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Removal of Heavy Metals from Industrial Wastewaters Using Local Alum and Other Conventional Coagulants-A Comparative Study

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Abstract: The present study aimed at effective management and purification of industrial wastewaters using cheaper and locally available local alum for removal of heavy metals as a substitute to convectional coagulants. The effect of local alum, aluminum sulphate and ferric chloride on the metal contents of industrial wastewaters was investigated in the pH range of 5.9-7.5. Wastewater samples from battery, paint and textile industries were treated with different doses of locally available alum, aluminum sulphate and ferric chloride in order to determine and compare their effectiveness in removing heavy metal contents of the wastewaters. The percentage removal of the metals from the industrial wastewaters increased with mg L^{-1} dosage of the coagulants used with optimal performance generally at a slightly alkaline pH. Local alum proved to be equally effective in removing heavy metals from the industrial wastewater samples compared with the conventional aluminum sulphate and ferric chloride.

Key words: Local alum, aluminum sulphate, ferric chloride, heavy metals, industrial wastewaters, coagulants

INTRODUCTION

Many coagulants are widely used in conventional water-treatment processes for tap water production. These coagulants can be classified into inorganic coagulant, synthetic organic polymer and naturally occurring coagulant (Pitt *et al*., 2004). These coagulants are used for various purposes depending on their chemical characteristics. An inorganic polymer, polyaluminum chloride and inorganic salt alum (aluminum sulfate) are the most widely used coagulants in water treatment (Boisvert *et al*., 1997; Okuda *et al*., 2001). Coagulation is a two-step process, the first step is destabilization of particles (by chemical addition), followed by amalgamation (by mixing) to form faster sinking particles (Fatoki and Ogunfowokan, 2002; WTA's Word Wide Water, 2002; Pitt *et al*., 2004). Coagulation is also useful in removing hydrophilic (water-loving) particles like organic colour forming compounds which are usually difficult to remove by some other means. Chemicals commonly used in the coagulation process are inorganic salts of iron and aluminum such as aluminum sulphate, ferric chloride, ferric sulphate, cationic polymers, potash alum, and chlorinated copper (Jiang and Graham, 1998).

Effluents generated from the industries contain pollutants especially heavy metals that are hazardous to the environment. Vast quantities of these heavy metals are discharged into the environment and are not biodegradable; they are also involved in a complex biogeochemical cycle. Due to toxicities caused by these heavy metals and pollutants in general to humans and the environment, it is important that wastewaters have to be treated before disposal. The principal objective of wastewaters treatment is generally to allow industrial effluents to be disposed off without causing unacceptable damage to the natural environment.

Springing small-scale and even some large-scale industries in developing countries seldom have their effluents properly treated before discharging them into water bodies. This is done either in an attempt to cut cost since the conventional chemicals for wastewater treatment are considered costly or because the appropriate technology used by the developed countries (Johnson *et al*., 2003; USInfrastructure, 2003) is expensive. As a result, the pollution level of streams and rivers around industrialized locations has tremendously increased. A review by Nriagu (1992) has shown that the limited water resources available in Africa are now being contaminated with metals.

Somashekar *et al*. (1982) observed that the chemical composition of water results from different environmental factors operating simultaneously but with differing influences and efficiency. According to Ang *et al*. (1989), heavy metals are present in sea water as a result of both natural and anthropogenic activities. Apart from natural factors like weathering, human activities towards improving conditions of living have inadvertently introduced harmful substances into the environment WHO, (1971; 1977). Even though some trace elements are

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essential to man, essential as well as non-essential elements at elevated levels can cause morphological abnormalities, reduced growth, increased mortality and mutagenic effects in humans (Pier and Bang, 1980; Friberg *et al*., 1986; DWAF, 1996). Thus, efforts geared towards effective management and purification of wastewaters using cheaper and easily available materials are quite important.

