



Kinetics and Particle Removal Profile of Pulverized Snail Shell – Alum Induced Coag-Flocculation of Quarry Effluent

C. Nnaji Patrick^{1*}, I. Okolo Bernard¹, Menkiti Matthew Chukwudi²,
Ume Cyril Sunday³ and Chizoba Chinelo Agu⁴

¹Department of Chemical Engineering, Michael Okpara University of Agriculture Umudike, Nigeria.

²Department of Chemical Engineering, Nnamdi Azikiwe University Awka, Nigeria.

³Department of Chemistry, Federal University Ndufu Alike Ikwo (FUNAI), Ebonyi State, Nigeria.

⁴Center for Environmental Management and Control (CEMAC), University of Nigeria, Enugu, Nigeria.

Authors' contributions

This work was carried out in collaboration between all authors. Authors CNP, IOB and MMC designed the study, performed the statistical analysis, wrote the protocol, and wrote the first draft of the manuscript and managed literature searches. Authors UCS and CCA managed the analyses of the study and literature searches. All authors read and approved the final manuscript.

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ABSTRACT

This study investigated the coag-flocculation performance of alum, pulverized snail shell coagulant (PSSC), and their blends under varying pH and coagulant dosage in removing turbidity from quarry effluent (QE) at room temperature. A laboratory bench-scale jar test was employed for the experiments. Coag-flocculation parameters such as coagulation rate constant, K11, coagulation half-time, $\tau_{(1/2)}$ etc were determined. The optimum pH was observed at 6.0, while the blend of 200.0 mg/L alum and 800.0mg/L PSSC achieved the optimum turbidity removal. Turbidity removal efficiency was recorded between 87.9% and 98.5% for various dosages and pH studied. The coagulation rate half-time, $\tau_{(1/2)}$ range from 13.8s to 972.45s for various dosages and pH studied.

*Corresponding author: E-mail: nnaji_pat@yahoo.com;

The use of PSSC blended with alum showed high level of potential, for the treatment of quarry effluent.

Keywords: Snail shell; quarry effluent; coagulation/flocculation; jar test.

1. INTRODUCTION

The issue of providing sufficient quantities of high quality water to take care of our domestic, industrial and agricultural needs has been a global concern. The ever increasing population size, climate change and environmental pollution exacerbate the situation.

It is a clear knowledge that there is no shortage of water on earth. It covers 70% of the globe. However, 97% of the world water is saline and non-drinkable. A 2% is locked in glacier and polar ice caps. This leaves 1% to meet social needs [1]. Human activities and climate change cause turbidity and other pollution, in the 1% which is left resulting in the need for treatment before use. Turbidity in water is caused by suspended and colloidal matters such as clay, silts, finely divided organic and inorganic matter, plankton and other micro-organism [2].

Coag-flocculation process has been used in the removal of turbidity from wastewater. This process is achieved by the addition of coagulants to wastewater in order to cause destabilization of the colloid dispersion and agglomeration of the resulting individual colloidal particles [3,4]. Among other factors, temperature, pH, effluent quality, concentration and type of coagulant influence the coag-flocculation process [5,6,7].

Coag-flocculation process using inorganic coagulants has been well documented, but less attention has been given to the use of organic coagulant of animal origin like snail shell. The high cost, post usage handling and health issues associated with the inorganic coagulants like alum continually present serious challenges. It has been reported that extensive dosage of alum causes Alzheimer's disease [8]. Researchers have shown interest in the use of natural organic derivative like oxidized starch [5], cocoyam [9], Periwinkle shell [10], etc for the treatment of waste water. The recent search for better alternatives to conventional coagulants, such as those of biological origin has become extremely vital, considering their environmental friendliness. These natural organic derivatives are non-toxic, and biodegradable [11,12] which makes their application very probable.

The study in this work is focused on snail shell. Snails belong to the phylum molluscs and to the class gastropods which; includes the slugs and snails. The shell has a brownish color with a characteristics stripe pattern. The main constituent of the shell is calcium carbonate which consists of two crystalline forms such as calcite and aragonite. The other is organic matrixes which constitute of a protein known as conchiolin which consists 5% of the shell. A study on the fine structure of molluscs shells, using various techniques, including scanning electron microscope of broken surfaces revealed blocks or stripes of calcium carbonate separated by a thin layer of conchiolin [13].

Based on these constituents, snail shell can be used as either a coagulant or adsorbent. In West African countries, particularly, Nigeria, snails are found virtually everywhere in the surroundings. Snail serves as food but their shells are not edible and form waste. This waste can be converted to a commodity by using it for wastewater treatment. Apart from wastewater treatment can be extended to quarry effluent.

Quarry effluent was analysed and the result indicated the presence of high suspended colloidal particles with average turbidity of 2840.0 NTU. It has pH > 11.0, TSS of 450.0 mg/L, TDS > 1000.0, alkalinity > 1000.0, total hardness > 10000.0, calcium > 9000.0 and COD of 66.20 mg/L (see methodology- 3.1).

In the present work, investigation was conducted to determine coagulant efficiency. This demonstrates the potential of application of pulverized snail shell coagulant (PSSC) for clarification of quarry effluent. It also investigated the effect of dosage, pH as well as blend of PSSC with alum. It further studied the process kinetics.

