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Assessment of Polyacrylamide and Aluminum Sulphate Coagulants in Turbidity Removal in Wastewater

Luke N. Ukiwe* • Jude I. Alinnor

Department of Chemistry, Federal University of Technology, P.M.B 1526, Owerri, Nigeria

Corresponding author: * luggil2002@yahoo.com

ABSTRACT

Turbidity adds an objectionable appearance to wastewaters. The present research investigates the application of a polymer (non-ionic polyacrylamide) and aluminum sulphate ($AI_2(SO_4)_3$) in coagulation-flocculation to improve the quality of domestic wastewater. Results of the "jar test" conducted indicated that by adding 150 mg L^{-1} of $AI_2(SO_4)_3$ at a contact time of 20 min, turbidity of the solution was reduced to about 90%. However, applying 200 mg L^{-1} of the polyacrylamide at a contact time of 35 min resulted in 89% turbidity removal. Evaluating the efficiency of turbidity removal of both coagulants, no significant differences were noted in the ability of both coagulants to clarify wastewater.

Keywords: adsorption, color, colloidal, ferric sulphate, precipitation, synthetic polymers

INTRODUCTION

Coagulation-flocculation is an essential part of drinking water and wastewater treatment. Wastewaters as well as surface waters must be treated to remove turbidity, color, and bacteria. Direct filtration of surface and wastewaters is largely ineffective in removing bacteria, viruses, soil particles and color.

The objective of coagulation-flocculation is to turn the small particles of color, turbidity and bacteria into larger flocs, either as precipitates or suspended particles. These flocs are then conditioned so that they will be readily removed in subsequent processes (Weiner and Matthews 2007). Technically, coagulation applies to the removal of colloidal particles (Weiner and Matthews 2007). During coagulation, a positive ion is added to water to reduce the surface charge to the point where the colloids are not repelled from each other. A coagulant is the substance (chemical) that is added to the water to accomplish coagulation. There are three key properties of a coagulant (Sharma 2002):

(i) Trivalent cation: the colloids most commonly found in natural waters are negatively charged; hence a cation is required to neutralize the charge. A trivalent cation is the most effective cation.

(ii) Nontoxic: this requirement is needed for the production of safe water.

(iii) pH: the coagulant that is added must precipitate out of solution at a neutral pH so that high concentrations of the ion are not left in the water. Such precipitation highly assists the colloid removal process.

The commonly used metal coagulants fall into two general categories (Sharma 2002) namely; those based on aluminum of which aluminum sulphate ($AI_2(SO_4)_3$), aluminum chloride ($AICI_3$) and sodium aluminate ($NaAIO_2$) are members and those based on iron which include ferric chloride ($FeCI_3$), ferrous sulphate ($FeSO_4$) and ferric sulphate ($Fe_2(SO_4)_3$).

The effectiveness of aluminum and iron coagulants arises principally from their ability to form multi-charged polynuclear complexes with enhanced adsorption characteristics. The nature of the complexes formed may be controlled by the pH of the system. When metal coagulants are added to water, the metal ions hydrolyze rapidly but in a somewhat uncontrolled manner forming a series of metal hydrolysis species. The efficiency of rapid mixing, pH, and the coagulant dosage determine which hydrolysis species is effective for treatment (Bratby 2006). There has been considerable development of pre-hydrolyzed inorganic coagulants, based on both aluminum and iron to produce the correct hydrolysis species regardless of the process conditions during treatment (Bratby 2006). These include aluminum chlorohydrate, polyaluminum chloride (PAC), polyaluminum sulphate chloride and forms of PAC with organic polymers. Iron forms include polyferric sulphate and ferric salts with polymers. Polymerized forms of aluminum-iron blends also exist.

