J Mater Cycles Waste Manag (2017) 19:1228–1234
DOI 10.1007/s10163-016-0513-0

ORIGINAL ARTICLE

Influence of sludge characteristics on coagulant recovery from water treatment sludge: a preliminary study

Abhilash T. Nair[1] · M. Mansoor Ahammed[2]

Received: 22 October 2015 / Accepted: 9 May 2016 / Published online: 25 May 2016

Abstract This study investigated the effect of raw water quality and coagulant type and dosage on the efficiency of aluminium recovery from water treatment sludge by acidification process. One of the features of this study is to prioritise several factors affecting aluminium recovery using demonstrating experiments and analysing the result using model. The previous studies comparing the aluminium recovery from PACl and alum sludge had collected the sludge from treatment plants treating different source water. However, this study compares aluminium recovery from PACl and alum sludge obtained from treating same source water. Water treatment sludge was prepared in laboratory by coagulating raw water with different inorganic and organic contents using aluminium sulphate (alum) and polyaluminium chloride (PACl) as coagulants. The effect of the different factors on the aluminium recovery efficiency was evaluated using a 2^3 full factorial experimental design. The aluminium recovery efficiency was higher for PACl sludge compared to alum sludge with recovery ranging from 62.5 to 74.5 % for alum sludge and from 70.7 to 84.0 % for PACl sludge. At higher concentrations of organic matter and turbidity in the raw water, lower aluminium recovery efficiency was observed for both the sludges. The study thus shows the important effects of raw water quality and coagulant type and dose on the recovery process.

✉ Abhilash T. Nair
nairabhilasht@gmail.com

[1] Civil Engineering Department, Dr. D. Y. Patil Institute of Engineering and Technology, Pimpri, Pune 411018, India

[2] Civil Engineering Department, Sardar Vallabhbhai National Institute of Technology, Surat 395007, India

Keywords Aluminium sulphate sludge · Coagulant recovery · Polyaluminium chloride sludge · Sludge management · Water treatment sludge

Introduction

Coagulation–clarification using iron or aluminium salts, is a widely used water treatment process. The large quantity of water treatment sludge (WTS) generated during the coagulation–clarification process is usually disposed into the water bodies or to landfill or applied on land considering it as non-hazardous and inert material [1, 2]. However, landfill disposal increases the land requirement while the WTS disposal into water bodies and land has a damaging effect on fish and plants due to its large Al and Fe contents [3].

WTS formed after coagulation process has a gelatinous structure with feathery and bulky nature [4]. Although WTS settles easily, it is barely possible to dewater it without any pre-treatment [4]. The biological digestion or incineration is not possible in case of WTS due to its low nutritional or calorific value [2]. The production of WTS will be increasing due to population explosion and increase in potable water demand. Hence the proper management of WTS is a serious environmental concern. The composition of aluminium-WTS (Al-WTS) is similar to soil with SiO_2 and Al_2O_3 as the main constituents along with aluminium hydroxide precipitates formed during coagulant process [5, 6]. The aluminium hydroxide present in WTS has amphoteric characteristics and is highly soluble in both acidic and alkaline pH. This facilitates coagulant recovery from WTS by simply adjusting the pH of WTS [7]. As the pollutant level is generally low in WTS due to use of good quality source water, coagulant recovery is an attractive option [8]. The water molecules bound within the sludge flocs are also displaced in the coagulant recovery process. This reduces the sludge volume after the coagulant recovery process and lowers the sludge handling cost [9, 10].

Acidification is the most popular coagulant recovery method due to its low cost and high efficiency in comparison to alkalization [8, 10–12]. Majority of the past researchers had focused on the coagulant recovery and sludge volume reduction from Al-WTS sludge as aluminium salts such as aluminium sulphate (alum) and polyaluminium chloride (PACl) are commonly used in the water treatment process [4, 6, 8, 11, 13].

