

Performance intensification of the coagulation process in drinking water treatment

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Surface water pollution has increased, owing to industrial development and population growth. Consequently, it is important to find alternative drinking water treatment strategies, which cater for changes in the quality of raw water. This study compared the efficiency of different coagulants in treating raw water that feeds a drinking water treatment plant (WTP). Using jar testing equipment and a number of physicochemical parameters, an investigation was conducted to establish optimum conditions for aluminium chloride (A), ferric chloride (B), and chitosan (C), and their performance compared with aluminium sulphate (D), which is the coagulant used at the WTP. The turbidity removal efficiencies for the single coagulants were in the order: B (95.7%) > A (94.7%) > C (94.4%), at optimum coagulant doses of 60, 50, and 0.6 mg/L, respectively. The coagulants achieved high removal efficiencies for turbidity, total dissolved solids (TDS), ultraviolet absorbance at 254 nm (UV₂₅₄) and conductivity under acidic conditions. For dual coagulants, there was a gradual increase in the removal efficiencies of the tested parameters with increasing pH. Combined coagulants were more effective compared to single coagulants, with highest removal efficiencies being exhibited by the A/C combination. Overall, the coagulants proved to be suitable alternatives to D, since they had comparable performances.

INTRODUCTION

Factors such as population growth and economic development have resulted in elevated concentrations of pollutants in water bodies (Wang and Yang, 2016; Zhao et al., 2018; Lv et al., 2019). A variety of natural phenomena, like weathering, volcanicity, and human activities such as agriculture, mining, and manufacturing, produce pollutant-laden effluents that contaminate water bodies (Howladar, 2017; Rashid et al., 2019). Poor microbial water poses a threat to human health as this can potentially cause water-borne diseases like cholera, dysentery and typhoid (Wang and Yang, 2016; Rashid et al., 2019). In addition, toxic metals are cumulative toxins with public health risks. Consequently, surface water needs to be treated in order to produce safe drinking water.

The main stages of the conventional water treatment process are coagulation, flocculation, sedimentation, filtration, and disinfection (Bairwa, 2016). Coagulation is the destabilization of suspended particles using inorganic salts or polymers so that they may aggregate and form larger flocs (Renault et al., 2009; Wei et al., 2018). The coagulation process removes turbidity, organic particles, and suspended particles in water, and its efficiency depends on the characteristics of raw water, and the coagulant type and dosage used (Alshikh, 2007; Sillanpaa et al., 2018; Lv et al., 2019). Coagulation parameters such as pH and coagulant dose differ for each water source; hence the optimization of coagulants is necessary for full-scale implementation. The various pollutants in raw water can decrease the efficiency of coagulants; therefore it is important to characterize raw water so as to determine the optimum coagulation conditions (Yonge, 2011; Lv et al., 2019).

Iron-based and aluminium-based coagulants are commonly used in water treatment processes (Justina et al., 2018). In aqueous solution, iron and aluminium salts dissociate and react to form hydrolysis products that act as the coagulant species responsible for removing turbidity and colour (El Samrani et al., 2004). Other materials such as chitosan are efficient in removing turbidity and suspended particles in water (Renault et al., 2009; Loganathan et al., 2018). The advantage of each coagulant may be maximized by combining different coagulants (Renault et al., 2009). Comparison data on the performance of chitosan, aluminium chloride and ferric chloride, either singly or in combination, basing on real raw water quality in Zimbabwe is scarce. Hence there is a need to generate information based on raw water quality of local water treatment plants (WTPs). Owing to the vast pollutant burden in raw water, WTPs are operating under compromised efficiency, necessitating the investigation of the performance of alternative coagulants. Combining coagulants allows for the treatment process to take advantage of the different properties and efficiencies of each coagulant, and this can potentially result in synergistic performance (Xu et al., 2018). Therefore, there is a need to investigate the optimum addition sequences to achieve optimum pollutant removal.

A previous study demonstrated that adding polyferric chloride before polyamine favoured efficient colour removal for synthetic disperse yellow 201, while the reverse dosing order favoured synthetic reactive red 24 (Chen et al., 2010). However, optimal colour removal efficiency (97.5%) was achieved at pH 6 for both dyes. Another study compared the performance of aluminium and ferric-based coagulants as pre-treatment for UVC/H₂O₂ treatment of wastewater (Umar et al., 2016).

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DATES

Received: 5 June 2020

Accepted: 15 April 2021

KEYWORDS

chitosan
coagulation
ferric chloride
raw water
water pollution

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The results indicated that ferric-based coagulants were generally more effective than aluminium sulphate. Other researchers have investigated the effectiveness of individual and dual coagulants in the removal of microalgae from seawater using chitosan-aluminium sulphate/ferric chloride (Loganathan et al., 2018). Ferric chloride coagulants had superior process performance in comparison to aluminium sulphate and chitosan-based individual coagulants. In terms of dual coagulation, using aluminium sulphate as coagulant and chitosan as flocculant improved microalgae removal efficiency.

