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Pre-treatment of toxic element and cationic dye onto natural biomass: characterization and optimization

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Abstract

This research describe wastewater pre-treatment that contaminated with Methylene Blue dye (MB) and Ni(II) ion by *Athelia Bombacina fungus dead biomass* (*ABFDB*). Researches finding on *ABFDB* characterization by SEM, XRD, CHNS and FT-IR analysis show that *ABFDB* can be used as efficient sorbent, because *ABFDB* cellular wall consist of Chitin, β-Glucan and Cellulose biopolymers, generally. Results show that removal of MB and Ni(II) ion by *ABFDB* sorbent is more than 86.41 and 66.2%, respectively. So, after parameters investigation of MB and Ni(II) ion sorption process by *ABFDB*, the response surface method was employed for optimization and study the interaction of operational parameters on the sorption of pollutants. This low-cost and natural environmental friendly biosorbent can be utilized for pretreatment process in the first step of wastewater treatment project.

1. Introduction

Nowadays, environment pollution is one the biggest global issues [1]. Increasing in the level of color reproduction and toxic elements application, much uses to produce them globally has been contaminated wastewater which in turn has caused concerns about the environment [2, 3]. Environmental pollution with the discharge of hazardous effluent containing toxic elements and organic dyes was increased water contamination [4]. Effluents from various industries contain harmful coloring agents, which must be removed to maintain environmental quality. Using dyes and toxic elements in various industries may cause harmful effects on living organisms in a short period of exposure [5]. One of the major global concerns is to save the planet and secure the future of mankind by saving natural water resources [6]. It is estimated that more than 100,000 commercially available dyes with high volume of dyestuff produced annually [7]. The negative effects of acute exposure to methylene blue dye (MB) may include increase heart rate, nausea, Heinz body formation, headache and gastritis [8].

Also, nickel is an essential trace element present in many species [9]. Nickel occurs naturally in the earth inner core, sea water and tea leaves. Nickel is an essential element to all the biotic life at lower concentrations. However, excessive levels of exposure make it quite carcinogenic in addition to various ailments related to heart and thyroid [10]. Direct contact of nickel may sometimes cause nickel dermatitis with symptoms of skin rashes. Nickel emissions from various industrial activities become air-borne affecting the public health [11].

Different techniques were used for removal of noxious dyes and toxic elements from industrial effluents that are based on the decolorization by activated sludge containing some additives such as powdered activated carbon, bentonite, clays [12], oxidation processes with ozone and hydrogen peroxide [13], photodecomposition catalyzed by titanium dioxide [14], microbiological decomposition using mixtures of

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aerobic and anaerobic microorganisms [15], electrochemical decomposition [16] and biosorption [17]. In particular, biosorption is the efficient method, because the hazardous species be transferred from the wastewater effluent to a solid phase and diminishing the effluents hazard. Also, the adsorbent can be regenerated afterwards or kept in a dry place without direct contact with the environment [18-20].

Many biosorbents and biomaterials were used for environmental application that can some of them cited in scientific reports such as bagasse pitch, carbonized bark, carbonized sewage sludge, tree-fern, wood sawdust, tree and eucalyptus bark, peat, biogas waste slurry, carbon slurry waste, diatomaceous earth, fly-ash, moss, orange peel, banana pitch, water hyacinth roots, peanut hull, microorganisms such as fungus, bacteria, algae, chitin, chitosan, rice husk and rice hull ash [21]. Therefore, the aim of present study is the preparing the *Athelia Bombacina* fungus dead biomass (*ABFDB*) and application of this biomaterial for adsorptive separation of Methylene Blue (MB) and Ni(II) ion pollutants from aqueous solution. Also, the mechanism of pollutants removal were modeled as kinetically, equilibrium and thermodynamically.

2. Materials and methods

2.1. Devices and chemicals

All Merck chemicals consist of $NiSO_4.6H_2O$ salt, Buffered Peptone water, sabouraud dextrose agar, HCl and NaOH with high purity were provided. MB cationic dye, were buy from Sigma-Aldrich Co. Also, double distilled water was also used in the experiment. Also, some of analytical techniques that have been used to study the biosorption process include atomic absorption spectrophotometry (AAS), UV-Vis spectrophotometry, scanning electron microscopy (SEM), Fourier-transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD) and CHNS analysis. These techniques may complement each other in giving insights into the mechanisms of biosorption process [22]. So, a pH meter, Temp.meter.86502 model was employed for measuring and adjusting of pH values in the aqueous phase. Solar-SS AAS was used for heavy metal ions determination. Unico-UV Visible/2100 spectrophotometer was used for dye determination. FT-IR spectra as KBr discs were recorded at 400-4000 cm^{-1} wavenumber range using a Spectrum One Fourier transform infrared spectrometer. LEO-1455VP SEM was used for consideration of morphology and particle size of biosorbent. Also, Vario EL model CHNS analyzer was used for determine the chemical composition.

