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A Combined Experimental and Kinetic Study on Crystal Violet Removal from Wastewater using Polyalkyd/Salvadora Persica Composite

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Abstract

In this study, a novel bio-composite was synthesized from inexpensive raw materials e.g., Salvadora persica, fatty acid, and glycerin. Polyalkyd/Salvadora persica bio-composite was evaluated as a new and effective sorbent for removing the hazardous cationic dye e.g., crystal violet from wastewater. The factors affecting the crystal violet removal process were examined e.g., the Polyalkyd/Salvadora persica bio-composite dosage, crystal violet dye concentration, contact time, pH, and wastewater temperature. The results indicated that the Polyalkyd/Salvadora persica bio-composite was highly efficient for removing crystal violet dye from acidic and alkaline solution (pH 2-13) within 30 minutes at room temperature (25 °C). To examine the sorption mechanism for the removal process, the results and experimental data were analyzed using several diffusion and kinetic models e.g., Morris–Weber, Reichenberg, Bangham, In addition to pseudo-first-order, pseudo-second-order. From the results obtained the pseudo-second-order equation best represented for the removal of crystal violet from wastewater. The thermodynamics parameters (ΔH, ΔG, and ΔS) were also calculated, and the results showed that the removal process of crystal violet using Polyalkyd/Salvadora persica bio-composite was spontaneous and exothermic, suggesting a decrease in randomness during the removal process.

Keywords: Bio-composite; Polyalkyd; Salvadora persica; Crystal violet; Removal; Kinetic.

Introduction

The Salvadora persica tree is also called arak, miswak, and toothbrush tree, it is a small tree with a crooked trunk, the root bark is like sand, and the inner surface color is an even lighter shade of brown. The sticks of Salvadora persica have been used for centuries as a natural toothbrush and have recently been promoted by the World Health Organization (Alili et al., 2014). The sticks of salvadora persica contain lignin, cellulose, and hemicelluloses. Also, it includes several medically beneficial properties including abrasives, enzyme inhibitors, astringents, antiseptics, detergents, and fluoride

(Amoian, et al., 2010). Additionally, Salvadore persica was employed as an effective sorbent for dyes removal (Moawed and Abulkibash, 2012; Moawed, 2013). The leaching of some of its components necessitates the preparation of a chemically stable salvadora persica for the removal process.

Alkyd resin is a thermoplastic polyester polymer that was modified using fatty acid (Lawandy et al. 2017). Alkyd resin was prepared through condensation (esterification) of three types of monomers: polyhydric alcohols (polyol: glycol, glycerol), polybasic acids, and fatty acids (stearic and palmitic acids). Alkyd resin characteristics as low cost, availability, eco-friendly, excellent wetting properties, and good adhesion. Alkyd resins consist of functional groups e.g., ester group, carbonyl site, and allylic methylene group (Albadarin et al. 2017).

discharge of industrial wastewater from various sectors, including paints, plastics, paper, leather, and textiles, has emerged as a critical environmental concern (Jawad et al., 2018). Dyes affect the physicochemical characteristics of freshwater, including color and pH (Al-Tohamy, et al., 2022; Zhou *et al*., 2011). Basic Violet 3, Gentian Violet, and Methyl Violet 10B are the names of Crystal Violet. It is a chemically synthesized dye that belongs to the triphenylmethane family. It is used in industries such as textiles, dyeing, ink, paper, additives, leather, cosmetics, and biochemistry (Mittal, Mariyam, et al., 2021). The wastewater containing crystal violet has become a focus of remediation studies (Dmitrieva *et al*., 2022). For this reason, a range of biological and physicochemical methods, including adsorption, nanofiltration, electrokinetic coagulation, coagulation and precipitation, advanced chemical oxidation, electrochemical oxidation, ozonation, supported liquid membranes, liquid-liquid extraction, and biological processes (Liang *et al*., 2020; Tee *et al*.,2022; Moawed *et al*.,2013). The adsorption process offers selectivity, recyclability, cost-effectiveness, ease of operation, and broad applicability (Tee *et al*., 2022; Munagapati *et al*., 2021; Mittal, *et al*.,2021). Several types of sorbents were used for removing crystal violet) such as sulfonated pomegranate peel biochar (Jawad et al., 2021), chitosan/carbon-doped TiO_2 composite (Abdulhameed et al., 2022), Ag-doped MnO_2 (AgMC) nanocomposites (Kumar et al., 2020), immobilized chitosan (Pitchay et al., 2022), and TiO_2 Degussa P25 (Mubarak et al., 2020). Activated carbon is one of the most widely used adsorbents for removing dyes (Somasekhara et al., 2017). Alternative materials like natural and plant-based adsorbents have been adopted as cost-effective, efficient, and renewable solutions (Gayathiri et al., 2022). Biosorbents such as pyracantha coccinea (Akar et al., 2010) orange peel (FENG et al., 2009), mango seed (Wang et al., 2022), rice bran, wheat bran (Wang et al., 2008), Moringa seeds (Soliman et al., 2019), and skin almond waste (Atmani et al., 2009) also were used.

