

Sorption kinetics of organic dyes methylene blue and malachite green on highly porous carbon material

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Abstract: In the article, the purpose of the research was to determine the important parameters of the organic compounds sorption – synthetic dyes methylene blue (MB) and malachite green (MG), on mesoporous carbon (MPC) from aqueous solutions in a limited volume. For the sorbent used, the elemental composition and the BET surface area were determined by nitrogen adsorption, which amounted to $2360 \text{ m}^2 \cdot \text{g}^{-1}$. Adsorption mechanisms were analyzed using kinetic dependences and sorption isotherms, for which empirical equations of pseudo-first and -second order, the Elovich equation and the intraparticle diffusion equation, as well as Langmuir, Freundlich, Dubinin-Radushkevich equations were used. In the course of kinetic studies, it was found that equilibrium occurs after 15 min of extraction with an adsorption capacity of $2446.6 \text{ mg} \cdot \text{g}^{-1}$ for MB and $2043.1 \text{ mg} \cdot \text{g}^{-1}$ for MG. It should be noted that the extraction of dye molecules proceeded predominantly by a mixed-diffusion mechanism and was controlled by a second-order reaction by a pseudo-second order model. The activation energy was $0.016 \text{ kJ} \cdot \text{mol}^{-1}$ for MG molecules, and $0.013 \text{ kJ} \cdot \text{mol}^{-1}$ for MB ones, which confirmed the physical mechanism of dye uptake. Thus, in the course of experimental studies, the high efficiency of the developed sorbent for the purification of aqueous systems from organic compounds was confirmed.

Keywords: carbon nanotubes; mesoporous carbon; synthetic dyes; methylene blue; malachite green; adsorption; kinetics; isotherms.

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Кинетика сорбции органических красителей метиленового синего и малахитового зеленого на высокопористом углеродном материале

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Аннотация: В статье целью исследований являлось определение важных параметров сорбции органических соединений – синтетических красителей метиленового синего (МС) и малахитового зеленого (МЗ), на мезопористом углероде (МПУ) из водных растворов в ограниченном объеме. Для используемого сорбента определены элементный состав и площадь поверхности по БЭТ по адсорбции азота, которая составила $2360 \text{ м}^2/\text{г}$. Проведен анализ механизмов поглощения с помощью кинетических зависимостей и изотерм сорбции, для чего применялись эмпирические уравнения псевдо-первого и псевдо-второго порядка, Еловича и внутривещичной диффузии, а также уравнения Ленгмюра, Фрейндлиха, Дубинина–Радускевича. В ходе кинетических исследований установлено, что равновесие наступает после 15 мин извлечения при величине адсорбционной

емкости 2446,6 мг/г по МС и 2043,1 мг/г по МЗ. Следует отметить, что извлечение молекул красителей проходит преимущественно по смешанно-диффузионному механизму и контролируется реакцией второго порядка согласно модели псевдо-второго порядка. Энергия активации имеет значение 0,016 кДж/моль по молекулам МЗ и 0,013 кДж/моль для МС, что подтверждает физический механизм поглощения красителей. Таким образом, в ходе экспериментальных исследований подтверждена высокая эффективность разработанного сорбента для очистки водных систем от соединений органической природы.

Ключевые слова: углеродные нанотрубки; мезопористый углерод; синтетические красители; метиленовый синий; малахитовый зеленый; адсорбция; кинетика; изотермы.

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1. Introduction

Water pollution is a serious environmental problem, which has reached an alarming level as a result of industrial development [1, 2]. Organic contaminants include dyes, which have a high solubility, thereby complicating the process of their removal from water [3]. In addition, due to the difficulty of biodegradation, water-soluble pigments are highly toxic and carcinogenic compounds, causing a serious environmental impact with serious problems for animals, plants and human health. On the other hand, textile, paper-printing, paint and varnish, leather, food and cosmetic factories are the largest consumers of dyes, since dye production is estimated at one million tons per year [4, 5]. Among these dyes, methylene blue (MB) as a cationic dye causes many health problems such as: allergic dermatitis, skin irritation, cancer, and mutations [6, 7]. Malachite green (MG) belongs to the same group of triphenylmethane dyes as crystal violet, which has been shown to be carcinogenic, so it can be assumed that MG also has these carcinogenic properties. Laboratory tests have also shown that MG can damage DNA after metabolic activation in vitro, although no genotoxicity has been demonstrated in in vivo tests [8].

