MESOPOROUS SILICA-BASED SENSOR FOR COLORIMETRIC DETERMINATION OF BASIC YELLOW 28 DYE IN AQUEOUS SOLUTIONS

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The textile industry is one of the largest water-consuming industries in the world. The wastewater generated by the textile industry is a major source of pollution, containing mostly dyes, but also detergents, and heavy metals. Since dyes can have a negative impact on the environment, human and aquatic life, it is really important to find proper solutions for removal of these pollutants from wastewater. In addition, special attention is given to the discovery of new and fast “in situ” methods for identification and quantification of pollutants from wastewater. In this paper, SBA-15, mesoporous silica material, was used as sorbent for removal of Basic Yellow 28 from aqueous solutions. In addition, colored SBA-15 after sorption process was subjected to image analysis, to evaluate if it was possible to quantify sorbed dye on sorbent based on intensity of SBA-15 coloration. The obtained results revealed that SBA-15 could remove 99% of dye within 5 min. The highest efficiency of dye removal was at pH8, using 200 mg of sorbent. According to the Langmuir isotherm model, the theoretical maximum sorption capacity was 909 mg/g. Image-Pro software confirmed that it would be possible to quantify sorbed dye onto SBA-15 with accuracy of 0.98. Overall, SBA-15 demonstrated to be efficient sorbent in removal of Basic Yellow 28 from water, but also to be potential carrier as a sensor for detection of colored pollutants in water.

Keywords: SBA-15, dye sorption, Basic Yellow 28, image analysis

Introduction

Water pollution by colored industrial waste is one of the most serious social and environmental problems nowadays. The use of color is widespread in all developed industrial sectors: textile, plastics, food and pharmaceutical industries, which has led to a growing amount of colored wastewater. Hence, synthetic colors have become one of the biggest pollutants, affecting human health and the aquatic ecosystem equally. Even extremely small concentrations of dyes show visible coloration of the water, reducing the penetration of sunlight, thereby hindering the process of photosynthesis, and some of them are even toxic and carcinogenic to humans and marine life. The very strict laws on the removal of dyes from wastewater before its immersion into waterways, and difficult removal of low concentrations of dyes, have emerged new requirements for the development of technologies for wastewater purification [1,2].

Adsorption is found to be the most common method for purification of wastewater, due to high efficiency and easy processing [3,4]. Up to date, the most investigated adsorbents for removal of pollutants from wastewater are activated carbons [5], zeolites [6] and clays [7,8]. Recently, mesoporous silica has gained attention due to its high order structure, high specific surface area and good hydrothermal stability [9]. Due to outstanding properties, mesoporous silica has found application in many fields, such as catalysis [10], adsorbents [11,12], and sensors [13,14]. Among all investigated mesoporous silica, SBA-15 is particularly interesting for sorption processes because it has specific surface area above 800 m^2/g, which is significantly higher than commercial activated carbon [15,16]. So far, SBA-15 is demonstrated to be an efficient sorbent for various gases [17,18], metal ions [19,20], pesticides [21], and synthetic dyes [22–24].

In this paper, SBA-15 was used to evaluate its capacity to remove Basic Yellow 28 dye from aqueous solutions. In addition, Image-Pro software was used to analyze coloration of SBA-15 after sorption process, in order to determine if a detection and quantification of dye could be obtained without use of instrumental methods.

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Materials and methods

Reactants

SBA-15 was synthesized according to the previously published method [25]: SBA-15 samples were synthesized using Pluronic P123 (non-ionic triblock copolymer, EO20PO70O20, BASF) as a surfactant and tetraethoxysilane (TEOS, 98%) as a source of silica. The obtained SBA-15 has specific surface of 710 m^2/g. Basic Yellow 28 (BY28) was obtained by CHT-Bezema.