2. COAG-FLOCCULATION THEORETICAL AND MATHEMATICAL PRINCIPLES

The clusters-size distribution of colloidal particles as time evolves is described by the Smolunchowski equation [14],

$$\frac{dN_n}{dt} = \frac{1}{2} \sum_{j=n-i} K_{ij} N_i N_j - N_n \sum_{i=1}^{\infty} K_{in} N_i \quad (1)$$

Where $N_n(t)$ is the time-dependent number concentration of n-fold clusters, t is the time and K_{ij} are the elements of the rate kernel which control the rate of coagulation between an i-fold and an j-fold clusters [14]. However, from Smolunchowski approximation, the coagulation is entirely controlled by Brownian diffusion and the coagulation rate constant for dimer formation of an initially monodisperse suspension is then given by [14],

$$K_{11} = \frac{8K_B T}{3\eta} \quad (2)$$

Where K_B is the Boltzmann constant, T is the temperature, and η is the viscosity of the medium. It has been reported that assuming a constant kernel i.e $K_{ij} = K_{11}$, the Smolunchowski Equation 1 can be solved to obtain the expression [14],

$$\frac{N_n(t)}{N_0} = \frac{(K_{11} N_0 t / 2)^{n-1}}{(1 + K_{11} N_0 t / 2)^{n+1}} \quad (3)$$

Where N_0 is the initial particle concentration. For $n = 1$, i.e monomer, the following linear functions in time for the inverse square root of monomer concentration N_1 is obtained.

$$\frac{1}{\sqrt{N_1}} = \frac{1}{\sqrt{N_0}} \left(1 + \frac{K_{11} N_0 t}{2} \right) \quad (4)$$

From Equation 4, a plot of inverse square root of monomer concentration N_1 vs t should give a straight line graph. The coagulation rate constant can be evaluated from the slope provided the initial concentration, N_0 , is known.

Equation 3, for monomer, gives;

$$N_1 = \frac{2N_0}{(2 + K_{11} N_0 t)^2} \quad (5)$$

For dimer, Equation 3 gives;

$$N_2 = \frac{2 K_{11} N_0^2 t}{(2 + K_{11} N_0 t)^3} \quad (6)$$

For trimer, Equation 3 gives;

$$N_3 = \frac{2N_0^3 (K_{11} t)^2}{(2 + K_{11} N_0 t)^4} \quad (7)$$

Also, the rapid coagulation rate constant, K_R is given by Smoluchowski as:

$$K_R = \frac{4K_B T}{3\eta} \quad (8)$$

Combining Equation 2 and 9 gives,

$$K_R = 1/2 K_{11} \quad (9)$$

Furthermore, from the constant kernel solution of Equation 3, it become necessary to introduce a coagulation time, when the total number concentration is reduced by a factor of 2,

$$\tau_{1/2} = \frac{2}{K_{11} N_0} \quad (10)$$

This coagulation time represents a useful time scale for the identification of the early stages in the coagulation process. Denoting the number of concentration of n- fold aggregates at reduced time, $\tau_{1/2}$ as, $N_n(\tau_{1/2})$ and $\tau_{1/2} = \frac{2}{K_{11} N_0}$, Equation 5 – 7 becomes;

$$N_1 = N_0 \left[\frac{1}{(\tau_{1/2} + t)^2} \right] \quad (11)$$

$$N_2 = 2N_0 \left[\frac{t/\tau_{1/2}}{(\tau_{1/2} + t)^3} \right] \quad (12)$$

$$N_3 = 4N_0 \left[\frac{(t/\tau_{1/2})^2}{(\tau_{1/2} + t)^4} \right] \quad (13)$$

Turbidity (NTU) was converted to TSS (mg/L) using Equation 14 and Coag-flocculation performance was evaluated with Equation 15.

$$\text{TSS (mg/L)} = (\text{TSS}_t) \cdot T \quad (14)$$

Where T = Turbidity in NTU; (TSS_t) = Conversion factor to TSS = 2.35 [2]

$$E(\%) = \left(\frac{N_0 - N_1}{N_0} \right) \times 100 \quad (15)$$

Where

E = Coag-flocculation Performance in percentage.

N_0 = Average Initial Particle Concentration = 6674.0mg/L

3. MATERIALS AND METHODS

3.1 Collection and Methods of Analyses of Quarry Effluent

The quarry effluent was collected from quarry factory located in Abakaliki, Nigeria. The chemical oxygen demand was determined at the Regional Water Laboratory, Enugu. Other parameters were determined at Enugu State Water Corporation Laboratory. The pH, electrical conductivity and turbidity were determined using Mettler Toledo Delta 320 pH Meter, El Digital Conductivity Meter (model number 161) and El Digital Turbidity Meter (model no. 337), respectively.

3.2 Preparation of Coagulants Stock Solutions

3.2.1 Preparation of pulverized snail shell coagulant (PSSC)

Snail shells were collected from popular snail meat shop in Enugu, Nigeria. The shell samples collected were washed, dried properly and homogenized into fine powder to ensure large surface area. The ground snail shell was sieved using 0.1 mm diameter sieve. The sieved snail shell was processed into a pulverized snail shell coagulant (PSSC) using standard methods reported by Fernandez - Kim [15].

