Application of Experimental Design Methodology for Adsorption of Brilliant Blue onto Amberlite XAD-4/*Agaricus campestris* as a New Biocomposite Adsorbent

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***Abstract*–This research presents a new biocomposite adsorbents using response surface methodology (RSM) to find the best conditions for highest adsorption of Brilliant Blue G250 (BBG) from aqueous solution by Amberlite XAD-4/*Agaricus campestris.* The most effective parameters are determined by Plackett–Burman design (PBD) with specific ranges initial dye concentration (5–150 mg.L^{-1}), temperature (20–50°C), contact time (5–100 min), pH (3–11), shaking speed (150–300 rpm), sample volume (5–75 mL), and adsorbent dosage (0.05–0.6 g). Then, in the second step, the optimum condition of effective factors is predicted using steepest ascent design. Finally, optimal medium conditions of effective parameters with central composite design are located. According to RSM, the best adsorbent amount, contact time, initial dye concentration, and sample volume for maximum removal% of BBG (96.72%) are 0.38 g, 60.78 min, 107.13 mg.L^{-1}, and 28.6 mL, respectively. The adsorption of brilliant blue is approved by scanning electron microscopy. Under optimum conditions, it is concluded that XAD-4/*A. campestris* biocomposite is a suitable adsorbent for removing BBG from aqueous solution.**

***Index Terms*—Adsorption; Agaricus campestris; brilliant blue; biocomposite material; optimization.**

I. Introduction

Water is essential to almost all life forms on the planet, and it is thought that life began in water. Although water covers more than 70% of the earth's surface, the majority of it is unfit for human consumption, and only a limited amount of potable water is available. The widespread use of chemicals for a variety of purposes in daily life, combined with increasing industrialization, has resulted in the unintentional contamination of our natural resources through the release of a variety of organic and inorganic pollutants into the water system (Mallampati, 2013).

Among the various pollutants found in industrial wastewaters, dye is regarded as a significant esthetic factor and is classified as a visible pollutant. Dyes are generally applied to color the products in the paper, food, textile, cosmetics, leather, and plastic industries. They are typically synthetic in origin and have complex aromatic molecular structures, making them hard to biodegrade and more stable (Aljebori and Alshirifi, 2012).

Many different types of adsorbents are used to remove dyes, but nowadays researchers are looking for the cheapest and most effective materials. Composite materials are one of the selected materials for adsorption because of the cost and high percent of removal dyes. They are made up of two materials that have distinct physical and chemical properties (Elhajjar, Valeria and Anastasia, 2013; Fazeli, Florez and Simão, 2019).

To remove dyes from wastewater, a variety of treatment methods have been used, including physical, biological, and chemical methods. One of the most efficient and cost-effective process is adsorption for dye removal from textile wastewater among physicochemical methods. In the literature, a number of natural adsorbents for dye removal have been reported (Beyene, 2014).

Empirical or statistical methods could be used to optimize the process. The empirical method takes time and does not always result in a successful optimization. Response surface methodology (RSM) is a statistics-based procedure and a useful tool for studying the interactions between two or more independent parameters (Ahmad and Alrozi, 2010; Öztürk and Şahan, 2015; Yönten and Aktaş, 2014).

The present study was intended to discover a new low cost biocomposite adsorbent material (XAD-4+*Agaricus campestris*) and its impact on removing Brilliant Blue G250 (BBG) dye from aqueous solutions. In batch adsorption techniques, the effects of pH, temperature, contact time, shaking speed, adsorbent dose, sample volume, and dye concentration were investigated, and RSM evaluated the

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equilibrium effective conditions for these parameters (Ince et al., 2016, and Yonten et al., 2016).

II. Materials and Methods

A. Chemicals and Apparatus

Sigma-Aldrich provided the Amberlite XAD-4 resins (20–60 meshes) (LOT: BCCB0646, Germany). BB G-250 (Table 1) was purchased from Fisher BioReagents (LOT: 154716, United States). In addition, Merck supplied the rest of the chemicals used in the study (Germany). To adjust the pH of BBG solutions, Thermo Orion digital pH meter (Germany) was applied. All chromatographic measurements were taken with a UV spectrometer (WTW 6100 UV spectrometer, Germany). The experiments were done in chemical engineering laboratories, Faculty of Engineering, Van Yuzuncu Yil University, Turkey.

