

Adsorption of dyes onto activated carbon prepared from olive stones

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Abstract: Activated carbon was produced from olive stones (OSAC) by a physical process in two steps. The adsorption character of this activated carbon was tested on three colour dyes molecules in aqueous solution: Methylene blue (MB), Rhodamine B (RB) and Congo Red (CR). The adsorption equilibrium was studied through isotherms construction at 30 °C, which were well described by Langmuir model. The adsorption capacity on the OSAC was estimated to be 303 mg/g, 217 mg/g and 167 mg/g respectively for MB, RB and CR. This activated carbon has a similar adsorption properties to that of commercial ones and show the same adsorption performances. The adsorption kinetics of the MB molecule in aqueous solution at different initial concentrations by OSAC was also studied. Kinetic experiments were well fitted by a simple intra-particle diffusion model. The measured kinetics constant was influenced by the initial concentration and we found the following correlation: $K_{id} = 1.55 C_0^{0.51}$.

Keywords: activated carbon; olive stones; adsorption; dyes; isotherms; kinetic

Introduction

The liquid-phase adsorption process using activated carbon is a known efficient technique widely used to remove organic pollutants from different effluents (Hsieh, 2000; Nouri, 2002; Arriagada, 1999; Garcia-Araya, 2003). Activated carbon is a highly porous solid, its internal total pores volume is commonly in the range of 0.5 to 1.0 cm³/g and can reach 2 cm³/g (Hu, 2001). Activated carbon porosity is essentially microporous with mean size lower than 2 nm, generating a high specific surface usually ranging from 900 to 1500 m²/g and can some times exceed 1500 m²/g (Hu, 2001; Pelekani, 2000). Even though high surface area, only a limited fraction of this surface is accessible to a given molecule of adsorbate. This is due to steric effects which control the diffusion of the solute molecules in the pores and to the behaviour in adsorption process related to the nature of chemical functional groups.

More recently, some investigators are interested not only in the surface area and porosity but also in the physical and chemical nature of the surface of activated carbon in order to understand and explain observed results of adsorption experiments (Laszlo, 2001; Peirera, 2003).

The structural characteristics of the activated carbon, porosity, pore size distribution, specific area, are determined by mercury porosimetry or by adsorption isotherms. In this last technique, the well-known molecules used are N₂, CO₂, CH₄ or C₃H₈. The nitrogen is an excellent standard adsorbate to estimate the specific surface (Sing, 1989) and porosity (Sonwane, 2000). When the activated carbon is used as adsorbent of molecules in liquid solutions, these structure allow just a qualitative interpretation. It is also difficult to specify for a given activated carbon an exhaustive test on all the potential adsorbed molecules and in all mixtures. Published work is limited to carry out tests on the typical molecules taken as models of certain families of the matter to be treated (Lin, 1995; Chinn, 1999; Rahman, 2003; Guzel, 2002). It is noted that some properties of these molecules (pK_a, functionality, polarity, molecular weight and size) can explain partially the interaction forces which took place on the surface of activated carbon. In another way, the adsorption process at the industrial scale, in particular in continuous operations, requires process kinetic control. Independently of the contact mode between fluid and

solid, the adsorption process can be described by three successive steps: external transport of the solute through the boundary layer surrounding the particle, internal diffusion in the particle, and adsorption of solute on the internal surface. Usually, the adsorption step is supposed instantaneous compared to the kinetics of the other two mass transfer steps (Alopaeus, 2000). Consequently the adsorption kinetic is often limited by the rate of the first two steps. Kinetic analysis is based on experiments carried out in a laboratory scale and the kinetic studies are often limited to a description of the adsorption dynamics under a given operating conditions (Lin, 1995; Gupta, 1997).

The present study reported the characterisation of an activated carbon, prepared in our laboratory from olive stones (OSAC), using adsorption of dyes in aqueous solution. This activated carbon is produced in two steps: carbonisation followed by activation with steam. Adsorption of three organic dyes of textile industry: Methylene Blue (MB), Rhodamine B (RB) and Congo Red (CR) were tested. These molecules were selected as models to estimate the adsorption capacity of the prepared activated carbon in aqueous solution. They belong to two chemical classes of water soluble organic dyes: cationic and anionic. The OSAC adsorption capacities of these dyes can give information about the interactions of adsorbent-adsorbate. These dyes have different molecular sizes, therefore, we expect different steric effects. The principal static properties are deduced from adsorption isotherms. The OSAC performances were compared with those of two commercial activated carbons. Kinetic adsorption analysis in a discontinuous system of MB onto OSAC was also performed.