In our preliminary work on the effect of coagulant treatment on the metal composition of raw water, two coagulants-$Al_2(SO_4)_2$ and $Fe_2(SO_4)_3$ were used. The results showed that apart from removing suspended matters both coagulants have additional advantage of being capable of removing metals with percentage removal enhanced with increased mg L^{-1} dosage of either coagulant used (Fatoki and Ogunfowokan, 2002). The present study is concerned with the application of coagulants-aluminum sulphate, ferric chloride and a locally available and widely used alum from South Western Nigeria (the local alum is used mainly for removal of suspended matter from water}, in the removal of heavy metals viz: Mn, Cr, Cd, Zn, Ni and Pb from industrial wastewaters from textile, battery and paint industries, respectively and evaluation of their effectiveness in the removal of metals and the extent to which local alum may be used as a substitute to the conventional and more costly coagulants for the removal of toxic heavy metals from industrial waste waters. Interest in coagulation and hence removal of these metals stems from the fact that most of these metals may occur at levels that can cause deleterious effect on the health of humans if discharged into the environment especially into a receiving stream which serves as drinking water for rural dwellers downstream.

MATERIALS AND METHODS

Sample collection and treatment: Industrial wastewater samples from paint, textiles and battery industries from industrialized part of Western Nigeria were collected separately into polyethylene plastic bottles, which had been previously washed with detergent solution, soaked in 10% HNO_3 and finally rinsed thoroughly with triply distilled water. The samples were transported to the laboratory and refrigerated at 4°C prior to analyses.

Chemicals and reagents: All the chemicals used were of Analytical grade. The anhydrous reagents were placed in an oven at 105°C for 12 h to remove traces of water and then placed in a desecrator to prevent absorption of moisture from the atmosphere prior to usage. Triply distilled water was used for all reagent preparations.

Quality control study

Recovery work: One hundred milliliter of triply distilled water was placed in each of six 250 mL beakers. Continuous stirring using a magnetic stirrer for each of the 100 mL distilled water was carried out at a speed of 100 rev min^{-1} for 1 min. pH adjustment of the triply distilled water to 7.5 was done using 0.1 MH_2SO_4 or 0.2 MNaOH as necessary. Each of the distilled water was, respectively spiked with 10 ppm of Zn^{2+}, Cd^{2+}, Cr^{6+}, Mn^{2+}, Pb^{2+} and Ni^{2+} mixture. Stirring of the spiked solutions at a speed of 100 rev min^{-1} was carried out for 15 min. About 75 mL of the stirred solution was placed in the pretreated sample bottle separately for analysis by Atomic Absorption Spectrophotometry (AAS) available at the Center for Energy Research and Development of the Obafemi Awolowo University, Nigeria. A 10 ppm each of the standards Zn^{2+}, Cd^{2+}, Cr^{6+}, Mn^{2+}, Pb^{2+} and Ni^{2+} were also analysed by AAS and the results obtained were compared with those of spiked water samples to obtain the recovery of the analytes.

A blank experiment was carried out with triply distilled water using the same procedure above omitting the standards to establish blank levels DWAF (1992).

Treatment of industrial wastewaters with coagulants: The pH of the wastewater samples was measured using a digital pH meter mode H (Hanna Instrument). The industrial wastewater samples were filtered through a 0.45 µm filter paper to remove large particulates. Two hundred and fifty milliter of the filtered wastewater samples were placed in 500 mL beakers and stirred continuously using a magnetic stirrer at a speed of 100 rev min^{-1}. 0.1 MH_2SO_4 or 0.2 MNaOH was added to achieve the desired pH. The filtered water samples were then coagulated with 3, 6 and 10 ppm doses of $Al_2 (SO_4)_3$ within the pH range of 5.9-7.5. Similarly, 3, 6 and 10 ppm of $FeCl_3$ and local alum were, respectively used to coagulate wastewater samples within the same pH range mentioned above. After the addition of each coagulant, the stirring rate was accelerated to 300 rev min^{-1} for 3 min and then reduced to 100 rev min^{-1} for 15 min. The flocs formed were allowed to settle for 30 min after stirring had been completed. 25 mL of the resultant solution was taken from 10 mL below the solution surface and then filtered through a 0.45µm filter paper. The filtrate was placed in the pretreated polyethylene sample bottle and analyzed by AAS.