2.2. *ABFDB* synthesis

Fungus and Oomycete have many and several differences that can study by Kotrba et al. (2011), but the main difference that is important and significant in line with this research is relevant to their cell walls [22]. Cell wall of fungus is chitin biopolymers [23]. According to Yan and Viraraghavan, (2001) used method which has been modified here, firstly *ABFDB* in the Sabouraud dextrose agar medium were cultivated and after seven days kept in the shaker incubator at 26 °C [24]. Thereupon, the spore suspension of the funguses with 1×10^5 spore/ml population were provide by Hemocytometer and added to liquid YPG medium. The YPG medium consist of 3 g/l Yeast, 10 g/l Peptone and 20 mg/l Glucose that was prepared as liquid, autoclaved and spore funguses added to it after cooling. Sabouraud dextrose agar medium consist of 40 g dextrose, 10 g/l peptone and 15 g agar that was added to 1 L distilled water and also, autoclaved for 20 min. After 24 hours and cooling be ready to use. Then sample in 23 °C on the shaker was kept at 125 rpm. After 4-6 days and inoculation of the fungus grown organs can be filtered using paper filter and was separated from other compounds. Then 50 mg of wet biomass with 0.5 M sodium hydroxide solution was treated for 30 min and then washed with distilled water to reach neutral pH (6.8-7.2). Finally, the synthesized *ABFDB* was autoclaved for 20 min. Obtained biomass, after autoclaving at a temperature of 60 °C for 24 hours in oven dried and then with planetary ball mill was powdered.

2.3. Biomass characterization

As it was introduced, various analytical techniques consist of SEM, FT-IR, XRD and CHNS elemental analysis were used to characterization of *ABFDB* biosorbent.

2.3.1. SEM and particle size analysis

Scanning electron microscopy SEM is an extremely useful method for the visual confirmation of the morphology and physical state of the surface. SEM was used to study the surface morphology, micro porosity and pores size of the *ABFDB* biosorbent. So, general structure of the *ABFDB* biosorbent were showed in Figure 1.

The results of the SEM analysis indicated that the particle size of biosorbent is less than 500 nm. The morphology of *ABFDB* is granular and has sharp edges structure.

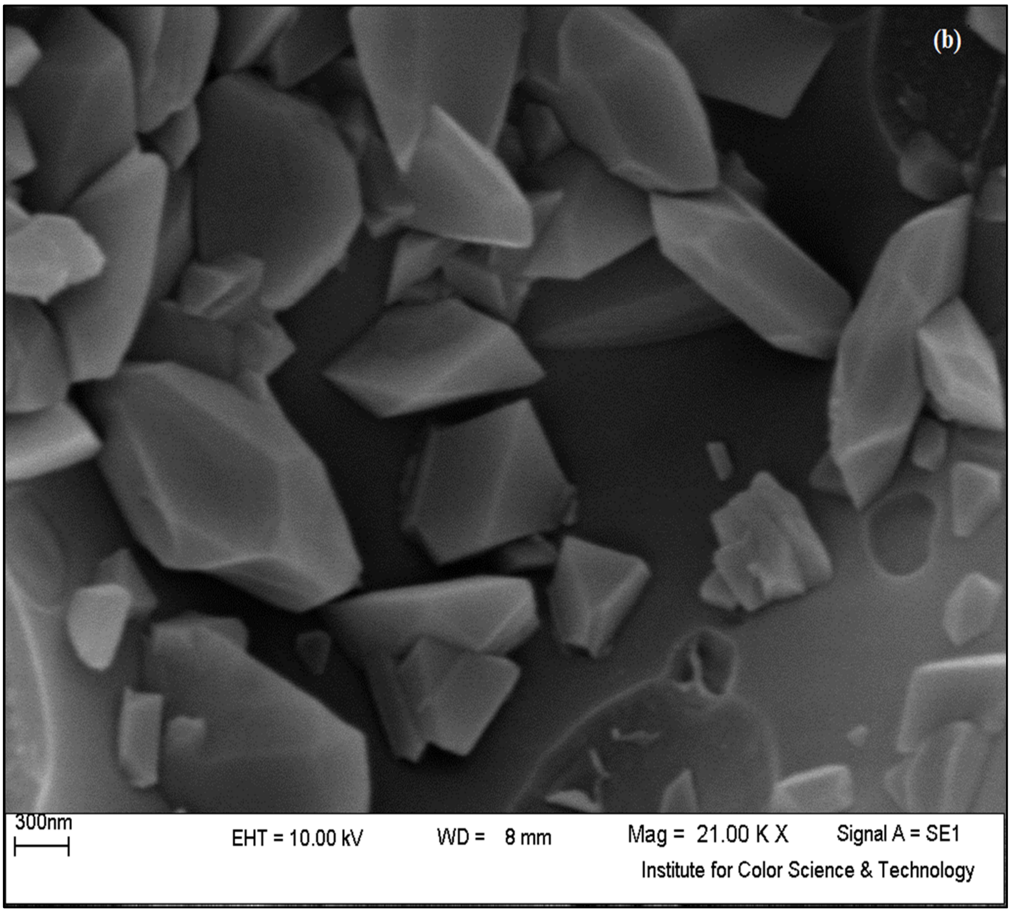


Figure 1. SEM image of ABFDB sorbent.

