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Removal of anionic surfactant from residential laundry wastewater using jackfruit (*Artocarpus heterophyllus*) seeds

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ABSTRACT

The study presented in this article investigated the removal of a long chain anionic surfactant from residential laundry wastewater using jackfruit (*Artocarpus heterophyllus*) seeds. The main ingredients of laundry wastewater are the surfactants. Therefore, great attention should be given to the treatment and disposal of laundry wastewater. The use of natural substitutes in treating wastewater has no harmful effects, and it is considered an effective step towards protecting the environment and promoting sustainability. Jar test experiments were conducted in order to determine the optimum conditions for the removal of surfactants, chemical oxygen demand (COD), biological oxygen demand (BOD), turbidity in terms of effective dosage, and pH control. The surfactant, COD, BOD, and turbidity removal efficiencies were 91.66%, 82.86%, 77.66%, and 85.14% at the optimum initial pH value of 6, the optimum dose of 2.5 g/L, and optimum mixing time of 25 minutes, respectively. It can be concluded that *Artocarpus heterophyllus* seed powder was a feasible and cost-effective natural coagulant for the removal of anionic surfactant from laundry wastewater. The results showed that the pseudo-second-order equation is the suitable model for this system.

1. Introduction

Surfactants are organic compounds that contain both hydrophobic groups and hydrophilic groups. The hydrophobic part (their tails) can be aliphatic, aromatic, or a combination of both. The hydrophilic part (their head) gives the basic classification to surfactants, which are non-ionic, anionic, cationic, or amphoteric [1-2]. Surfactants tend to exist at phase boundaries, where they are connected with both polar and non-polar media; thus, they lower the surface tension of a liquid, the interfacial tension between two liquids, or between a liquid and a solid [3]. Anionic surfactants are widely used in domestic and industrial applications. Laundry is the major consumer of the surfactants. Due to industrial development and an increase in population, a large number of surfactants have been consumed and released, resulting in a serious environmental problem [4-5]. They cause foaming in the receiving rivers and other water sources and also accumulate in the environment due to their non-biodegradable nature [6-8]. As a result of these, environmental and public health regulatory authorities have set permissible limits for anionic detergent in drinking water and water for other purposes [9]. There are different physical, chemical, and biological processes employed to remove surfactants from surfactant contaminated wastewater [10-14]. A coagulation-flocculation process is a widely used treatment method for water contaminated with ionic surfactants due to its high pollutant removal efficiency [7,15-16]. Coagulation and flocculation are processes in which the colloidal and coarse suspended solids of wastewaters are destabilized, aggregated, and finally removed [17]. The discharges of anionic surfactants, such as soap containing wastewater, have a high organic pollution load, high total suspended solid, conductivity,

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apparent color, turbidity, alkalinity, and phosphate [18]. Soap is still the surfactant of choice in many countries in the world. In Ethiopia, large amounts of soap (sodium and potassium salts of long-chain carboxylic acids) have been used as surfactants. Increasing the alkyl chain length in the hydrophobic group will generally increase toxicity [19]. However, there have been limited attempts to treat laundry discharge contaminated with soap using natural coagulants. Thus, this work aims to use naturally available material such as *Artocarpus heterophyllus* (jackfruit) seeds as an alternative coagulant to treat laundry wastewater.

2. Material and methods

2.1. Wastewater sample collection

The sample used for the experiment was laundry wastewater, which was collected from residential laundry effluents from the Mendera Kochi Kebele condominiums, Jimma town, South West Ethiopia. After the sample was collected, a composite was made by mixing the collected samples and placed in a plastic container. It was then transported to a laboratory at the Jimma University, Institute of Technology, Department of Environmental Engineering, and stored in a refrigerator at 4° C for further analysis. After the sample was collected, it was analyzed to determine its turbidity, pH, BOD, conductivity, COD, and anionic surfactant concentration. The turbidity was measured using a turbidity meter. The pH of the wastewater was measured with the aid of a digital pH meter. The BOD, conductivity, and surfactant removal efficiency were measured via the probe method, a multiprobe, and the methylene blue index method, respectively. Its chemical oxygen demand (COD) was measured using the potassium permanganate consumption method, while the nitrate and phosphate were determined using a kit, and the chloride by means of the titration method.

