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Research article

Optimization and modeling of methyl orange adsorption onto polyaniline nano-adsorbent through response surface methodology and differential evolution embedded neural network

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ABSTRACT

Presence of pigments and dyes in water bodies are growing tremendously and pose as toxic materials and have severe health effects on human and aquatic creatures. Treatments methods for removal of these toxic dyes along with other pollutants are growing in different dimensions, among which adsorption was found a cheaper and efficient method. In this study, the performance of polyaniline-based nano-adsorbent for removal of methyl orange (MO) dye from wastewater in a batch adsorption process is studied. Along with this to minimize the number of experiments and obtain optimal conditions, a multivariate predictive model based on response surface methodology (RSM) is developed. This is compared with data-driven modeling using the artificial neural network (ANN) which is integrated with differential evolution optimization (DEO) for prediction of the adsorption of MO. The interactive effects on MO removal efficiency with respect to independent process variables were investigated. The fit of the predictive model was found to good enough with $R^2 = 0.8635$. The optimal ANN architecture with 5-12-1 topology resulted in higher R^2 and lower RMSE of 0.9475 and 0.1294 respectively. Pearson's Chi-square measure which provides a good measurement scale for weighing the goodness of fit is found to be 0.005 and 0.038 for RSM and ANN-DEO respectively, and other statistical metrics evaluated in this study further confirms that the ANN-DEO is very superior over RSM for model predictions.

1. Introduction

Water pollution is considered as one of the most important concerns of human society in recent years. Most process industries discharge their wastewater to the environment without treatment which could lead to a lot of environmental problems and health issues. Thus, the wastewater from these industries should be treated before discharging to the environment including the rivers and flowing water. Presently, many process industries including the paper, textile, plastic, and leather industries use pigments, and dyes for the colouring their products, and excess of these colours end up in the discharge which ultimately end-up as industrial effluents. The presence of dyes poses as toxic materials, resistant to a chemical reaction in wastewaters leads to cancer, mutagenesis, and other severe problems in human and aquatic creatures. The complicated chemical structures of dyes make these materials highly resistant to biodegradation (Fu et al., 2015; Mokhtari et al., 2016). One among the most commonly used dyes in process industries is methyl orange (MO), which belongs to a group of azo dyes, and poses higher toxic nature. These dyes are dangerous for aquatic creatures, due to the presence of an azo group (-N=N-) in their chemical structure and hence need to be removed from the aqueous environment (Agarwal et al., 2016; León et al., 2016).

This biggest concern has driven many researchers and scientists to develop technologies and remedial methods for its removal. A number of methods have already been in use to remove the coloured compounds from effluent water and wastewater. These methods include photocatalytic degradation (He et al., 2017), separation by membrane (Bouazizi et al., 2017), coagulation process (Li et al., 2016),electrolysis (Zou and Wang, 2017), liquid-liquid extraction (Bukman et al., 2017),and adsorption (El Essawy et al., 2017; Hu et al., 2016; Karri

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et al., 2017a, 2018; Karri and Sahu, 2018; Zhou et al., 2017). Among these technologies, the application of adsorption has found many applications owing to the fact that this method is easier, cheaper and highly efficient for dye removal (Kumari et al., 2017; Li et al., 2018; Wang et al., 2017; Zhang et al., 2016).

Recently, researchers have discovered that few polymeric materials (as an adsorbent) can remove a variety of pollutants from effluent and wastewater (Agcaoili et al., 2017; Jeon et al., 2008; Karimi et al., 2016; Kono et al., 2016; Yang and Ni, 2012). Polyaniline is one of the conventional conductive polymers which can remove various pollutants such as organic materials from aqueous solutions due to the presence of amine and imine groups in its chemical structure. These groups in the polyaniline could bind with the functional groups of the pollutant material leading to the removal of pollutant from the aqueous solution. This conductive polymer can be an excellent adsorbent to remove the pollutants due to advantages such as cheaper aniline monomer, the simplicity of its synthesis and thermal stability of polyaniline against the ambient conditions (Liu et al., 2017; Sun et al., 2017; Tanzifi et al., 2017b, 2018; Yan et al., 2015). Investigating the performance of newly developed material as an adsorbent under different operating conditions was highly time-consuming and costly. Notably, each process variables have a different impact on the adsorption efficiency and these variables also have some interaction effects and overall adsorption efficiency. A systematic experimental procedure is needed to identify the influence of each parameter on the adsorption process and finding their optimal values to achieve maximum removal efficiency. Particularly to analyze and understand the system behaviour, a various number of experimental runs has to be carried out. This process can be tedious and highly time-consuming. In order to accomplish these tasks, the researcher has to properly plan and design the experimental model which uses limited resources. Design of experiments approach is adapted to design and plan optimal experimental runs, to get valuable insights of the process.