Instrumentation: The analyses of the treated and untreated samples were performed with Flame Atomic Absorption Spectrometer (FAAS) (Chemtech Analytical U. K.) at the Center for Energy Research and Development of the Obafemi Awolowo University, Ile-Ife, Nigeria. Data were acquired on a Gateway 2000 PC system using the appropriate software. Six elements: Mn, Cr, Cd, Zn, Ni and Pb were determined using the above instrumentation and the percentage removal (% Removal) of each metal was estimated.

RESULTS AND DISCUSSION

The percentage recovery (% Recovery) ranged from (60%) Pb to (85%) Mn (Table 1). Results of the blank experiments for the metals in the study gave clean blank levels. These results thus validated the experimental procedure used for the chemical analysis. Table 2 shows the results of the heavy metal concentration of the raw industrial wastewaters from three industries investigated in this study together with the total metal load in mg L^{-1}. The levels of the heavy metals in the raw industrial waste waters before treatment with the coagulants were: 3.01 mg Mn L^{-1}, 0.26 mg Cr L^{-1}, 4.01 mg Cd L^{-1}, 129.64 mg Zn L^{-1}, 1.88 mg Ni L^{-1} and 0.12 mg Pb L^{-1} in battery industrial wastewater; 0.39 mg Mn L^{-1}, 0.01 mg Cr L^{-1}, 0.23 mg Cd L^{-1}, 0.59 mg Zn L^{-1}, 0.15 mg Ni L^{-1} and 0.11 mg Pb L^{-1} in the effluent from paint industry and 0.28 mg Mn L^{-1}, 0.00 mg Cr L^{-1}, 0.20 mg Cd L^{-1}, 0.36 mg Zn L^{-1}, 0.05 mg Ni L^{-1} and 0.00 mg Pb L^{-1} in textile industry wastewater, respectively. The total metal load was highest in battery industry (138.92 mg L^{-1}) and the least in textile industry (0.89 mg L^{-1}) Table 2. This shows that the wastewater from battery industry was the most polluted of the selected heavy metals in this study. This is unfortunate as these metals may be extremely toxic to humans and aquatic biota even at low concentrations (Leonard, 1991), if they are discharged continuously into fresh water bodies directly without their removal from the wastewaters and such water is consumed untreated by rural dwellers downstream.

Presently, only the effluent from textile industry met with the effluent limitation guidelines stipulated by Federal Environmental Protection Agency (FEPA, 1991) for all the metals analyzed (Table 2). There were violations of these guidelines for Cd and Pb in paint industry while in battery industry, violations of guidelines occurred for Cd, Zn, Ni and Pb (Table 2). This calls for concern more so that most industries in the study area discharge effluents into receiving surface waters either untreated or partially treated in spite of the campaign for proper treatment of effluents prior to their discharge into the environment by the Nigerian Ministry of Environment regulatory body (FEPA). Furthermore, the high levels of some of these metals reported in the raw wastewaters especially for paint and battery industries show that interest in the coagulation processes and hence the removal of the heavy metals from the industrial wastewaters prior to their discharge into surface waters is of great importance because these metals may constitute a threat to human health particularly that levels of most of the metals in the wastewaters for all the categories of industries studied were presently above the minimum allowable threshold in water intended for human consumption (Table 3). Two

Table 1: Recoveries of Metals from spiked water sample

	Concentration (mg L^{-1})		
Element	Amount added	Amount recovered	Recovery (%)
Mn	10.0	8.50±0.08	85
Cr	10.0	8.40±0.07	84
Cd	10.0	8.00±0.13	80
Zn	10.0	7.30±0.11	73
Ni	10.0	6.30±0.15	63
Pb	10.0	6.00±0.09	60

