Synthesis and characterization of modified activated carbon (MgO/AC) for methylene blue adsorption: optimization, equilibrium isotherm and kinetic studies

Vahab Ghalekhkhondabi, Alireza Fazlali and Keyhan Ketabi

ABSTRACT

Methylene blue (MB) is the cationic dye that is widely used for coloring cotton, wool, and silk. Since MB is harmful to human beings and toxic to microorganisms, there is the need to find cheap and efficient methods for removal of MB from wastewater prior to disposal into natural waters. In the present study, MB adsorption potential of MgO/AC prepared using a sol-gel-thermal deep-coating method was compared with the activated carbon (AC). The central composite design (CCD) as a method of the response surface methodology (RSM) was applied to minimize the number of runs and process optimization. The characterization of the microporous MgO/AC composite showed that the magnesium oxide nanoparticles were successfully coated on the AC and the BET specific surface area of AC and MgO/AC were 1,540 and 1,246 m²/g, respectively. The MB removal efficiency and the maximum adsorption capacity of AC and MgO/AC were 89.6, 97.5% and 571.7, 642.3 mg/g, respectively under optimum operational conditions of initial dye concentration of 100.9 mg/L, the adsorbent dosage of 69.4 mg/100 mL, pH of 10.2 and contact time of 149.1 min. According to an analysis of variance (ANOVA), the initial dye concentration and its interaction with the other effective factors have a large impact on adsorption efficiency. Furthermore, the mechanism of adsorption followed the Langmuir isotherm \( (R^2 = 0.9935, \Delta q_e = 2.9\%) \) and adsorption kinetics fitted by the pseudo-second-order model \( (R^2 = 0.9967, \Delta q_e = 6.6\%) \). Finally, our results suggest that the prepared MgO/AC is an efficient and promising material for dye wastewater treatment.

Key words | activated carbon, adsorption, correlation, magnesium oxide, methylene blue removal, optimization

HIGHLIGHTS

- MgO/AC composite \( (BET = 1,246 \text{ m}^2/\text{g}) \) prepared with the MB removal efficiency and the maximum adsorption capacity of 97.5% and 642 mg/g, respectively.
- The optimum operating conditions were determined with low error by the RSM.
- The CCD revealed that predicted data were in agreement with observed ones \( (R^2 = 0.9969) \).
- The data of this adsorption fits greatest to the isotherm model of Langmuir and pseudo-second-order kinetic model.
INTRODUCTION

Dyes and pigments are released into wastewaters from various industrial units, mainly from the dyestuffs, textile, paper, and plastics (Crini 2006). These dyes are primarily of synthetic origin and have complex aromatic structures, which make them more stable to light, heat and oxidizing agents, and are usually biologically non-degradable (Wang et al. 2013). Dyes even in low concentrations affect aquatic life and the food web. Some dyes are carcinogenic and mutagenic. Hence, ways and means are required to remove the dyes from wastewater (Namasivayam & Kavitha 2002).

The methods of color removal from industrial effluents include adsorption, biological treatment, coagulation, flotation, hyperfiltration, and oxidation (Iqbal & Ashiq 2007). Among the treatment options, adsorption has become one of the most effective and comparable low-cost methods for the decolorization of wastewater (Mall et al. 2005). Several non-conventional, low-cost adsorbents have been tested for dye removal. These include activated sludge, coir pith, minerals, neem leaf, peat, red mud, tree fern, and waste organic peel (Lorenc-Grabowska & Gryglewicz 2007). However, some raw agricultural by-products have many disadvantages, and one of them is the leaching of some organic pollutants, which may result in further pollution. This problem could be simply avoided by the carbonization of the raw materials or chemical modification. Activated carbons, prepared from agricultural materials, had good adsorption capacities for heavy metals, phenol, and dyes (Ioannidou & Zabaniotou 2007; Belaid et al. 2013).

As the most important cationic dye, methylene blue (C_{16}H_{18}ClN_{3}S, MB) is widely used in chemical indicators and biological dyes. A large amount of organic dye wastewater is produced in the processes of the printing and dyeing industries (Xavier et al. 2012; Kuang et al. 2020). It has various harmful effects on human beings, so it is of utmost importance to be removed from wastewater (Ahmad et al. 2012). This dye has been studied because of its known strong adsorption onto solids, and it often serves as a model compound for removing organic contaminants and colored bodies from aqueous solutions. Among the adsorbents, the ACs have the highest dye removal efficiency due to their large surface area and the functional groups on their surface that facilitate interactions with various dyes (Hameed et al. 2007b). Over the last decade, several studies have been shown that activated carbons obtained from various resources, including acrylic fibrous (Naeem et al. 2016), banana pith (Kadirvelu et al. 2003), coconut coir dust (de Souza Macedo et al. 2006), coir pith (Kavitha & Namasivayam 2007), eucalyptus bark and Cnetaeus oxyacantha core (Zazouli et al. 2016), novolac resin and olive stone biomass (Ioannou & Simitzis 2013), oil-palm kernel shells, sawdust and tannery leather scraps (Montoya-Suarez et al. 2016), rattan sawdust (Hamdaoui 2006), rice husk residue (Li et al. 2016), sepiolite (Alkan et al. 2008), tobacco stems (Mudyawabikwa et al. 2013) and walnut shells (Aygün et al. 2006) can be effectively used for the removal of MB dye from aqueous solution. Although the adsorption of dye removal from solutions was sufficiently studied by many authors, it was observed that these methods suffer from a major drawback of sludge formation and also adsorbent regeneration (Daniel & Shoba 2013). In order to improve the adsorbent’s ability for removal of refractory pollutants, metal oxides-coated materials have been studied because of their potential application as effective adsorbents (Zhang et al. 2014a).

