**Effect Of Mucuna Seed Coagulant On Total Dissolved Solid Particles Removal Efficiency In Pharmaceutical Effluent Medium By Coag-flocculation Process**

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**Abstract.:**In this study, total dissolved solid particles removal from pharmaceutical effluent by coag-flocculation process has been investigated at room temperature using various dosages of Mucuna Seed Coagulant. A conventional standard jar test method was employed for the turbidity measurement while Mucuna Seed Coagulant was prepared in line with the procedure reported by Adebowale and Adebowale (2007). The influence of coag-flocculation kinetic parameters such as coagulation period τ1/*2*, coag-flocculation reaction order α, and αth order of coag-flocculation constant K, etc were determined. The maximum Mucuna Seed Coagulant kinetic parameter performances were all obtained at (α = 2), K of 6.0 x 10-3 m3 k/kg. S, dosage of 0.3 x 10-3 kg/m3, pH = 10 and τ1/2 of 0.24 Secs. The maximum coag-flocculation efficiency E (%) obtained is 92%, dosages of (0.1, 0.2) x 10-3 kg/m3.Conclusively,Mucuna Seed Coagulant is very effective in the treatment of pharmaceutical effluent sample tuned to alkaline medium.

[V.I Ugonabo, M.C Menkiti, O.D Onwukwuli. **Effect Of Mucuna Seed Coagulant On Total Dissolved Solid Particles Removal Efficiency In Pharmaceutical Effluent Medium By Coag-flocculation Process.** *N* *Y Sci J* 2012;5(9):75-85]. (ISSN: 1554-0200). <http://www.sciencepub.net/newyork>. 9

**Keywords:** Particle Removal, Pharmaceutical Effluent, Mucuna, Coag-Flocculation.

**1.Introduction**

 Coagulation and flocculation are an essential part of drinking water treatment as well as wastewater treatment (Bratby, 2006). This process also plays a core role in surface water treatment by reducing turbidity, bacteria, algae, organic compounds and clay particles (Bina, et al., 2009; McCarthy and Zachara, 1989)

Coagulation and flocculation are the processes that involves the addition of substances usually metal salts to aqueous effluents in order to destabilize colloidal materials. This result, in particles with oppositely charged ions to attract together (Coagulate) and subsequently, aggregating, into larger, more easily removed flocs (Stephenson and Duff, 1996).

 The effectiveness of the process is influenced by raw effluent quality, coagulant dosage, pH, temperature, etc (Randtke, 1988;Nedabigengesere,et al, 1998; Jin, 2005). It has been reported that in conjunction with other treatment processes is regarded as a visible option for the treatment of aqueous effluent such as coal washery etc (Menkiti, et al,2010). In addition, it is recognize that, treating industrial sewage and wastewater by using coagulants and flocculants of different types and dosages reduces toxic matters from these waste (Bordrezou, et al, 1993r; FeoFanov and Smernova, 1995; Abdelaal, 2004). Coagulation/Flocculation can be attained by addition of any of the common coagulant such as metal salts (Al, Fe), starch etc. Coagulation/flocculation of these common compounds have been widely researched with little attention given to agro -products. It is against this back drop that motivated the author to the study of mucuna been seed as a precursor to mucuna seed coagulant. Mucuna pruriens is an annual twinning tropical plant with pods containing bean seed, non- toxic, bio -degradeable, cost effective and safe to human health.

 To this end, this work endavours to evaluate the performance and mechanism of mucuna seed coagulant through examination of variables such as initial turbidity, pH, temperature, dosage etc.

**2.0 Theoretical principles and model development.**

 The general model for Brownian coagulation of mono dispersed particles at early stage (t≤30), is given as (Smoluchowski, 1917).

= ∑αβ (Vi, Vj)ninj - ∑αβ(Vi, Vj)nink 1.

 i+j=k i=1

Where = is the rate of change of concentration of particle size *k* (conc./ time)

Where α is the particle collision efficiency (fraction of collisions that result in particle attachment, β is the collision function (rate that particles are brought into contact by Brownian, shear, ad differential sedimentation), n is the particle number concentration in a size interval and i j are subscripts designating particle size class.

