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**Research Article**

# INVESTIGATION OF THE REMOVAL OF REACTIVE BLUE 220 DYE BY GUM EXTRACTED FROM TARAMIND SEEDS

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**ABSTRACT**

*In this study, gum was successfully extracted from tamarind seeds (gum) and used as a material for the removal of reactive dye (Reactive Blue 220-RB220). The performance of decolorization and COD reduction of gum was investigated using the one-factor-at-a-time method. The factors include pH, mixing time, agitating speed, color concentration, and gum concentration. Under optimal conditions, color and COD removal efficiency of gum was roughly 74.4% and 83.3%, respectively. These results indicate that gum is a "green" and "eco-friendly" coagulant and has great potential for application in textile dyeing wastewater treatment.*

***Keywords:*** coagulant; gum; Reactive Blue 220; tamarind seeds; textile dyeing wastewater

## 1. Introduction

Currently in Vietnam, the textile industry has an increasingly important role in the national economy to meet the needs of domestic consumption and export. However, the production of the textile industry also bears many negative impacts on the ecological environment, particularly the effects of textile wastewater. Textile wastewater is wastewater generated in different stages such as cooking, bleaching, dyeing, or printing. The main pollutants in textile wastewater are persistent organic substances with functional groups chromophore, surfactant, and compounds of organic halogen(Holkar, Jadhav, Pinjari, Mahamuni, & Pandit, 2016). According to estimates, about 700,000 tons per year of different colors are produced from nearly 100,000 thousand kinds of commercial dyes worldwide (Rafatullah, Sulaiman, Hashim, & Ahmad, 2010). Among these, Reactive Dyes (pigments activity) is one kind of dyes that are widely used in the dyeing of cellulose fibers and cotton, and especially colorants containing components difficult to treat such as azo, formazan, triarylmethane, and phthalocyanine (Benkhaya, M 'rabet, & El Harfi, 2020; Gupta & Suhas, 2009). Therefore, their residues in sewage water pollute the environment severely, affecting aquatic flora and fauna, and is a carcinogen for humans (Berradi et al., 2019).

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Several methods of dyeing wastewater treatment technologies are applied such as filters, advanced oxygen processes such as UV/Fenton, electron beam irradiation, photocatalysis, biological processes such as aerobic, anaerobic, combined aerobic and anaerobic (Gupta & Suhas, 2009). However, these technologies have certain disadvantages i.e. high investment costs, maintenance costs, complex operation as well as high technical skills. Nowadays, coagulation is the method most commonly used owing to several advantages as low investment costs, simple operation procedures with high processing efficiency. However, the drawbacks of this method are that the used chemical coagulants are not capable of re-use; the effluent pH needs to be controlled; treated water still contains many toxic substances, creating sludge after the treatment process and, thus, requiring sludge management and increasing the costs incurred in the process of sludge treatment (Crini & Lichtfouse, 2018). One remedy is to conduct blemishes to replace chemical coagulants with natural-based ones. Compared with chemical coagulants such as alum (aluminum sulfate), alkaline iron (ferric sulfate), PAC (poly aluminum chloride), biological coagulants are polymeric safe, biodegradable, and the biggest advantage is no secondary pollution. In Vietnam, there are many seeds of plants that can be used to produce eco-friendly coagulant (gum) such as Cassia fistula and county moringa oleifera for the treatment of seafood and textile wastewater (Dao, Bui, Ngo, & Nguyen, 2016; Dao Tran, Nguyen, Ngo, & Nguyen, 2017). Worldwide, there are studies about types of gums such as guar gum, xanthan gum for processing dye Congo red (Ghorai, Sarkar, Panda, & Pal, 2013; Gupta, Agarwal, Ahmad, Mirza, & Mittal, 2020). In addition, the tamarind seed, a byproduct of industrial food processing, can be reused to feed livestock and to use as a binder, antioxidant, cosmetics, processed textile wastewater as well to make gum additive, stabilizer (stabilizer), antimicrobial agents (antimicrobial agent) in the industry of food and applications in medicine in recent years (Kumar & Bhattacharya 2008; & Suresh Rana, 2017; Rawooth et al., 2020). Nevertheless, studies on gum extracted from tamarind seeds for textile wastewater treatment have not been paid enough attention.

