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Enhancement of Pumpkin Seed Coagulant Efficiency Using a Natural Polyelectrolyte Coagulant Aid

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Abstract : Recent studies have pointed out several serious drawbacks of using chemical coagulants such as aluminium and iron salts. These drawbacks include Alzheimer's disease occurring as a result of residual aluminium normally present in treated water and production of large sludge volumes [1]. To eliminate the problems associated with these chemical coagulants, the use of natural coagulants produced from microorganisms, animals, or plants have been found promising. Based on that, in this work, coagulation process has been used to treat dairywastewater that was synthesized by dissolving instant powdered milk in borehole water for turbidity removal. The coagulant used, which was aided with tamarind tree bark, was prepared from pumpkin seed.In order to investigate turbidity removal efficiency of the coagulants and coagulation process kinetics, three sets of experiments were carried out using conventional jar method. The first and second experimental sets were carried out to determine the optimum pH and optimum time for the coagulation. In the third set of experiments, the coagulant and the aid concentration were varied while keeping pH and time at their optimum values already found. The results showed that the optimum operating conditions for turbidity removal from the dairy wastewater were pH of 5, coagulation time of 15 min and concentrations of both coagulant and its aid were 0.5 g/L aid each. Under these optimum conditions, 93.67% turbidity removal was found to be achieved. The results of this work were found to be better than those of [2] in which the maximum efficiency obtained was 71.09%. Therefore, it can be said that the efficiency of the coagulant has been enhanced with the addition of the tamarind tree bark, which is a polyelectrolyte coagulant aid. Furthermore, the kinetics study carried out on the process at the obtained optimum conditions revealed that the process reaction order and rate constant were 3.48 and $2.35\times10^{-6}min^{-1}NTU^{-2.48}$, respectively.Based on the results obtained, the use of pumpkin seed as natural coagulant and tamarind tree bark as aid for treatment of industrial wastewater is recommended as a pre-treatment step for industries because of its promising environmental friendliness.
Keywords: Coagulation, pumpkin seed, polyelectrolyte coagulant aid, dairy wastewater, coagulation kinetics.

1.0 Introduction

Contamination of drinking water sources by fecal matter, inorganic and organic substances represents a major health hazard in many parts of the developing world ([3],[4]). In the treatment of this contaminated water, removal of turbidity is of paramount importance because suspended particles represent transport vehicles for undesirable organic and inorganic contaminants, taste, odour and colour–causing compounds as well as pathogenic organisms ([4],[5]).

Generally, methods applied for treatment of wastewater includephysical processes principally comprising screening, sedimentation, floatation and filtration. Chemical methods of this process utilize the chemical properties of the impurities, and the commonly used ones are precipitation, coagulation and disinfection. Other physical and chemical processes used in water treatment such as air stripping, carbon adsorption, oxidation and reduction, ion-exchange and membrane processes like reverse osmosis and electrodialysis are also important in some certain cases. In addition, biological processes that are employed in water treatment utilize biochemical reactions [6].

Treatment of industrial wastewater is an important issue in environmental protection as it (the wastewater) normally contains pollutants that, if not efficiently treated, can cause serious hazards to the environment. Among the currently employed chemical processes in wastewater treatment, coagulation has received a considerable attention because of its high impurity removal efficiency.

Coagulation is a commonly used process in water and wastewater treatment in which coagulants such as ferric chloride and/or polymers are added to the wastewater in order to destabilize the colloidal materials and cause the small particles to agglomerate into larger settleableflocs [7].

Many coagulants are widely used in conventional water treatment processes based on their chemical characteristics. These coagulants are classified into inorganic, synthetic organic polymers, and natural coagulants.Alum has been the most widely used coagulant because of its proven performance, cost effectiveness, relatively easy handling and availability. However the incidence of an incurable disease in human, characterized by incapacitating and progressive disorder of thought, reasoning and memory, has been linked to amount of aluminium in drinking water, usually added during the water treatment ashydrated aluminum potassium sulfate $KAl(SO)\cdot 12H_2O$ or alumas a clarification and "finishing" agent ([8],[9],[10],[11]).It is also regarded as an important poisoning factor in dialysis encephalopathy[12]. Besides, reaction of alum with water alkalinity reduces water pH and its efficiency in cold water for various operations. Other chemical coagulants like synthetic organic polymers (for example, acrylamide) have neurotoxic and strong carcinogenic effects[13].Furthermore, the use of alum salts and some of these chemical coagulants is inappropriate in some developing countries because of the high costs of their importation and low availability. This is the reason why these developing countries need low-cost methods of wastewater treatment that require low maintenance and skill [6].