2.2. Determination of chemical oxygen demand (COD)

In the potassium permanganate consumption method, 100 mL of the wastewater sample was transferred into a conical flask, and 5 mL of sulphuric acid was added; then, it was covered and boiled for 5 min. While boiling the mixture, 20 mL of potassium permanganate ($KMnO_4$) solution was added from a pipette. The solution was allowed to simmer for 10 min. and then, 20 mL of oxalic acid was added; the entire mixture was heated until the color completely disappeared. The temperature of the mixture was allowed to drop to about 80 °C before titrating it against the $KMnO_4$ solution until a pink color appeared and was persistent for at least 30 sec. A blank with 100 mL of dilution water was analyzed in parallel. The total consumption of $KMnO_4$ was calculated using the expression given in Equation 1:

$$\text{Total KMnO}_4\text{ consumption} = \frac{(\text{A} - \text{B}) \times \text{F} \times 316\text{ mg}}{\text{V}} \qquad (1)$$

where *A* is the $KMnO_4$ consumption by the sample (mL), *B* is the $KMnO_4$ consumption by the blank (mL), *F* is the titration factor of the $KMnO_4$, and *V* is the volume of sample used (mL) [20].

2.3. Determination of biological oxygen demand (BOD)

The biological oxygen demand (BOD) was measured based on the amount of oxygen consumed in the 5-day test period (5-d BOD or BOD_5) at 20^0 C after the arrival of the sample to the laboratory [21].

2.4. Analysis of surfactant

The determination of surfactants was done by measuring the methylene blue index (MBAs) [22]. According to the Ethiopian Protection Agency, the MBAs represent the spectrophotometric method for the determination of anionic surfactants by measuring the methylene blue index in a water environment (EPA, Ethiopian Protection Agency) [23-24]. The efficiency of removal is defined as a percentage of absorbance decrease for surfactant derivatives according to Equation 2:

$$Decoloration = \frac{A_0 - A}{A_0} \times 100 \qquad (2)$$

where A_0 presents the absorbance at the wavelength, which shows the maximum absorption of the surfactant derivative (λ_{max}) while *A* presents the absorbance at λ_{max} of the surfactant derivatives after the plasma treatment. The spectrophotometric measurements were done 5 min. after the plasma treatment with quartz cuvettes that had a 1 cm long optical path [25].

2.5. Collection and preparation of seed powder

The jackfruits were bought from the local market in Jimma, South West Ethiopia, and the rind, fleshy pulps, and seeds were separated. The seeds were washed with distilled water, chopped into small pieces, and dried in an oven at 70° C. After drying, the chopped seeds were pulverized using a domestic blender and sieved to a size less than 355 µm; then they were stored in desiccators until further use [26].

2.6. Jar test experiment procedure

The coagulation-flocculation test of this work was carried out using the jar test experiment method. The experiments were carried out in four parts, viz. coagulation at varying pH, times, coagulant dosage, and surfactant concentrations. Initially, a constant coagulant dosage of 2.5 g of the coagulant was used to optimize the pH value. The optimum pH value was established by a series of experiments using different initial pH ranging from 5.4 - 7.4. A constant mixing speed of 140 rpm was applied for 3 min in order to have rapid mixing, and 40 rpm for 25 min was applied for slow mixing and the final settling step for 1 hr. The pH that gave the highest surfactant removal was taken as the optimum pH. Then, the optimum mixing time was established by keeping the pH and coagulant dose constant. The rapid mixing speed of 140 rpm for 3 minutes was used for all the jars; the slow mixing of 40 rpm was used at time intervals of 5, 10, 15, 20, 25, 30, 35, and 40 minutes and the final settling step for 1 hr to determine the optimum mixing time that would give the highest surfactant removal. For the optimization of the coagulation dose, the optimum pH and the optimum mixing time were kept constant while the coagulant dosage was varied from 0.5 g, 1.0 g, 1.5 g, 2.0 g, 2.5 g, 3.0 g, 3.5 g, and 4.0 g. The dosage that gave the highest percentage removal of surfactant was taken as the optimum coagulant dosage. For the effect of surfactant concentration, the optimum pH, the optimum mixing time, and optimum coagulant dosage were kept constant while the surfactant concentrations were varied [20].