Although numerous studies have been reported on the use of coagulants, there are no comparative studies on their performance on raw water, particularly in Zimbabwe. This study aimed to investigate optimum conditions for ferric chloride, chitosan and aluminium chloride in raw water samples from a local WTP, and compare their performance with aluminium sulphate, which is used at the WTP. The specific objectives were: (i) to determine the optimum dosage and pH for ferric chloride, aluminium chloride and chitosan, singly and in combination, (ii) to determine the most effective dosage strategy for combined coagulants (order of adding coagulants), and (iii) to compare the performance of the coagulants with that of aluminium sulphate.

MATERIALS AND METHODS

Materials

Chemical reagents used in the study were of analytical grade and were used without further purification. Aluminium chloride (A) (99%, Associated Chemical Enterprises), ferric chloride (B) (66%, Associated Chemical Enterprises), and chitosan (C) (LMW, Sigma Aldrich) were used as coagulants in this study. The acids used were acetic acid (99%, Merck), hydrochloric acid (32%, Glassworld), and nitric acid (55%, Skylabs), and the alkali was sodium hydroxide (98%, Acechem). Calcium carbonate (99%, Acechem) and magnesium metal (99%, Glassworld) were used to prepare stock solutions for determining the water hardness.

Sampling and preparation of coagulant stock solutions

The study area was described in detail elsewhere (Chaukura et al., 2020). Briefly, the WTP is fed from a river and two dams, uses the conventional water treatment process, and has a daily output of 14.4 ML. On average, the raw water had a pH of 6.65, conductivity 307 $\mu\text{S}/\text{m}$, turbidity 15.34 NTU, and total dissolved solids (TDS) 156 mg/L. Using 5-L HDPE bottles, water samples were collected from the raw water feed point at the WTP. The water samples were transported to the laboratory for experimentation and stored at 4°C. Prior to jar test experiments, stock solutions of the coagulants were freshly prepared by dissolving a certain amount of coagulant in a suitable solvent, and making up to the mark in a 1 000 mL volumetric flask. Specifically, 1 000 mg/L stock solution of A (pH = 2.6) was prepared using 1 g of AlCl_3 , while B (1 000 mg/L, pH = 2.1) was prepared using 1 g of FeCl_3 . A stock solution of C (100 mg/L, pH = 2.5) was prepared by dissolving 0.1 g of chitosan in 1% acetic acid and topping up to the mark in a 1 000 mL volumetric flask with de-ionized water.

Jar tests

The coagulation experiments were carried out on raw water samples in triplicate using jar testing apparatus (Fig. 1a) following the method described by Ayekoe et al. (2017). Briefly, each jar was filled with 500 mL of raw water and a specified amount of coagulant stock solution was added to each jar, resulting in the desired dosage (40, 50, 60, 70, 80, 90 mg/L for A and B, and 0.2, 0.4, 0.6, 0.8, 1.0, 1.2 mg/L for C) at room temperature. The choice

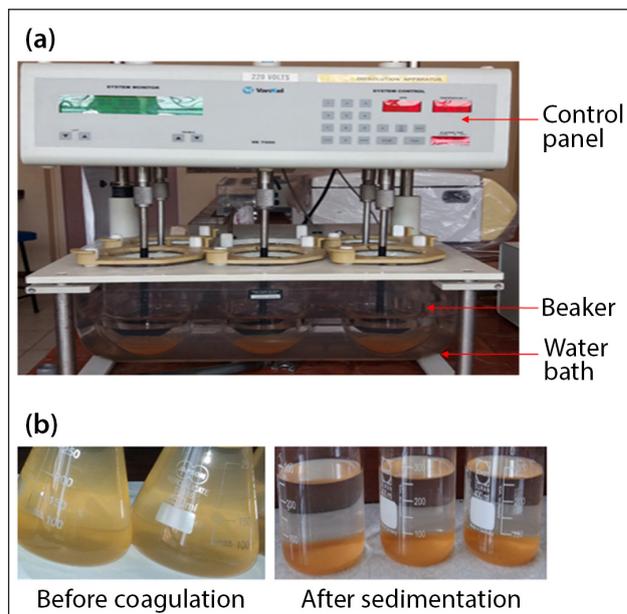


Figure 1. Coagulation experiment set up showing (a) jar-testing apparatus, and (b) water sample before coagulation and after sedimentation

of experimental values was guided by literature (Ammary and Cleasby, 2004) and preliminary studies not reported here. The samples were mixed rapidly at a velocity gradient of 10^2 s^{-1} for 3 min, following which the velocity gradient was reduced to $2 \times 10^1 \text{ s}^{-1}$ for a further 15 min. Thereafter the flocs were allowed to settle for 30 min (Fig. 1b), and an aliquot of the supernatant solution was withdrawn from approximately 3 cm below water surface, filtered and analysed for different parameters.