Metal oxides nanoparticles were synthesized and used for removal of color from the wastewater effluent. The nanoparticles tend to agglomerate and thus lower the color removal capacity. To prevent the aggregation irreversibly, the nanoparticles were coated on substrates such as activated carbon (Sakulchaicharoen et al. 2010). Iron and manganese oxide are widely used to coat zeolite (Zhao et al. 2010), diatomite (Al-Ghouti et al. 2005, 2007) or other materials for removal of dyes, but magnesium compounds...
are seldom used. Recently, to synthesise MgO/AC composites, some methods, including aerogel procedure (Vu et al. 2016) and evaporation-induced self-assembly (EISA) (Zhou et al. 2018), have been suggested. It has been reported that the magnesium oxide introduced to the activated carbon improves the adsorption capacity of CH4 (Othman et al. 2017), CO2 (Wan Isahak et al. 2013; Shahkarami et al. 2016; Heo & Park 2017), H2S (Siriwardane et al. 2017), heavy metals (Wang et al. 2005) and acid red B (Baowei 2011).

The objective of this work is to compare the potential of MgO/AC composite with the AC as an adsorbent for the removal of a model dye, MB from wastewater. The novelty of this research is the impregnation of magnesium oxide nanoparticle to activated carbon using a sol-gel-thermal deep-coating method to synthesize the adsorbent with high surface area and pore volume to improve adsorption capacity and dye removal efficiency. The reason for choosing nano magnesium oxide is that it has a higher affinity for cations compared with Al, Fe, or Mn oxides (Daniel & Ramsundram 2019). The characterization of the MgO/AC composite was determined by Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and Brunauer–Emmett–Teller (BET) method. The influence of operation variables including initial dye concentration, adsorbent dosage, pH, and contact time was optimized on the MB removal efficiency by response surface methodology (RSM) technique. A further aim is to describe equilibrium data using adsorption isotherms and kinetic models.

**MATERIALS AND METHODS**

**Material**

Walnut shell-based AC was purchased from Part Chemical Company (Tuyserkan, Hamedan, Iran). The physicochemical characteristics of the adsorbent are given in Table 1. MB stock solution used in the study was prepared from MB (C16H18ClN3S) with a concentration of 1,000 mg/L purchased from Merck KGaA (Darmstadt, Germany). Working solutions and standard solutions were prepared by diluting from stock solution using distilled-deionized water (18.2 μΩ). Mg(NO₃)₂·6H₂O supplied from Merck KGaA (Darmstadt, Germany). 0.01 M HCl and 0.05 M NaOH solutions were used to adjust the pH of MB solutions.

**Preparation of modified activated carbon (MgO/AC)**

The sol-gel-thermal deep-coating method (Moussavi et al. 2013b, 2014; Rezaei et al. 2016; Rashidi et al. 2019) was used to prepare the MgO/AC composite. Magnesium oxide was loaded on AC as the support media using magnesium nitrate hexahydrate as the precursor. In this procedure, 5 g of Mg(NO₃)₂·6H₂O was dissolved in 50 mL deionized water and 3 mL of NaOH (1 N) was then added under constant stirring (50 rpm). The mixture was stirred for 10 min to form a homogeneous gelatinous suspension of Mg(OH)₂, then 5 g of AC (Mg/AC: 10 wt.%) was added and stirred gently for about 1 h. The suspension was then allowed to settle for 1 h and supernatant of the suspension was then discharged. The AC covered with Mg(OH)₂ gel was dried in air at 100 °C for 24 h. To convert Mg(OH)₂ to MgO nanocrystals, it was calcinated in the air at 500 °C for 2 h.

**Table 1 | Characteristics of activated carbon (AC)**

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk density (g/mL)</td>
<td>0.5</td>
</tr>
<tr>
<td>BET (m²/g)</td>
<td>1,540</td>
</tr>
<tr>
<td>Iodine number (mg/g)</td>
<td>1,550</td>
</tr>
<tr>
<td>Moisture (%)</td>
<td>3.7</td>
</tr>
<tr>
<td>Volatile matter (%)</td>
<td>2.2</td>
</tr>
<tr>
<td>Ash (%)</td>
<td>5.7</td>
</tr>
<tr>
<td>Fixed carbon (%)</td>
<td>88.4</td>
</tr>
</tbody>
</table>

**Batch adsorption experiments**

In order to investigate the effect of initial dye concentration, adsorbent dosage, pH, and contact time on MB adsorption, the experiments were conducted in a set of 250 mL of Erlenmeyer flask containing AC and MB solution. The flasks were agitated in an isothermal water-bath shaker at 150 rpm and 25 ± 1 °C until the equilibrium is reached. The pH was adjusted by adding either a few drops of diluted hydrochloric acid or sodium hydroxide. The flasks were then removed from the shaker and the final concentration of dye in the solution was analyzed. The samples were filtered before analysis in order to minimize interference of the carbon fines with the analysis. Each experiment was performed at least in triplicate and the results were presented to be their averages. The percentage removal of MB was calculated using the following equation:

\[
\text{MB removal efficiency} (\%) = \left(1 - \frac{C_e}{C_i}\right) \times 100 \tag{1}
\]

where \(C_i\) and \(C_e\) are initial and equilibrium concentration of MB (mg/L), respectively.
Regeneration of saturated AC

The spent activated carbon which had inappreciable adsorption capacity was filtered, washed by distilled water three times, and then dried in an oven at 105 °C to remove the water content. Afterward, chemical–physical regeneration (Wang et al. 2013; Xin-hui et al. 2014; Ghasemzadeh et al. 2017; Cundari et al. 2019) was carried out by using 7 N HCl for 30 min immersion, then heating at 190 °C for 1 h. The regeneration efficiency can be obtained by the following equation:

\[
\text{Regeneration efficiency (\%)} = \frac{q_{e,0}}{q_{e,1}} \times 100
\]

(2)

where \(q_{e,0}\) and \(q_{e,1}\) are the adsorption amount of MB before and after regeneration, respectively.

Analytical measurements

The synthesized adsorbents of AC and MgO/AC were characterized for surface functional groups, surface morphology, and specific surface area of the particles. The functional groups on the surface of AC and MgO/AC were analyzed by Fourier transmission infrared spectroscopy (FTIR, Alpha II, Bruker) using the KBr pellet technique at the wavenumbers ranging from 4,000 to 400 cm\(^{-1}\). The morphology of the particles was visualized using scanning electron microscopy (SEM, Philips–XL30). The specific surface area was determined using a nitrogen gas adsorption analyzer by the BET method (BELSORP, Mini II). The pH of the liquid samples was measured using a pH meter (model pH-03(1), Shanghai Yong Cheng Scale Co.). Samples of solutions before and after treatment were analyzed for the concentration of MB using a UV–vis spectrophotometer (Perkin-Elmer, Lambda 25 model) by finding out the absorbance at 668 nm.

