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Research Article

REMOVAL OF REACTIVE YELLOW 145 COLOR AND REACTIVE RED COLOR 194 BY GUM EXTRACTED FROM MORINGA OLEIFERA SEEDS

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ABSTRACT

In this study, a biological coagulant (gum) extracted from moringa seeds is produced as a coagulant to treat Reactive Yellow 145 (RY145) and Reactive Red 194 (RR194). The material's properties are determined by scanning electron microscopy (SEM) and FTIR. SEM images show that the surface structure of the gum has the appearance of relatively fine pores and homogenously distributed gum particles on the surface. FTIR spectra indicate that the prepared material contains surface functional groups such as –OH, –HC=O and C-O of the xyloglucan ring. The factors affecting the coagulation process of RY145 and RR194 colors include pH, gum concentration, initial color, stirring speed, and stirring time. based on the results, it is suggested that the bio-flocculating agent (gum) prepared from moringa seeds, an eco-friendly material, can decolorize and reduce COD and, thus, is potential for the treatment of textile dyeing wastewater. The RY145 and COD removal efficiency was 87.77% and 76.92%, respectively, at optimal points of pH = 9, gum concentration (2000 mg/L), color concentration (40 mg/L), stirring speed (60 rpm)and time stirring (55 min). The RR194 and COD removal efficiency was 89.31% and 80%, respectively, at optimal points of pH = 7, gum concentration (3000 mg/L), color concentration (20 mg/L), stirring speed (45 rpm), and time stirring (65 min).

Keywords: coagulant; gum; Moringa seeds; Reactive Yellow 145; Reactive Red 194; textile dyeing wastewater

1. Introduction

Nowadays, with the increasingly advanced and developed life, the problem of environmental pollution is increasing exponentially. Every day, thousands and tons of waste, gas, etc., are discharged into the environment. Environmental pollution then became a global concern. In recent years in Vietnam, the strong development of the textile and dyeing industry has contributed a great deal to the overall economic development of the country. More and more textile dyeing companies have been built across the country. However, due to the specificity of the industry, the textile industry is considered one of the industries

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causing serious environmental pollution, especially water pollution. With a complex technological chain, including many different stages, the wastewater after production contains many types of toxic compounds, such as dyes, surfactants, organic halogen compounds, and other chemicals color, that are difficult to decompose since these do not adhere to all to the fabric, thus leaving a certain amount of residue. Therefore, they cause severe pollution to the environment, affecting the animals, aquatic plants, and human health (Nguyen, Dang, Nguyen, & Dao, 2016). The development of industrialization and modernization of the country has indirectly polluted the environment, while the balance between developing the economy and protecting the environment is a matter of concern that needs to be overcome.

Textile dyeing wastewater treatment system includes many different methods. Each method has a certain effect on the corresponding pollutant. The applied wastewater treatment technology requires the removal of components such as temperature, color, suspended solids SS, COD, BOD_5, and heavy metals. Reverse osmosis, cation exchange, adsorption, and flotation are considered possible techniques to degrade toxic dyes (Jawad, Mamat, Hameed, & Ismail, 2019; Adeleye et al., 2016). Researchers found that adsorption is the most favorable direction for dye removal from water because of its high efficiency, moderate approach, ease of handling, and high selectivity (Munagapati, Yarramuthi, Kim, Lee, & Kim, 2018; Foroutan et al., 2019; García, Plaza-Cazón, Montesinos, Donati, & Litter, 2018).

Coagulation-flocculation method is one of the methods of dye treatment with high efficiency and simple operation. Commonly used chemicals are chemical coagulants such as iron alum, aluminum alum, and PAC.. However, the disadvantages of this method are that the used coagulant chemicals cannot be reused, the wastewater pH should be controlled, post-treatment water still contains many toxic substances, sludge produced after treatment, requires management and increases treatment costs (Crini & Lichtfouse, 2019). Therefore, a material that is a biological coagulant can be a new alternative to replace chemical coagulants to reduce pollution and solve the problem of sludge discharge after wastewater treatment. Currently, biological coagulants (gum) derived from the endosperm of seeds have been studied.

In Vietnam, many types of seeds have been studied as biological coagulants, such as *Cassia Fistula L.* seeds applied to improve the quality of aquatic wastewater (Dao, Bui, & Nguyen, 2016). In the world, there have been many studies showing that the endosperm of seeds can act as a flocculating agent, such as *Cassia Angustifolia* seeds used to decolorize dye solutions (Sanghi, Bhatttacharya, & Singh, 2002). Therefore, in this study, the removal efficiency of Reactive Yellow 145 and Reactive Red 194 with gum extracted from *Moringa oleifera* seeds is investigated to create a-novel biological coagulant (gum) that contributes to the treatment of textile dyeing wastewater, which can improve environment quality.