The principal advantages of pre-polymerized inorganic coagulants are that they are able to function efficiently over wide ranges of pH and raw water temperatures. They are not influenced by low water temperature. Lower dosages are required to obtain water treatment goals, less chemical residuals are produced and lower chloride or sulphate residuals are produced resulting in lower final water total dissolved solid (Weiner and Matthews 2007). Comparative study between chemical coagulation/precipitation and coagulation/dissolved air flotation for pre-treatment of personal care products has been studied (EI-Gohary *et al*. 2010). $FeCI_3$, alum and $FeSO_4$ were used as coagulants. Results showed that alum produced higher chemical oxygen demand (COD) removal compared to $FeCI_3$, and $FeSO_4$. Huerta *et al*. (2011) suggested that coagulants such as $FeSO_4$ could be used successfully to remove pharmaceuticals and hormones through drinking water treatment. Other methods for treating waters and wastewaters using inorganic coagulants have been investigated by Verma *et al*. (2010) who studied the use of $FeCI_3$ in coagulation and flocculation pre-treatment of petrochemical wastewaters. Kushwaha *et al*. (2010) also reported on the treatment of simulated dairy wastewater using inorganic coagulants such as PAC, $FeSO_4$ and potash alum. They reported that optimum COD removal was obtained using PAC. However, Zhang *et al*. (2010) used combined hydrolysis acidification and dynamic membrane bioreactor coagulation process as a novel treatment

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method of treating water from polymer flooding. It was noted that COD removal efficiency could reach 89.41% with $AI_2(SO_4)_3$ at 140 mg L^{-1}. Silicate colloids have been investigated for their capacity to absorb heavy metals such as Pb and Cr in effluent and after adsorption, the colloids are separated by coagulation using $AI_2(SO_4)_3$. The removal ratio of heavy metals was as high as 99% (Li *et al*. 2004). Lu and Wei (2011) demonstrated that using a zerovalent iron/EDTA/air treatment system, removal efficiency of hydrolyzed polyacrylamide (PAM) and COD in oilfield wastewater containing polymer could be up to 45-66%.

Polymers are a large range of natural or synthetic water-soluble macromolecular compounds that have the ability to destabilize or enhance flocculation of the constituents of a body of water. Natural polymers such as crushed nuts from the Nirmali tree (*Strychnos potatorum*) have been used as flocculants for clarifying wastewaters (Bratby 2006). The advantages of natural polymers are that they are virtually free of toxins, biodegradable in the environment and the raw products are often locally available. Recently, synthetic polymers have gained acceptance in water purification (Amuda *et al*. 2006). Synthetic polymers are more effective as flocculants due to their high level of control of unreacted monomers attained during manufacture of the substance. Important mechanisms that relates to polymers during treatment include electrostatic and bridging effects. Polymers are available in various forms including solutions, powders, beads, oil or water-based emulsions. The polymer charge density influences the configuration in solution for a given molecular weight. Increasing charge density stretches electrostatic repulsion between charged units thereby increasing the viscosity of the polymer solution. A major concern about synthetic polymers relates to their potential toxicity, arising from residual unreacted monomers (Amuda *et al*. 2006). As earlier stated this problem is controlled during manufacture of the polymer and the quantity present after manufacture of the substance is rather too low to cause toxicity in water.

Attempts have been made to examine the effectiveness of polymer addition to coagulation process during treatment of beverage industrial wastewater to remove some of its trace metals content such as Pb, Cd, total Fe, total Cr, Ni and Zn. Results of standard "jar test" experiments conducted to determine the performance of both $FeCI_3$ and organic polymer (a non-ionic polyacrylamide) individually and $FeCI_3$-polymer combination showed that optimal removal efficiency for total Cr in the wastewater was about 300 mg L^{-1} for ferric chloride and 65 mg L^{-1} for the polymer. Addition of $FeCI_3$ resulted in significant removal of the metals 91, 72, and 54% of total Cr, Zn and total Fe respectively, while addition of polymer also achieve about 95, 87 and 88% total Cr, Zn and total Fe respectively. The study also noted that combination of ferric chloride and polymer at different ratio achieved better removal efficiency of the above mentioned metals (Amuda *et al*. 2006). The efficiency of water treatment with organic coagulants (cationic polymers) and calcium hydroxide (CaOH) in combination with $FeSO_4$ (mineral coagulant) as influenced by the nature and concentration of a mineral coagulant was also studied by Kurenkov *et al*. (2008). The study noted that in the absence of mineral coagulant the salt content in purified water would be lower and the concentration of organic coagulants required for water treatment would be significantly smaller than those of mineral compounds. Studies have been carried out for possible exploitation of *Cassia javahikai* seeds as potential source of commercial gum for textile wastewater treatment. Graft copolymerization with acrylamide was done to modify the seed gum for favorable properties. *C. javahikai* seed gum and its copolymer grafted with acrylamide were synthesized in the presence of oxygen using potassium persulphate/ascorbic acid redox system. Both *C. javahikai* seed gum and its grafted PAM were found to be good working substitutes as coagulants aids in conjunction with PAM (Sanghi *et al.* 2006). Preparation of liquid-phase polymerization of acrylamide acrylic acid to form polyelectrolytes used in wastewater cleaning using accelerated electron beam and microwave irradiation methods was studied by Radolu *et al.* (2004). The flocculation capacity of the obtained polymers was tested using two wastewaters, one sample from a slaughterhouse and the other from a vegetable oil plant. It was found that the combined treatment with polymers and $Al_2(SO_4)_3$ increased the degree of purification of both wastewaters up to 99%. Improvement of coagulation-flocculation process using anionic PAM as coagulant aid to improve the settling velocity of flocs formed when the coagulant was applied to treat wastewaters was studied by Aguilar *et al*. (2005). Results revealed that anionic PAM when added with $Fe_2(SO_4)_3$ or PAC led to a significant increase in the settling speed.

