Studies on the Absorption of Dyes Used in the Textile Industry Using Metallosilicate Beads as Adsorbents

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The purpose of this paper is to synthesize porous zeolitic materials in the form of macrospheres which are intended to be used as adsorbent for the adsorption of cationic dyes from aqueous solutions. The synthesized sample was characterized by water sorption technique, SEM and EDAX analyses. The adsorption capacities of iron silicate macrospheres (IYC) at various contact times and effect of pH were investigated. The experimental data were analysed using the Langmuir and Freundlich adsorption isotherms. The intraparticle diffusion model was used to describe the kinetic data.

Key words: porous material, basic dyes, adsorption isotherms, intraparticle diffusion model

Many industries such as paper, textile, leather and plastic materials use dyes and pigments to color their products. Discharge of wastewaters resulted from the dyeing process into the natural streams and rivers rises serious environmental problems. The presence of dyes in water affects not only the aesthetic nature but also reduces light penetration and the photosynthesis process. Because of their toxicity (even in very low concentration) and persistence after the releasing into the natural water, these coloring agents may affect the aquatic ecosystem. Therefore, their removal from the wastewater is required. Various methods have been used for the removal of dyes from the wastewaters such as membrane based separation processes, aerobic and anaerobic degradation using various microorganisms, chemical oxidation, photodegradation, coagulation and flocculation, electrochemical treatment and adsorption using different kind of adsorbents [1-7]. The adsorption is a promising technique for the removal of dyes from wastewater streams due to its low cost, simple design, easy operation and due to the possibility of recycling the adsorbents. The adsorption process typically uses low cost adsorbents such as clays, agricultural solid wastes, fly ashes, industrial by-products, biomass and zeolites [8-15]. Among these adsorptive materials, the zeolites which constitute an important class of crystalline microporous materials, have been accepted as appropriate adsorbents for the removal of cationic dyes from different types of residual wastewaters. Because the small pore sizes of zeolites restrict the adsorption of bulky molecules (due to the diffusional limitations) many strategies that aimed to shorten the diffusion pathway and/ or to broaden the diffusion space have been proposed. Templating technique constitutes a general method used to obtain hierarchical materials with controlled porosity [16-19]. In this purpose a variety of hard and soft templates has been employed to generate meso and macropores [20-22]. However, the hierarchical porous materials

synthesized via conventional techniques are usually obtained as powder or film. The synthesis of the monolithic zeolites with an appreciable of mechanical strength still remains a problem incompletely solved.

The aim of this work was to synthesize a porous hierarchically zeolitic material under macrospherical shape tusing as adsorbent for Basic Blue 3 and Basic Red 22 dyes.

Experimental part

Materials

Commercial basic dyes Astrazon Blue BG (C.I. Basic Blue 3) and Astrazon Red F3BL (C.I. Basic Red 22) used as adsorbates were obtained from Dye Star Company and were used without any further purification. The structures of the dyes are shown in figure 1.



Fig. 1. Chemical structures of the basic dyes

The IS macrospheres were prepared as follows: Solution A: 0.922 g Fe(NO₃), 9H₂O, 9 mL distilled water, 1.4 mL HCl 37% and 3 mL TEOS were kept under stirring for 6 h at room temperature. Gel B: A suspension of 2.7 g yeast cells dispersed in 4 mL distilled water was heated at 80°C under stirring for 6 h. After cooling, 9 g chitosan solution (4%) were introduced and the resulting gel was mixed for 2 h. Solution A was poured over the gel B and the mixture was kept under stirring for 2 h.

Finally the gel was transformed into macrospheres according to the procedure described in our previous papers [23].

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Characterization techniques of IYC beads SEM/EDX analysis

The scanning electron micrograph image and chemical composition of the synthetized sample were recorded on A VegaTescan LMHII (VegaTescan) scanning electron microscope coupled with an energy dispersive X-ray analyzer (Bruker XFlash 6/10 SDD with Esprit Software).

Water adsorption-desorption isotherms

Water adsorption-desorption isotherms were recorded on automated gravimetric analyzer IGAsorp produced by Hiden Analytical, Warrington (UK).