Therefore, this study surveyed the ability to treat reactive dye Reactive Blue 220 of tamarind gum extracted from seeds, thereby contributing to the development of a friendly environment substance as well as for further development of gum extracted from tamarind seeds.

## 2. Experiment

### *2.1. Chemicals*

Materials and chemicals utilized in this work include tamarind seeds (Tan Trung Market, Mo Cay Nam Town, Ben Tre Province), sodium chloride (NaCl), acid acetic ($CH_3COOH$), ethanol ($C_2H_5OH$), sodium hydroxide (NaOH), hydrochloric acid (HCl), deionized water (Puris-Evo water system), Reactive Blue 220 dye, sulfuric acid ($H_2SO_4$), potassium dichromate ($K_2Cr_2O_7$), and ammonium iron (II) sulfate hexahydrate ($Fe(NH_4)_2(SO_4)_2.6H_2O$).

### 2.2. Extracting gum from tamarind seeds

A total of 100 g of tamarind seeds was boiled with distilled water for two hours, and the skins were removed from the seeds. Then, the seeds were ground in 0.1 M NaCl solution to a fine powder and continued to be magnetically stirred in 1% CH3COOH solution for four hours. Next, the above sample mixture was centrifuged to take the liquid portion and precipitated in 99% ethanol solution. Finally, the precipitate was dried at 80 $^oC$ for 8 hours.

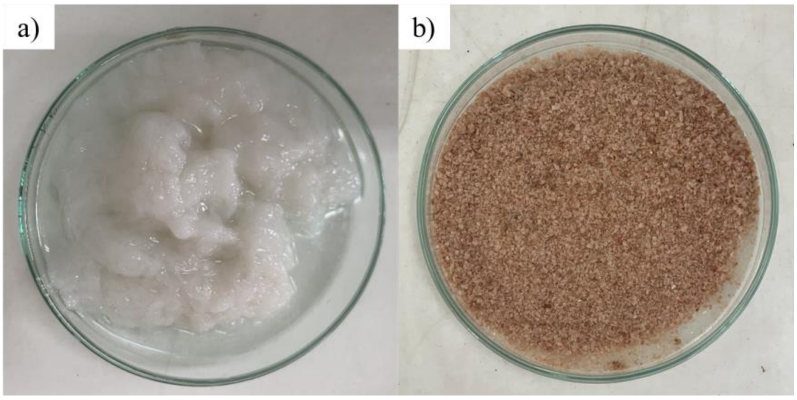


***Figure 1.*** *Gum extracted from tamarind seeds a) before drying and b) after drying*

### 2.3. Investigation of RB220 color treatment ability by gum extracted from tamarind seeds

The experiment to evaluate the color processing ability of RB220 was carried out by investigating five factors affecting the color processing process, including pH (3; 7; 10 and 12), mixing time (15-90 minutes), agitating speed (25-90 rpm), gum concentration (150-500 mg/L), and color concentration RB220 (20-140 mg/L) using Jartest method. The highest absorption peak of RB220 is measured on UV-vis spectrometer (UV5100) to calculate treatment efficiency and COD is determined according to Standard Method SMEWW 5220C:2017.