2. Experiment

2.1. Chemicals

Materials and chemicals used in this study include dried moringa seeds, sodium chloride (NaCl), sodium hydroxide (NaOH), acetic acid (CH_3COOH), sulfuric acid (H_2SO_4), hydrochloric acid (HCl), ethanol (C_2H_5OH), hexane (C_6H_{14}), Reactive Yellow 145 dye (RY145), and Reactive Red 194 dye (RR194) (99% purity purchased at Azelis Company at 1489 Nguyen Van Linh Street, Tan Thuan Dong Ward, District 7, Ho Chi Minh City) and certain chemicals used in analyzing textile wastewater samples.

2.2. Extracting gum from moringa oleifera seeds

The following steps were conducted:

- Dry moringa seeds at 100ºC for 2 hours
- Remove the seed coat
- Grind 60 g of seeds
- Separate the fat in the granules with n-Hexane for 10 hours and decolorizes the granules with 99.8% ethanol for another 10 hours.
- Mix the defatted seed powder and colorants with 800 mL of 1% acetic acid solution using a magnetic stirrer for 4 hours.
- Soluble polysaccharides were then recovered by centrifugation.
- Precipitate with 99.8% ethanol.
- Centrifuge to separate the solvent and collect the precipitate.
- The polysaccharide powder was recovered by drying at 80ºC for 2 hours.

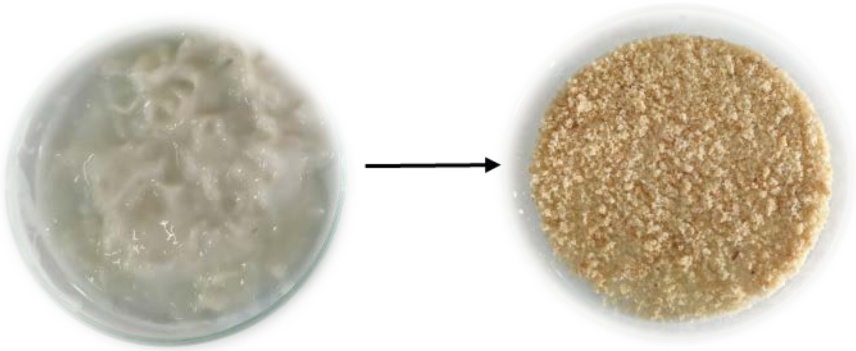

Figure 1. *Gum extracted from moringa oleifera seeds (a) before and (b) after drying*

2.3. Remove RY15 and RR194 by the gum

This study was designed to investigate the ability to treat RY145 and RR194, a synthetic textile dyeing wastewater. The prepapration was conducted individually for each color with the gum based on five factors: pH (3, 7, 9, 12), gum concentration (1000 - 4000 mg/L), initial color (20 – 80 mg/L), agitating speed (15 – 90 rpm), and mixing time (25 – 75 minutes) on the Jar-test apparatus. The Standard Methods for Examination of Water and Wastewater (SMEWW 5220B: 2017 and SMEWW 5220C: 2017) were used to calculate the maximum absorption peaks of the colors RY145 and RR194 on a UV-vis spectrophotometer for the efficiency and COD. .

3. Results and discussion

3.1. Absorbance spectrum and standard absorption line of RY145 and RR194

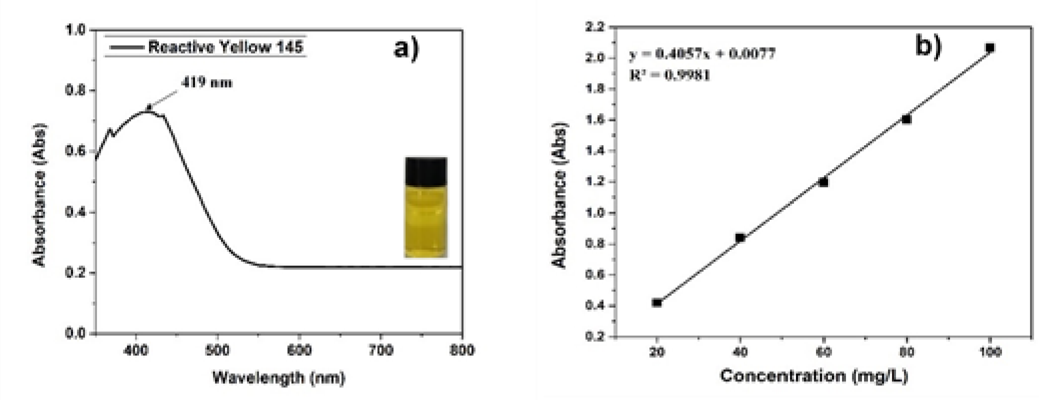

Figure 2. *(a) Absorption spectrum and (b) standard absorption line at 419 nm of RY145*