Aluminium recovery of 60–100 % has been reported in the pH range of 1–3 in different studies [7, 11]. Although coagulant recovery is a simple process, its efficiency is governed by a number of factors such as sludge characteristics, raw water characteristics, type and dose of coagulant used which in turn depends on raw water characteristics and on concentration of type of solids present [11, 13].

Previous studies which compared aluminium recovery from alum and PACl sludges concluded that Al recovery from PACl sludge was greater when compared to that from alum sludge [7, 11]. However, the sludge for these studies were obtained from water treatment plants which treated water from different sources with varying quality and had used different coagulant dosages. Thus there is a need to study the influence of raw water quality and coagulant type and dose on coagulant recovery in a more controlled environment. One factor at a time (OFAT) approach is generally used to study the effect of different factors involved in acidification process [4, 8, 11, 13]. However, this involves large number of experiments, consuming time and resources with little information on interaction effect among the factors using design of experiments (DoE) approach [14, 15]. A better way would be to perform statistically planned experiments as an effective way to extract maximum information with a limited number of experiments using design of experiments (DoE) approach [14, 15]. In DoE, the most commonly used first-order design is the two-level factorial which experimentally studies each factor at only two levels. The number of experiments involved in two-level full factorial design is 2^k where k is the number of factors, each tested at two levels (-1 for low level and $+1$ for high level) [15, 16]. Two-level factorial designs are useful for either preliminary studies or initial optimization process [14]. Hence testing the model for more number of points in between would provide more reliable data and help us to understand the process in between the two points.

In the present study, the effect of raw water characteristics and coagulant type and dose on the coagulant recovery by acidification process was investigated using design of experiments approach. The factorial experimental design was applied to prepare Al-WTS in the laboratory with different raw water quality and coagulant dose using alum and PACl as coagulants.

Materials and methods

Factorial experiments

Influence of three variables, organic content (added as humic acid), coagulant dose and turbidity (added as clay) on aluminium recovery was investigated in this study. Each factor was tested at two different levels, correspondingly coded as (-1) for lower and ($+1$) for higher level. The factorial design used is given in Table 1. Turbidity levels and humic acid levels in the range of 25 NTU (-1) to 50 NTU ($+1$) and 1 mg/L (-1) to 5 mg/L ($+1$), respectively, were used as these are the typical range found in the settled surface water in the developing countries [17]. Coagulant dosage of 3 mg Al/L (-1) to 6 mg Al/L ($+1$) were used as these doses were the approximate optimum coagulant doses found from the preliminary tests for the raw waters used in this study. The experiments were performed in randomised order to minimise possible systematic errors.

Sludge preparation and aluminium recovery

Different sets of raw water were prepared by spiking required amounts of humic acid (HA) and turbidity in 20 L tap water (pH = 7.9, alkalinity = 62 mg/L as $CaCO_3$ and hardness = 36 mg/L as $CaCO_3$) as given in Table 1. Sludge samples were prepared after coagulation of raw water with a certain amount of alum or PACl. The coagulation procedure comprised of flash mixing for 2 min followed by slow mixing for 30 min and 30 min of settling. The sludge settled at the bottom of the container was collected and was oven dried at 105 °C for 24 h. It was crushed with mortar pestle and aluminium recovery was performed at pH 2.

For aluminium recovery, 1.0 gm of powdered sludge was added to 100 mL deionised water and the pH of the solution was adjusted to 2.0 using 1 N sulphuric acid. The solution was then mixed for 30 min at 100 rpm and then kept quiescent for 15 min. These conditions were selected based on the reported studies. Cheng et al. [11] reported that when mixing speed increased beyond 80 rpm the film diffusion control did not influence the Al leaching rate. Similarly, 30 min reaction time and pH 2 was found optimum for maximum Al recovery [7, 13]. After 15 min of quiescent condition, solutions were analysed for Al concentration. The aluminium recovery was calculated as:

Table 1 Factorial design for aluminium recovery from synthetic sludge

Run	Humic acid (mg/L) (A)	Coagulant dose (mg Al/L) (B)	Turbidity (NTU) (C)	Observed Al recovery (%)	
				Alum sludge	PACl sludge
1	1 (−1)	3 (−1)	25 (−1)	68.36	76.94
2	5 (+1)	3 (−1)	25 (−1)	62.55	70.67
3	1 (−1)	6 (+1)	25 (−1)	74.54	83.99
4	5 (+1)	6 (+1)	25 (−1)	65.03	73.22
5	1 (−1)	3 (−1)	50 (+1)	67.88	76.19
6	5 (+1)	3 (−1)	50 (+1)	62.94	70.87
7	1 (−1)	6 (+1)	50 (+1)	70.73	79.84
8	5 (+1)	6 (+1)	50 (+1)	61.25	68.88

Numbers in parentheses represent the coded values

$$\text{Al recovery efficiency (\%)} = \frac{\text{Al in supernatant after acidification (mg Al/g dry sludge)}}{\text{Al in raw sludge after acid digestion (mg Al/g dry sludge)}} \times 100 \quad (1)$$

Analysis

Aluminium concentration was measured using inductively coupled plasma atomic emission spectrometry (ICP-AES) (ARCOS Spectro, Germany). The raw water characteristics were analysed as per standard methods [18]. pH was measured using a pH metre (Hanna 209) and turbidity was measured using turbidimeter (Hach 2100P). The chemicals (RANKEM Fine Chemicals Ltd., India) used were of analytical grade. Design and statistical analysis of the 2^3 factorial experiments were carried out with Design Expert v 8.0 (Stat-Ease, USA) DoE package.

Results and discussion

Aluminium recovery

$Al(OH)_3$ was the dominant species present in the WTS flocs which reacted with sulphuric acid to produce aluminium sulphate. The chemical reaction occurring in the acidification process is expressed by the following equation [6]:

$$2Al(OH)_3 + 3H_2SO_4 \rightarrow Al_2(SO_4)_3 + 6H_2O \quad (2)$$

The percentage of Al recovered from alum and PACl sludge is shown in Table 1. As observed from Table 1 the best combination of the factors for highest Al recovery from WTR was from run 3 where humic acid concentration and turbidity was the lowest and coagulant dose was highest. This result was applicable for both alum and PACl sludge. It was also observed that Al recovery from PACl sludge was higher than the one from alum sludge by 7–10 % (Figs. 1, 2, 3). Previous studies have also reported higher Al recovery from PACl sludge compared to alum sludge [7, 11]. This may be due to the PACl-induced interparticle bridging resulting in more aluminium ions being adsorbed on the solid phase of sludge and released by acidification [11]. However, the acidification of WTS to recover Al is non-selective process and other impurities present in WTS are also dissolved into the solution along with Al [11].

Main effects

The main effect plots of each factor on the Al recovery efficiency from alum and PACl sludge are shown in Figs. 2 and 3 respectively, where *A* represents humic acid concentration, *B* represents coagulant dose and *C* represents turbidity. These plots were generated in order to present the results of the regression analysis. The deviations of the mean values for each factor are also presented in the main effect plots. From Figs. 2 and 3 it can be seen that humic acid concentration, when compared to turbidity and coagulant dose, had higher influence on the Al recovery efficiency from both alum and PACl sludge. The effects of humic acid concentration and turbidity were negative, that is Al recovery efficiency reduced when these factors move from low level to high level. The opposite was true for the coagulant dose. The presence of organic matter may offer an inert surface coating on the sludge and thus obstructs alum resolubilization [19, 20]. Li et al. [9] reported that the organic matter present in the sludge might consume H^+ through protonation reaction thus reducing the Al recovery efficiency. The presence of inert solids causing turbidity may have limited the reagent contact which reduced the Al recovery efficiency.