Table 2: Trace metal concentration of raw industrial wastewater samples

	Concentration of trace metals (mg L^{-1})						
Industry	Mn	Cr	Cd	Zn	Ni	Pb	(Total load (mg L^{-1})
Battery	3.01±0.54	0.26±0.13	4.01±0.29	129.64±5.33	1.88±0.11	0.12±0.07	138.92
	(5.0)	(<1.0)	(0.01)	(<1.0)	(0.05)	(0.01)	
Paint	0.39±0.11	0.01±0.00	0.23±0.12	0.59±0.19	0.15±0.06	0.11±0.04	1.48
	(5.0)	(1.0)	(0.01)	(1.0)	(1.0)	(0.01)	
Textile	0.28±0.10	ND	0.20±0.08	0.36±0.16	0.05±0.01	ND	0.89
	(5.0)	(<0.10)	(<1.0)	(<1.0)	(<1.0)	(<1.0)	

Values in parenthesis are Nigerian national effluent limitation guidelines for industries for effluents for discharge into surface water (FEPA, 1991). ND = Not detected

Table 3: Tolerance limits of some trace metals in drinking water

Metal	Upper limit of concentration (mg L^{-1})
Mn*	0.05
Cr	0.05
Cd	0.01
Zn*	5.00
Ni	-
Pb	0.05
Hg	0.001

Source: WHO, 1971.*Report of the committee on water quality criteria federal water pollution administration, Us department of interior, Washington DC

Table 4: Removal of heavy metals from industrial wastewaters by aluminum sulphate [$Al_2(SO_4)_3$].

Industry	Coagulant dosage (mg L^{-1})	pH	Change in concentration (mg L^{-1})						Removal (%)					
			Mn	Cr(VI)	Cd	Zn	Ni	Pb	Mn	Cr	Cd	Zn	Ni	Pb
Battery	0	5.94	3.01±0.07	0.26±0.3	4.013±0.243	129.640±2.741	1.88±0.21	0.12±0.03	0.00	0.00	0.00	0.00	0.00	0.00
	3	7.20	1.43±0.21	0.03±0.01	0.004±0.001	0.008±0.002	1.40±0.11	0.08±0.02	52.50	88.50	99.90	99.90	25.50	33.30
	6	7.30	1.35±0.10	0.02±0.00	0.00	0.00	1.20±0.07	0.01±0.00	55.10	92.30	100.00	100.00	36.20	91.70
	10	7.50	1.34±0.12	0.01±0.01	0.00	0.00	1.20±0.03	0.00±0.00	55.50	96.20	100.00	100.00	36.20	100.00
Paint	0	5.94	0.39±0.09	0.01±0.00	0.230±0.006	0.591±0.019	0.15±0.08	0.11±0.06	0.00	0.00	0.00	0.00	0.00	0.00
	3	7.20	0.15±0.05	0.00	0.00	0.00±0.01	0.00	0.05±0.02	61.70	100.00	99.80	99.80	100.00	95.50
	6	7.30	0.06±0.00	0.00	0.00	0.00	0.00	0.00	84.70	100.00	100.00	100.00	100.00	100.00
	10	7.50	0.04±0.01	0.00	0.00	0.00	0.00	0.00	89.80	100.00	100.00	100.00	100.00	100.00
Textile	0	5.94	0.28±0.11	0.00±0.00	0.197±0.094	0.360±0.083	0.05±0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	3	7.20	0.01±0.01	0.00	0.00	0.00	0.00	0.00	96.50	0.00	100.00	100.00	100.00	0.00
	6	7.30	0.00	0.00	0.00	0.00	0.00	0.00	100.00	0.00	100.00	100.00	100.00	0.00
	10	7.50	0.00	0.00	0.00	0.00	0.00	0.00	100.00	0.00	100.00	100.00	100.00	0.00

Table 5: Removal of heavy metals from industrial wastewaters by ferric chloride ($FeCl_3$)

