

Original Research Article**KINETICS AND PARTICLE REMOVAL PROFILE OF SNAIL SHELL – ALUM INDUCED COAG-FLOCCULATION OF QUARRY EFFLUENT.****ABSTRACT**

This study investigated the coag-flocculation performance of alum, snail shell coagulant (SSC), 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, K_{11} , coagulation half-time, $\tau_{1/2}$ etc were determined. The optimum pH was observed at 6.0, while the blend of 200.0mg/L alum and 800.0mg/L SSC 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. The use of SSC blended with alum showed high level of potential, for the treatment of quarry effluent.

Keywords: Snail shell, Quarry effluent, Coagulation/flocculation, Jar test

1.0 INTRODUCTION

The problem of providing sufficient quantities of high quality water to satisfy our domestic, industrial and agricultural needs have been a global concern. The continuous increase in population size, climate change and environmental pollution only exacerbate the situation.

It is worthy to note that there is no physical shortage of water on the planet earth. It covers 70% of the globe. However, 97% of the world water is saline and is thus, non-drinkable. 2% is locked in glacier and polar ice caps. This leaves 1% to meet humanity needs [1]. Out of this 1% left, human activities and climate change introduced turbidity and other pollution, resulting on 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 addition of coagulants to wastewater to promote destabilization of the

colloid dispersion and agglomeration of the resulting individual colloidal particles [3], [4]. Among other factors, temperature, pH, effluent quality, coagulant concentration and type influence the coag-flocculation process [5], [6], [7]

The 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. Also, 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 intake of alum causes Alzheimer's disease [8]. Thus, 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, especially those of biological origin has become extremely vital, considering their environmental friendliness. These natural organic derivatives are non-toxic, and biodegradable [11], [12] and these made their application very promising.

In present work, snail shell provides the focus of the study. Snails belong to the phylum molluscs and to the class gastropods; this class 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 is either of two crystalline forms: calcite and aragonite. The other is organic matrixes which constitute of a protein known as conchiolin. This makes up to 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 existing results, Snail shell can be used as either a coagulant or an adsorbent. Also, in West African countries, particularly, Nigeria, snails are found virtually everywhere in our surroundings. Snail serves as food but the shell are not edible and it becomes waste. This waste can be converted to wealth by using it for wastewater treatment among many other applications. Its application for 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 2840.0NTU. It has pH > 11.0, TSS of 450.0mg/L, TDS > 1000.0, alkalinity > 1000.0, total hardness > 10000.0, calcium > 9000.0 and COD of 66.20mg/L

In the present work, attempt was made to determine coagulant efficiency, and thus, was able to demonstrate the effectiveness of the application of SSC for clarification of Quarry effluent. It also investigated the effect of dosage, pH as well as blend of SSC with alum. It further studied the process kinetics.

2.0 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.

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

Where T = Turbidity in NTU; (TSS_f) = 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

3.0 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

Preparation of Snail Shell Coagulant (SSC)

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 0.1mm diameter sieve. The sieved snail shell was processed into a snail shell coagulant (SSC) using standard methods reported by Fernandez - Kim [15].

Preparation of alum solution

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

3.3 Jar Test

The conventional jar test procedure was employed. 2min of rapid mixing at 300rpm, followed by 20 min of slow mixing at 100rpm was adapted. The volume of wastewater used for the study was 500mL. After stirring, the solution was carefully poured into 500mL cylinder and allowed to settle for 30mins. 20mL of the clear upper layer of the settling solution was pipetted at 2cm depth at 5mins interval. The turbidity of these supernatants were measured and recorded. 100, 200, 300, 400, 500, and 200, 400, 600, 800, 1000mg/L alum and SSC were respectively, used.

4.0 RESULT AND DISCUSSION

4.1 Coag-flocculation Performance Plots

The coag-flocculation performances of alum, SSC, 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 SSC from 100 to 500 and 200 to 1000mg/L respectively are presented in **Figure 1**. In **Figure 1a**, it was seen that the minimum and the maximum percentage removal of turbidity achieved was 75.1% for 100mg/L and 94.8% for 200mg/L alum. **Figure 1b** recorded minimum and maximum turbidity removal of 76.4% for 400mg/L and 83.7% for 600mg/L SSC, respectively.

