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Natural coagulants derived from *Cassia fistula* and tamarind seed for the removal of copper ions

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ABSTRACT

Coagulants derived from plants have shown great potential in wastewater treatment, with good coagulation–flocculation performance, economic benefits and high environmental friendliness. Therefore, the coagulation–flocculation processes conducted by coagulants prepared from *Cassia fistula* and tamarind seeds were studied for the removal of Cu^{2+} ions. Fourier-transform infrared spectra, scanning electron microscopy images and nitrogen adsorption–desorption analysis of the synthesized coagulants indicated the presence of many important functional groups along with preferable surface characteristics for the removal of heavy metal ions. The effects of different variables, including initial solution pH and coagulant dosage, were also estimated to reveal the optimal conditions for Cu^{2+} removal as pH 5.0 and a dosage of 0.6 g/L of *C. fistula* seed-derived coagulant and 1.5 g/L of tamarind seed-derived coagulant. A further pilot-scale study at a volume of 30 L revealed the great efficiency of *C. fistula* coagulant for removing Cu^{2+} ions from real wastewater, with 89.45% removal at a dosage of 0.884 g/L. The obtained results have proved the great potential of these bio-coagulants.

Keywords: Coagulation; Flocculation; Natural coagulant; *Cassia fistula* seeds; Tamarind seeds; Copper removal

1. Introduction

Heavy metals have been an essential part of the development of human society [1]. However, industrial activities, for instance metal coating, excavation or battery production [2], contaminate aquatic environments with non-biodegradable, carcinogenic and highly toxic metals [3,4]. This not only diminishes water quality in rivers, lakes or artesian wells but also leads to great health hazards for living beings. Even at low concentrations, the accumulation of heavy metals in the human body can cause serious diseases like diarrhea and cancer or severe damage to kidneys, bones, nerves and reproductive systems [2,4–6]. Therefore, to sufficiently scavenge those, a variety of heavy metal treatments have been developed. Despite various limits, such techniques as solvent extraction, ion exchange, deploying adsorbents (e.g., biochar, zeolite, active carbon, hydroxyl-apatite, iron oxide), electro-floatation or biodegradation, coagulation–flocculation and membrane filtration are still effective in specific circumstances [3,6–8].

Recently, the coagulation–flocculation process has become widespread because of its simplicity in operation, cost effectiveness and exclusive performance in water treatment, especially in controlling drinking water quality [9,10]. By dispersing chemical reagents called coagulants and flocculants, insoluble particles or dissolved organic matter within aquatic systems can combine into larger aggregates, making their removal favorable by subsequent sedimentation, floatation, or filtration procedures [11]. This process is preferable to reducing water turbidity [12], dissolving natural organic

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matter [13] or rejecting hazardous and harmful agents, such as toxic metals, aggressive anions [13], odor, color [14] and cyanobacteria [15]. Nevertheless, common coagulants are sulfate and chloride salts of aluminum or iron and synthetic polymers that might notoriously affect human health [16]. Moreover, chemical coagulants are quite expensive and their performance strongly depends not only on their concentration but also on the solution's temperature, pH level and composition [2,11].

Therefore, coagulants derived from eco-friendly materials have drawn a lot of attention among scientists recently. In comparison to conventional coagulants, organic alternatives possess many preeminent advantages, including biodegradability, low toxicity, wide variety and abundant sources [16,17]. Some of the popular natural coagulant sources are plants like *Moringa oleifera* [18,19], *Jatropha curcas* [20], maize [21], *Cactus opuntia* [17], Orka gum (*Abelmoschus esculentus*) [16] or chitosan from marine creatures like crab and shrimp [11,22]. Among them, seeds of *Cassia fistula*, known as golden rain tree, and tamarind have been considered as potential materials. Coagulants prepared from these sources have been applied in the removal of toxic dyes [23–25], the treatment of raw and toxic paint industry wastewater [23,26] and for reducing the turbidity and chemical oxygen demand of industrial wastewater [27,28]. Despite both *C. fistula* seed and tamarind seed being common solid waste in Vietnam, their applications in preparing low-cost natural coagulants for heavy metal treatment are quite scarce. To the best of our knowledge, there is no previous report on tamarind seed-derived coagulant in heavy metal removal. For *C. fistula* seed, only studies on its adsorptive removal of Pb(II) and Cu(II) as surface-modified adsorbents [29,30] and of zinc as modified activated carbon from aqueous environments [31] have been conducted at bench scale. Moreover, evaluation of the efficiency of *C. fistula* seed-derived coagulant in practical treatment of heavy metals through pilot-scale studies has not been reported.

