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Investigation of the Effect of pH in Aqueous Media on Reactive Red 195 Textile Toxic Dye Using Different Adsorbents: pH Optimization for the *Crataegus*

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Abstract –The removal of Reactive Red 195(RR195) textile dye in aqueous medium was investigated with different bio-adsorbents *Crataegus* (*Crataegusspecies* (hawthorn) (Cr), coconut (*Cocos nucifera(*Cn)), grapefruit (*Citrus paradisi*(Ci)), peach seed(*Prunus persica L.*(P)) and SBA-15 material. First experiments were carried out at solution pH and pH=1. Other experimental parameters were determined as adsorbent dosage; 0.1 g, experimental temperature; room temperature, solution amount; 50mL, dye concentration; 100ppm and contact time; 60 min. Experiments were carried out in two parallels and average values were used in the calculations of removal (%) and qe values. In addition, initial concentration values for each sample were determined at 541 nm with UV-VIS. According to the results obtained, it was calculated that especially hawthorn fruit (fruit + seed) used as bio-adsorbent has an efficiency of over 74% in the removal of RR195 dye. However, it was determined that bio-adsorbents obtained only from *Crataegus* seeds were more efficient in the removal of RR 195 dye. It can be stated that the reason for this is especially due to the particle size distribution and organic content difference of bio-adsorbents. Because it can be said that the homogeneity and smallness of the particle size provide the necessary surface areas for the dye to be held on the surface.

Keywords – pH optimization, adsorbents, Reaktif Red 195, Wastewater

I. INTRODUCTION

The world population is expected to reach 12 billion by 2100 [1] and people's habits (preferences) have many positive and negative effects on ecological life in the world. Therefore, societies and countries need to make an effort to keep toxic outputs at a minimum level for sustainable agriculture, healthy society and sustainable ecology. In particular, an increase in toxic chemicals used parallel to industrialization and the increase in world population is observed. One of these toxic chemicals is dyes used in textiles. Textile industry wastewater can pose serious risks to the environment and human health due to the wide variety of chemicals it contains. This problem arises from the widespread use of harmful chemicals, especially dyes. Dyes used in the textile industry are one of the most dangerous chemical compound classes found in industrial wastewater [2]. These dyes must be disposed of in wastewater because they prevent photosynthesis of aquatic flora and reduce light penetration [3,4]. They are also undesirable in terms of aesthetics, drinking and other purposes [5].

In addition to causing allergies, dermatitis and skin irritation [6], they are carcinogenic [7] and mutagenic [8]. However, the textile industry has a significant global importance despite the water consumption and ecotoxicity it causes.

Reactive dyes, one of the industrial dyes, represent a significant part of commercial and synthetic dyes. Reactive dyes are widely used especially in the textile industry and cause an increase in environmental problems due to their release in the ecosystem, toxicity, mutagenicity and non-biodegradability [9-11]. In addition, reactive dyes are in the most problematic group in terms of environmental health since they are not affected much by traditional disposal systems [11,12]. In addition, reactive dyes are resistant to natural biodegradation due to the aromatic rings in their structures [13]. Reactive Red-195 is a textile azo dye distinguished by the azo group (N = N) acting as a chromophore group and SO_3H (sulfonate) acting as an auxochrome group containing a reactive group. More than 750,000 L of clean water is needed to produce just 1 kg of textile dye [14]. In addition, textile wastewater poses a significant ecological problem globally due to its large volume [15]. On a global scale, it is estimated that the textile industry produces more than 50 billion m3 of wastewater per year. Large volumes of wastewater combined with pollutants such as dyes, salts, heavy metals and other chemicals pose serious environmental and health risks if not properly treated before disposal [16]. Moreover, it is very important for the health of living beings to keep such wastewater at a certain level before being discharged into aquatic ecosystems. It is estimated that there are approximately 10,000 different types of dyes and the production amount of dyes on the market today is approximately 700,000 tons per year [17]. The textile industry is the industrial sector that releases the highest amount of dyes to ecosystems with 54% [18]. Other major industries are; dye industry (21%). Textile wastewater contains many different organic and inorganic compounds, especially dyes (azo, anthraquinone, etc.), various chemicals (indigo derivatives, phthalocyanines, xanthene, diphenylmethane and triphenylmethane, cyanines and metals) [19]. Many different methods are used in the removal of textile dyes from wastewater, including adsorption [20, 21], coagulation [22], nanofiltration [23], biosorption [24] and oxidation [25]. The biosorbents used in the biosorption method (any biomass source obtained from bio-adsorbent) are preferred in the treatment of wastewater because they are easy to obtain, cheap, environmentally friendly, and can be used directly without any complicated process and often without the need for chemicals.