Design of experiment (DoE)

Design-Expert software (version 12.0.3.0, Stat-Ease Inc., USA) was applied for design and optimization. The parameters (independent variables) used in this study were initial dye concentration (\(X_1\)), adsorbent dosage (\(X_2\)), pH (\(X_3\)), and contact time (\(X_4\)). Adsorption of MB (\(Y_1\)) was considered as the dependent factor (response). Temperature and agitation speed were fixed at 25 °C and 150 rpm respectively to reduce the number of variables and simplify the experimental design. The central composite design (CCD) was applied in this work for statistical calculations and the selected independent variables were converted into the dimensionless codified data. Table 2 shows independent variables and their levels for the CCD used in the present study. According to the statistical analysis by the software (DoE), the experiments were conducted in sixteen factorial, eight axial, and four central points.

### MATHEMATICAL MODELING

Adsorption equilibrium isotherm

Generally, the capacity of an adsorbent for a particular adsorbate involves the interaction of three properties: the concentration of the adsorbate in the fluid phase (\(C_e\)), the concentration of the adsorbate in the solid phase (\(q_e\)) and the temperature of the system (T). If T is kept constant, \(C_e\) and \(q_e\) can be graphed to represent the equilibrium. Such a plot gives an adsorption isotherm (Al-Qodah 2000; Ho et al. 2002; Okada et al. 2003).

Amount of MB adsorbed per gram of AC were calculated by the following equation (Sivrikaya et al. 2012):

\[
q_e = \frac{(C_i - C_e)V}{m}
\]

(3)

where \(q_e\) is the amount of MB adsorbed per gram of AC (mg/g), \(C_i\) is initial MB concentration (mg/L); \(C_e\) is the concentration of MB that remained unadsorbed in the solution (mg/L); \(V\) is the volume of MB solution (L); \(m\) is the amount of AC (g).

Three adsorption isotherm models of two parameters were used in this work, Langmuir, Freundlich, and Temkin isotherms. They differ in the basic assumptions, the shape of the isotherm, and the nature of the adsorbent surface (Liu et al. 2019). The theoretical model most appropriately that describes the experimental data of the

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Factor</th>
<th>Coded levels of variables</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Initial MB concentration (mg/L)</td>
<td>-1 100 400 700</td>
</tr>
<tr>
<td>B</td>
<td>AC dosage (mg/100 mL)</td>
<td>30 50 70</td>
</tr>
<tr>
<td>C</td>
<td>pH</td>
<td>3 7 11</td>
</tr>
<tr>
<td>D</td>
<td>Contact time (min)</td>
<td>30 90 150</td>
</tr>
</tbody>
</table>

Table 2 | independent variables and their levels for the CCD
MB–AC system was chosen from the adsorption capacity ($q_m$), correlation coefficient ($R^2$) values, and the normalized standard deviation ($\Delta q_e$) that was calculated by (Vargas et al. 2011):

$$\Delta q_e (%) = 100 \sqrt{\frac{\sum (q_{e,\exp} - q_{e,\cal})^2}{q_{e,\exp}}}$$  \hspace{1cm} (4)

where $N$ is the number of data points, $q_{e,\exp}$, and $q_{e,\cal}$ (mg/g) are the experimental and calculated equilibrium adsorption capacity value, respectively.

**Langmuir isotherm model**

Adsorption is treated as specific to a homogenous site, incules monolayer coverage without adsorbates on the surface of the adsorbent plane transmigrating due to uniform energy. The reversibility of the adsorption and presence of a certain number of vacant sites describes the nature of the model. The expression for Langmuir isotherm is given as follows (El Qada et al. 2006):

$$q_e = \frac{q_mK LC_e}{1 + K LC_e}$$  \hspace{1cm} (5)

where $q_e$ (mg/g) and $q_m$ (mg/g) are MB concentration at equilibrium onto biosorbent and when monolayer forms on biosorbent, respectively. $K_L$ is Langmuir constant (L/mg) related to the affinity of binding sites and the free energy of sorption. $C_e$ (mg/L) refers to the concentration of adsorbate at equilibrium.

The shape of this isotherm can also be expressed in terms of separation factor ($R_L$), which is given as follows (Foo & Hameed 2012):

$$R_L = \frac{1}{1 + K LC_i}$$  \hspace{1cm} (6)

For the Langmuir type adsorption process, the influence of the isotherm shape on whether adsorption is ‘favorable’ or ‘unfavorable’ can be classified by a dimensionless constant ‘$R_L$’. The separation factor ($R_L$), an important parameter of the Langmuir isotherm, can be used to verify if the adsorption in the system studied is unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L < 1$), or irreversible ($R_L = 0$).

**Freundlich isotherm model**

Freundlich sorption isotherm model views adsorption to be non-ideal, takes place in a heterogeneous multilayer surface with no limited degree of adsorption sites and energies exponentially or non-uniformly distributed. This is given by (Hameed et al. 2007b):

$$q_e = K_F C_e^{1/n}$$  \hspace{1cm} (7)

where $K_F$ (mg/g) (L/mg)$^{1/n}$ and $n$ are Freundlich constants, which are related to sorption capacity and the sorption intensity of the system. The magnitude of the term (1/n) indicates the favorability of the sorbent/adsorbate systems. The $n$ parameter, known as the heterogeneity factor, can be used to indicate whether the adsorption is linear ($n = 1$), whether it is a chemical process ($n < 1$), or whether a physical process is favorable ($n > 1$). On the other hand, the values of $1/n < 1$ and $1/n > 1$ indicate a normal Langmuir isotherm and cooperative adsorption, respectively (Malik 2003; El Nemr et al. 2009).