In accordance with the shortage of proper research, this study focuses on preparing coagulants from Vietnamese *C. fistula* seeds and tamarind seeds along with appraising their capability to remove Cu(II) ions. The effect of important variables like initial pH and coagulant dosage was revealed in a bench-scale study, and a pilot-scale study, according to Vietnamese environmental standards, was conducted for practical evaluation.

2. Experimental set-up

2.1. Chemical and plant material

The *n*-hexane, acetone, ethanol, HCl, NaOH and $CuSO_4{\cdot}6H_2O$ used were of analytical grade. *C. fistula* seeds and tamarind seeds were collected in Binh Duong Province, Vietnam. The real wastewater was collected from Nam Tan Uyen Industrial Park, Binh Duong Province, Vietnam.

2.2. Preparation of coagulant from C. fistula seeds

The collected *C. fistula* seeds were directly dried under sunlight and ground into fine powder. Using the Soxhlet system, extraction of the obtained powder was conducted in *n*-hexane. Then, by adding ethanol, in 48 h, the fibrous mass was yielded from the precipitation of the extracted solution. Finally, it was carefully washed (with distilled water) and dried at 50°C for 2 h to obtain the coagulant [32].

2.3. Preparation of coagulant from tamarind seeds

Similar to the preparation of *C. fistula*, the fine powder of tamarind seeds was submitted to the Soxhlet system using ethanol for 16–18 h so that color and fat could be removed. Then it was dried at 50°C for 2 h to completely discharge the water. Subsequently, a solution of 20 g of seed powder in 200 mL of distilled water was added to 800 mL of boiled distilled water while being stirred. The final solution was stirred at room temperature before being centrifuged at 5,000 rpm for 15 min. Next, 90% acetone (1:1, v/v) was introduced to the solution to proceed with the precipitation. Finally, the obtained solid material was washed (with acetone) and dried at 30°C to obtain the purified coagulant [33,34].

2.4. Material characterizations

The Fourier-transform infrared (FT-IR) spectra of the prepared materials were recorded using a Nicolet Nexus 670 FT-IR Spectrometer (America). Scanning electron microscopy (SEM) using a Hitachi S-4800 Microscope (Japan) was performed to reveal the surface morphology of the materials while their surface area was measured by the N_2 adsorption–desorption method using a TriStar 3030 Instrument (Micromeritics, USA) with N_2 adsorbed at 77 K.

2.5. Coagulation–flocculation process

In order to evaluate the capability of the prepared materials to remove Cu(II) ions, the coagulation–flocculation process using them was performed with a jar test apparatus system equipped with six 1 L-capacity beakers. The process was conducted at room temperature with the initial concentration of Cu^{2+} ions in the treated solution set at 25 mg/L. The standards TCVN 5999:1995 and TCVN 4556:1988 were consulted for setting up and preserving samples, respectively, while 0.1 M HCl and 0.1 M NaOH were used to adjust the pH of the solution.