The optimum pH for the coagulants was determined by applying a fixed coagulant dose to the raw water basing on the results obtained after the dosage optimization. The optimum coagulant dosage obtained at the coagulation stage was used as the fixed dosage. The pH of the solution at optimum dosage for A, B, and C was 7.09, 7.13 and 6.97, respectively. The pH of raw water was adjusted to 4, 5, 6, 7, 8 and 9 using 0.1 M HCl or 0.1 M NaOH as appropriate. The same procedure used for dosage and pH optimization was used for combined coagulants. The dual coagulants were added in succession, 2 min apart at the rapid mixing stage. The coagulants were added in different orders and the operations coded differently. For example, when A was added before B the process was coded A/B, and B/A when B was fed before A. Different coagulation combinations were investigated (Table 1).

The removal efficiencies (r) for the water parameters in the study were calculated using Eq. 1 (Ang et al., 2016):

$$r = \frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

where C_i and C_f are the initial concentration of parameters in untreated, and the final concentration in treated water samples, respectively.

Table 1. Experimental design for coagulant combinations

	A	B	C
A	A/A	B/A	C/A
B	A/B	B/B	C/B
C	A/C	B/C	C/C

A = aluminium chloride, B = ferric chloride, C = chitosan

Analytical methods

To assess the performance of coagulants, the physicochemical characteristics of water were determined before and after the coagulation process for each run. The turbidity and pH were measured using a turbidity meter (Orbeco-Hellige, 966, USA), and pH meter (pH 700, Eutech Instruments, Singapore), respectively. Conductivity and TDS were determined using a multimeter (Cyberscan con 11, Eutech Instruments, Singapore), and optical absorbances were measured in the wavelength range 200–850 nm using a UV-Vis spectrophotometer (EVO300PC, Thermo Fisher Scientific, USA). Water hardness (Ca and Mg) was determined using a flame atomic absorption spectrometer (FAA-6701F, Shimadzu, Japan).

Statistical analysis

Optimization studies were carried out by investigating the effect of 2 variables – coagulant dosage and pH – using two-way analysis of variance. Minitab 18 software was used to design and analyse the response, in order to measure the effect of turbidity on the coagulation process. The coagulation process was optimized using response surface methodology (RSM) and the adequacy of the optimization process was evaluated using analysis of variance (ANOVA).

RESULTS AND DISCUSSION

Effect of coagulant dosage

The turbidity removal efficiency for the coagulants increased with increasing coagulant dosage until it reached a plateau, beyond which there was a decrease in the removal efficiencies (Fig. 2). This decrease was attributed to the re-stabilization of colloidal particles when coagulant dosage exceeded the optimum value (Sun et al., 2010). The optimum dosages for A, B, and C were 50, 60 and 0.6 mg/L, achieving turbidity removal efficiencies of 94.7,

95.7 and 94.4%, respectively. In all cases, the conductivity and TDS removal efficiencies decreased gradually as the coagulant dose increased. Since ionic species act as charge carriers in solution, a drop in conductivity confirmed removal of these ions by the coagulation process. Similar observations are reported in literature (Matilainen et al., 2010). The UV₂₅₄ removal efficiency increased moderately up to 60 mg/L as the dosage of A increased, then decreased slightly thereafter (Fig 2a). There were fluctuations in the UV₂₅₄ removal efficiencies for B and C (Fig. 2b,c) as the doses were increased. Coagulant C achieved a UV₂₅₄ removal efficiency of 58.2% with an optimum dosage of 0.6 mg/L, while B attained a UV₂₅₄ removal efficiency of 62.2% with a dosage of 50 mg/L. UV₂₅₄ is an important parameter for characterizing aromatic organics in water (De Oliveira et al., 2018). The results indicate that the coagulants can effectively reduce the concentration of aromatic compounds in the source water.

For dual coagulants, the order of performance in turbidity removal was: turbidity: A/C > A/B > C/B > B/A > C/A > B/C, conductivity was removed in the order: C/B > A/C > C/A > B/C > A/B > B/A, TDS: B/C > C/A > C/B > A/C > A/B > B/A, and UV₂₅₄: B/C = B/A > A/B = A/C > C/A > C/B. According to the findings, the order in which coagulants are added has an effect on the coagulation performance. Addition of inorganic coagulants prior to polymers neutralizes the negative suspended particles, leading to the formation of micro-flocs which are then adsorbed to the polymer particles (Ammary and Cleasby, 2004; Sun et al., 2010). However, the addition of polymeric coagulants before inorganics provides a base structure for the formation of flocs while the inorganic coagulants remove the remaining particles (Ammary and Cleasby, 2004). Overall, the results indicated that best performance was achieved by adding inorganic coagulants first, followed by the polymeric coagulant (C), with conductivity being the exception. Chitosan is reported in literature to be a good coagulant aid (Hesami et al., 2014).