1 Experimental

1.1 Materials

1.1.1 Activated carbon

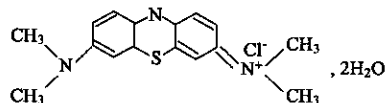
The activated carbon used was prepared from olive stones. This precursor is a by-product disposed in a considerable amount by oleic industries. The process follow-up for the preparation of activated carbon consists of the carbonisation of olive stones in a nitrogen atmosphere followed by steam activation. A precursor sample weighting 25 g, with particle size in the range 1.25–4 mm, was introduced in a vertical tubular reactor heated by electric furnace and fed with a continuous nitrogen flow rate of 9 L/h. The carbonisation time and temperature were optimized to 2 h and 600 °C

respectively. The char obtained in the first step with a yield of 30% was activated in the same reactor by the steam streamed in nitrogen at 70% volumic fraction at the same flow rate of nitrogen used in the carbonisation step. The activation temperature was fixed to 700°C during 8 h. A granular activated carbon was obtained with a global massic yield of 20%.

To carry out a comparative adsorption study, two commercial granular activated carbons were also tested in the same conditions: Norit and Chemviron. Their structural characteristics are close to those of the OSAC and they are recognized to be effective for the sorption of organic matters in solution. These granular activated carbons were crushed to obtain a granulometric range of 63–100 μm with an average diameter of 80 μm .

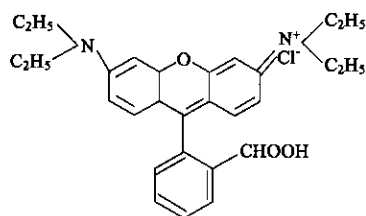
1.1.2 Dyestuffs

Methylene Blue (3,9-bisdimethylamino-phenazothionium chloride), was supplied by the Labosi Company. It belongs to thiazine dye class. It has a chemical formula $\text{C}_{16}\text{H}_{18}\text{ClN}_3\text{S}$, $2\text{H}_2\text{O}$ and a developed formula:



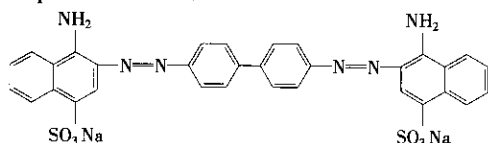
The molecular weight of the anhydrous product is 319.86 g. It absorbs visible light at a characteristic wavelength of 660 nm. The largest section of this molecule is estimated to 1.30 nm^2 (Santamarina, 2002). This dye is used in several fields, in particular in the industry of paper, dyeing and leather (Perrin, 1993).

Rhodamine B (Basic violet 10), was supplied by the Labosi company, it belongs to Xanthenes class. It is a basic dye of $\text{C}_{28}\text{H}_{31}\text{N}_2\text{O}_3\text{Cl}$ formula and developed as follows:



It has a molecular weight of 479 g. It absorbs the visible light at a characteristic wavelength of 550 nm. Among its applications, we indicate paper colouring, cotton, silk and leather dyeing (Perrin, 1993).

Congo Red was supplied by the Merck Company. It belongs the direct dyes. Its formula is a $\text{C}_{32}\text{H}_{22}\text{N}_6\text{Na}_2\text{O}_6\text{S}_2$ and developed as follows:



It has a molar weight of 696.66 g. It absorb the visible light at 500 nm wavelength. The largest section of this molecule is 1.94 nm^2 (Pelekani, 2001). It is used in dyeing processes (Perrin, 1993).

