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EVALUATION OF DYNAMIC ADSORPTION OF DYES FROM AQUEOUS SOLUTIONS BY APPLYING MATHEMATICAL MODELLING WITH THE VS2TDI PROGRAM AND EXPERIMENTAL STUDIES

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Abstract. Dyes, heavy metals, sulphates, chlorides, petroleum products and other substances can be found in industrial wastewater. Synthetic dyes are divided into different groups according to their chemical composition. It is known that synthetic dyes have toxic and mutagenic properties. Adsorption is considered a reliable method for the removal of various pollutants. The adsorbent synthesized from paper waste, which has a lot of air gaps in its composition, is called aerogel. The maximum sorption capacity reached the value of 0.25–0.28 and 0.12–0.16 mg/g. It is characterized by low density and large surface area. Evaluation of dynamic adsorption of dyes from aqueous solutions was made by using the VS2TDI mathematical modelling program.

Keywords: adsorption, circular economy, textile industry, synthetic dyes, wastewater.

Introduction

Providing clean water is extremely important issue that has received much attention (Rahman et al., 2021, p. 2). Domestic and agricultural activities generate various effluents containing different contaminants and release them into the environment (Ardila-Leal et al., 2021, p. 2). Numerous contaminants could be found in textile wastewater. Dyes, heavy metals, petroleum products, sulphur, nitrates were linked to adverse health effects (Slama et al., 2021, p. 3). Those materials have low degradability and detrimental impacts on animal and human health. Dyes are one of the most critical contaminants produced in various industries like plastic, cosmetics or food industry, paper or textile (Wei et al., 2020, p. 5). The textile industry is one the biggest consumers of water in the globe. Synthetic dyes are non-biodegradable, bio-accumulative products and toxic materials which are the origin to considerable potential threat to the safety of the environment (Kishor et al., 2021, p. 7). Dyes are stable and resistant to microbial damages that is why elimination them from textile wastewater using conventional methods has become a major challenge for scientists (Samsami et al., 2020, p. 4). According to the ionic charge carried by artificially synthetic dye structures, there are three main classes of synthetic dyes: disperse dyes (non-ionic), acids dyes (anionic) and basic dyes (cationic). The last group is considered to be more toxic than the other two classes (Elmegied et al., 2021, p. 3).

There are mane techniques to remove the contaminants from wastewater (Musa & Idrus, 2021, p. 1). Although, there is no textile wastewater treatment method which is called the best. Each method has its advantages and disadvantages. Classical methods for the treatment of textile effluents are ion exchange, membrane filtration, photocatalytic process, electrochemical and chemical oxidation, biological methods (Zhang et al., 2021, p. 2). Adsorption was considered one of the most promising methods for removing contaminants from textile wastewater. Adsorption has various benefits such as great performance, low cost, flexibility and no sludge production (Sočo et al., 2021, p. 2). Numerous adsorbent materials, including nanoparticles, agricultural wastes, industrial or urban wastes, clay minerals or natural polymers (e.g., chitin, chitosan, alginate, etc.), activated carbon or even coconut charcoal could be used in the adsorption process (El-Zawahry et al., 2021, p. 2). The use of natural polymer-based adsorbents has received recently much attention in removing various contaminants from wastewater (Esmaeili et al., 2021, p. 2).

Furthermore, many parameters which affect the adsorption process were identified, including dosage of adsorbent, adsorption contact time, pH, pollutant concentration (Malatji et al., 2021, p. 3). As a result, scientists

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are constantly looking for new materials that could be cheap, ecological, and environmentally friendly. In this case, agricultural or other waste is often used as adsorbents.

Adsorption is a physical treatment method in which dissolved molecules in wastewater are chemically and physically bonded to the adsorbent surface (Yang et al., 2021, p. 3). Because of higher pollutant removal efficiency, adsorption technology was described as a potential method for pollutants removal from wastewater. Adsorption is known for being a cost-effective method (Guo et al., 2018, p. 2). The process of dyes adsorption could be divided into three main steps. The first one involves molecules diffusion or convection. The second is about the molecules of dyes diffusion through a diffusion boundary layer. The third step describes the molecules diffusion from the surface into the adsorbent material interior part. The concentration of dye may affect the second step. The last step could entail two phenomena: the first one is called porous diffusion, in which the adsorbate diffuses through the liquid and fills the pores of adsorbent. In this case adsorbate could be adsorbed. The next phenomena is related to the surface diffusion, in which the adsorbate is first adsorbed and then diffused from one site to another.

To sum up it could be stated that adsorption is largely implemented in the wastewater treatment are due to its elevated removal efficiency, small operating cost and simplicity of the application process. The scientists have been focused to find new materials to remove coloured substances from wastewater. Many commercial adsorbents have been utilized to minimize the toxic dye concentrations from wastewater (Guo et al., 2018, p. 3). A little-studied adsorbent, aerogel, which is a solid aggregate material with low density, is believed to be useful for the removal of dyes from textile wastewater (Luo et al., 2021, p. 3). Aerogel has a high surface area and it is very lightweight (Liang et al., 2020, p. 2).