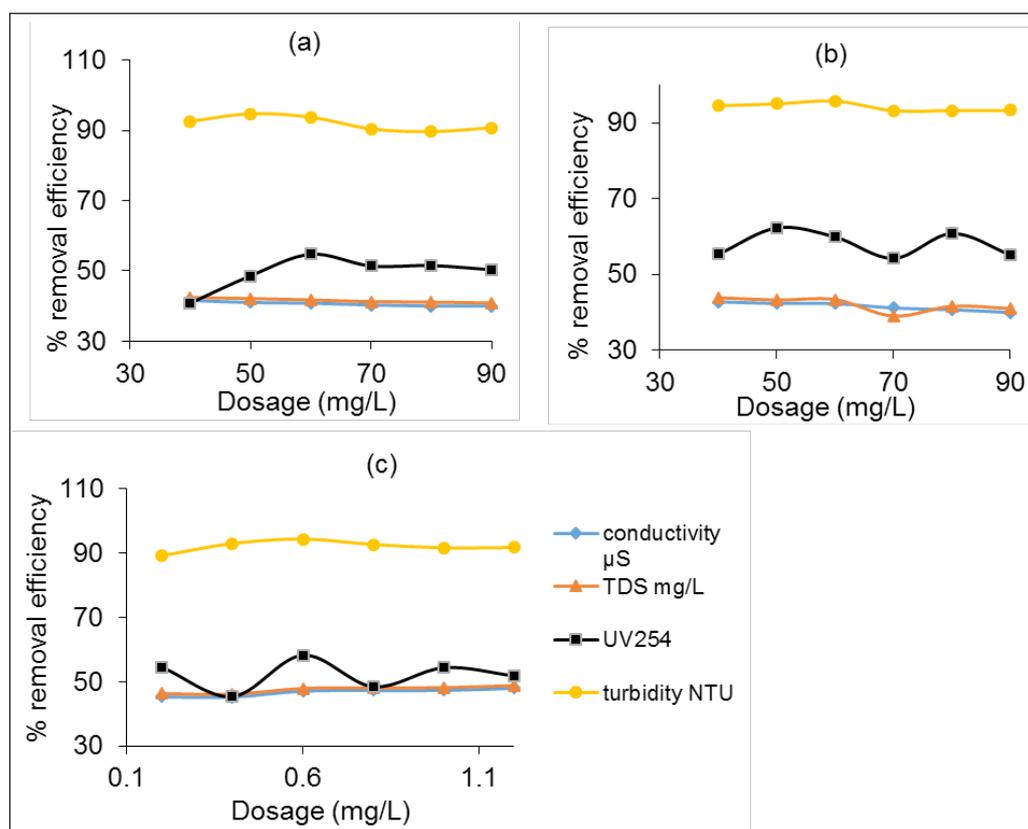


Figure 2. Effect of coagulant dose change on turbidity, UV₂₅₄, conductivity and TDS removal efficiencies using (a) A, (b) B, and (c) C

Effect of pH on coagulation

The raw water pH was 6.65. There was a decrease in pH with increasing dosage for single coagulants (Fig. 3a,b), while for dual coagulants, final pH was dependent on the order in which coagulants were added, especially for the inorganic/organic coagulant combinations (Fig. 3c). The final pH for dual coagulants rose as the initial pH was increased from 4 to 9. It is reported in literature that the resultant pH is dependent on the type of coagulant (inorganic or organic) and the dosage used. Coagulants (such as inorganics) which consume alkalinity can lead to a reduction in pH. When using dual coagulants, the order in which they are added plays a key role in the resultant pH (Naceradska et al., 2019).

According to WHO guidelines, the acceptable pH range for drinking water is between 6.5 and 8.5, hence the pH of water is usually adjusted to fall in this range (WHO, 2017). The final water pH decreased with increasing coagulant dosage for single coagulants (Fig. 4c). For dual coagulants, the pH after coagulation is consistently below this range. This could be due to the combined effect of metal-based coagulants in the case of A/B and B/A, and the combination of an organic coagulant and inorganic coagulant in the case of A/C, C/A, B/C, and C/B (Chen et al., 2018). It further points to the need for pH adjustment following coagulation. The decrease in supernatant pH when metal-based coagulants were used, is due to formation of hydrolysis species, leaving the H⁺ ions in solution (Yonge, 2011). The formation of hydroxide precipitates suggests that sweep flocculation was likely to be the main coagulation mechanism in this investigation (Eslami et al., 2019). Bridging was probably the main coagulation mechanism when chitosan was used since charge neutralization is supported by a rise in pH due to protonation of C, therefore consuming the H⁺ ions (Rustøen, 2015). The resulting supernatant pH for combined coagulants was in the range of 5.4 to 6.0 and the order for pH decrease was: B/C > A/B > C/B > A/C > C/A > B/A. The pH for treated samples in pH optimization studies fluctuated when single coagulants were used (Fig. 4a), while a steady increase was observed for dual coagulants (Fig. 4b).