Adsorption studies

In the adsorption experiments the following calibration curves $y = 0.0715 \cdot x$, $R^2 = 0.998$ for Astrazon Blue BG and respectively $y = 0.0805 \cdot x$, $R^2 = 0.9943$ for Astrazon Red F3BL were used for determination of the unknown dye concentrations from the residual solutions.

The amount of dye adsorbed per unit mass of adsorbent q. (mg/g) at any time t, was calculated using the mass balance relationship equation as follows:

$$\mathbf{q}_{t} = \frac{\left(\mathbf{C}_{0} \cdot \mathbf{V}_{0} - \mathbf{C}_{t} \cdot \mathbf{V}_{t}\right)}{\mathbf{w}}$$
(1)

where: C_0 (mg/L) is the initial concentration of the dye in solution; C_t (mg/L) is the liquid phase concentration of the dye at any time t; V_0 – is the initial volume of solution, mL;

 $V_t = V_o \cdot n \cdot \Delta V$ is the volume of solution at time t; n – is the number of withdraws carried out; $\Delta V = 0.5$ mL is the volume of sample withdrawn at any time t; and w is the mass of the adsorbent (g). At equilibrium, $q_t = q_e$ and $C_t = C_e$; therefore the amount of dye adsorbed per unit mass of adsorbent at equilibrium, q_e (mg/g), was calculated by the following expression:

$$\mathbf{q}_{e} = \frac{\left(\mathbf{C}_{0} \cdot \mathbf{V}_{0} - \mathbf{C}_{e} \cdot \mathbf{V}_{e}\right)}{\mathbf{w}}$$
(2)

where C_0 and C_e are the initial and respectively the equilibrium dye concentrations in the bulk solutions (mg/L), V is the volume of the solution (L) and w is the mass of the adsorbent (g).

Adsorption experiments were carried out at different initial dye concentrations (30, 60, 90, 120 and 150 mg/L for AR dye and respectively 60, 90, 120, 150 and 180 mg/ L for AB dye) using a 5g/L adsorbent loading. In all the experiments the temperature was kept at 30°C and the stirring speed was 250 r.p.m. The adsorption tests were performed until the equilibrium concentrations were attained.

Results and discussions

Characterization of the adsorbent

Energy dispersive X-ray analysis (EDX) The elemental composition of the IYC beads (after

calcination and washing) determined by EDX analysis is shown in figure 2.

The results obtained from the EDAX analysis confirm the presence of Fe in the synthesized sample.

Scanning electron microscopy

The surface morphology and pores size of the IS sample were investigated by SEM analysis (fig. 3).

Figure 3 indicates that the iron silicate macrospheres contain wide pores resulted through removal of chitosan and yeast cells templates by calcination.



Fig 2. Elemental analysis of the calcined IS beads



Fig. 3. SEM image of the calcined IS sample

Determination of the textural properties from the water sorption data

The adsorption-desorption isotherm for the IYC beads is shown in figure 4.

The isotherm shows a less common allure. It is a hybrid between type II and type IV isotherms which has also some particular features. The H, type hysteresis loop is typical for the materials which have pores of different sizes and shapes. The broad hysteresis loop exhibiting a horizontal plateau with a steep desorption branch is an indicative for the existence of meso and macropores.

The low value of the upper limit of the isotherm, the horizontal plateau and the sharp raising of the adsorption branch indicate that the sample contain large pores whose volume cannot be assessed only by water adsorption technique. The values found for the textural characteristics of the sample, calculated from the adsorption branch of the isotherm were: BET surface area $S_{BET} = 234.5 \text{ m}^2 \cdot \text{g}^{-1}$,



Fig. 4. The adsorption-desorption isotherm



the micropore volume $V_{\text{micropore}} = 0.028 \text{ cc}\cdot\text{g}^{-1}$ and the mesopore volume $V_{\text{mesopore}} = 0.229 \text{ cc}\cdot\text{g}^{-1}$. The pore size distribution expressed as derivative of pore volume with respect to pore diameter versus pore diameters is shown in figure 5.

It can be noticed that the sample has a large distribution of mesopore diameters starting from 20Å until 160Å.