2.3.2. FT-IR spectroscopy

Infrared spectroscopy is a technique used to obtain an infrared spectrum of absorption or emission of a solid, liquid or gas. The FT-IR spectra of *ABFDB* sorbent are illustrated in Figure 2.

According to Socrates (2004) studies [25], the FT-IR spectra of *ABFDB* show eleven bands stretching vibrations between 400 and 4000 cm^{-1} (B_1-B_{11}). For *ABFDB* biosorbent FT-IR, the observed bands at 1029 and 1148 cm^{-1} (B_9-B_{11}) correspond to the organic phosphate groups and P– O bond. The band of 1239 cm^{-1} (B_8) related to COO^{-} anions. The bands of 1374 cm^{-1} (B_7) are assigned to the CH_3 group. The bands observed at 1548 cm^{-1} (B_6) correspond to the N-H bending vibrations. The bands observed in 162 and 1745 cm^{-1} (B_4 and B_5) related to C=O chemical bond. The bands observed at 2855 and 2922 cm^{-1} (B_2 and B_3) related to alkyl C-H group stretch and the bands observed at 3419 cm^{-1} (B_1) correspond to the hydroxyl group.

2.3.3. XRD and CHNS analysis of *ABFDB* sorbent

The nature and chemical composition of the *ABFDB* sorbent was studied by XRD and CHNS analysis. The XRD pattern of *ABFDB sorbent* are presented in Figure 3.

X-ray diffraction is a tool in the investigation of the fine structure of matter and it was used for determination of crystallizing the structure. This analysis was applied to detect the crystallinity of the *ABFDB* that consist of chitin component and the XRD analysis show that the biosorbent has crystalline structure. According to Figure 3, the *ABFDB* biosorbent exhibit two characteristic crystalline peaks at $2\theta \approx 9.7^{o}$ and $2\theta \approx 19.4$-22^{o} that were corresponded with chitin biopolymer, which is fit with obtained results by El Knidri et al. (2016) [26].

The results of *ABFDB* sorbent CHNS analysis were showed 40.15% carbon, 3.11% hydrogen, 3.60% nitrogen, 2.95% sulfur. The loss of ignition for *ABFDB* sorbent is 50.19%.

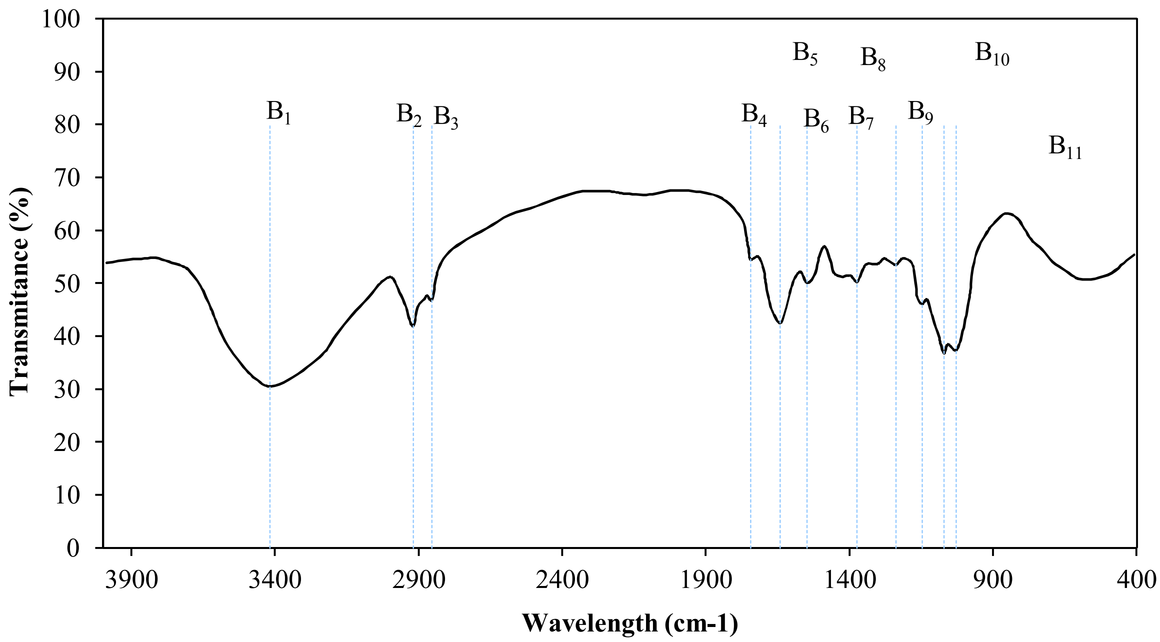


Figure 2. FT-IR spectra of ABFDB sorbent.