1.2 Characterisation techniques of activated carbon

1.2.1 Nitrogen isotherms adsorption of the activated carbons

The porous texture characterisation of the three carbons used was accessed by nitrogen adsorption isotherm at 77 K

and carried out on an automatic apparatus Quantachrome Autosorb AS1C. Samples were first outgassed at a temperature of 250–300°C under vacuum lower than 0.03 torr. The nitrogen adsorption isotherm data at 77 K was used to estimate the specific surface by using BET model fitting. The micropore size and volume are calculated by the Dubinin-Radushkevich (DR) method using nitrogen isotherm data. The total pore volume is estimated from the nitrogen quantity adsorbed at a relative pressure near the unity. The mesopore volume is obtained by the difference between the total pore volume and that of the micropore determined by the DR method.

1.2.2 Iodine number

The iodine number gives an indication about the accessible specific surface area of activated carbon (Karpel, 1991) and on its microporosity (Usmani, 1994). It is expressed in milligrams of iodine adsorbed per gram of activated carbon, this number is considered similar to the specific surface area expressed in m^2/g (Arriagada, 1995). It is determined by using the standard method (Environmental Agency Protection, 1973). A sample of 1 g of activated carbon, with a particle size lower than 0.1 mm and dried during 3 h, was suspended in 10 ml of 5% wt HCl in a glass vessel. The latter is heated by a sand bath for 30 s. After cooling the vessel to ambient temperature, 100 ml of iodine solution (0.1 mol/L) is introduced into the vessel. The suspension is then shaken during 30 s and filtered using Whatman paper filter. The free iodine in the filtrate solution is titrated by a sodium thiosulfate solution (0.1 mol/L).

1.2.3 Phenol number

Phenol adsorption was selected as a technique to characterise the micropore (San, 1998). This number is an indication of the adsorption capacity of aromatic compounds. It is determined by a standard specific technique (Ruiz-Bevila, 1984). Adsorption experiments were conducted by dispersing a sample of 0.3 g of dry activated carbon in 100 ml of phenol solution (0.1 g/L) in a glass vessel. After 24 h under continuous agitation at 25°C, the residual phenol concentration was measured by adsorption at a characteristic wave length of 270 nm.

1.2.4 Acidic and basic surface groups

Acidic and basic surface groups were determined by using the Boehm method (Julien, 1994). The acidic functions are measured by mixing 1 g of activated carbon, dried at 110°C during 48 h, with 50 cm^3 of sodium hydroxide solution during 72 h, under continuous agitation. The suspension is then filtered and titrated by 0.1 mol/L HCl solution. The basic functions of the surface are carried out by mixing 1 g of the activated carbon with 50 ml of 0.1 mol/L HCl solution. After an agitation time of 72 h, the suspension is filtered and titrated by 0.1 mol/L sodium hydroxide solution.

1.2.5 Ash content

The relative weight of mineral matter resulting from calcination of the activated carbon, is the ash content. It was measured according to a standard method (Environmental Protection Agency, 1973): the residual weight of a sample of 5 g of the solid was calcinated at a temperature of 600°C under an oxidising atmosphere.

1.2.6 Measurement of pH and conductivity

The pH is one of the most important factors controlling the adsorption of dye. The pH and conductivity parameters were measured in a filtrated suspension of activated carbon of a granulometry lower than 250 mm at a concentration of 2 g/L in distilled water during 6 h.

1.3 Characteristics of activated carbons

The nitrogen adsorption-desorption isotherms at 77 K of the three studied activated carbons are reported in Fig. 1. These isotherms are of type I according to the BDDT classification indicative of highly microporous materials. The existence of desorption hysteresis loops shows the existence of mesoporous cavities.

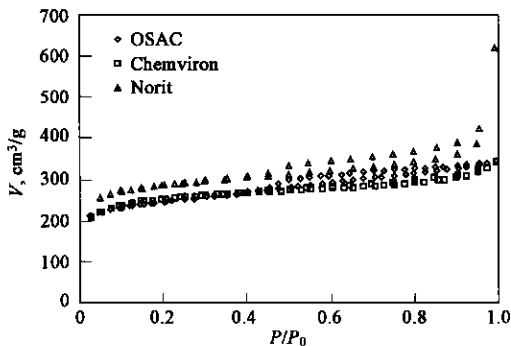


Fig.1 Nitrogen(77 K) adsorption-desorption isotherms onto three activated carbons

The measured characteristics of the three carbons used in this work are reported in Table 1. It can be observed that the OSAC has the highest ash content, its conductivity is important due to the mineral content and OH⁻ concentration resulting in pH elevation of distilled water to 8.3 when it is contacted with activated carbon. Chemviron and Norit have respectively an acid and a neutral pH.