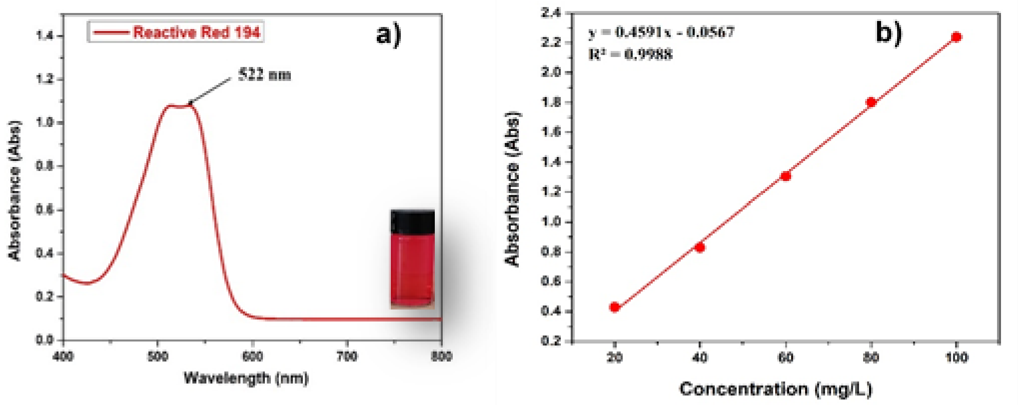

Figure 3. *(a) Absorption spectrum and (b) standard absorption line at 522 nm of RR194*

Figures 2(a) and 3(a) show that the characteristic peak of RY145 is at 419 nm while that of RR194 is at 522 nm. This result is similar to previous studies (Gül & Özcan-Yıldırım, 2009). In addition, Figures 3(a) and 3(b) show a linear equation (standard curve), where *y* is the absorbance value, and *x* is the concentration of the color. Based on the equation, the color concentration can be calculated directly from the characteristic absorbance value at the maximum wavelength at the initial and treatment stages, thereby determining the decolorization efficiency during the investigation.

3.2. Surface morphology and gum structure extracted from moringa oleifera seeds.

3.2.1. Scanning electron microscopy image

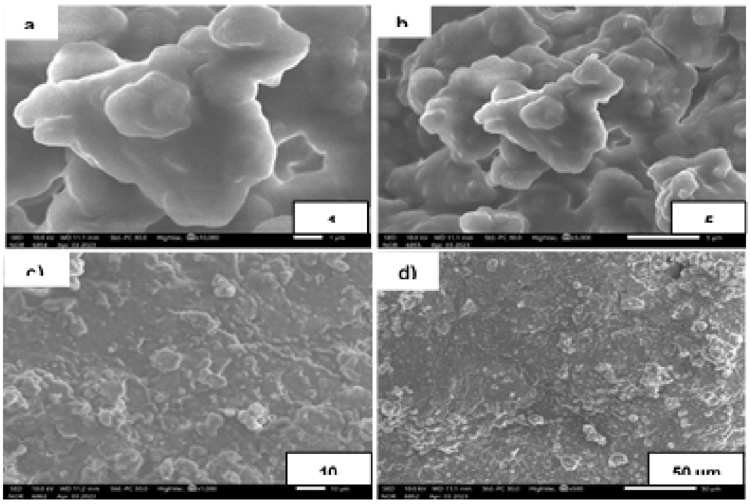

Figure 4. *SEM images of gum at different scales*

The scanning electron microscopy (SEM) images illustrate the surface structure of the gum synthesized at different ratios. The moringa gum has the appearance of relatively fine pores, and the gum particles are relatively homogenously distributed on the surface.