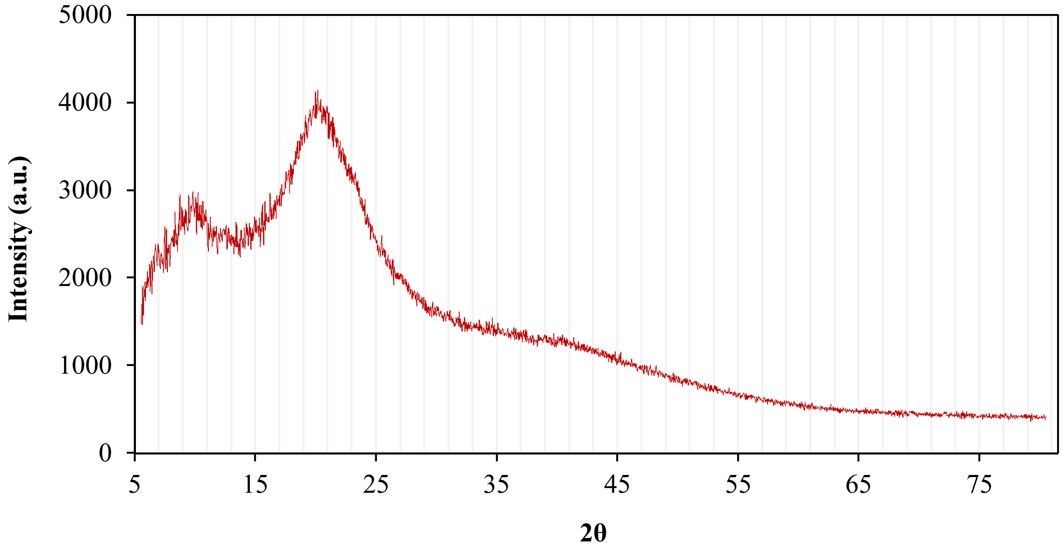


Figure 3. XRD patterns of ABFDB sorbent.

2.4. Pollutants biosorption

Stock synthetic solutions of MB and Ni(II) ion (1000 mg/l) were prepared by dissolving appropriate quantities of dye and metal salt powders in distilled water. The solutions of different concentrations were obtained by adequate dilution of the respective stock solutions.

The adsorption studies for evaluation of the *ABFDB* for removal of MB and Ni(II) ion from aqueous solutions was carried out using the batch adsorption procedure. For these experiments, the change of major parameters consist of pH, initial concentration, sorbent dosage, temperature and time was done, which set as Table 1.

Table 1. The values of major parameters that changed for studying the sorption process.

Pollutant	pH	Initial Concentration (mg/l)	Bio sorbent Dosage (mg)	Temperature (°C)	Time (min)	Volume (ml)
MB dye	2-11.5	2-25	1–7	10, 20 and 30	0–120	50
Ni(II) ion	3-6.5	2-40	5–20	12, 22 and 32	0 – 60	100

The efficiency and adsorption capacity of MB dye and Ni(II) ion pollutants on *ABFDB* biosorbent by using Equation 1 and 2 can be calculated, respectively:

% Pollutant Removal =

$$\frac{(C_0 - C_t)}{} \times 100 \quad (1)$$

$$q = \frac{(C_0 - C_t)}{m} \times V \quad (2)$$

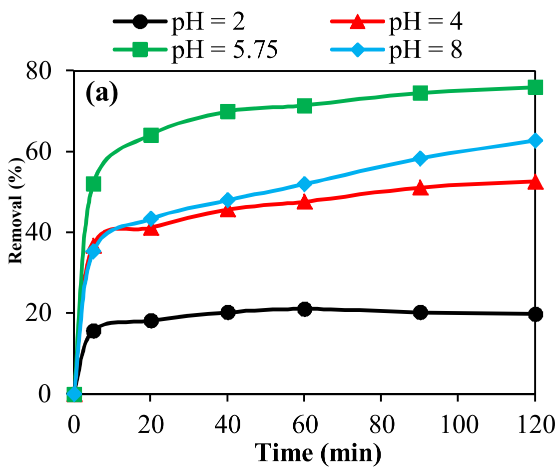
Where, C_0 and C_t are the initial concentration and concentration at time t (mg/l), respectively. V is the solution volume (l) and m is the weight of the bio sorbent (g).

3. Results and discussion

3.1. Effect of pH

The effect of the initial pH value of the sample solution on the adsorption of MB and Ni(II) onto the surface of *ABFDB* was investigated at different pH values ranging from 2.0 to 11.5 for MB and from 3.0 to 6.5 for Ni(II) ion, respectively. The initial concentrations of MB and Ni(II) were all set at 100 mg l^{-1}. The results are depicted in Figure 4.

