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Adsorption of Triamterene on multi-walled and single-walled carbon nanotubes: Artificial neural network modeling and genetic algorithm optimization

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ABSTRACT

Rapid adsorption of Triamterene using multi-walled carbon nanotube (MWCNT) and single-walled carbon nanotube (SWCNT) was well investigated and elucidated. The impact of influential variables such as temperature, amount of adsorbent, initial drug concentration, contact time was modeled using multiple linear regression (MLR) and artificial neural network (ANN) and the influential variables were optimized using genetic algorithm (GA). The adsorption equilibrium and kinetic data was well fitted and found to be in good agreement with the Langmuir monolayer isotherm model and pseudo second order kinetics mechanism. The maximum adsorption capacity of SWCNT and MWCNTs for the removal of Triamterene was found to be 25.77 mg g^{-1} and 33.14 mg g^{-1} respectively. The negative value of adsorption enthalpy (ΔH°) reveals towards the exothermic nature of the adsorption process. Based on the results comparison of the proposed models, results revealed that the applicability of the ANN model is more appropriate in comparison to the MLR model for predicting the adsorption efficiency of the process. The coefficient of determination (R^2) of 0.980 and mean squared error (MSE) of 0.002 for adsorption on SWCNT 0.986 and 5.4e-04 on MWCNT were obtained, respectively using the optimal ANN model.

1. Introduction

Triamterene (2,4,7-triamino-6-phenylpteridine) is a yellow crystalline powder. Triamterene in combination with hydrochlorothiazide and propranolol hydrochloride is used in therapeutics as diuretic medicines, in oedema (water retention), hypertension (high blood pressure) and chronic insufficiency of heart muscle [1,2]. The drugs are unavoidably discharged into the aquatic and soil environments. In recent years, the environmental risks of pollutants and removed of them from the environment have been interested for researchers [3].

There are many pollutant removal pathways. Among treatment techniques for wastewater treatment, the adsorption methods are widely applied to remove classes of chemical contaminants from wastewater. These wide application concerns due to high efficiency, capacity adsorbent usable for large scale application, while the adsorbent is generable, safe and non-toxic and/or lower toxicity [4]. Carbon nanotubes (CNTs), including single-walled carbon nanotubes (SWCNT) and multiwalled carbon nanotubes (MWCNT) have been widely studied since their discovery [5,6]. CNTs have been widely used in electronic transistors [7], biosensors [8], optical component [9], because of their unique and prominent chemical, mechanical and electronic properties [10].

Problem engineering solving are typically implemented using a trial and error experiments which are normally inaccessible of inputs and outputs because it is a costly, dangerous and time-consuming work. Modeling is a proven and well-accepted engineering method that it can help us to better understand the system. Adsorption is a complex and nonlinear process. Therefore modeling of the adsorption is a significant topic. In recent years, various intelligent systems (IS) including artificial neural networks (ANN) [4,11], adaptive Neuro-Fuzzy inference system (ANFIS) [12,13], support vector machine (SVM) [14] and random forest (RF) [15] approaches have been widely employed to solve a set of problems in engineering applications such as adsorption. Genetic

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algorithm (GA) according to evolutionary algorithms is utilized for optimization of the objective function by means of the parameter space coding. This technique used in complex spaces of different scientific disciplines following admit to maximize or minimize an objective function [4, 12]. The GA can achieve acceptable results using operators, including reproduction, crossover and mutation. The theory and application of ANN and GA to chemical problems can be obtained in literature [16].

From this point of view, we have decided to evaluate the potential usage of SWCNT and MWCNT as a new, renewable, inexpensive, green, and environmentally friendly absorbent for the removal of Triamterene. The effects of important experimental parameters including aqueous solution pH, contact time, adsorbent concentration, temperature and initial drug concentration were studied. The SWCNT and MWCNT are applicable for the removal of the high amount of Triamterene in a very short contact time with high adsorption capacity. In order to figure out the adsorption mechanism, the kinetics and equilibrium data were analyzed and several models were employed to fit the experimental data. Study of the impact of temperature on the adsorption makes possible to compute the thermodynamic parameters. In addition, the MLR and ANN have been applied for consideration of linear and nonlinear relationships exist among variables while GA employed to optimize the impact of variables concern to the adsorption of Triamterene by SWCNT and MWCNT.

2. Experimental

2.1. Instruments and reagents

Chemicals, such as HNO_3, NaOH, and HCl, with the highest purity available purchased from Merck (Dermasdat, Germany). The Triamterene (Fig. 1) [Chemical formula $=C_{12}H_{11}N_7$, Mol. Mass $= 253.263\ g\ mol^{-1}$] is produced for the Iran DARU Company (Tehran, Iran) and single walled carbon nanotubes (SWCNTs) and multiple walled carbon nanotubes (MWCNTs) are prepared from Research Institute of Petroleum Industry (RIPI) (Tehran, Iran). The stock solutions ($100\ mg\ L^{-1}$) of Triamterene prepared by dissolving 10 mg of pure compound in 100 mL of double distilled water, while working solution prepared freshly. The pH measurements were carried out using pH/Ion meter model-691 (Metrohm, Switzerland, Swiss) and thermometer Metrohm, international ASTM sieves and Stirrer model UKA were used in this study.