Some technologies have been developed for industrial wastewater treatment, including adsorption, ion exchange, membrane filtration, chemical/electrochemical, chemical precipitation, ozonation, oxidation, photodegradation, and bioactive sludge [9–11].

Adsorption is an inexpensive method in which activated carbon is most often used as sorption materials due to its porous structure, a large number of surface functional groups, and ease of regeneration [12–15]. However, ordinary activated carbon mainly contains a large number of micropores with a pore size of less than 2 nm, which cannot allow large diameter dye molecules to access the inner surface of the carbon, resulting in low adsorption capacity, so its application is highly limited [16].

Many research teams are developing new sorbents based on nanomaterials – carbon nanotubes (CNTs), graphene oxide (GO), etc. They show very high efficiency in the removal of organic dyes from liquids. The paper [17] presents the results on the adsorption capacity of a porous material based on modified carbon, 2555 mg·g⁻¹, upon removal of dye MB molecules. In the article [18], the sorption capacity of a nanocomposite material based on graphene oxide modified with lignosulfonate was studied. As a result of the extraction of MB dye molecules, the authors of the article revealed the adsorption capacity of the adsorbent – 1822.3 mg·g⁻¹. The authors of [19] used a graphene aerogel to remove MB dye molecules. As a result of research, the authors of the work revealed the adsorption capacity of graphene aerogel equal to 420 mg·g⁻¹.

Mesoporous carbon (MPC) with pore sizes (2–50 nm) has a higher adsorption capacity with respect to some macromolecular substances, such as a dye, an antibiotic, some natural organic substances, etc. [20], since such bulky molecules of pollutants easily diffuse in volume and are adsorbed on the surface of pores. Mesoporous materials are promising sorption materials that are quite effectively used by scientific communities around the world for the adsorption removal of various contaminants, including MB molecules [21, 22].

The paper aims are to study the adsorption of organic contaminants – MB and MG dyes – on a mesoporous carbon material, as well as to reveal the absorption mechanism using kinetic and isothermal models.

2. Materials and Methods

2.1. Material synthesis

The procedure for the MPC synthesis was described in detail in [23]. The initial components for preparing MPC were aqueous solutions of dextrin, phenol-formaldehyde resin and multi-walled carbon nanotubes (CNTs) (NanoTechCenter Ltd., Tambov). The mixture was evenly stirred at 300 °C, then mixed

with potassium hydroxide and activated at 750 °C. The material was washed stepwise with an aqueous solution of hydrochloric acid and distilled water. The resulting aqueous paste was dried at 110 °C to constant weight.

2.2. Analytical methods

The MPC surface morphology was studied using a MERLIN scanning electron microscope (SEM) (Carl Zeiss, Jena, Germany) with Oxford Instruments X-ray microanalysis attachments. Surface area and porosity were measured by nitrogen adsorption using an Autosorb-1 analyzer (Quantachrome, Odelzhausen, Germany).

2.3. Kinetic study

To determine the kinetic parameters of MB and MG adsorption, experiments were carried out under static conditions; for this, 0.01 g of MPC was taken, the initial concentration of solutions of dyes MB and MG was 1500 mg·L⁻¹, the volume of the solution was 30 mL. The solutions were shaken for 5, 10, 15, 30 and 60 min at 100 rpm and room temperature on a Multi Bio RS-24 rotator (Biosan) and then filtered.

2.4. Isothermal study

Equilibrium studies were carried out using 0.01 g of MPC and 30 mL of solutions of MB and MG with initial concentrations of 150–1500 mg·L⁻¹. The tubes with solutions were shaken for 15 min at 100 rpm and room temperature on a programmable rotator Multi Bio RS-24 (Biosan). In all sorption experiments, the amount of dye in the liquid phase before and after adsorption was determined spectrophotometrically (Ekros, St. Petersburg, Russia) at wavelengths of 815 and 710 nm for MB and MG, respectively.

3. Results and Discussion

3.1. MPC characteristics

According to the obtained micrographs (Figs. 1a, b), mesoporous carbon is a composite, in the structure of which there are individual CNTs (10–20 nm in diameter) deformed as a result of alkaline activation, covered with a rather uniform layer of a certain shell.

According to Fig. 2, the sample under consideration is a carbon material with an amount of C = 94.09 wt. % with an admixture of oxygen O = 5.67 wt. %. The presence of oxygen is associated with the chemisorption of atmospheric oxygen on the surface of the obtained material due to the fact that the activation is carried out in a reducing medium containing metallic potassium.