3.2.2 Preparation of alum solution

A 2.5 g of aluminum sulphate was weighed and dissolved in 100 mL of distilled water and made up to a liter to give 2.5 g/L stock solution.

3.3 Jar Test

The conventional jar test procedure was employed using 2 mins of rapid mixing at 300 rpm, followed by 20 mins of slow mixing at 100 rpm. The volume of wastewater used for the study was 500 mL. The solution was poured into 500 mL cylinder after stirring and allowed to settle for 30 mins. A 20 mL of the supernatant was pipetted at 2 cm depth at 5 mins interval. The turbidity of the supernatants was measured and recorded. The concentrations of 100, 200, 300, 400, 500, and 200, 400, 600, 800, 1000 mg/L alum and PSSC were dosed respectively.

4. RESULTS AND DISCUSSION

4.1 Coag-flocculation Performance Plots

The coag-flocculation performances of alum, PSSC and the blend of the two were determined using Jar test. This was aimed at obtaining the optimum pH value and dosage.

The results of coag-flocculation performance of varying dosages of alum and PSSC from 100 to 500 and 200 to 1000mg/L respectively are presented in Fig. 1. In Fig. 1a, it was observed that the minimum and the maximum percentage turbidity removal were achieved to 75.1% with 100 mg/L and 94.8% with 200 mg/L alum. Fig. 1b shows minimum and maximum turbidity removal of 76.4% with 400mg/L and 83.7% with 600mg/L PSSC, respectively.

The coag-flocculation performance in Fig. 1a showed steady increase of turbidity removal with time at the optimum dosage of 200mg/L alum. This could be attributed to charge neutralization and sweep flocculation; a mechanism where particles can be destabilized so that agglomeration could occur [16]. The coag-flocculation performance in Fig. 1b at the optimum dosage of 600mg/L PSSC initially showed slow flocculation with enhanced performance observed between 25 to 30min. This could be due to relaxation and re-conformation of the polymer molecules on the surface of PSSC, thereby providing favourable spatial environment [17].

The coag-flocculation performances of optimum dosages of alum (200 mg/L) and PSSC (600mg/L) were investigated while varying quarry effluent pH values. The results were plotted in Fig. 2. It was observed in Fig. 2a that the minimum and maximum percentage turbidity removal of 84.4% and 95.0% for QE pH values of 2.0 and 6.0 respectively were recorded using optimum alum dosage. Fig. 2b showed that the minimum and maximum percentage turbidity removal of 81.5% and 84.9% for QE pH values of 2.0 and 8.0 respectively were observed using optimum PSSC dose. This clearly highlighted the effect of pH in coag-flocculation, considering the fact that pH determines the solubility and the formation of cation species like Al^{3+} , $Al(OH)^{2+}$ etc [2]. The formation of these cations avails positive charged ions that quickly neutralize the negatively charged particles. The result further affirms the reported effective pH for alum to be

5.0 to 7.0 [18,19]. The steady increased and improved turbidity removal in Fig. 2b compared with Fig. 1b showed that the pH could have quickened the polymer relaxation and re-conformation process.

The coag-flocculation performances of the blend of alum and PSSC and the optimum blend varying QE pH values were investigated. The results are shown in Fig. 3. Figs. 3a and 3b showed similar trends throughout the graphs and revealed an increased turbidity removal when compared to the use of only alum or PSSC. In Fig. 3a, it was observed that the minimum and maximum percentage turbidity removal of 80.3% with 100 mg/L alum and 1000 mg/L SSC and 95.2% with 200 mg/L alum and 800 mg/L PSSC, respectively were achieved. Fig. 3b indicated minimum and maximum turbidity removal of 87.8% and 98.5% for QE pH of 2.0 and 6.0 respectively for optimum blend of 200mg/L alum and 800mg/L PSSC.

Figs. 3a and 3b showed that at optimum blend and pH, the coag-flocculation achieved more than 95% turbidity removal in 5 mns. This could be associated with the sweep floc caused by charge neutralization and inter-particle bridging. These resulted in fast agglomeration and settling [2,20,21].

The greater increase in turbidity removal witnessed for the blend of alum and PSSC highlighted the potential of PSSC as coagulant aid for the conventional coagulant alum.

Fig. 4 showed the bar chart presentation of coag-flocculation performance for optimum values

after 30 mns settling time. It showed that upon pH variation an increase in coag-flocculation performance was observed. The blend showed the best performance.

4.3 Coag-flocculation Kinetics

The kinetic plots for the coag-flocculation performance of alum, PSSC and their blends are shown in Figs. 5-7. The plots were based on Equation 4. The rate constant, K_{11} and regression coefficient, R^2 were obtained from the plot of $1/\sqrt{N_1}$ vs t. Both K_{11} and R^2 were recorded in Tables 1-6. Tables 1-6 showed maximum and minimum coag-flocculation rate constant, K_{11} , of $2.957E-5$ at pH of 6.0 for 200 mg/L alum and 800 mg/L PSSC and $5.310E-7$ with 1000 mg/L PSSC, respectively. The coagulation time, $\tau_{1/2}$, which is time taken for the initial concentration of colloidal particles to reduce by half was calculated from Equation 10. It was observed that the coagulation rate constants vary inversely with the coagulation time i.e higher K_{11} goes with lower $\tau_{1/2}$. This is an established relationship between rate constant and $\tau_{1/2}$ [9,10,22].