Single coagulants

The turbidity removal efficiency for A (Fig. 5a) increased moderately as the pH of water was increased from 4 to 7, then decreased gradually thereafter. The optimum pH for A was 7. A similar trend was observed for B (Fig. 5b), which attained a turbidity removal efficiency of 95.6% at pH 6. Using C, the

turbidity removal efficiency decreased slightly between pH 4 and 5 (Fig. 5c) then increased gradually up to pH 7 before dropping again. Coagulant C is effective in acidic conditions (Rustøen, 2015), a result confirmed by this study. The UV₂₅₄ removal efficiency for A increased sharply between pH 4 and 5, while the opposite trend was observed for TDS and conductivity removals (Fig. 5d). Similar trends were observed for B and C in the pH range 4 to 6, and 4 to 5, respectively (Fig. 5e,f). The results suggest attachment of the polymer chains of C to the pollutant forming bridges, a process called bridging, could be the dominant coagulation mechanism for C, while aggregation through entrapment of particles, called sweep flocculation, dominated in the case of inorganic coagulants (Zhang et al., 2018; Eslami et al., 2019).

Dual coagulants

For the dual coagulants A/B, there was a gradual increase in turbidity, conductivity and TDS removal efficiencies as the pH of the water increased from pH 4 to 8. Beyond pH 8, there was a steady decrease in the removal efficiencies (Fig. 5d). Solution pH influences the charge of the solute, and this determines the efficiency of the coagulation process. The optimum pH for the dual coagulants was higher compared to either single coagulant. A similar trend was observed in the conductivity and TDS removal efficiencies using A/C combination (Fig. 5e). However, the highest turbidity removal efficiency (95.9%) was achieved at pH 6 for the A/C combination, perhaps because C is more effective under acidic conditions (Bergamasco et al., 2009; Lurling et al., 2007). With the B/C combination, the optimum pH was 7 (Fig. 5f). At this pH, the protonated amino groups of C interact with negatively charged components of the raw water allowing effective coagulation (Lurling et al., 2007). At alkaline pH there is interference from anions in water, which will surround the positively charged amino groups preventing coagulation (Lurling et al. 2007). Overall, combined coagulants performed better than single coagulants, most likely due to their synergistic effect.

One of the most important factors that influence coagulation performance as well as mechanisms is pH. When the pH of the system is acidic, the H⁺ may compete with the coagulants forming bonds with the negatively charged colloids, while at basic pH the OH⁻ ions may compete with the negatively charged colloids to bond with coagulant, hence reducing the concentration of coagulants which bond with colloids (Lek et al., 2018).

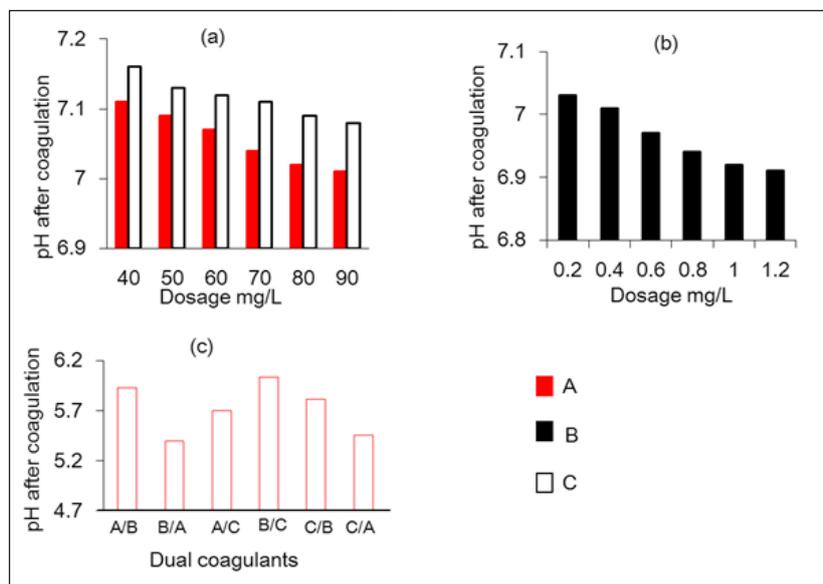


Figure 3. Changes in pH of water treated with single coagulants, (a) aluminum chloride and ferric chloride, and (b) chitosan; and dual coagulants, (c) A/B, A/C, and B/C