Industry	Coagulant dosage (mg L^{-1})	pH	Change in concentration (mg L^{-1})						Removal (%)					
			Mn	Cr	Cd	Zn	Ni	Pb	Mn	Cr	Cd	Zn	Ni	Pb
Battery	0	5.94	3.01±0.05	0.26±0.01	4.013±0.110	129.640±5.021	1.88±0.30	0.12±0.02	0.00	0.00	0.00	0.00	0.00	0.00
	3	7.20	1.71±0.22	0.03±0.01	0.008±0.002	0.018±0.001	1.60±0.22	0.00	43.20	88.50	99.80	99.90	14.90	100.00
	6	7.30	1.54±0.02	0.00	0.008±0.002	0.007 ±0.003	1.50 ±0.09	0.00	48.80	100.00	99.80	100.00	20.20	100.00
	10	7.50	1.42±0.16	0.00	0.005±0.002	0.00	1.30±0.15	0.00	52.80	100.00	99.80	100.00	30.90	100.00
Paint	0	5.94	0.39±0.11	0.01±0.01	0.230±0.081	0.591±0.062	0.15±0.04	0.11±0.06	0.00	0.00	0.00	0.00	0.00	0.00
	3	7.20	0.07±0.01	0.01±0.01	0.003±0.001	0.005±0.001	0.00	0.02 ±0.01	82.10	0.00	98.70	99.20	100.00	81.80
	6	7.30	0.06±0.02	0.00	0.00	0.00	0.00	0.00	84.70	100.00	100.00	100.00	100.00	100.00
	10	7.50	0.04±0.02	0.00	0.00	0.00	0.00	0.00	89.80	100.00	100.00	100.00	100.00	100.00
Textile	0	5.94	0.28±0.07	0.00	0.197±0.019	0.360±0.008	0.05±0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	3	7.20	0.03 ±0.01	0.00	0.003±0.01	0.008±0.001	0.00	0.00	89.40	0.00	98.50	87.70	100.00	0.00
	6	7.30	0.01±0.01	0.00	0.00	0.00	0.00	0.00	96.50	0.00	100.00	100.00	100.00	0.00
	10	7.50	0.00	0.00	0.00	0.00	0.00	0.00	100.00	0.00	100.00	100.00	100.00	0.00

Table 6: Removal of heavy metals from industrial wastewaters by local alum

Industry	Coagulant dosage (mg L^{-1})	pH	Change in concentration (mgL^{-1})						Removal (%)					
			Mn	Cr	Cd	Zn	Ni	Pb	Mn	Cr	Cd	Zn	Ni	Pb
Battery	0	5.94	3.01±0.10	0.26±0.02	4.013±0.150	129.640±0.112	1.88 ±0.14	0.12±0.05	0.00	0.00	0.00	0.00	0.00	0.00
	3	7.20	0.04±0.01	0.19±0.05	3.182±0.234	67.672±0.157	0.70±0.25	0.06 ±0.01	98.70	26.90	20.70	47.80	62.80	50.00
	6	7.30	0.02±0.01	0.15±0.07	2.243±0.211	67.154±0.374	0.67±0.33	0.04±0.00	99.40	42.30	44.10	48.20	64.40	66.70
	10	7.50	0.01±0.00	0.00±0.00	0.542±0.098	45.892±2.321	0.66±0.20	0.03±0.01	99.70	100.00	86.50	64.60	64.90	75.00
Paint	0	5.94	0.390±0.015	0.010±0.001	0.230±0.07	0.591±0.065	0.15±0.01	0.11±0.02	0.00	0.00	0.00	0.00	0.00	0.00
	3	7.20	0.002±0.001	0.008±0.003	0.208±0.097	0.489±0.08	0.10±0.08	0.07±0.00	99.50	20.00	9.60	17.30	33.30	36.40
	6	7.30	0.002±0.000	0.007±0.002	0.100±0.08	0.434±0.20	0.09±0.02	0.05±0.01	99.50	30.00	56.50	26.60	40.00	54.40
	10	7.50	0.001±0.001	0.00	0.061±0.005	0.321±0.019	0.08±0.03	0.01±0.00	99.70	100.00	73.50	45.70	46.70	90.90
Textile	0	5.94	0.280±0.121	0.00	0.197±0.027	0.360±0.026	0.05±0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	3	7.20	0.002±0.001	0.00	0.155±0.061	0.330±0.093	0.03±0.01	0.00	99.30	0.00	21.30	8.30	40.00	0.00
	6	7.30	0.002±0.001	0.00	0.070±0.008	0.194±0.10	0.027±0.01	0.00	99.30	0.00	64.50	46.10	46.00	0.003
	10	7.50	0.00	0.00	0.029±0.004	0.119±0.032	0.026±0.011	0.00±0.00	100.00	0.00	85.50	66.90	47.90	0.00

or more poisons present together in water may exert additive, antagonistic or synergistic effects (Mason, 1996). For example, Ni and Cr act synergistically while Zn and Cu have negative additive effects on marine organism.