Interaction effects

Interaction plots are effective to understand the change in response of a factor from low to high level depending on

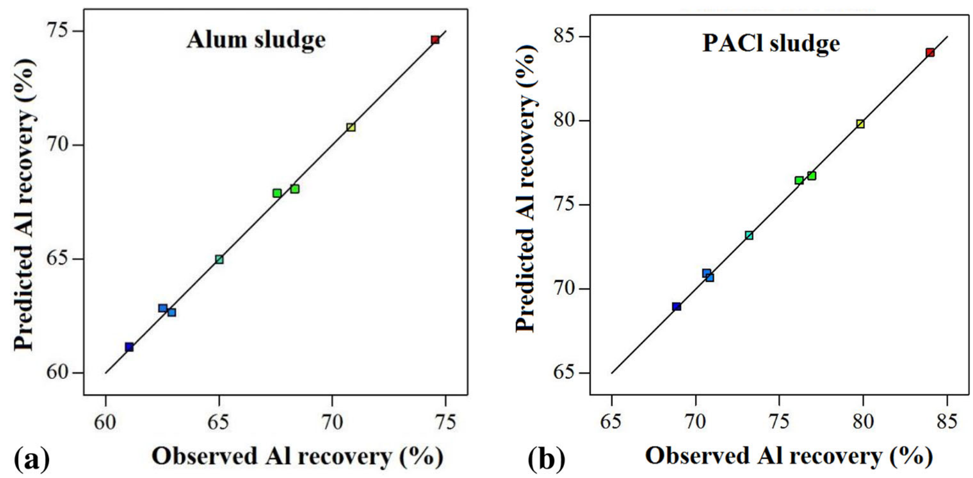

Fig. 1 Observed and predicted Al recovery from (**a**) alum and (**b**) PACl sludges