## 3. Results and discussion

### 3.1. Absorbance spectrum and standard absorption line of RB220

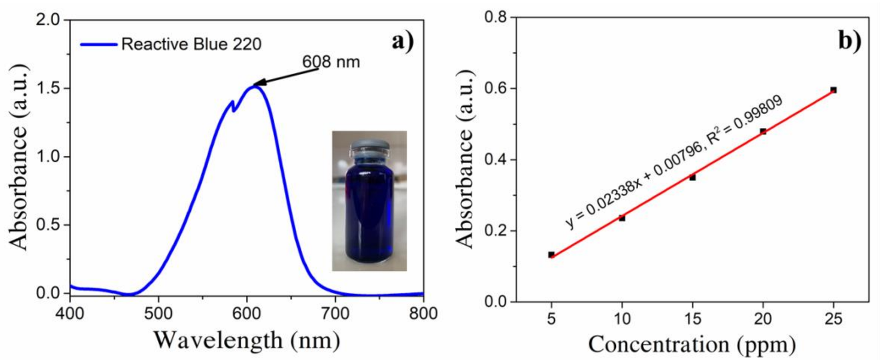


***Figure 2.*** a) *Absorption spectrum and b) standard absorption line at 608 nm of RB220*

Figure 2a shows that the characteristic peak of RB220 color stands at 608 nm. This result is similar to those of previous studies determining the characteristic absorption peak of the color RB220 (Khanna & Shetty, 2014; Patel, Bhatt, & ÌBhatt, 2013). In addition, Figure 2b shows a linear equation (standard curve): y = 0.02338x + 0.00796 (*), $R^2$ = 0.99809, where y is the absorbance value and x is the concentration of RB220 (ppm). Based on equation (*), the RB220 concentration can be calculated directly from the characteristic absorbance peak value at 608 nm at the initial and processing stages and, hence the determination of the decolorization efficiency during the survey.

### *3.2. Surface morphology and gum structure extracted from tamarind seeds*

#### *3.2.1. Scanning electron microscopy image*

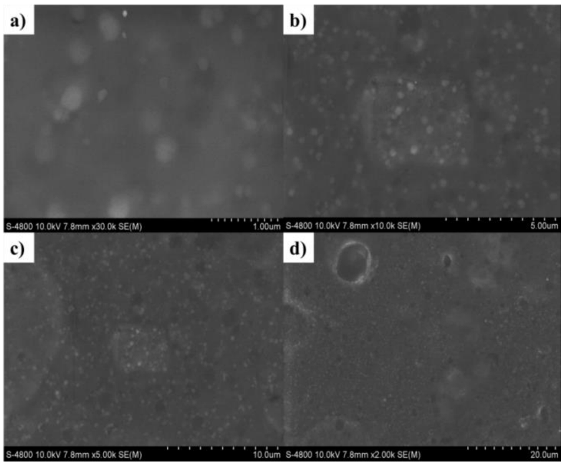


***Figure 3.*** *SEM images of gum at different scales: a) 1 µm, b) 5 µm, c)10 µm và d) 20 µm*

The surface morphology of the gum material extracted from tamarind seeds is shown in Figure 3. Gum has the appearance of pores of different sizes and the gum particles are relatively uniformly distributed on the surface. The SEM image results of gum extracted from tamarind seeds in this study are similar to the SEM results of previous studies (Meenakshi & Ahuja, 2015; Paul et al., 2017).

