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Research article

Reuse of sustainable materials for xylenol orange dye and copper (II) ion ammoniacal removal

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A B S T R A C T

Water pollution caused by heavy metals and organic compounds is an environmental problem with negative impact, making the restoration of water quality a priority. In this paper, the adsorption of xylenol orange dye (XO) on vitreous tuff mineral (VT) was studied. It was established that the adsorption capacity of VT was 45.17 mg/g. The removal was carried out by interactions between active sites on the surface of the material and the functional groups of the dye. The solid waste obtained from this process (VTXO) was reused as adsorbent material for Cu removal in the form of the complex Cu-NH_3 because this process was done in an ammoniacal medium. It was found that the adsorption capacity of this new material was 33.09 mg/g. In a previous research, VT mineral was used to remove crystal violet (CV) instead of XO. The solid waste of this last process (VTCV) was also applied for Cu-NH_3 removal, in order to compare the adsorption capacity of VT after the adsorption of two different kinds of dyes. The adsorption capacity of VTXO was lower than that of VTCV (71.23 mg/g). In both processes, adsorption kinetic was well described by a chemical adsorption onto a heterogeneous surface. The equilibrium time for XO removal was 50 min and 80 min for Cu-NH_3. The experimental design stated that the maximum adsorption capacity was reached when the initial concentration was 6400 mg/L and the solid-liquid ratio was 10 g/L. The system that requires the least amount of adsorbent was the counter flow batch. Finally, it was possible to estimate the behavior of the system on a higher scale. This research provides an efficient and economical alternative to treat water contaminated with dyes and cooper in an ammoniacal medium using the same material in both processes, one after the other.

1. Introduction

Nowadays, water consumption is growing in every economic sector worldwide. In recent years water pollution has increased with the industrial development, population growth and unplanned urbanization (Ávila, 2011). Many different kinds of pollutants could be found in wastewater, like dyes and heavy metals ions, coming from textile and paper industries mainly. These pollutants have negative impact on the environment and the living beings. Therefore it is essential the development of efficient, adequate, cheap and environmental friendly new technologies (Wu et al., 2014).

Dyes are discharged in textile effluents, some of them are difficult to remove from aqueous medium due to its chemical complex structure, formed by aromatic rings with heteroatoms. Dyes in aquatic ecosystems cause concern because its toxicity, carcinogenic and mutagenic effects. Furthermore, dyes reduce light

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penetration and photosynthesis processes in aquatic plants (Aguiar et al., 2017). For this reason, dyes removal is an important challenge for the environmental science and technology.

Xylenol orange is an organic reagent, most commonly used as a salt for heavy metals determination. It is applied in some processes of manufacturing and textile industries. This dye is toxic not only for aquatic life but also for human beings. It is known to exert its toxicity through complex formation with heavy metals. Inorganic toxic pollutants, like heavy metals, are generated from mining and galvanizing industries. In contrast to organic compounds, heavy metal are not biodegradable, they are very difficult to destroy and can bioaccumulate entering to the trophic chain (Ge et al., 2015). Effluents from metal plating and printed circuit contain high concentrations of cooper in aqueous and ammoniacal medium (Metwally et al., 2013). In large quantities, copper may cause gastrointestinal problems, ulcers, kidney damage, or even death. Most water treatments, for this kind of effluent, involve precipitation, but this method is usually ineffective because it brings the pollutant from the water to the solid phase named waste sludge. The final disposal of sludge becomes another environmental problem.

Organic and inorganic pollutants removal can be achieved by adsorption process. In the literature, there are only few reports of XO removal by this process (Zhang et al., 2016). However, it is the most attractive method for wastewater treatment because its simplicity, low cost of operation and ability to provide efficient processes (Feng et al., 2017). Normally, a high adsorption capacity is proportional to its surface area and superficial functional active sites. For this reason, in some cases the surface of materials need to be modified by adding functional groups. For this purpose, it is necessary the use of reactants or additional processes, these may increase costs. However, some pollutants, previously adsorbed in the surface material, have these functional groups already and the process with this reused material may be a promising option. Traditionally, the adsorption of more than one pollutant implies the application of more than one adsorbent materials. But, if the same material could be reused for another process the amount of employed material could be significantly reduced.