In detail, the procedure was conducted as follows. In the beginning, prepared coagulants and the Cu^{2+} solution were introduced to each beaker. Subsequently, rapid mixing at 200 rpm was performed for 5 min as the coagulation followed by slow mixing at 20 rpm for 5 min as the flocculation. After 30 min of layer formation, an atomic absorption spectrometer (ContraAA800D, Germany) was applied to determine the Cu^{2+} concentration within the supernatant layer. From these data, the efficiency of the derived coagulants to remove Cu^{2+} from the treated solution was estimated using Eq. (1), where C_0 and C_e are the initial and equilibrium concentration of Cu^{2+} ions in the treated solution, respectively:

$$\text{Removal}(\%) = \frac{(C_0 - C_e)}{C_0} \times 100$$

To study the effect of solution pH and coagulant dosage (initial Cu^{2+} concentration of 25 mg/L and volume of 10 mL) on the performance of the coagulants, the experiment was performed at different pH values of 2.0, 3.0 and 5.0, as well as at different coagulant dosages ranging from 0.3 to 1.5 g/L.

The pilot-scale study was further conducted to evaluate the practical efficiency of the potential materials [32]. The simulation included three consecutive stages of rapid mixing (200 rpm, 5 min), slow mixing (20 rpm, 5 min) and quiescent gravity separation (30 min), similar to the jar test experiments. The supernatant was then collected for assessing and revealing the removal efficiency of the pilot-scale treatments using Eq. (1).

3. Results and discussion

3.1. Characterization of derived coagulants

Functional groups of the coagulants derived from *C. fistula* and tamarind seeds were identified based on FT-IR spectra recorded in the range of 4,000–400 cm^{-1}. Spectral analysis revealed that both prepared coagulants possess various important functional groups which were similar to the data reported in our previous research. The FT-IR spectrum of *C. fistula* seed coagulant was reported in our previous research, whose characteristic vibration peaks at 3,448; 2,935; 1,735–1,573 and 1,462–1,369 cm^{-1} and the broad band between 1,180 and 960 cm^{-1} confirmed the presence of hydroxyl (–OH) groups, aliphatic C–H bonds, absorbed water and n_{sym} (COO$^-$), C=C bonds (aromatic) and n_{asym}, and C–O bonds (in C–OH bonds), in that order [32]. Noteworthily, the peaks at 876 and 814 cm^{-1}, respectively, confirmed the presence of two important units, β-D-mannopyranose and α-D-galactopyranose [32]. In terms of the tamarind seed coagulant, the material contains several functional groups, namely hydroxyl (–OH) group (3,439.92 cm^{-1}), aliphatic –CH$_2$ groups (2,931 and 2,926 cm^{-1}), carboxyl groups –COO$^-$ (1,648 cm^{-1}) and C–N bonds (of aliphatic amine group) (1,037 cm^{-1}) (Fig. 1) [35]. From the depicted information, activity of the prepared coagulants in the coagulation–flocculation process can be ensured.

The surface morphology of the two coagulants was further investigated from their SEM images. As observed in Figs. 2 and 3, both coagulants displayed an irregular surface along with fibrous networks with a rough surface and porosity. These characteristics can provide the materials with a greater specific surface area and more adsorption sites, and hence significantly enhance their adsorption and bridging ability, some of the main mechanisms occurring in the coagulation–flocculation process [32]. Moreover, the specific surface areas of *C. fistula* seed and tamarind seed coagulants were also elucidated by the Brunauer–Emmett–Teller

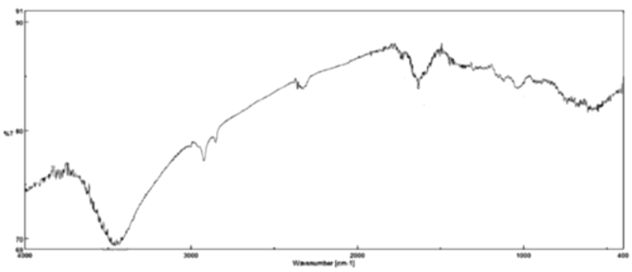

Fig. 1. Fourier-transform infrared spectrum of coagulant prepared from tamarind seeds.