#### *3.2.2. FT-IR infrared spectrum of tamarind seed gum*

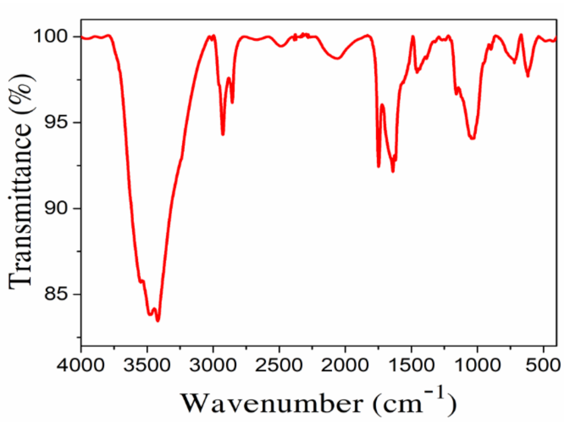


***Figure 4.*** *FT-IR infrared spectrum of tamarind seed gum*

The results of FT-IR spectroscopy in the wavenumber region of 400-4000 $cm^{-1}$ of the gum extracted from tamarind seeds are shown in Figure 4. The peaks appearing in the region of 3000-3500 $cm^{-1}$ are derived from the oscillations of the oscillations -OH radicals. In addition, the peaks appearing at wave number 2855.35 $cm^{-1}$ and 2926.75 $cm^{-1}$ belong to the stretching vibration of the C–H bond (Crispín-Isidro et al., 2019; Mali, Dhawale, & Dias, 2017). In particular, the presence of a peak at 1639.28 $cm^{-1}$ is caused by carbonyl bonds (–HC=O) in the monomers of glucose, galactose, and xylose of tamarind seed gum (Paul et al., 2017). Moreover, the peak appearing at 1037 $cm^{-1}$ belongs to the C–O bond of the xyloglucan ring in the gum structure (Alpizar-Reyes et al., 2017). Thus, the morphological and structural results demonstrate the presence of gum after extraction from tamarind seeds.

### *3.3. Determination of optimal factors in RB220 color treatment with gum*

#### *3.3.1. Determine the optimal pH*

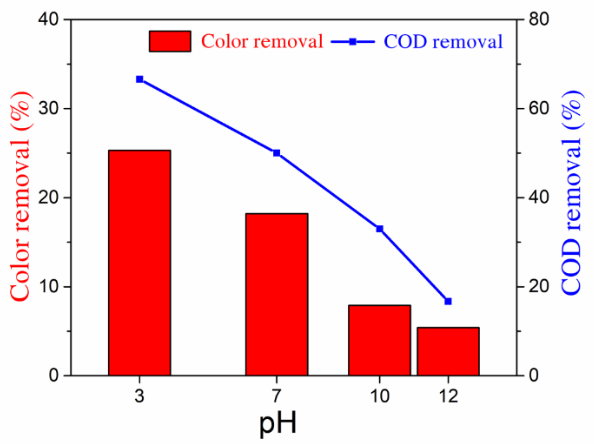


***Figure 5.*** *Effect of pH on the color treatment efficiency of gum*

The effect of pH on the effectiveness of gum color treatment is shown in Figure 5. Results show that at pH = 3, the best decolorization efficiency of 25.3% is, better than the neutral pH at 7 (18.2%) and alkaline pH at 10 and 12 (7.9% and 5.4%, respectively). In addition, COD removal efficiency also shows similar results to the color treatment efficiency, in which at pH = 3, COD removal efficiency is 66.7%, then the efficiency decreases gradually through other pH conditions at 7, 10, and 12. Based on the previous research of Blackburn (Blackburn, 2004), the dye color treatment using coagulation process is explained through 3 main mechanisms: (i) electrostatic interaction; (ii) van der Waals forces, and (iii) hydrogen bonding between gum and dye. The two factors that play a major role in removing RB220 staining are van der Waals forces and hydrogen bonding, similar to those found in Blackburn's (2004).

In an alkaline environment (pH= 10 and 12), tamarind seed gum decomposes into several components with smaller structures such as glucose and xylose, which breaks the gum structure (Whistler & BeMiller, 1958). This is the reason why gum interacts poorly with RB220 pigment and reduces processing efficiency.

Meanwhile, at pH = 3, the acidic environment helps to increase the dye removal efficiency better than other pH conditions. Because in addition to the two mechanisms (ii) and (iii) mentioned above and the nature of the RB220 dye, there are negatively charged functional groups such as (–O–, –C=O, –NH, $–SO_3$) (Boduroglu, Kilic, & Donmez, 2014). Therefore, the acidic environment ($H^+$) enhances the electrostatic interaction between the gum and RB220 dye by mechanism (i) and improves the coagulation ability of the gum. Since then, the dye processing performance improves but not ~~too~~ significantly.