2.7. Adsorption kinetic studies

Coagulation-flocculation is a time-dependent process, and it is very important to know the rate of coagulation for designing and evaluating the coagulant's potential in removing the surfactant wastewater. The amount of adsorption at time *t*, q_t (mg/g), was calculated using the following relation [27].

$$q_t = \frac{(C_0 - C_t)v}{w} \tag{3}$$

The amount of equilibrium adsorption, q_e (mg/g), was calculated using

$$q_e = \frac{(C_0 - C_e)v}{w} \tag{4}$$

$$\%removal = \frac{C_0 - C_e}{C_0} \times 100 \tag{5}$$

where C_t (mg/L) is the liquid phase concentrations of the surfactant at any time, C_0 (mg/L) is the initial concentration of the surfactant in solution, C_e (mg/L) are the concentrations of surfactant at equilibrium, *v* is the volume of the solution (L), and *w* is the mass of dry adsorbent (g).

In many cases, the kinetics of adsorption based on the overall adsorption rate by the adsorbents is described by the first order Lagergren model and pseudo-second-order. [28] The first-order rate expression of Lagergren is given as:

$$\frac{dq}{dt} = k_1(q_e - q_t) \tag{6}$$

where q_e and q_t are the amount of surfactants adsorbed on the adsorbent at equilibrium and time *t*, respectively (mg/g), and k_1 is the rate constant of the first order adsorption (min^{-1}). Integrating Equation (6) for the boundary conditions t = 0 to t = t is as follows:

$$log(q_e - q_t) = logq_e - \left(\frac{k_1}{2.303}\right)^t \tag{7}$$

The plot of $\log(q_e - q_t)$ versus t will give a straight line, and the value of k_1 can be obtained from the slope of the graph. The second-order kinetic model is expressed as:

$$\frac{dq}{dt} = k_2(q_e - q_t)^2 \tag{8}$$

where k_2 is the pseudo-second-order rate constant of adsorption (g $mg^{-1}min^{-1}$). The linearized integrated form of Equation (8) is given as:

$$\frac{1}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e t} \tag{9}$$

If the pseudo-second-order kinetics is applicable to the system, then the plot of t/q_t versus *t* of Equation 9 will give a linear relationship with $1/q_e$ and $1/k_2q_e^2$ as a slope and intercept, respectively.

3. Results and discussion

3.1. Effect of pH

Generally, the pH of the solution changes the coagulant surface charge, and it is likely that the extent of coagulation will also be affected by a change in pH. Therefore, pH values were varied between 5.4 and 7.4 to determine its influence on surfactant removal; the experimental data is presented in Figure 1A. As it can be seen at a fixed dose of coagulant (2.5 g of jackfruit seed), the surfactant removal increases as the pH increased up to 6, but it tends to be less effective as the pH becomes higher. The surfactant removal at a pH of 6 was 91.66%. The surfactant anionic character cannot be reduced by lowering the pH; also, the cationic form of the coagulant would be higher at an acidic pH. So, the electrostatic attraction between the cationic coagulant and negatively charged surfactant active centers is reinforced [29]. In acidic pH, the active sites become positively charged owing to the protonation of its amine functional group according to the FTIR analysis [29,30]. As the pH increases, due to increased competition between OH^- ions and anions and also because of deprotonation of the surface, the electrostatic force of attraction between the coagulant and anionic species decreases, leading to the reduction in percentage removal [31-32].

3.2. Initial surfactant concentration

The removal of the surfactant onto the jackfruit seed was studied for different concentrations (5, 10, 20, 30, 40, 50, 60 , 80, and 100 mg/L) of surfactant solution; the result are shown in Figure 1B. The experiment was conducted under the optimum conditions of a 2.5 g adsorbent dosage and a pH of 6. The percentage removal decreased from 94 to 64.1%, with an increasing initial concentration of surfactant from 5 to 100 mg/L. The maximum surfactant removal occurred for low initial surfactant concentration that showed a gradual reduction when the initial concentration of the surfactant was raised. It could be ascribed to the fixed concentration of the adsorbent dosage. With an increase in the initial surfactant concentration, the adsorption sites were fixed and achieved saturation at low surfactant concentration. Hence, with an increase in the surfactant concentration, no further adsorption could be achieved and resulted in the reduced removal of the surfactant with an increase in surfactant concentration [33-34].