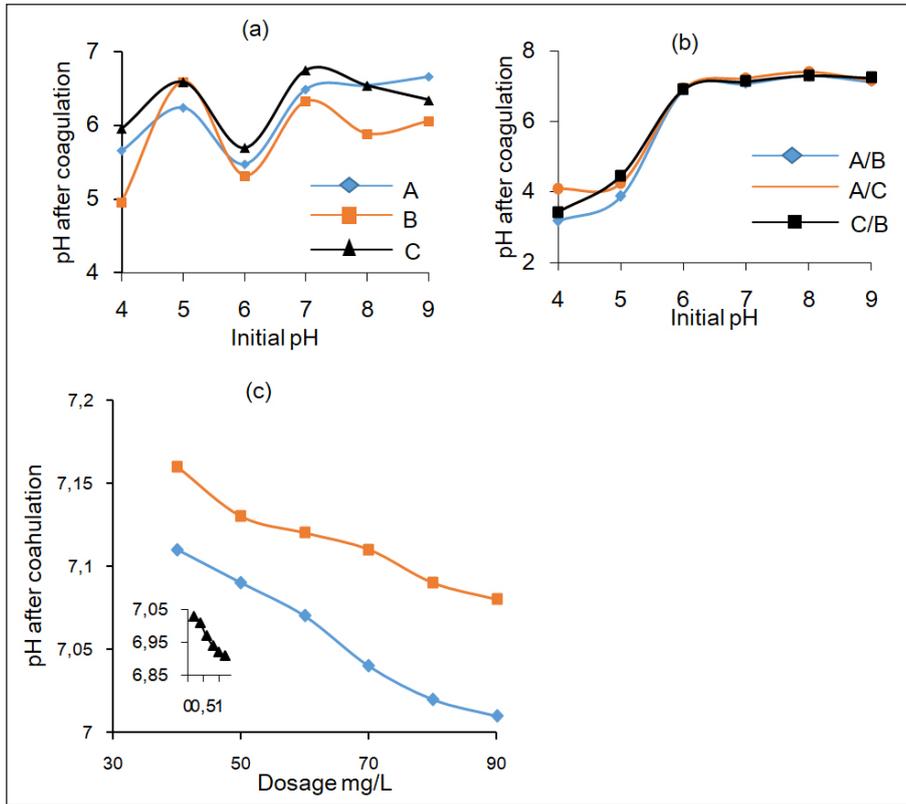


Figure 4. Changes in final pH with increase in initial pH using (a) single coagulants, (b) dual coagulants, and (c) the variation of pH with dosage

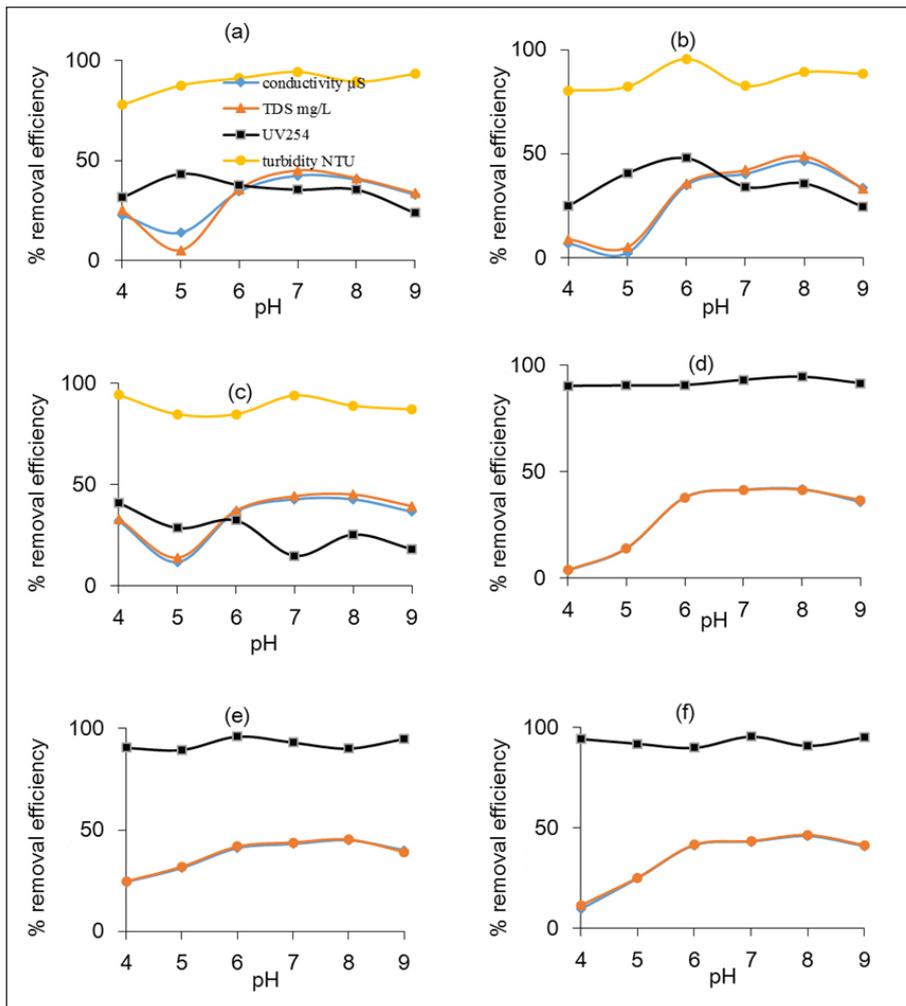


Figure 5. Effect of pH on turbidity, UV₂₅₄, conductivity and TDS removal efficiencies using single coagulants: (a) A, (b) B, and (c) C as coagulants, and dual coagulants: (d) A/B, (e) A/C, and (f) B/C

The pH of water also affects the hydrolysis products of coagulants; therefore, it has a significant influence on the coagulation process. Although the chemistry of ferric-based coagulants in aqueous systems is complex, since many species are generated at different pH values, solubility diagrams may possibly be useful in estimating the optimum coagulant dosage under different operating conditions (Johnson and Amirtharajah, 1983; Silanpaa et al., 2018). Ideally, the pH range for the hydrolysis of alum is 5.5–7.7 (Johnson and Amirtharajah, 1983). Under optimum pH, there is minimal solubility and most of the coagulant forms floc. At pH lower than optimum pH, positively charged dissolved Al species form. When the pH exceeds the optimal value, more negatively charged Al species are generated. Outside the optimal pH range, particularly at pH 3 and 11, the destabilization potential will be greatly lowered, hence the smaller particulates will not coalesce to form large flocs, but rather remain in solution (Silanpaa et al., 2018). It is intriguing to note that at an initial pH of 6 all coagulant reduces pH marginally but when dosed together in 5b at the same initial pH, they increase pH. This deserves further research.