Therefore, it is very important to treat such wastewater and to use environmentally friendly techniques during treatment. The main objective of this study is to investigate the removal pH optimizations of bioadsorbents and SBA-15 adsorbent by biosorption method to decrease the value of RR 195 dye used in the textile industry in the fresh, and ecosystem of salt water without discharging it into the clean water media, and to protect the sustainability of natural life.

II. Materials and Method

2.1. Materials

Adsorbents used in the removal of RR195 textile dye (Figure 1 and Table 1); bio-adsorbents (*Crataegus* (*Crataegus species* (hawthorn), coconut (*Cocos nucifera*), small peach (*Prunus persica L.*), grapefruit (*Citrus paradisi*) and silica-based mesoporous adsorbent (SBA-15; Santa Barbara Amorphous) were used (Figure 2). Other materials and devices; 10-magnetic mixer, centrifuge device, beaker, volumetric flask, magnetic fish and UV-VIS (T80 UV/VIS Spectrometer PG Instruments Ltd) device.

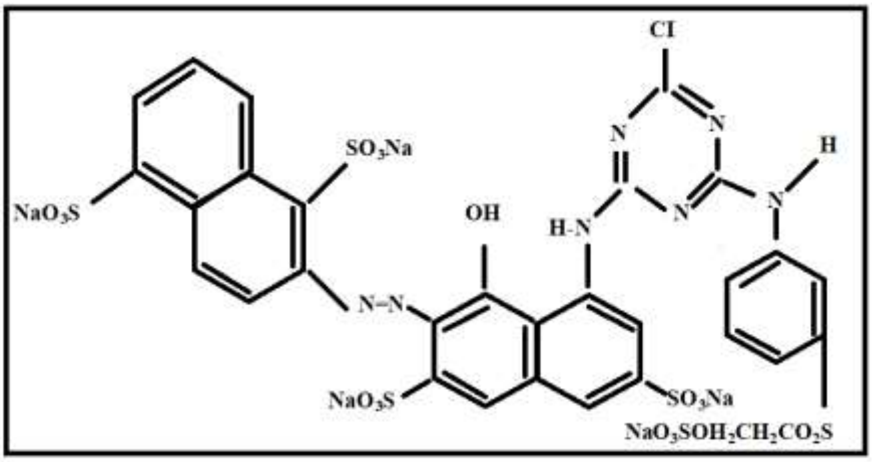

Fig. 1 Molecular structural of RR195

Table 1. RR195 molecular formula and weight

Molecular Formula	$C_{31}H_{19}ClN_7Na_5O_{19}S_6$
Molecular Weight	1136.32 g/mol