The studies of  Pal et al. (2015) (Pal et al., 2015) have fabricated a hybrid material between guar gum and silica ($SiO_2$) to apply in the color treatment of Reactive Blue 4 (RB) and Congo Red (CR). When investigating the effect of pH on the ability to process these two colors, RB and CR color treatment ability reaches the best performance at pH=2 and pH=3, respectively, then decreases in the alkaline environment. This could be explained that in an acidic environment, electrostatic attraction exists between the positively charged surface of the hybrid material and the negatively charged surface of the dye molecules (RB and CR), increasing strong dye adsorption and high processing efficiency. When the pH value increases, especially in an alkaline environment, the excess of OH- ions on the surface of the material leads to the competition of these OH- ions with the anionic dye molecules, causing repulsion on the surface of the material. The surface of the material is negatively charged and the anionic dye (CR/RB) reduces the treatment efficiency.

Thus, in terms of both economic benefits and treatment efficiency, a neutral environment (pH = 7) is optimal for RB220 color treatment using gum extracted from tamarind seeds.

*3.3.2. Determining the optimal mixing time*

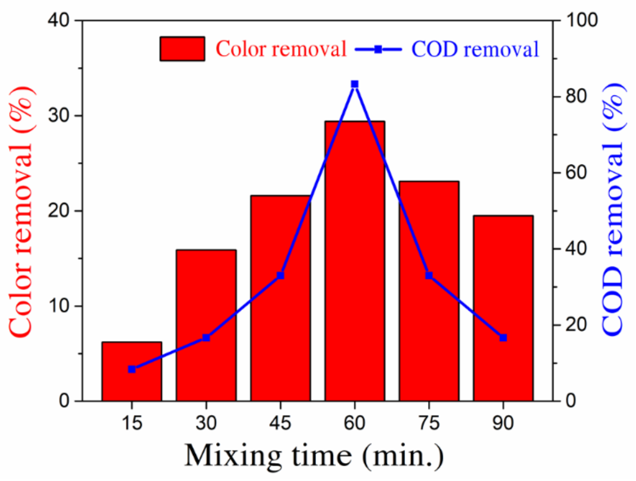


***Figure 6.*** *Effect of mixing time on the color treatment efficiency of gum*

Figure 6 shows the influence at different mixing time intervals: 15, 30, 45, 60, 75, and 90 minutes on the RB220 color treatment efficiency. The above results suggest that the decolorization efficiency increases gradually from 6.2% to 29.4% when the mixing time increases from 15 minutes to 60 minutes. In addition, COD removal efficiency also rises gradually from 15 minutes to 60 minutes and reaches the optimal performance after 60 minutes of mixing with an efficiency of 83.3%. After that, the decolorization and COD removal efficiency both gradually decrease with the efficiency of 19.5% and 16.7%, respectively, after 90 minutes of mixing. Thus, the optimal mixing time is suggested to be 60 minutes.

