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# Simultaneous bioremoval of two unsafe dyes from aqueous solution using a novel green composite biosorbent

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ABSTRACT

Simultaneous bioremoval of Basic red 46 (Br46) and Basic violet 3 (Bv3) from binary mixture aqueous system onto novel green composite biosorbent made from a combination of *Spirogyra* sp. and *Rhizoclonium* sp. green filamentous algal biomasses was investigated in this study. The impacts of various influential parameters such as pH, biosorbent amount, dye concentration and contact time were evaluated using a batch biosorption method. Biosorption equilibrium experiments indicated that the best fit was achieved with Sips isotherm model. The maximum values of biosorption from binary component system were estimated to be 53.303 and 37.734 mg $g^{-1}$ for Br46 and Bv3, respectively. Kinetic modeling studies showed that the co-biosorption process of dyes from dual solution was acceptably described by the pseudo-second-order model. All these findings proposed that the algal composite biosorbent could be used for simultaneously bioremediating an effluent polluted with such dyes.
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## 1. Introduction

Synthetic-based dye applications in various industries like leather, textile, paper, plastics, cosmetics and food are an important contamination source especially for the aquatic environment. Most of these dyes and their degradation products cause serious damage to the ecosystem due to their toxicological properties which pose a major threat to the biota [1,2]. The extensive use of synthetic-based dyes has led scientists to develop new ways of eliminating dyes from impacted media. Various conventional treatment processes such as coagulation-flocculation, activated sludge, ozonation and ion exchange, are either highly expensive or ineffective, and may also cause secondary negative impacts on the ecosystem [3]. Compared with these processes, biosorption technique has been the main focus for remediation of dye pollution due to its effectiveness, low cost and eco-safety in recent years [4–6].

Numerous biosorbents have been studied for removal of dye molecules from water systems. Development of novel effective and eco-friendly biosorbents for treatment of dye impurity is always getting special interest of worldwide researchers [7]. In this context, many species of algae have been used as potential biosorbents due to their ubiquitous occurrence in nature, low cost and high biosorption potential towards dyes [8–12]. Algae are primary producers in ecological systems, widely distributed around the world and closely connected with human life. Algal cell walls consist of different functional groups such as hydroxyl, carboxylate, amino and phosphate which can play an important role to remove pollutants from aqueous solution [13,14].

On the other hand, all of previous dye removal studies using algal biomasses have focused on single algal biomass. The novelty of our study is using phyco-composite material composed of *Spirogyra* sp. and *Rhizoclonium* sp. filamentous green algal biomasses for bioremediation of dye pollution. The important difference between previous studies and this work is gaining the advantages and also using the biosorptive potentials of these biosorbents in a combination. The purpose of blending algal biomasses is having all potentials of biosorbents for dye removal. We chose *Spirogyra* sp. and *Rhizoclonium* sp. as a composite biosorption material due to their similar habitat distribution and close taxonomic grouping. Both are benthic filamentous algae belonging to division of Chlorophyta. These species are naturally abundant throughout the world and ease of harvesting. In recent times, many studies have applied both algae for removal of different pollutants [15–19]. These algal biomasses have also been adequately characterized various analyses including Scanning electron microscopy, Fourier transform infrared spectroscopy and X-ray diffraction [18–21]. Apparently, these two genera of algae have high developmental potential as biosorbent.

Besides, up to now, biosorption studies have mainly focused on the biotreatment of single dye. However, actual wastewater treatment systems often have to deal with a mixture of dyes, and the researches in the literature concerning biosorption in multi-dye systems are scarce [5,22]. Hence, we evaluated co-biotreatment of Basic red 46 (Br46) and Basic violet 3 (Bv3) model dyes from binary mixture aqueous solution onto novel green composite biosorbent composed of *Spirogyra* sp. and

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*Rhizoclonium* sp. in this paper. These synthetic dyes are widely used in various industries, and have harmful effects on biota [23,24]. To the best of our knowledge, there are no reports on the mutual biosorption of Br46 and Bv3. The effects of important operating variables like pH, amount of biosorbent, dye concentration and contact time were studied through bath biosorption experiments. The biosorption mechanism of dual dye onto the composite biosorbent was also evaluated in terms of kinetics, equilibrium. On the basis of the available literature, we hypothesized that this novel green composite biosorbent may be a promising candidate for such biotreatment applications.

## 2. Material and methods

### 2.1. Materials

The mix biomasses of *Spirogyra* sp. and *Rhizoclonium* sp. filamentous green algae was supplied from a local freshwater pool, Sinop, Turkey. These algae belong to division of Chlorophyta. The composite biosorbent was first washed with tap water, followed by several washings with distilled water to remove extraneous materials. It was dried at 80 °C in an oven until a constant weight was achieved. The dried composite biomass was then grounded in a domestic mixing grinder. The resulting material was sieved by using 0.5 mm standard sieve and kept at room temperature.