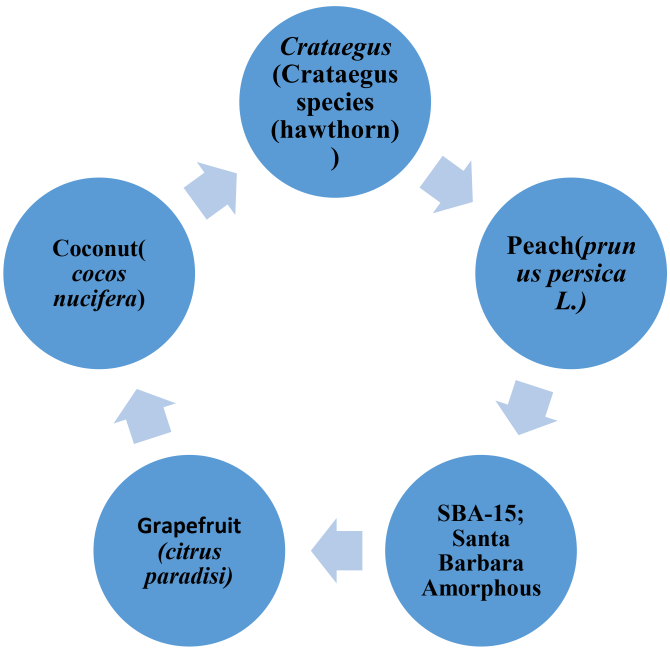

Fig. 2 Adsorbent types

2.2. Methods

The preparation of selected bio-adsorbents was firstly carried out by drying the fruits (biomass) and their wastes (core parts(seed)) collected in their seasons under natural conditions. Then, the grinding processes were carried out. After the grinding process, the bio-adsorbents were sieved to bring them to a certain particle size. The hydrothermal synthesis method was used in the synthesis of SBA-15 material (SBA-15 was synthesized based on the recipe used in the study conducted by Şimşek [26]. Second, 100 ppm stock solution (100 mg/L in deionized water) of RR195 textile dye was prepared. Then, the removals (adsorptions) of the selected Reactive Red 195 textile dye with the obtained bio-adsorbents and SBA-15 material were carried out at the solution pH and pH=1 values, at room temperature, for 1 hour contact time, in 50 mL solution, 100ppm dye concentration and 0.1 gr substance dosage(the samples are named as follows; solution pH: Cr(*Crataegus* seed),Cr*(*Crataegus* fruit and seed), Cn(*Cocos nucifera*), P(*Prunus persica L.*), Ci(*Citrus paradisi*) and SBA-15, pH= 1: Cr_1(*Crataegus* seed),Cr*_1(*Crataegus* fruit and seed), Cn_1(*Cocos nucifera*), P_1(*prunus persica L.*), Ci_1(*Citrus paradisi*) and SBA-15_1).The solution samples were separated from each other by the dye-containing bio-adsorbent and SBA-15 by centrifugation method (4000rpm for 5 minutes). Then, the concentrations of the RR195 solution separated from the bio-adsorbents were determined using a UV spectrophotometer device (541nm). Finally, the best removal result and cheap bio-adsorbent material was selected and pH optimization studies (pH: 1,2,3,4, 6, 8 and 10) were carried out under the same experimental conditions. The experimental setup is shown in Figure 3.

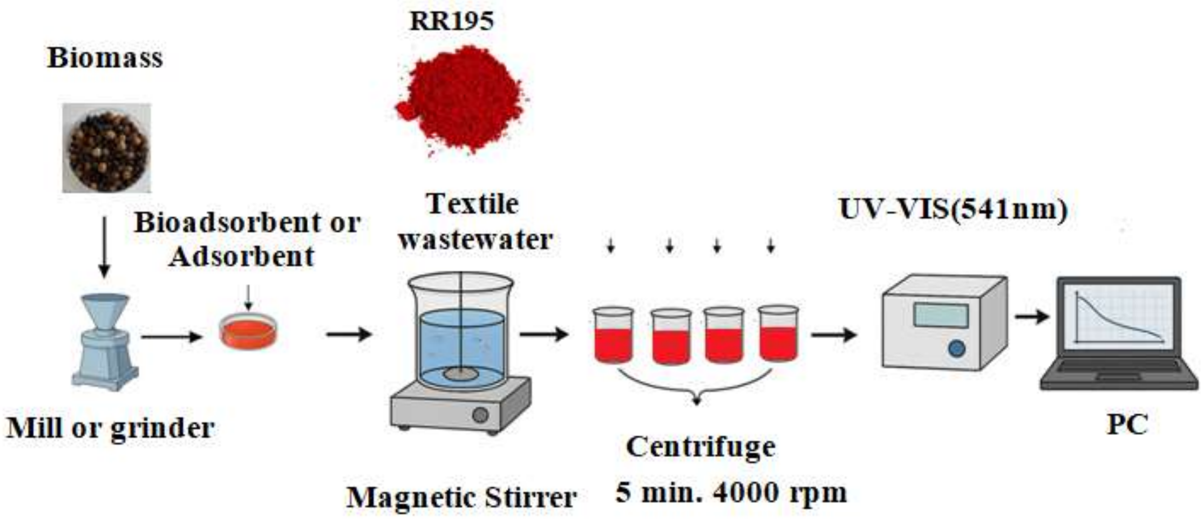

Fig. 3 The experimental setup

III. RESULTS

First, the calibration curve for dye concentrations of 5, 10 and 25 ppm (RR195 dye concentration) was drawn using a wavelength of 541 nm. The R^2 value was obtained as 0.9999 (Figure 4).