Table 1 Principal characteristics of studied activated carbons			
Activated carbon	OSAC	Chemviron	Norit
Apparent density, cm ³ /g	0.54	0.70	0.30
Ash content, % ww	7.5	6.4	5.8
pH	8.3	6.2	7.15
Conductivity, μS/cm	167	17	15
BET specific surface, m ² /g	920	951	1058
Total pore volume, cm ³ /g	0.53	0.49	0.96
DR micropore volume, cm ³ /g	0.38	0.36	0.55
Mesopore volume, cm ³ /g	0.15	0.13	0.41
Mean pore diameter, nm	2.80	3.87	3.13
Phenol number, mg/g	30.05	32.33	32.04
Iodine number, mg/g	1060	964	998
Acidic surface quantity, meq/g	0.83	0.66	0.81
Basic surface quantity, meq/g	1.40	0.28	0.29

The specific surface areas of the three carbons are in the same order. We note that the Norit carbon has the most important total pore volume.

The phenol and iodine numbers are of the same order of magnitude for the three adsorbents, however, the OSAC reveals an iodine number relatively high. This demonstrates the prospective potential of this product for the adsorption of low molar weight species(Juang, 2001).

The titration of the surface functions gives comparable acid quantities for the three carbons. However, the concentration of the basic functions is more important on the surface of the OSAC.

1.4 Adsorption isotherms construction and kinetic studies

The equilibrium isotherms were determined by mixing 10 mg of a dry activated carbon with 200 ml of a dye solution at different concentrations in a sealed glass bottle. All solutions were then left in a bath maintained at 30°C and shaken for 3 d. This time was fixed on the basis of preliminary tests, which show that equilibrium was reached in 24 h. The suspension was then filtered through an organic membrane of porosity 0.45 mm. This membrane does not adsorb the dye and retain perfectly the powdered of activated carbon. The residual concentration of the dye was then titrated in the filtrate using a spectrophotometer UV-visible at a specific wavelength. The optical density varies linearly with the dye concentration according to the Beer-Lambert law in the diluted concentration range. The adsorbed quantity, q_e , was calculated from the final concentration(C_e): $q_e = V(C_0 - C_e)/m$. m is a weight of carbon added to the same (V) volume of the dye solution of initial concentration(C_0). The kinetic measurement of adsorption was carried out with varying only contact time in similar equipment and conditions.

2 Results and discussion

2.1 Adsorption equilibrium of the three dyes in aqueous solution on the OSAC

The equilibrium adsorption quantity(q_e) of the three dyes on the OSAC versus the concentration of the aqueous solution at equilibrium(C_e) are given in Fig.2. The shape of these isotherms shows that they are of type I, indicating that the adsorption takes place in monolayer. Langmuir model is used to describe this isotherms, the model equation is expressed by the following linear form:

$$\frac{C_e}{q_e} = \frac{1}{bq_{\infty}} + \frac{C_e}{q_{\infty}}$$

Where q_{∞} is the maximum adsorption capacity and b is the equilibrium constant. The Langmuir equation fit correctly the experimental isotherms data of the OSAC(Fig.3). The evaluated maximum specific adsorption capacities of the OSAC and the equilibrium constants are reported by Table 2. The Freundlich model is also tested in fitting the isotherms data, it fails to describe correctly this results.

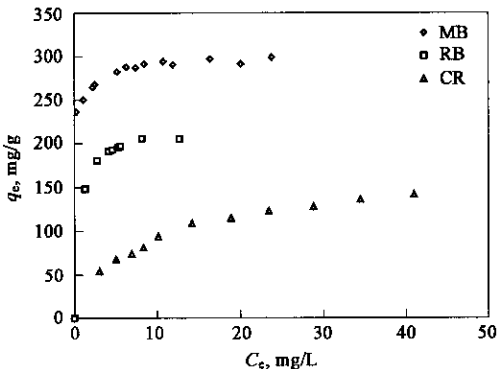


Fig.2 Three dyes adsorption isotherms onto olive stones activated carbon($T = 30^{\circ}\text{C}$)