The coag-flocculation performance recorded in **Figure 1a**, showed steady increase of turbidity removal with time at the optimum dose 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 recorded in **Figure 1b** particularly the optimum dose of 600mg/L SSC, showed initial slow flocculation, but enhanced performance was observed between 25 to 30min. This could be due to relaxation and re-conformation of the polymer molecules on the surface of SSC, thereby providing favourable spatial environment [17].

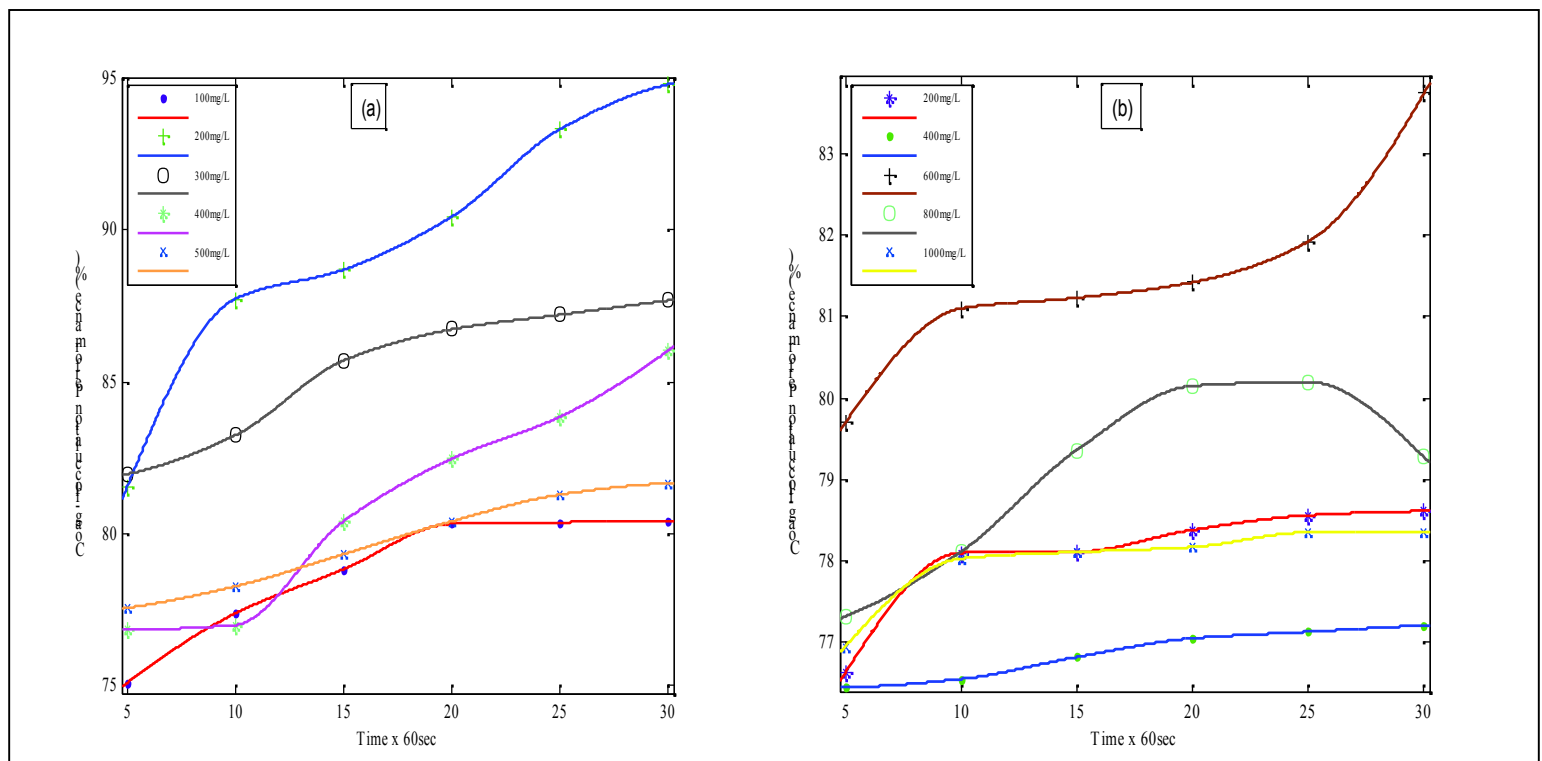


Figure 1:(a) Coag-flocculation performance as a function of Time for varying alum doses, (b) Coag-flocculation performance as a function of time for varying SSC doses