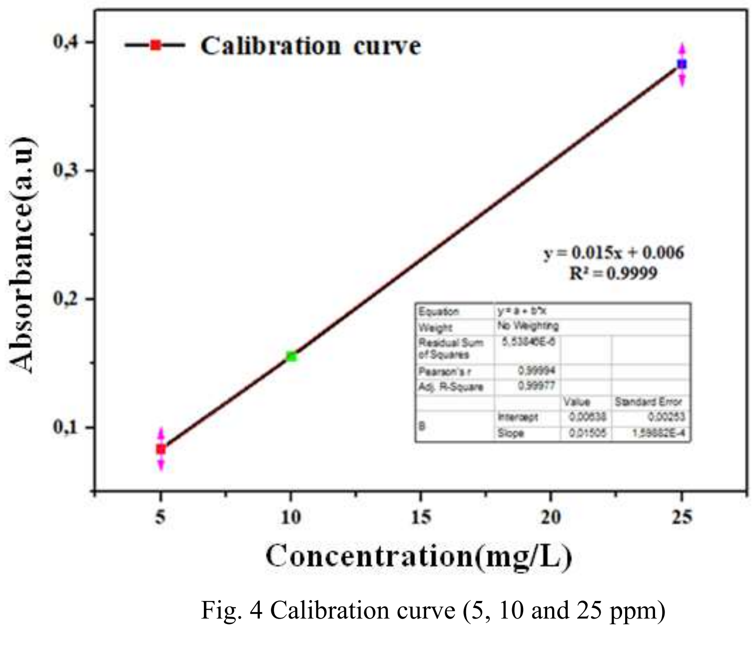

Fig. 4 Calibration curve (5, 10 and 25 ppm)

The experiments were carried out in two parallels (solution pH and pH:1, at room temperature, adsorbent dosage; 0.1g, contact time;1 hour, solution; 50mL, and RR195 concentration; 100ppm).

In addition, the initial concentrations of each pH solution were determined at 541 nm (to reduce the error margin) and then used in the calculation of removal (%) and qe (mg/g) values. The equations used to calculate the removal (%) and absorbance capacity (qe) values are given in Eq 1 and 2, respectively. The removal results of RR195 dye obtained at solution pH and pH:1 values are shown in Figure 5. The results show that the biosorbent formed from *Crataegus* fruit and seed is more efficient in RR195 removal. However, since one of the main objectives of this study is to evaluate bioadsorbents obtained from organic waste biomass, pH optimization studies were carried out on bioadsorbents from *Crataegus* seeds. In addition, it was calculated that *Crataegus* seeds also have high removal values. Moreover, it is predicted that modifications to be made in the experimental conditions (for example, improvements in the homogeneity and size reduction of the bioadsorbent) will have a positive effect on RR195 removal efficiency. Therefore, pH optimization studies of RR195 dye were carried out with bioadsorbents obtained from hawthorn seeds. The obtained results are shown in Figure 6. It was determined that the highest removal efficiency for this textile dye in aqueous media would be achieved at pH values between 1 and 2.

$$\%(Removal) = \frac{C0 - Ce}{C0}\, X100 \tag{1}$$

Here; C_0 and Ce (mg/L) are respectively the liquid concentrations of initial and at equilibrium dye.

$$qe = \frac{(C0 - Ce) * V}{W} \tag{2}$$

Here; V is the volume of the solution (L), W is the mass of adsorbent (g) and qe(mg/g).