It shows that the pH of the sample solution could significantly affect the extent of adsorption of the MB and Ni(II). Adsorption of MB onto *ABFDB* at low pH, the quantity adsorbed was 19.7%, later on it slowly increased with mounting pH up to pH 5.75 (75.98%). The cationic dyes give positively charged ions when dissolved in water. Thus, in acidic conditions (at lower pH), the positively charged surface of adsorbent tends to resist the adsorption of cationic dye molecules. It was found that the maximum adsorption by *ABFDB* at the optimal pH= 5.75 and 6.5, are 76 and 58.5%, respectively. At low pH values (3–5), there are no changes in their properties; their binding properties are modified only when the pH of the system is increased, due to the dissociation of hydrogen from the acid functional groups. H^+ ions would compete strongly with Ni(II) ions for the active sites since adsorbent would be protonated in strong acidic solutions.



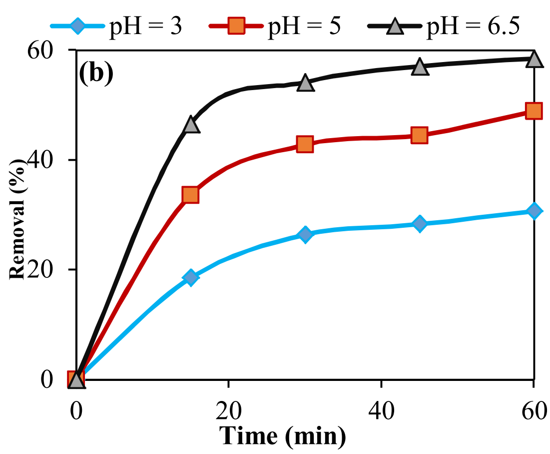


Figure 4. The effect of pH on the adsorption of MB (a) and Ni(II) (b) by ABFDB sorbent.

3.2. Effects of adsorbent dosage

Effects of *ABFDB* adsorbent dosage on removal efficiency were investigated at 10 mg/l concentration of MB and Ni(II) shown in Figure 5.

The study of adsorbent dosages for removal of the MB and Ni(II) from aqueous solution was carried-out using masses of *ABFDB* sorbent ranging from 1 to 20 mg, fixing the initial concentration at 10.0 mg/l and pH 5.75 and 6.5, respectively. The adsorption of MB and Ni(II) initially increased, as the adsorbent dose increased and the maximum adsorption efficiency for 78.35 and 64.8% was achieved. However, the efficiency increased by increasing the adsorbent dose for both pollutants. Increase in biosorption yield with increase in biosorbent dosage can be attributed to an increase in the biosorbent surface area and availability of more biosorption sites.

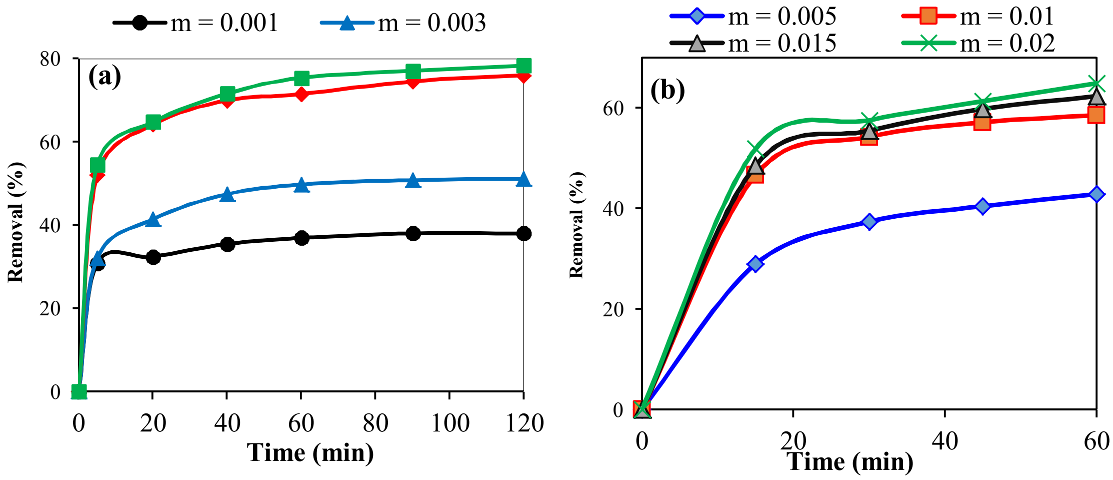


Figure 5. The effect of adsorbent dosage on the sorption of MB (a) and Ni(II) (b) by ABFDB sorbent.

3.3. Effect of initial concentration

The effect of the initial MB and Ni(II) concentration on adsorption was investigated and is shown in in Figure 6.

To evaluate the effect of MB and Ni(II) ions initial concentration on adsorption, the concentration range of 2–25 and 2–40 mg/l was studied, respectively. The amount of adsorbed MB and Ni(II) ions *ABFDB* adsorbent increased from 43.22 to 85.56% and 41.3 to 72.5% with an increase in the initial concentration and time 90 and 60min, respectively. At higher concentrations of MB, the removal decreased significantly due to the saturation of the adsorbent surface. general tendency is that high dye concentrations will cause a slower decolorization rate. The mechanism underlying this inhibiting effect may be higher dye concentration could inhibit the growth of adsorbent and production of ligninolytic enzymes. The decrease in percentage metal removal with increase in initial concentration was due to the exhaustion of the sorption sites available on the biomass, for a given biomass dosage.