**Temkin isotherm model**

This model which is co-operatively applied under the condition of an intermediate mode of sorbate concentration considers the fall in the heat of sorption of molecules in the layer to decrease linearly with an area of coverage owing to the interactions that exist between the dye and activated carbon molecules in this case instead of logarithmic. It is given by (Kavitha & Namavisayam 2007):

$$q_e = \frac{RT}{b} \ln(K_T C_e)$$  \hspace{1cm} (8)

where $K_T$ (L/mg) and $b$ (J/mg) are Temkin constants. $R$ (8.314 J/mol. K) and $T$ are the universal gas constant and the absolute solution temperature in Kelvin, respectively.

**Adsorption kinetic study**

The procedures of kinetic experiments were identical to those of equilibrium tests. The aqueous samples were taken at preset time intervals, and the concentrations of dye were similarly measured. The amount of adsorption at...
time $t$, $q_t$ (mg/g), was calculated by:

$$q_t = \frac{(C_i - C_t)V}{m}$$  \(9\)

where $C_i$ and $C_t$ (mg/L) are the liquid-phase concentrations of MB at initial and any time $t$, respectively. $V$ is the volume of the solution (L), and $m$ is the mass of dry adsorbent used (g).

In order to understand the dynamics of adsorption in relation to time in the MB–AC system, pseudo-first order, pseudo-second order, and Avrami kinetic models were used. All models were fit employing the non-linear fitting method by MATLAB R2016a software. The advantage of using the non-linear form lies directly on the fact that we don’t need to know the equilibrium capacity ($q_e$) from experience since it can be determined from the model.

**Pseudo-first-order model**

The rate constant of adsorption is determined from the pseudo-first-order equation given by (Malash & El-Khaiary 2014):

$$q_t = q_e[1 - \exp\left(-k_1 t\right)]$$  \(10\)

where $q_e$ and $q_t$ are the amounts of MB adsorbed (mg/g) at equilibrium and at time $t$ (min), respectively, and $k_1$ the rate constant adsorption (1/min).

**Pseudo-second-order model**

Pseudo-second-order equation based on equilibrium adsorption is expressed as (Dogan et al. 2009):

$$q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t}$$  \(11\)

where $k_2$ (g/mg.min) is the rate constant of second-order adsorption.

**Avrami model**

The Avrami model is used to verify specific changes of kinetic parameters as functions of the temperature, initial concentration, and adsorption time. Avrami model is expressed mathematically by the following equation (Lopes et al. 2003):

$$q_t = q_e[1 - \exp\left(-k_{AV} t^n\right)]$$  \(12\)

where $k_{AV}$ (1/min) and $n$ are the Avrami kinetic constant and fractional adsorption order.

### RESULTS AND DISCUSSION

**Characterization of AC and MgO/AC**

The bulk density affects the rate of adsorption onto AC. In the present study, the bulk density was less than 1.2 (0.5 g/mL) (Table 1) indicating that the activated carbon materials are fine in nature and hence enhanced the adsorption of MB from aqueous solution (Moyo et al. 2015). Iodine number is usually used to roughly estimate the surface area of activated carbon at room temperature conditions. It is used as an indicator of the porosity and adsorbent capacity of the activated carbon. The higher iodine number of AC (1,350 mg/g) (Table 1) has been attributed to the presence of a large micropore structure and to the great probability of carbons to have a large surface area due to enlargement of their pore structure (Ekpete et al. 2017).

The functional groups on the surface of AC and MgO/AC were characterized using the FTIR technique. The spectra reveal several intense peaks indicating the presence of several main functional groups on the surface of the adsorbent. The FTIR spectra shown in Figure 1 indicates several main peaks at 3,441, 2,363, 1,630, 1,434, 1,063 and 600 cm$^{-1}$ on the spectra which correspond to hydroxyl
groups (Tang et al. 2012), carbonyl group (Lee et al. 2002), aromatic C = C stretching (Liu et al. 2009), carboxylic groups (Gaur et al. 2005), C-O stretching (Moussavi et al. 2013a) and aromatic C-H bending and carboxyl–carbonate structures (Mashayekh-Salehi et al. 2017), respectively. Although the broadband at 1,434 (Dhal et al. 2015) and 600 cm$^{-1}$ (Zhang et al. 2014b) can be attributed to MgO stretching vibrations. Referring to Figure 1, the increase in peak intensity is greater in MgO/AC than in AC, which indicates the increased functional groups (particularly carboxylic groups) on the surface of MgO/AC as compared with the AC. Therefore, the lower infrared (IR) transmittance percentages and the increased functional groups on the surface of AC can be related to the impregnation of the MgO and modification process (Li et al. 2011).

The surface morphology of AC and MgO/AC was characterized using SEM is illustrated in Figure 2. As shown in Figure 3, the surface of the AC (Figure 2(a)) is heterogeneous and grainy textured (Liu et al. 2012), while MgO/AC (Figure 2(b)) has a smoother surface with relatively small and uniform holes due to the consolidation of MgO on activated carbon (Biglari et al. 2018). A comparison of SEM micrographs of AC and MgO/AC composite depicts that the MgO nanoparticles are almost evenly dispersed on the surface of AC and the surface morphology of the AC was noticeably modified after effective coating procedure with MgO. These results follow the results of other researchers (Moussavi & Mahmoudi 2009; Mohandes et al. 2010), which is fully confirmed in the BET experiment.

The specific surface area and pore volume of AC and MgO/AC composite was determined from the N$_2$ adsorption/desorption isotherms. The BET measurements taken indicated that the pore surface and volume of AC were reduced after the stabilization of MgO on it. The specific
surface area and the total pore volume of AC were 1,540 m²/g and 0.6737 cm³/g, respectively, which are changed to 1,246 m²/g and 0.5441 cm³/g in MgO/AC, respectively. The decrease in BET and total pore volume of the AC upon loading with MgO can be attributed to the blockage of the small pores on AC by MgO (Chuang et al. 2003).

ANOVA analysis

According to the software, analysis of variance (ANOVA) can be used for the data analysis. The quality of the fit polynomial model was expressed by the coefficient of determination $R^2$. Furthermore, the statistically significant was checked by the student t-test in the same program. The model terms were also evaluated by the probability value. One factor and 3D surface plots were obtained based on the effects of three factors at three levels.