Recently, some studies have pointed out that the introduction of natural coagulants and coagulant aids as substitutes for metal saltsmay eliminate the problems associated with the use of chemical coagulants. Natural macromolecular coagulants are promising and have attracted the attention of many researchers because of their abundant source, low price, multi-purposeness, and biodegradation. Okra, rice, and chitosan are natural compounds which have already been used in turbidity removal from water. The extracts of the seeds have been mentioned for drastically reducing the amount of sludge and bacteria in sewage [14]. Other used natural coagulants include Nirmali seed, maize, mesquite beanand *Moringaoleifera*seed. The main advantages of using natural plant-based coagulants as water treatment material are apparent; they are cost effective, unlikely to produce treated water with extreme pH and highly biodegradable. Naturally occurring coagulants are usually presumed safe for human health[13].

Due to the health risk involved in the use of chemical coagulants in thecontaminated water treatment and high cost of the chemicals in some countries, it is important to use healthfully safe, cheap and readily available natural coagulant for the treatment of industrial wastewater.

Some researchers have applied some of these different natural coagulants, and some even supported them with aids in treating water. Among them,[15]applied a coagulant made from snail shell to treat pharmaceutical wastewater. It was observed that the turbidity removal efficiency of the coagulant was affected by time, but the magnitude was found to vary for different pH and dosage. It was concluded from the work that 90.82% total dissolved solid particle was able to be removed at alkaline pH. [16] worked on the treatment of synthetic turbid water using a coagulant produced from herbs of *Cicer arietinum* (chickpea) with alum as an aid.They were able to observe that natural coagulants were effective on high turbid water because 95.5% turbidity removal efficiency was achieved in their work. [7] used ferric chloride and polyelectrolyte as a coagulant with clay minerals as an aid for the treatment of detergent wastewater and discovered that maximum removal efficiency of turbidity could be obtained with the addition of polyelectrolyte as an aid to the coagulant. [2] used a coagulant produced from pumpkin seed to treat dairy wastewater that was synthesized by dissolving instant powdered milk in tap water, and they were able to achieve a turbidity removal efficiency of approximately 71.09%. The turbidity removal efficiency value attained in the work of [2] has been found not to be very satisfactory. It was presumed to be associated with the fact that no coagulant aid was used in their work.

Therefore, in this work, it is aimed to treat a dairy wastewater for turbidity removal using pumpkin seed aided with tamarind tree bark in order to evaluate the effects of initial pH of the wastewater, coagulant dosage and coagulation time on the treatmentas well as carry out one-factor-at-a-time optimization of the process to obtain the values of the factors that will give very high turbidity removal efficiency.

2.0 Methodology

2.1 Coagulant Preparation

The moist pumpkin seeds(Figure 1) used in this work were obtained from Muda Lawal Market located in Bauchi, Bauchi State, Nigeria. After obtaining them (the seeds), they were shelled, washed and dried in an oven at a temperature of 60^{o}C for 6 hr to turn to the form shown in Figure 2. Thereafter, the seeds were ground and processed further in order to remove the oil content present in them. To achieve this, a 250 ml Soxhlet extractor (Figure 3) was employed and placed on a thermostatic water bath that was used to heat the extraction solvent (n-hexane) to its boiling point. The dried pumpkin seeds were packed inside a thimble bag and placed inside the thimble chamber of the Soxhlet extractor. A round bottom flask containing 250 ml of n-hexane was fixed to one end of the extractor while a condenser was tightly fixed to the other end. The round bottom flask was heated with a heater set to a temperature of 50 ^{o}C. The solvent then vaporized to extract the oil and later condensed as a mixture of solvent and oil. The mixture obtained (solvent and oil) moved directly into the round bottom flask. The process was made to continue for the specified time of 4 hr. The cake obtained after the oil had been extracted was exposed to sunlight for about 15 min to yieldthe powder that was used as the coagulant. Shown in Figure 4 are the cake obtained from the pumpkin before and after the extraction of the oil content.