3.2.2. FTIR infrared spectrum of moringa oleifera seeds gum

The FTIR infrared spectroscopy shows that there are peaks at wavelengths in the region of 3000 - 3500 cm^{-1}, originating from the -OH radical oscillation. Besides, the peaks appearing at wavelengths of 2858 cm^{-1} and 2926 cm^{-1} belong to the stretching oscillation of the C–H bond (Crispín-Isidro et al., 2019; Mali, Dhawale, & Dias, 2017). The appearance of the peak at 1658 cm^{-1} is caused by carbonyl bonds (–HC=O) in the glucose and galactose monomers of moringa seed gum. In addition, the peak appearing at 1000 - 1114 cm^{-1} is related to the C–O bond of the xyloglucan ring in the gum structure (Alpizar-Reyes et al., 2017). The above results prove the existence of gum after extraction from moringa seeds.

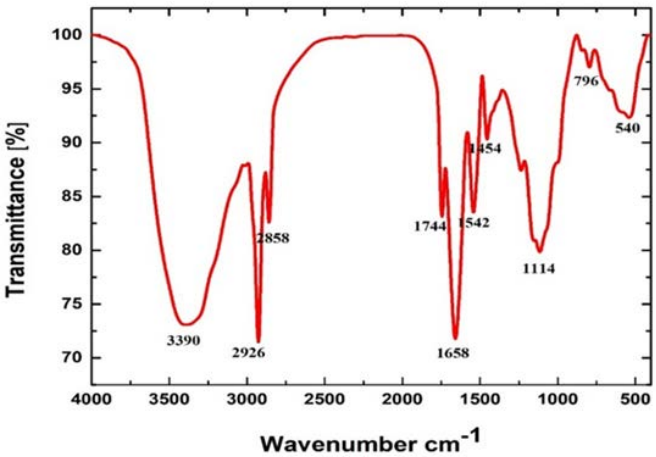

Figure 5. *FTIR infrared spectrum of moringa oleifera seeds gum*

3.3. Optimal factors in RY145 and RR194 treatment with the gum

3.3.1. Optimal pH

Galactomannan is a flocculating agent, a settling aid which is a polysaccharide composed of mannose and galactose. Based on another research, moringa seeds are not only rich in protein (35.37 ± 0.07 g/100 g), fat (43.56 ± 0.03 g/100 g), and minerals (Mg^{2+} and Zn^{2+}) but also glucose in moringa seeds, fructose, and sucrose are the main carbohydrates (Compaoré, Nikièma, Bassolé, Savadogo, & Mouecoucou, 2011), which are the main determinants of gum coagulant properties.

From the research results, it is shown that pH plays an important role in the decolorization of gum. The removal efficiency of color and COD of RY145 at a pH of 9 (basic medium) are the highest at 50.61% and 62.50%, respectively, as illustrated in Figure 6(a). The results for RR194 at a pH of 7 (neutral medium) are at 44.62% and 45.46%, respectively (Figure 6(b)).

Based on the previous research by Blackburn (2004), the flocculation mechanism for dye treatment is explained through three main mechanisms: (i) electrostatic interaction, (ii) van der Waals forces, and (iii) hydrogen bonding between gum and dye. Since moringa seed gum has no electrical charge, the two factors that play a major role in removing RY145 and RR194 are van der Waals forces and hydrogen bonding, similar to those found in Blackburn (2004). Backburn found that hydrogen bonds were formed in two ways: (a) the bond between the -OH group in the polysaccharide molecule and the unpaired electrons on the nitrogen atom and (b) the bond between the -OH group in the polysaccharide molecule with an aromatic ring of the dye molecule.