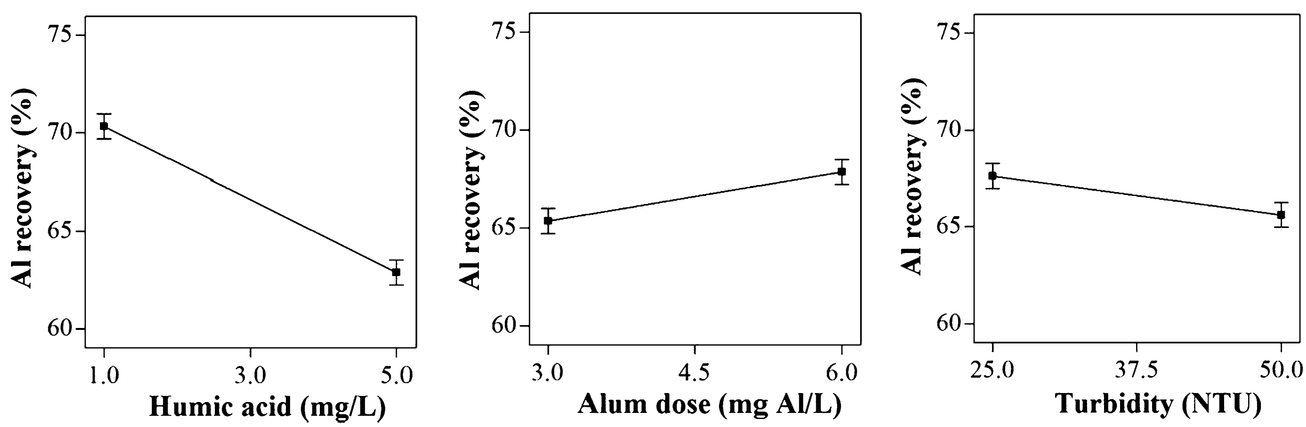

Fig. 2 Main effects of the factors on Al recovery efficiency from alum sludge

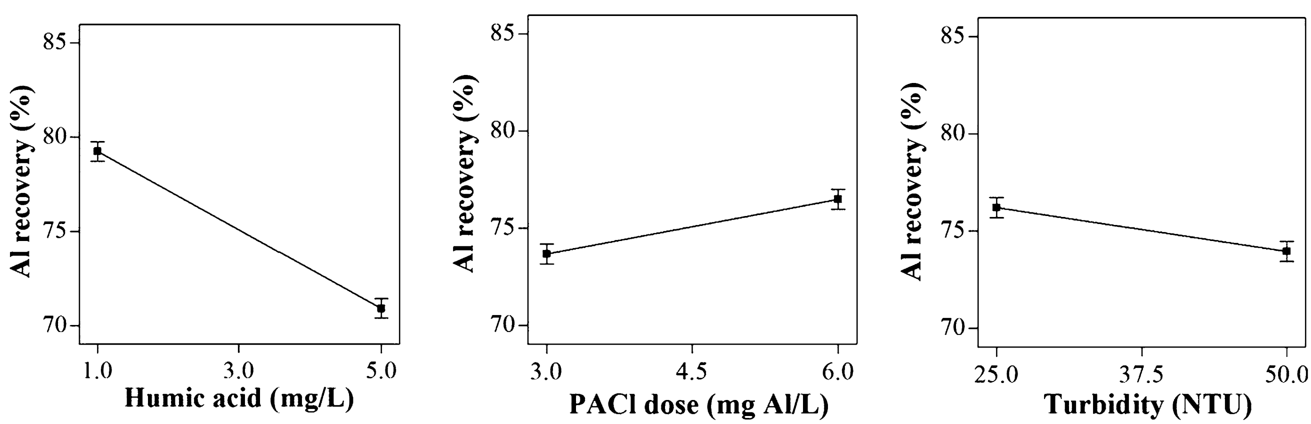

Fig. 3 Main effects of the factors on Al recovery efficiency from PACL sludge

the level of a second factor [16]. Figures 4 and 5 show interaction between the factors *AB* and *BC* for alum and PACl sludge, respectively, where *AB* represents humic acid concentration–coagulant dose interaction, *BC* represents coagulant dose–turbidity interaction and *AC* represents humic acid concentration–turbidity interaction effect on Al recovery. These plots clearly indicated that interaction between humic acid and coagulant dose (*AB*) was stronger than the interaction between coagulant dose and turbidity (*BC*). The interaction between humic acid and turbidity