Figure 1. Raw pumpkin seeds used

Figure 2. Dried pumpkin seeds used

Figure 3. Soxhlet extractor setup for oil extraction

(a) (b)

Figure 4. Pumpkin seed cake preparation: (a) before oil extraction, (b) after oil extraction

2.2 Coagulant Aid Preparation

In order to prepare the material used as an aid to the coagulant, the bark of tamarind treeobtained from the Faculty of Sciences premises of Abubakar Tafawa Balewa University, Bauchi,Nigeria was, obtained, washed and cut into pieces before being dried under atmospheric temperature first and, then,in an oven for about eight (8) hours at 50 °C. This was carried out like that so as to make the tamarind tree barkeasy to be crushed. Further, the tree bark pieces were crushedto yield the tamarind tree bark powder. The powder was later sieved to form medium fine powder, which was used as the coagulant aid.

2.3 Synthesis of Dairy Wastewater

The wastewater used in this research work was synthesized by dissolving instant powdered milk in borehole water. The solution was thoroughly shaken and left at room temperature in a tightly covered container for about 24 hr. The turbidity of the synthesized dairy wastewater was measured before and after the treatment (experiment) using DR/890 Colorimeter (Figure 5). A pH meter (shown in Figure 6) was also used to measure the pH of the synthesized wastewater. Prior to the commencement of the experiment, pH adjustment was done by adding 1 M H_2SO_4or KOH, as the case demanded, to the wastewater.

Figure 5. Turbidity meter (DR/890)

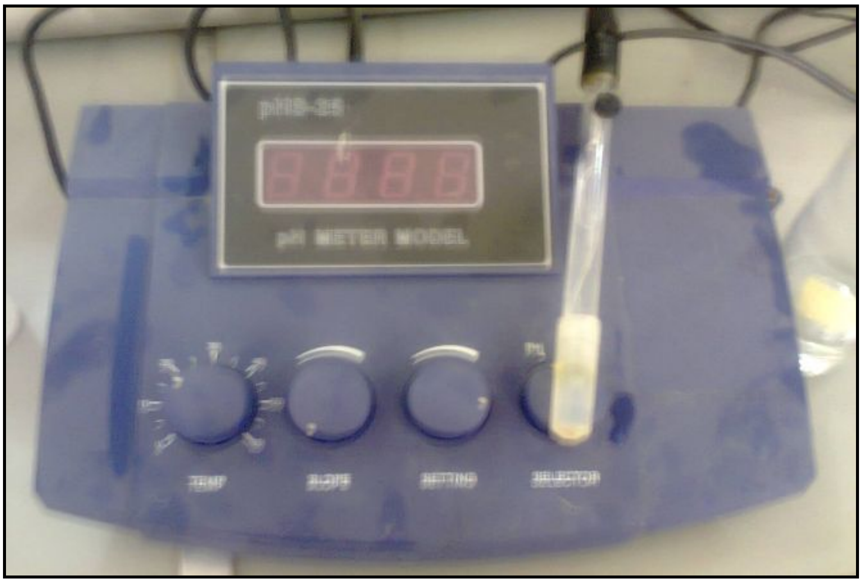

Figure 6. pH meter (pHS-25)

2.4Jar Test Procedure

Coagulation/flocculation studies were performed in a six-place conventional jar-test apparatus, shown in Figure 7, equipped with 6 beakers each of 500 mL volume.

Figure 7. Jar test machine

The jar test experimental studies were carried out in three steps. In each of the steps, 500 ml of synthesized dairy wastewater was poured into each beaker labelled from 1-6.

In the first step, the optimum pH for the treatment was determined. This study was carried out using pH value range of 5 – 9. In each case, the desired pH value of the wastewater was adjusted by using 1 M H_2SO_4 and KOH. The added doses of the coagulant and the aid were at 1 g/L (that is, 0.5 g each) and the resulting mixture was mixed for 30 min at 150 rpm.

In the second step, coagulation was performed to obtain the optimum time, and it was carried out between the time values ranging from 5-30 min. In this case, the doses of the coagulant and the aid were fixed at 1 g/L.

In the last (third) step, coagulant dose was varied between 1 g/L and 6 g/Lwhile the coagulant aid was also fixed between 1 g/L and 6 g/Lfor each experiment. Furthermore, coagulation was performed six (6) times using the optimum pH and time obtained from the first and second steps of this jar test experimental proceduresat the speed of 150 rpm to determine the optimum turbidity removal efficiency of the coagulant dose at each stage. After each coagulation experiment, the resulting mixture was allowed to get settled for 30 minutes and, then, filtered to obtain the treated dairy wastewater. Samples were taken from the mixture for analyses.