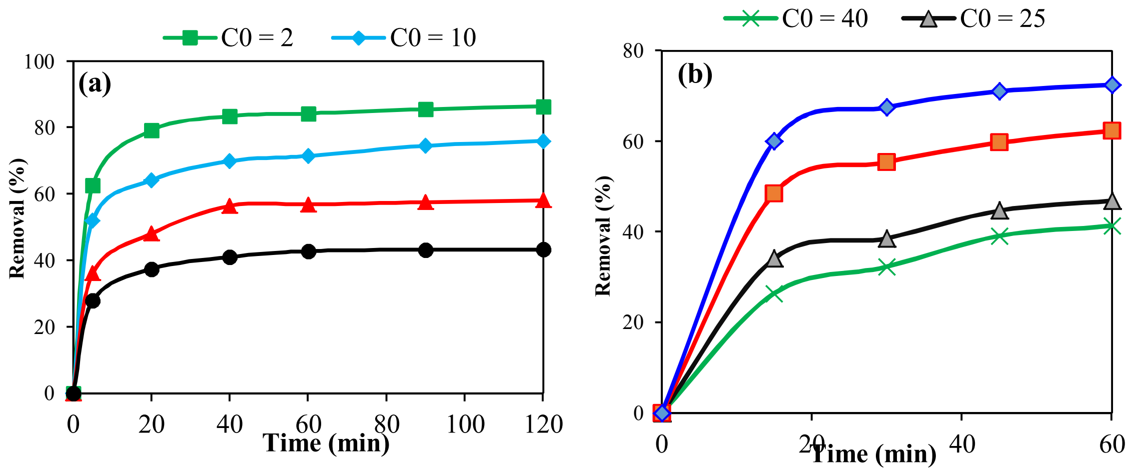


Figure 6. The effect of contaminant initial concentration on the sorption of MB (a) and Ni(II) (b) by ABFDB sorbent.

3.4. Effect of temperature

Temperature is a significant parameter affecting the adsorption capacity of sorbent and the transport/kinetic process of dye and metal adsorption. Figure 7 shows the effect of temperature for the adsorption of MB and Ni(II) onto *ABFDB* sorbent.

The effect of temperature on the adsorption of MB and Ni(II) was investigated at the temperature ranged of 10–30 and 12–32 ˚C at initial concentration of 10 mg/l. The increasing the temperature will change the equilibrium capacity of the adsorbent for a particular adsorbate. The low adsorption of MB on *ABFDB* (10 ˚C) might be due to the weakening of sportive forces between the active sites on adsorbent surface and adsorbed ions. The rate of diffusion of the MB molecules across the external boundary layer and in the internal pores of the adsorbent could be increased by increasing the temperature; the increase may have also stemmed from the negligible viscosity of the MB solution. Furthermore, the equilibrium capacity of the adsorbent for a particular adsorbate was changed by the increasing temperature. With the temperature increase, the mobility of the Ni(II) ions increases and the retarding forces acting on the diffusing ions decrease, thereby increasing the adsorption capacity of adsorbent.

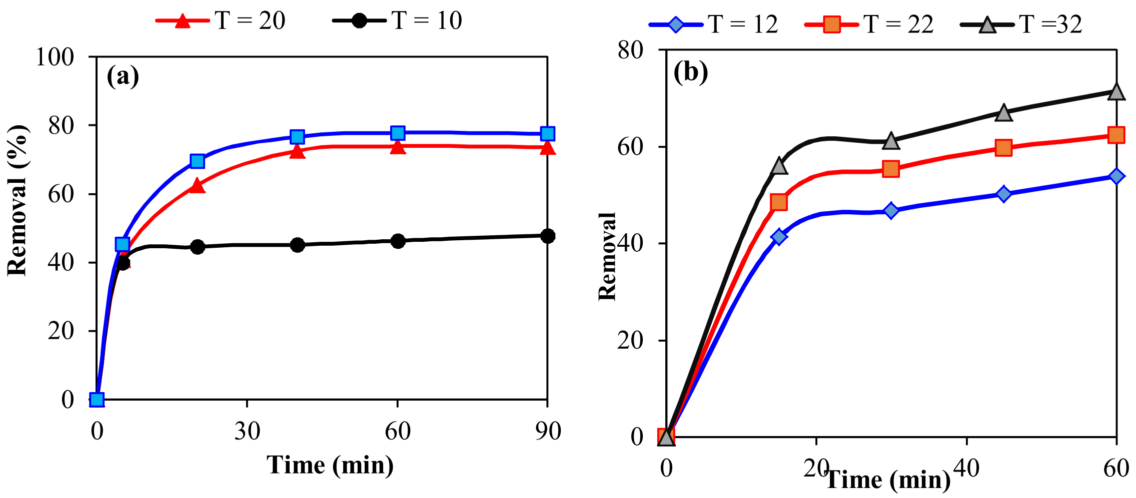


Figure 7. The effect of temperature on the sorption of MB (a) and Ni(II) (b) by ABFDB sorbent.