B. Fungus Preparation as an Adsorbent

The aboriginal fungus (*A. campestris*) was obtained from Van province in turkey to be used as a biosorbent. To remove contaminants, the fungus was washed with distilled water 2 times then dried at room temperature. A fine powder was made by grinding the dried fungus in a porcelain mortar. To certain that the dried cells of fungus had died completely, it was put in oven for 24 h at 70°C. The viability was tested by inoculation the cells into Sabouraud Dextrose Agar medium for 24 h at 27°C (Aksu et al., 2015).

C. Biocomposite Material Preparation by Immobilization

The following procedure was used to immobilize the fungus (*A. campestris*) on the substrate: 1 g of Amberlite XAD-4 was mixed with 0.1 g of fungus powder. The mixture was wetted several times with 2 mL of ultra-pure water to improve immobilization efficiency and ensure thorough mixing. Then, put on a hot plate magnetic stirrer, set the temperature at 50°C and the stirring speed at 150 rpm. Following these steps, the mixture was dried in a 50°C oven for 24 h, for improving immobilization efficiency. The obtained sample was milled to minimize the particle size to <120 micrometer and used as an adsorbent for BBG. The turbidity analysis indicates that all of the fungus (0.1 g) immobilized on XAD-4 (Yonten et al., 2016).

D. Characterization

Characterization of biocomposite materials is critical to determine the controlling mechanism of interaction betwixt the biocomposite material and BBG. Scanning electron microscopy-energy dispersive X-ray (SEM-EDX) was used to characterize the morphological structures of XAD-4/*A. campestris* before and after the biosorption process (Nicolet, Protege 460, Madison, WI, USA).

Table I
Properties of Brilliant Blue g-250.

No.	Properties	Description
1	Chemical formula	$C_{47}H_{48}N_3O_7S_2Na$
2	CAS Number	6104-58-1
3	Molar mass	854.02 g/mol
4	Storage Temperature	15–25°C
5	Synonyms	Brilliant Blue G, CBB G-250, Acid Blue 90

E. Adsorption Studies

A biocomposite material was used in this study to remove BBG from aqueous solution. Batch adsorption was performed under various conditions. The adsorbent was treated with brilliant blue solution concentrations ranging from 5 to 150 mg L^{-1} in sample volumes ranging from 15 to 75 mL. A digital pH meter (Jenco 6173) was used to adjust the pH from 3 to 11 using 0.1 M hydrochloric acid and sodium hydroxide. The dye solutions were stirred between 5 and 100 min at 150 and 300 rpm (WiseStir MSH-20D) and at temperatures ranging from 20 to 50°C. Before analysis, samples were centrifuged. The BBG in the supernatant was measured using a UV spectrometer set to 600 nm. The blanks were processed without any adsorbent.

F. Evaluation Method of Chromatographic Experimentation

Before starting the experimental work the UV spectrophotometer calibrated regularly in some period. In addition, before experiments, a scan of dye solution was taken and compared with the literatures for checking the absorbance, when it is true, the analysis of supernatants was done. Furthermore, at the first of starting of researches, the dye solutions (known ppm) were scanned by the UV device to find maximum wavelength (nm) then compared to another UV device and literatures. Besides the continuing with calibration, the deviation of UV device reading also checked.

G. Experimental Design

Plackett–Burman design (PBD)

The outputs of PBD were used in RSM to optimize the efficient factors influencing BBG removal percentage by biocomposite material. PBD performed an initial screening of the most important independent parameters affecting BBG removal by naturally powdered fungus/XAD-4. This technique assessed the determination of the factors that significantly influenced the specific response. The method is founded on the first-order polynomial model, as shown in Equation (1).

$$Y = \beta_0 + \sum(\beta_i X_i) \quad (1)$$

Where, Y represents the response (removal of BBG), β0 is the models intercept, β_i symbolizes the linear coefficient, and X_i denotes the level of the independent factor (Plackett and Burman, 1946). These seven parameters (temperature, shaking speed, pH, dye concentration adsorbent amount, contact time, and sample volume) were all investigated to determine the key factors influencing BBG removal. Based on PBD, the each factors prepared in the levels (−1: Low level and +1: High level). The PBD design model with seven parameters and responses is shown in Table 2.

Table II
Plackett-Burman Design and Responses.