2.5 Turbidity Removal Efficiency Calculation

The efficiency of the coagulant in reducing the turbid content of the water was calculated using Equation (1).

$$\%Reduction=\frac{T_1-T_2}{T_1}100\% \qquad (1)$$

where T_{1}= final turbidity

T_{2}= initial turbidity

2.6 Rate and Order of Reaction

The rate and order of the reaction occurring in the water treatment process was deduced by applying kinetics equation given in Equation (2), which was modified to become the form given in Equation (3).

From

$$-r_A = kC_A^{\ n} \qquad (2)$$

Taking the log of both sides,

$$log(-r_A)=log\,k+n\,log(C_A) \qquad (3)$$

where, r_A = Rate of reaction
k = Rate constant
n= Order of reaction
C_A= Turbidity

3.0 Results and Discussion

The analyses of the synthesized dairy wastewater used in this work revealed that its initial turbidity and pH were 600 NTU and 7.45, respectively.

Numerous jar tests were carried out in order to establish a practical understanding of the coagulation performance and to find optimum pH, time, coagulant dosage and coagulant aid dosage.In coagulation process, pH has been found to be very important since it (coagulation) occurs within a specific pH range for each coagulant, and the influence of pH on the removal of suspended solids and colour is important in coagulation process [17]. In considering the influence of pH, a range between 5and 9 was selected, and the results obtained from the experiments carried out on the effect of pH on turbidity removal efficiency when pumpkin seed and tamarind tree bark were used as the coagulant and the coagulant aid respectively are presented in Table 1. As can be seen from the table, to determine the optimum pH value for the process, it was varied while all the other parameters were held constant. Specifically, pumpkin seed and tamarind tree bark dosage weremade to be 1g/Lwhile the coagulation time was set to 30 min with rapid mixing of 150 rpm. According to the results given in Table 1, when pH was increased from 5 to 9, turbidity removal efficiency was found to decrease from approximately 76.50 to 18.17%.As can be observed from the results given in Table 1, the higher the pH value, the lower the turbidity removal efficiency of the coagulant with its aid. It was found clearly from the results that the optimum pH for the process was established to be 5.The information obtained from a previous research showed that the optimum pH of turbidity removal was pH 6.84 [2] The difference in the optimum pH obtained in this study in comparison with that of the previous study can be attributed to the coagulant aid used in this work.

Table 1.Jar test results obtained for the coagulation process carried out by varying the pH of the wastewater

Run no	pH	Coagulant dose (g/L)	Coagulant aid dose (g/L)	Coagulation time (min)	Residual turbidity (NTU)	Turbidity removal efficiency (%)
1	5	1.0	1.0	30	141	76.50
2	6	1.0	1.0	30	305	49.17
3	7	1.0	1.0	30	381	36.50
4	8	1.0	1.0	30	478	20.33
5	9	1.0	1.0	30	491	18.17

Another important parameter considered for the coagulation process of this work was time because it is the factor showing the actualtime required for the coagulation to occurwhen a particular coagulant is used. The results obtained from this study are given in Table 2. From the table, it can be seen that a wide range of time between 5 and 30 min was selected and other parameters (pH, concentrations of coagulant dose and coagulant aid dose) were made constant. The optimum value of time required for the coagulation was obtained to be 15 min (that is, when the residual turbidity of the wastewater was 47 NTU). With these conditions, the maximum turbidity removal from the dairy wastewater was estimated to be 92.17%. Also observed from the results obtained was that increase in time from the optimum value led to increase in the residual turbidity of the water.

Table 2. Jar test result for optimum time determination

Run no	pH	Coagulant dose (g/L)	Coagulant aid dose (g/L)	Coagulation time (min)	Residual turbidity (NTU)	Turbidity removal efficiency (%)
1	5	1.0	1.0	5	59	90.17
2	5	1.0	1.0	10	49	91.83
3	5	1.0	1.0	15	47	92.17
4	5	1.0	1.0	20	55	90.83
5	5	1.0	1.0	25	58	90.33
6	5	1.0	1.0	30	60	90.00

The dosage of the coagulant used on a coagulation process is as important as other parameters (pH and coagulation time) considered because it has been established to be one of the most important factors having influence on the mechanism of coagulation. Given in Tables 3 – 8 are the results obtained when the coagulant dosage was varied while other parameters were kept constant.