The coag-flocculation performances of optimum doses of alum (200mg/L) and SSC (600mg/L) were investigated while varying QE pH values. The results were depicted in **Figure 2**. It was observed in **Figure 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 dose. **Figure 2b** recorded 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 SSC 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+} , $\text{Al}(\text{OH})^{2+}$ etc[2]. The formation of these cations, makes available positive charged ions that quickly neutralizes 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 **Figure 2b** as against **Figure 1b** suggested that the pH could have quickened the polymer relaxation and re-conformation process.

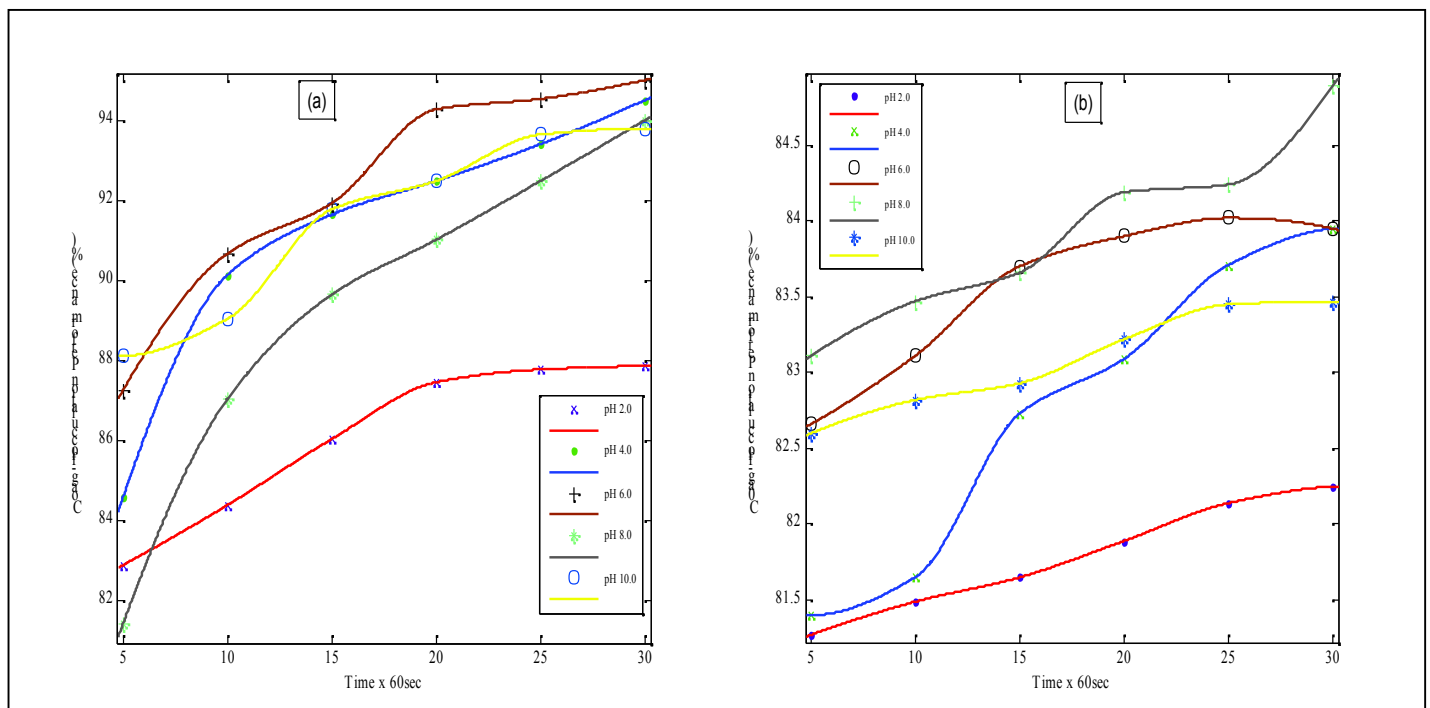


Figure 2: (a) Coag-flocculation Performance as a function of time for 200mg/L alum @ varying QE pH, (b) Coag-flocculation Performance as a function of time for 600mg/L SSC @ varying QE pH

The coag-flocculation performances of the blend of alum + SSC and the optimum blend varying QE pH values were investigated. The results were recorded in **Figure 3**. **Figures 3a and 3b** showed similar trends throughout the graphs and revealed an increased turbidity removal when compared to the use of only alum or SSC. In **Figure 3a**, it was observed that the minimum and maximum percentage turbidity removal of 80.3% for 100mg/L alum + 1000mg/L SSC and

95.2% for 200mg/L alum + 800mg/L SSC, respectively were recorded for the blends. **Figure 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 + 800mg/L SSC.