Monitoring of dye sorption

The sorption of BY28 was monitored through the analysis of the immersion solutions and through analysis of colored SBA-15 powder. An accurately weighed sample (0.2 g) of the mesoporous silica SBA-15 was added to 0.1 dm^3 of single-dye solution (BY28), with different initial concentrations (10-40 mg/dm^3). The sample glass bottles were shaken at a speed of 150 rpm for 5 minutes days at a constant temperature of 25 °C. The mixtures were centrifuged at 2000 rpm for 3 minutes with a Biofuge model centrifugation machine. The residual concentration of dye solutions was analyzed by UV–Vis spectrophotometer, where the maximum absorbance of the dye at λmax = 530 nm was measured, respectively. Various experimental parameters such as pH (2–8), adsorbent mass (0.01–0.2 g) and contact time (30 s, 1, min, 3 min and 5 min) were investigated.

The capacity of sorbed dye was calculated according to the following equation:

$$q_e = (C_0 - C_e)\frac{V}{m} \qquad (1)$$

where q_e (mg/g) is the amount of dye adsorbed on the membrane, C_0 (mg/dm^3) is the initial concentration of dye in solution, C_e (mg/dm^3) is the equilibrium concentration of dye in solution, V (dm^3) is volume of the used dye solution and m (g) is weight of the used membrane.

The percent of dye removal was calculated as follows:

$$ADS = \frac{C_0 - C_e}{C_0} \cdot 100\% \qquad (2)$$

The colored SBA-15 samples were dried and collected for further image analysis by Image-Pro software. The Image-Pro software was applied to determine the coloration intensity of SBA-15 and to compare the quantification of this parameter with amount of dye entrapped into SBA-15. Prior the image analysis, the colored SBA-15 samples obtained after sorption of Basic Yellow 28 and referent uncolored SBA-15 sample were scanned by HP Scan jet G311. The 24-bit color images of scanned samples were subjected to Image-Pro software for further analysis. In order to apply the image analysis method to these samples, the images were first converted into 8-bit grey scale images and then divided into three filter channels of primary colors: red, green and blue. Gray scale was used for separation into primary filter channels, because in that way every filter channel gives one intensity value for each pixel. In the case of 24-bit-colored images, three color intensity values are given for each pixel, which complicates the whole data processing. The pixel intensity at 50 different positions was collected from each image and corelate to initial dye concentration used for sorption, in order to estimate possibility to make calibration curve using only image analysis method.

Results and discussion

Influence of various parameter conditions on Basic Yellow 28 removal by SBA-15

The dependence of the contact time on the sorption capacity of Basic Yellow 28 dye by SBA-15 is presented in Figure 1. The initial concentration of the dye solution was 20 mg/dm^3, whereas the SBA-15 was exposed to the dye solution at a time range between 30 seconds and 5 minutes. Based on the presented results, it can be seen that the sorption reaction of Basic Yellow 28 dye onto SBA-15 occurs really fast (in less than 5 minutes), and that its capacity is independent of the contact period. The maximum achieved sorption capacity is 80 mg/g. The fast sorption time of other cationic dyes onto SBA-15 was reported previously, namely for the Basic Red 46 [26] (5 min), Basic Violet [27] (5 min) and Methylene Blue (10 min), whereas equilibrium time for sorption of anionic dyes was longer than in this study, for example for Reactive Red 2 and Safranin O was 30 min, and for Congo and Neutral Red 80 min [28].

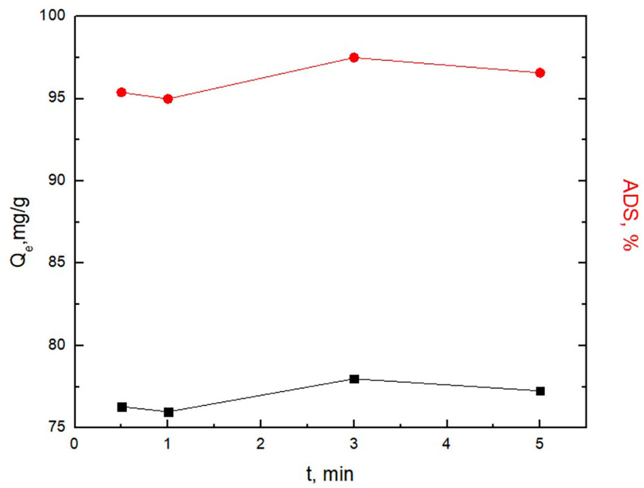

Figure 1. The influence of contact time on sorption capacity (■) and sorption percentage (●) of Basic Yellow 28 dye onto SBA-15. The initial dye concentration was 20 mg/dm^3 at pH 8, and mass of sorbent 0.2 g.