The first term of (1), represents the formation of particle size K by collision of particle size i and j. The second term represents the loss of particle size k by collision with all other particles. The value of β for Brownian transport mechanism is given as [13].

βвr = εΡ 2.

Where Boltzman’s constant (j / k)

η - is the viscosity of the fluid (effluent medium)

εp - is collision efficiency

T - is the absolute temperature (k)

The general equation representing aggregation rate of particles is obtained by solving the combination of (1 and 2), analytically to yield.

 = α 3.

Where is the total particle concentration at time t, = ηk (mass/volume)

 K is the αth order coagulation-flocculation constant

 α is the order of coagulation-flocculation.

 And K = βBR 4.

 Where βBR is collision factor Brownian transport

 Also, βBR  = εp kR  5.

 Combining (3, 4 and 5), yields

 εp α 6.

 Where is the Von smoluchowski rate constant for rapid coagulation [13]

1 7

 = 2a 8.

 Where D1 is particle diffusion coefficient, a is particle radius

 From Einstein’s equation, particle Diffusion coefficient is given

 [14-15]

 D1 = KBT

 B 9.

 Where B is the friction factor from strokes equation:

 B = 6πղa 10.

 Where η is viscousity of the fluid (coagulating and flocculating effluent medium)

 combining (6 to 10), gives

 α 11.

 Comparing (3 and 11), show that k = 12. For perikinetic aggregation α Theoretically equals 2 (i.e. α = 2) as reported [14,16,17].

 From fick’s law

 Jf = D4πRp2 13.

 Where Jf is flux – number of particles per unit surface entering sphere with radius r

 Re-arranging and integrating (13), at initial condition Nt = O, Rp = 2a

 Jf  Rp

4πD1 ∫o  = Nt 14.

Jf  = 8πD1aNo  15.

For central particle of same size undergoing Brownian motion, the initial rate of rapid coagulation - flocculation is

 - = Jf No 16.

 On substitution of (15 into 16), yields

 - dNt = 8πaD1 No εp 17.

 dt

On substitution of (9 and 10 into 17), gives

 - dNt = 8πa KBT No εp 18.

 dt 6πηa

 Thus -dNt = 4 εpKBT N02 19.

 dt 3 η

 Similarly at t > 0

 - 2  20.

 3 η

 Hence (20), has confirmed the theoretical value α= 2 For α = 2, (3), yields

 = -KNt2 21.

 Re – arranging and integrating (21), yields

t  = - K  22.

 Nt2

 = Kt + 23

 Plot of lin VS.t gives a slops of K and intercept of

 From (23), making Nt the subject matter yields a relation for the evaluation of coagulation period, τ½

 Thus Nt = No

 1 + No Kt 24.

Similarly, Nt = No

 1 + 25.

 Let τ = 26.

 Putting (26 into 25), produces

 Nt = No

 1 + 27.

 t 1 m-1

 2 KNo

 Nm(t) 28.

 No = 1 + t m+1

 2

 Let τ1/2 = τ = τ1 29

 m-1

Similarly Nm(t) =

 No m+1 30 1 + t/τ1

 ( 30), gives a general expression for particle of mth order

 Where “m” ranges from 1 to 3 for singlets, doublets and triplets respectively.

 Evaluation of coagulation – flocculation efficiency is given as

 E (%) = No - Nt x 100

 No 31.

**3.0 Materials and Method**

**3.1 Material Sampling, Preparation and Characterization.**

**3.1.1 Pharmaceutical Effluent**

The effluent was taken from a pharmaceutical industry located at Ogidi, Anambra State, Nigeria. The characterization of the effluent was based on standard method (WST, 2005;AWWA, 2005).

**3.1.2 Mucuna Bean Sample**

The sample of mucuna bean seed sample was sourced from Oye Oba Market, Nsukka, Nigeria and processed to mucuna seed coagulant (MSS) based on the work reported by Adebowale and Adebowale (2007).