A careful look at **Figures 3a and 3b** showed that at optimum blend and pH, the coag-flocculation achieved more than 95% turbidity removal in 5min. This could not be unconnected with the sweep floc occasioned by charge neutralization, colloids entrapment and interparticle bridging. These resulted in fast agglomeration and settling [2], [20], [21].

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

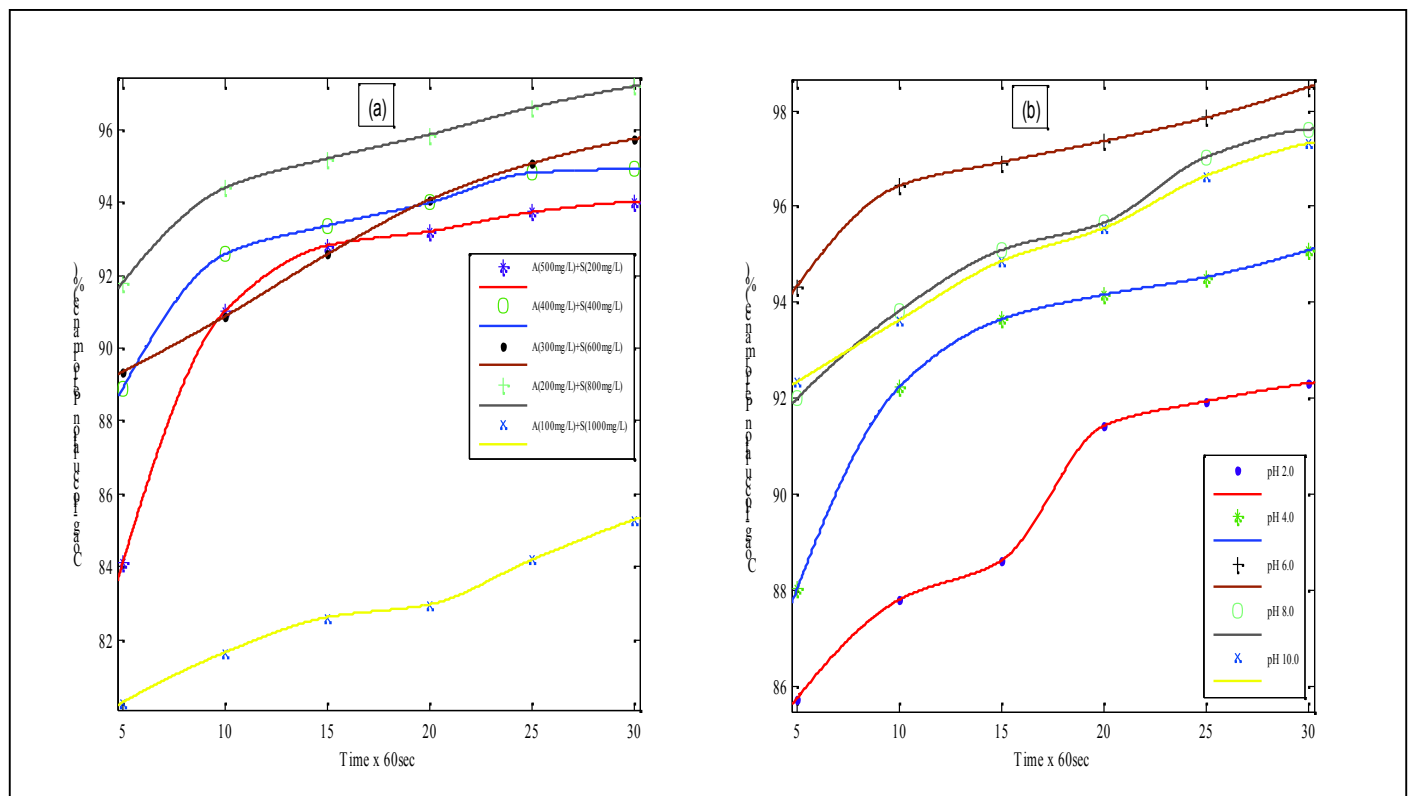


Figure 3: (a) Coag-flocculation performance as a function of time for blends of alum and SSC doses, (b) Coag-flocculation Performance as function of time for blend of 200mg/L alum + 800mg/L SSC @ varying QE pH