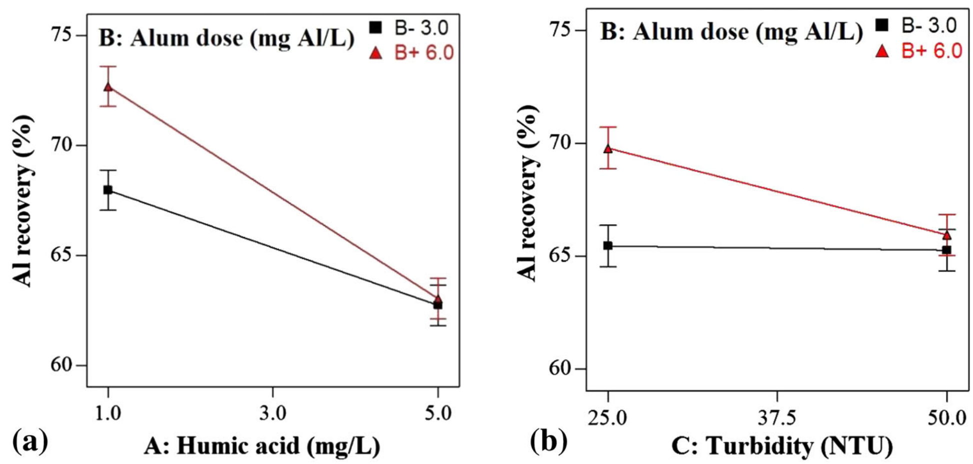

Fig. 4 Interaction plots of Al recovery from alum sludge

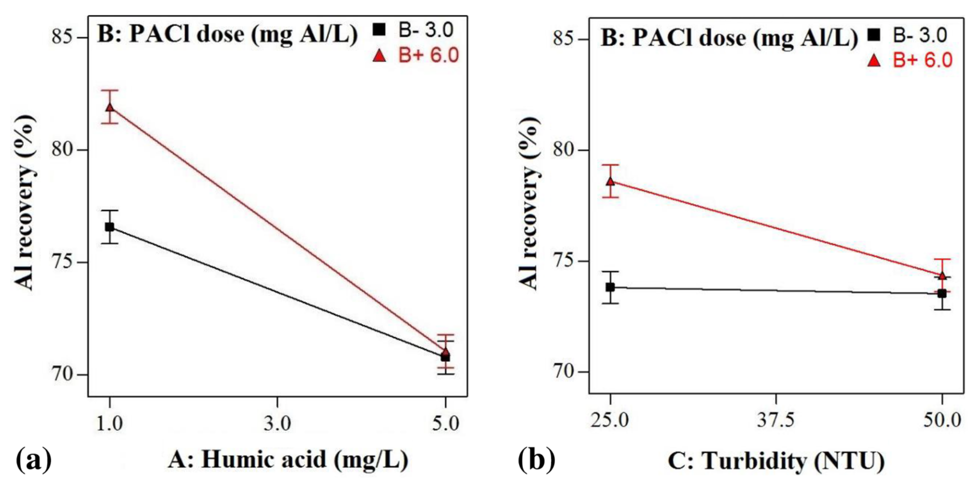

Fig. 5 Interaction plots of Al recovery from PACl sludge

(*AC*) was not statistically significant and hence not presented. The interaction plots shown in Figs. 4 and 5 also suggested that compared to lower coagulant dose, at higher coagulant dose, the effect of humic acid and turbidity was more remarkable on Al recovery from both alum and PACl sludge.

2^3 factorial regression models

The factors which are significant in the regression model were determined by carrying out an analysis of variance (ANOVA) of the data from Table 1. ANOVA results for Al recovery from alum and PACl sludge along with with the p values for each factor are presented in Tables 2 and 3. The factors with p value greater than 0.05, are considered to be statistically insignificant at 95 % confidence interval [14].

The final model after excluding the statistically insignificant terms for Al recovery from alum and PACl sludge is presented in Eqs. (2) and (3). There exist a statistical relation between the response and the factors selected at 95 % confidence level as the p values for both the models were lower than 0.05.

$$\begin{aligned}&\text{Al recovered (alum) (\%)}\\&= 66.66 - 3.72A + 1.23B - 0.96C - 1.03AB - 0.94BC\end{aligned} \quad (3)$$

$$\begin{aligned}&\text{Al recovered (PACl) (\%)}\\&= 75.08 - 4.17A + 1.41B - 1.13C - 1.27AB - 0.99BC\end{aligned} \quad (4)$$

where A = humic acid concentration, B = coagulant dose and C = turbidity.

Equations (3) and (4) described how the experimental factors and their interactions influence the Al recovery from alum and PACl sludge, respectively. Humic acid concentration (A) had the greatest effect on Al recovery process followed by coagulant dose (B), humic acid concentration–coagulant dose interaction (AB), turbidity (C) and coagulant dose–turbidity interaction (BC) in that order for both the sludge. The positive sign of the factors indicate that they

Table 2 ANOVA table for aluminium recovery from alum sludge

Source	Sum of squares	Degree of freedom	Mean square	*F* value	*p* value	
Model	145.63	5	29.13	304.87	0.0033	Significant
A—Humic acid	110.65	1	110.65	1158.27	0.0009	
B—Alum dose	12.11	1	12.11	126.71	0.0078	
C—Turbidity	7.35	1	7.35	76.97	0.0127	
AB	8.47	1	8.47	88.65	0.0111	
BC	7.05	1	7.05	73.76	0.0133	
AC	0.072	1	0.072	0.45	0.6227	

$R^2 = 0.9987$, $R^2_{\text{adjusted}} = 0.9954$, $R^2_{\text{predicted}} = 0.9790$