This work describes simple and inexpensive techniques for the preparation of a new biosorbent (Polyalkyd/Salvadora persica Composite). Parameters controlling the removal of crystal violet, including pH, initial concentrations, shaking time, and solution temperature were optimized.

Materials and Methods

Materials and Reagents

Using 0.15 g of pure crystal violet dye was dissolved in 100 mL of distilled water to produce a stock solution of crystal violet (Molecular Weight: 407.99). The source of Salvadora persica was a Saudi Arabian plantation near El-Khtrichia. Salvadora persica sticks were cleaned using tap and distilled water, dried for 24 hours at 100 °C then blended the dried sticks. Salvadora persica powder was sieved to produce a biosorbent with uniformly sized particles. Polyalkyd/Salvadora persica Composite was prepared as follows: The salvadora persica powder was treated with 100 mL of 0.1 mol L^{-1} $KMnO_4$ in 0.1 mol L^{-1} H_2SO_4 for 3 h at room temperature and then refluxed with 2 g of Stearic acid, 1g of phthalic acid, and 2 g of glycerol for 2 h. After being washed with methanol and distilled water, the product was dried at 25 °C.

Apparatus

The absorbance measurements were taken with a JASCO UV/VIS Spectrometer V630 (Japan). The pH values were measured with a JENWAY 3510 pH meter (UK). The electronic balance SHIMADZU TW423L (Japan) was employed.

Methods

The removal of crystal violet dye using Polyalkyd/Salvadora persica Composite was evaluated using batch experiments. A 0.1 g of Polyalkyd/Salvadora persica Composite was shaken with 25 mL of the crystal violet solution for 1 h at room temperature. The solution samples were filtered, and the concentration of the remaining dye was determined spectrophotometrically at λmax 590 nm. The percentages of crystal violet removal (%E) were calculated from the following equations: $\%E = \left(\frac{C_0 - C_e}{C_0}\right) \times 100$ Where C_o is the initial crystal violet concentration, C_e is the concentration of crystal violet in solution at equilibrium.

Results

To evaluate the efficiency of the Polyalkyd/Salvadora persica Composite for removing hazardous organic dye from wastewater, the numerous factors affecting the removal process were evaluated (Abdulhameed et al., 2022). The contact time between the polyalkyd/Salvadora persica composite and crystal violet dye solution was first examined at pH=7 at room temperature (Figure 1). The initial removal rates of crystal violet are very rapid, 87 % of the total amount of crystal violet was removed within the first 5 minutes. Then the rates became slower with the increase in time until reaching 98 % at 30 minutes. It can be noted that the removal rate for crystal violet is significantly higher than those reported for many sorbents (Moawed et al. 2019).

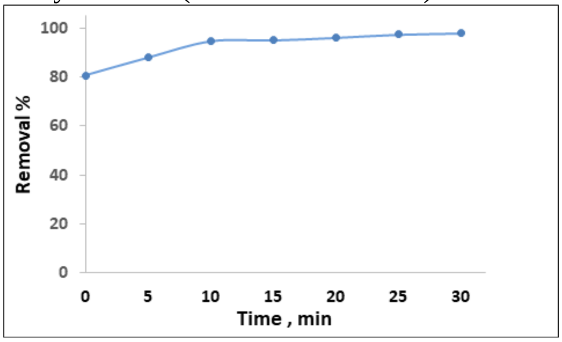

Figure 1. Effect of time on the removal of crystal violet using Polyalkyd/Salvadora persica Composite

A sorption kinetic study is essential in investigating the removal mechanism, which is necessary to evaluate the dye uptake efficiency.

To examine the sorption mechanism and the rate-controlling processes involved in the sorption process, pseudo-first-order ($Log(q_e - q_t) = log\ q_e - k_1/2.303$) and pseudo-second-order ($t/q_t = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$) calculations were employed.