Rapid coagulation constant K_R was calculated from Equation 9 and the variation in K_R values were minimal due to insignificant change in temperature and viscosity of the effluent medium.

R^2 was used to evaluate the adequacy of the experimental data to the main model as expressed in Equation (4). Most of the R^2 values in Tables 1-6 were very high (> 0.9), showing high level of correlation between the variables (N_1 and t), hence the adequacy of the model.

Table 1. Coa-flocculation parameter for alum – dosage variation

Parameter	100 mg/L	200 mg/L	300 mg/L	400 mg/l	500 mg/L
R^2	0.8757	0.9579	0.9585	0.9675	0.9913
K_{11} (L/mg.s)	$2.714E-6$	$1.990E-5$	$5.300E-6$	$6.350E-6$	$2.475E-6$
K_R	$1.357E-6$	$9.950E-6$	$2.650E-6$	$3.175E-6$	$1.238E-6$
$\tau_{1/2}$ (s)	150.3	20.0	76.95	64.25	164.83

Table 2. Coa-flocculation parameter for PSSC – dosage variation

Parameter	200 mg/L	400 mg/L	600 mg/L	800 mg/l	1000 mg/L
R^2	0.7387	0.9184	0.8484	0.5924	0.5838
K_{11} (L/mg.s)	$7.973E-7$	$4.196E-7$	$2.157E-6$	$1.259E-6$	$5.310E-7$
K_R	$3.987E-7$	$2.098E-7$	$1.079E-6$	$6.295E-7$	$2.655E-7$
$\tau_{1/2}$ (s)	511.73	972.45	189.18	324.1	768.38

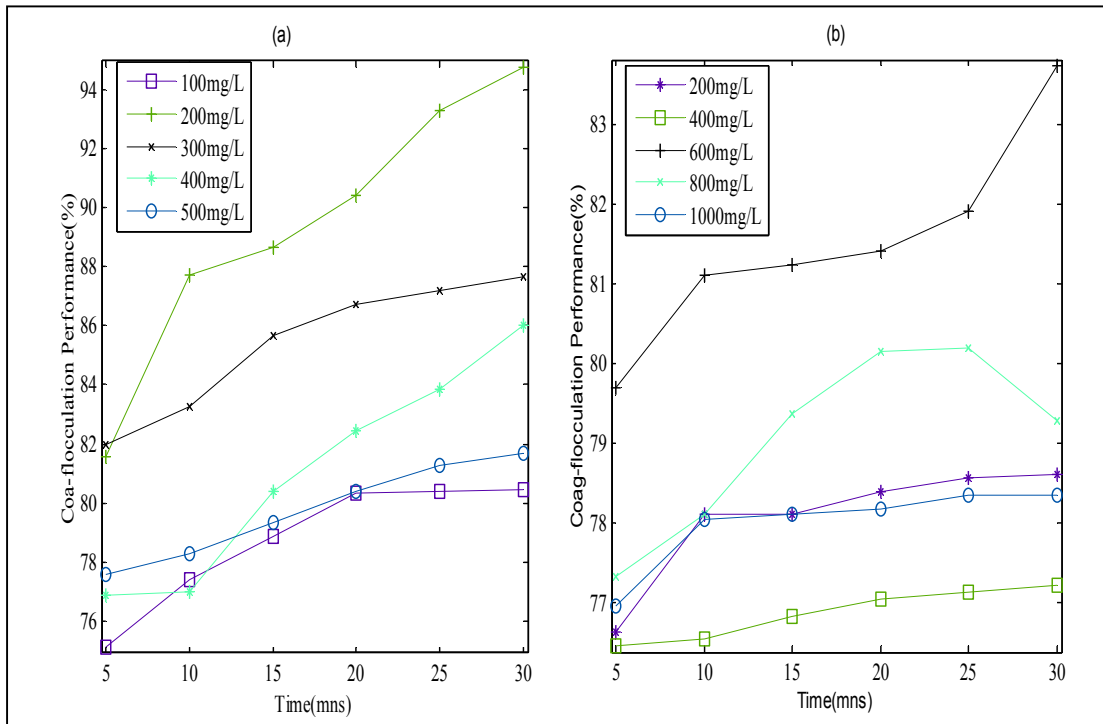


Fig. 1. (a) Coag-flocculation performance as a function of time for varying alum dosages, (b) Coag-flocculation performance as a function of time for varying PSSC dosages