**3.2 Coagulation – Flocculation Experiment**

Experiments were carried out using conventional Jar test apparatus. Appropriate dose of MSS in the range of (0.1 – 0.6 x 10-3 kg/m3 was added to 250ml of pharmaceutical effluent sample. The suspension, tuned to pH range 1 – 13 by addition of 10M HCL/NaOH ,was subjected to 2 minutes of rapid mixing (120 rpm), 20 minutes of slow mixing (10 rpm) using APPNo 688644A Gullenhamp magnetic mettler Toledo Delta pH meter, followed by 30 minutes of settling. During settling, samples were withdrawn from 2cm depth and changes in TDSP concentration measured for kinetic analysis (Lab-Tech. Model 212R Turbidimeter) at various time intervals of 2, 4, 6, 10, 20 and 30 minutes. The whole experiment was carried out at room temperature. The data obtained were subsequently fitted in appropriate kinetic models for evaluation.

**Table 1: Coagulation – flocculation Kinetic parameters for varying pH and constant 0.1 x 10-3 kg/m3 MSS dosage**

Parameter pH = 1 pH = 3 pH = 5 pH = 7 pH = 10 pH = 13

α 2 2 2 2 2 2

R2 0.938 0.963 0.896 0.860 0.822 0.933

K(m3/kg.S) 3.0x10-5 1.0x10-5 1.0x10-5 5.0x10-5 7.0x10-5 3.0x10-4

βBR(m3/kg.S) 6.0x10-5 2.0x10-5 2.0x10-5 1.0x10-4 1.4x10-4 6.0x10-4

KR(m3/S) 1.542x10-19  1.547x10-19 1.575x10-19 1.534x10-19 1.570x10-19 1.572x10-19