The sorption capacity at equilibrium is indicated by Qe and Qt, while the pseudo-first-rate constant and pseudo-second-order rate constant are denoted by k1 and k2, respectively, and t is the time in minutes. The pseudo-second-order sorption model's R2 value of 0.999 (Figure 2) is greater than the pseudo-first-order sorption model's R2 value of 0.845 (Figure 3). The pseudo-second-order is hence more prevalent. A chemical adsorption mechanism governs the elimination process (Moawed et al., 2013).

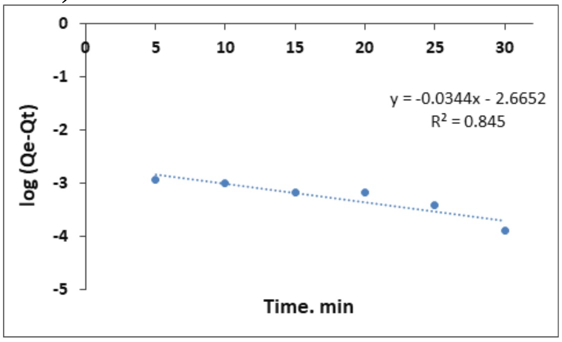

Figure 2. Pseudo first-order model for removal of crystal violet using Polyalkyd/Salvadora persica Composite

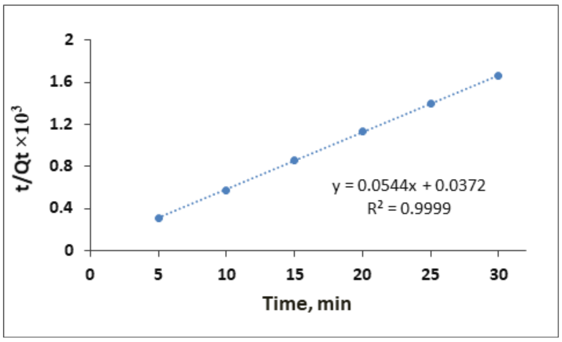

Figure 3. Pseudo second order model for removal of crystal violet dye by using Polyalkyd/Salvadora persica Composite

The Morris–Weber method was used to examine the diffusion mechanism ($Q_t = k_m\sqrt{t}$), Reichenberg ($B_t = -0.497 - Ln\ (1-f)$) and Bangham ($loglog(C_0 - C_t) = \log(k_b m/2.303v) + \alpha log t$) equation.
where Qt is the quantity of sorbed crystal violet at time t and km is the particle diffusion rate constant (mmol/g $min_{1/2}$). The mathematical formula for the B_t value is $F = Q_t/Q_e$, where α, k_b is constant.

Plots of Qt versus $t_{1/2}$ for the diffusion of crystal violet using Polyalkyd/Salvadora persica Composite according to the Morris Weber model (Figure 4) provide straight lines that do not go through the origin and have an R2 value of 0.844. The diffusion rate constant is found at 0.0005 mmol/g min1/2. Figure 5 shows the Bangham equation's double logarithmic plots beside the time yield linear curve. The correlation coefficient R^2 for the removal of crystal violet onto Polyalkyd/Salvadora persica Composite is 0.907, This finding indicates that each sorbent's pores allow CV to diffuse, which contributes to the rate-controlling step. The values represent 0.0404 for α. The correlation coefficient between Bt and the Reichenberg diffusion model (Figure 6) for crystal violet removal using Polyalkyd/Salvadora persica Composite is 0.054.

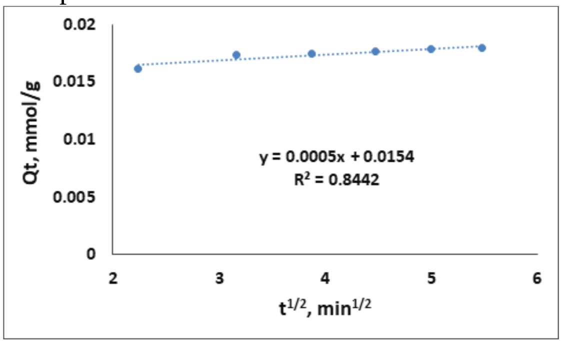

Figure 4. Morris-Weber diffusion model for sorption of crystal violet using Polyalkyd/Salvadora persica Composite