3.5. Modeling by response surface method (RSM)

RSM as one of the useful designs covers a class of mathematical procedures according to the study of the correlations and dependency among the independent factors and the obtained response in order to fit the empirical models to the investigational data obtained from experimental design [27]. RSM contains many types of design which two designs extensively utilized in chemical methods such as biosorption process [28]. Design Expert 11 software was employed to obtain the experimental design for determining the interaction between operational independent parameters consist of pH, time, temperature, sorbent dosage and initial concentration. The variation of operational parameters values are based on Table 1. The interaction of the five operational parameters which obtained by RSM illustrated in Figure 8.

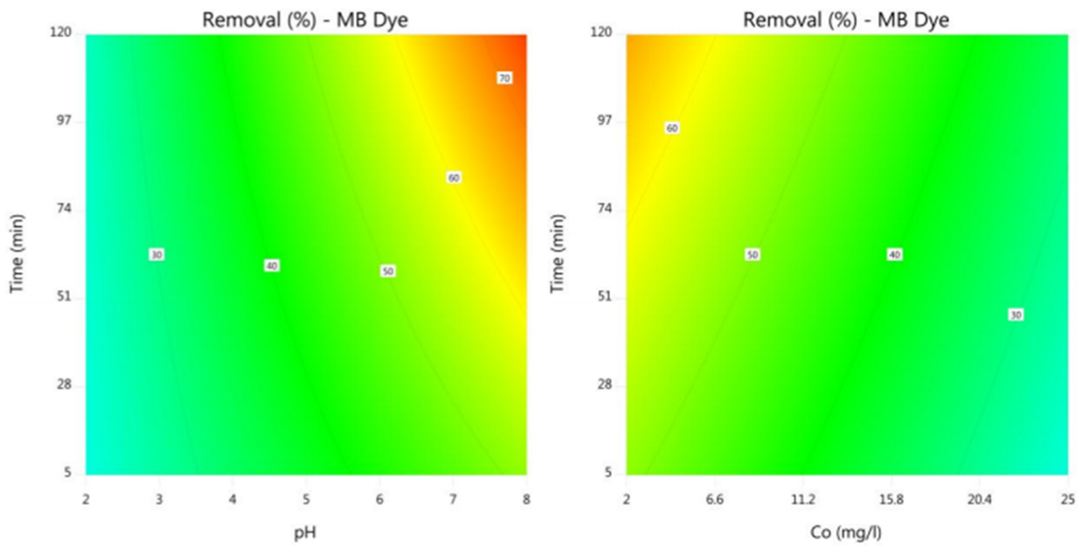


Figure 8. The interaction of operational parameters consist of pH, temperature, amount of adsorbent, time and initial concentration on the removal (%) of MB dye an Ni(II) ion.