Runs	pH	Temperature (°C)	Adsorbent (g)	Shaking speed (rpm)	Concentration (mg L^{-1})	Contact time (min)	Sample volume (mL)	Y (%)
1	3	20	0.05	300	5	100	75	73.77
2	3	50	0.6	150	150	100	75	71.27
3	11	20	0.05	150	150	5	75	54.51
4	11	50	0.6	150	5	5	75	86.37
5	3	50	0.05	300	150	5	75	55.27
6	11	20	0.6	300	150	5	15	94.20
7	3	50	0.6	300	5	5	15	59.05
8	11	50	0.05	150	5	100	15	82.87
9	11	50	0.05	300	150	100	15	94.65
10	11	20	0.6	300	5	100	75	84.62
11	3	20	0.05	150	5	5	15	86.73
12	3	20	0.6	150	150	100	15	96.97

H. Steepest Ascent Design (SAD)

This design model used to determine the greatest increases in response. Finding points at the upper limit are more difficult in optimization. It depicts the relative amounts that should vary to maximize the yield of the direction and factors. SAD experiments were used to screen the impact of four effective parameters that were found by PBD to locate the optimum region in terms of response (Myers, Douglas and Christine, 2016). Table 3 represents the SAD experimental design and corresponding responses.

Table III
Steepest Ascent Design with Responses.

Runs	Initial concentration (mg L^{-1})	Sample volume (mL)	Time (min)	Adsorbent dosage (g)	Y (%)
0	77.5	45	52.5	0.325	86.38
0+Δ1	85.25	41.30	54.78	0.339	90.09
0+Δ2	93	37.61	57.06	0.353	91.58
0+Δ3	100.75	33.92	59.35	0.367	97.38
0+Δ4	108.5	30.23	61.63	0.381	98.03
0+Δ5	116.25	26.53	63.92	0.395	97.51

I. Central composite design (CCD)

RSM entails designing and testing experimental response surfaces through regression and optimization. The goal is to find the process's optimum operating conditions or a region that meets the operating requirements. This study used CCD with four factors at five levels. The total number of experiments was 30 = 2k + 2^k + 6, where k represents the number of factors. Thirty experiments were supplemented at the center points to assess pure error. CCD experimental design is shown in Table 4. In quadratic models, the response can be related to selected factors during the optimization process. Equation (2) represents a quadratic model.

$$Y = \beta_0 + \sum_{i=1}^{3}\beta_i X_i + \sum_{i=1}^{3}\beta_{ii} X_i^2 + \sum_{i<1}^{3}\sum_{i=1}^{3}\beta_{ij} X_i X_j \quad (2)$$

Where, Y is response, β_0 is constant factor, X_i (i = 1–3) are variables, β_i is the linear, $\square_{ii}$ is the quadratic, and β_{ij} (i and j = 1–3) are second-order interaction factors. Design-Expert 7.0 was used to refine all of the data of PBD, SAD, and CCD (Ghaedi et al., 2015; Ince et al., 2016; Yonten et al., 2016).

III. Results and Discussion

A. PBD

In statistical analysis, many scientists have recently carried out the impacts of the most important and independent factors (Öztürk and Şahan, 2015; Palanivelan, Ayyasamy and Ramya, 2019; Yönten and Aktaş, 2014). The minimum

Table IV
Parameters, Intervals and Results Perform in ccd.

Runs	Initial concentration (mg L^{-1})	Sample volume (mL)	Time (min)	Adsorbent dosage (g)	Y	
					Actual removal	Predicted %
1	116	26	59	0.39	97.07	95.28
2	100	26	63	0.36	99.55	97.73
3	108	29.5	61	0.375	96.00	96.00
4	100	33	59	0.39	96.44	95.80
5	108	29.5	61	0.345	97.37	96.39
6	116	26	59	0.36	95.16	97.02
7	108	29.5	61	0.375	96.00	96.00
8	124	29.5	61	0.375	96.34	95.02
9	108	29.5	61	0.375	96.00	96.00
10	108	29.5	61	0.375	96.00	96.00
11	92	29.5	61	0.375	99.78	98.74
12	108	29.5	61	0.375	96.00	96.00
13	108	36.5	61	0.375	95.60	92.88
14	100	33	59	0.36	93.91	96.01
15	116	33	63	0.36	94.20	95.08
16	100	33	63	0.36	93.27	95.04
17	100	26	59	0.39	97.50	99.04
18	116	26	63	0.36	97.94	98.55
19	108	29.5	57	0.375	97.04	95.10
20	100	26	59	0.36	99.08	99.09
21	100	26	63	0.39	98.27	99.26
22	116	33	59	0.36	94.20	93.18
23	108	29.5	65	0.375	97.65	97.22
24	108	29.5	61	0.405	97.40	96.01
25	116	33	63	0.39	94.80	94.75
26	108	29.5	61	0.375	96.00	96.00
27	100	33	63	0.39	95.83	96.40
28	116	26	63	0.39	98.06	98.39
29	108	22.5	61	0.375	99.25	99.59
30	116	33	59	0.39	87.02	91.27

and maximum values for the variables are given in Table 2 and their effects on the removal of BBG. The most effective response factors were determined on the basis of the Pareto graph (Fig. 1) and the half-normal plot (Fig. 2) which were the adsorbent dose (C), the dye concentration (E), the contact time (F), and the volume sample (G).