The specific surface area of the material was determined by the BET method and its value was 2360 m²·g⁻¹ for nitrogen.

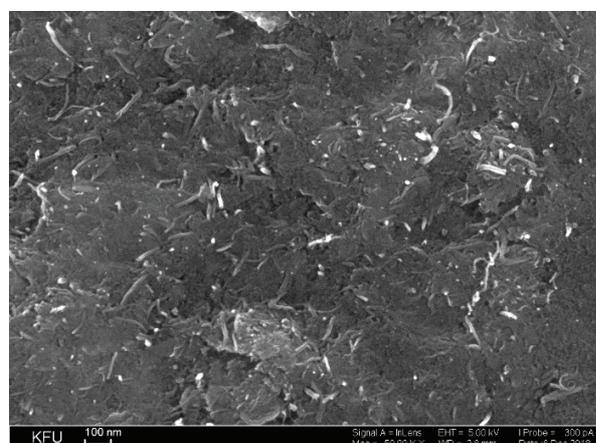
3.2. Kinetic study

The equilibrium adsorption capacity (Q_e , mg·g⁻¹) of the material was determined from the following expression:

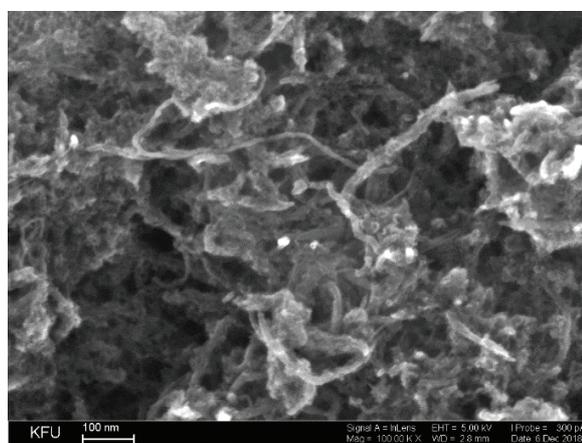
$$Q_e = \frac{V(C_0 - C_e)}{m}, \quad (1)$$

where C_0 (mg·L⁻¹) is initial solution concentration; C_e (mg·L⁻¹) is equilibrium concentration of dye in solution (after adsorption); V (L) is the solution volume; m (g) is the adsorbent weight.

The adsorption kinetics depends on many factors. To describe the adsorption rate, it is necessary to take into account the limiting stage of



(a)



(b)

Fig. 1. SEM micrographs of MPC: a – magnification 50.00 kX; b – magnification 100.00 kX

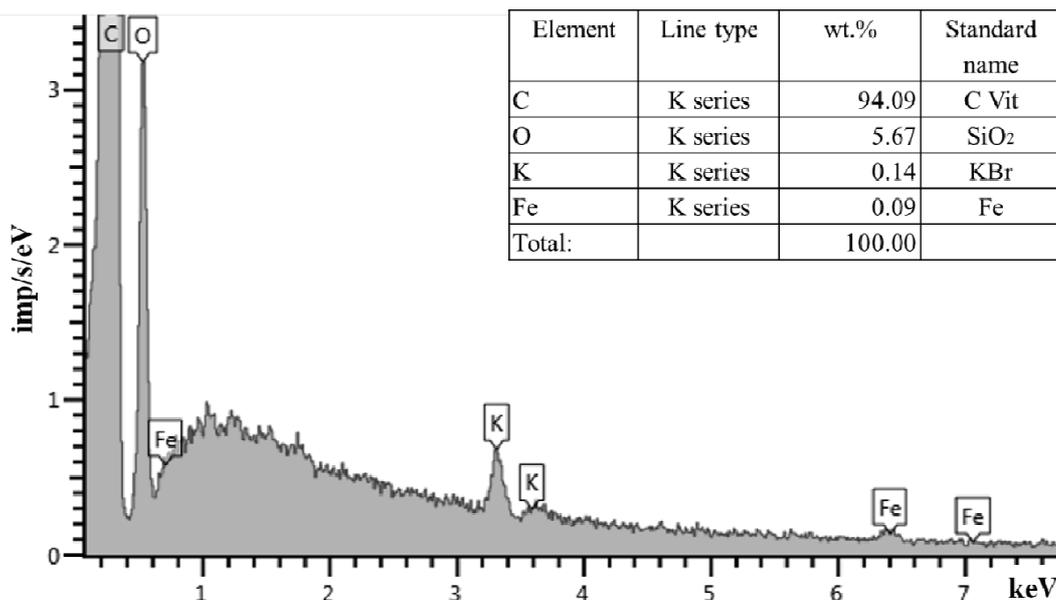


Fig. 2. Elemental analysis of MPC

the process. As for any heterogeneous chemical reaction, the adsorption reaction can proceed in the diffusion region, when the adsorption limiting step is the delivery of the adsorbate to the adsorbent surface, and in the kinetic region, when the rate of chemisorption for the process is the limiting step.

