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Research article

High-efficiency extraction of bromocresol purple dye and heavy metals as chromium from industrial effluent by adsorption onto a modified surface of zeolite: Kinetics and equilibrium study

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ABSTRACT

Tannery industrial effluent is rich in heavy metals and basic dyes as bromocresol purple (BCP), poses an economic problem and a serious danger to the environment. This research had evaluated the importance of the adsorption properties of a modified clinoptilolite (CL) (a type of zeolite) for the removal of BCP dye and some heavy metals as total chromium (tCr) in the ammoniac phase. The modified adsorbent was prepared by mixing solid waste (SW) and CL in a ratio 10:1. The CL, SW, and CL-SW materials were characterized and the adsorption behavior of the later to BCP and tCr was completely studied. The batch removal showed the optimal conditions for BCP adsorption: pH (6.5), time (t) (60 min), temperature (T) (303.15 K), sorbent dosage (m) (60.4 mg), and initial concentration (C_o) (11.7 mg/L). Moreover, the optimum conditions for tCr removal were: pH (8.8), t (55 min), T (303.15 K), m (400 mg), and C_o (16.0 mg/L). Cr desorption mechanism was an ion exchange reaction. The experimental data of tCr were best fitted by the Freundlich isotherm and the pseudo-second-order kinetic model. The maximum adsorption capacities of BCP and tCr onto the CL-SW were 175.5 mg/g and 37 mg/g, respectively. Thermodynamic studies revealed that the adsorptions were spontaneous and endothermic with an increase of entropy. The CL modified adsorbent seems to be a good and efficient for the removal of dyes as BCP and such heavy metals including Cr. Surprisingly, this treatment has largely improved the physicochemical properties of the industrial wastewater and proved a new concept “Polluter Cleans Polluters (PoClPos)”.

1. Introduction

Water consumption has increased worldwide in every economic sector. In recent years, water pollution has improved by industrial development. Therefore, population increases without any planning urbanization. Several kinds of pollutants could be found in industrial wastewater (IWW) in huge amounts. Some of them are dyes as alizarin red (AR), congo red (CR), crystal violet (CV), fuchsin, malachite green (MG), shembl 17538738, thymol blue (TB), and triphenylmethane (TPM) (Özdemir et al., 2015). The latter dye has different structures namely phenol red (PR), bromophenol blue (BPB), bromochlorophenol blue (BCPB), bromocresol blue (BCB), and bromocresol purple (BCP) (Chmelová and Ondrejovič, 2015). For instance, BCP (molecular weight = 540.22 g/mol) produces from pharmaceutical, textile, and paper industries.

Some organic and inorganic toxic compounds as heavy metals are derived from mining and galvanizing industries. All these pollutants disturb both the environment and the living beings causing many critical diseases. Therefore, it is essential that the development of alternative technologies may be efficient, adequate, cheap and eco-friendly. The aquatic ecosystem is suffering from piles of organic pollutants including dyes causing toxic and carcinogenic effects on the human beings. For example, just textile industry uses around 10,000 different types of dyes; its annual production is higher than 7×10^5 tons (Sadegh et al., 2015). It has been estimated that 10–15% of dyes are lost in dying textile water (Burkinshaw and Salihu, 2018). Some of them are hard to remove from aqueous phase because of their solubility. For this reason, dyes removal is an important challenge for the environmental chemistry. Recently, the concern about finding other methods for organic compounds degradation has been substantially increased. BCP ($C_{21}H_{16}Br_2O_5S$) is known as a pH indicator. In its sultone form, it has a pKa 6.3 and is usually prepared as a 0.04% aqueous solution (Ueno et al., 2013). A researching group (Airoudj et al., 2008) has designed and characterized an optical sensor for gaseous ammonia based on evanescent wave absorption at room temperature using different carrier gases like argon (Ar), nitrogen (N_2) and air. BCP was deposited on fiber's core through the sol-gel process. It was reported that the described sensor showed the best response time and sensitivity

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when air used as carrier gas. Besides, BCP is used in medical laboratories to measure albumin, and as an addition to acid stop baths used in photographic processing as an indicator that the bath reaches neutral pH and needs to be replaced.

Furthermore, the manufacturing industries use BCP dye as part of some processes (Bousnoubra et al., 2016). This dye causes several problems to human and aquatic life not only by itself but also when binds to heavy metals. Thus, it is highly recommended for economical and environmental reasons to extract and recycle BCP from industrial waste streams.

In the literature, there are little reports describing BCP dye as a removal agent retained on adsorbent materials as activated carbon (AC) (Geetha and Belagali, 2013).

On the other side, it is well-known that mining and industrial activities generate polluted water effluents and streams with toxic heavy metals; for example: arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), and mercury (Hg). In contrast to organic compounds, heavy metals are not biodegradable. These are very difficult to destroy, but, can bioaccumulate causing them penetrate the trophic chain (Aljerf, 2018). Besides, textile industries release their effluents with great quantities of such heavy metals (i.e. Cr, Cu) in the ammoniac medium (Metwally et al., 2013). Commonly, this effluent is not treated before its discharge to surface water causing serious pollution and reaching underground water. Treating this kind of effluent involves also a precipitation. Sometimes this method is not efficient because it can bring the pollutant from the ammoniac phase to the solid phase as waste sludge. The final disposal of sludge becomes another environmental problem.