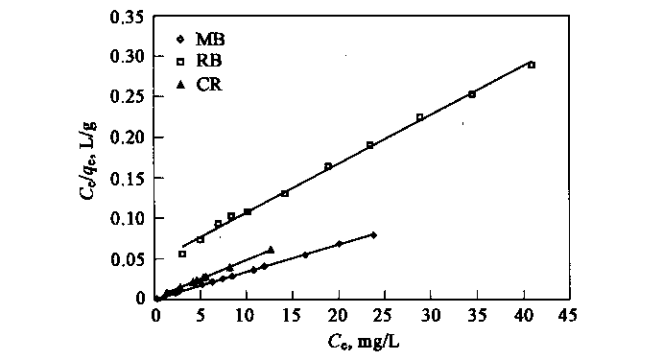


Fig.3 Langmuir fitting of the adsorption isotherms of three dyes on OSAC

Table 2 Langmuir parameters determined for the adsorption of MB, RB, CR by three activated carbons

Adsorbent	S_{BET} , m^2/g	Adsorbate	q_{∞} , mg/g	q' , mg/m^2	b , L/mg
OSAC	920	MB	303	0.329	4.12
		RB	217	0.236	1.84
		CR	167	0.182	0.13
Chemviron	951	MB	238	0.250	2.62
		RB	178	0.187	11.2
		CR	167	0.175	0.77
Norit	1058	MB	312	0.295	2.67
		RB	303	0.286	1.65
		CR	172	0.163	0.26

Fig.2 shows that OSAC presents a relative high affinity to the cationic dyes(MB and RB). This phenomenon can be attributed to the negative surface charge of carbon. The same observation is reported by other work using activated carbons from wood and others cellulosic adsorbents (Marmier-Dussoubs, 1991; Sun, 1997). Compared to RB, MB is better adsorbed; in fact, MB molecule has a smaller size, therefore this must be favourable to its accessibility to internal micropores.

2.2 Comparison with two commercial activated carbons

The adsorption of each of three dyes by the OSAC is compared to its adsorption on two commercial activated carbons: Norit and Chemviron. The adsorption isotherms at 30°C of the three dyes on the three activated carbons and under the same conditions are plotted in Fig. 4, 5 and 6.

The isotherms are of the same shape indicating that there is a similarity in the relative solute-adsorbent affinity. Isotherms are close to type I, indicating a monolawer adsorption process. These isotherms are well described by the Langmuir model from which the specific adsorption capacities are estimated(Table 2).