4.3 Coag-flocculation Kinetics

The kinetic plots for the coag-flocculation performance of alum, SSC and their blends are shown in **Figures 4-6**. 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** indicated maximum and minimum coag-flocculation rate constant, K_{11} , of $2.957\text{E-}5$ at pH of 6.0 for 200mg/L alum + 800mg/L SSC and $5.310\text{E-}7$ for 1000mg/L SSC, 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 insignificant change in temperature and viscosity of the effluent medium.

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

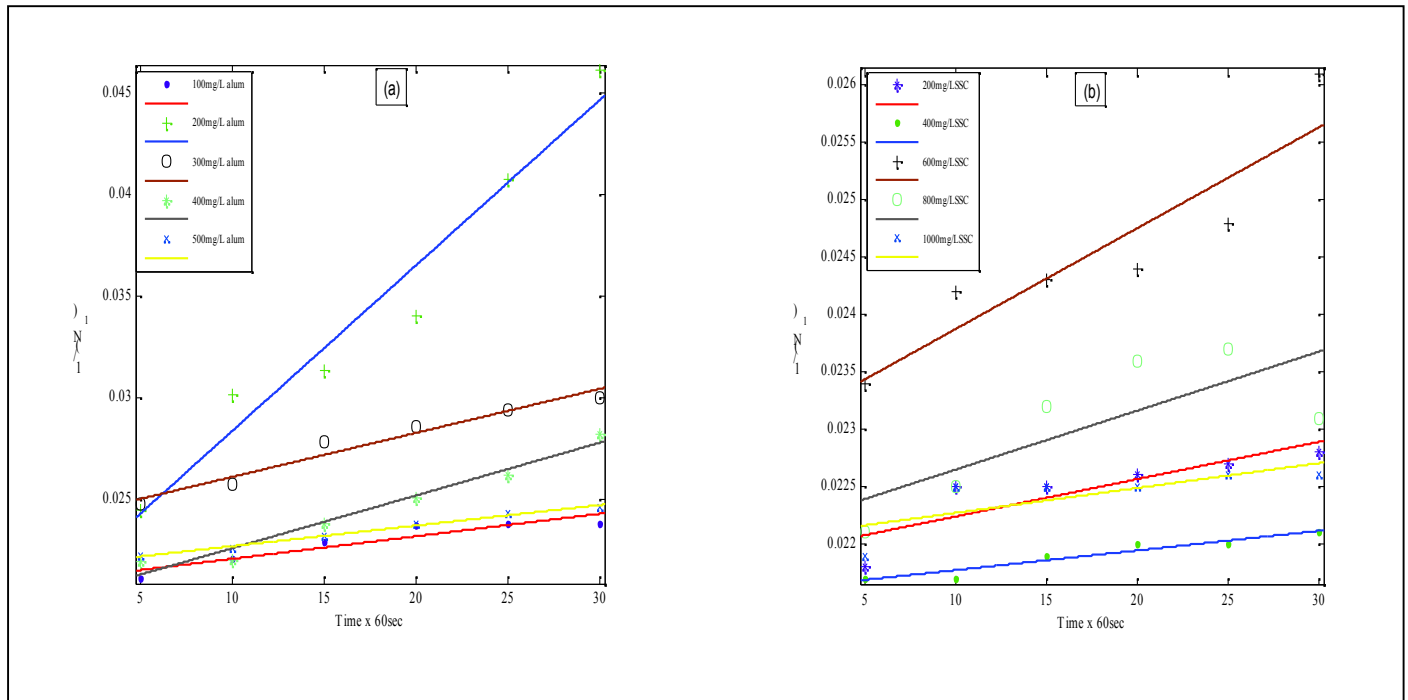


Figure 4: (a) Inverse square root of number of monomer N_1 vs Time for varying alum dose, (b) Inverse square root of number of monomer N_1 vs Time for varying SSC dose

