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Analysis of synergistic and antagonistic adsorption of heavy metals and acid blue 25 on activated carbon from ternary systems

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A B S T R A C T

This article reports the antagonistic and synergistic adsorption involved in the multicomponent removal of acid blue 25 dye (AB25) and heavy metal ions (Zn^{2+} , Ni^{2+} , Cd^{2+}) from ternary systems using an activated carbon modified with wastes of egg shell. Adsorption experiments were performed in ternary mixtures: Zn^{2+} - Ni^{2+} -AB25, Ni^{2+} - Cd^{2+} -AB25 and Zn^{2+} - Cd^{2+} -AB25 where Taguchi experimental designs have been used to study the adsorbent performance and to identify the antagonistic and synergistic adsorption caused by the simultaneous presence of these water pollutants. Results indicated that these systems might show both competitive and synergistic adsorption. Dye AB25 enhanced the removal of these metallic species and reduced the competitive adsorption between the metal ions present in multicomponent solution especially for Cd^{2+} . This synergistic adsorption caused by AB25 depends on the concentrations of dye and heavy metals, and the improvement of metal adsorption is also dependent on the metal ion. This improvement of the adsorption capacity of activated carbon could be caused mainly by the ion exchange between the heavy metals and the ion Na^+ from the auxochrome functional group of the dye molecule. Traditional multicomponent isotherm models and a model based on the response surface methodology have been employed and compared in the modeling of multicomponent adsorption data. This study provides new experimental data and new findings on the multicomponent adsorption of water pollutants using activated carbon.

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1. Introduction

Adsorption process is the most used and effective technique for treatment of wastewaters polluted by inorganic and organic toxic compounds (Dias et al., 2007; Tongpoothorn et al., 2011; Li et al., 2013; Monsalvo et al., 2012). Several studies have shown that wastewater treatment based on adsorption process is an effective and low-cost purification technology if the proper adsorbent is used. In particular, adsorption may offer more advantages for removing priority water pollutants than those obtained with other water purification methods,

e.g., electrochemical treatment and ultrafiltration, coagulation and ion exchange (Babel and Kurniawan, 2003; Gupta and Suhas, 2009; Bhatnagar and Sillanpaa, 2010; Fu and Wang, 2011; Wang Ngah et al., 2011).

In real-life applications, the system to be treated is likely to have more than a single adsorbate in solution. Therefore, multicomponent systems are useful to exemplify the practical situations because industrial effluents usually contain several pollutants, which may show different chemical and physical properties (Erto et al., 2012). The operation and design of adsorption units for water treatment must be based on

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the study of multicomponent adsorption data to identify and characterize the adsorbent performance under competitive conditions (Reynel-Avila et al., 2011). Literature indicates that the presence of several pollutants in the solution to be treated may decrease, increase or may not affect the removal performance of the adsorbent used in the purification process (Mohan and Chander, 2006; Srivastava et al., 2008, 2009). As a consequence, there may be a great variation in the adsorbent performance depending on the characteristics (e.g., type of pollutants and its concentration) of the multicomponent system under study. In particular, the pollutant concentration in multicomponent systems is a crucial parameter for adsorption because it can determine the occurrence of unexpected effects when different solutes are present in the same solution. Therefore, a proper understanding of the impact of pollutant concentration on competitive or synergistic adsorption that can take place in multicomponent systems is an important topic in adsorption research and is fundamental to identify the best operating conditions for wastewater treatment.