Equations (12) and (13) show two correlations for MB removal efficiency and adsorption capacity:

\[
\text{MB removal efficiency} \, (\%) = +50.80 - 21.34A + 6.43B + 2.51C + 8.23D - 2.59AB - 1.21AC - 5.49AD + 0.26BC + 0.44BD + 0.39CD - 0.64A^2 - 1.54B^2 - 0.69C^2 - 3.99D^2
\]  
\[
\text{Adsorption capacity} \, (\text{mg g}^{-1}) = +405.90 + 106.26A - 71.68B + 15.01C + 52.42D - 25.69AB + 5.63AC + 22.21AD - 4.04BC - 14.62BD + 2.59CD - 151.83A^2 + 34.12B^2 - 4.63C^2 - 31.03D^2
\]

where A, B, C, and D are initial MB concentration, AC dosage, pH, and contact time, respectively. According to the ANOVA analysis, MB removal efficiency increased with the adsorbent dosage, pH, and contact time while the initial dye concentration decreased it. A considerable effect of the interaction between the variables of AB, AC, and AD was observed.

The quadratic model statistical results for the response surface are summarized in Table 3. The model F-values of 302.72 implies the model is significant for percentage MB removal. There is only a 0.01% chance that model F-values occur due to noise. Values of probability $>$ F less than 0.0500 indicate model terms are significant. In this case A, B, C, D, AB, AC, AD, and $D^2$ are significant model terms. Table 3 indicates high reliability in the estimation of MB removal efficiency ($R^2 = 0.9969$). A high $R^2$ coefficient ensures a satisfactory adjustment of the quadratic model to the experimental data. In optimizing a response surface, an adequate fit of the model should be achieved to keep away from poor outcomes. The Predicted $R^2$ of 0.9857 is in reasonable agreement with the Adjusted $R^2$ of 0.9936. The adequate precision (AP) value is a measure of the signal to noise ratio and was found to be 65.755, which indicates an adequate signal. AP values higher than four are desirable and confirm that the predicted models can be used to navigate the space defined by the CCD.

Figure 3(a) shows a good agreement between the predicted data (for the responses) and the observed ones, the data points are distributed relatively close to the straight line. The models could navigate the design space. The residuals from the least-squares fit are important for judging the model adequacy. Figure 3(b) presents studentized residuals vs. normal percentage of probability as a straight line.

### Effects of initial dye (MB) concentration

The effect of initial MB concentration on MB removal efficiency was investigated in the concentration range of 100–700 mg/L and the results are given in Figure 4. Since the amount adsorbed is a function of concentration at equilibrium, increasing initial MB concentration caused an increasing amount of MB adsorbed, as expected, which suggests filling of available adsorption sites on the AC surface (Barka et al. 2011; Iqbal et al. 2011).

Figure 4(a)–4(d) shows that dye concentration had a negative effect on removal efficiency. This is due to the fact that with increased dye concentration, the driving force for mass transfer also increases. At low concentrations, there will be unoccupied active sites on the adsorbent surface. Above optimal MB concentration, the active sites required for the adsorption of dye will lack. This retards the overall MB adsorption by activated carbon. MB removal decreased from 74.9 to 25.7% when the initial concentration

<table>
<thead>
<tr>
<th>Variable</th>
<th>Value</th>
<th>Variable</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Standard deviation</td>
<td>1.60</td>
<td>$R^2$</td>
<td>0.9969</td>
</tr>
<tr>
<td>Mean</td>
<td>46.40</td>
<td>Adjusted $R^2$</td>
<td>0.9936</td>
</tr>
<tr>
<td>Coefficient of variance</td>
<td>3.45</td>
<td>Predicted $R^2$</td>
<td>0.9857</td>
</tr>
<tr>
<td>Predicted residuals error sum of squares</td>
<td>155.99</td>
<td>Adequate precision</td>
<td>65.755</td>
</tr>
<tr>
<td>Lack of Fit</td>
<td>3.33</td>
<td>Pure Error</td>
<td>0.000</td>
</tr>
<tr>
<td>F Value</td>
<td>302.72</td>
<td>$p$-value</td>
<td>&lt;0.0001</td>
</tr>
</tbody>
</table>

<Figure 3(a)> shows a good agreement between the predicted data (for the responses) and the observed ones, the data points are distributed relatively close to the straight line. The models could navigate the design space. The residuals from the least-squares fit are important for judging the model adequacy. Figure 3(b) presents studentized residuals vs. normal percentage of probability as a straight line.

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<Table 3> Analysis of variance for response surface quadratic model terms for MB removal

<table>
<thead>
<tr>
<th>Variable</th>
<th>Value</th>
<th>Variable</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Standard deviation</td>
<td>1.60</td>
<td>$R^2$</td>
<td>0.9969</td>
</tr>
<tr>
<td>Mean</td>
<td>46.40</td>
<td>Adjusted $R^2$</td>
<td>0.9936</td>
</tr>
<tr>
<td>Coefficient of variance</td>
<td>3.45</td>
<td>Predicted $R^2$</td>
<td>0.9857</td>
</tr>
<tr>
<td>Predicted residuals error sum of squares</td>
<td>155.99</td>
<td>Adequate precision</td>
<td>65.755</td>
</tr>
<tr>
<td>Lack of Fit</td>
<td>3.33</td>
<td>Pure Error</td>
<td>0.000</td>
</tr>
<tr>
<td>F Value</td>
<td>302.72</td>
<td>$p$-value</td>
<td>&lt;0.0001</td>
</tr>
</tbody>
</table>

<Figure 3(a)> shows a good agreement between the predicted data (for the responses) and the observed ones, the data points are distributed relatively close to the straight line. The models could navigate the design space. The residuals from the least-squares fit are important for judging the model adequacy. Figure 3(b) presents studentized residuals vs. normal percentage of probability as a straight line.

<Figure 4(a)>–<Figure 4(d)> shows that dye concentration had a negative effect on removal efficiency. This is due to the fact that with increased dye concentration, the driving force for mass transfer also increases. At low concentrations, there will be unoccupied active sites on the adsorbent surface. Above optimal MB concentration, the active sites required for the adsorption of dye will lack. This retards the overall MB adsorption by activated carbon. MB removal decreased from 74.9 to 25.7% when the initial concentration
Figure 4 | Three-dimensional surface of MB removal as a function of (a) Ci & m; (b) Ci & pH; (c) Ci & t; (d) m & pH; (e) m & t and (f) pH & t.
increased from 100 to 700 mg/L (m: 50 mg/100 mL, pH: 7 and t: 90 min). According to a report, by increasing the initial concentration from 500 to 700 mg/L, the dye removal percentage decreases from 85 to 60% (Pathania et al. 2017).