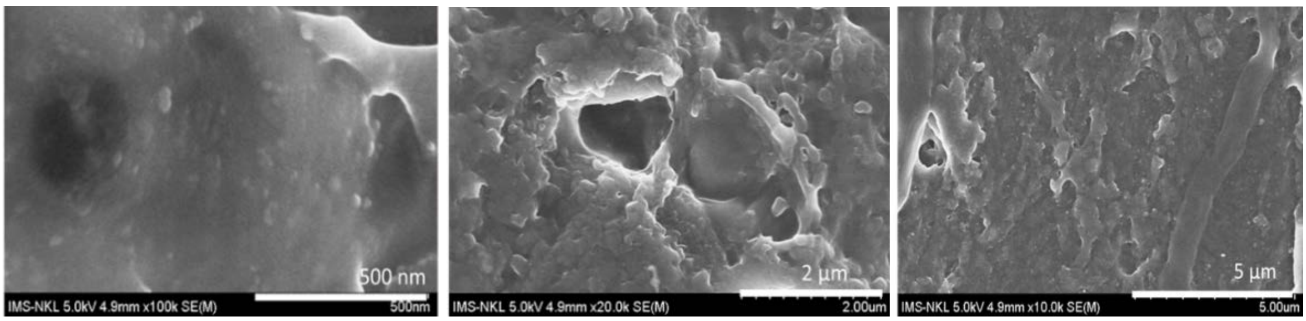

Fig. 2. Scanning electron microscopy images of *Cassia fistula* seed coagulant.

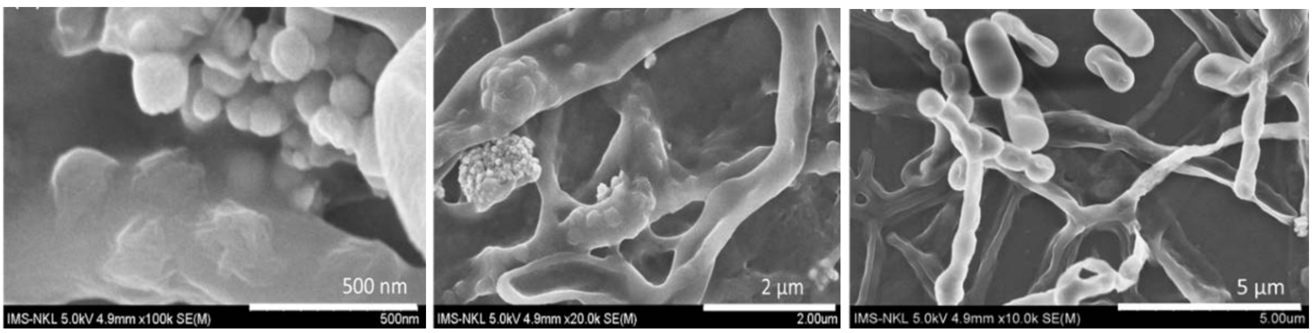

Fig. 3. Scanning electron microscopy images of tamarind seed coagulant.

method as 5.1 and 5.072 m^2/g, respectively. The impediment of the physical adsorption between the solid materials and the nitrogen gas molecules could be the reason behind the modest specific surface areas. However, unlike solid-gaseous absorption, the materials' performance was evaluated in aqueous media. Being suspended in the treated solution allows the fibrous networks of the coagulants to preferably diffuse and interact with the treated component.

3.2. Copper ion removal

3.2.1. Effect of solution pH on copper ion removal efficiency

In the coagulation–flocculation process, the efficiency of the coagulants to remove polluted components is affected by the pH value. Therefore, the experiments studying the effect of this variable on the performance of the derived coagulants in the removal of Cu^{2+} ion were conducted with the initial pH varied from 2.0 to 5.0. As depicted in Fig. 4, the removal of Cu^{2+} ions performed by *C. fistula* seed and tamarind seed coagulants depends strongly on pH level: their efficiencies were enhanced as the pH value increased, which was also quoted in the research of Vinod et al. [36]. In particular, with *C. fistula* seed coagulant the highest removal of Cu^{2+} ions of 45.43% was obtained at pH 5.0 compared to only 17.15% at pH 2.0. Similar was observed for the process performed using tamarind seed coagulant, 53.49% and 23.45% removal being recorded at the same pH levels, respectively. This tendency can be explained as the removal being based on the interaction between electron-rich functional groups, displayed on the bio-coagulants' surface, and the heavy metal cations. From this perspective, a higher pH level means the coagulant surface becomes more negative since its electrostatic interaction with positive Cu^{2+} cations can be facilitated.