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

Parameter	100mg/L	200mg/L	300mg/L	400mg/L	500mg/L
R^2	0.8757	0.9579	0.9585	0.9675	0.9913
$K_{11}(\text{L/mg.s})$	$2.714\text{E-}6$	$1.990\text{E-}5$	$5.300\text{E-}6$	$6.350\text{E-}6$	$2.475\text{E-}6$

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 SSC – dosage variation

Parameter	200mg/L	400mg/L	600mg/L	800mg/l	1000mg/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

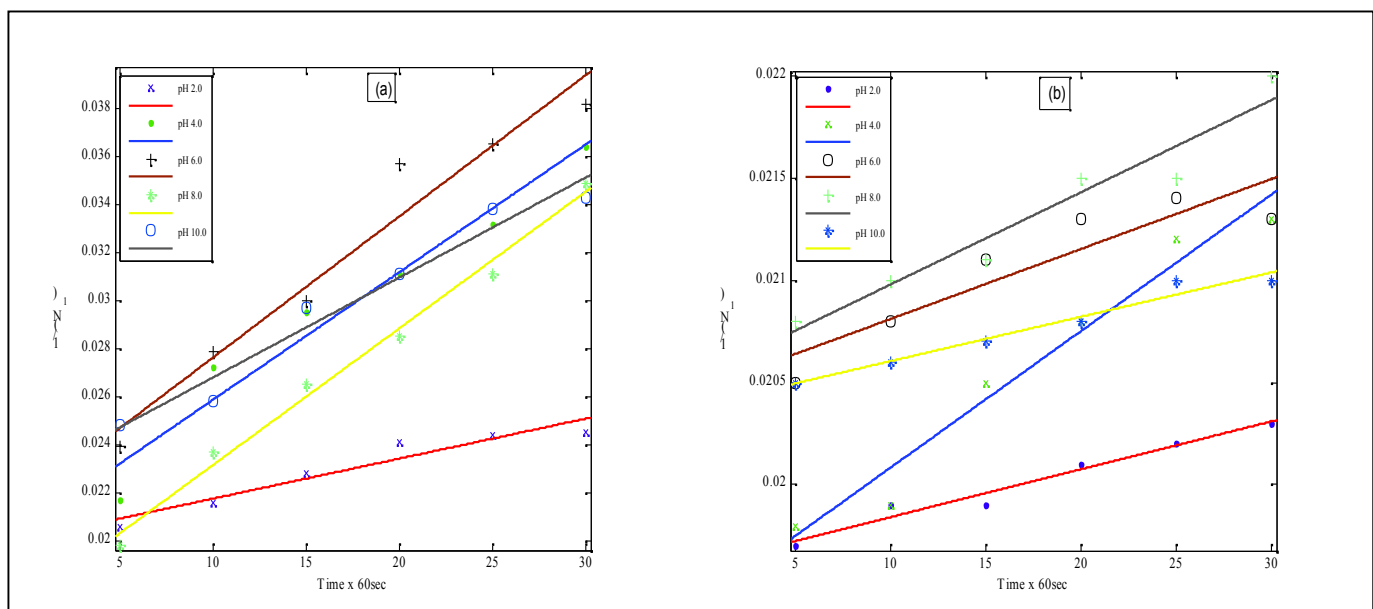


Figure 5: (a) Inverse square root of number of monomer N_1 vs Time for alum @ 200mg/L varying QE pH , (b) Inverse square root of number of monomer N_1 vs Time for SSC @ 600mg/L varying QE pH

Table 3: Coa-flocculation parameter for alum @ 200mg/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
$K_{11}(\text{L/mg.s})$	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
$\tau_{d,2}(s)$	99.88	31.33	28.33	29.27	40.02

Table 4: Coa-flocculation parameter for SSC @ 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
$K_{11}(\text{L/mg.s})$	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
$\tau_{d,2}(s)$	249.7	711.47	486.32	369.97	767.68