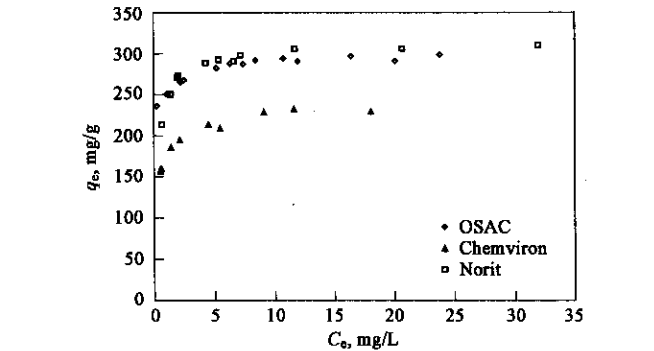


Fig.4 Methylene Blue adsorption isotherms onto three activated carbons

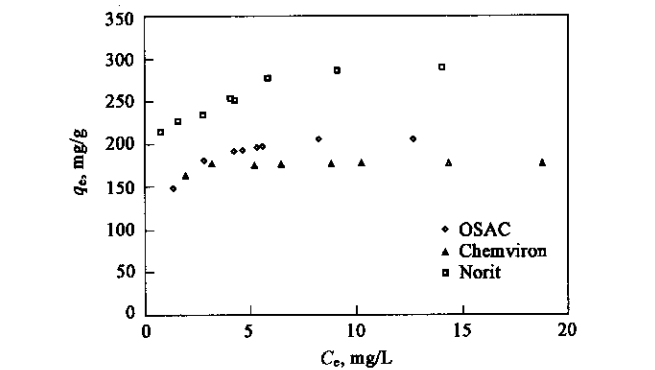


Fig.5 Rhodamine B adsorption isotherms onto three activated carbons

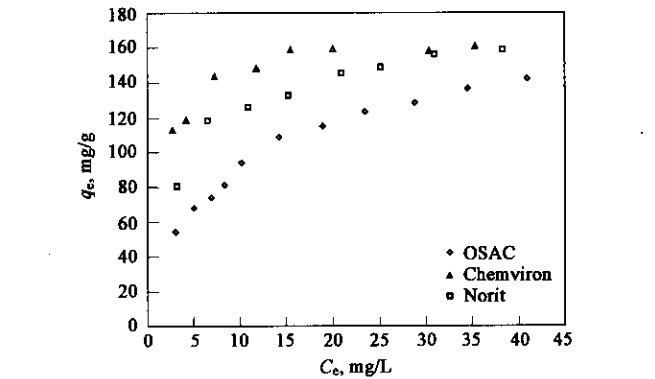


Fig.6 Congo Red adsorption isotherms onto three activated carbons

Fig.5 reveals very little difference between the adsorption isotherms of the MB on the OSAC and on the Norit activated carbon. Close adsorption capacities, 303 mg/g of OSAC and 312 mg/g of Norit are deduced. The affinity of the Chemviron carbon to the MB is less important and its adsorption capacity is 238 mg/g. The adsorption capacities of CR onto the three carbons are in the same range: 167 mg/g OSAC, 167 mg/g activated carbon Chemviron and 172 mg/g activated carbon Norit. Using the specific surface areas of the various activated carbons and the molar weights of the three dyes, the adsorption capacities are expressed into molar adsorption capacities in $mmol/m^2$ of adsorbent surface(Table 3). Table 3 also reports the micropore volume and the pH values of the three activated carbons. First, it is noted that the molecular capacities follow the order MB > RB > RC. Such order of adsorption capacities is probably due to the steric effect, knowing that these molecules have sizes which are classified as follows: CR > RB > MB. Second, the comparison of the three activated carbons shows that the adsorption capacity of the MB is maximum on the OSAC ($0.823\text{ mmol}/m^2$), that of RB is maximum on the activated carbon Norit($0.476\text{ mmol}/m^2$) and Chemviron carbon has the maximum adsorption capacity of the CR ($0.223\text{ mmol}/m^2$). These relative affinities depend essentially on the chemical and electric interactions surface-molecule (acido-basic behaviour, electronegativity etc.). By considering the acido-basic character of the activated carbon, we can say that the MB is more influenced by the acido-basic behaviour of adsorbent. Indeed, it is noted that the MB is more adsorbed by the activated carbon which has the higher aqueous suspension pH.

Table 3 Specific molar adsorption capacities (mmol/m²)

Carbon	V_{mic} , cm ³ /g	MB	RB	CR	pH
Chemviron	0.36	0.690	0.344	0.223	6.2
OSAC	0.38	0.823	0.392	0.208	8.3
Norit	0.55	0.735	0.476	0.197	7.15

2.3 Kinetic adsorption of MB on OSAC

The adsorption mechanism of the MB in aqueous solution by a porous solid takes place essentially in three steps: mass transfer of solute towards the external surface of the particle—external transport; mass transfer in the internal porosity of solid by a diffusion process—intraparticle transport; adsorption on the internal surface of solid.

The kinetic study aims principally to identify the limiting steps in the adsorption process. It should be noted that the majority of work suppose that adsorption is very fast and that the equilibrium is instantaneous in the interface liquid-solid inside the particle. In order to examine the mechanism of adsorption process, a suitable kinetic model is needed to analyse the rate data. If the agitation of the activated carbon suspension is considered sufficiently intense, the effect of exterior particle mass transfer is neglected. Consequently, the internal diffusion can be the kinetic limiting step (Baudu, 2001). For that, a kinetic simple model based on the intraparticle diffusion to describe the adsorption kinetic in aqueous solution will be tested. This model is proposed in 1962 by Weber and Morris, it is expressed by the equation: $q = k_{id} t^{0.5}$ (Wu, 2001; Namasivayam, 1993) where q is the amount of solute adsorbed at time t and k_{id} is the intraparticle kinetic constant diffusion.