Table 3 ANOVA table for aluminium recovery from PACl sludge

Source	Sum of squares	Degree of freedom	Mean square	*F* value	*p* value	
Model	185.59	5	37.12	321.68	0.0031	Significant
A—Humic acid	138.84	1	138.84	1203.21	0.0008	
B—PACl dose	15.81	1	15.81	137.03	0.0072	
C—Turbidity	10.21	1	10.21	88.51	0.0111	
AB	12.86	1	12.86	111.45	0.0089	
BC	7.87	1	7.87	68.21	0.0143	
AC	0.10	1	0.10	0.39	0.6450	

$R^2 = 0.9988$, $R^2_{\text{adjusted}} = 0.9957$, $R^2_{\text{predicted}} = 0.9801$

have constructive effect on the Al recovery process while the negative sign of the factors indicate that they have adverse effect. Accordingly, the coagulant dose had positive effect while humic acid concentration and turbidity had negative effect on the Al recovery process. Equations (2) and (3) facilitate the prediction of Al recovery (%) from alum and PACl sludge as a function of humic acid concentration, coagulant dose and turbidity along with their interactions.

The coefficient of determination (R^2) describes the overall prediction performance of the model and should be close to 1.0 [14, 15]. R^2 is the ratio of sum of square of regression to total sum of squares [14, 15]. However, R^2 increases with the addition of model terms which may not be significant. Hence R^2_{adjusted} should be considered for model adequacy check. The sum of square used in R^2 calculation is divided by degree of freedom to obtain R^2_{adjusted}. The value of R^2_{adjusted} often reduces if statistically insignificant variables are included in the model [15]. Tables 2 and 3 shows high R^2 values of 0.99 for both the models. In addition, the models had a high R^2_{adjusted} value of 0.99, fitting the statistical model quite well. The prediction ability of the model for new responses is described by $R^2_{\text{predicted}}$. Hence a large variation between R^2_{adjusted} and $R^2_{\text{predicted}}$ cannot be accepted. Tables 2 and 3 show that R^2, R^2_{adjusted} and $R^2_{\text{predicted}}$ are in close agreement. Extensive details of the model adequacy check can be referred from the literatures [14, 15]. Similarly Fig. 1 showed that the experimental data and the predicted data do not differ significantly for both the models. Accordingly the models were found suitable for prediction within the range of the selected factors.

It may be noted that the amount and composition of WTS generated from a water treatment plant vary because of the variations in raw water quality, coagulant used, dosage applied and treatment efficiency. High levels of colour and turbidity in the source water required higher coagulant dosages, influencing sludge quantity and quality. Hence the characteristics and quantities of WTS generated vary from treatment plant to plant and also within the treatment plant. The present study considered three parameters, humic acid concentration, turbidity and coagulant dosage on aluminium recovery from the sludge formed. The empirical models developed in this study clearly depicted the effect of these parameters on the recovery. More parameters can be included in further studies which would result in improved models. Controlled laboratory experiments were conducted in the present study, and more studies based on the field data should also be conducted to optimise the recovery process.

Conclusions

The present study compared the aluminium recovery from sludge prepared with alum and PACl coagulants using raw water with different characteristics and coagulant dosages.

Controlled laboratory experiments, with design of experiments approach was used to prepare sludges from raw water with different turbidity and humic acid concentration and coagulant dosages. The results of the study showed that the Al recovery efficiency was higher from PACl sludge than from alum sludge. Increase in organic matter (humic acid) and turbidity reduced the Al recovery efficiency from the sludge for both the coagulants while increase in coagulant dose increased the Al recovery efficiency. Compared to lower coagulant dose, at higher dose, the effect of humic acid and turbidity was more remarkable on Al recovery from both alum and PACl sludge. The models developed in the study can be used for predicting aluminium recovery from sludges produced from raw waters of different quality.