Three important factors in coagulant addition are pH, contact time and dose (Sharma 2002). Kurenkov *et al*. (2002) and Kabsch (2005) studied the effect of dosage of PAM and $Al_2(SO_4)_3$ in wastewater treatment. These studies noted that given dosage conditions, application of $AI_2(SO_4)_3$, PAC and PAM improve water quality, though, the influence of characteristics such as chemical composition, conformation of flocculant macro-molecules, concentration and nature of the coagulant as well as quality of the original water are factors to consider for effective treatment of water and wastewaters.

The present study compares the effectiveness of PAM and $AI_2(SO_4)_3$ in improving water quality when the dosage and reaction contact times of these two coagulants are varied with a view to corroborate recent advances in this area.

MATERIALS AND METHODS

The wastewater sample used in this study was collected three times a week from Hostel A of the Federal University of Technology, Owerri, Nigeria, exactly at 8.30 am of each sampling day from the pipe conveying all wastewater out of the hostel. Each time sampling was done, the rate of flow of wastewater was determined using a flow meter (WMFT 100, HPZ Ltd., Lagos, Nigeria). At the end of the third sampling day, a composite turbidity value was obtained from "jar test" experiments conducted on samples from the second and third sampling days. The amount of wastewater generated by residents in the hostel was also determined by adding the values obtained from the flow rate of the first, second and third sampling days respectively.

The conventional "jar test" method was employed for the coagulation-flocculation process. A 50-L plastic container was used to collect the wastewater sample from the sampling point. For each sampling day 50 L wastewater sample was fetched and taken to the Project Laboratory of the Department of Chemistry, Federal University of Technology, Owerri, Nigeria and allowed to stand for 2 hrs for debris to settle. About 2 L of this sample was measured into a 5-L beaker and further screened to remove other suspended particles. A 2-L sample from this screened wastewater sample was measured ion to another 5-L beaker and from this stock solution, sample for "jar test" was continuously drawn. 1-L sample from the stock was measured into a 2-L beaker and 50 mg of $Al_2(SO_4)_3$ was added to the sample solution and the pH adjusted to 5.0 using NaOH/HCL as appropriate. Rapid mixing at 150 rpm for 2 min followed by gentle mixing at 20 rpm for another 3 min was performed using a centrifuge (MC 5415C, AKSON Scientific Instruments, Awka, Nigeria), then the resultant mixture was allowed to stand for 5 min to allow flocs formed to settle. After this period, the clear mixture was decanted and filtered using a Whatman No. 42 filter paper. About 10 ml of this filtrate was used for turbidity measurement using a turbidimeter (WGZ-1B, Shanghai Xinrui Instruments, China).

The above procedure was repeated allowing the mixture to stand for flocs to settle at 10, 15, 20, 25, 30, and 35 min. The above experiment was repeated for day two and three sampling days allowing the mixture to stand for 5, 10, 15, 20, 25, 30, and 35 min contact time for flocs to settle. However, using the above protocol, the experiment was repeated for 100, 150, and 200 mg $Al_2(SO_4)_3$ and also using the above weights, the entire experiment was repeated using PAM.

Analysis of variance (one-way ANOVA) was employed to determine the difference between turbidity values at various co-

Table 1 Turbidity values of various weights of $Al_2(SO_4)_3$ coagulant.

