation into sludge, then transferred to the recovered solutions again. In other words, the organic matter accumulation is directly proportional to the number of coagulant recoveries. Therefore, the increase in THMs is not due to THMs in sludge because they were already decomposed during the drying process in the preparation step (WHO 2005). The main cause is due to the organic content in sludge, which leached to the recovered solutions by acidification and acts as a precursor for THM formation during the disinfection process. This indicates that the maximum possible sequential times for coagulant recovery from ESZ-WTP sludge is three times, and then a virgin coagulant should be used to avoid accumulation of organic pollutants in the treated water. Thus, the recovery process will be applied on the sludge for three cycles of operation. The repeated recovery of aluminum coagulant from the sludge will reduce the amount used of virgin coagulant by about 66%. In addition, it will reduce the area needed for drying lagoons, as less sludge will be dumped into these lagoons.

CONCLUSION

The recovery of aluminum from sludge generated in water treatment can be achieved using hydrochloric acid. The aluminum recovery using the optimum conditions obtained through the study by adding hydrochloric acid of 2.0 N to dry sludge at an acid to sludge mass ratio of 1.83 and mixing them at 80 rpm for 40 minutes at a temperature of 40 °C results in a recovery efficiency of 82.4%. The recovered coagulant solution was analyzed and found to be similar in composition to the commercial alum. According to our study, the maximum possible sequential recoveries for coagulant from ESZ-WTP sludge are three times, then a noticeable increase in THMs will appear in the resulting water, so a virgin coagulant should be used after three times' recovery to refresh the sludge before applying the recovery process again. Recovered coagulant performance shows no significant deviation from commercial coagulant with three times' sequential recoveries, which empowers its usage on a large scale in Egypt.

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