εp(kg.-1) 3.891x1014 1.293x1014 1.270x1014 6.519x1014 8.917x1014 3.817x1015

τ½ (Sec) 45.66 90.91 90.91 28.99 20.70 4.83

(- r) 3.0x10-5Nt2 1.0x10-5Nt2 1.0x10-5Nt2 5.0x10-5Nt2 7.0x10-5Nt2 3.0x10-4Nt2

**Table 2: Coagulation – flocculation Kinetic parameters for varying pH and constant 0.2 x 10-3 kg/m3 MSS dosage**

Parameter pH = 1 pH = 3 pH = 5 pH = 7 pH = 10 pH = 13

α 2 2 2 2 2 2

R2 0.918 0.890 0.619 0.745 0.710 0.962

K(m3/kg.S) 2.0x10-5 1.0x10-5 1.0x10-5 5.0x10-5 5.0x10-5 3.0x10-4

βBR(m3/kg.S) 4.0x10-5 2.0x10-5 2.0x10-5 1.0x10-4 1.4x10-4 6.0x10-4

KR(m3/S) 1.542x10-19 1.547x10-19 1.575x10-19 1.534x10-19 1.570x10-19 1.572x10-19

εp(kg.-1) 2.594x1014 1.293x1014 1.270x1014 6.519x1014 6.369x1014 3.817x1015

τ½ (Sec) 68.49 90.91 90.91 20.70 28.99 4.83

(- r) 2.0x10-5Nt2 1.0x10-5Nt2 1.0x10-5Nt2 5.0x10-5Nt2 5.0x10-5Nt2 3.0x10-4Nt2

**Table 3: Coagulation – flocculation Kinetic parameters for varying pH and constant 0.3 x 10-3 kg/m3 MSS dosage**

Parameter pH = 1 pH = 3 pH = 5 pH = 7 pH = 10 pH = 13

α 2 2 2 2 2 2

R2 0.052 0.878 0.930 0.557 0.790 0.920

K(m3/kg.S) 7.0x10-6 2.0x10-5 3.0x10-5 3.0x10-5 6.0x10-3 2.0x10-4

βBR(m3/kg.S) 1.4x10-5 4.0x10-5 6.0x10-5 6.0x10-5 1.2x10-2 4.0x10-4

KR(m3/S) 1.544x10-19 1.547x10-19 1.578x10-19 1.537x10-19 1.570x10-19 1.572x10-19

εp(kg.-1) 9.067x1013 2.586x1014 3.802x1014 3.904x1014 7.643x1016 2.545x1014

τ½ (Sec) 195.69 45.45 30.30 48.31 0.24 7.25

(- r) 7.0x10-6Nt2 2.0x10-5Nt2 3.0x10-5Nt2 3.0x10-5Nt2 6.0x10-3Nt2 2.0x10-4Nt2

**Table 4: Coagulation – flocculation Kinetic parameters for varying pH and constant 0.4 x 10-3 kg/m3 MSS dosage**

Parameter pH = 1 pH = 3 pH = 5 pH = 7 pH = 10 pH = 13

α 2 2 2 2 2 2

R2 0.936 0.911 0.833 0.924 0.906 0.879

K(m3/kg.S) 2.0x10-5 1.0x10-5 2.0x10-5 5.0x10-5 9.0x10-5 1.0x10-4

βBR(m3/kg.S) 4.0x10-5 2.0x10-5 4.0x10-5 1.0x10-4 1.8x10-4 2.0x10-4

KR(m3/S) 1.544x10-19 1.547x10-19 1.578x10-19 1.537x10-19 1.571x1 1.573x10-19

εp(kg.-1) 2.591x1014 1.293x1014 2.535x1014 6.506x1014 1.146x1015 1.271x1015

τ½ (Sec) 68.49 90.91 45.45 28.99 16.10 14.49

(- r) 2.0x10-5Nt2 1.0x10-5Nt2 2.0x10-5Nt2 5.0x10-5Nt2 9.0x10-5Nt2 1.0x10-4Nt2

**Table 5: Coagulation – flocculation Kinetic parameters for varying pH and constant 0.5 x 10-3 kg/m3 MSS dosage**

Parameter pH = 1 pH = 3 pH = 5 pH = 7 pH = 10 pH = 13

α 2 2 2 2 2 2

R2 0.798 0.817 0.911 0.933 0.676 0.982

K(m3/kg.S) 2.0x10-5 2.0x10-5 2.0x10-5 6.0x10-5 1.0x10-4 1.0x10-4

βBR(m3/kg.S) 4.0x10-5 4.0x10-5 4.0x10-5 1.2x10-4 2.0x10-4 2.0x10-4

KR(m3/S) 1.544x10-19 1.547x10-19 1.578x10-19 1.537x10-19 1.571x10-19 1.573x10-19

εp(kg.-1) 2.591x1014 2.586x1014 2.602x1014 7.807x1014 1.273x1015 1.271x1015

τ½ (Sec) 68.49 45.45 45.45 25.15 14.49 14.49

(- r) 2.0x10-5Nt2 2.0x10-5Nt2 2.0x10-5Nt2 6.0x10-5Nt2 1.0x10-4Nt2 1.0x10-4Nt2

**Table 6: Coagulation – flocculation Kinetic parameters for varying pH and constant 0.6 x 10-3 kg/m3 MSS dosage**

Parameter pH = 1 pH = 3 pH = 5 pH = 7 pH = 10 pH = 13

α 2 2 2 2 2 2

R2 0.529 0.886 0.884 0.830 0.875 0.679

K(m3/kg.S) 4.0x10-5 2.0x10-5 2.0x10-5 7.0x10-5 8.0x10-5 7.0x10-5

βBR(m3/kg.S) 8.0x10-5 4.0x10-5 4.0x10-5 1.4x10-4 1.6x10-4 1.4x10-4

KR(m3/S) 1.547x10-19 1.547x10-19 1.578x10-19 1.537x10-19 1.571x10-19 1.573x10-19

εp(kg.-1) 5.171x1014 2.586x1014 2.535x1014 9.109x1014 1.018x1015 8.900x1014

τ½ (Sec) 34.25 45.45 45.45 20.70 18.12 20.70

(- r) 4.0x10-5Nt2 2.0x10-5Nt2 2.0x10-5Nt2 7.0x10-5Nt2 8.0x10-5Nt2 7.0x10-4Nt2

Figure 1: Plot of Efficiency (E %) Vs Coag-flocculation time for 0.1x10-3kg/m3 MSS at varying pH

Figure 2: Plot of Efficiency (E %) Vs Coag-flocculation time for 0.2x10-3kg/m3 MSS at varying pH

Figure 3: Plot of Efficiency (E %) Vs Coag-flocculation time for 0.3x10-3kg/m3 MSS at varying pH

