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### Simple and facile sonochemical synthesis of lead oxide nanoparticles loaded activated carbon and its application for methyl orange removal from aqueous phase

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#### ABSTRACT

The efficiency and performance of lead oxide nanoparticles loaded activated carbon (PbO-NP-AC) which was fully characterized by different techniques including FTIR and SEM analysis were described. The influence of variables including pH, contact time, MO concentration and mass of adsorbent was investigated and optimized by artificial neural network-partial swarm optimization (ANN-PSO). At optimal conditions predicted by ANN-PSO, the coefficient of determination ( $R^2$ ) and mean square error (MSE) which corresponds to test data was 0.9685 and 0.00093, respectively. The maximum removal percentage (approximately 98%) was observed at conditions set at: 0.02 g of PbO-NP-AC, 15 mg L<sup>-1</sup> of MO at pH 2.0 following mixing and stirring for 30 min. The experimental data were efficiently adopted by Langmuir model at all conditions with maximum adsorption capacity of 333.33 mg g<sup>-1</sup>. Kinetic studies at various adsorbent mass and initial MO concentrations reveal that maximum MO removal was achieved within 15 min, while experimental data follow the pseudo-second-order rate equation in addition to interparticle diffusion model.

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#### 1. Introduction

The industrial and food staff pollutants have more contribution on different ecosystem media such as aqueous media [1]. Dyes and their breakdown products generate high toxicity for living organisms following their arrival to rivers and other aqueous media [1]. Azo reactive dyes with high brightness in color due to the presence of azo (-N=N-)groups in cooperation with substituted aromatic structures [2,3] have more toxicity to living organisms. Unique properties viz. high stability to light, heat and oxidizing agents cause their hard and difficult degradation, while in some times more toxic compounds were appeared [4]. The previously reported disadvantages and drawbacks such as the requirement of a large amount of supporting material, generation of a large amount of secondary waste in expensive and tedious stages and production of a large amount of sludge [5–11] encourage the researchers to develop and design efficient methods such as adsorption. Adsorption is especially based on search for novel and non-toxic very reactive material candidate adsorption as an efficient and most prominent approach for dye removal [10–15]. The application of nanoparticles is associated with advantages including high specific surface area (high

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adsorption capacity) [16]. Their performance is significantly affected by their size, surface area and interparticle interaction affinity. Methyl orange (MO; anionic category) despite of its extensive usage in textile, printing, paper manufacturing, pharmaceutical and food industries [17, 18] leads to health hazards (breathing problem, vomiting, diarrhea and nausea) which are corroborated due to its application as a weak acid-base indicator.

Adsorption process (complex and nonlinear process) encourages the researchers to apply nonlinear models including least squares support vector machines (LS-SVM), random forest (RF), adaptive neurofuzzy inference system (ANFIS) and artificial neural network (ANN) for the prediction of the adsorption process. Artificial neural networks (ANNs) are widely used as alternative mathematical methods to solve various problems with minor adaptations [19–25]. Back propagation neural network (BPNN) with high efficiency is able to solve non-linear and complex problems. The improvement in reliability achieved by using heuristic optimization algorithm such as genetic algorithm (GA), imperialist competitive algorithm (ICA) and particle swarm optimization (PSO) is combined with BPNN to avoid local minimum and achieve global convergence quickly and correctly [26,27]. A hybrid of artificial neural network and particle swarm optimization (ANN-PSO) is used for the prediction of pollutant removal from water samples [28–30].

Another objective of this study is to focus on the application of the ANN-PSO model to predict the MO adsorption behavior onto PbO-





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Fig. 1. Chemical structure of methyl orange.

NP-AC, which in routine and high efficiency protocol was synthesized in our laboratory. The PbO-NP-AC was characterized via different techniques including FTIR, XRD and SEM analysis. Variables (pH, MO concentration, amount of adsorbent and contact time) affected on the removal process were optimized possible to achieve reasonable removal percentage (more than 98%) in reasonable time (lower than 30 min).

#### 2. Experimental

#### 2.1. Instruments and reagents

The stock solution (100 mg L<sup>-1</sup>) of MO (Fig. 1; formula weight: 327.34 g mol<sup>-1</sup> with formula of C<sub>14</sub>H<sub>14</sub>N<sub>3</sub>NaO<sub>3</sub>S) was prepared by 20 mg pure MO in 200 mL of double distilled water and its suitable dilution to desired concentration was used as working solution. All chemicals including NaOH, HCl, KCl and MO were purchased from Merck (Dermasdat, Germany). The experimental measurements for measurements of MO uptake at optimization section, kinetic and

isotherm studies were carried out according to conditions and instruments previously reported elsewhere [13,15,19].

#### 2.2. Hybrid neural networks and PSO

The classical ANN training methods (back-propagation (BP)) were simply extended by particle swarm optimization (PSO) algorithm. The training starts with PSO based on global search on the net weight range to refine an initial random set of weights to achieve a better estimate close to the global optimum. Subsequently, the BP algorithm progresses the training and refines the solution provided by the PSO to approach the optimum solution as efficient, powerful and simple optimization algorithm [31–33]. Fig. 2 indicates the flow chart of the PSO that is composed of several steps such as [34,35] generation and initialization of particles array, evaluation of objective function until obtaining better position, determines new g best value, calculation of particles' new velocity, updates particle's position and repeating steps (1) and (2).

PSO has several controllable parameters such as the acceleration coefficients (Personal Learning Coefficient  $= c_1$  and Global Learning Coefficient  $= c_2$ ), inertia weight (w), velocity clamping and swarm size (n Pop). Wrong initialization of these parameters may lead to divergent or cyclic behavior [32].

In this study, MATLAB R2013a software (ANN-PSO) models were used to predict the removal percentage of MO as a function of three input variables including concentration (mg  $L^{-1}$ ), adsorbent dosage



Fig. 2. Flowchart of the PSO algorithm.

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