The primary objectives of this research study is to (i) find the performance of polyaniline-based nano-adsorbent for removal of MO from wastewater in a batch adsorption process, (ii) obtain a predictive multivariate model based on response surface methodology (RSM) and compare with data-driven modeling using artificial neural network (ANN) which is integrated with differential evolution optimization (DEO). Implementation of ANN has large applications; whereas implementation of ANN-DEO in the field of adsorption are very few (Karri et al., 2017b; Lingamdinne et al., 2018; Maheshwari and Gupta, 2016); however, as per the author's knowledge, there is no application in open literature so far for application of ANN-DEO for the removal of dyes from wastewater. Therefore, this is a novel approach investigating the performance of polyaniline-based nano-adsorbent for removal of MO from wastewater.

2. Material and methods

2.1. Materials

The aniline monomer which was used in this study was purchased from Merck Company, Germany, and distilled before use. Ammonium peroxydisulfate (APS) as the oxidant, sodium dodecylbenzene sulfonate (DBSNa) as a stabilizer, methyl orange pigment, sodium hydroxide, and sulfuric acid were all purchased from Merck Company, Germany.

2.2. Synthesis of adsorbent

The method of preparing the polyaniline nano-adsorbent was explained in detail in our other article (Tanzifi et al., 2017a) and hence presented briefly here. To synthesize the polyaniline nano-adsorbent based on DBSNa, 0.4 g of DBSNa was first added to 25 mL of water and placed on a magnetic stirrer (the first solution). In another container, 2.5 g of APS was added to 25 mL of distilled water, and after stirring for 15 min, the first solution was slowly added and placed on the magnetic stirrer for another 15 min. Finally, 50 mL of 2 M sulfuric acid was added to the above solution, let for 15 min of stirring and 1 mL of aniline monomer was injected resulting in the polymerization reaction observed by the change of solution colour to black. This polymerization reaction was let to go for 5 h, and the product was then filtered and rinsed with distilled water for several times to remove the impurities until the solution passing the filter became colourless. Then, the remaining product on the filter was dried in the oven at 40 °C for about 48 h till it is anhydrous. Finally, it was powdered and used as the adsorbent.

2.3. Characterization

The morphology and surface form of pure polyaniline and polyaniline nano-adsorbent before and after the process of MO dye adsorption were studied by SEM (model KYKY-EM3200). In order to study the synthesized nano-adsorbent structure, the FTIR (Broker, vertex 70) was used. Furthermore, the properties of pure polyaniline and polyaniline nano-adsorbent including the surface area, the total volume of pores, and average pore diameter were studied by BET and BJH analyses (Belsorp mini analyzer, Japan).

The morphology and form of pure polyaniline particles and polyaniline nanoparticles synthesized based on DBSNa with different magnifications are presented in Fig. 1. In the SEM image of pure polyaniline (Fig. 1a), a structure with large, non-uniform, and hulk particles was observed which can be converted into a uniform structure with round and fine particles with particles size about nano by using DBSNa (Fig. 1b). In fact, DBSNa acted as a stabilizer material, was bonded with polyaniline chain, and did not allow the polymer chain to grow more resulting in the small and uniform size of polyaniline particles. Moreover, Fig. 1c displays the SEM image of polyaniline nano-adsorbent after the adsorption with two different magnifications in which changing the surface form of adsorbent was observed towards before the MO dye adsorption process. After the adsorption process, the adsorbent particles were entirely stuck to each other and lost their uniformity and sphericity (Tanzifi et al., 2017a). The FTIR spectrum at 450-4000 cm^{-1} wavelength was used to study the chemical structure of polyaniline nano-adsorbent, and details regarding the FTIR spectrum is presented in extra supplementary material (ESM).

The physical properties of polyaniline nano-adsorbent surface such as BET surface area, the total volume of pores, and average pore diameter were studied by adsorption-desorption isotherms of nitrogen. Whereas, the conventional BJH method, was used for studying the distribution of nano-adsorbent pore size. In this method, the surface area in contact with nitrogen and the volume of adsorbed nitrogen into the adsorbent were obtained from the pores size based on the single-layer and multi-layer adsorption theories, respectively. The distribution was obtained by determining the parameter of adsorbed nitrogen volume changes with respect to the pore size. The results indicate that the values of the total pore volume, average pore diameter and surface area for polyaniline nano-adsorbent based on DBSNa was enhanced about 30%, 20%, and 30%, respectively, compared with pure polyaniline. The surface area, average pore diameter, and the total pore volume of synthesized adsorbent was 10.441 m^2/g, 13.824 nm, and 2.3988 m^3/g, respectively (Tanzifi et al., 2017a).

2.4. Batch adsorption experiments and experimental design

The experimental studies of MO dye adsorption onto polyaniline nano-adsorbent were conducted in a batch process, wherein the stirring speed and dye volume were maintained at 1200 rpm and 50 mL respectively in all the experimental runs. As the adsorption efficiency depends on the process parameters (variables), in this study, the effect of five significant parameters namely solution pH, process temperature, initial concentration, the required time for adsorption process and