B. Pareto Plots

For regression analysis, the effects of response parameters are important. The positive sign means increasing the response, whereas the negative sign reducing in response (Öztürk and Şahan, 2015; Yonten et al., 2016). There are positive effects of adsorbent quantity, pH and adsorbent quantity interaction, initial concentration and adsorbent quantity interaction, and negative impacts of pH and initial concentration interaction (Fig. 1). Pareto analysis clarified the results of the experiments as it will provide more important information on the percentage effect on the rate of adsorption (Bazrafshan et al., 2013; Haaland, 2020). Naturally, the weakness of bonds between dye molecules and adsorbing process can clarify this. The solution temperature effect is decreased and corrected by greater pH (Hassani et al., 2014).

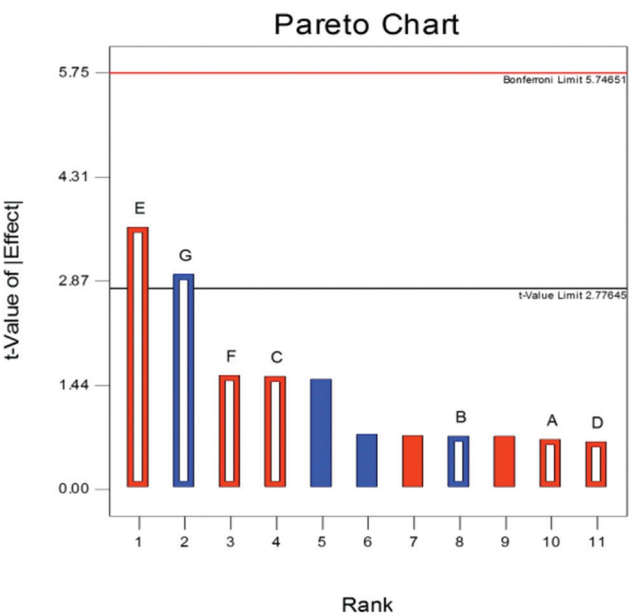

Fig. 1. Pareto graph.

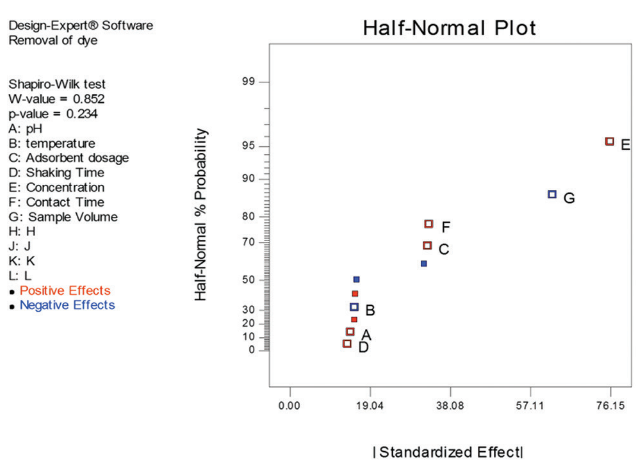

Fig. 2. Half-normal plots.

Figs. 1 and 2 were shown in the Pareto graphical analysis. Efficient parameters for removing dye by XAD-4/*A. campestris* are the linear effect of initial dye concentration (40.57%), sample volume (27.31%), adsorbent dosage (7.55%), and contact time (7.68%).

C. SAD

The SAD method was determined by increasing (initial concentration (C), contact time (F), and adsorbent dosage [C]) and decreasing sample volume to find the correct direction for changing variables (G). The SAD path and the results obtained are shown in Table 3. When dye concentration, adsorbent dose, sample volume, and contacting time were 108.5 mg L^{-1}, 0.381 g, 30.23 mL, and 61.63 min, respectively, the plateau of the response was obtained as a 98.03% removal of BBG. The optimal points in that region were discovered to be.

D. CCD

Table 4 illustrates the CCD and responses. The optimization by a linear model related to factors takes the response.