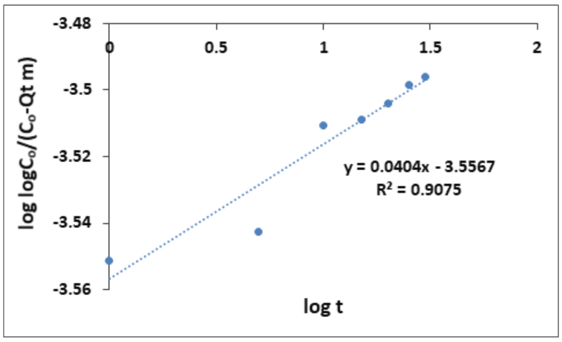

Figure 5. Bangham model for removing crystal violet using Polyalkyd/Salvadora persica Composite

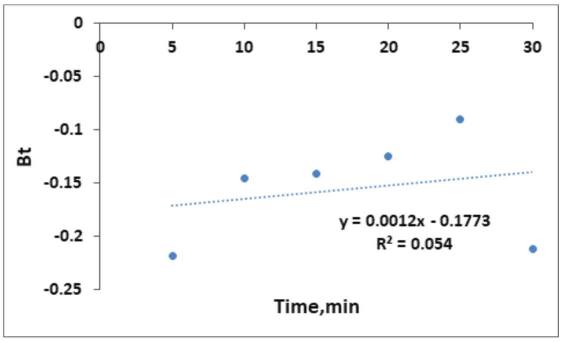

Figure 6. Reichenberg diffusion model for sorption of crystal violet using Polyalkyd/Salvadora persica Composite

The effect of initial crystal violet concentration was studied for different crystal violet concentrations at pH=7 (Figure 7). The removal rates of crystal violet using Polyalkyd/Salvadora persica Composite increased with increasing crystal violet concentrations from 12 to 36 mg/L. The maximum Polyalkyd/Salvadora persica Composite sorption capacity was 30 mg/g.

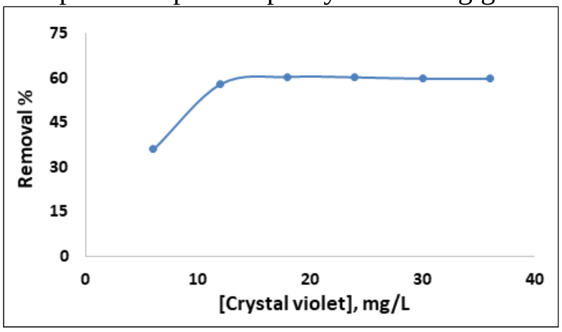

Figure 7. Effect of crystal violet concentration for removal by Polyalkyd/Salvadora persica Composite

The effect of solution pH on the percentage of crystal violet removal from aqueous solution by Polyalkyd/Salvadora persica Composite was illustrated in Figure 8. The result shows removal rates of crystal violet increase from 53% to 96%. by increasing the pH values from 2 to 13

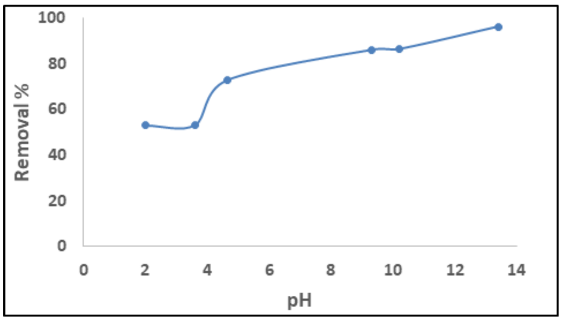

Figure 8. Effect of pH on crystal violet removal using Polyalkyd/Salvadora persica Composite

Effect of Polyalkyd/Salvadora persica Composite dosage of the sorption of crystal violet is usually increased by increasing the amount of the sorbent due to increasing the number of sorption sites. It was found that only 0.9 g is enough to reach 95 % sorption (Figure 9).

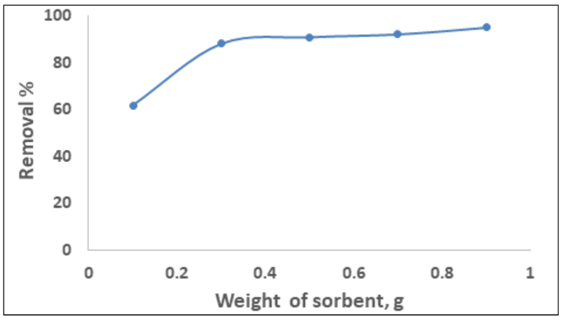

Figure 9. The plot of crystal violet & weight of Polyalkyd/Salvadora persica Composite

Using Polyalkyd/Salvadora persica Composite, the impact of temperature on crystal violet sorption was examined (Figure 10). The removal rates of crystal violet were decreased gradually with increasing temperature from 25ºC to 60ºC. The removal rates of crystal violet using Polyalkyd/Salvadora persica Composite were decreased from 30% to 17% with increasing temperature.