Effect of the initial dye concentration

The effect of initial dyes concentrations on the adsorption capacity of iron silicate beads was investigated in the range from 60 to 180 mg/L for AB dye and respectively in the range from 30 to 150 mg/L for AR dye. The adsorption capacity increased with the increase of the initial dye concentration from 14.84 mg/g to 19.85 mg/g for AB dye, and respectively from 4.85 mg/g to 15.07 mg/g for AR dye (fig. 6 and fig. 7).

The adsorption process was faster at higher concentrations due to the increased concentration gradient, which acts as a driving force at the transfer of the dye molecules from the bulk solution to the bead



Fig. 6. AB adsorption uptake versus adsorption time at various initial dye concentrations (T = 303 K, adsorbent dose 5 g/L)



Fig. 7. AR adsorption uptake versus adsorption time at various initial dye concentrations (T = 303 K, adsorbent dose 5 g/L)

surface. The adsorption rate is high at the early adsorption period due to the availability of a large number of vacant sites and decreases in time due to the lowering of the number of available adsorption sites. The differences that occur between the two dyes can be



Adsorption isotherm	Linear equation	Plots	Slope	Intercept			
Langmuir	$\frac{C_e}{q_e} = \frac{1}{q_m} \cdot C_e + \frac{1}{K_L \cdot q_m}$	$\frac{C_e}{q_e}$ vs C _e	$\frac{1}{q_m}$	$\frac{1}{K_L \cdot q_m}$			
Freundlich	$\log q_e = \log K_F + \frac{1}{n} \cdot \log C_e$	logq _e vs logC _e	$\frac{1}{n}$	logK _F			
q _m (mg/g) is the maximum amount of adsorbate per unit weight of adsorbent required to form a complete monolayer on the adsorbent surface; K _L (L/mg) is the Langmuir isotherm constant which is a measure of the affinity between adsorbate and adsorbent and is related to the free energy of adsorption.							

 K_F (mg/g) is a measure of the adsorption capacity (being related to the bonding energy) and 1/n corresponds to the strength of adsorption and to the surface heterogeneity. The value of n describes the adsorption characteristics as follows: if n = 1 the Freundlich expression reduces to a linear adsorption isotherm, which means a homogenous adsorption without interactions between the adsorbed species; n > 1, indicates that the adsorption is a favourable process; n < 1, indicates an unfavourable adsorption process.

Table 1ISOTHERM MODELS

Model	Cationic dyes					
	AB	AR				
Langmuir	1					
K _L (L mg ⁻¹)	0.1152	0.0429				
gm (mg g ⁻¹)	21.834	19.048				
R ²	0.9915	0.9958				
Freundlich						
n	5.0660	2.3397				
K _F (mg g ⁻¹)	8.2110	2.5404				
R ²	0.9921	0.9314				

Table 2ISOTHERM CONSTANTS ANDCORRELATION COEFFICIENTS FORTHE ADSORPTION OF AB AND AR DYESON IS BEADS

explained based on their chemical and steric structural differences (the position of positive charge, length and symmetry of the dyes molecules).

Effect of pH

In order to investigate the effect exerted by *p*H on dyes adsorption, the *p*H of the dyes solutions was varied over a *p*H range from 2 to 10, by adding either 0.1N HCl or 0.1N NaOH solution (fig. 8). The experiments were carried out at an initial dye concentration of 120 mg/L, contact time of 420 min, adsorbent dose of 5 g/L and temperature of 303 K.

The initial *p*H of dye solution controls the adsorption process, particularly the adsorption capacity, because it influences the surface charge of the sorbent and the ionization degree of adsorbate molecules in the solution. Surface functional groups of the metallosilicate beads may dissociate at higher *p*H values thus creating a higher number of anionic sites that contribute to the cationic dyes uptake. As the *p*H of the aqueous system increases, the number of negatively charged sites increases slightly favouring the adsorption of cationic dyes due to the electrostatic attraction between the negatively charged sites of the adsorbent and the positively charged dye molecules. At lower pH values the H^yions compete with the cations of dye for the occupation of vacant adsorption sites and consequently the amount of adsorbed dye decreases. If the pH of solution is changed from 2 to 12, the equilibrium adsorption capacity increases from 11.482 to 18.2 mg dye/g adsorbent for AB dye and respectively from 9.2844 to 15.51 mg dye/g adsorbent for AR dye. It can be noticed that the adsorption process is optimal in the *p*H range 6-10.