The figures below depict the parameters impact on BBG removal, as well as the interaction of four variables with the effect of the parameters. The linear model was used to organize the 3D response surface. In Fig. 3 which is three-dimensional response surface plot (TDRSP), some experiments were designed with sample volume (26–33 mL) and initial concentration (100–116 mg L^{-1}), fixed contact time amounts (61 min), and adsorbent amount (0.38 g) to explore the impact of dye concentration and sample volume on adsorption effectiveness. The removal of BBG has been strongly influenced by both variables in this figure.

The certain increase in initial concentration from 100 to 116 mg L^{-1} had no statistically significant effect on the

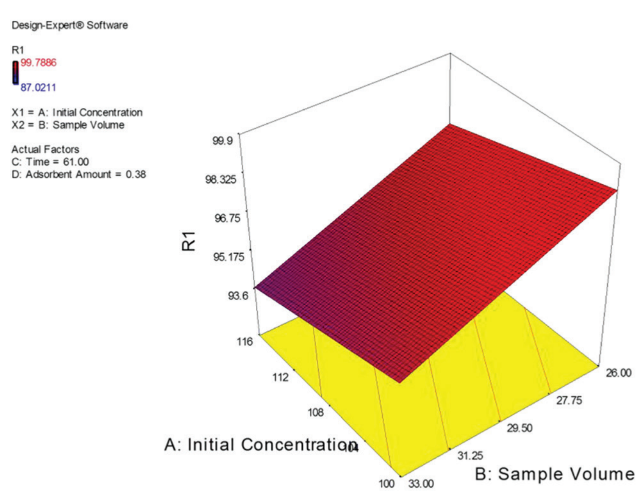

Fig. 3. TDRSP of the impact of the sample volume-dye concentration ratio on BBG removal percentage.

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percentage of BBG removal, which fell from 97.20% to 95.36% (Fig. 4). The optimum initial concentration was found to be 107.13 mg L^{-1}. The impact of sample volume is similar to the initial dye concentration. As shown in Fig. 5, any significant increase in sample volume from 26 to 33 mL reduced the percentage of BBG removal from 97.97% to 94.61%. The optimum volume for BBG removal was discovered to be 28.6 mL.

The same pattern has been achieved in some works. Adsorbent adsorption capacity was enhanced as the dye concentration increased. This might be due to a high mass transfer force. However, because the binding sites on the adsorbent are saturated, the percent removal decreases (Amin, Alazba and Shafiq, 2015). It can also be connected to the fact that the initial dye concentration had a limited impact on dye removal ability; at the same time, the adsorbent media had a specified range of active sites, which would have become saturated at a specific concentration. This has resulted in an increase in the number of dye molecules competing for available function groups on the adsorbent material's surface (Abbas, 2013). The removal percentage decreased as the initial dye concentration increased because the lower concentration solution contained fewer dye molecules than the higher concentration solution.

Furthermore, as the dye concentration increased, so also do the adsorbent's adsorption capacity. This influence could be caused by rising in the driving force of the ionic gradient with increasing dye concentration (Soni et al., 2012), which is justified by an increment in sorption capacity due to an increase in the amount of dye anion-adsorbent interactions. Santhi et al. (2010) discovered a similar phenomenon when they used agricultural waste (*Annona squamosa* seed) as an adsorbent to adsorb methyl red dye.

The effect of sample volume and contacting time on BBG removal% indicated by three dimensional plot as shown in Fig. 6. BBG removal percent tilted simultaneously with an increase in sample volume from 26 mL to 33 mL and contact time achieved a maximum of approximately 98.4% BBG removal at sample volume of approximately 33 mL and contact time of approximately 59 min and a slight increase at higher contact time values, as shown in Fig. 7.

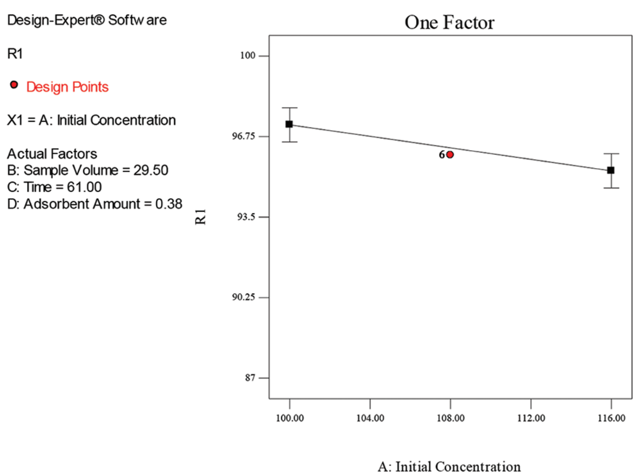

Fig. 4. Effect of initial concentration on removal % at mean value of other factors.