The influence of the mass of sorbent was tested at initial concentration of the dye solution 20 mg/dm^3 and pH 8 in a period of 5 minutes, varying the sorbent mass in the range of 0.01 g to 0.2 g. Figure 2 displays the effect of the sorbent mass on the sorption capacity of BY28 dye onto SBA-15 mesoporous silicates. An increase in sorbent mass leads to a linear increase in the sorption capacity of dye onto SBA-15. Maximum capacity of sorption and percentage of BY28 dye removal is achieved by using 0.2 g of absorbent and therefore during the presentation of other experiments this mass of sorbent was used. The maximum percentage of removal of BY28 is 96.57%.

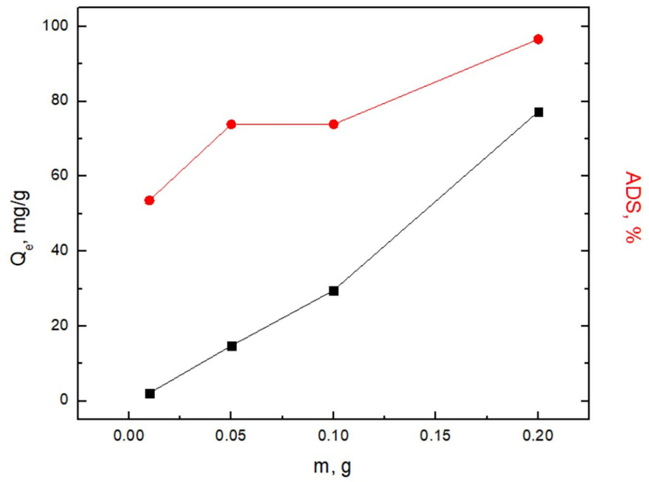

Figure 2. The mass sorbent influence on sorption capacity (■) and sorption percentage (●) of Basic Yellow 28 dye onto SBA-15. The initial dye concentration was 20 mg/dm^3, pH 8 and contact time of 5 min.

The pH of the dye solution can show a significant influence on the surface properties of the sorbent, as well as the ionization and dissociation of Basic Yellow 28 dye. The influence of pH on the sorption capacity of Basic Yellow 28 using SBA-15 mesoporous silicate is shown in Figure 3. Solutions of dyes with the initial concentration of 20 mg/dm^3 were in contact with the sorbent material (0.2 g) over a period of 5 minutes at a temperature of 25 °C. The results demonstrate that change in pH from acidic to alkaline nature slightly influences the increase in sorption capacity of Basic Yellow 28 onto SBA-15. Namely, sorption capacity varies from 67 (at pH 2) to 77 mg/g (at pH 8). It was reported in literature that sorption of cationic dyes onto SBA-15 rose with an increase of pH from 2 to 8 [26,29], while for the anionic dyes the opposite trend was detected [28]. The high sorption capacity of cationic dyes in alkaline environment was described by electrostatic and hydrogen bond interactions since SBA-15 is charged negative at high pH.

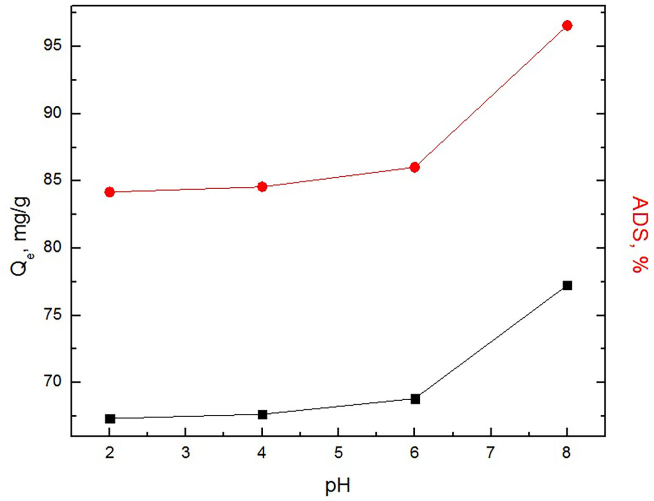

Figure 3. The pH influence on sorption capacity (■) and sorption percentage (●) of Basic Yellow 28 dye onto SBA-15. The initial dye concentration was 20 mg/dm^3, mass of sorbent 0.2 g and contact time of 5 min.

