Chemometric assisted sonochemical dyes adsorption in ternary solutions onto Cu nanowires loaded on activated carbon

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A B S T R A C T

The aim of this research is the evaluation of ultrasonic assisted adsorption of basic dyes in ternary mixtures onto Cu nanowires loaded on activated carbon (Cu-NWs-AC) via the normal and derivative spectrophotometric methods. These are considered as simple, inexpensive, rapid, and accurate methods for the dyes determination in multi-components mixtures. The shape, size, and specific surface area of Cu-NWs-AC was characterized by FESEM, XRD, and BET techniques. The main parameters affecting the simultaneous dyes removal were studied, and subsequently optimized through central composite design (CCD), one of the most widely used multivariate procedures. Because of high $R^2$ values and being close to adj-$R^2$ values, the predicted and experimental responses were in the reasonable agreement with each other. In optimal conditions consisted of 20 mg of adsorbent dosage, 4 min of sonication time, pH 6, 26 mg/L of BB41 dye, 18 mg/L of BR46 dye, and 15 mg/L of BY13 dye; high removal percentages (higher than 95%) with logical desirability of 0.95 were obtained. Among the conventional isotherms and kinetic models, extended Freundlich isotherm and pseudo-second-order model showed reasonable fit to the adsorption equilibrium data, respectively.

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1. Introduction

Synthetic dyes, as the main category of colorants, have the most consumption in various industries such as textiles, leather, cosmetics, paper, printing, plastics, and so on. They generally possess complex aromatic structures with various chromophores, color-bearing groups, like azo, monoazo, diazo, nitro, azine, thioazine, anthraquinone, quinoline, sulfur, xanthene, phthalocyanine, nitro, and nitroso. They mainly carry net negative or positive charges in the aqueous solution due to the presence of sulfonate or protonated amine groups, respectively. Among the synthetic dyes, basic and reactive dyes with azo chromophores are extensively used in the textile industry. They have favorable characteristics such as bright color, good solubility in water, being cheaper to produce, and easier to apply to fabric. Unfortunately, during the dyeing fibers, 10%–15% of the dyes amount is lost in the effluent [1]. Azo dyes can be more toxic, because of production hazardous aromatic amines under anaerobic conditions. Therefore, discharging of the dye effluents into the hydrosphere before their treatments can be dangerous for the environment. They can damage aquatic organisms, flora and fauna by preventing the light penetration to water, varying the pH value of water, and increasing the chemical oxygen demand. Also they can potentially possess toxic, carcinogenic, and even mutagenic effects for the human health and cause many diseases like nausea, hemorrhage, allergic dermatitis, ulceration of the skin, and severe irritation of respiratory tract. Moreover, they may have adverse effects from visual endpoints due to strong colors in the wastewaters [2–5].

The physicochemical techniques including oxidation, flocculation, coagulation, membrane filtration, photocatalysis, electrochemical techniques, and adsorption are usually applied for the removal of colored contaminants from industrial wastewaters [6–14]. Adsorption generally has higher efficiency, without production of any unwanted by-products. Also, ease of operation and usage of the large variety of effective, cheap, and nontoxic materials as adsorbents, are other advantages of this method. Among various adsorbents, activated carbon is more popular because of high porosity, various reactive functional groups, and ease of surface modification by different nanomaterials [15–21]. According to shapes and morphologies, nanomaterials can be classified to nanowires, nanorods, nanofibers, and nanoparticles. Due to unique physical and chemical properties like good dispersibility and high ratio of surface-to-volume, they can provide better kinetic and greater adsorption capacity. Besides, they can apply for several times without lowering the efficiency of adsorption [22–29].