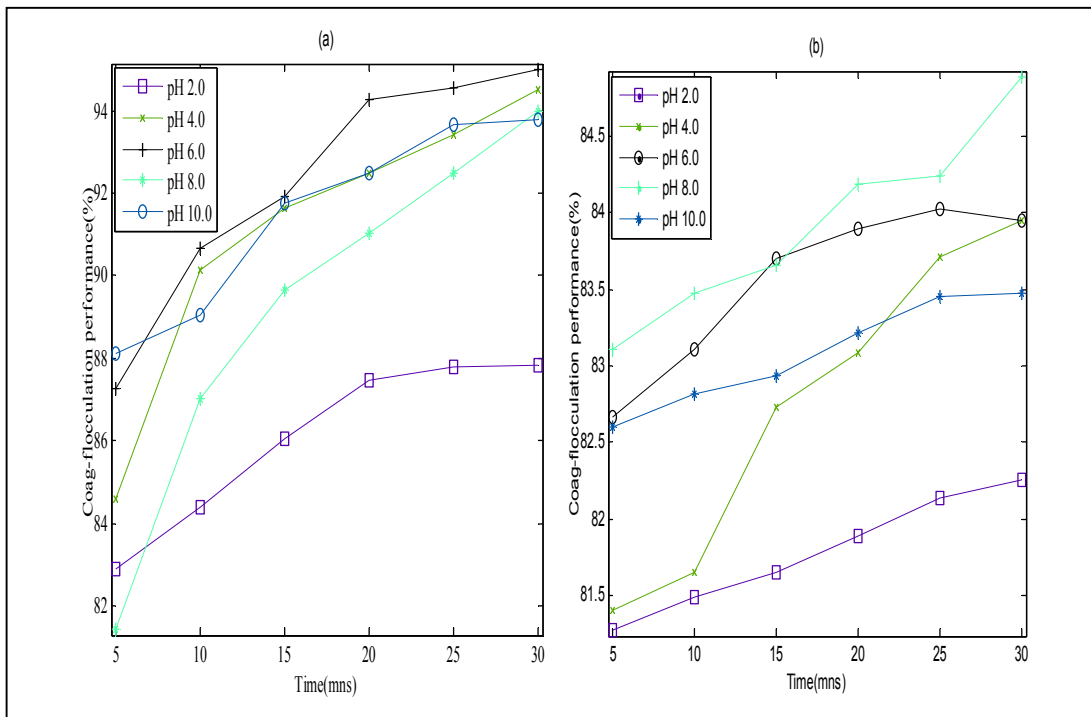


Fig. 2. (a) Coag-flocculation performance as a function of time for 200 mg/L alum at varying QE pH, (b) Coag-flocculation performance as a function of time for 600 mg/L PSSC at varying QE Ph

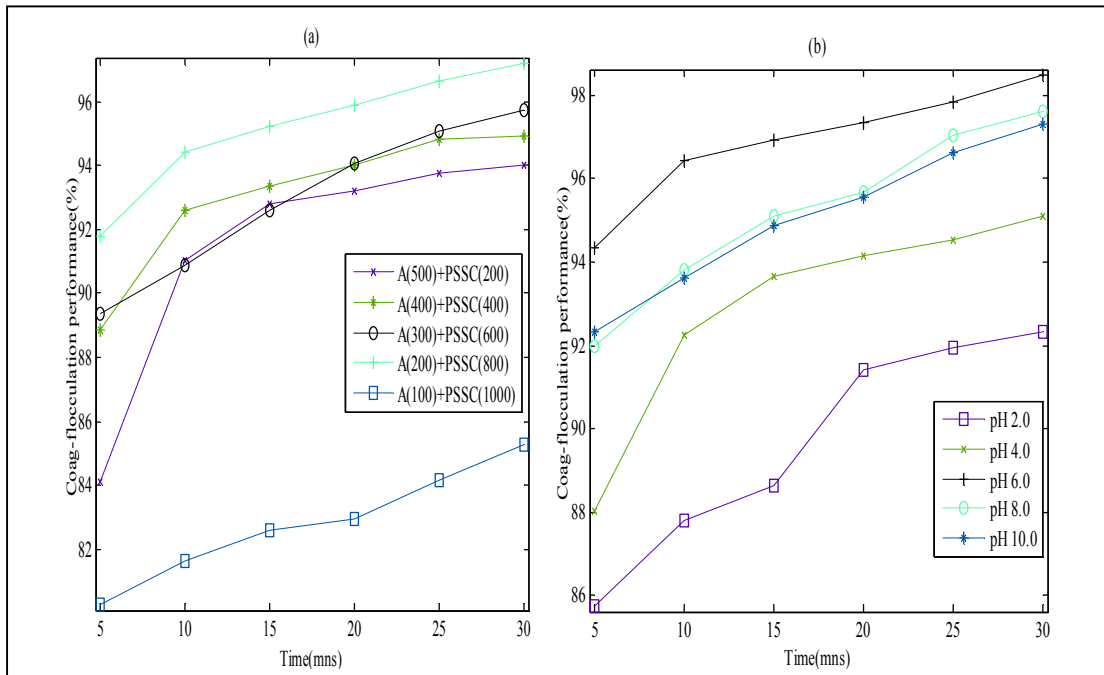


Fig. 3. (a) Coag-flocculation performance as a function of time for blends of alum and PSSC dosages, (b) Coag-flocculation performance as function of time for blend of 200 mg/L alum and 800 mg/L PSSC at varying QE pH