Effect of adsorbent (AC) dosage

The adsorbent dosage varied from 30 to 70 mg/100 mL. It is evident from Figure 4(a), 4(d) and 4(e) that the MB removal increased sharply with an increase in the adsorbent concentration from 30 to 50 mg/100 mL. This may be due to the availability of more adsorbent sites as well as greater availability of specific surfaces of the adsorbents. However, no significant changes in removal efficiency were observed beyond 70 mg/100 mL adsorbent dose. Due to the conglomeration of adsorbent particles, there is no increase in the effective surface area of AC (Benadjemia et al. 2011). MB removal increased from 41.6 to 57.2% when the adsorbent dosage increased from 30 to 70 mg/100 mL (C_i: 400 mg/L, pH: 7 and t: 90 min). According to a report, by increasing the adsorbent dosage from 30 to 70 mg/100 mL, the dye removal percentage increases from 65.3 to 80.7% (Hassan et al. 2014).

Effect of pH

The pH of the solution from which adsorption occurs may influence the extent of adsorption. pH affects adsorption in that it governs the degree of ionization of the acidic and basic compounds. In general, initial pH value may enhance or depress the uptake. This is attributed to the change of the charge of the adsorbent surface with the change in pH value. The effect of pH can be described based on the influence of pH on the zero points of charge, which is the point at which the net charge of the adsorbent is zero (Ghaedi et al. 2013; Gokce & Aktas 2014).

Results of the effect of initial pH on the adsorption of MB on AC are shown in Figure 4(b), 4(d) and 4(f). It is apparent that the amount of adsorbed MB increases with increasing pH from 3 to 7 and does not significantly increase at pH values higher than 7. The basic dye gives positively charged ions when dissolved in water. Thus, in the acidic medium, the positively charged surface of sorbent tends to oppose the adsorption of the cationic adsorbate. When the pH of dye solution is increased the surface acquires a negative charge, thereby resulting in increased adsorption of MB due to an increase in the electrostatic attraction between the positively charged dye and negatively charged adsorbent. MB removal increased from 47.4 to 53.1% when the pH increased from 3 to 11 (m: 50 mg/100 mL, C_i: 400 mg/L and t: 90 min). According to a report, by increasing the pH from 3 to 11, the dye removal percentage increases from 45 to 82% (Hassan et al. 2014).

Effect of contact time

The effect of contact time on the removal of MB is shown in Figure 4 ((c), (e), and f). Dye removal increased from 37.6 to 56.3% when contact time increased from 30 to 150 min (m: 50 mg/100 mL, C_i: 400 mg/L and pH: 7). The change in the rate of adsorption might be due to the fact that initially all the adsorbent sites are vacant and the solute concentration gradient is very high. Later, the lower adsorption rate is due to a decrease in the number of vacant sites of adsorbent and dye concentrations. The decreased absorption rate, particularly toward the end of experiments, indicates the possible monolayer formation of MB on the adsorbent surface. This may be attributed to the lack of available active sites required for further uptake after attaining the equilibrium (Abd El-Latif & Ibrahim 2009; Kadirova et al. 2014). According to a report, by increasing the contact time from 5 to 45 min, the dye removal percentage increases from 58 to 69% (Ghaedi et al. 2014).

Optimization and validation

The software can automatically find optimum conditions. It is necessary to validate these conditions by comparing the theoretical removal data with the experimental ones. Numerical optimization was used to determine the optimum parameters for maximum removal efficiency. The optimum conditions for MB removal were obtained based on the response surface and desirability function. In this case, all variables were targeted to be in range.

In order to confirm the accuracy of the predicted models and the reliability of the optimum conditions, an additional experiment was carried out. Table 4 illustrates the observed data (obtained from the experiment) under the optimum conditions compared with the predicted ones (Equations (13) and (14)). Low errors (0.89%) between the observed and predicted data validated both models. Furthermore, the adsorption efficiency of modified activated carbon (MgO/AC) at the optimum point was examined. It was found that it has a 9% increase in MB removal (97.5%) compared to unmodified AC (89.6%). Also, in optimum conditions and after three chemical–physical regeneration cycles (7 N HCl, 190 °C, 1.5 h), the regeneration efficiency
remains 96% indicating that the adsorbent has desirable regeneration ability.

However, both the AC and MgO/AC adsorbents were effective in the removal of MB, such that the adsorption capacity of the MgO/AC (642.3 mg/g) increased in comparison with the AC (571.7 mg/g) in adsorbing MB at optimum condition (Table 4). The greater adsorption capacity of MgO/AC than AC was related to the destructive sorption of MgO impregnated on the AC. In addition, the comparison of the maximum adsorption capacity of MB on various adsorbents in the literature such as Norit (400 mg/g) (Szlachta & Wójtowicz 2013), betel nut husk (381.6 mg/g) (Bardhan et al. 2020), coals (344.8 mg/g) (Purevsuren et al. 2015), lignocellulosic biomass (286 mg/g) (Namazi et al. 2010), AC-Degla Beida stones (163.6 mg/g) (Gherbia et al. 2019), coffee pulp (150 mg/g) (Gonçalves et al. 2013), AC-ZnO (149.7 mg/g) (Cruz et al. 2018), coconut leaves (149.3 mg/g) (Jawad et al. 2016), sewage sludge (125 mg/g) (Guo et al. 2017), iron impregnated AC (30.61 mg/g) (Shah et al. 2015), grinding palygorskite (111.7 mg/g) (Zhang et al. 2015), attapulgite/bentonite (168.6 mg/g) (Liu et al. 2014), pea shell (246.91 mg/g) (Geçgel et al. 2012), titanate nanotubes (133.3 mg/g) (Xiong et al. 2010), rattan sawdust (294.12 mg/g) (Hameed et al. 2007a), palm fiber (277.78 mg/1) (Tan et al. 2007), jute fiber (225.64 mg/g) (Senthilkumara et al. 2005), mango seed kernel (142.8 mg/g) (Kumar & Kumaran 2005), date pits (123.10 mg/g) (Banat et al. 2003), pistachio shells (129 mg/g) (Attia et al. 2003), and groundnut shell (164.9 mg/g) (Kannan & Sundaram 2001) shows that the adsorbent studied in this work has a very large adsorption capacity.