Adsorption isotherms

Adsorption isotherms describe the relationship between the amount of solute adsorbed onto the solid at equilibrium (q_e) and the equilibrium concentration of the solute remained in solution (C_e) at a given temperature [24, 25]. For the solid-liquid systems, the study of adsorption isotherms is very important to supply informations about the adsorption capacity of adsorbents. In order to quantify the affinity of the iron silicate beads for the basic dyes and to analyse the experimental data, the Langmuir and Freundlich isotherm models were used. The linear equations of the investigated adsorption isotherms are shown in table 1.

The models parameters calculated from the slopes and the intercepts of the corresponding linear plots are listed in table 2.

The Langmuir adsorption isotherm has been successfully used in many homogeneous adsorption processes wherein only a monolayer of dye can cover the surface of the adsorbent and a rapid decrease of the intermolecular attractive forces with the increase of distance is assumed [26].

The validity of the Langmuir adsorption process is assessed by means of a dimensionless constant called separation factor R_{t} which is calculated using:

$$R_{L} = \frac{1}{1 + K_{L} \cdot C_{0}}$$
(5)

where C_0 is the initial concentration of dye (mg/L) and K_L is the Langmuir adsorption isotherm constant. The value of R_L indicates whether the isotherm is irreversible $(R_L = 0)$, favourable $(0 < R_L < 1)$, linear $(R_L = 1)$ or unfavourable $(R_L > 1)$. The calculated R_L values of AB dye are found to lie between 0.12 and 0.046 at 30°C for the initial concentration values in the range 60 – 180 mg/L, while the calculated R_L values of AR dye are found to lie between 0.106 at 30°C for the initial concentration values in the range 30 – 150 mg/L. The obtained R_L values show that the adsorption of dye on metallosilicate beads is favourable irrespective of the initial dye concentration.

The Freundlich isotherm is an empirical model employed to describe the adsorption on heterogeneous systems (assumes a heterogeneous energetic distribution of the active sites on the adsorbent surface and takes into account the interactions occurred between the adsorbed species). This model is not restricted to the formation of the monolayer [27]. The values of *n* are 5.066 for AB dye and respectively 2.3397 for AR dye indicating the favourability of the adsorption process. The higher *n* value obtained for AB dye shows that the adsorption process is more favourable for this dye.

Comparing the fitting of the experimental adsorption data with the calculated values (summarized in table 2), in terms of \mathbb{R}^2 correlation coefficient values the following conclusions can be drawn regarding the applicability of the Langmuir and Freundlich models: for AB dye the best matching with the experimental data was obtained for the Freundlich isotherm, while for AR dye the Langmuir isotherm shows the best agreement with the experimental data. The molecular structures of the two dyes are responsible for different adsorption behaviour.

The mechanism of the basic dyes adsorption on the ironsilicate beads

The dye adsorption process from aqueous solution onto porous adsorbents involves several steps: (i) transfer of dye from the bulk solution to the external surface of the liquid film surrounding the adsorbent particle; (ii) diffusion of dye through the liquid boundary layer to the external surface of the adsorbent bead,

C ₀ (g/L)	60	90	120	150		
	AB dye					
k _{i,1}	0.859	1.104	1.399	1.536		
\mathbb{R}^2	0.99	0.99	0.99	0.99		
ki,2	0.441	0.582	0.696	0.728		
R ²	0.99	0.95	0.99	0.99		
ki,3	0.06	0.084	0.151	0.2433		
R ²	0.99	0.99	0.99	0.99		
	AR dye					
k _{i,1}	0.856	1.139	1.485	1.766		
\mathbb{R}^2	0.99	0.99	0.99	0.99		
k _{i,2}	0.553	0.669	0.803	0.805		
R ²	0.99	0.99	0.99	0.99		
ki,3	0.213	0.237	0.300	0.316		
R2	0.99	0.99	0.99	0.99		
♠ 60 mg/I	= 90 mg/I	4.120 mg/I				

 Table 3

 RATE CONSTANTS OF THE

 INTRA-PARTICLE MODEL



Fig. 9. Plot of intraparticle diffusion kinetic model for the adsorption of AB basic dye on IS beads at various initial dye concentrations (T = 303 K, adsorbent dose 5 g/L).

known as external diffusion; (iii) adsorption of the dye on the sites localized at the external surface of the adsorbent (external sites); (iv) transfer of the dye from the external surface of adsorbent to the internal adsorption sites, known as internal diffusion or intraparticle diffusion; (v) adsorption on the surface of the interior sites [28, 29]. One or more of the abovementioned steps may control the rate at which the dye is adsorbed and the amount of dye that is adsorbed onto the adsorbent.