Based on the results of the experiments, the kinetic dependences of the adsorption of MB and MG were plotted (Fig. 3). According to the data obtained, the adsorbent exhibits high adsorption activity with respect to organic molecules: 2446.6 mg·g⁻¹ according to MB and 2043.1 mg·g⁻¹ according to MG. Adsorption equilibrium is reached within 15 minutes.

To describe the process of sorption of MB and MG molecules, namely, the mechanisms involved in the transfer of sorbate to the surface and inside the structure of sorbents, the obtained experimental data were processed by equations of known kinetic models (pseudo-first and pseudo-second order, Elovich and intraparticle diffusion) [24–26].

Phenomenological models are often used that imitate mass transfer processes with the help of formal equations of chemical kinetics. This is due to the complexity of the quantitative description of diffusion processes using simple models. This approach most often involves the use of pseudo-first and pseudo-second order models.

The Lagergren equation is called a pseudo-first order equation and is used to describe kinetic processes that depend on the concentration of a solution and the sorption capacity of a solid. This equation is widely used in the literature to describe the sorption kinetics and is the first rate equation developed to describe sorption in liquid/solid systems [24].

Currently, the pseudo-second order equation is used to describe and correlate the kinetic data of solid/solution sorption systems [25, 26]. The applicability of this model indicates the occurrence of a reaction that limits the sorption kinetics. This is obviously due to the fact that the theoretical substantiation of the pseudo-second order model is based on fundamental theories of surface reactions. There are studies in which the pseudo-second order model is considered as a rather flexible mathematical formula capable of modeling the sorption kinetics of internal mass transfer (diffusion) for systems with flat and spherical particles [26].

The intraparticle diffusion equation was developed for sorbents with a system of pores, the size of which allows the sorbate molecules to penetrate inside and settle there. In this case, the rate of the internal mass transfer stage is taken into account, i.e. diffusion of the sorbate in the pores of

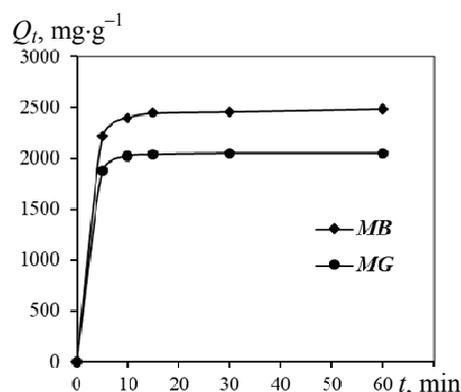


Fig. 3. Adsorption kinetics of MB and MG molecules on MPC

the sorbent with spherical particles [26]. If the dependency Q_t vs. t is a straight line passing through the origin ($C = 0$), then we can assume that internal diffusion is the limiting stage of adsorption. If, when constructing Q_t vs. t , a straight section was obtained that does not pass through the origin of coordinates; this may be due to the difference in the mass transfer rate at the initial and final stages of sorption. In addition, such a deviation of straight lines from the origin of coordinates may indicate that diffusion in pores is not the only limiting stage [26].

According to Fig. 4, rather low values of the coefficients ($R^2 = 0.8483$; 0.421) of the pseudo-first order model (Fig. 4a, Table 1) suggest that there is no chemical interaction of the dye molecules MB and MG with the functional groups of the sorbent. The pseudo-second order model with $R^2 = 0.9999$ for both dyes (Fig. 4b, Table 1) describes the absorption well, which indicates the occurrence of a second-order reaction on the surface of the sorbent. Satisfactory correlation of experimental data in the coordinates of the Elovich model indicates the energy inhomogeneity of the sorbent surface, which, in turn, promotes chemical adsorption [27].