The goal of this experimental research falls within the framework of environmental protection. The aerogel, as an adsorbent, was used for removing synthetic dyes from the water. This work is subdivided into two parts: the first one is dedicated to an experimental study and the second one is devoted to a theoretical-modelling study.

In the experimental part, effects of several parameters, including the dye concentration, adsorbent dosage, pH value of the solution and temperature on the dye removal from aqueous solution were studied.

Adsorption kinetics describes the course of the process reaction, the time. The models indicate the dependence of the sorption process on the physical and chemical properties of the adsorbent material, which also affect the adsorption mechanism (Xue et al., 2023, p. 2).

A model is a simplified description/representation of a more complex entity or process. It is often used to simplify and analyse a complex subject, usually focusing on one or more aspects of the subject being studied (Kheradmand et al., 2022, p. 3). We use models to better understand mechanisms of action, explore alternatives, model, simulate, evaluate or validate (Ayawei et al., 2017, p. 6).

In practice, engineers use many different models to study various aspects of the thing being modelled, such as input data, process behaviour or progress, process duration, reliability, performance, quality, usability (Al-Ghouti & Razavi, 2020, p. 5). The examples help to gain a better understanding of processes that occur in the environment. Modelling of environmental processes and contaminant transport is important for the modern scientist and engineers.

The use of computer modelling programs in practice facilitates the examination of environmental processes and helps to predict the outcomes of the processes (Bujdak, 2020, p. 3). Since large-scale research, sample collection and analysis are expensive, the application of computer models can be significantly cheaper and, in most cases, faster, to obtain reliable process data and results.

The accuracy of the output of each model is directly proportional to the accuracy of the input data. The more initial input data is provided, the more accurate the predictive results of the investigated process or phenomenon are.

In the theoretical modelling part, the VS2TDI modelling program was used in order to determine the effective contact time. This program is designed to simulate prevalence of pollution. The use of this program makes it possible to reduce research costs, obtain reliable results and assess the further spread of pollutants. The program also allows you to change several parameters and monitor ongoing changes. It was studied how long it takes to filter aqueous solutions through an adsorbent of different thicknesses. The aim of this study is to evaluate the dynamic adsorption of dyes from aqueous solutions by applying mathematical modelling with the VS2TDI program and experimental studies.

1. Materials and methods

1.1. Materials

All chemical products used in the preparation and the adsorption studies were of analytical grades. For the production of the aerogels, office paper was produced by The Navigator Company (Portugal), brownish 3-ply corrugated cardboard was purchased from UAB Dekpaka (Lithuania), and MoTip polyester resin (MOTIP DUPLI Group, Wolvega, The Netherlands) were used as base materials.

The dyes particles used in this experimental work are a commercial product and were supplied by Sigma-Aldrich. Methylene blue is a heterocyclic aromatic chemical compound with the molecular formula $C_{16}H_{18}ClN_3S$. The dye is widely used in biology and chemistry. At room temperature, methylene blue appears as a solid, odourless dark green powder that dissolves in water to form a blue solution (Kayabaşı & Erbaş, 2020, p. 137). The hydrated form has 3 water molecules per molecule of methylene blue. Methylene blue can damage red blood cells and reduce the blood's ability to carry oxygen. The dye has antioxidant properties. It is well soluble in water, chloroform, ethanol and glacial acetic acid.

Congo red is a dye belonging to the group of azo dyes. The molecular formula of the dye is $C_{32}H_{22}N_6Na_2O_6S_2$. It is a water-soluble substance, when the dye dissolves it forms a red colloidal solution; its solubility in organic solvents is better than in water. Due to its colour change from blue to red at pH 3.0–5.2, Congo red can be used as a pH indicator.

Rhodamine B is often used to determine the speed and direction of flow. The molecular formula of the dye is $C_{28}H_{31}ClN_2O_3$. Rhodamine B is also widely used in fluorescence microscopy, flow cytometry, fluorescence correlation spectroscopy etc (Awoniyi et al., 2021, p. 7). Rhodamine B can exist in equilibrium between two forms: open (fluorescent form) and closed (non-fluorescent Spiro lactone form). In the acidic state, the "open" form predominates, while in the basic state, the "closed" form is colourless. The fluorescence intensity of Rhodamine B decreases with increasing the temperature.

Naphthol green B is a coordination iron complex used as a dye. Its molecular formula is $C_{30}H_{15}FeN_3Na_3O_{15}S_3$. The dye is soluble in water. Naphthol green B is used in histology to stain collagen, as well as wool, nylon, paper and soap (Gunasundari et al., 2020, p. 359).