Figure 4: Plot of Efficiency (E %) Vs Coag-flocculation time for 0.4x10-3kg/m3 MSS at varying pH

Figure 5: Plot of Efficiency (E %) Vs Coag-flocculation time for 0.5x10-3kg/m3 MSS at varying pH

Figure 6: Plot of Efficiency (E %) Vs Coag-flocculation time for 0.6x10-3kg/m3 MSS at varying pH

 Figure 7: Plot of E % Vs pH at 30mins for varying MSS dosages

 Figure 8: Plot of E % Vs Dosage at 30mins for varying pH

Figure 9: Selected Linear plot of 1/TDSP as a function of time for 0.1x10-3kg/m3 MSS dosage

Figure 10: Selected Linear plot of 1/TDSP as a function of time for 0.2x10-3kg/m3 MSS dosage

Figure 11: Selected Linear plot of 1/TDSP as a function of time for 0.3x10-3kg/m3 MSS dosage

 Figure 12: Particles distribution behavior for half life of 4.83S

 Figure 13: Particles distribution behavior for half life of 0.24S

**4.0 Results and Discussion**

**4.1 Efficiency E (%) Vs Time (S) plots.**

 These are shown in figures 1 to 6. These figures actually depicts the performances of Mucuna Seed coagulant with passage of time at varying pH. The important feature of the figures indicates that the coagulant have high degree of solubility in alkaline medium. It is noticeable that the higher the degree of solubility of the coagulant, the greater the probability of their collision with the total dissolved solid particles (TDSP). This is supported by the best performance recorded for pH = 13, though the performance recorded for pH = 10 is equally satisfactorily. The maximum efficiency recorded is 92%, thus satisfies the theory of rapid coagulation which is actually obtained in real situation of coag-flocculation process. This result is in agreement with previous similar work (Menkiti, et al; 2010). Another significant features in the figures is that efficiency of the Mucuna Seed Coagulant increases with time. This phenomenon is an indication that at longer time more ions/particles (in form of TDSP) are adsorbed onto the coagulant. Thus, confirming that coag-flocculation process employed, is controlled by surface charge neutralization and bridging mechanism.

**3.2 Plot of Efficiency E% Vs pH**

 This is presented in figure 7. It illustrate the performance of Mucuna Seed Coagulant (MSS) at varying pH. As can be observed from figure 7, the performance of the coagulant in removing TDSP in pharmaceutical effluent is affected by the pH of the medium. It can be inferred from figure 7 that the performance of the coagulant reached the optimum value at the highest pH medium. The result obtained indicate that at pH = 13, optimum efficiency, E is recorded at 30 minutes. Thus, it can be deduced that coagulant dosage has negligible effect on efficiency, E. In addition, the general features in the figure indicate that the coagulant efficiency decreases downwards to pH = 1. This phenomenon could be that the coagulant is less soluble in acidic medium.

**4.3 Plots of Efficiency E% Vs Dosage (Kg/m3)**

 This is presented in figure 8. It indicate that addition of the coagulant caused a substantial increase in the TDSP removal, but decreases after the coagulant dosage of 0.2 x 10-3 kg/m3 at 30mins. This phenomenon supports the observations made from figures 1 to 7. This indicate that there is negligible change in the efficiency E%, value at pH of 13 for the dosage range of (0.1 to 0.3) x 10-3kg/m3. The best performance of MSS coagulant is recorded at pH = 13 for the dosages of (0.1 to 0.2) x 10-3kg/m3

**4.4 Effect of Coag-flocculation Parameters.**

The results obtained from the experiment were evaluated as the coag-flocculation functional parameters. These were presented in tables 1 – 6. Assuming perikinetic aggregation condition, where, α = 2 solving equation 23 by integration method, gives equation 25, which is presented in the selected linear plots in figures 9 – 11. The plots of 1/Nt Vs t yields, K and 1/No, as the slope and intercept respectively. The results presented in tables 1 – 6, indicate that optimal K values are recorded for all coagulant dosages and pH. But the coag-flocculation performance at pH = 10 is high, followed by pH = 13. These facts are supported by the low values of τ1/2 recorded. In addition, the results show that low values of τ1/2 corresponds to high values of K, which favours fast coag-flocculation process (Smoluchowski, 1917).