3.2.2. Effect of coagulant dosage on copper ion removal efficiency

Another prominent variable is the coagulant dosage. Besides evaluating a coagulant's efficiency in the removal of polluted particles, this factor is also involved in estimating the coagulant's economic benefits and environmental friendliness, as the excessive use of this type of material during wastewater treatment may cause secondary pollution [37]. The Cu^{2+} ion removal efficiency at the pH value of 5.0 achieved with both prepared coagulants is shown in Fig. 5. For the treatment using tamarind seed coagulant, a steady increase in removal efficiency was witnessed, from nearly 50% to 64.57% as the dosage used rose from 0.3 to 1.5 g/L. This enhancement was mainly because the higher dosage of coagulant provided better accessibility of exchangeable sites for Cu^{2+} ions, and hence significantly improved the collision between this component and the applied coagulant [32,36]. In terms of *C. fistula* seed coagulant, the removal efficiency fluctuated. At first, it rose and reached the highest value of 85.41% at the dosage of 0.6 g/L. However, when the dosage increased from 0.6 to 1.5 g/L, the removal efficiency of *C. fistula* seed coagulant rapidly decreased to around 58%. This could be attributed to the re-stabilization of coagulant flocs at excess coagulant dosage reducing the effectiveness of the solid–liquid separation process [38]. In summary, the optimal dosage of *C. fistula* seed and tamarind seed coagulants was 0.6 and 1.5 g/L, respectively. Since *C. fistula* seed-derived coagulant performed more efficiently at a significantly lower dosage compared to that of tamarind seed, the former material was chosen for the pilot-scale study.

3.3. Pilot-scale study using C. fistula seed coagulant

According to preferable pollution parameters of Vietnamese environmental standards, a pilot-scale study of *C. fistula* seed coagulant was performed for more detailed and practical evaluation. In particular, the allowable concentration of copper ions in industrial effluent discharged into the water sources serving tap water supply is assigned at 2 ppm (Column A) by the national technical regulation QCVN 40:2011/BTNMT [39]. The removal percentage and coagulant dosage required to meet the regulated concentration were calculated as 92% and 0.884 g/L, respectively, based on the linear plot of those parameters (Fig. 6) resulting from data collected in the bench-scale experiments. As depicted in Table 1, using the calculated dosage, the pilot treatment was carried out with moderately better results, as the final concentration of Cu^{2+} ions obtained was 2.64 ppm while the

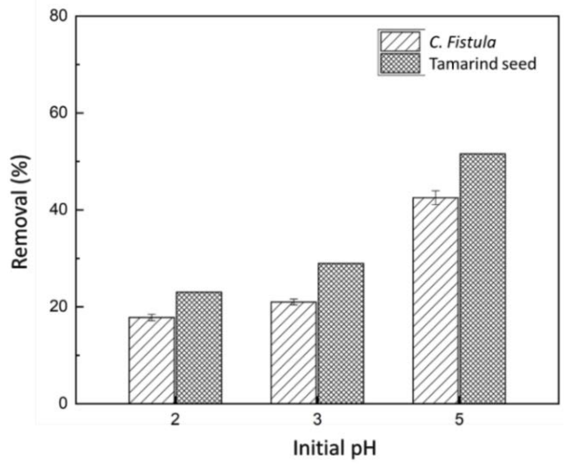

Fig. 4. Effect of pH on the removal of Cu^{2+} ions.