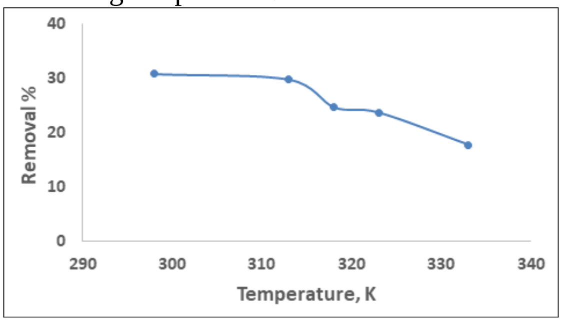

Figure 10. Effect of temperature on removing crystal violet using Polyalkyd/Salvadora persica Composite

The thermodynamic parameter for the sorption process of crystal violet (Figure 11) can be determined using the following equations: $lnK = -\Delta H/RT + \Delta S/R$ and $\Delta G = \Delta H - T\Delta S$, Where K is the equilibrium constant for sorption, T is the temperature (K), and R is the gas constant (8.314 J/mol K). The value of ΔH and ΔS Were calculated from the slope and the intercept of ln K versus 1/T was calculated. The enthalpy (ΔH) was -15.51 kJ/mol, and the negative value of ΔH revealed that the removal process of crystal violet using Polyalkyd/Salvadora persica Composite is exothermic. The Gibbs free energy (ΔG) was -11.86 kJ/mol; these values attributed to the sorption process are spontaneous. Finally, the entropy (ΔS) of removal crystal violet using Polyalkyd/Salvadora persica Composite was -12.5 J/K mol, the negative value suggests a decrease in randomness during the removal process.

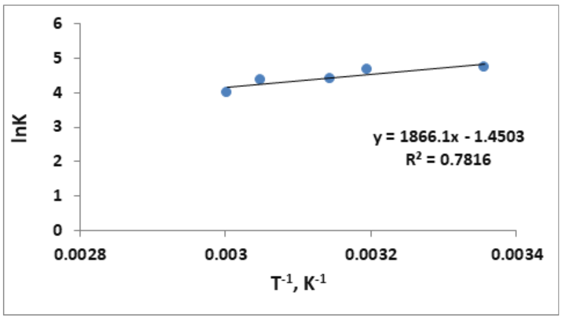

Figure 11. Thermodynamic for removal of crystal violet dye using Polyalkyd/Salvadora persica Composite

Discussion

Using agricultural by-products like Salvadora persica as a cost-effective way to clean wastewater that contains dyes is quite interesting. Miswak is an extremely sensitive and selective adsorbent for removing crystal violet dye from aqueous solutions, according to our research. Polyalkyd/Salvadora persica was produced using glycerol, phthalic acid, stearic acid, and oxidized miswak. It has advantageous properties such as great chemical stability, affordability, and rapid extraction (Ofomaja and Unuabonah 2011), the removal process is most likely a two-step mechanism that involves the sorption of crystal violet from solution onto the Polyalkyd/Salvadora persica Composite surface and the subsequent electrostatic attraction between the anionic Polyalkyd/Salvadora persica Composite and the positive dye molecules.

To determine the ideal conditions for removing dye from wastewater, we have examined the sorption behavior of crystal violet dye onto Polyalkyd/Salvadora persica. The sorption percentage of crystal violet dye increases as pH levels increase. The Polyalkyd/Salvadora persica Composite's surfaces have a positive charge when the pH is less than 4. Because the Polyalkyd/Salvadora persica Composite's positively charged surface electrostatically repels the crystal violet molecules in an acidic medium (pH 1-4), the crystal violet molecules are thus difficult to remove. However, the electrostatic attraction between the positively charged dye and the negatively charged Polyalkyd/Salvadora persica Composite surface was responsible for the elimination process at pH 4–13.