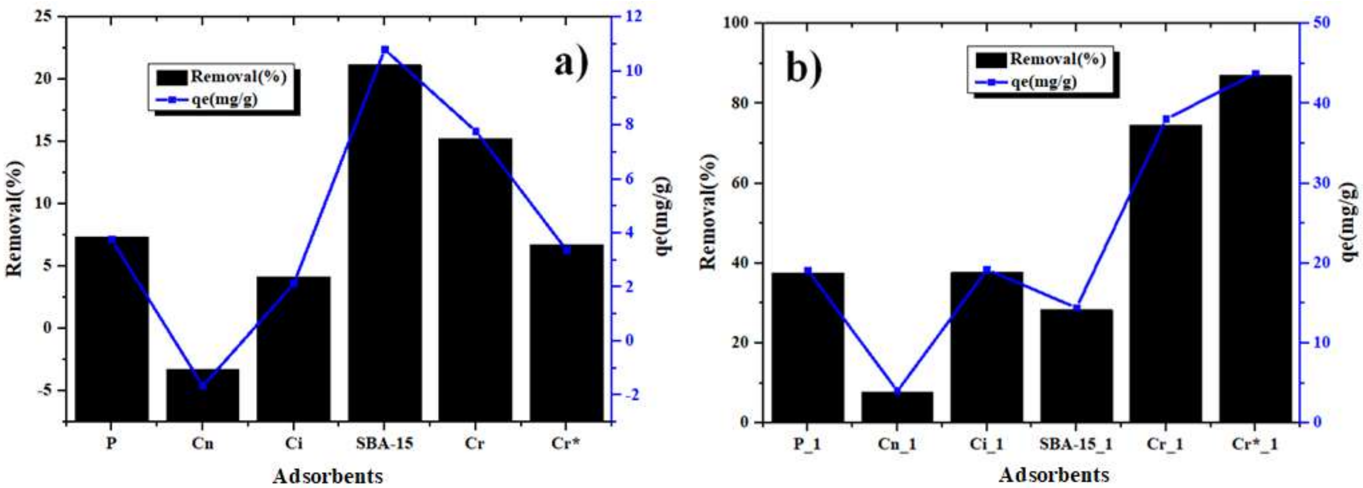

Fig. 5 Removal and adsorption (biosorption) capacity results of RR195 dye with different bio-adsorbents and adsorbent at room conditions, (a) solution pH and (b) pH=1

pH is an important parameter affecting chemical adsorption in terms of controlling the ionization degree of the adsorptive molecule and the surface charge of the adsorbent [27].

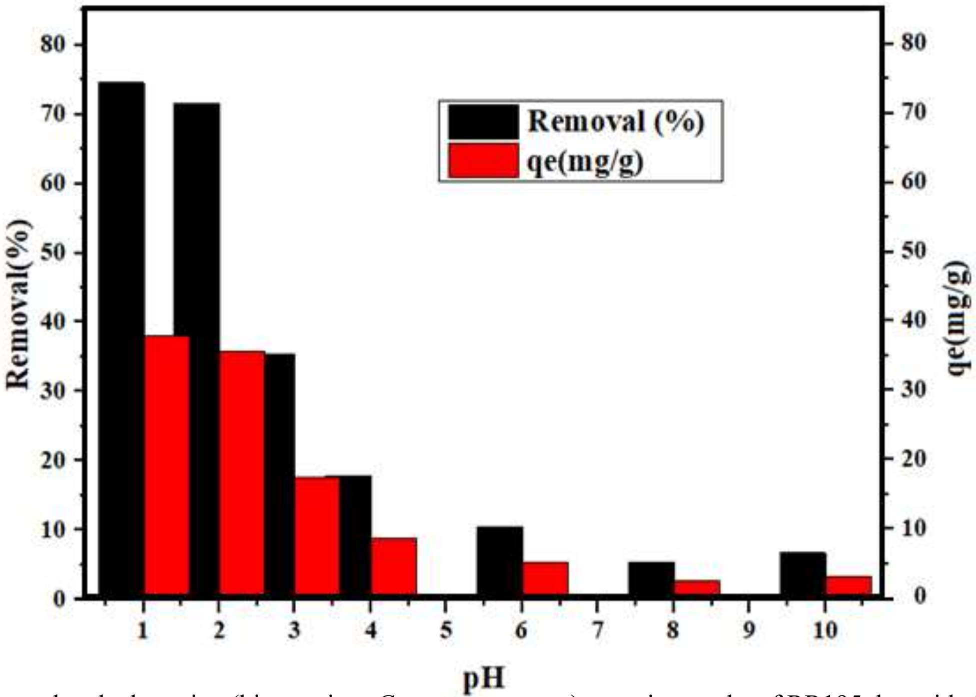

Fig. 6 Removal and adsorption (biosorption; *Crataegus pattern*) capacity results of RR195 dye with different pH

IV. DISCUSSION

These preliminary studies have provided important data for the removal of RR 195 textile dye. Especially the fact that cheap, eco-friendly bio-adsorbents obtained from organic biomass sources have higher removal values makes these studies valuable. In addition, these studies will be detailed in the future with bio-adsorbent size, contact time, dosage amount, and dye concentration parameters. It is foreseen that especially particle size will play an important role in textile dye removal. The dosage amount used may be higher than actived carbons. On the other hand, one of the points that makes this study important is that no chemical is used in the adsorbent production (i.e. it completely includes green chemistry). These features make this study much superior to other adsorbents (such as activated carbons, silica-based sieves, etc.).

As a result, it can be predicted that it can make a significant contribution to the continuity of sustainable freshwater and salt water ecosystems.

V. CONCLUSION

In this study, the removal of RR195, a commonly used azo dye in the textile industry, from aqueous media was investigated using natural bioadsorbents and SBA-15. RR195 removal experiments in aqueous medium were performed in solution, and pH 1 (room temperature, 100 ppm dye concentration, 50 ml solution volume, 1 hour, using a magnetic stirrer). Comparing the results, the best result was obtained with the bioadsorbent prepared from hawthorn waste at pH 1. Furthermore, pH optimization was performed under the same experimental conditions with the hawthorn sample. The results showed that the best removal value was obtained at pH 1.

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