Ascan be seen in Table 3 that is showing the results obtained from the process when the pH, coagulant aid dose and coagulation time were 5, 1.0g/L and 15 min, respectively, the optimum coagulant dose was achieved to be 1.0 g/L that gave a residual turbidity of 38 NTU and a turbidity removal efficiency of 93.67%, when the already estimated optimum pH and optimum time of 5 and 15 min respectively were applied. It can also be seen from the table that increasing the coagulant dose beyond 1.0g/Lled to an increase in the residual turbidity of the water. This was found to be an indication that residual turbidity could increase with increase inthe dose of the coagulant used.

Table 3.Jar test result for varying coagulant dose but fixed coagulant aid dose (1.0g/L)

Run no	pH	Coagulant dose (g/L)	Coagulant aid dose (g/L)	Coagulation time (min)	Residual turbidity (NTU)	Turbidity removal efficiency (%)
1	5	1.0	1.0	15	38	93.67
2	5	2.0	1.0	15	59	90.17
3	5	3.0	1.0	15	62	89.67
4	5	4.0	1.0	15	65	89.17
5	5	5.0	1.0	15	67	88.83
6	5	6.0	1.0	15	75	87.50

Looking at Table 4 showing the results of the coagulation process using 2.0 g/L of coagulant aid, it was observed that the optimum dose of the coagulant was 4g/L, which yielded a residual turbidity of 51 NTU when the optimum pH and optimum time were respectively5 and 15 min at their constant values. It can also be seen from the results that the trend between the coagulant dose and residual turbidity as well as turbidity removal was not linear. As can be seen from the table, the efficiency of the coagulant in removing turbidity from the dairy wastewater was estimated to be 91.50%.

Table 4.Jar test result for varying coagulant dose but fixed coagulant aid dose (2.0 g/L)

Run no	pH	Coagulant dose (g/L)	Coagulant aid dose (g/L)	Coagulation time (min)	Residual turbidity (NTU)	Turbidity removal efficiency (%)
1	5	1.0	2.0	15	89	85.17
2	5	2.0	2.0	15	66	89.00
3	5	3.0	2.0	15	56	90.67
4	5	4.0	2.0	15	51	91.50
5	5	5.0	2.0	15	69	88.50
6	5	6.0	2.0	15	98	83.67

From Table 5 in which the results of the coagulation process carried out by varying the coagulant dose while keeping other parameters constant and setting the dosage of the coagulant aid to be 3.0g/L, it was discovered that the optimum value of the coagulant dose required for the process was 5.0g/L, and the residual turbidity of the wastewater was obtained at those conditions to be 102 NTU. Furthermore, the maximum turbidity removal efficiency was obtained under these conditions as 83.00%.

Table 5.Jar test result for varying coagulant dose but fixed coagulant aid dose (3.0g/L)

Run no	pH	Coagulant dose (g/L)	Coagulant aid dose (g/L)	Coagulation time (min)	Residual turbidity (NTU)	Turbidity removal efficiency (%)
1	5	1.0	3.0	15	150	75.00
2	5	2.0	3.0	15	130	78.33
3	5	3.0	3.0	15	115	80.83
4	5	4.0	3.0	15	104	82.67
5	5	5.0	3.0	15	102	83.00
6	5	6.0	3.0	15	169	71.83

Looking at Table 6, it can be seen that the optimum value of the coagulant dose obtained was 1.0g/L with coagulant aid dose and coagulation time being 4.0 g/L and 15 min respectively. Under these conditions, the value of the residual turbidity of the water was found to be 154 NTU, and the turbidity removal from the dairy wastewater was estimated to be 74.33%.

Table 6.Jar test result for varying coagulant dose but fixed coagulant aid dose (4.0 g/L)

Run no	pH	Coagulant dose (g/L)	Coagulant aid dose (g/L)	Coagulation time (min)	Residual turbidity (NTU)	Turbidity removal efficiency (%)
1	5	1.0	4.0	15	154	74.33
2	5	2.0	4.0	15	209	65.17
3	5	3.0	4.0	15	209	65.17
4	5	4.0	4.0	15	213	64.50
5	5	5.0	4.0	15	218	63.67
6	5	6.0	4.0	15	249	58.50

The results of the experiments carried out by varying coagulant dose from 1.0 to 6.0 g/L while maintaining the mass concentration of the coagulant aid at 5.0g/L and the coagulation time at 15 min are given in Table 7. Based on the information obtained from the results given in the table, the maximum turbidity removal efficiency was found to be 74.83%, which was obtained when the coagulant dose was 4.0 g/L.