*3.3.3. Determine the optimum agitating speed*

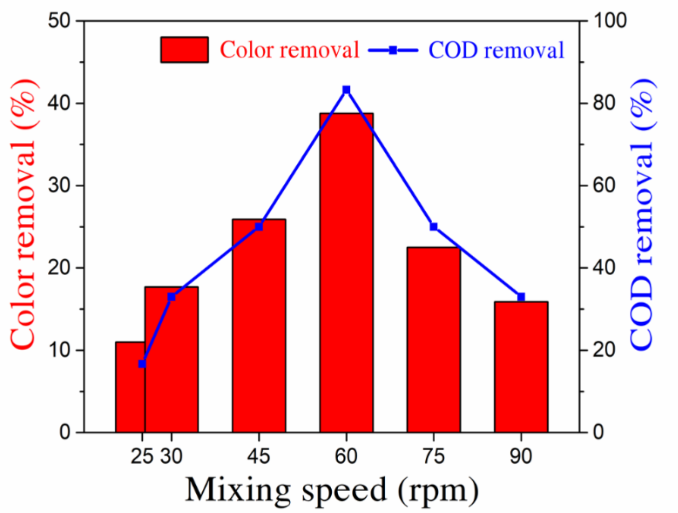


***Figure 7.*** *Effect of agitating speed on the color treatment efficiency of gum*

This study conducted a rapid mixing for the first two minutes, then slowed down the speed to investigate the change of agitating speed from 25 rpm to 90 rpm to determine the optimal agitating speed in the treatment of RB220 color of the RB220 using tamarind seed gum. The results are shown in Figure 7, showing that the agitating speed affects the color processing of the gum. When increasing the agitating speed from 25 rpm to 60 rpm, the decolorization and COD removal efficiency gradually increase and reach the optimal value at 60 rpm with the efficiency of 38.8% and 83.3%, respectively. Then, while the agitating speed increases up to 75 rpm and 90 rpm, the decolorization efficiency decreases to 22.5% and 15.9%, respectively. Meanwhile, COD removal efficiency decreases to 50% and 33.3% at 75 rpm and 90 rpm, respectively.

The reduction in treatment efficiency under high agitating speed conditions is due to excessive collision between the gum and colloidal cotton particles, causing them to break. Therefore, stirring conditions at 60 rpm are found to be optimal in this investigation.

*3.3.4. Determination of optimal gum concentration*

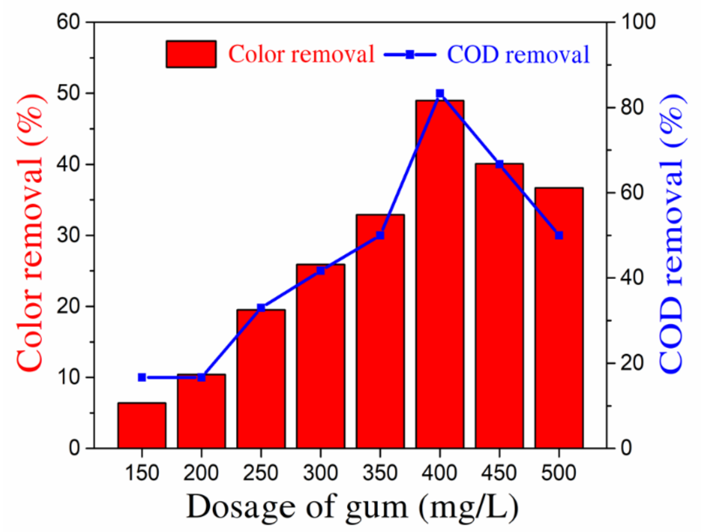


***Figure 8.*** *Effect of gum concentration on color treatment efficiency*

Through the color treatment results shown in Figure 8, it is found that when the gum concentration increases from 150 mg/L to 400 mg/L, the decolorization efficiency increases gradually and the best color treatment was achieved at the concentration of 400 mg/L with an efficiency of 49%. However, when increasing the concentration to 450 mg/L and 500 mg/L, the decolorization efficiency starts to decrease gradually and reaches 40.1% and 36.7%, respectively. Meanwhile, with gum concentrations of 150 and 200 mg/L, the COD removal capacity is the same with an efficiency of 16.7%. Then, when gradually increasing the concentration of gum to 400 mg/L, the COD removal as well as the decolorization efficiencies increase and reach the optimum at this concentration with an efficiency of 83.3%. When increasing the concentration beyond the optimal threshold, COD removal efficiency tends to decrease and reaches 50% at the gum concentration of 500 mg/L.

The mechanism of color processing with the change of gum concentration is explained as follows: when the concentration of coagulant increases, the larger the number of charges added to the system, the greater the sweeping effect, resulting in the Zeta potential = 0. Combined with the sweeping effect, it leads to good coagulation ability and increased coagulation efficiency. But when the threshold is reached, if the coagulant concentration is too much, the Zeta potential ≠ 0 will increase the repulsive force between the molecules, reducing the coagulation ability and, thus, reduces treatment efficiency.

Thus, in this experiment, the optimal gum concentration is found to be 400 mg/L to rationally use the gum concentration to avoid waste of material during processing.