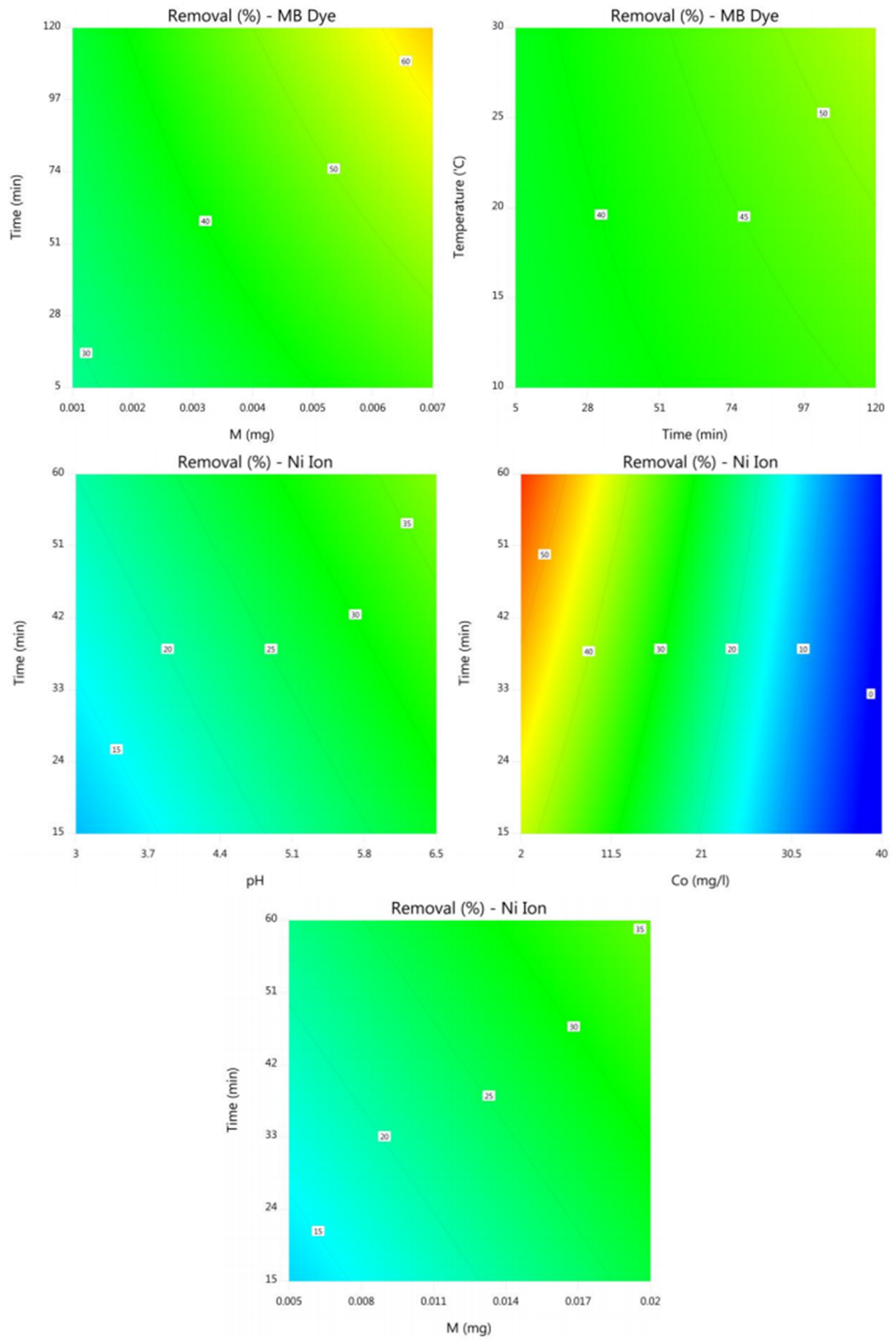


Figure 8. Continued.

According to the Figure 8, the obtained results of RSM modeling show that MB dye an Ni(II) ion sorption has direct relation with pH, temperature, amount of adsorbent and time parameters and has inverse relation with initial concentration parameter. Fot removal of MB dye an Ni(II) ion, the optimum conditions in pH 7.98 and 6.5, time 120 and 59.9 min, temperature 29.99 and 32 °C, initial concentration 24.99 and 30.34 mg/l and amount of adsorbent 6.9 and 1.99 mg/l were achieved by RSM which resulted in maximum sorption of 72.68 and 50.2%, experimentally which was near to the RSM predicted sorption of 76.25 and 48.78%, respectively.

4. Conclusions

Fungal biomass used successfully for the adsorption of Ni(II) and MB from aqueous media. The physicochemical characteristic of *Athelia Bombacina* fungus biomass confirmed that it is very interesting material containing numerous of functional groups in its structure that can easily adsorb positively charged dye and metal ions in the pre-treatment project. Fungus is an abundant and low-cost renewable resource with a wide variety of properties and applications depending on its source, treatment, functionalization, blending, and other factors. According to the obtained results, it was explicit that the maximum removal of MB by Athelia bombacina is 86.41%. The maximum adsorption of Ni(II) by Athelia Bombacina are 66.2%. Athelia Bombacina as effective and economic adsorbent for the pre-treatment of low concentration of MB and Ni(II) ions from water and wastewater.

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پیش‌تصفیه عناصر سمی و رنگزای کاتیونی با استفاده از بیومس طبیعی: شناسایی و بهینه‌سازی

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چکیده:

این پژوهش به بررسی فرآیند پیش‌تصفیه پساب آلوده به رنگزای کاتیونی متیلن آبی و یون نیکل با استفاده از بیومس مرده آتلیا بومبا سینا پرداخته است. یافته‌های پژوهش مربوط به شناسایی جاذب با استفاده از آنالیزهای تصویر میکروسکوپ الکترونی، پراش اشعه ایکس، سی اچ ان اس و تبدیل فوریه فرو سرخ نشان می‌دهد که جاذب متفاوت از سایر جاذب‌ها است، این تفاوت به تفاوت دیواره سلولی جاذب برمی‌گردد. دیواره سلولی جاذب از کیتین، گلوکان بتا و سلولز تشکیل شده است. نتایج فرآیند جذب نشان می‌دهد رنگزای متیل آبی و یون نیکل به ترتیب ۸۶/۴۱ و ۶۶/۲۰ درصد در فرآیند پیش‌تصفیه حذف شده‌اند. پس از بهینه‌سازی پارامترها برای جاذب مذکور و آلاینده‌های آلی و معدنی فوق، روش سطح پاسخ به منظور بهینه‌سازی و بررسی تأثیر متقابل پارامترها به کار گرفته شد. نتایج نشان می‌دهد که این جاذب طبیعی، در دسترس و ارزان قیمت می‌تواند به شکل کاملاً هدفمند در فرآیندهای پیش‌تصفیه پساب استفاده شود.

کلمات کلیدی: بیومس آتلیا بومبا سینا، جذب سطحی، رنگزای متیل آبی، یون نیکل.