1.2. Preparation of aqueous solutions of dyes

All chemicals were used as received without further purification. For all experiments, deionized water was used to prepare the aqueous solutions.

The concentration of solutions of dyes was 10 mg/L. The pH values of the solutions were fixed between 6 and 7 continuously agitated at room temperature of 23 °C. The solutions of dyes were prepared using aqueous solutions of 0.1 N H_2SO_4 and 0.1 N NaOH. Solutions of each dye (congo red, naphthol green B, rhodamine B and methylene blue) were made individually.

1.3. Filtration test

In order to investigate the nature of the sorption process, a series of studies on the removal of dyes on aerogels were carried out. The adsorption experiments of naphthol green B, congo red, methylene blue and rhodamine B were carried out using a column. The height of the column was 50 cm, diameter 4.6 cm. Only one aerogel plate with a height of 1 cm was used in the experimental research.

Filtration of prepared solutions (100 mL) used in the textile industry through the column was carried out at two speeds: the first filtration test was done by pouring the dyes solution a little at a time, the speed of filtration was 10 mL/min., and the second filtration was done by pouring the entire volume of the solution (100 mL) at once, the speed of filtration was 30 mL/min.

The mass of the adsorbent used in the study was 2 g. The maximum sorption capacity of aerogels is a physical property of the aerogel to absorb maximum amount of liquid into its pores at any given time.

First of all, aerogels samples were weighed (m_0). Then aerogels samples were immersed in solutions of dyes (congo red, naphthol green B, rhodamine B and methylene blue) during the filtration test. The bottom layer of the column was filled with gravel. The upper layer of the column was filled with adsorbent (disc of aerogel). In order to leave the aerogel under the solution, the grid was added. The samples were then taken out from the solutions and drained for 1 min and weighed again (m_1). The maximum sorption capacity of the solutions of dyes was calculated as follows:

$$Q = \frac{m_1 - m_0}{m_0}, \tag{1}$$

where: m_0 – the mass of aerogel before filtration test, m_1 – the mass of aerogel after filtration test, Q – maximum sorption capacity, mg/g.

Triplicate experiments were performed and the average values were taken into consideration.

1.4. Effect of the thickness of adsorbent

The VS2TDI computer program is designed to model the spread of pollution. It was developed by scientists Paul A. Hsiekh, William L. Wingle and Richard W. Healy of the CCU.S. Geological Survey's. The program consists of two separate parts: a preprocessor and a postprocessor. The first on is dedicated to the information about the conditions of the task. It is necessary to create input data about adsorbent properties, concentration, limitations, background pollution. The second part is devoted for displaying the data graphically.

The movement of pollutants in soil and water takes place under the influence of convection, molecular diffusion and mechanical dispersion. This phenomena is complicated by physical, chemical and biological processes. Various physical and chemical reactions related to mass transfer in soils and water are very complex and difficult to process. In order to better understand these processes, they need to be modelled.

The maximum sorption capacity of the solutions of dyes was calculated via the filtration method. In order to find out how long it would take to filter aqueous solution through a adsorbent of different thicknesses the calculations with VS2TDI were performed.

It is necessary to create the area (domain) of adsorbent and filtration column: the aerogels, which were used in experimental research, consisted of round shape disc.

The height of the adsorbent is 1 cm, the diameter of the disc is 5 cm. Adsorbent, synthesized from paper waste, is composed of many cellulose fibers. The structure of aerogel could be seen in Figure 1.

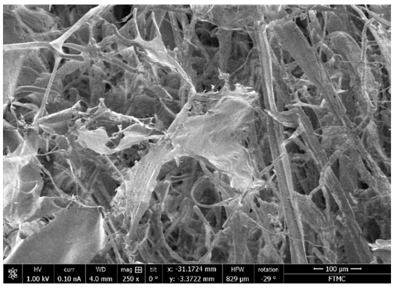

Figure 1. Surface area of aerogel captured before sorption process with SEM Helios Nanolab 650, 100 µm

Scanning electron microscopy (SEM) was used to visualize the morphology of the cellulose aerogel. The unique internal structure of the prepared sorbent is expected to improve the properties of the absorption process. As can be seen in Figures 1, the aerogel is composed of many fibers with angular edges. This surface property improves sorption properties because the surface area takes up more space.

The parameters of the column were as follows: the height was 3 meters, the width was 20 centimeters. The diameter of the column was increased almost four times in order to better see the changes in model. It was decided to use 4 different thicknesses of adsorbent filler: 1 cm, 10 cm, 50 cm and 100 cm. The liners of the filled columns with aerogel are provided in Figure 2.