The effect of the initial concentration of the dye solution Basic Yellow 28 (range between 10 and 40 mg/dm^3) at pH 8 on the sorption capacity of SBA-15 was studied. The increase in the initial concentration of the Basic Yellow 28 dye solution has emerged an increased sorption capacity onto SBA-15. This result is the outcome of raised driving force of sorption within an increase of dye concentrations, thus leading to the faster diffusion of dye through SBA-15 and increased sorption capacity. The maximum sorption capacity is obtained when 40 mg/dm^3 of dye solution is used, and reaches a value of 153 mg/g. The obtained data were plotted through Langmuir and Freundlich mathematical models to determine the type of sorption. The linear forms of both models are presented by the following equations:

Langmuir model:

$$\frac{1}{q_e} = \frac{1}{q_{\max}} + \left(\frac{1}{q_{\max} K_L}\right)\frac{1}{C_e} \qquad (3)$$

Freundlich model:

$$\ln q_e = \ln K_F + \frac{1}{n}\ln C_e \qquad (4)$$

where q_e (mg/g) is sorption capacity, C_e (mg/dm^3) the equilibrium dye concentration in solution, q_{max} (mg/g) the theoretical maximum sorption capacity, K_L Langmuir adsorption constant related to the free energy constant, K_F (dm^3/g) represents Freundlich constant for the sorption capacity and n is parameter related to the intensity of sorption.

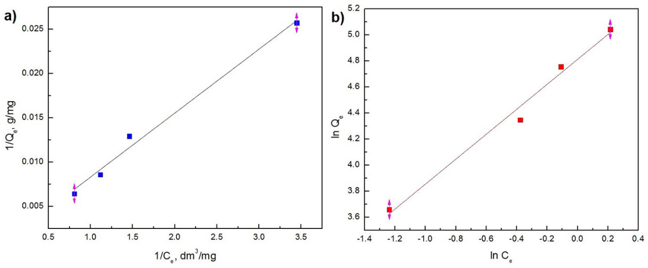

Figure 4. a) Langmuir and b) Freundlich isotherms for the sorption of Basic Yellow 28 onto SBA-15.

Table 1. Isotherm parameters.

Langmuir isotherm			Freundlich isotherm		
R^2	K_L, dm^3/g	Q_{max}, mg/g	R^2	n	K_F, dm^3/g
0.98	0.152	909	0.98	1.04	119.7

The calculated parameters from obtained Langmuir and Freundlich plots (Figure 4) are presented in Table 1. The linear correlation coefficients implies that the Langmuir isotherm model has a better agreement with the experimental data and is more suitable model for describing the equilibrium of the sorption of Basic Yellow 28 on SBA-15 mesoporous silicate in the observed range of concentrations. In accordance with Langmuir's model, sorption process occurs as a monolayer coverage of the material surface with the investigated dye molecules and the interaction between neighboring molecules is negligible. The Langmuir isotherm model represents a chemisorption process. According to the Langmuir model, the maximum theoretical sorption capacity of Basic Yellow 28 on SBA-15 mesoporous silicate is 909 mg/g. Comparing the obtained results in this study with the data from literature, it can be concluded that SBA-15 is more efficient sorbent than activated carbon (769.23 mg/g) [30], activated carbon from persea species (400 mg/g) [31], clinoptilolite (59.6 mg/g) [32], recycled newspaper pulp (91.21 mg/g) [33], reed (181 mg/g) [34].

Image analysis

The colored SBA-15 samples collected after sorption (influence of different concentrations test) were scanned and analyzed using Image Pro software, to evaluate the accountability of this software for determination of concentrations of sorbed dye onto SBA-15. The conversion of colored samples into gray scale and dependence of the average intensity in the different primary filter channels within different initial dye concentration are presented in Figure 5. The average pixel intensity was calculated from 50 different positions along the diameter of the colored samples. Gray scale goes from 0 to 255, where 0 represents total black, and 255 total white.