References

1. Babatunde AO, Zhao YQ (2007) Constructive approaches toward water treatment works sludge management: an international review of beneficial re-uses. Crit Rev Environ Sci Technol 37:129–164
2. Keeley J, Jarvis P, Judd SJ (2014) Coagulant recovery from water treatment residuals: a review of applicable technologies. Crit Rev Environ Sci Technol 44:2675–2719
3. Muisa N, Hoko Z, Chifamb P (2011) Impacts of alum residues from Morton Jaffray Water Works on water quality and fish, Harare, Zimbabwe. Phys Chem Earth Parts A/B/C 36:853–864
4. Abdo MSE, Ewida KT, Youssef YM (1993) Recovery of alum from wasted sludge produced from water treatment plants. J Environ Sci Health Part A Environ Sci Eng Toxicol Toxic/Hazard Subst Environ Eng 28:1205–1216
5. Wu CH, Lin CF, Chen WR (2004) Regeneration and reuse of water treatment plant sludge: adsorbent for cations. J Environ Sci Health Part A Toxic Hazard Subst Environ Eng 39:717–728
6. Cheng WP, Fu CH, Chen PH, Yu RF (2012) Dynamics of aluminium leaching from water purification sludge. J Hazard Mater 217–218:149–155
7. Nair AT, Ahammed MM (2014) Coagulant recovery from water treatment plant sludge and reuse in post-treatment of UASB reactor effluent treating municipal wastewater. Environ Sci Pollut Res 21:10407–10418
8. Ishikawa S, Ueda N, Okumura Y, Lida Y, Baba K (2007) Recovery of coagulant from water supply plant sludge and its effect on clarification. J Mater Cycles Waste Manag 9:167–172
9. Li C-W, Lin J-L, Kang S-F, Liang C-L (2005) Acidification and alkalization of textile chemical sludge: volume/solid reduction, dewaterability, and Al(III) recovery. Sep Purif Technol 42:31–37
10. Huang S, Chen J-L, Chiang K-Y, Wu C-C (2010) Effects of acidification on dewaterability and aluminum concentration of alum sludge. Sep Sci Tech 45:1165–1169
11. Chen YJ, Wang WM, Wei MJ, Chen JL, He JL, Chiang KY, Wu CC (2012) Effect of Al-coagulant sludge characteristics on the efficiency of coagulant recovery by acidification. Environ Tech 33:2525–2530
12. Xu GR, Yan ZC, Wang YC, Wang N (2009) Recycle of alum recovered from water treatment sludge in chemically enhanced primary treatment. J Hazard Mater 161:663–669
13. Xu GR, Yan ZC, Wang N, Li GB (2009) Ferric coagulant recovered from coagulation sludge and its recycle in chemically enhanced primary treatment. Water Sci Technol 60:211–219
14. Montgomery DC (2010) Design and analysis of experiments, 7th edn. Wiley India Pvt, New Delhi
15. Nair AT, Makwana AR, Ahammed MM (2014) The use of response surface methodology for modelling and analysis of water and wastewater treatment processes: a review. Water Sci Technol 69:464–478
16. Bingol D, Tekin N, Alkan M (2010) Brilliant Yellow dye adsorption onto sepiolite using a full factorial design. Appl Clay Sci 50:315–321
17. Nair AT, Ahammed MM, Davra K (2014) Influence of operating parameters on the performance of a household slow sand filter. Water Sci Technol Water Supply 14:643–649
18. American Public Health Association (APHA), American Water Works Association (AWWA), Water Environmental Federation (WEF) (1998) Standard methods for the examination of water and wastewater, 20th edn (Clescerl L, Greenberg A, Eaton A, eds). American Public Health Association, Washington, DC
19. Wang LK, Yang JY (1975) Total waste recycle system for water purification plant using alum as primary coagulant. Resour Recover Conserv 1:67–84
20. Okuda T, Nishijima W, Sugimoto M, Saka N, Nakai S, Tanabe K, Ito J, Takenaka K, Okada M (2014) Removal of coagulant aluminum from water treatment residuals by acid. Water Res 60:75–81