The extremely intense sorption of crystal violet onto Polyalkyd/Salvadora persica composite has been confirmed by kinetics and isotherm studies; the developed procedure is high-tolerance to interfering ions, simple, quick, and inexpensive, and the faster adsorption kinetic data may be useful for environmental technologies in the removal of crystal violet from wastewater.

Different models were used to analyze the diffusion and kinetic curve data (Moawed et al. 2019). The results indicate that the diffusion rate is rapid and reflects the formation of partial film along with the intraparticle diffusion. The correlation coefficient values or the removal of crystal violet dye by using Polyalkyd/Salvadora persica Composite due to Morris Weber, Bangham, and Reichenberg diffusion model were 0.844, 0.907, and 0.054, respectively. These results demonstrate that the diffusion of crystal violet molecules into the pores of each sorbent participates in the rate-controlling step due to Bangham.

Batch extraction mode was used to measure the rate at which crystal violet was extracted onto Polyalkyd/Salvadora persica Composite. According to the results, it takes 30 minutes to fully extract crystal violet onto Polyalkyd/Salvadora persica Composite. Crystal violet is extracted at an extremely fast speed at first, then as time passes, the rate gradually slows down. The outcome shows that the rate of crystal violet sorption was quick.

Strong bond formation with the Polyalkyd/Salvadora persica composite is the cause of this crystal violet sorption rate. To fit the experimental results for the sorption of crystal violet onto the Polyalkyd/Salvadora persica composite, the pseudo-first-order and pseudo-second-order kinetic models are assessed. Compared to the pseudo-first-order model, the pseudo-second-order sorption model has a greater R2 value. This suggests that the pseudo-second-order sorption mechanism is more prevalent. The Langmuir model explains the experimental data well, according to the equilibrium isotherms.

The negative value of ΔG indicates that the sorption process was spontaneous. The exothermic character of the sorption of crystal violet dye is indicated by the negative value of ΔH. Salvadora persica/polyalkyd Under ideal conditions, composite biosorbents demonstrated their effectiveness in removing crystal violet dye from wastewater. The Polyalkyd/Salvadora persica composite's reusability for repeated adsorptions was validated by the desorption investigation, which also raises the opportunity for dye desorption. The Polyalkyd/Salvadora persica Composite is an economical adsorbent for removing cationic dyes from wastewater, according to the results.

Conclusion

Polyalkyd/Salvadora persica Composite was evaluated for the removal of crystal violet dye the maximum sorption capacities are found to be 30 mg/g, within 30:35 min over a wide pH range (2-13). In addition, the equilibrium, kinetics, and thermodynamics of the crystal violet sorption onto Miswak were studied. The negative values of ΔG and ΔH suggest that the sorption of crystal violet dye is spontaneous and exothermic. This study suggests that Miswak can remove dye from wastewater.

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الملخص العربي

عنوان البحث: دراسة تجريبية وحركية مشتركة لازالة صبغة البنفسج الكرستالية من مياه الصرف الصحي باستخدام مركب مسواك/ بولى ألكيد

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فى هذه الدراسة تم تطوير مادة ماصة متجددة و حيوية من المسواك والتى تعمل كمادة ماصة لازالة صبغة البنفسج الكرستالية من مياه الصرف الصحى وتم التحقق من العوامل المؤثرة على الصبغة مثل تركيز الصبغه و درجه حراره المحلول و درجه حرارة المحلول و كمية الماده الماصة المضافة وايضا وقت حدوث الامتصاص
كانت الممتزات الحيوية)المسواك(عالية الكفاءة في إزالة صبغة البنفسج الكرستالية عند مستويات الأس الهيدروجيني ٢-١٣ في زمن يتراوح من ٣٠ إلى ٣٥ دقيقة عند درجة حرارة ٢٥ درجة مئويةتم تحليل البيانات التجريبية الحركية من الدرجة الاولى و الثانية و أشارت هذه النتائج إلى أن امتصاص صبغة البنفسج الكرستالى كان أفضل تمثيل من خلال معادلة الدرجة الثانية .تم حساب المعاملات الديناميكية الحرارية (ΔH) و ΔG و (ΔS وأظهرت النتائج أن عملية الامتزاز ل (MA – Resin) كانت تلقائية وطاردة للحرارة ، مما يشير إلى انخفاض في العشوائية أثناء عملية الإزالة.
كمادة ماصة لازالة صبغة البنفسج الكرستالية(Poly alkyd Miswak) بالتالى يمكن استخدام مركب.