Table 7.Jar test result for varying coagulant dose but fixed coagulant aid dose (5.0g/L)

Run no	pH	Coagulant dose (g/L)	Coagulant aid dose (g/L)	Coagulation time (min)	Residual turbidity (NTU)	Turbidity removal efficiency (%)
1	5	1.0	5.0	15	253	57.83
2	5	2.0	5.0	15	208	65.33
3	5	3.0	5.0	15	152	74.67
4	5	4.0	5.0	15	151	74.83
5	5	5.0	5.0	15	207	65.50
6	5	6.0	5.0	15	208	65.33

Given in Table 8 are the results obtained from the experiments carried out using a fixed coagulant aid dose of 6.0 g/L and a constant time of 15 min while varying the coagulant dose from 1.0 to 6.0 g/L. From the results given in the table, it was discovered that 82.33% of the turbidity of the synthesizedwastewater could be removed by using a coagulant dose of 5.0g/L together with the other constant conditions of coagulant aid dose and coagulation time.

Table 8. Jar test result for varying dose, fixed aid (6.0 g/L)

Run no	pH	Coagulant dose (g/L)	Coagulant aid dose (g/L)	Coagulation time (min)	Residual turbidity (NTU)	Turbidity removal efficiency (%)
1	5	1.0	6.0	15	269	55.17
2	5	2.0	6.0	15	198	67.00
3	5	3.5	6.0	15	152	74.67
4	5	4.0	6.0	15	118	80.33
5	5	5.0	6.0	15	106	82.33
6	5	6.0	6.0	15	349	41.83

The results obtained were observed to be indications of good performance of the pumpkin seed used as the coagulant and that of the tamarind tree bark used as the coagulant aid. Actually, the good performance of the combination of the pumpkin seed and tamarind tree bark was found to be due to the fact that the tamarind tree bark used as the coagulant aid is a polyelectrolyte material, and the coagulation potential of polyelectrolytes increases with increasing initial turbidity of raw water ([18],[14]). According to [19], a high rate of interparticle contacts occurs in high turbid water that influences the destabilization of particles by adsorption and interparticles bridging whereby along polymeric chain absorbs to more than one particle to form a particle bridge that settles efficiently. Looking at the graphs shown in Figure 8, it can be seen that all the curves follow some certain trends that are in line with the literature findings.

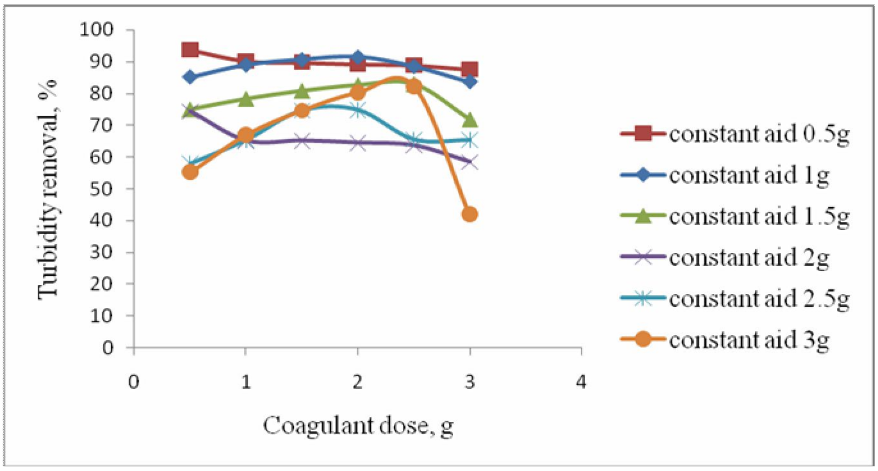

Figure 8. Representative of total turbidity removal (%) versus total coagulant (g)

Another study carried out in this work was studying the kinetics of the coagulation system of dairy wastewater using pumpkin seed coagulant aided with tamarind tree bark coagulant aid. This was achieved using constant coagulant dose and coagulant aid dose of 1.0 g/L each, pH of 5 and varying time from 5 to 15 min. The results obtained from this kinetics study analysis were evaluated as the coagulation functional parameters and are presented in Figures 9 and 10.