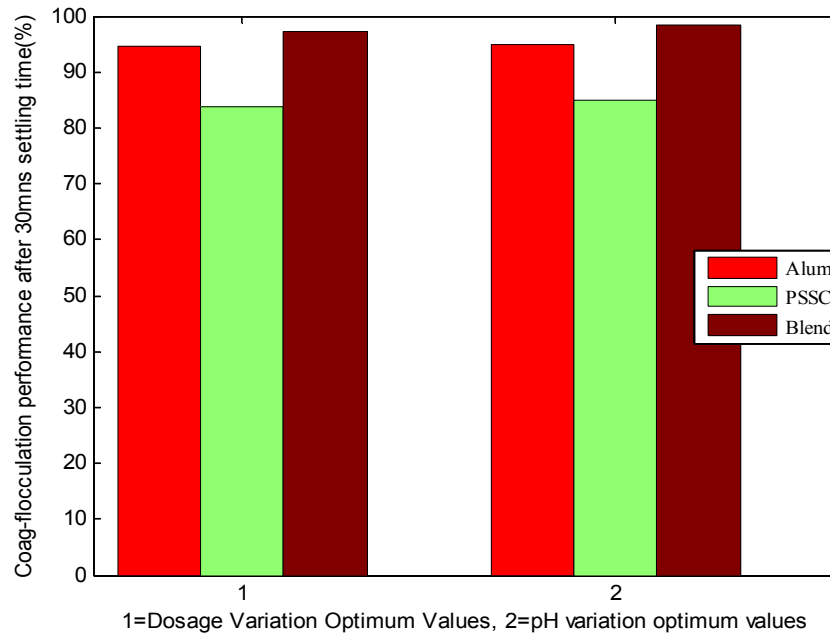


Fig. 4. Bar chart presentation of coag-flocculation performance for optimum values after 30 mns settling time showing effect of dosage and pH variation

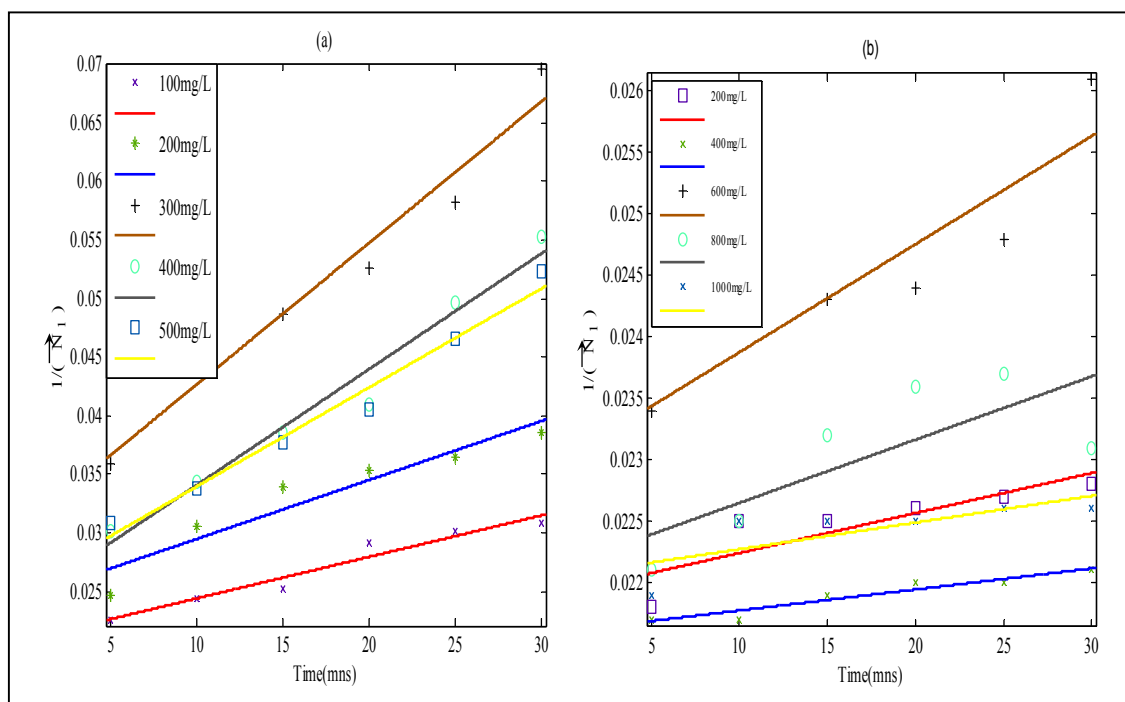


Fig. 5. (a) Inverse square root of number of monomer N_1 vs time for varying alum dosage, (b) Inverse square root of number of monomer N_1 vs time for varying PSSC dosage