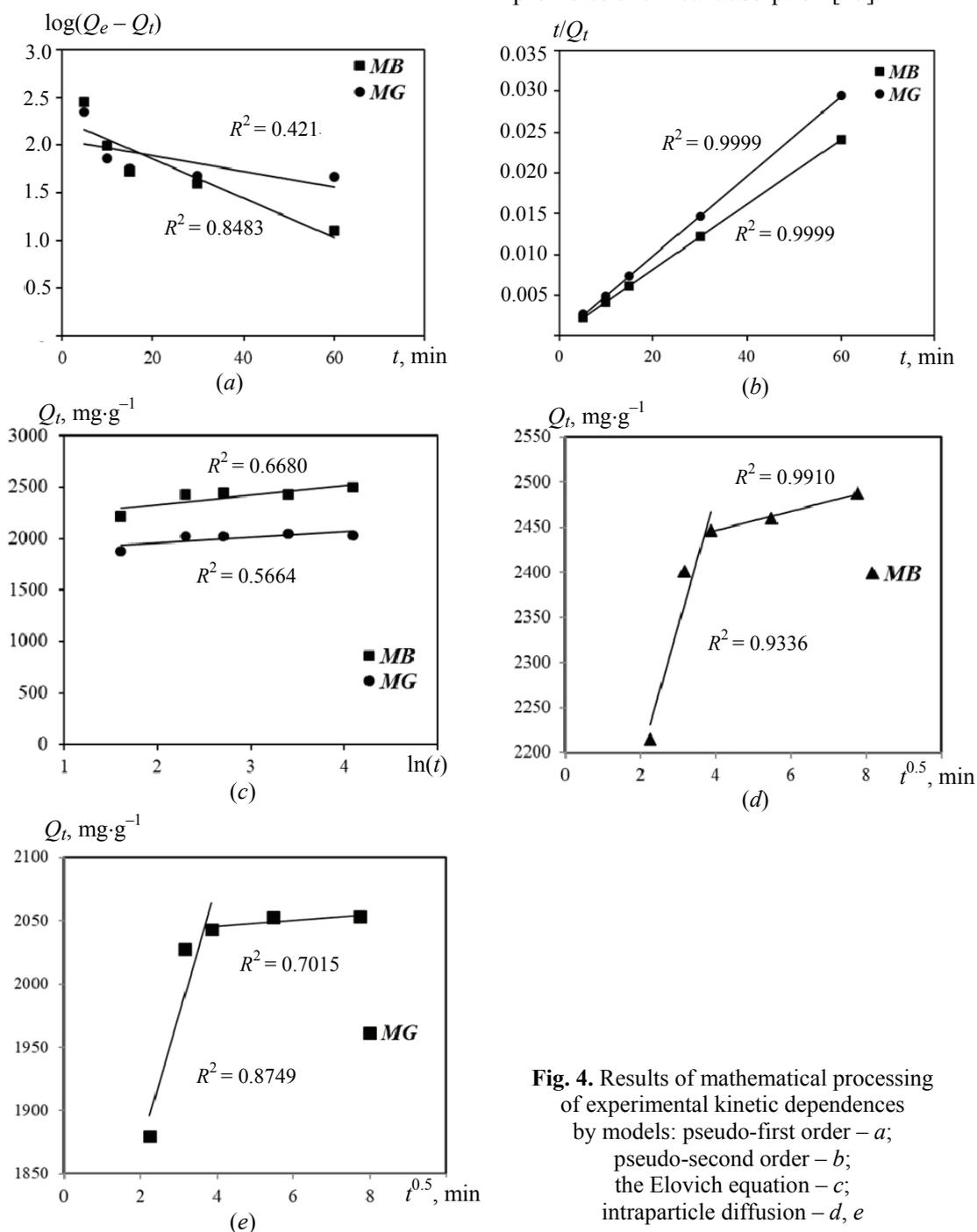


Fig. 4. Results of mathematical processing of experimental kinetic dependences by models: pseudo-first order – a; pseudo-second order – b; the Elovich equation – c; intraparticle diffusion – d, e

Table 1. Kinetic parameters of organic dye sorption*

Dye	Model parameter					
	Pseudo-first order			Pseudo-second order		
	$\log(Q_e - Q_t) = \log Q_e - \frac{k_1}{2.303} t$			$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{1}{Q_e} t$		
	Q_e	k_1	R^2	Q_e	k_2	R^2
MB	187.28	0.047	0.8483	2500	0.0003	0.9999
MG	114.18	0.008	0.4210	2000	0.0028	0.9999
	Elovich equation			Intraparticle diffusion equation		
	$Q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln t$			$Q_t = k_{id} t^{0.5} + C$		
	α	β	R^2	k_{id}	C	R^2
MB	$8.67 \cdot 10^{11}$	0.0107	0.6680	144.3/10.57	1908.3/2404.3	0.9336/0.991
MG	$2.16 \cdot 10^{16}$	0.0182	0.5664	102.5/2.42	1666.7/2035.7	0.8749/0.7015