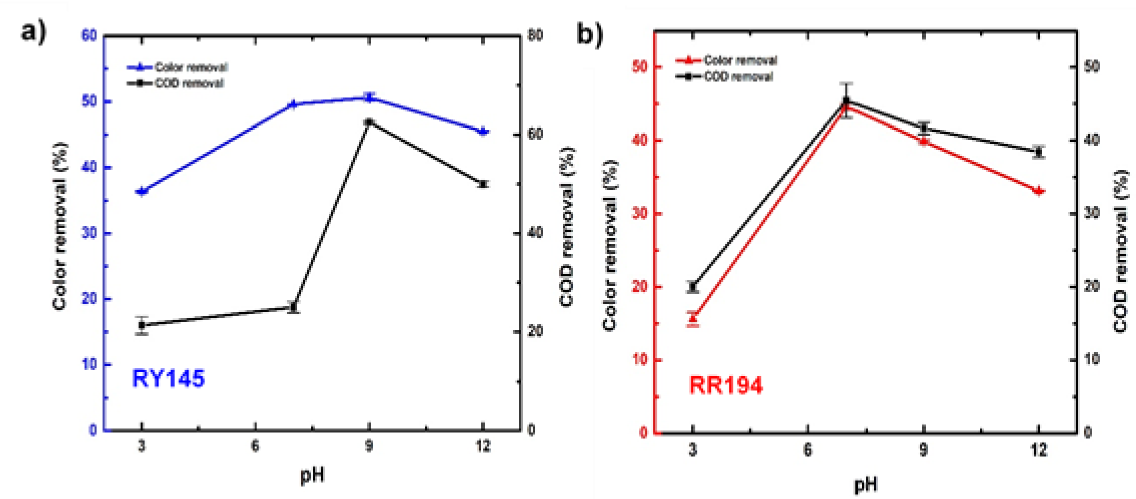

Figure 6. *Effect of pH on the treatment of RY145(a) and RR194(b)*

Each dye color has a different chemical structure and color binding force, so the ability to bind and adsorb gum materials will be different at different pH environments. In the case of highly alkaline media (pH = 12), the carbohydrate bond of the gum may be partially degraded (Whistler & BeMiller, 1958). This makes it difficult for the gum to interact with the dye even if its present form remains hydroxyethyl sulfone at this pH value. Therefore, the decolorization efficiency of gum tends to decrease at a pH of 12.

3.3.2. Optimal gum concentration

The volume of gum is one of the factors that have a direct impact on color treatment. Using excessive gum mass not only affects the treatment efficiency but also affects the cost-effectiveness of the treatment system. Determining the optimal gum mass for flocculation is essential. This experiment was performed by varying the gum concentration from 1000 mg/L to 4000 mg/L and determining the remaining factors to find the optimal gum concentration for the optimal treatment.

Figures 7(a) and 7(b) show that when the concentration of gum increases from 1,000 mg/L to 4,000 mg/L, the decolorization efficiency increases gradually and achieves high efficiency. The best gum concentrations for RY145 and RR194 treatment are 2,000 mg/L and 3,000 mg/L, respectively. The RY145 decolorization and COD removal efficiencies were 62.74%, and 57.14%, and forRR194, they are 50.73% and 54.55%, respectively. However, when the concentration increases beyond the optimal threshold, the decolorization efficiency begins to decrease gradually.

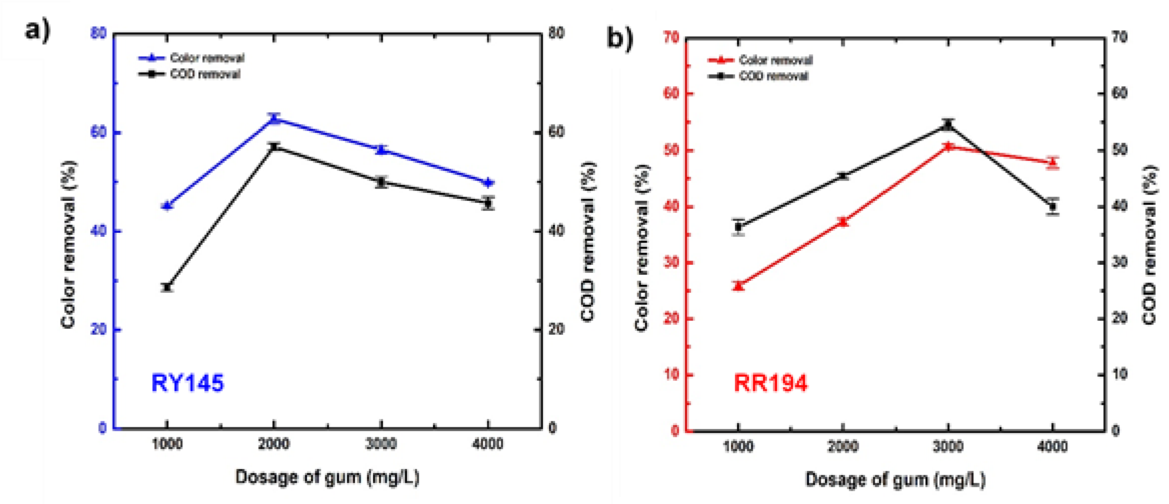

Figure 7. *Effects of gum concentration on the treatment efficiency of RY145(a) and RR194(b)*

The mechanism of color removal with the change of gum concentration can be explained as follows: when the higher concentration of coagulant, the larger the number of charges added to the system, the greater the sweeping effect. Then, the Zeta potential is 0,. combined with the sweeping effect, ? (what) leads to good flocculation ability and increased flocculation efficiency. After the coagulant concentration surpasses a certain threshold, the Zeta potential ≠ 0 will increase the repulsive force between the molecules, reducing the flocculation, and leading to a decrease in the treatment efficiency (Nguyen, Nguyen & Duong, 2021). Too much of the coagulant density will lead to imbalancewith the amount of textile dyeing wastewater. The coagulants will become excess substances that affect the decolorization and COD treatment process.