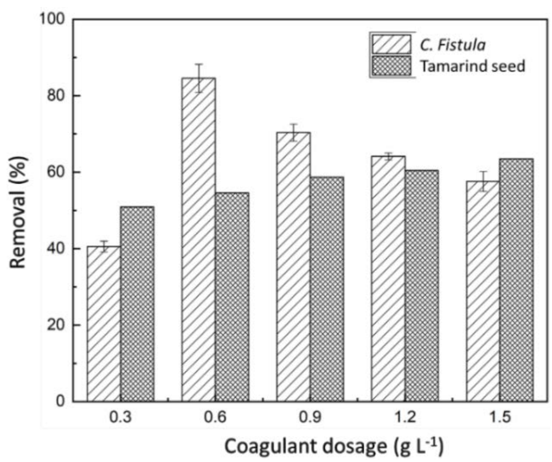

Fig. 5. Effect of coagulant dosage on the removal of Cu^{2+} ions.

Table 1
Pilot-scale study of real industrial wastewater treatment using *Cassia fistula* seed coagulant

		Final concentration of copper ions according to the limitations of Vietnamese standards Column A[a] (2 ppm)
Calculated values	Removal[b], %	92
	Cassia fistula seed coagulant dosage, g/L	0.884
Pilot-scale study results	Real pilot-scale percentage removal of copper ions, %	89.45
	Final concentration of copper ions, ppm	2.64

[a]National technical regulation on the limits for industrial wastewater (QCVN 40:2011/BTNMT) discharging into water sources serving tap water supply [39].
[b]Required percentage removal to ensure meeting the national technical regulation on the limits for industrial wastewater.
[c]Coagulant dosage for the pilot-scale study calculated based on linear plot of percentage removal and dosage found in the bench-scale experiments.

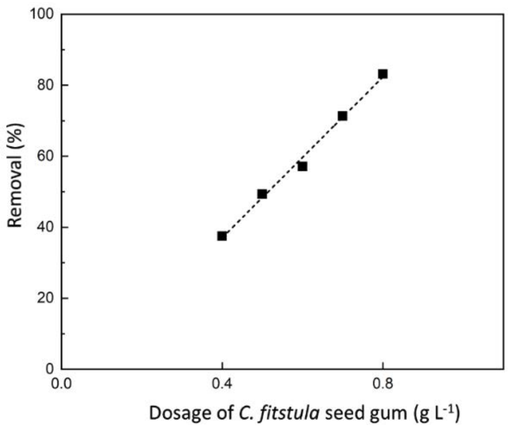

Fig. 6. Linear plot of percentage removal vs. coagulant dosage.

removal percentage was 89.45%. Those slight discrepancies seem reasonable based on coagulation–flocculation usually being considered as a pre-treatment method. In general, this technique can be used prior to others like membrane or biological processes [40,41] to dilute the load of contaminants, and hence reduce the cost of deploying complicated treatments [41,42]. All in all, the pilot results strongly indicate the great potential of *C. fistula* seed coagulant as an effective material for removing heavy metal ions within industrial wastewaters.

4. Conclusion

Two types of coagulants were successfully derived from *C. fistula* and tamarind seeds, possessing several important functional groups and a rough-shaped fibrous network with porosity. The high efficiency in Cu^{2+} removal of the prepared coagulants has been proved with significant dependence on various prominent factors like the solution's pH level and the coagulant dosage. The optimal conditions for the treatment of Cu^{2+} based on the coagulation–flocculation process using the studied materials were determined as an initial pH of 5.0 and applied dosage of 0.6 g/L for *C. fistula* seed coagulant and 1.5 g/L for tamarind seed coagulant. With higher efficiency and a lower dosage, *C. fistula* seed coagulant was further introduced to the pilot-scale study which strongly proved its application in the removal of heavy metal ions as a primary technique. In conclusion, the coagulants derived from the two common solid wastes show great potential in heavy metal treatment. Further development of those materials or similar ones is necessary to provide more economic, eco-friendly and effective treatment of real industrial wastewater.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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