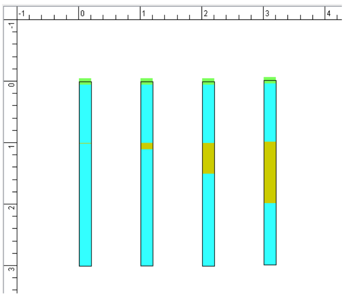

Figure 2. The columns are filled with a different thickness layer of aerogel

After this it is necessary to adjust the model's boundary conditions: initial, maximum and minimum time steps should be added. It is also necessary to create the grid and add the value of initial concentration of solution. It is important to mention that one type of solution was used in the simulation, which represented the solution of dye.

2. Results

The research results are divided into two parts: the data from experimental research are given in Table 1 and the results from modelling are given in Table 2.

Results of calculations of maximum sorption capacity of the dyes are given in Table 1.

Table 1. The maximum sorption capacity of aerogels

Dye	m_0, g	m_1, g	Q, mg/g
The first filtration test (10 mL/min)			
Congo red	2	2.54	0.27
Naphthol green B	2	2.52	0.26
Rhodamine B	2	2.49	0.25
Methylene blue	2	2.55	0.28
The second filtration test (30 mL/min)			
Congo red	2	2.32	0.16
Naphthol green B	2	2.25	0.12
Rhodamine B	2	2.28	0.14
Methylene blue	2	2.30	0.15

It could be seen that after adsorption process the values of aerogels mass have changed: in all cases the values of mass have increased from 2 g to 2.49–2.55 g during the first filtration test and from 2 g to 2.25–2.32 g during second filtration test. Meanwhile the maximum sorption capacity reached the value of 0.25–0.28 and 0.12–0.16 mg/g accordingly. It can be observed that the longer the solution is in contact with the adsorbent, the more the adsorbent is absorbed by the volume of the solution.

In the Figure 3, it can be seen that the spaces between the cellulose fibers have decreased, the surface has become smoother. It is likely that the adsorption capacity should decrease if the sorbent is used several times.

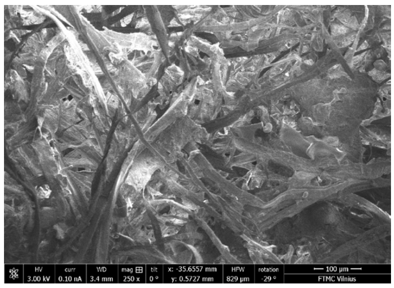

Figure 3. Surface area of aerogel captured after sorption process with SEM Helios Nanolab 650, 100 µm

It is believed that keeping the adsorbent in solution for a longer time would flatten the surface even more.

In order to determine the time it takes for the dye solution (100 mL) to filter through an aerogel filler of different thicknesses, simulations were performed using VS2TDI computer program.

During the first filtration test, the filtration time through the adsorbent varied from 140 seconds to more than 10 000 seconds. During first test of filtration modelling it was difficult to determine the filtration time (sec.), because it was reached the maximum value of modelling

Table 2. Filtration time using different thickness of aerogel

Thickness of aerogel, cm	Filtration	Filtration time, sec
1	First test	140
	Second test	120
10	First test	>10 000
	Second test	340
50	First test	>10 000
	Second test	430
100	First test	>10 000
	Second test	560

time. By slowly adding the dye solutions, the filtrate very slowly filtered through the adsorbent layer made from discs of aerogel. The uptake efficiency of an adsorbent and the extent of the contaminant's adsorption largely depend on its specific surface area and porosity. An adsorbent, aerogel, possessing a high surface area bears greater active surface sites resulting in higher adsorptive competency.

During the second filtration test, the filtration time through the adsorbent varied from 120 seconds to 560 seconds. As the thickness of the adsorbent increased, the filtration time also increased. The difference between 1 cm and 10 cm of adsorbent filler is not great. Although the difference between 1 cm and 50–100 cm is quite significant. When planning experimental studies with a large amount of adsorbent, a longer duration of the sorption process should be planned.

Conclusions

During experimental research it was found out that the longer the solution is in contact with the adsorbent, the more the adsorbent is absorbed by the volume of the solution. The spaces between the cellulose fibers have decreased after the adsorption process. The surface area of aerogel has become smoother. The maximum sorption capacity reached the value of 0.25–0.28 mg/g during the first filtration test and 0.12–0.16 mg/g during the second filtration test. After adsorption process the values of aerogels mass have changed: in all cases the values of mass have increased from 2 g to 2.49–2.55 g during the first filtration test and from 2 g to 2.25–2.32 g during second filtration test.

The thicker the column packing is, the longer the filtration took time. Then the filtration speed was 30 mL/min, the filtration time took place from 120 sec. (when the thickness of adsorbent was 1 cm) to 560 sec. (when the thickness of adsorbent was 100 cm). After creating the models it should be easier to plan the experimental studies: if it is planned the experimental research with a large amount of adsorbent or the smaller speed of filtration, a longer duration of the sorption process should be planned.

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