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In one-component colored solutions, the concentration of a dye can be easily determined by normal spectrophotometric method. However, it is not suitable for simultaneous quantitative determination of two or more components in the mixtures, due to spectral overlapping. Many techniques such as high performance liquid chromatography, voltammetry, capillary electrophoresis, derivative spectrophotometric methods, and etc. have been applied for the simultaneous dyes determination at multi-components mixtures. Most of them mainly require separation or purification steps as well as high volumes of organic solvents; while derivative spectrophotometric methods are simple, sensitive, safe, and inexpensive. The most widely used derivative spectrophotometric procedures consist of zero-crossing, peak to peak, peak to zero, and ratio derivative methods. Via zero-crossing method, the simplest and the most applicable derivative spectrophotometric methods, quantitative measurement of one analyte in the mixture can be performed at a wavelength in which the amounts of absorbance derivations of other components are zero. For achievement to reasonable accuracy and precision, the absorbance derivation amount should be sufficiently large \[30–34\].

Univariate and multivariate procedures have been used for the optimization and investigation of the parameters involved in the ultrasound assisted adsorption method. In univariate method, one variable at a time method, the effect of each parameter is investigated while the values of others are held at constant levels. In addition to the high numbers of tests, the interaction effects between parameters are ignored in this method. So, it is not proper for finding real optimum conditions. Alternatively, utilization of multivariate methods, designs of experiments, have been encouraged in the simultaneous optimization of several variables; because of rapidity, more affordability, and effective performance. Response surface methodologies (RSMs) are the most applicable multivariate methods. They are combination of the mathematical and statistical methods used for the identification of significant variables and optimum conditions. Major, efficient, and applicable types of the response surface methodologies consist of central composite design (CCD), Box- Behnken design (BBD) and Doehlert design. These expanded forms of two-level full (or fractional) factorial designs offer accurate models for curvature surfaces by considering interaction and quadratic terms. Despite the numbers of experiments in BBD are lower, if values of factors can be extend in five levels, CCD is more effective \[35–41\].

In this research, CCD was applied for the optimization of the effective variables on the ultrasonic assisted adsorption of basic dyes in ternary solution by using Cu-NWs-AC. Simultaneous analysis of BR46, BB41, and BY13 dyes in ternary solutions was successfully performed via zero-crossing method by using zero and first order derivative spectra. Finally, for assaying the dyes equilibrium adsorption in ternary solutions, the commonly modified isotherms and kinetics models were applied.

2. Experimental

2.1. Materials and instruments

Basic Blue 41 (BB41), Basic Red 46 (BR46), and Basic Yellow 13 (BY13) dyes with commercial names of Astraron Blue FFGG, Astrazon Red FBL, and Astrazon Yellow 8G, respectively were supplied from Daystar Firm. The chemical structures and other properties of these dyes are shown in Table 1. All of used chemicals including hydrazine, ethylenediamine, copper nitrate, HNO₃, and NaOH with the highest purity available were purchased from Merck (Darmstadt, Germany).

A Shimadzu, UV–vis 1650 PC spectrophotometer was used to take the absorbance spectra of BB41, BR46, and BY13 dyes in the range of 200–700 nm by a pair of quartz cells having optical paths of 1 cm. PHS-3BW pH-meter (Bell, Italy) with a combined glass-Ag/AgCl electrode was used for adjustment of pH values of test solutions. An ultrasonic bath at 50/60 kHz of frequency (SW3, Switzerland) was used to accelerate the dyes adsorption process. Shape and size of Cu-NWs was determined via X-ray diffraction (XRD) pattern, collected by an automated Philips X’Pert X-ray diffractometer (Netherlands) with Cu Kα radiation (40 kV and 30 mA) in 2θ values over 10°–100°.

Table 1

<table>
<thead>
<tr>
<th>Chemical structure</th>
<th>Commercial name</th>
<th>CAS NO.</th>
<th>λ_max (nm)</th>
<th>M_w (g/mol)</th>
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</thead>
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<tr>
<td><img src="image" alt="Chemical structure" /> Basic Blue 41</td>
<td>12270-13-2</td>
<td>607</td>
<td>482</td>
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<tr>
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<td>530</td>
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<td>12217-50-4</td>
<td>413</td>
<td>343</td>
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