* Q_t ($\text{mg} \cdot \text{g}^{-1}$) the number of adsorbed dye molecules on the adsorbent surface at a time t ; k_1 (min^{-1}) pseudo-first order adsorption rate constant; k_2 ($\text{g} \cdot (\text{mg} \cdot \text{min})^{-1}$) pseudo-second order adsorption rate constant; α ($(\text{min} \cdot \text{mg} \cdot \text{g}^{-1})^{-1}$) adsorption constant; β ($\text{g} \cdot \text{mg}^{-1}$) degree of surface coverage and chemisorption activation energy; k_{id} ($(\text{mg} \cdot (\text{g} \cdot \text{min})^{-1})^{-1}$) internal diffusion coefficient; C ($\text{mg} \cdot \text{g}^{-1}$) boundary layer thickness.

The adsorption of MB and MG under the conditions under consideration does not obey the Elovich model (Fig. 4c, Table 1).

The sorption rate can be limited both by the stage of chemical interaction and by the sorbate diffusion. In this case, transport processes (the movement of sorbate molecules from the solution to the active centers of the sorbent) play an important role in the sorption system. It is possible to estimate the contribution of the diffusion process to the sorption kinetics using the intraparticle diffusion model. The constant C in the equation of this model is proportional to the thickness of the boundary layer. If it is equal to zero, that is, the straight line passes through the origin, then the sorption process is limited by internal diffusion [26]. According to the data obtained (Fig. 4d, e), the dependences in the coordinates Q_t vs. $t^{0.5}$ are bilinear – the sorption of dyes is accompanied by two stages. The first sharper step is associated with diffusion in the boundary layer; the second one corresponds to the stage of pore filling. In addition, the first linear section does not pass through the origin for all dyes. Consequently, the process is not limited by internal diffusion and has a mixed-diffusion character [26].

3.3. Isotherm study

The construction of a sorption isotherm makes it possible to determine the maximum sorption capacity for the extracted component. The nature of the isotherm (curvature of the course, the presence of kinks, the length of the linear section, etc.) helps to interpret the sorption interactions between the active surface of the sorbent and the extracted components and to establish the key features of sorption.

The well-known models for studying the adsorption process are the Langmuir and Freundlich models. The Langmuir isotherm (Table 2) is an empirical isotherm describing monolayer adsorption, i.e. adsorption occurs on a homogeneous surface covered with adsorption centers. Adsorption is considered as a “chemical” equilibrium between a free adsorption center, a substance in solution, and an adsorbed substance (substance + center, adsorbate). On the surface, free centers and the adsorbate form an ideal solution [28].

Experimental isotherms are not always described by the Langmuir equation. The theoretical concepts developed by Langmuir idealize and simplify the true picture of adsorption. In fact, the surface of the adsorbent is inhomogeneous, there is an interaction

between the adsorbed particles, active centers are not completely independent of each other, etc. Some of these factors are taken into account by the Freundlich isotherm. The Freundlich isotherm (see Table 2) is used to describe non-ideal sorption equilibrium data, which is often an initial surface adsorption followed by a condensation effect resulting from extremely strong interactions. Furthermore, it is contemplated that after the surface is covered, additional adsorbed particles can still be accommodated. In other words, this equation describes polymolecular adsorption [28].

To describe adsorption on homogeneous and heterogeneous surfaces and to determine the nature of adsorption interaction by activation energy (Table 2), the Dubinin–Radushkevich isotherm is used [29].

The experimental results obtained are shown in Fig. 5, Table 2.

The experimental sorption isotherm of MG was satisfactorily described by the Langmuir and Freundlich equation (Fig. 6a, b). According to the calculated values, the maximum adsorption capacity of MPC for MG was 3333.3 mg·g⁻¹. The activation energy of sorption of MG molecules was 0.016 kJ·mol⁻¹ (Table 3). The adsorption of MB molecules obeys the theoretical models of Freundlich and Dubinin–Radushkevich (Fig. 6b, c), which indicates the layer-by-layer filling of the pore space of the sorbent. The value of the activation energy of the sorption of MB molecules, as well as the MG, corresponds to physical sorption and has a value of 0.013 kJ·mol⁻¹ (Table 3).