The intra-particle diffusion model was used to investigate the mechanism of the basic dyes adsorption on the ironsilicate beads and to predict the rate-limiting step for the adsorption of cationic dyes onto IS beads. According to the theory of Weber and Morris the intraparticle diffusion equation can be written as follows:

$$\mathbf{q}_{t} = \mathbf{k}_{id} \cdot \mathbf{t}^{1/2} + \mathbf{C}_{i} \tag{6}$$

where k_{id} (mg·g⁻¹ min^{-1/2}) is the rate constant of stage i (obtained from the slope of the straight line q_i versus $t^{1/2}$), C_i is the intercept which is proportional with the boundary layer thickness. The larger is the intercept the greater is the contribution of the surface sorption in the rate controlling step. If the plot of q_i versus $t^{1/2}$ gives a straight line which passes through the origin the sorption process is controlled only by intra particle diffusion. When the sorption process is controlled by more than



Fig. 10. Plot of intraparticle diffusion kinetic model for the adsorption of AR basic dye on IS beads at various initial concentrations (T = 303 K, adsorbent dose 5 g/L)

one mechanism then the plot of q_t versus $t^{1/2}$ will show multi linearity. According to our experimental data four stages are identified (fig. 9 and fig. 10): (1) the first sharper portion corresponds to the external surface adsorption or instantaneous adsorption stage. In this stage the external resistance to mass transfer plays a significant role. The increase of q, with time is steeply due to the fast coverage of external surface; (2) the second straight portion is attributed to the gradual adsorption stage, where the intraparticle diffusion is the rate limiting step. Since the plots don't pass through the origin (at each concentration investigated) the pore diffusion is not the sole rate controlling step; (3) in the third stage the solute molecules move slowly from larger pores into narrower pores causing a slowdown of the adsorption rate. The slopes of the straight lines corresponding to this portion of the plot are lower than those calculated for the straight lines from the previous linear portions, which indicate that the diffusion into narrow pores is the slowest adsorption step (the rate limiting step); (4) the fourth linear portion attributed to the final equilibrium stage indicates the saturation of the adsorbent surface.

The slope of the third portion of the plot (the rate limiting step) was defined as the intraparticle diffusion rate constant $k_{1,3}$. The values of k_1 increase with the increasing of initial dye concentrations for all three stages investigated (table 3), indicating the absence of micropores.

Regeneration and reuse of the adsorbent

In view of potential industrial applications, reuse is an important parameter to evaluate the efficiency of an adsorbent. The IYC beads deposited automatically on the bottom of the vial a few seconds after the stirring stopped) were easily separated from the dye solution. After each use the adsorbent was regenerated by calcination at 600°C for 4 h in order to eliminate the dye adsorbed around the active sites. Iron silicate beads were reused 20 times without any loss of adsorptive capacity.

Conclusions

Eco-friendly iron silicate beads synthesized by a multi templating method (using yeast cells and chitosan as pore generators) were used for the adsorption of cationic dyes Basic Blue 3 and Basic Red 22. This novel adsorbent was characterized by SEM, EDAX and water adsorption techniques. SEM and water adsorption prove the high porosity of the samples, while the EDAX analysis confirms the presence of Fe within the synthesized beads. Langmuir isotherm model fitted very well with the equilibrium data of AR dye, while the Freundlich isotherm showed the best agreement with the adsorption experimental data of ÅB dye. The intraparticle diffusion model was used to understand the sorption mechanism of the two basic dyes onto IYC beads. Due to their advantages (better accessibility, long lifetime) these green materials can be used as adsorbents.

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