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Artificial neural network optimization for methyl orange adsorption onto polyaniline nano-adsorbent: Kinetic, isotherm and thermodynamic studies

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ABSTRACT

This study aims to synthesize a polyaniline nano-adsorbent and use it to adsorb methyl orange dye from the aqueous solution. The average particle size of nano-adsorbent was about 70 nm. The effects of various parameters have been analysed, as are pH, temperature, adsorption time, initial concentration and adsorbent dosage, and they were optimized by an artificial neural network model. The multilayer feed forward neural network with five inputs and one output has been trained with eight neurons in the hidden layer. A comparison of the experimental data with the dye adsorption efficiency predicted by the artificial neural network model showed that this model can estimate the behavior of the adsorption process of methyl orange dye on the polyaniline nano-adsorbent under different conditions. The study yielded the result that dye adsorption capacity of the nano-adsorbent increased from 3.34 to 32.04 mg/g and from 3.28 to 30.28 mg/g as the dye initial concentration was increased from 10 to 100 mg/L, at 65 °C and 25 °C, respectively. Also, dye adsorption equilibrium was achieved in 60 min for methyl orange dye. The adsorption kinetics was studied based on pseudo-first-order, pseudo-second-order, intraparticle diffusion and Elovich models. The results showed that the adsorption data at all levels of initial concentration have the best consistency with the pseudo-second order equation. Furthermore, two-parameter isotherm models (Langmuir, Freundlich and Temkin) and three-parameter isotherm models (Hill, Sips and R-P) were used to ascertain the nature of the adsorption isotherm. Based on this study, the maximum adsorption capacity was estimated to be 75.9 mg/g. Thermodynamic studies indicated that methyl orange dye adsorption was endothermic on the polyaniline nano-adsorbent.

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1. Introduction

Many factories and industries, such as textile, paper making, food processing, leather, and plastics and rubber use synthetic organic dyes in their various processes. The dyes discharged into the environment through wastewater have undesirable effects on living organisms [1,2]. This material is toxic and hazardous and can cause irreparable illnesses and diseases, such as cancer and genetic mutation in humans and other creatures [3,4]. Therefore, the dyes must be removed from different industrial wastewaters in order to prevent various possible damages to living organisms and to reduce soil and water pollution.

Over the years, several methods have been used to conduct dye containing wastewater treatment, as are photocatalytic degradation of dyes [5], nanofiltration [6], degradation of dyes with microorganisms [7] and adsorption [8–12]. Due to lower prices, simpler design and operation, and high dye removal efficiency, the adsorption process is highly considered compared to other methods of water and wastewater treatment [13,14]. Recently, several adsorbents have been used for removing dyes from water and wastewater, as are activated carbon produced by chitosan flakes for removal of methylene blue dye [15], iron composite nanoparticles for uptake of congo red dye [3], waste pea shells for removal of malachite green [1],polyaniline/titanium dioxide nanocomposite and polypyrrole/titanium dioxide nanocomposite in the removal of congo red dye [16], TLAC/Chitosan composite for removal of crystal Violet [17], polyacrylonitrile/activated carbon composite for removal of acid Yellow 99 [18] and Hexagonal shaped nanoporous carbon for adsorption of methyl orange [19].

One of the most important conductive polymers is polyaniline, which has many advantages, such as low cost, ease of synthesis, stability

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against air and moisture and high efficiency for the removal of pollutants [20,21]. This conductive polymer has wide applications in various fields including anti-corrosion coatings [22], sensors and biosensors [23], supercapacitors [24] and also the removal of various pollutants from water and wastewater [25,26].

Methyl orange dye is regarded as one of the harmful dyes for aquatic ecosystems and human life [27]. Robati et al. used graphene oxide for the simultaneous removal of methyl orange dyes and BR-12 from the aqueous phase. This study analysed the effect of various parameters, such as pH, adsorption time, initial concentration of dye solution and temperature. The optimum time for the adsorption process is 100 min and the maximum adsorption capacity of methyl orange dye is obtained at pH = 3, 16.83 mg/g [28]. Furthermore, Haldorai et al. used chitosan/MgO composite for removing methyl orange dye. In their study, the maximum adsorption capacity was calculated as 60 mg/g [29].

The artificial neural network (ANN) is regarded as one of the most powerful tools to predict the adsorption process [30], which is rarely used to model the process of dye adsorption from water and wastewater. Ghaedi et al. used artificial neural networks to predict the adsorption of methyl orange dye by gold nanoparticles loaded on activated carbon and Tamarisk. They studied and modeled the effect of four parameters, namely, initial concentration of dye solution, adsorbent dosage, contact time and stirrer speed, on the efficiency of dye adsorption. A comparison between the experimental results with the adsorption efficiency predicted by artificial neural networks indicated that the artificial neural network is a suitable predictor to estimate the behavior of methyl orange dye adsorption on gold nanoparticles loaded on activated carbon and Tamarisk under different experimental conditions [31].