Table 4-6 show the results of removal of heavy metals from the various industrial wastewaters using $Al_2(SO_4)_3$ $FeCl_3$ and local alum respectively. It is evident from these Tables that as the dosage and pH increase, percentage removal of the heavy metals from the industrial wastewaters generally increased reaching an optimum level at pH 7.5 and dosage 10 mg L^{-1} of the various coagulants used.

In the Battery Industry, local alum gave 99.70% efficiency of Mn removal and proved to be the best coagulant for removing Mn at 10 mg L^{-1} dosage. Both local alum and Ferric Chloride gave significant removal of Cr (VI) (100.00%) while aluminum sulphate removed 96.20% Cr (VI) from the waste water at 10 mg L^{-1} dosage of the coagulants. The results for the Cd removal from Battery industry wastewater sample showed that local alum gave the least percentage removal of 86.50%, while 100.00% and 99.80% percentage removal were obtained respectively for aluminum sulphate and ferric chloride at pH 7.50 and 10 mg L^{-1} coagulant dosage. Local alum was best suitable for removing Ni having the highest percentage removal of 64.90% while the percentage removal of Ni by ferric chloride and aluminum sulphate were 30.90% and 36.20% respectively at 10 mg l^{-1} coagulant dosage (Table 4-6). Present results further showed that local alum even at lower dosages of 3-6 mg L^{-1} is the best coagulant for removing Ni from the battery industrial wastewater compared to other conventional coagulants. Treatment of the wastewater with $FeCl_3$ best removed Pb followed by $Al_2(SO_4)_3$ and local alum, respectively.

In the paint industry, local alum gave the best percentage removal of Mn (99.70%) compared to $Al_2(SO_4)_3$ (89.80%) and $FeCl_3$ (89.80%) at 10 mg L^{-1} coagulant dosage and pH 7.50 (Table 4-6). This observation is consistent with what was observed for the removal of Mn in battery wastewater. Its efficiency of removal for Zn and Ni from the wastewater of paint industry however, was less than those of the other two coagulants investigated in this study. Furthermore, local alum gave 90.90% percentage removal of Pb from Paint Industrial wastewater whereas other coagulants gave 100% removal.

Although $Al_2(SO_4)_3$ and $FeCl_3$ showed significant removal of Mn, Cd, Zn and Ni (100%) from textile industrial wastewater, local alum also showed an acceptable percentage removal of Mn (100%), Cd (85.50%) and Zn (66.90%) from the wastewater at 10 mg L^{-1} dosage of the coagulants. It is worth mentioning however that the removal of Ni from the textile wastewater by the local alum was small (47.90%).

This study has shown that there is a general increase in the percentage removal of metals from industrial wastewaters with increased dosage of coagulants. Apart from the fact that local alum was the best coagulant for the removal of Mn and Ni from all the three industrial wastewaters, its performance for the removal of all other metals is equally effective compared with the other conventional coagulants used in this study. Judging from these results local alum may therefore be used as a substitute to the conventional and more costly coagulants for the removal of heavy metals from industrial waste waters.

CONCLUSIONS

In this study, local alum, $Al_2(SO_4)_3$ and $FeCl_3$ were investigated for their effectiveness in removing heavy metals from wastewaters from battery, paint and textile industries. The percentage removal of the metals from the industrial wastewaters increased with the dosage of the coagulants used with optimal performance at a slightly alkaline pH. All the coagulants apart from removing turbidity from the wastewaters also showed their capabilities of removing heavy metals. The local alum proved to be a cheap and effective coagulant in the removal of heavy metals from industrial wastewaters and may be used as a substitute to the conventional coagulants.

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