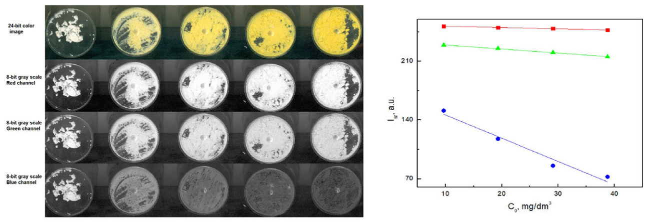

Figure 5. The conversion of 24-bit colored image of SBA-15 samples into 8-bit gray scale with 3 primary filter channels (R ■, G▲ and B●) and the influence of different initial concentrations of dye on average pixel intensity of colored SBA-15.

As it can be seen from Figure 5, the visual color intensity of SBA-15 increases with an increase of initial dye concentration from 10 to 40 mg/dm^3 (left figure). On the other side, the average pixel intensity decreases with increased initial concentration of the dye solution, which is in line with the expected results. Namely, lower values of pixel intensity correlate with darker samples, whereas higher values of intensity correlate to the brighter samples. The obtained linear dependence between average intensity and dye concentrations is obtained with the correlation coefficient R^2=0.98. These results indicate that concentration of colored pollutants from wastewater can be predicted by Image-Pro software and without use of instrumental methods, just based on the intensity of sorbent coloration after exposure to the dye wastewater.

Conclusion

SBA-15, as a representation of mesoporous silica, was used for sorption studies of Basic Yellow 28 in aqueous solutions. The influence of different dosages of sorbent, contact time, pH and concentration on sorption capacity was investigated. It was shown that SBA-15 had the highest efficiency in alkaline conditions. The sorption of Basic Yellow 28 occurred fast, in 5 minutes, with 99% removal of dye from water. After plot of experimental data through isotherm models, it was concluded that Langmuir model fits better, and that Basic Yellow 28 dye was chemically sorbed onto SBA-15. In addition, colored SBA-15 was tested by image analysis. The average pixel intensity was estimated for different colored samples, and it was confirmed that it is possible to obtain calibration curve and quantify dye on sorbent by coloration intensity. Hence, SBA-15 represents the promising candidate as a sorbent and sensor for fast and efficient removal of dyes from water.

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Izvod

SENZOR NA BAZI MEZOPOROZNE SILIKE ZA KOLORIMETRIJSKO ODREĐIVANJE BOJE BASIC YELLOW 28 U VODENIM RASTVORIMA

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Tekstilna industrija je jedna od najvećih potrošača vode u odnosu na druge industrijske grane. Otpadne vode iz tekstilne industrije su jedan od glavnih izvora zagađenja i uglavnom sadrže boje, ali takođe i deterdžente i teške metale. Pošto boje mogu imati negativan uticaj na životnu sredinu, ljude i vodeni svet, važno je pronaći odgovarajuća rešenja za uklanjanje ovih zagađivača iz otpadnih voda. Pored toga, posebna pažnja je posvećena otkrivanju novih i brzih „in situ" metoda za identifikaciju i kvantifikaciju zagađivača iz otpadnih voda. U ovom radu kao sorbent za uklanjanje Basic Yellow 28 boje iz vodenih rastvora korišćen je SBA-15, mezoporozni silika materijal. Pored toga, obojeni SBA-15 nakon procesa sorpcije podvrgnuti su analizi slike, kako bi se procenilo da li je moguće kvantifikovati sorbovanu boju na sorbentu na osnovu intenziteta obojenosti SBA-15. Dobijeni rezultati su pokazali da SBA-15 može ukloniti 99% boje u roku od 5 minuta. Najveća efikasnost uklanjanja boje bila je pri pH 8, korišćenjem 200 mg sorbenta. Prema Langmuirovom modelu, teoretski maksimalni kapacitet sorpcije bio je 909 mg/g. Softver Image-Pro je potvrdio da je moguće kvantifikovati boju na SBA-15 sa tačnošću od 0,98. SBA-15 se pokazao kao efikasan sorbent u uklanjanju Basic Yellow 28 boje iz vode, ali i kao potencijalni senzor za detekciju obojenih zagađivača u vodi.

Ključne reči: SBA-15, sorpcija boja, Basic Yellow 28, analiza slike