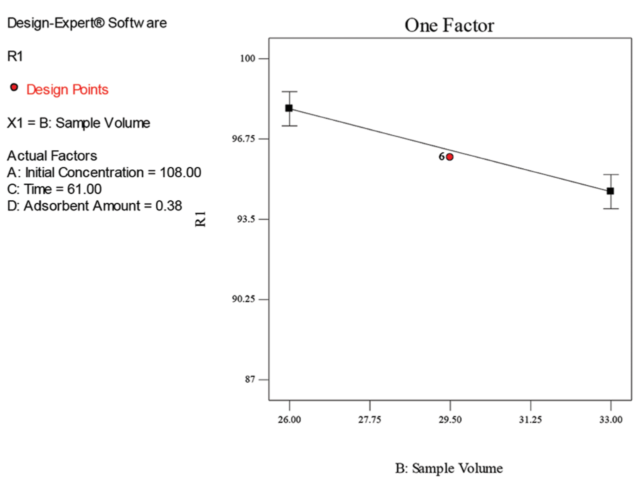

Fig. 5. Effect of sample volume on removal% at mean value of other factors.

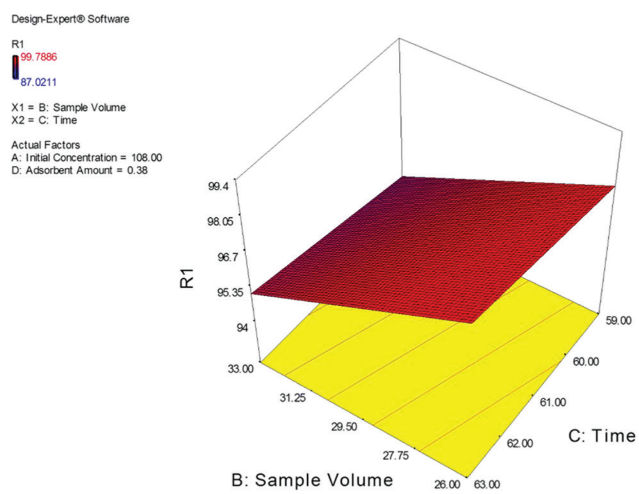

Fig. 6. TDRSP of the impact of sample volume-contact time ratio on removal% of BBG.

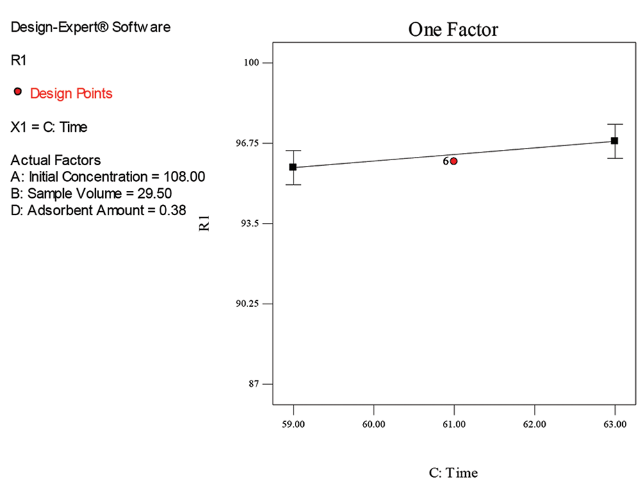

Fig. 7. Effect of contact time on removal% at mean value of other factors.

Moreover, enhancing the BBG concentration after reaching the saturated point, which can be attributed to a lack of vacant active sites, has little effect on the adsorption process. According to the Pareto graph results (Fig. 1), the removal process has a higher t-value of the initial concentration (E) than the other factors when compared to the initial BB concentration.

The interaction effect of contact time and adsorbent quantity ratio is given in Fig. 8. The removal percentage of BB increases as contact time increases from 59 to 63 min. The maximum contact time for the optimum yield was maintained at approximately 59.99 min (Daâssi et al., 2012; Şahan and Öztürk, 2014). The increase in removal percentage by adsorbent dose could be due to an increase in available surface area for sorption as well as the availability of additional adsorption sites. The increase in the removal of the adsorbent dose of methylene blue dye and linked it to an increase in surface area and sorption sites (Altaher and ElQada, 2011).