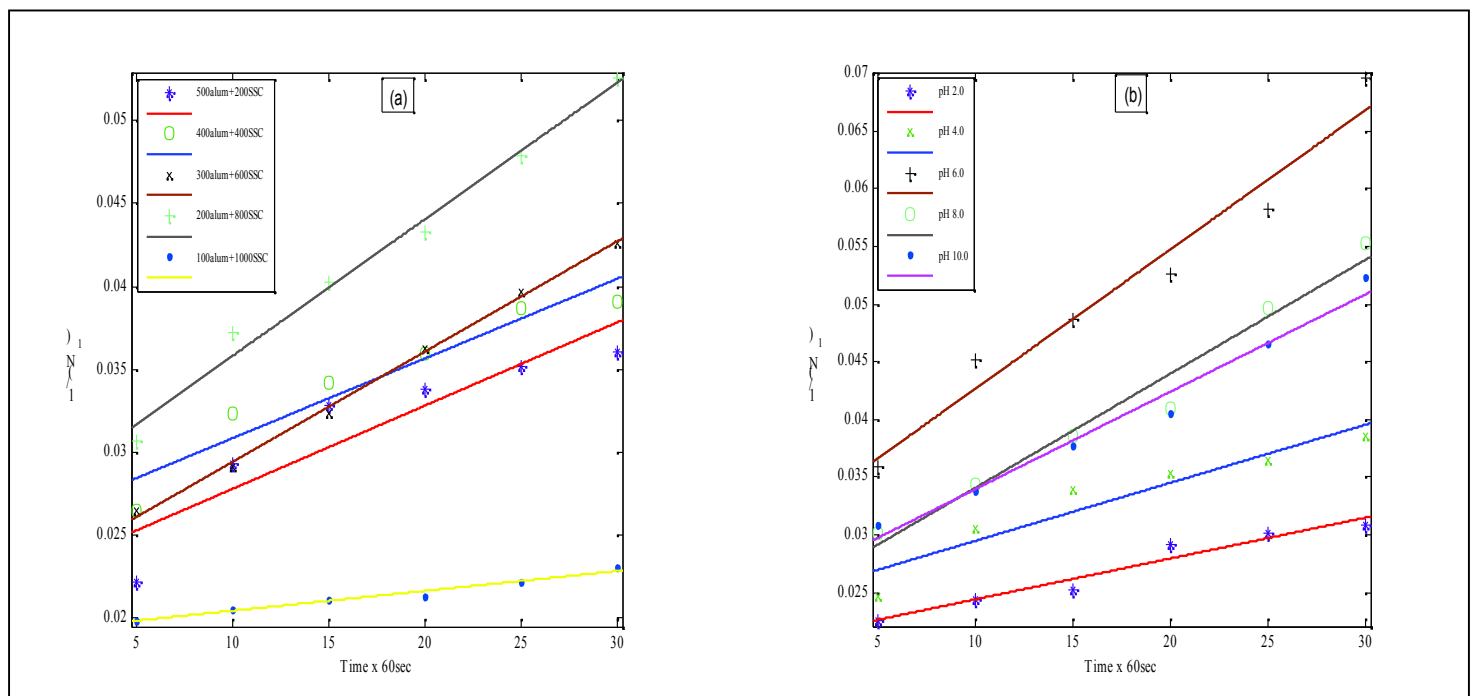


Figure 6: (a) Inverse square root of number of monomer N_1 vs Time for blend of alum and SSC (b) Inverse square root of number of monomer N_1 vs Time for blend of alum(200) + SSC(800) @ varying QE

Table 5: Coa-flocculation parameter for blend of alum and SSC – dosage variation

Parameter	800/ 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 (200mg/L) and SSC (800mg/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

4.4. Particle Distribution Plot

Equations 11 –13 were used to predict the particle aggregation with time for alum, SSC, and their blends at varying dosages. Also, the particle aggregation with time for varying pH using optimum dosages of alum, SSC, and the blend were studied. The particle distribution N_1 , N_2 and N_3 represent monomer, dimmer and trimer particles, respectively. **Figure 7** indicates the plots of the lowest and highest half-time recorded. In **Figure 7a**, the curves represent the expected particle distribution in a typical fast coag-flocculation process [22], [23], [24].