Ultraviolet-visible absorbance

A steady decrease in UV-vis absorbance was observed from raw water to treated water for all samples, indicating the decrease in organic compounds during the coagulation processes. A decrease in UV-vis absorbance indicates removal of chromophoric substances such as organic matter, which contribute to turbidity (Matilainen et al., 2010). Different wavelengths represent different types of organic particles; for example a wavelength of 254 nm represents aromatic compounds, 272 nm shows the potential for DBPs formation and 456 nm indicates color (Ng, 2013; Tshindane et al., 2019). A pH of 6 resulted in the largest organic compound removal in the case of both A and B, while the least removal was observed at pH 9 (Fig. 6a,b). At this pH, most organic compounds are protonated, and can thus interact with the charged coagulants to form flocs (Chen et al., 2010). Figure 6c shows the results obtained for Coagulant C under pH optimization studies, where

pH 4 gave the least removal while the highest removal was observed at pH 7. Coagulant C does not carry an ionic charge, and therefore depends on dipole interactions with the organic matter (Ang et al., 2016). The dosage of 90 mg/L resulted in the largest decrease of organic content when Coagulant A was used, while the lowest removal was observed at 40 mg/L (Fig. 6d). For Coagulant B, the lowest removal was observed at a dosage of 40 mg/L while the highest removal efficiency was observed at 60 mg/L (Fig. 6e). The lowest removal efficiency for Coagulant C was observed 0.2 mg/L, while the highest removal was at 1.2 mg/L (Fig. 6f). This shows that Coagulant C is more effective than both Coagulants A and B, mainly because, being organic, Coagulant C has more interaction with small charged particles and facilitates aggregation into flocs more efficiently (Sillanpaa et al., 2018). Previous studies have used Coagulant C as a flocculant aid in combination with Flocculant A and B separately (Loganathan et al., 2018; Sillanpaa et al., 2018). For dual coagulants, the organic content removal varied with wavelength; therefore the results may be used to deduce the effective coagulant combination at a particular wavelength. In all the cases, pH 7 resulted in the highest organic content removal for dual coagulants during the pH optimization studies while pH 4 produced the lowest removal (Fig. 6g–i).

Coagulant performance evaluation

The order for removal efficiencies were (Table 2): turbidity: D > B > A > C. TDS: C > D > B > A; conductivity: C > B > D > A. UV₂₅₄: D > B > C > A. The optimum dose for D (80 mg/L) was higher than that of B (60 mg/L), and C (0.6 mg/L). Basing on the findings, the performance of A, B, and C were satisfactory and comparable with that of D. In a related study, other researchers established optimum coagulant doses of 40, 80, and 1 mg/L for aluminium sulphate, ferric chloride and chitosan, respectively, at pH 6. These variations in coagulant doses are related to the quality of raw water used. In their study, the raw water had high alkalinity (>180 mg CaCO₃/L) and low total organic carbon (TOC) (<4.0 mg/L) (Rizzo et al., 2008). Hence, it is of great importance