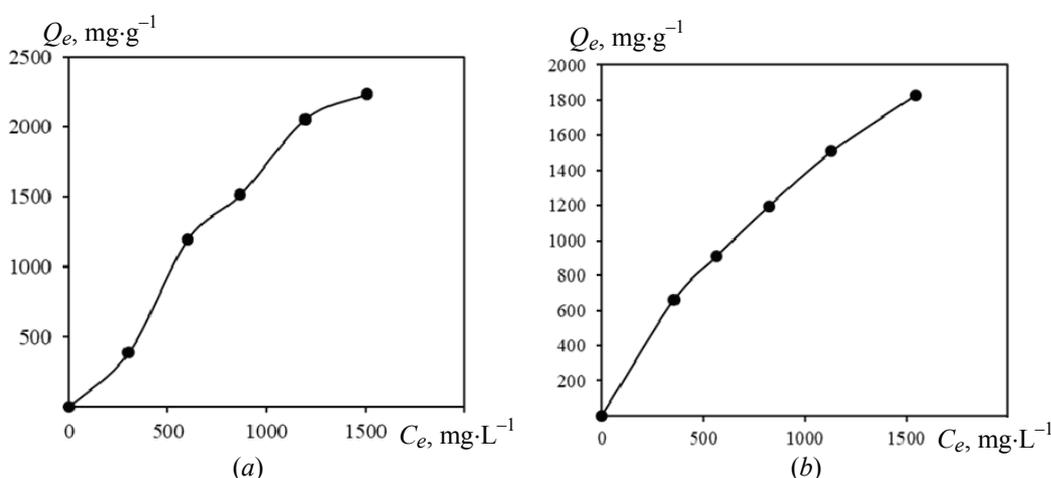


Fig. 5. Adsorption isotherms of MB (a) and MG (b) dyes on MPC

Table 2. Adsorption equations based on Langmuir, Freundlich, Dubinin–Radushkevich models [28, 29]*

Langmuir	Freundlich	Dubinin–Radushkevich
$Q_e = Q_{\max} \frac{K_L C_e}{1 + K_L C_e}$	$Q_e = k C_e^{1/n}$	$Q_e = Q_{\max} \exp(-K_{ad} \varepsilon^2)$
<i>Linear form of equations</i>		
$\frac{1}{Q_e} = \frac{1}{Q_{\max}} + \frac{1}{Q_{\max} K_L} \frac{1}{C_e}$	$\ln Q_e = \frac{1}{n} \lg C_e + \lg k$	$\ln Q_e = \ln Q_{\max} - K_{ad} \varepsilon^2$
<p>* Q_{\max} (mg·g⁻¹) is maximum adsorption; K_L is adsorption rate constant; k, $1/n$ are Freundlich constants indicates how favorable the adsorption process; k (mg·g⁻¹ (mg⁻¹)) shows the adsorption capacity of the sorbent; K_{ad} (mol²·kJ⁻²) Dubinin–Radushkevich isotherm constant; ε (kJ·mol⁻¹) is the Polanyi potential.</p>		