Adsorption data on multi-pollutant systems are still limited especially for mixtures containing different types of pollutants. However, this type of studies is required to design and to optimize adsorption units for the multicomponent removal of priority water pollutants. Heavy metal ions and dyes are common pollutants of the wastewater discharged by different industries (Gupta and Suhas, 2009; Fu and Wang, 2011; Wang Ngah et al., 2011; Lo et al., 2012). Both chemical species are dangerous pollutants of great importance due to their wide usages in different industrial sectors, toxicological properties and negative impact on ecosystems and live organisms including the human beings (Gupta and Suhas, 2009; Fu and Wang, 2011). Several studies on the adsorption of both heavy metals and dyes have emphasized on the uptake of single pollutant (i.e., single component) using a number of different adsorbents. These adsorbents include zeolites, activated carbons, agricultural and industrial by-products, natural and synthetic polymers, biomasses and clays, among others (Babel and Kurniawan, 2003; Saha et al., 2003; Srivastava et al., 2006; Dias et al., 2007; Gupta and Suhas, 2009; Bhatnagar and Sillanpaa, 2010; Wang Ngah et al., 2011; Fu and Wang, 2011; Moussavi and Khosravi, 2011; Moreno-Virgen et al., 2012). However, the effluents generated by several industrial activities may simultaneously contain both metallic ions and dyes. For example, the wastewaters of textile industries contain a variety of inorganic and organic pollutants including heavy metals and acid dyes, which are generated from different textile process operations, e.g., scouring, bleaching and dyeing (Ozturk et al., 2009). Despite this fact, studies on the simultaneous adsorption of dyes and heavy metals have been mainly focused on binary dye-metal mixtures (Wang and Ariyanto, 2007; Visa et al., 2010; Tovar-Gómez et al., 2012). Furthermore, results reported in other studies indicate that this type of multicomponent systems (i.e., dye + heavy metals) may show both competitive and synergistic adsorption depending mainly on the dye chemical nature. For example, Wang and Ariyanto (2007) studied the simultaneous removal of Pb^{2+} ions and malachite green using a natural zeolite. These authors concluded that both pollutants exhibited an antagonistic adsorption on the natural zeolite. Visa et al. (2010) reported the multicomponent adsorption of Ni^{2+} , Cd^{2+} , Cu^{2+} and methylene blue on a modified fly ash. Results showed that adsorption of these metals decreased with metal ion concentration. Finally, Tovar-Gómez et al. (2012) analyzed the

binary adsorption of heavy metals and dye acid blue 25 (AB25) on an activated carbon modified with wastes of egg shell. Adsorption experiments in binary systems (i.e., dye + heavy metal) showed that the heavy metal removal was significantly enhanced by the presence of AB25, i.e., there was a synergistic adsorption. However, studies on adsorption in ternary or higher order multicomponent systems, containing dyes and heavy metals, do not seem to be available in the existing literature. In addition, the analysis and the interpretation of results of multicomponent adsorption from mixtures of three or more components are more complex and, consequently, suitable tools for processing multicomponent adsorption data should be employed to establish reliable conclusions.

In this article, we have studied the multicomponent adsorption of AB25 dye and heavy metal ions Ni^{2+} , Cd^{2+} , Zn^{2+} in ternary systems using an activated carbon that has been chemically modified with the wastes of egg shell (Guijarro-Aldaco et al., 2011; Tovar-Gómez et al., 2012). The performance of this adsorbent for removing multiple adsorbates has been studied using the ternary mixtures: AB25- Zn^{2+} - Ni^{2+} , AB25- Zn^{2+} - Cd^{2+} and AB25- Ni^{2+} - Cd^{2+} . Note that anthraquinonic dyes are among the most important commercial dyes and AB25 has been selected in this study because of it has several applications in dyeing industry and can be considered as a reference compound for studying dye adsorption in the multicomponent environment (Ghodbane and Hamdaoui, 2009). We have analyzed the multicomponent adsorption of both AB25 dye and metallic ions on this adsorbent using Taguchi's experimental designs. For data analysis, the signal-to-noise (S/N) ratio has been proposed to determine both competitive and synergistic adsorption in the simultaneous removal of heavy metals and AB25 using activated carbon. Results indicated that these multicomponent systems may show both competitive and synergistic adsorption processes. Specifically, AB25 may enhance the removal of metallic ions and reduces the competitive adsorption between the metals present in multicomponent solution especially for Cd^{2+} . In summary, this article reports new findings on the multicomponent adsorption of these water pollutants on activated carbon.

2. Methodology

2.1. Adsorbent description and physicochemical characterization

Adsorption studies were performed with a commercial bituminous carbon provided by Clarimex Company (Mexico). The surface chemistry of this adsorbent was chemically modified for enhancing its adsorption properties using wastes of egg shell (Guijarro-Aldaco et al., 2011; Tovar-Gómez et al., 2012). Fig. 1 provides the general procedure used for the preparation of this adsorbent and details of this experimental methodology have been reported by Guijarro-Aldaco et al. (2011) and Table 1 contains its physicochemical properties. Results of previous studies indicated that this adsorbent has improved adsorption properties for the removal of dyes and heavy metal ions. Note that the multicomponent removal of metallic ions and AB25 using this adsorbent has been studied in binary solutions dye-metal and metal-metal (Guijarro-Aldaco et al., 2011; Tovar-Gómez et al., 2012). Herein, this modified adsorbent has been used in adsorption experiments with ternary systems dye-metal-metal. Then, this manuscript reports new experimental data for the multicomponent adsorption of these pollutants on this activated carbon using ternary systems.

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