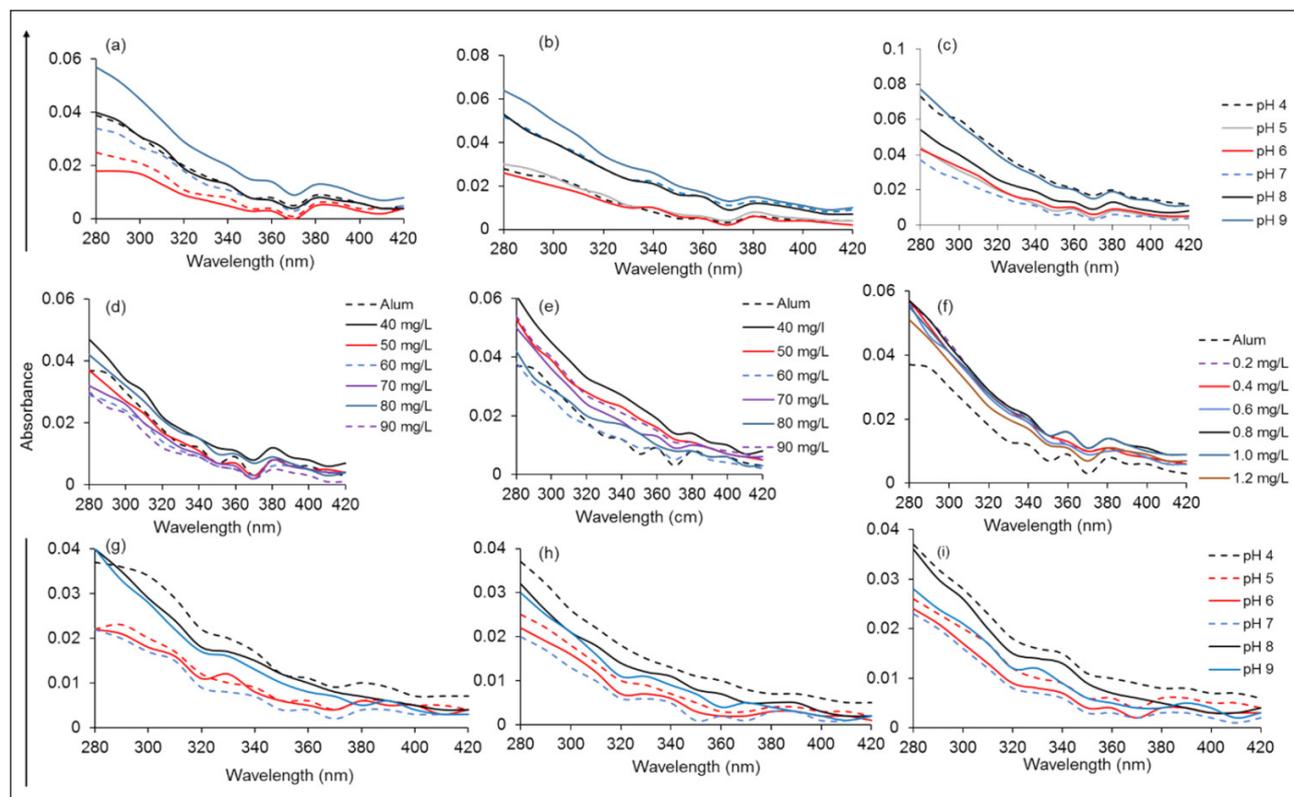


Figure 6. Ultraviolet absorbance spectra showing effect of pH using (a) A, (b) B, (c) C; dosage optimization for (d) A, (e) B, (f) C; and pH optimization for (g) A/C, (h) A/B, and (i) C/B

Table 2. Removal efficiencies (%) for Coagulants A, B, C, and D at their optimum dosages

	D	C	B	A
Dose mg/L	80.0	0.6	60.0	50.0
Turbidity (NTU)	96.7±0.42	94.4±0.38	95.7±0.33	94.7±0.40
TDS (mg/L)	45.8±0.30	47.9±0.26	43.3±0.41	42.3±0.35
Conductivity (µS)	41.7±0.32	47.1±0.37	42.2±0.35	41.0±0.39
UV ₂₅₄	73.5±0.44	58.2±0.41	59.8±0.36	48.6±0.34

to know the raw water quality in order to establish the most suitable conditions for effective coagulation. The present study revealed that a much lower dose of ferric chloride is required in comparison to aluminium sulphate, and this potentially lowers the costs of operation. However, a detailed cost-benefit analysis is required to support results-based decision-making and conclusions. Used as dual coagulants, ferric chloride and chitosan resulted in the highest TDS and U₂₅₄. Previous studies have reported that chitosan serves as a good coagulant aid, and in particular was observed to reduce the dose of ferric chloride required in the treatment process (Hesami et al., 2014).

CONCLUSION

This study investigated the performance of three coagulants: aluminium chloride (A), ferric chloride (B), and chitosan (C) in drinking water treatment in order to improve efficiency of the coagulation process. The results demonstrated removal efficiency decreased in the order: B > A > C. The optimum pH for the single coagulants was acidic. Combined coagulants gave better removal efficiencies than single coagulants with maximum removal efficiencies obtained with the A/C combination. The optimum pH for combined coagulants was acidic, except when using the A/B combination. Overall, the coagulants presented satisfactory removal efficiencies, hence they are viable alternatives to D in treating drinking water. Aluminium coagulants are known to generate secondary products in treated water, and these have adverse human health effects. Since C is non-toxic and safe to ingest, it can potentially be used at household level for point-of-use water treatment in combination with other techniques.

The study showed that, in comparison to aluminium sulphate, a lower dose of ferric chloride is required, and this potentially reduces operation costs. However, a detailed cost-benefit analysis is required to support decision making. Future research should investigate the efficiency of combining the three coagulants and discovering the best dosing strategy. The removal of organic matter has been indicated by UV₂₅₄ in this study. Dissolved organic matter is an influencing factor in the coagulation process, thus the effectiveness in DOC removal by the different coagulant schemes would be interesting to investigate further.

AUTHOR CONTRIBUTIONS

NC conceptualized the idea, TAM collected and analysed data, HHM helped in data analysis, and RTT carried out statistical analysis. All authors contributed equally to manuscript writing.

FUNDING

This work was partially supported by the National Research Foundation of South Africa [Grant UID: 112013, 2018-2019].

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