Coagulant weights (mg) / Contact time (min)	50[(a)]		100[(b)]		150[(c)]		200[(d)]	
	Mean (NTU) ± SD × 10^{-1}	SEM	Mean (NTU) ± SD × 10^{-1}	SEM	Mean (NTU) ± SD × 10^{-1}	SEM	Mean (NTU) ± SD × 10^{-1}	SEM
5.0	19.61 ± 1.9	1.0	7.86 ± 4.1	2.3	2.06 ± 1.8	1.0	9.70 ± 6.7	3.8
10.0	18.62 ± 0.0	0.0	1.62 ± 3.9	2.2	1.90 ± 2.5	1.4	3.11 ± 5.7	3.2
15.0	19.41 ± 0.5	0.2	1.12 ± 3.0	1.7	2.66 ± 1.8	1.0	6.29 ± 4.4	2.5
20.0	17.76 ± 9.1	5.2	0.17 ± 0.0	0.0	0.04 ± 0.0	0.0	1.63 ± 4.8	2.7
25.0	19.20 ± 0.0	0.0	3.36 ± 1.5	0.8	0.67 ± 0.0	0.0	9.66 ± 0.2	0.1
30.0	16.12 ± 0.1	0.5	2.59 ± 1.9	1.0	2.86 ± 4.8	2.7	6.26 ± 7.6	4.3
35.0	13.04 ± 1.5	0.8	0.62 ± 0.0	0.0	4.19 ± 8.8	5.0	4.31 ± 3.5	2.0

SEM: Standard Error of the Mean; *F*-test between (a) and (b) = 1.15; (b) and (c) = 1.95; (c) and (d) = 2.78

Table 2 Turbidity values of various weights of polyacrylamide coagulant.

Coagulant weights (mg) / Contact time (min)	50[(a)]		100[(b)]		150[(c)]		200[(d)]	
	Mean (NTU) ± SD × 10^{-1}	SEM	Mean (NTU) ± SD × 10^{-1}	SEM	Mean (NTU) ± SD × 10^{-1}	SEM	Mean (NTU) ± SD × 10^{-1}	SEM
5.0	15.06 ± 6.9	3.9	6.86 ± 3.3	1.9	4.90 ± 5.8	3.3	2.90 ± 3.6	2.0
10.0	13.08 ± 5.9	3.4	8.12 ± 1.9	1.0	4.76 ± 0.0	0.0	3.16 ± 6.8	3.9
15.0	16.17 ± 4.9	2.8	5.14 ± 5.8	3.3	5.15 ± 7.7	4.4	2.67 ± 4.9	2.8
20.0	12.06 ± 5.9	3.4	9.19 ± 6.7	3.8	4.97 ± 4.6	2.6	3.44 ± 0.0	0.0
25.0	12.18 ± 3.9	2.2	4.56 ± 8.9	5.1	3.88 ± 8.8	5.0	3.76 ± 3.8	2.1
30.0	13.65 ± 7.9	4.5	3.11 ± 4.7	2.7	4.26 ± 1.5	8.6	4.29 ± 3.5	2.0
35.0	11.44 ± 5.6	3.2	2.56 ± 5.8	3.3	2.64 ± 3.9	2.2	2.14 ± 3.8	2.1

SEM: Standard Error of the Mean; *F*-test between (a) and (b) = 1.12; (b) and (c) = 1.31; (c) and (d) = 1.49

agulant weights. BMDP statistical software was used for data analysis as data were presented as arithmetic mean, standard deviation and standard error of the mean. However, the *F*-test was used to observe for significance in turbidity values between coagulant weights.

Table 1 show values of the standard error of the mean and the *F*-test between coagulant mean weights. These values were tested at 6 degrees of freedom, $P < 0.01$.

RESULTS

Tables 1 and **2** give mean turbidity values at 5 to 35 min range contact time for various $Al_2(SO_4)_3$ and PAM weights. For the $Al_2(SO_4)_3$ coagulant, it was observed that at 20 min contact time, about 99% clarification of the wastewater was achieved at 150 mg $Al_2(SO_4)_3$ weight while adding 200 mg PAM coagulant to the wastewater at 35 min contact time, resulted in 89% clarification of the wastewater, the highest percentage value obtained for PAM. Interestingly, 35 min contact time using PAM gave the best clarified solutions at 50-200 mg coagulant weights as the weights were ascended, though this trend was not replicated at other contact times observed. The present study noted that assessing both coagulants in turbidity removal efficiency, the results obtained did not reflect the fact that any of the coagulants has edge over the other in its ability to clarify wastewater.