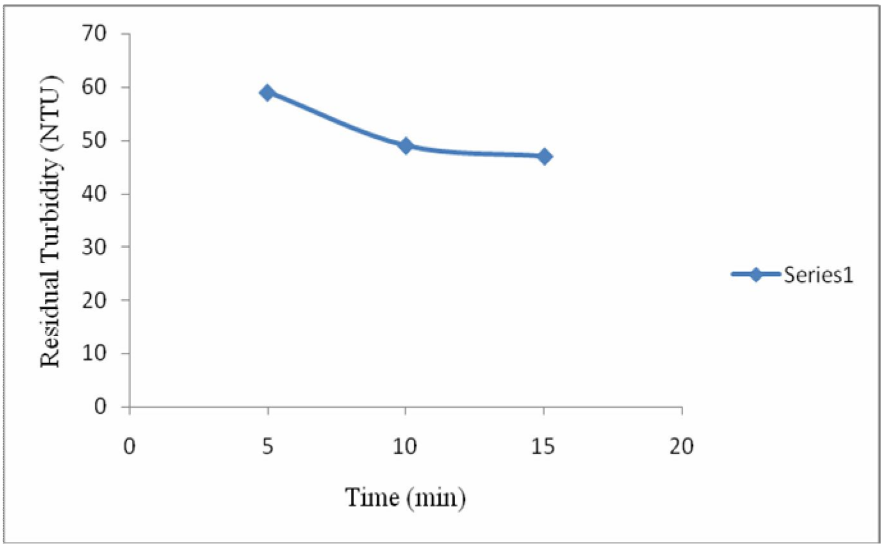

Figure 9. Kinetics plot of residual turbidity (NTU) versus time (min)

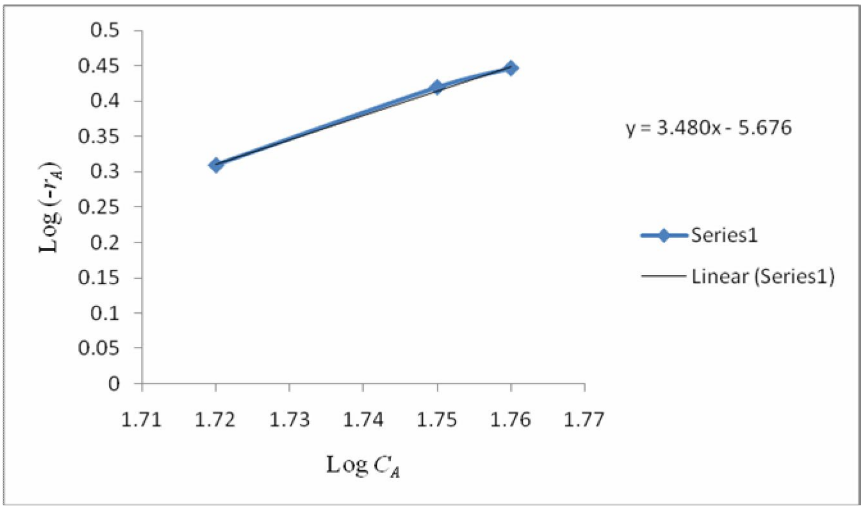

Figure 10.A plot of log (-r_A) against C_A

Using differential method toobtain the slope of the plot of turbidity (C_A)against time (see Figure 9) at each point of the turbidity yielded the values that were used to obtain the values of rate of reaction (-r_A) and concentration (C_A). Those values were later plotted against each other as shown in Figure 10 to estimate the reaction order (n) to be 3.48 and the rate constant (k) as 2.35×10^{-6}(min^{-1}NTU$^{-2.48}$).

4.0 Conclusion

The results obtained from the experiments carried out to evaluate the effectiveness and efficiency of pumpkin seed and tamarind tree bark for removal of turbidity in synthesizeddairywastewater by coagulation method showed that the system was able to achieveoverall maximum efficiency of 93.67% at 15 min coagulation time, 1.0 g/L each of the coagulant and coagulant aid dose, and pH of 5. However, the minimum residual turbidity of 5 NTU recommended by World Health Organization [20] could not be achieved in this study owing to some constraints.It has, thus, been discovered that coagualtion using pumpkin seed as coagulant and tamarind tree bark as coagulant aid can be used for pretreatment of wastewater in industries owing to its environmental friedliness. It is recommended that further study should be carried out to investigate the effectiveness and efficiency of other available natural plants that can be used as coagulants.

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