Fig. 7 illustrates the concentration profiles of the MB dye in the solution against contact time for various initial concentrations. It shows that the initial adsorption rate is relatively high, this rate falls considerably with time. This is probably due to the mass transfer resistance inside of the activated carbon particle (Bagane, 2000). From the results of the Fig. 7, we plot the adsorbed quantity at time t , $q(t)$, versus square root of the time $t^{0.5}$ (Fig. 8). We obtain a straight lines with correlation coefficients close to unity which makes it possible to confirm that the diffusion of the MB inside the particle is a limiting step of the kinetic process. Therefore, these lines do not pass through the origin, which proves that intraparticle diffusion is not the only process, which controls the kinetic adsorption (Namasivayam, 1993; Houas, 1999). The values of kinetic constant determined from the slopes of the lines of Fig. 8 increase with initial concentrations of dye (Fig. 9) and follow the correlation: $k_{id} = 1.55 C_0^{0.51}$. A similar relation is found by Houas *et al.* (Houas, 1999) for the adsorption of MB onto CECA 40 carbon.

3 Conclusions

This experimental work shows that the activated carbon prepared from olive stones can be used for the adsorption of three dyes from textile industry in aqueous solution. The isotherms obtained are well fitted by Langmuir model showing the maximum capacities of 303 mg for the MB, 217 mg for RB and 167 mg for the CR per gram of activated carbon. Compared to commercial activated carbons, the olive stone

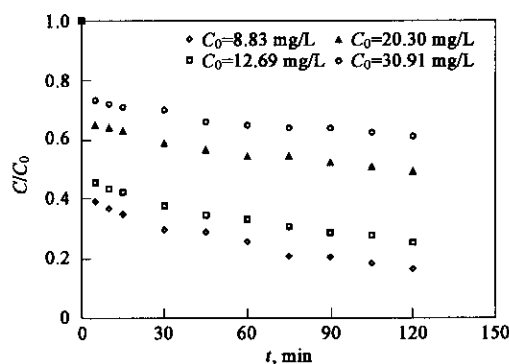


Fig. 7 Adsorption kinetic of MB onto OSAC for different initial concentrations of MB $T = 30^{\circ}\text{C}$, activated carbon concentration 50 mg/L of solution

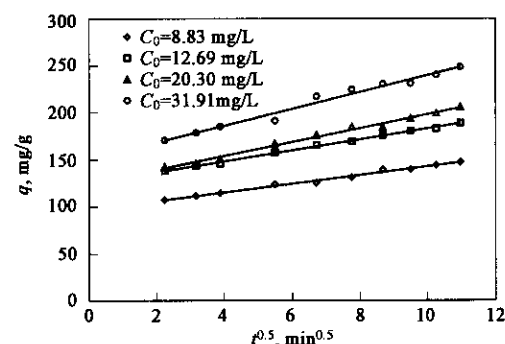


Fig. 8 Intraparticle diffusion model for the adsorption of MB at different initial concentrations ($T = 30^{\circ}\text{C}$)

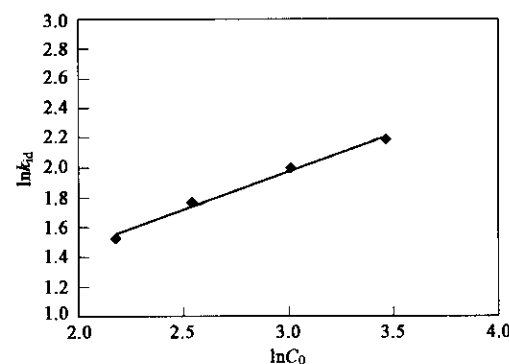


Fig. 9 Variation of intraparticle diffusion constant with MB initial concentration ($T = 30^{\circ}\text{C}$)

activated carbon can be considered effective for the elimination of these dyes in aqueous solution. The application of the intraparticle diffusion model to the MB adsorption on the OSAC shows a good fit of the experimental points. The kinetic intraparticle diffusion constant increases with initial concentration according to the relation $k_{id} = 1.55 C_0^{0.51}$.

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