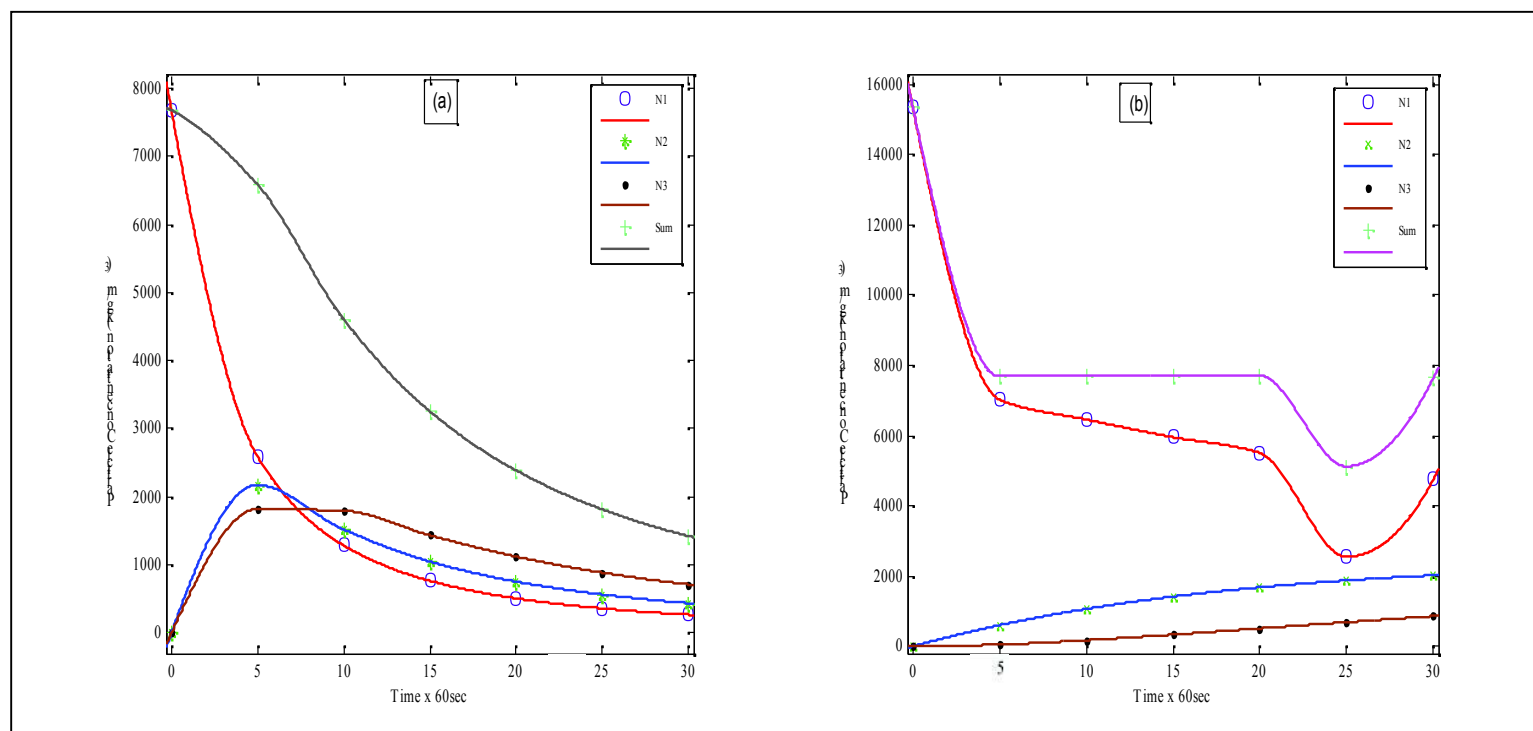


Figure 7: (a) Particle Distribution for $\tau = 13.8\text{sec}$, (b) Particle Distribution for $\tau = 972.45\text{sec}$.

The sharp aggregation of monomer particle that occurred in $< 5\text{min}$ of the process suggests instant charge neutralization and destabilization mechanism leading to fast agglomeration. This is evident in the rise of dimmer and trimmer in the first 5 minutes since the formation of the two resulted from the charge neutralization and destabilization.

Figure 7b indicated very small increase in dimmer and trimmer formation with time. This suggested very low charge neutralization and destabilization 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 SSC has strong potential for treatment of effluent particularly Quarry effluent. It also performed better when combined with alum. This implies that SSC 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 200mg/L alum and 800mg/L SSC at pH 6.

6.0 References

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