The absorbance measurements were carried out using PerkinElmer UV–Vis spectrophotometer model Lambda 25 (USA) at a wavelength of 375 nm. The Raman spectra were determined using the FT-Raman accessory kit (FRA/106-S) of a Bruker Equinox 55 FT-IR spectrometer. A continuous-wave Nd: YAG laser working at 1064 nm utilized for Raman excitation. The morphology of the MWCNTs followed by field emission scanning electron microscopy (FE-SEM; Hitachi S-4160, Japan) under an acceleration voltage of 15 kV. The morphology and size distribution of the MWCNTs and SWCNTs were determined by a transmission electron microscopy (TEM; Hitachi H-800, Japan) at an operating voltage of 200 kV. A BET surface analyzer (Quantachrome NOVA 2000, USA) was employed to determine nitrogen adsorption–desorption isotherm at 77 K. Before each survey, the samples were degassed by helium purging at 553 K for 3 h. The BET experiments present the useful data on the adsorbent properties, namely surface area, total pore volume and micropore area.

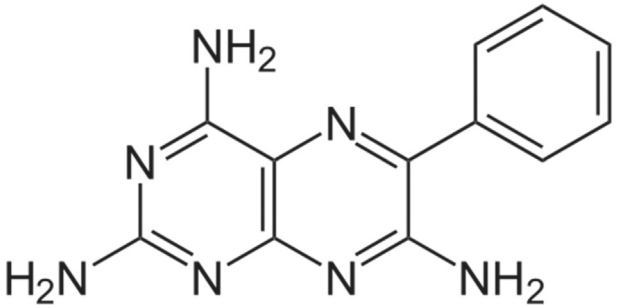

Fig. 1. Structure of Triamterene.

2.2. Methods

The effect of important parameters such as pH, contact time, initial drug concentration and temperature on the removal of Triamterene, was investigated and studied using a batch experiment. For each experimental run, 50 mL of drug solution of known concentration was completely mixed with optimum value. This mixture at room temperature with constant speed was stirred. Samples at various time intervals (5–60 min) were gathered and then filtration, and determination of concentration were done. The impact of pH (in the range of 2–10) on Triamterene removal on both adsorbents was studied and further experiments were carried out at pH 7 for SWCNT and pH 8 for MWCNT. The percentage removal of Triamterene was calculated using the following relationship:

$$\text{Drug removal}\ (\%) = ((C_0 - C_t)/C_0) \times 100 \quad (1)$$

where C_0 and C_t ($mg\ L^{-1}$) are the initial drug concentration and concentration at time t, respectively.

Adsorption isotherms considered by conducting a set of similar experiments according to the described method at various concentrations of Triamterene in the range ($10–40\ mg\ L^{-1}$) while agitated with 0.005–0.02 g of MWCNT and SWCNT. After equilibrium, the adsorption capacity was calculated by:

$$q_e = (C_o - C_e)\ V/W \quad (2)$$

where q_e ($mg\ g^{-1}$), is the equilibrium adsorption capacity, C_e is the drug concentration at equilibrium, V (L) is the volume of solution and W (g) is the weight of adsorbent.

2.3. Modeling and optimization

2.3.1. Artificial neural network (ANN) model

The Matlab R2012a was applied to predict the adsorption efficiency. A three layer ANN (Fig. 2), with a tangent sigmoid transfer function (tansig) at hidden layer, Levenberg–Marquardt back-propagation algorithm with 1000 iterations and a linear transfer function (purelin) at output layer employed. The data were randomly divided into three groups (70% for training, and 30% for the testing set). Therefore 45 and 19 samples used for training, and testing subsets, respectively. Three neurons (concentration ($mg\ L^{-1}$), adsorbent dosage (g) and contact time (min)), 1 to 25 neurons in the hidden layer and one neuron (removal percentage) in the output layer were applied. The data were normalized between 0.1 and 0.9 to avoid numerical overflows because of very large or small weights. The following normalization equation used for modeling:

$$y = (x_i - x_{min}/x_{max} - x_{min}) \times 0.8 + 0.1 \quad (3)$$

where y is the normalized value of x_i, the x_{max} and x_{min} are the maximum and minimum values of x_i, respectively. The performance of the ANN models conducted according to the mean squared error (MSE) and the coefficient of determination (R^2) which can be defined as follows:

$$MSE = \frac{1}{N}\sum_{i=1}^{N}\left(\left|y_{prd,i} - y_{exp,i}\right|\right)^2 \quad (4)$$

$$R^2 = 1 - \frac{\sum_{i=1}^{N}\left(y_{prd,i} - y_{exp,i}\right)}{\sum_{i=1}^{N}\left(y_{prd,i} - y_m\right)} \quad (5)$$