The results indicate that the rate of dye removal increased with contact time. For the higher dye concentration solution used, the longer balance time was required. Because of the large surface area of the adsorbent available for dye ion adsorption, the rate of dye removal is higher at first. Because the adsorbent surface has few active sites, only a poor increasing trend in color removal has been observed after a certain period (Soni et al., 2012).

The interaction of initial concentration and adsorbent quantity is depicted in Fig. 9. The removal percentage of BBG decreases as the initial concentration increases from 100 to 116 mg L^{-1} and the amount of adsorbent increases from 0.36 to 0.39 g. As shown in Figs. 9 and 10, the maximum percentage of BBG removal was 96.38%.

According to the graphs, the optimum adsorption of 96.72% occurs at a sample volume of 28.60 mL, adsorbent quantity of 0.38 g, initial dye concentration of 107.13 mg. L^{-1}, and contact time of 60.78 min in accordance with the model, as shown in Fig. 11.

E. Characterization of Biocomposites

EDX data (Table 5) and SEM micrographs (Fig. 12a-d) of pre- and post-sorption Amberlite XAD-4/*A. campestris* are presented. The results show that any of the elements, such as C, O, Na, Cl, Cu, and K, are present before adsorption in the biocomposite material with a percentage of weight (Table 5) of 81.40, 7.80, 3.37, 2.60, 3.72, and 1.11. After adsorption, all elements except K were present with a different percentage of weight (86.63, 7.71, 1.80, 0.62, and 3.23).

Fig. 12a and c shows Amberlite XAD-4 and fungal images at 100 and 300 µm before adsorption after bonding

Table V
Parameters of Amberlite XAD-4/*Agaricus campestris* Composite Material.

Before the adsorption			After the adsorption		
Chemical elements	Weight %	Atomic %	Chemical elements	Weight %	Atomic %
C	81.40	89.50	C	86.63	91.98
O	7.80	6.44	O	7.72	6.15
Na	3.37	1.94	Na	1.80	1.00
Cl	2.60	0.97	Cl	0.62	0.22
Cu	3.72	0.77	Cu	3.23	0.65
K	1.11	0.38	-	-	-

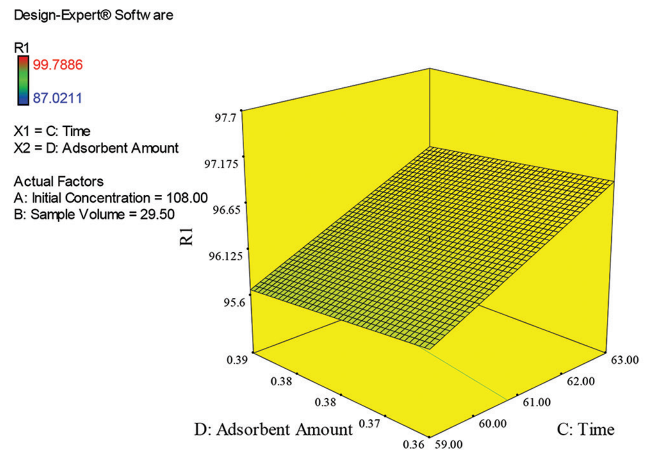

Fig. 8. TDRSP of the impact of adsorbent amount-contact time ratio on BBG removal %.

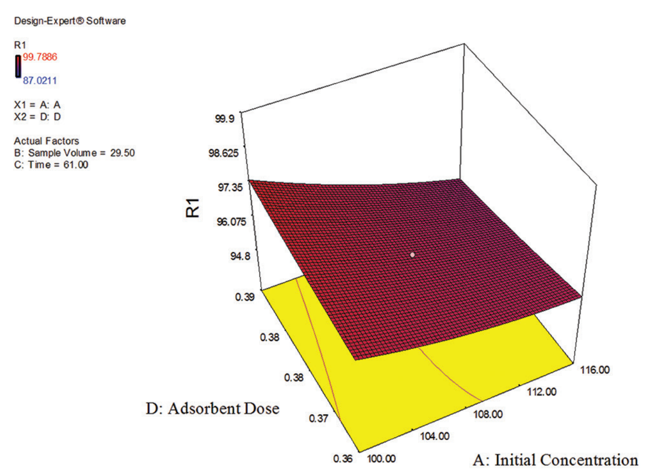

Fig. 9. TDRSP of the impact of initial concentration-adsorbent amount ratio on BBG removal %.