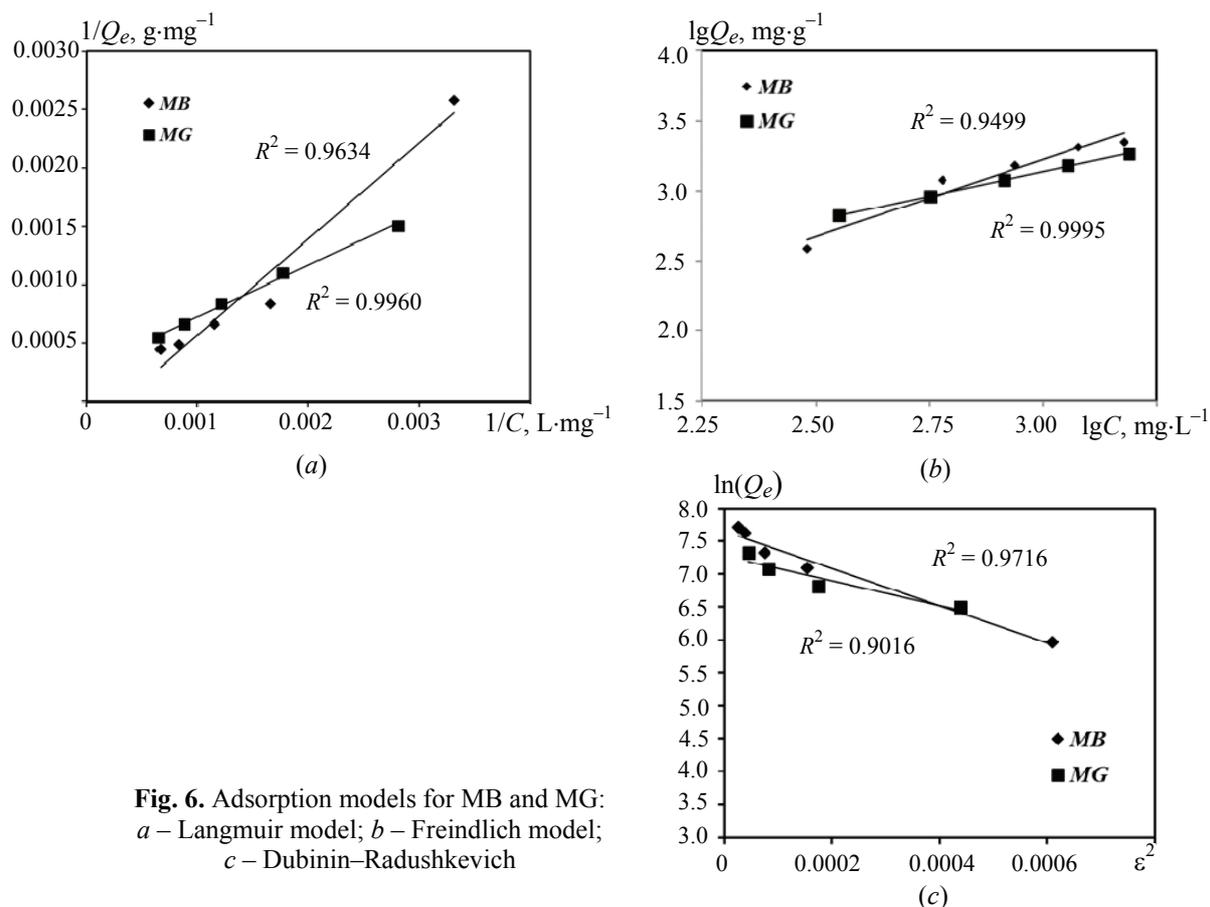


Fig. 6. Adsorption models for MB and MG: a – Langmuir model; b – Freundlich model; c – Dubinin–Radushkevich

Table 3. The parameters of MB and MG molecules sorption on MPC by isotherm equations

Dye	Model parameter			
<i>Langmuir</i>				
	K_L	Q_{\max}		R^2
MB	-0.0004	-3333.3		0.9634
MG	0.4436	3333.3		0.9960
<i>Freindlich</i>				
	n	$1/n$	k	R^2
MB	0.918	1.089	0.901	0.9499
MG	1.435	0.697	11.07	0.9995
<i>Dubinin–Radushkevich</i>				
	k_{ad}	Q_{\max}	E	R^2
MB	2837.5	2118.01	0,013	0.9716
MG	1884.7	1453.16	0.016	0.9016

4. Conclusion

The authors studied the adsorption of synthetic organic dyes from aqueous solutions on mesoporous carbon. The morphological features, elemental

composition and parameters of the porous space of the studied material are determined. The BET specific surface area was $2360 \text{ m}^2\cdot\text{g}^{-1}$ for nitrogen. Kinetic and isothermal studies of the absorption of

dye molecules – methylene blue and malachite green – were carried out in a static mode. It has been established that sorption is rapid – adsorption equilibrium is reached in 15 min at the adsorption activity of MPC: 2446.6 mg·g⁻¹ for MB and 2043.1 mg·g⁻¹ for MG. Adsorption kinetics has been described using pseudo-first order, pseudo-second order, Elovich and intraparticle diffusion models. The adsorption process is satisfactorily described by the pseudo-second order model and the intraparticle diffusion model, thus indicating a "sorbate-sorbate" interaction along with a contribution to the overall rate of the internal diffusion process. Empirical data obtained in the course of isothermal studies were processed using the Langmuir, Freundlich, Dubinin-Radushkevich models. The calculated data confirm the physical mechanism of absorption in accordance with the values of the activation energy $E = 0.016$ kJ·mol⁻¹ for MG molecules and $E = 0.013$ kJ·mol⁻¹ for MB.

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7. Conflict of interests

The authors declare no conflict of interest.

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