Organic and inorganic pollutants removal can be achieved by the adsorption process, with some materials have large superficial areas. Normally, the concern in this process is proportional to the adsorption capacity (q_e) of the adsorbent. The adsorption technology could be low-cost when the adsorption material has low price and is used with minimum or without any modification. The adsorption of more than one pollutant from IWW refers to the utilization of different kinds of adsorption materials. If the same material could be used in more than one process, then the quantity of the material employed could be reduced. However, when adsorption dye occurs, the solid disposal material becomes a problem. Usually, many researchers treat the solid adsorbent by two methods (Song et al., 2015): the first one starts with recovering the adsorbent from the treatment solutions then is recycled, while the contaminant is separated for another use. Meanwhile, the second one concentrates on clearance and incinerating facilities or even fixing it directly.

On the other hand, another type of waste is generated from these industries that called solid waste (SW) which comprises: waste fibers, residues from finishing chemicals, hydrocarbons (HCs), dyes and chemicals from solvent recovery systems, sludge from effluent plant, dye containers, chemical containers, pallets, fly ash (Aljerf, 2015) and general paper trash. Besides, SW has been invested to constitute a new adsorbent material with a modified surface structure so that other species could be adsorbable on it, using the affinity of some dye molecules as BCP dye to attract heavy metals as Cr. Cr ion in liquid tanning wastes occurs in two forms, trivalent (Cr^{3+}) and hexavalent (Cr^{6+}). The latter form is 500 times more toxic than the first one.

Zeolites are naturally occurring alumino-silicate minerals. Due to their compositions, they have the capability of adsorption in three ways: ion-exchange, physisorption (multilayer adsorption), and chemisorption (monolayer adsorption). The way adsorption is performed, occurs by the substitution of cations present in the zeolite as sodium (Na^+), potassium (K^+), calcium (Ca^{2+}), and magnesium (Mg^{2+}) by metal cations exist in the solution, by van der Waals forces or covalent bonds. In zeolites, a permanent negative charge is present due to aluminum (Al^{3+}) substitution for Si^{4+} in the mineral structure. The negatively charged zeolite surface attracts cations present in the solution (Mozgawa and Badja, 2005). Clinoptilolite (CL) is the zeolite used primarily because of its abundance, its permanent negative charge and its good performance as an adsorbent, containing silica (SiO_2)/alumina (Al_2O_3) molar ratio, between 8 and 12. The terminal groups of CL are ≡Si–OH, ≡Si–ONa, ≡Si–O–, and (≡ Si–O)$_3$Al–O–. The lower the ratio, the better the adsorption rate, since the presence of Al^{3+} promotes CL adsorption properties.

Despite, zeolites adsorption of metals and other substances is not recognized yet which leads to an important question: why these are not used to more extent for IWW treatment? A possible reason is that knowledge about the performance of the zeolites in liquid with multiple substances or pollutants is limited. Thus, no accurate behavior can be predicted, some isotherms, i.e. mathematical models showing how many adsorbates will be attached to the adsorbent, have been developed and no mathematical model is adopted for this purpose. Taking into consideration that some characteristics suggest utilization of zeolites for IWW or leachate treatment are abundant and easy extraction methods are available. These lead to fairly low price in the market (100 USD/ton) and save a good stability to the chemical and thermal processes which allow the possibility for the reutilization of it.

The potentiality of CL (negative charge ore) in combination with SW to remove BCP dye from the total industrial waste (both solid and liquid) is assessed. Then, the negative surface of the modified adsorbent (Mod: CL-SW) is used to reduce total chromium (tCr) from IWW in an alkaline (ammoniac) medium. An experimental design is obtained by response surface methodology (RSM) for the optimized conditions of tCr adsorption. A mathematical model is developed and the optimum conditions are predicted. These results will contribute to the characterization of a new material used in the reduction of some dyes and heavy metals from IWW.

2. Materials and methods

We studied at first the effectiveness to use natural zeolite to remove BCP dye from the industrial waste phase. In the treatment process, SW was separated from the effluent and participated in the investigation of the decreasing levels of tCr in the solution using adsorption properties. Therefore, tCr adsorption experiments were carried out to realize the best conditions to separate this toxic element from IWW using the adsorbent BCP-zeolite-SW phase.

2.1. Materials

All the solutions and reagents were prepared using analytical reagent grade chemicals and doubly distilled water unless otherwise specified (Supplementary Material (SM)-Chemicals and reagents, pp. 12; Reagent preparation, pp. 14).

2.2. Zeolite

CL was obtained from the Jordan Phosphate Mines Company PLC (Shmeisani, Amman, Jordan). The material was dried at 378.15 K for 24 h and stored in the desiccator, milled in a disc mill and sieved to 250 μm (60 mesh). Before experiments, the sample was washed with deionized water to remove the surface dust and dried again at 378.15 K. The ore is an aluminosilicate (Si/Al = 8.1, Table 1) with narrow mesopores, has a rough surface and is made up of the aggregation of different kinds of particles. Its BET surface area (S_{BET}) was determined by the nitrogen (N_2) adsorption at 77.15 K (−196 °C) with an Ar/N_2 ratio of 70/30.

2.3. IWW and SW sampling

IWW samples (N = 36, repetition: n = 3) and SW Samples (N = 36, n = 3) were filled in LDPE bottles previously cleaned by soaking in non-ionic detergent, rinsed with tap water, soaked in 10% v/v HNO_3 (Merck-Millipore Co., Massachusetts, USA) for 24 h and finally cleaned