3.3. Effect of coagulation time

In order to determine the effect of coagulation time, it was varied in the ranges of 5 to 40 min at a fixed coagulant dosage of 2.5 g and a pH of 6. As Figure 1C reveals, the surfactant removal efficiency and adsorption capacity were found to increase with contact time. Initially, within the first 25 min, a rapid rate of removal was observed, then a steady-state equilibrium was obtained. The initial rapid rate of removal of the surfactant could be due to the higher number of vacant sites available at the initial stage of removal. Therefore, the optimum coagulation time for the removal of the surfactant onto the coagulants was fixed at 25 min [35-36].

3.4. Effect of coagulant dose

An experimental series of tests was performed to determine the coagulant dosage influence on surfactant removal. A fixed-dose of 2.5 g/L of surfactant was evaluated to be removed with different doses of jackfruit seed in the range of 0.5 to 4.0 g. As can be seen in Figure 1D, the removal of surfactant increased as the coagulant dosage was increased from 0.5 g to 2.5 g, but after a further increase in the coagulant dosage, no significant change in surfactant removal was observed. The percent removal reached 91.66% with an increase of the coagulant dose from 0.5 to 2.5 g, then it remained constant around 91% of removal. This was due to the presence of more adsorption sites of coagulant, thus making for easier coagulation of the surfactant to the adsorption sites. After an adsorbent dosage of 2.5 g, the surfactant adsorption was not observed to increase significantly. On the other hand, the adsorption capacity decreased from 0.2-0.08 as the coagulant dosage increased. This could be due to the existence of an equilibrium surfactant concentration, which was highly difficult to remove. The adsorption capacity *q* tended to be lower as the flocculant dose became higher. The decrease in the adsorption capacity was due to the increased solid dose for the fixed solute load resulting in lower availability of the surfactants per unit mass of coagulants [35,37-38].

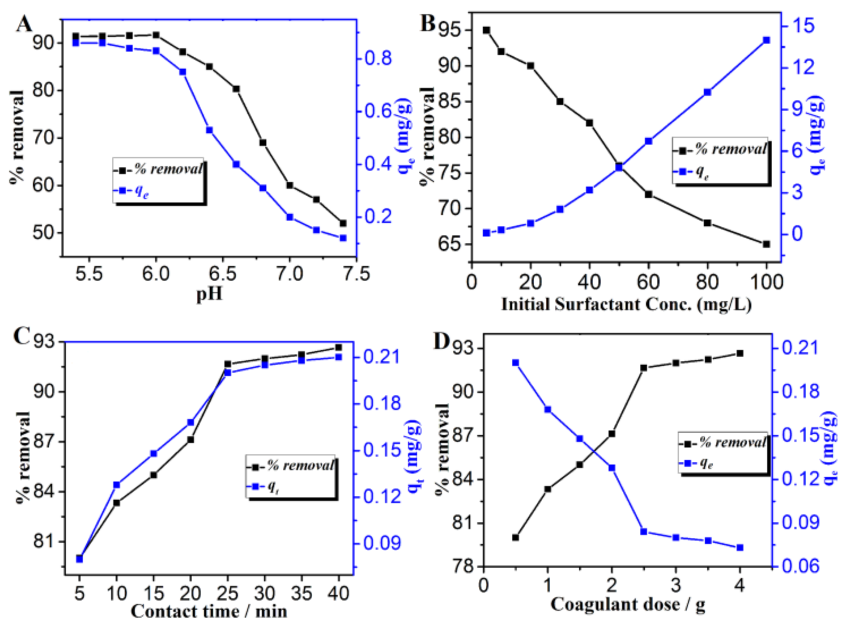

Fig. 1. Influence of variables: (A) pH, (B) Initial surfactant concentration, (C) Contact time, and (D) Coagulant dose.

3.5. Kinetics of the surfactant removal

The study of adsorption kinetics provides valuable information on the mechanism of the adsorption process. The pseudo first order and pseudo second order models have been used in this work for fitting the experimental data [39].The linear plots of the pseudo first and pseudo second order model are shown in Figure 2. The correlation coefficient, R^2, of the pseudo first order kinetics was 0.62, thus it can be concluded that it is not appropriate to use the pseudo first order kinetic model to predict the coagulation kinetics for the surfactant onto the jackfruit seed for the removal process (Figure 2A). On the contrary, the correlation coefficient, R^2, for the second order kinetic model is about 0.989, signifying the applicability of this model (Figure 2B).