Basic red 46 (Br46) and Basic violet 3 (Bv3) were provided by a local source. The chemical structures of dyes were shown in Fig. 1. The stock solutions of Br46 and Bv3 (1 g $L^{-1}$) were prepared by dissolving required amount of dyes in distilled water. For binary dye system, the desired combinations of dye molecules were obtained by diluting the stock solutions of dyes and mixing them in the test medium. The ratios of initial concentrations of Br46 and Bv3 were 1:1 over a concentration range of 10–50 mg $L^{-1}$. All chemicals utilized were of analytical reagent grade, and were used without further purification.

### 2.2. Experimental procedure

Batch biosorption studies were performed with the dual synthetic dye mixture at room temperature. The biosorption system pH was adjusted by sodium hydroxide (0.1 mol $L^{-1}$) and hydrochloric acid (0.1 mol $L^{-1}$). A known weight of biosorbent (10 mg) was added to a series of 100 mL Erlenmeyer flasks containing 100 mL mixture of Br46–Bv3 solution and the flasks were periodically shaken at a constant speed. After equilibration, to separate the biomass from solution, the solution was centrifuged and final concentrations of dyes were measured using UV–visible spectrophotometer (Thermo, Genesys 10 S) at wavelengths of 530 nm and 590 nm, respectively. Biosorption capacity of biosorbent, $q_t$ or $q_e$ (mg $g^{-1}$), was determined by:

$$q_t = \frac{(C_0 - C_t)V}{M} \quad (1)$$

$$q_e = \frac{(C_0 - C_e)V}{M} \quad (2)$$

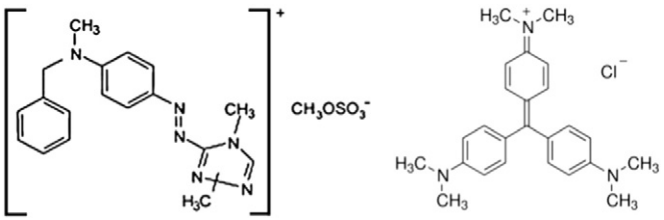


**Fig. 1.** Chemical structures of Br46 (a) and Bv3 (b) dyes.

Besides, biosorption yield (*BY*, %) of biosorbent was obtained by:

$$BY = \frac{C_0 - C_t}{C_0} \times 100 \quad (3)$$

where $C_0$, $C_t$ and $C_e$ (mg $L^{-1}$) are the concentrations of colorant at the initial, a time $t$ and equilibrium, respectively. $V$ (L) is the volume of aqueous colorant solution and $M$ (g) is the mass of biosorbent.

### 2.3. Impacts of operating variables on biosorption

In order to investigate the effect of pH on the biosorption process, the solution pH was adjusted to different values varied from 3 to 9 by using 0.1 mol $L^{-1}$ sodium hydroxide and 0.1 mol $L^{-1}$ hydrochloric acid, measuring them with a pH-meter. Biosorption of dyes from aqueous solution by various biosorbent amounts (10–50 mg) was performed at the optimum solution pH obtained. The dependence of biosorption on the concentrations of dyes was investigated at different dye concentrations which varied from 10 to 50 mg $L^{-1}$ as explained by the biosorption procedure above. Finally, the effect of contact time on the biosorption process was studied in the time range from 0 to 120 min with a fixed biosorbent amount.

### 2.4. Biosorption kinetics

Kinetic experiments were performed in 100 mL flasks containing 100 mL of Br46–Bv3 mixtures with 10 mg of algal biosorbents at room temperature. The flasks were periodically shaken at a constant speed. The samples were taken at predetermined time intervals, centrifuged and analyzed for the residual concentrations of dyes. The pseudo-first-order, pseudo-second-order, Elovich, logistic and intra-particle diffusion models were used to explain the biosorption kinetics of dyes by the composite biosorbent. The parameters of each models were evaluated by non-linear regression using SigmaPlot 12.0 software. The suitability of models was tested by the determination coefficient ($R^2$) and standard error (SE). A high $R^2$ and low SE value point out the best model.

### 2.5. Biosorption isotherms

Equilibrium experiments were conducted using biosorbent masses of 10 mg in Erlenmeyer flasks containing 100 mL of Br46–Bv3 mixtures for a period of time equal to the equilibrium at optimum pH value. The mixtures were periodically shaken at constant temperature and speed. After biosorption equilibrium, the samples were centrifuged and then the concentrations of residual dyes in the supernatant solutions were analyzed as described above. The equilibrium data obtained were evaluated by Freundlich, Langmuir, Sips and Dubinin–Radushkevich isotherm models. The characteristic constants of each isotherm model were estimated by non-linear regression using SigmaPlot 12.0 software. The suitability of models was also controlled by $R^2$ and SE. A value of high $R^2$ and low SE show the best equation.

### 2.6. Characterization works

Fourier transformed infrared spectroscopy (FTIR) was performed using PerkinElmer Spectrometer (Spectrum 400) to know about various bonds present in the biosorbent before and after biosorption. Scanning electron microscope (SEM) analysis was accomplished using Zeiss Evo Ls 10 to study the surface morphology of bare and dyes laden with biosorbent.

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