3.3.3. Optimal color concentration

After determining the optimal pH and gum concentration, this study continues to conduct a color concentration survey. This experiment investigates the influence of color concentration on the efficiency of the coagulation process. The color concentration varies between 20 mg/L and 80 mg/L, and the remaining factors are fixed.

Figures 8(a) and Figure 8(b) show that the decolorization and COD reduction of the gum were affected by the color concentration. During the survey, it is also observed that the higher the color concentration, the lower the decolorization and COD removal efficiency. Specifically, the decolorization and COD reduction efficiencies of RY145 and RR194 are optimal at 40 mg/L and 20 mg/L, respectively.

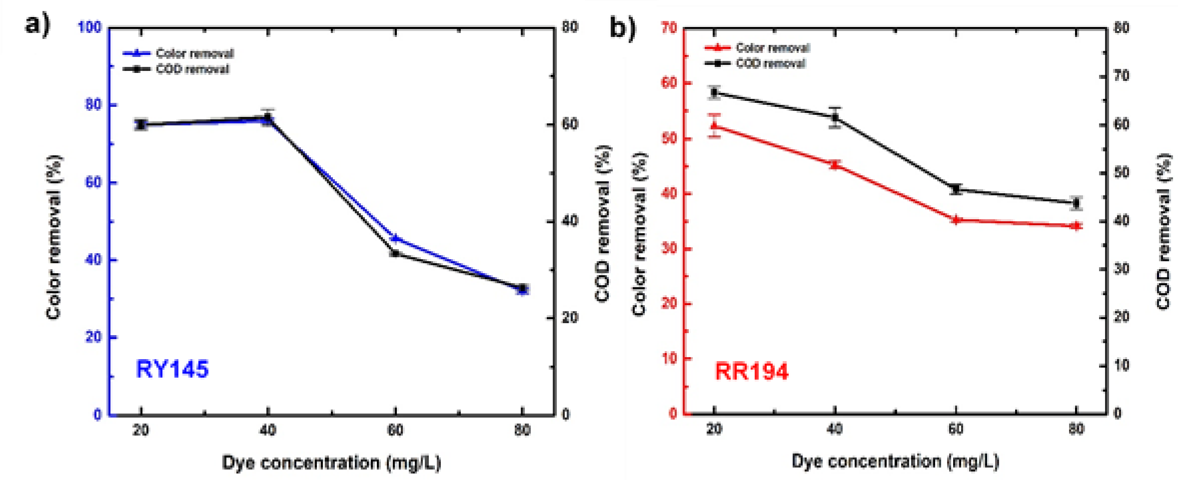

Figure 8. *Effect of color concentration on treatment efficiency of RY145(a) and RR194(b)*

The highest RY145 decolorization and COD removal efficiencies are at 40 mg/L concentration (75% and 60%, respectively). The results show that the decolorization of RY145 and COD removal efficiencies at 20 mg/L and 40 mg/L are both the highest and quite similar to each other. Therefore, it has been demonstrated that at a concentration of 40 mg/L, moringa seed gum can still be processed. The highest RR194 decolorization and COD removal efficiencies are at 20 mg/L concentration (52.37% and 66.67%, respectively).

When the color concentration is too high, the decolorization and COD removal efficiency of both dyes decrease. This is explained by the fact that the gum concentration is not enough to interact with the color concentration when the color concentration increases above the optimal threshold. Therefore, to improve the efficiency of color treatment at higher color concentrations, it is necessary to add gum to the treatment process.

3.3.4. Optimum agitating speed

The stirring speed determines the homogenous distribution of the coagulant in the solution. In addition, the stirring speed also determines the ability to form and maintain the "colloidal cotton." The appropriate mixing speed will help the glue balls collide and stick together to form stronger glue balls. This experiment is conducted with rapid stirring for the first 2 minutes at a speed of 180 rpm, then gradually increased from 15 rpm to 90 rpm to determine the optimal speed in the decolorization of gum. Figures 9(a) and 9(b) shows that the decolorization and COD removal efficiencies of the RY145 and RR194 dyes reach the optimal efficiency at 60 rpm and 45 rpm, respectively.