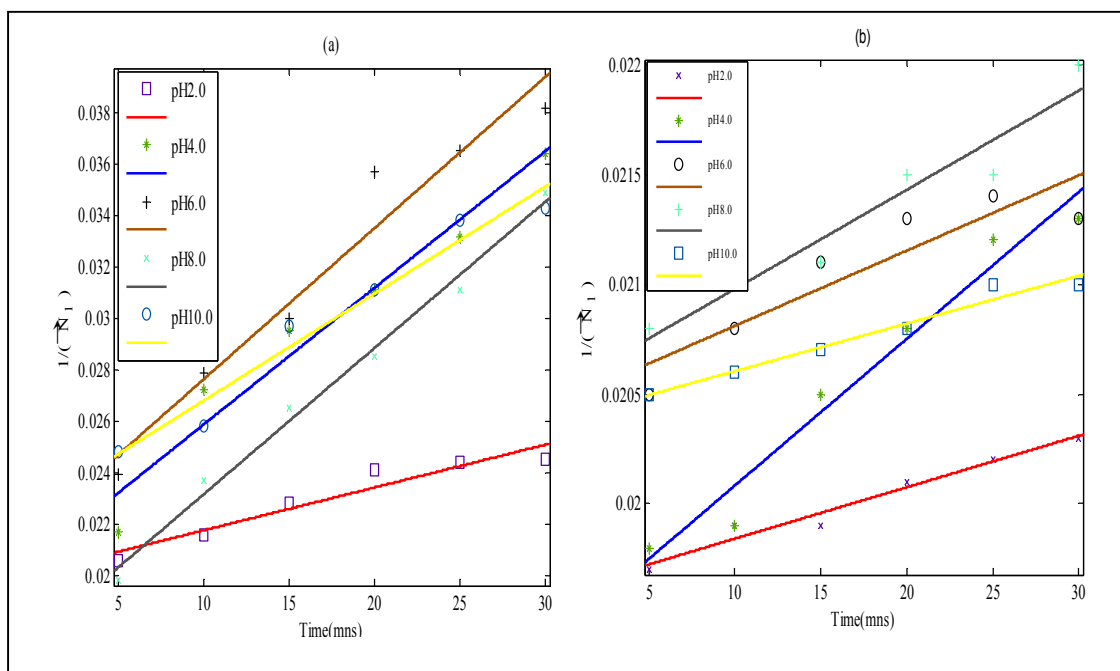


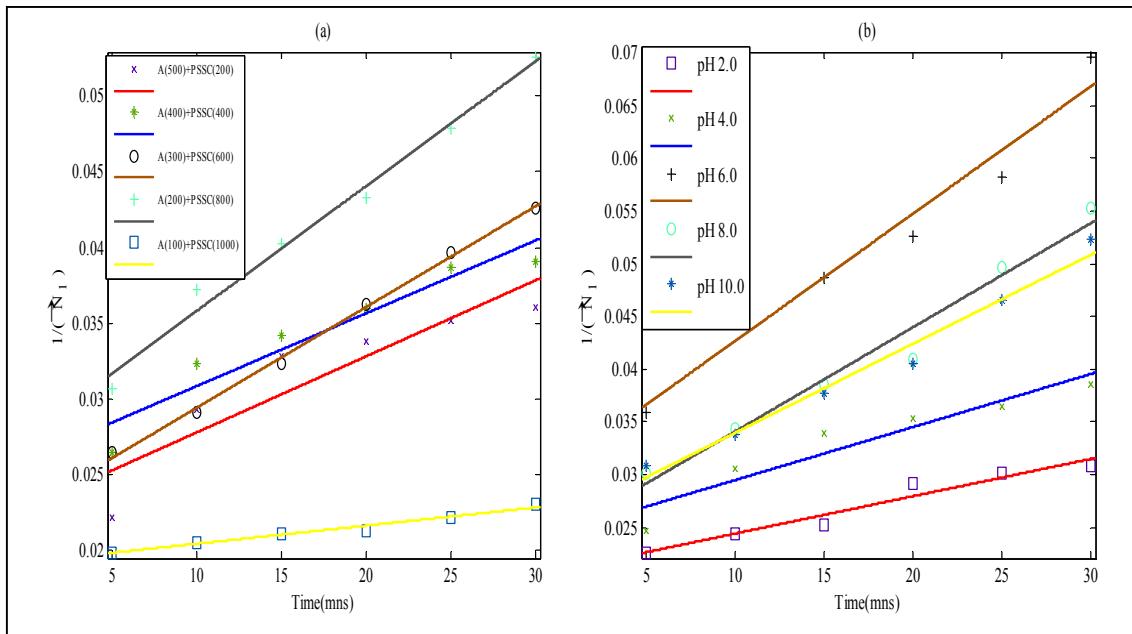
Fig. 6. (a) Inverse square root of number of monomer N_1 vs Time for alum at 200 mg/L varying QE pH , (b) Inverse square root of number of monomer N_1 vs Time for PSSC at 600 mg/L varying QE pH

Table 3. Coa-flocculation parameter for alum at 200 mg/L – pH variation

Parameter	pH 2.0	pH 4.0	pH 6.0	pH 8.0	pH 10.0
R ²	0.9242	0.9580	0.9544	0.9902	0.9626
K ₁₁ (L/mg.s)	4.085E-6	1.302E-5	1.440E-5	1.395E-5	1.020E-5
K _R	1.25E-6	6.51E-6	7.200E-6	6.975E-6	5.100E-6
τ _{1/2} (s)	99.88	31.33	28.33	29.27	40.02

Table 4. Coa-flocculation parameter for PSSC at 600mg/L – pH variation

Parameter	pH 2.0	pH 4.0	pH 6.0	pH 8.0	pH 10.0
R ²	0.9641	0.9670	0.8385	0.9401	0.9670
K ₁₁ (L/mg.s)	1.634E-6	5.735E-7	8.390E-7	1.643E-6	5.315E-7
K _R	8.170E-7	2.868E-7	4.195E-7	8.215E-7	2.658E-7
τ _{1/2} (s)	249.7	711.47	486.32	369.97	767.68



**Fig. 7. (a) Inverse square root of number of monomer N₁ vs time for blend of alum and PSSC
(b) Inverse square root of number of monomer N₁ vs time for blend of alum(200) and PSSC(800) varying QE pH**

4.4 Particle Distribution Plot

Equations 11 –13 were used to predict the particle aggregation with time for alum, PSSC, and their blends at varying dosages. It also shows the particle aggregation with time for varying pH using optimum dosages of alum, PSSC, and the blend. The particle distribution N₁, N₂ and N₃ represent monomer, dimer and trimer particles, respectively. Fig. 8 shows the plots of the lowest and highest half-time recorded. In Fig. 8a, the curves represent the

expected particle distribution in a typical fast coag-flocculation process [22,23,24].