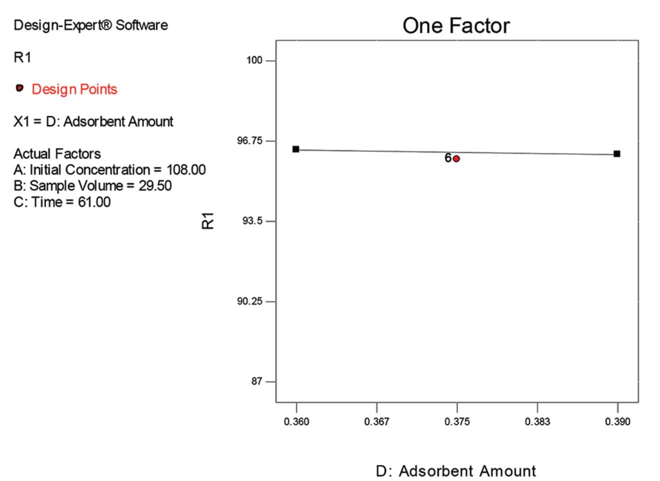

Fig. 10. Effect of adsorbent amount on removal% at mean value of other factors.

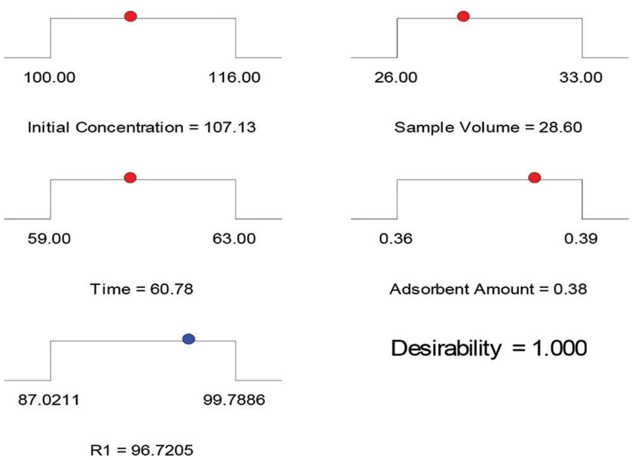

Fig. 11. Four variable parameters (adsorbent amount, contact time, initial concentration, and sample volume) numerical prediction graph.

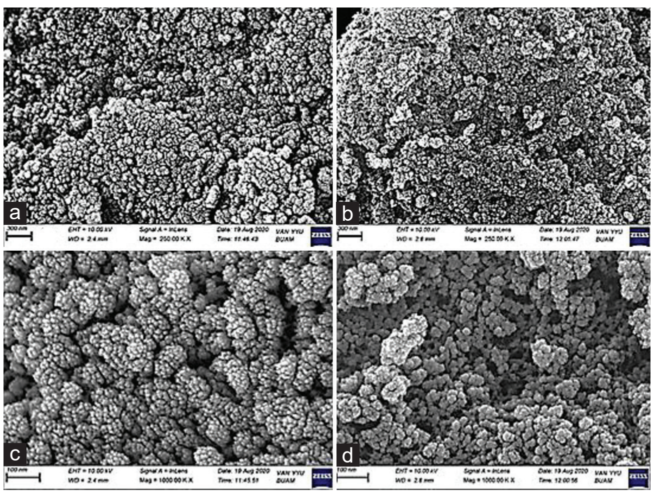

Fig. 12. SEM image at 100 and 300 µm (a and c) pre-adsorption and (b and d) post-adsorption.

with BBG, and these images are denser and BBG particles adhere to the surface more than the post-adsorption images (b and d). The simplicity observed before adsorption is visible in post-adsorption images, demonstrating the prevalence of adsorption (dye removal) in the present study.

IV. Conclusion

It is possible to conclude:

1. Because of its sediment-free operation, simplicity and flexibility of design, and complete deletion of dyes even in low concentration solutions, the adsorption process has been demonstrated to be highly effective for dye removal from polluted water. Adsorbent surface modification is very useful in enhancing adsorption capacity and adsorbent selectivity by taking advantage of specific interactions between adsorbents and target molecules in the adsorption technique sector to remove dye from wastewater streams.
2. Based on the findings, the biocomposite material (Amberlite XAD-4/*A. campestris*) is an effective adsorbent for the removal of Brilliant Blue G250 from aqueous media.

V. Recommendation

The following recommendations are made which can supplement the study performed in the future:

1. Using Amberlite XAD-4/*A. campestris* as a biocomposite adsorbent for the removal of other types of dyes, and it could be a viable alternative to low adsorption capacity materials.
2. RSM will be more economic and accurate way for finding the effective factors and optimizing the optimum point in adsorption process.

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