*3.3.5. Determination of optimal color concentration*

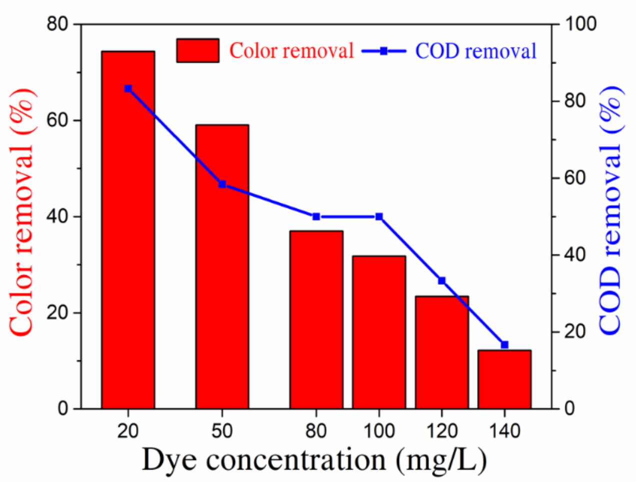


***Figure 9.*** *Effect of color concentration on the color treatment efficiency of gum*

Figure 9 shows that the decolorization and COD reduction efficiency of gum is affected by the color concentration of RB220. The results show that as the color concentration of RB220 increases, the decolorization and COD removal efficiency decreases. Specifically, the decolorization and COD removal efficiency at the concentration of 20 mg/L is optimal with the efficiency of 74.4% and 83.3%, respectively, when increasing the color concentration to 140 mg/L leads to a significant reduction of decolorization and COD removal efficiency to only 12.2% and 16.7%, respectively. This was explained by the insufficient gum concentration to interact with the color concentration. Therefore, to improve the efficiency of color treatment at higher color concentrations, it is necessary to add more gum to the treatment process.

Thus, apart from the other factors such as pH, mixing time, agitating speed, and gum concentration, the color concentration has a significant impact on the color treatment efficiency and corresponds to each different dye concentration. It is necessary to calculate the appropriate amount of gum to use.

## 4. Conclusion

The gum was successfully extracted from tamarind seeds and used as a coagulant to treat Reactive Blue 220. The results in this study proved that gum can decolorize and reduce COD of color RB220. Optimal conditions of RB220 are suggested as below: pH of 7; mixing time of 60 minutes; agitating speed of 60 rpm; gum concentration of 400 mg/L, and color concentration RB220 of 20 mg/L. Under these optimal conditions, the gum is proven to achieve decolorization and COD removal efficiency of 74.4% and 83.3%, respectively. Thus, gum extracted from tamarind seeds is a "green" coagulant, environmentally friendly, and has great potential for application in textile dyeing wastewater treatment.

❖ ***Conflict of Interest:*** *Authors have no conflict of interest to declare.*

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# NGHIÊN CỨU KHẢ NĂNG XỬ LÍ MÀU NHUỘM HOẠT TÍNH REACTIVE BLUE 220 BẰNG GUM TRÍCH LI TỪ HẠT ME

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## TÓM TẮT

*Trong nghiên cứu này, gum được trích li thành công từ hạt me và sử dụng làm vật liệu để xử lí màu nhuộm hoạt tính Reactive Blue 220-RB220. Hiệu quả khử màu và khử COD của gum trích li từ hạt me đã được khảo sát với các yếu tố ảnh hưởng như: pH, thời gian khuấy, tốc độ khuấy, nồng độ màu và nồng độ gum. Tại điều kiện tối ưu, gum trích li từ hạt me đạt hiệu suất khử màu và khử COD lần lượt là 74.4% và 83.3%. Như vậy, nghiên cứu này cho thấy gum trích li từ hạt me là một chất keo tụ* "xanh"*, thân thiện với môi trường và rất có tiềm năng ứng dụng trong xử lí nước thải dệt nhuộm.*

***Từ khóa:*** chất keo tụ; gum; Reactive Blue 220; hạt me; nước thải dệt nhuộm