The sharp aggregation of monomer particle that occurred in less than 5mns of the process suggests instant charge neutralization mechanism leading to fast agglomeration. This is evident in the rise of dimer and trimer in the first 5 minutes since the formation of the two resulted from the charge neutralization and destabilization.

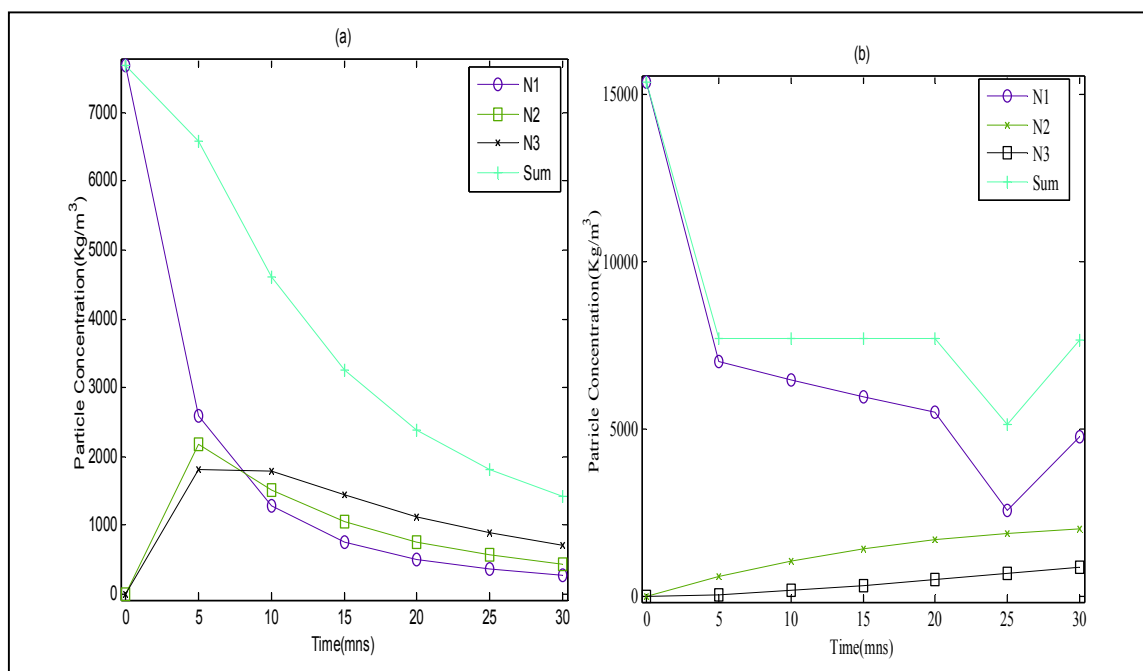


Fig. 8. (a) Particle distribution for $\tau = 13.8$ sec, (b) Particle distribution for $\tau = 972.45$ sec

Table 5. Coa-flocculation parameter for blend of alum and PSSC – dosage variation

Parameter	500/200	400/400	300/600	200/800	100/1000
R^2	0.8286	0.9145	0.9974	0.9877	0.8286
K_{11} (L/mg.s)	1.234E-5	1.180E-5	1.632E-5	2.024E-5	1.234E-5
K_R	6.170E-6	5.900E-6	9.660E-6	1.012E-5	6.170E-6
$\tau_{1/2}$ (s)	33.08	34.56	25.00	20.15	33.08

Table 6. Coa-flocculation parameter for blend of alum (200 mg/L) and PSSC (800 mg/L) – varying QE pH

Parameter	pH 2.0	pH 4.0	pH 6.0	pH 8.0	pH 10.0
R^2	0.9489	0.9024	0.9714	0.9610	0.9770
K_1 (L/mg.s)	8.660E-6	1.225E-5	2.957E-5	2.418E-5	2.067E-5
K_R	4.330E-6	6.125E-6	1.479E-5	1.209E-5	1.034E-5
$\tau_{1/2}$ (s)	47.12	33.1	13.8	16.87	19.73

Fig. 8b showed very small increase in dimmer and trimmer formation with time. This suggested very low charge neutralization mechanism with low bridging. Thus, these gave rise to slow particle aggregation and settling [23,24].

5. CONCLUSION

The results of the study indicate that PSSC has strong potential for treatments of effluent particularly quarry effluent. It also performed better when combined with alum. This implies that PSSC can be a good coagulant aid when combined with alum. This is supported by high

turbidity removal efficiency range of $87.8 \leq E \leq 98.5$. The optimum condition was observed with the blend of 200 mg/L alum and 800mg/L PSSC at pH 6.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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