In the present study, artificial neural networks were used to predict methyl orange dye adsorption on polyaniline nano-adsorbent. The effect of five parameters, i.e., pH, adsorbent dosage, adsorption time, initial concentration of dye solution and temperature, on the efficiency of methyl orange dye adsorption was analysed, and the data were modeled using artificial neural networks. Moreover, kinetics, isotherms and thermodynamics of methyl orange dye adsorption on the polyaniline nano-adsorbent were studied.

2. Materials and methods

2.1. Materials

The materials used in this study include aniline monomer, ammonium peroxodisulfate, sodium dodecylbenzenesulfonate, sulfuric acid, sodium hydroxide and methyl orange pigments, which are all laboratory products bought from Merck Company in Germany. Prior to the synthesis process, aniline monomer was distilled once. The deionized distilled water was used to wash and prepare the acid, alkali and dye solutions.

2.2. Synthesis of polyanilinenano-adsorbent

In order to prepare the polyaniline nano-adsorbent, 2.5 g ammonium peroxodisulfate oxidant were added to 25 mL of distilled water and the solution was stirred by a magnetic stirrer for 15 min. Then, the solution containing the stabilizer, i.e., 0.4 g of sodium dodecylbenzenesulfonate in 25 mL of distilled water, was added to the solution. Subsequent to 15 min, 50 mL of 2 M sulfuric acid solution were added to the solution in order to reach a final volume of 100 mL. The solution was stirred for 15 min with a magnetic stirrer. In the next step, 1 mL of once-distilled aniline monomer was added to the solution drop by drop. By adding aniline monomer, the solution color changed indicating the beginning of the polymerization. After 5 h, the final solution was filtered, and the polymer was then rinsed several times by distilled water and dried in an oven at 40 °C in order to remove oligomers and impurities in the synthesized polymer. Finally, the polymer was powdered in order to be used as a sorbent for methyl orange dye adsorption.

2.3. Methyl orange dye adsorption runs

A study was conducted on the effect various parameter, namely, pH, adsorbent dosage, temperature, initial concentration of dye solution and adsorption time, have on the efficiency of methyl orange dye adsorption on the polyaniline nano-adsorbent. The ranges analysed of pH, adsorbent dosage, temperature, initial concentration of solution and adsorption time were 2–10, 0.02–0.15 g, 25–65 °C, 10–100 mg/L and 1–60 min, respectively. Furthermore, the volume of dye solution in all experiments was 50 mL and the stirring speed 1200 rpm. Sulfuric acid solution and sodium hydroxide solution were used to change the dye solution pH. Visible light spectroscopy at a maximum wavelength of 464 nm was used to measure the dye concentration. In order to calibrate the device, a number of standard samples with concentrations of 0.25–10 mg/L were prepared and adsorption values for each concentration were measured by the device. Using the values of the standard solution concentration and their corresponding adsorption values, a calibration line was drawn. In each experiment, 50 mL of dye solution of given initial concentration, pH and temperature were stirred and a certain amount of adsorbent was added. The solution was stirred for the time required for dye adsorption. The resulting solution was filtered and the final concentration was measured by visible light spectroscopy. Dye adsorption efficiency was calculated by the following equation:

$$(\%) = \frac{C_i - C_f}{C_i} 100 \tag{1}$$

where C_i and C_f (mg/L) are the initial concentration of dye solution, and after the adsorption process, respectively.

Furthermore, q_e and q_t (mg/g) are the amount of methyl orange dye adsorbed on the polyaniline nano-adsorbent in equilibrium and at time t, respectively, which are obtained by the following equations:

$$q_t = (C_i - C_t)\frac{V}{m} \tag{2}$$

$$q_e = (C_i - C_e)\frac{V}{m} \tag{3}$$

where, C_e (mg/L) is the concentration of methyl orange dye in equilibrium and C_t (mg/L) the concentration of methyl orange dye at time t.

3. Results and discussion

3.1. Morphology and chemical structure of the nano-adsorbent

The morphology of the synthesized nano-adsorbent was studied by scanning electron microscope (SEM). Fig. 1a and b show the SEM images of the pure polyaniline and the polyaniline nano-adsorbent, respectively. As observed, the pure polyaniline is made up large size particles with an agglomerated structure, while the polyaniline nano-adsorbent has size-uniform 70 nm particles. The presence of sodium dodecylbenzenesulfonate in the synthesized environment caused the generated nano-particles to be small. Stabilizers can generate chemical binding with the polymer or can be physically absorbed by polymer chains, and so they do not allow an excessive growth of the polymer chains. As a result, they prevent the aggregation of particles during polymerization and products with more uniform and smaller particles are formed. The smaller size of the particle increases the contact area, thus increasing the efficiency of pollutant adsorption. Fig. 1c shows the SEM images of the polyaniline nano-adsorbent after methyl orange adsorption. As observed the adsorbent surface has changed and particles are stuck together and their pores filled.

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