After an adsorption process the material became a solid waste. Usually, this is subjected to a desorption process or discarding, incinerating and also fixing. However, this solid waste can be applied as a new adsorbent material. A surface structure modification allows the adsorption of other species. In addition the affinity of some dye molecules to attract heavy metals, like xylenol orange, could be exploited.

The aim of this work is to establish the potentialities and efficiency of a recycled material, coming from a dye adsorption process with VT, for cooper removal as Cu-NH_3 from an ammoniacal effluent of an industrial process. It provides important information about adsorption processes of cooper in ammoniacal medium that has been reported very few times. However, this kind of medium is very common in industrial processes.

2. Material and methods

For Cu-NH_3 adsorption experiments first, we studied the effectiveness of VT to remove XO. The material named VTXO was used to decrease levels of copper ions in an ammoniacal solution.

2.1. Vitreous tuff mineral

VT was obtained from a deposit in Cuba. The material was milled and sieved to 60 mesh.

2.2. Characterization of vitreous tuff mineral and modified material

TG/DSC analyses of VT mineral were realized in Netzsch model STA 449 F3 Jupiter, at 0–600 °C temperature range. The aluminium crucible was employed in the analyses under nitrogen atmosphere.

Textural properties measurements were done by using the nitrogen physisorption technique at 77 K in a Quantachrome Autosorb-1. The average pore diameter of VT was determined with the method of Barrett, Joyner and Halenda (BJH) and by the Kelvin equation. The total pore volume was obtained at 0.99 relative pressure. The sample was previously degassed out at 200 °C for 3 h to remove water and CO_2.

Concentrations of the acid–base groups of VT were determined according to the method described in previous paper (Blanco-Flores et al., 2016).

Scanning electron microscopy (SEM) images were acquired in a JEOL JSM-6510 microscope operated at 20 kV. Samples were fixed on a support with a carbon film and sputter-coated with gold to a thickness of ~200 Å. Elemental microanalyses were performed at randomly selected areas on the solid surfaces, in order to elucidate the atomic distribution of both materials, with an energy X-ray dispersive spectroscopy (EDS) system attached to JSM-6510 microscope. All micrographs were collected on the same experimental conditions with the backscattered electron detector.

2.3. Characterization of the wastewater used for the investigation

The wastewater was collected from a printed circuit production industry which generates Cu-NH_3 on a large scale. For the determination of conductivity and pH a Conductronic pH 120 equipment was used. Infrared spectra in the 4000-400 cm^{-1} range was recorded at room temperature using Bruker Tensor 27 FTIR. The sample did not require previous preparation. Chemical composition was determined by inductively coupled plasma-atomic emission spectrometry, using a Spectroflame FTMO8, Spectrophotometer.

2.4. Kinetics adsorption of XO dye and Cu (II) ion

The wastewater in ammoniacal medium was diluted to prepare a solution with initial concentration of 6400 mg/L. 25 mL of dye and metal solution (ion initial concentrations were 700 and 6400 mg/L respectively) were added to 25 mg of VT and VTXO separately. Then, the mixture was shaken at different times at 120 rpm and room temperature. Later, samples were centrifuged and decanted. Experiments were performed in duplicate. Pseudo first order, pseudo second order and second models were applied for experimental kinetic data in order to study the kinetic process (Blanco-Flores et al., 2016).

2.5. Adsorption isotherms

25 mg of each material were put in contact with 25 mL of solutions with different initial concentrations: 700–6400 mg/L for XO and 640–6400 mg/L for cooper ion in ammoniacal solution. They were stirring during equilibrium time at room temperature. The mixture was centrifuged and decanted. The dye and metal ion concentrations in the solutions were determined using an UV/Vis Perking Elmer Lambda 10 ultraviolet–visible spectrophotometer at 500 and 622 nm, respectively.

The amount of dye and metal ion adsorbed at time interval q_t (mg/g) and equilibrium q_e (mg/g) were calculated by equation (1):

$$(C_o - C) \cdot V/_m = q \quad (1)$$

where, C_o (mg/L) is the initial dye or metallic ion concentration, C