Generally, equation 3 implies that α is inversely related to K. K, being rate per concentration is associated with energy barrier (KT) and particle interactions potential, it is understandable that high K is a condition for high particle interactions (Fridkhsberg, 1984).

The values of K (= 0.5βBr) presented in tables 1 – 6 are constant for pH = 3 and 5 for the dosages of (0.1, 0.2, 0.5 and 0.6) x 10-3 kg/m3. This is an indication that it has minimal sensitivity for pH = 3 and 5.

Minimal variations in KR are observed in tables 1 – 6. This is because KR is a function of KB, T and ղ, both KB and ղ are constants, only T, which represents room temperature under which the experiment was performed is varying minimally. At nearly constant values of KR posted in the tables, εp is directly proportional to βBr = 2k and εp is associated with kinetic energy acquired by the colliding particles. The implication is that high εp result in high kinetic energy required to overcome the electrostatic repulsions existing between the intending particles.

This could be achieved by double layer compression or colloids destabilization to ensure low τ1/2, which favours rapid coag-flocculation (Smoluchowski, 1917, Lenntech, 2005). The best τ1/2 value obtained in this study which is in fractions of second lie within the range of previous works where milliseconds had been obtained (Smoluchowski 1917).

Finally, the variations observed in the parameters presented in the tables, especially εp, KR and K could be as a result of unrealistic assumptions, homogeneity of wastewater particles (TDSP) and coagulant throughout the coagulating period prior to flocs formation. The consequence of this could lead to uneven distribution of particles (TDSP)/ Coagulants radicals. Another limitation could be inadequate or excessive coagulant dosage. The effect of this may result to partial coagulation or returbidization of wastewater sample (PFRA, 2003).

**4.5 Particle Distribution Plots.**

These are presented in figures 12 and 13 for τ1/2 =4.83Sec and τ1/2 = 0.24 sec respectively. The curves exhibits similar trends. For figure 12, sum of the particles, ∑Nmi and number of singlet particles Nm1 are at maximum because they are absent at minimum (i.e. t = 0, N = 0).The number of singlet particles decreases more rapidly than the sum of particles. There is less energy barrier between the classes of particles at early time of 0 – 2 mins. This indicate that at early stage of the coag-flocculation is affected by colloidal destabilization and particle entrapment. Subsequently, from 2 – 10 mins, witnessed forces of repulsion between the classes of particles, though is more pronounced between ∑Nmi and other particles. After which, the presence of shear resistance and energy barrier between all classes of particles became negligible leading to total attraction and floc sweep among all classes of particles. In figure 13, the curves exhibits the same behavior as seen in figure 12 except that ∑Nmi and Nm1, are seen to decrease more rapidly than other classes of particles. There are also presence of shear resistance and considerable energy barrier between ∑Nmi and Nm1, Nm2 and Nm3 and between the pairs of particles (∑Nmi and Nm1, Nm2 and Nm3) at the coag-flocculation period of 0 – 4 mins, subsequently, the curves after 4 mins, followed the same trend as obtained in figure 12. This implies that the dominant mechanism in these figures are colloidal destabilization and low particle entrapment to ensure moderate speed of coag-flocculation.

**5. Conclusion.**

The experimental results indicate that maximum value of K is recorded for pH = 10 with the corresponding low value of τ1/2, (in fraction of secs). Also the maximum coag-flocculation efficiency obtained is 92% for pH = 13. These facts, presents MSS as plant-based-derived coagulant, which is very effective in alkaline medium. The results obtained agrees with previous similar works (Smoluchowski, 1917, Van–Zanten and Elimelech, 1992, Menkiti, et al; 2010).

**ABBREVIATIONS**

K : αth order coag-flocculation constant

βBr :Collision factor for Brownian Transport

εp :Collision Efficiency

R2:Coefficient of Determination

 E : Coag-flocculation Efficiency

 τ1/2 : Coagulation Period/Half Life

 ∝ : Coag-flocculation Reaction Order

-r : Coag-flocculation Reaction Rate

TDSP : Total Dissolved Solid Particles

KR : Smoluchowski Constant

Jf : Flux

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7/28/2012