MB adsorption isotherm

The adsorption isotherm indicates how the adsorption molecules distribute between the liquid phase and the solid phase when the adsorption process reaches an equilibrium state. The analysis of the isotherm data by fitting them to different isotherm models is an important step to find a suitable model that can be used for design purposes. The adsorption isotherm is important to describe how solutes interact with adsorbents and are critical in optimizing the use of adsorbents (El-Geundi 1991, 1992). Langmuir, Freundlich, and Temkin isotherms are widely recognized and have been successfully applied to defining many adsorption equilibriums and evaluate adsorption equilibrium of metal ions and dyes from aqueous solutions. Therefore, MB adsorption data were analyzed by these equations.

The Langmuir isotherm assumes that adsorption occurs on a homogeneous surface containing sites with equal energy and that are equally available for adsorption. This is valid for the complete monolayer of adsorption, on which there is no transmigration of adsorbate on the surface plane, while the Freundlich isotherm is an empirical equation that can be used for heterogeneous systems with the interaction between the molecules adsorbed. Like the Freundlich isotherm, the Temkin isotherm considers the interactions between adsorbates assuming that the adsorption heat of all molecules decreases linearly when the layer is covered and that the adsorption has a maximum energy distribution of uniform bond (Vargas et al. 2011).

The values of the maximum adsorption amount (q_m), correlation coefficient (R^2), normalized standard deviation (Δq_m), and the other parameters for all the isotherms are shown in Table 5. The q_m value of 657.9 mg/g obtained for the Langmuir isotherm is close to the experimental value of q_m (642.3 mg/g), and the value of R^2 of 0.9935 shows the good fitting of this isotherm to the experimental data (Figure 5). In the concentration range studied (100–700 mg/L), the values of R_L between 0.039 and 0.357 indicate favorable adsorption (0 < R_L < 1) in the MB-MgO/AC system.

The values of the heterogeneity factor of n = 3.481 and 1/n = 0.287 in Table 5 indicate that the physical process and the normal Langmuir isotherm. This is in great agreement with the findings regarding R_L values. The fitting of the Freundlich isotherm to the experimental data (R^2 = 0.9683) is shown in Figure 5.

The constant of Temkin isotherm (b_T) is related to the heat of adsorption, and the positive values found (b_T = 45.8, 57.9, and 65.8) indicates an exothermic process (Table 5). The fit to experimental data (R^2 = 0.8711) in Figure 5 shows that the Temkin isotherm is less adequate

Table 4 | Optimum conditions were obtained from the software and experiment for MB removal and adsorption capacity

<table>
<thead>
<tr>
<th>Response</th>
<th>C_0 (mg/L)</th>
<th>m (mg/100 mL)</th>
<th>pH</th>
<th>t (min)</th>
<th>AC</th>
<th>MgO/AC</th>
<th>Predicted (AC)</th>
<th>Error (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MB (%)</td>
<td>100.9</td>
<td>69.4</td>
<td>10.2</td>
<td>149.1</td>
<td>89.6</td>
<td>97.5</td>
<td>90.4</td>
<td>0.89</td>
</tr>
<tr>
<td>q (mg/g)</td>
<td>530.1</td>
<td>31.2</td>
<td>10.9</td>
<td>138.1</td>
<td>571.7</td>
<td>642.3</td>
<td>590.2</td>
<td>3.23</td>
</tr>
</tbody>
</table>

### The Analysis of the isotherm data

The adsorption isotherm indicates how solutes interact with adsorbents and are critical in optimizing the use of adsorbents (El-Geundi 1991, 1992). Langmuir, Freundlich, and Temkin isotherms are widely recognized and have been successfully applied to defining many adsorption equilibriums and evaluate adsorption equilibrium of metal ions and dyes from aqueous solutions. Therefore, MB adsorption data were analyzed by these equations.
to explain the adsorption of MB onto MgO/AC, as compared to the Langmuir and Freundlich isotherms.

MB adsorption follows Langmuir isotherm since its normalized standard deviation is low ($\Delta q_e = 2.9\%–5.3\%$) and correlation coefficients calculated for Langmuir adsorption isotherm are close to unity ($R^2 = 0.9765–0.9935$) as well as larger than those calculated for Freundlich and Temkin isotherms and non-linear lines corresponding to Langmuir isotherm are much closer to experimental points (Hameed et al. 2007b; Karaçetin et al. 2014).

**MB adsorption kinetic**

The kinetic adsorption data can be processed to understand the dynamics of the adsorption reactions in terms of the order of the rate constant. Since the kinetics parameters provide important information for designing and modeling the adsorption process. The kinetic data were analyzed using three different kinetic models: pseudo-first-order, pseudo-second-order, and Avrami equation.

The pseudo-first-order model (Gürses et al. 2006) is the earliest known equation describing the adsorption rate based on the adsorption capacity. The pseudo-second-order model (Al-Ghouti et al. 2009) describes that the adsorption process is controlled by chemisorption which involves valency forces through sharing or exchange of electron between the solvent and the sorbate. The Avrami kinetic equation determines some kinetic parameters, as possible changes of the adsorption rates in the function of the initial concentration and the adsorption time, as well