The RY145 decolorization and COD removal efficiency reach the highest at 60 rpm (85.53% and 69.23%, respectively), and those of RR145 reach the highest at 45 rpm (74.35% and 69.23%, respectively).

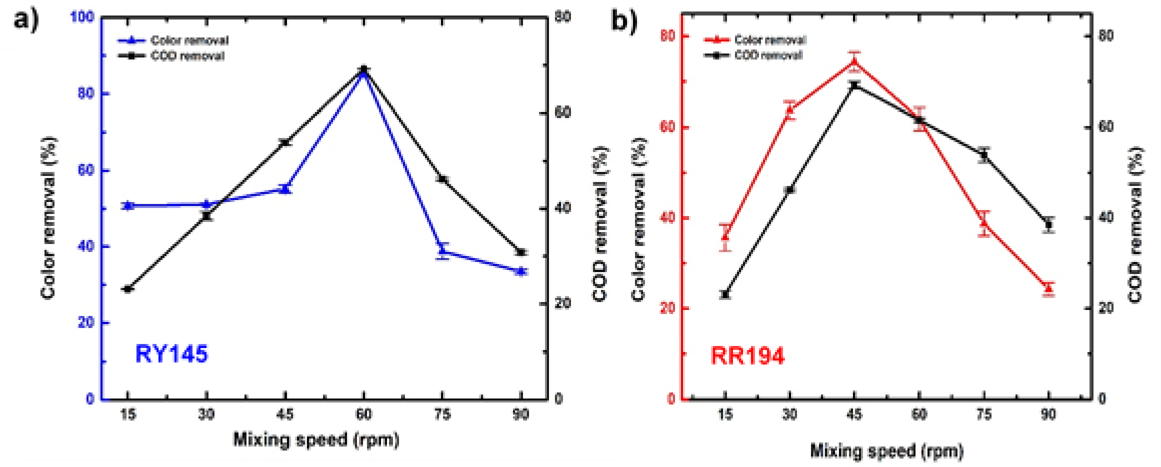

Figure 9. *Effect of stirring speed on treatment efficiency of RY145(a) and RR194(b)*

If the stirring speed continues to increase gradually from 75 rpm to 90 rpm, the decolorization efficiency decreases sharply. It is explained that with increasing stirring speed, the efficiency decreases sharply due to excessive collision between the gum and colloidal cotton particles, causing them to break.

3.3.5. Optimal mixing time

Stirring time mainly depends on the characteristics of the coagulant and the dye color. To determine the influence of stirring time on the flocculation process as well as the optimal stirring time for dye treatment, the experiment is conducted by using different stirring times: 15, 35, 45, 55, 65, and 75 minutes.

Figure 10(a) shows that the highest decolorization efficiency of RY145 and COD removal after 55 minutes are 87.77% and 76.92%, respectively, while the highest of RR194 and COD removal efficiency after 65 minutes are 89.31% and 80%, respectively (Figure 10(b)). However, as the stirring time continues to increase beyond the optimal threshold, the efficiency begins to decrease. Increasing the stirring time to 75 min reduces both decolorization and COD removal efficiency.