DISCUSSION

Many treatment technologies for wastewaters containing heavy metals have been developed in recent years, but these technologies have some disadvantages, such as poor removing efficiency and complex operation (Chang and An 2007). A macromolecular heavy metal coagulant polyethyleneimine sodium xanthogenate (PEX) was prepared by Chang and An (2007) by grafting a xanthogenate group to polyethyleneimine. It was demonstrated that PEX has function capable of removing both turbidity and copper ions, since copper ions and turbidity have cooperative removal effect with each other in the process of treating wastewater containing both turbidity and copper ions. Graft copolymerization has also been used as an important technique for modifying the physical and chemical properties of polymers. Sango starch graft-polyacrylamide (ss-g-PAM) was prepared by graft copolymerization of ceric ion-induced redox polymerization of acrylamide at room temperature and used for the treatment of turbidity of water. The ss-g-PAM coagulant was found to achieve water turbidity removal up to 96.6%. The result obtained from the study suggested that ss-g-PAM copolymer is a potential coagulant for reducing turbidity during water treatment (Qudsieh *et al*. 2008).

Polyferric sulphate and cationic PAM have been used as primary coagulant and coagulant aid for the pretreatment of diosgenin wastewater. The influence of the main operating parameters such as effluent pH and coagulant dosage in the coagulant performance were investigated. It was revealed that cationic PAM substantially promoted the reduction of residual turbidity with the appropriate coagulant dosage in the range of 2-5 mg L^{-1} substrate (Liang and Wang 2010). A recent study on the use of $FeCl_3$, $FeSO_4$, PAC and cationic PAM to improve the treatment of raw tannery wastewater at low temperature (10°C) also revealed that these polymeric and conventional coagulants were efficient in removing suspended solids and organic matter in tannery wastewaters (Zang *et al*. 2009).

Applying inorganic coagulants such as $Al_2(SO_4)$ and calcium oxide (CaO) have been shown to remove heavy metals from soil leachate (Ukiwe and Nwoko 2010). It was found that "jar test" experiments conducted by adjusting the pH of the leachate in the range of 2–5, about 98% Cd was coagulated out of the leachate at pH 3.0, while Cu (8%) was coagulated at pH 5.0. The study further concluded that coagulation of metals dependent on pH. Pulp and paper mill effluent is highly polluting and is a subject of environmental concern. A batch coagulation study conducted to treat paper and pulp mill effluent using coagulants such as $AlCl_3$, $CuSO_4$ and PAC revealed that the initial pH of the effluent had tremendous effect on color removal. PAC, $AlCl_3$, $CuSO_4$ removed 92, 84, and 78% color at pH 5.0, 4.0, and 6.0, respectively (Chaudhari *et al.* 2010). Garg *et al*. (2010) also studied the effectiveness of coagulation using aluminum and $FeSO_4$-based chemicals for the pre-treatment of diluted black liquor. The research noted that commercial alum was found to be the most economical among all the aluminum and ferrous salts used as coagulant with a color reduction of about 90% at pH 5.0. Other studies have also reported similar results (Johnson *et al*. 2008; Zazouli and Yousefi 2008; Ersoy *et al*. 2009).

Turbidity is due to colloidal and extremely fine dispersions. Its measurements are helpful to determine the extent of pollution of water and wastewaters. Different coagulants and flocculants have been applied to determine the optimum conditions for color and turbidity removal. Marañón *et al*. (2010), Hsu *et al.* (2009), and Matilainen *et al*. (2010) all reported on the use of inorganic and polymeric coagulant, biocoagulants, and natural organic matter in reduction of turbidity in sludge and superturbid drinking water respectively. These researchers all agreed that by applying single or dual coagulant options, waters and wastewaters could be treated efficiently and economically, however, Hsu *et al.* (2009) noted that this method of treating waters and wastewaters if effectively applied can be the solution by which water deficiency can be eliminated during typhoons and other such emergency situations.

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