### Table 5 | Isotherm parameters for MB adsorption by modified AC

<table>
<thead>
<tr>
<th>AC dosage</th>
<th>Langmuir</th>
<th>Freundlich</th>
<th>Temkin</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$q_m$</td>
<td>$K_L$</td>
<td>$R^2$</td>
</tr>
<tr>
<td></td>
<td>$K_F$</td>
<td>$n$</td>
<td>$R^2$</td>
</tr>
<tr>
<td></td>
<td>$K_T$</td>
<td>$b$</td>
<td>$R^2$</td>
</tr>
<tr>
<td>30</td>
<td>657.9</td>
<td>0.018</td>
<td>0.9935</td>
</tr>
<tr>
<td>50</td>
<td>502.5</td>
<td>0.022</td>
<td>0.9820</td>
</tr>
<tr>
<td>70</td>
<td>413.0</td>
<td>0.035</td>
<td>0.9765</td>
</tr>
<tr>
<td>107.7</td>
<td>3.481</td>
<td>0.9683</td>
<td>10.5</td>
</tr>
<tr>
<td>85.1</td>
<td>3.479</td>
<td>0.9380</td>
<td>18.9</td>
</tr>
<tr>
<td>90.9</td>
<td>3.941</td>
<td>0.9221</td>
<td>21.4</td>
</tr>
<tr>
<td>46.9</td>
<td>45.8</td>
<td>0.8711</td>
<td>30.2</td>
</tr>
<tr>
<td>38.3</td>
<td>57.9</td>
<td>0.8329</td>
<td>44.1</td>
</tr>
<tr>
<td>38.5</td>
<td>65.8</td>
<td>0.8476</td>
<td>43.4</td>
</tr>
</tbody>
</table>

**Figure 5 |** Fit of the isotherm models on the experimental data ($T = 25 \degree C \& \text{pH} = 7.0$) at AC dosage of (a) 30; (b) 50; (c) 70 mg/100 mL.
as the determination of fractional kinetic orders (Cestari et al. 2006). The conformity between the experimental data and the model-predicted values was expressed by correlation coefficients ($R^2$). Besides the $R^2$ values, the applicability of kinetics models was verified through the normalized standard deviation ($\Delta q_e$, %).

All kinetic parameters were calculated from fits of the three models (Figure 6), and they are shown in Table 6. According to Figure 6, the models represent initial stages where rapid adsorption occurs. The correlation coefficients for the pseudo-second-order model (Table 6) were greater than the other kinetic models and much closer to unity.

![Figure 6](image-url)  
**Figure 6** | Fit of kinetic models on the experimental data ($T = 25^\circ\text{C}$, $pH = 7.0$, and AC dosage of 30 mg/100 mL) at initial MB concentration of (a) 100, (b) 400, and (c) 700 mg/L.

<table>
<thead>
<tr>
<th>$C_i$ (mg/L)</th>
<th>$q_{e,\text{exp}}$ (mg/g)</th>
<th>Pseudo-first-order</th>
<th>Pseudo-second-order</th>
<th>Avrami</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>241.76</td>
<td>$q_e = 244.9$</td>
<td>$q_e = 304.0$</td>
<td>$q_e = 243.1$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$k_1 = 0.030$</td>
<td>$k_2 = 1.071 \times 10^{-4}$</td>
<td>$k_{AV} = 0.032$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$R^2 = 0.9780$</td>
<td>$R^2 = 0.9967$</td>
<td>$R^2 = 0.9801$</td>
</tr>
<tr>
<td>400</td>
<td>547.71</td>
<td>$q_e = 521.0$</td>
<td>$q_e = 588.8$</td>
<td>$q_e = 519.4$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$k_1 = 0.049$</td>
<td>$k_2 = 1.278 \times 10^{-4}$</td>
<td>$k_{AV} = 0.058$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$R^2 = 0.8846$</td>
<td>$R^2 = 0.9746$</td>
<td>$R^2 = 0.9139$</td>
</tr>
<tr>
<td>700</td>
<td>571.42</td>
<td>$q_e = 513.7$</td>
<td>$q_e = 604.8$</td>
<td>$q_e = 511.6$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$k_1 = 0.039$</td>
<td>$k_2 = 0.828 \times 10^{-4}$</td>
<td>$k_{AV} = 0.044$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$R^2 = 0.9039$</td>
<td>$R^2 = 0.9720$</td>
<td>$R^2 = 0.9255$</td>
</tr>
<tr>
<td>400–700</td>
<td>–</td>
<td>$\Delta q_e = 7.5%$</td>
<td>$\Delta q_e = 6.6%$</td>
<td>$\Delta q_e = 7.8%$</td>
</tr>
</tbody>
</table>
(R² = 0.9720–0.9967), which led to the belief that the pseudo-second-order kinetic model provided good correlation for the adsorption to different concentrations of MB onto MgO/AC. The Δqe values obtained in high concentrations (400–700 mg/L) for the pseudo-second-order model was 6.6%, which was the lower compared to the values of 7.5 and 7.8% obtained for the pseudo-first-order and Avrami kinetic models, respectively. Considering that the models presented high values of R², the applicability was based on the lowest Δqe values, so the pseudo-second-order model was the most suitable equation to describe the MB adsorption kinetics on modified AC (Hameed et al. 2007b; Karaçetin et al. 2014). Moreover, the experimental qe values agreed with the calculated values obtained from the no linear plots (Figure 6).

CONCLUSIONS

In this investigation, activated carbon was coated with magnesium oxide nanoparticles using the sol–gel-thermal deep-coating method and used as an efficient adsorbent for the removal of methylene blue. The removal of MB decreased with the increase of initial concentration, whereas increasing other parameters such as adsorbent dosage, pH, and contact time caused higher performance of adsorbent. Although impregnation of AC with MgO resulted in a 19% reduction in the BET specific surface area, the removal efficiency and the adsorption capacity of the AC composite improved by over 9% and 12% respectively for the removal of MB. Also, AC has a grainy and heterogeneous surface texture, while MgO/AC has a smoother surface with relatively small and uniform holes due to the consolidation of MgO on activated carbon. The statistical results indicated that the response surface quadratic model for our parameters was significant (p-value <0.0001). The isotherm evaluations revealed that the Langmuir model attained better fits to the experimental equilibrium data than the Freundlich model, which confirmed that the adsorption is heterogeneous and occurred through physicochemical interactions. Moreover, the rate of adsorption was found to obey the pseudo-second-order kinetic model with a good correlation coefficient, which indicated that the adsorption is done at a faster rate. It can be concluded that the MgO/AC composite absorbent prepared performed much better than AC and other previous works with the high removal efficiency and it can be a practical and promising option for enhancing the adsorption capacity for the treatment of wastewater containing dyes.

DECLARATION OF INTERESTS

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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