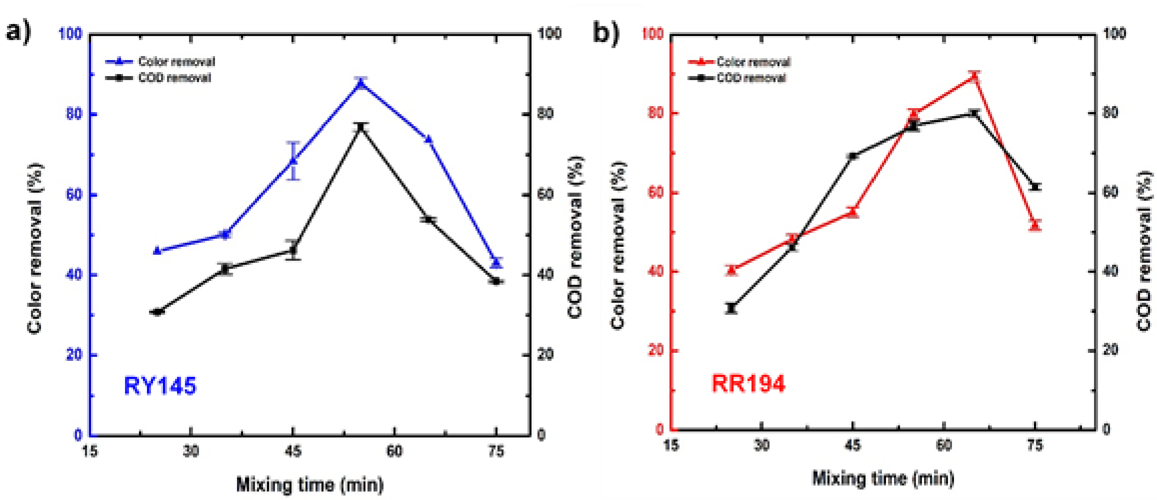

Figure 10. *Effect of stirring time on the treatment efficiency of RY145(a) and RR194(b)*

4. Conclusion

To solve the problem between developing the economy and the environment, biological pollutants can still be treated effectively, and the cost is not too high. In the past, chemical coagulants were used to treat wastewater, but the discharged sludge carrying chemicals was still a problem and incurs the cost of sludge treatment. The biological coagulant (gum) extracted from moringa seeds is then used to remove the RY145 and RR194 from textile wastewater samples.

The color removal efficiency of RY145 color and COD at optimal points of pH = 9, gum concentration (2000 mg/L), color concentration (40 mg/L), stirring speed (60 rpm), time stirring (55 min) are 87.77% and 76.92%, respectively. The color removal efficiency of RR194 color and COD at optimal points of pH = 7, gum concentration (3000 mg/L), color concentration (20 mg/L), stirring speed (45 rpm), time stirring (65 min) reaches 89.31% and 80%, respectively. Therefore, the biological flocculation material (gum) from the specific seed endosperm in this study, moringa seed, is considered a novel material that can replace chemical materials for textile dyeing wastewater treatment in the future. Study on the ability to treat dyes on actual wastewater samples from the home machine.

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

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KHẢO SÁT HIỆU QUẢ XỬ LÍ MÀU REACTIVE YELLOW 145 VÀ MÀU REACTIVE RED 194 BẰNG GUM TRÍCH LI TỪ HẠT CHÙM NGÂY (*MORINGA OLEIFERA)*

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TÓM TẮT

Trong nghiên cứu này, đã nghiên cứu tạo ra chất keo tụ sinh học (gum) được trích li từ hạt chùm ngây làm chất keo tụ xử lí màu Reactive Yollow 145 và màu Reactive Red 194. Đặc tính của vật liệu được xác định bằng các phương pháp kính hiển vi điện tử quét (SEM), phương pháp phân tích FTIR. Hình ảnh chụp SEM cho thấy vật liệu thấy cấu trúc bề mặt của vật liệu sinh học gum trích li từ hạt chùm ngây có sự xuất hiện của các lỗ rỗng tương đối mịn và các hạt gum phân bố tương đối đồng nhất trên bề mặt, phổ FTIR chỉ ra rằng vật liệu điều chế được có chứa các nhóm chức bề mặt –OH, –HC=O và C-O của vòng xyloglucan. Các yếu tố ảnh hưởng đến quá trình keo tụ xử lí màu RY145 và màu Reactive Red 194 bao gồm: pH, nồng độ gum, nồng độ màu, tốc độ khuấy và thời gian khuấy. Từ kết quả nghiên cứu cho thấy chất keo tụ sinh học (gum) được điều chế từ hạt chùm ngây có khả năng khử màu và COD, là vật liệu thân thiện với môi trường, có tiềm năng lớn trong việc ứng dụng vào việc xử lí màu nhuộm trong nước thải dệt nhuộm với hiệu suất khử màu RY145 và khử COD (tương ứng lần lượt: 87.77% và 76.92%) (tại pH = 9 , nồng độ gum (2000 mg/L), nồng độ màu (40 mg/L), tốc độ khuấy (60 vòng/phút), thời gian khuấy (55 phút)*, hiệu suất màu RR194 và khử COD (tương ứng lần lượt: 89.31% và 80%)* (tại pH = 7, nồng độ gum (3000 mg/L), nồng độ màu (20 mg/L), tốc độ khuấy (45 vòng/phút), thời gian khuấy (65 phút).

Từ khóa: chất keo tụ; gum; hạt chùm ngây; Reactive Yellow 145; Reactive Red 194; nước thải dệt nhuộm