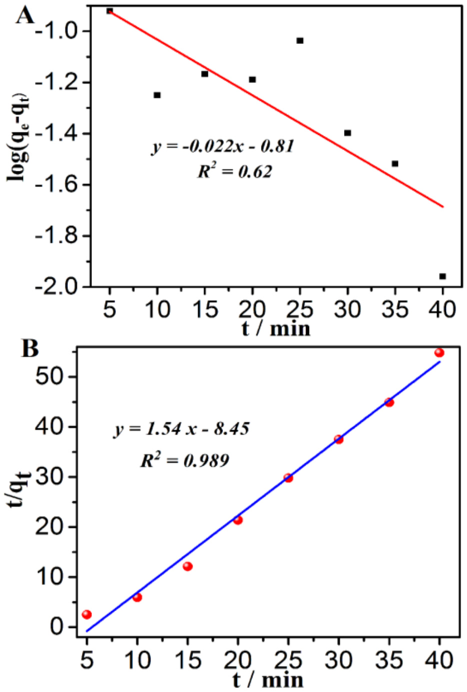

Fig.2. Plot of the pseudo-first order (A) and pseudo-second order (B) kinetics for the anionic surfactant coagulation at different removal rates.

3.6. The physicochemical parameters of laundry wastewater

The physicochemical characteristics of the wastewater and coagulated water are listed in Table 1. The wastewater is characterized by substantial amounts of turbidity, COD, BOD, anionic surfactants, and high pH; therefore, it could not be discharged into a river directly or reused for different purposes without treatment.

Table 1. Physicochemical parameters of laundry wastewater before and after treatment

Parameters	Raw wastewater	Coagulated wastewater
Turbidity (NTU)	9.76	1.45
pH	9.58	6.5
Conductivity (µs/cm)	889	1041
Surfactant concentration (mg/L)	38.9	3.24
Wastewater temperature (^{0}C)	23	22.6
DO (mg/L)	5.2	7.33
COD (mg/L)	3135	542
BOD (mg/L)	1546	345
SO_4^{-2} (mg/L)	44	44
NO_3^- (mg/L)	8.2	8.2
PO_4^{-3} (mg/L)	15.4	15.2
Cl^- (mg/L)	84.09	76.0
Color	Gray	Colorless

(NTU - nephelometric turbidity units)

At the optimum initial pH value of 6, the optimum dose of 2.5 g/L, and optimum mixing time of 25 minutes, the surfactant, COD, BOD, and turbidity removal efficiencies were 91.66%, 82.86%, 77.66% and 85.14%, respectively.

Table 2. Surfactant removal comparison with other coagulants

Coagulant	pH	Adsorption capacity (mg/g)	Ref.
Tanfloc	4.0	1.36	[29]
Moringa oleifera	5.0	0.61	[40]
SilvaFLOC	5.8	1.15	[41]
Jackfruit seed	6.0	0.21	This study

3.7. Surfactant and COD Removal Comparison with other Coagulants

The removal of anionic surfactant using jackfruit seed powder as a coagulant was compared with other reported methods, and the results are summarized in Table 2. It can be seen from the table that the proposed removal method is good and has a comparable performance with other natural coagulants to treat laundry wastewater.

4. Conclusion

The experimental results showed that coagulation using jackfruit seed powder as a coagulant demonstrated promising performance in anionic surfactant removal from laundry wastewater. The results revealed that the removal of anionic surfactant, turbidity, COD, and BOD was dependent on pH, coagulant dose, and mixing time. The highest removal efficiency obtained was 91.66% for anionic surfactant, 85.14% for turbidity, 82.86% for COD, and 77.66% for BOD from the laundry wastewater. The optimum dose, pH, and mixing time obtained from the experimental study were 2.5 g/L, 6, and 25 min., respectively. In the coagulation process, pH was found to be very important, since coagulation occurred within a specific pH range. Moreover, the existence of a cationic protein by protonation was the key component that contributed to the coagulation process. The obtained results showed that the pseudo-second equation model was found to be in good agreement with the experimental results. Jackfruit seed powder showed a relatively good and comparable performance with the other natural and chemical coagulants used to treat laundry wastewater. It can be concluded that jackfruit seed powder can be used as a safe, biodegradable, and environmental replacement for chemical coagulants such as alum and ferric chloride coagulant.

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