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Efficient removal of cadmium and 2-chlorophenol in aqueous systems by natural clay: Adsorption and photo-Fenton degradation processes

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ABSTRACT

Adsorption and photo-Fenton processes were used as handy tools to ascertain the capability of natural clays to remove cadmium (Cd) and 2-chlorophenol (2-CP) from aqueous solution. Natural Fe-rich clay collected from Tejera-Esgkira in Medenine area, south Tunisia, was used as a catalyst in the heterogeneous photo-Fenton oxidation of 2-CP in aqueous solution. Clay samples were acid activated to improve their adsorptive capacity for the removal of Cd. Experimental results indicated that the adsorption of Cd ions onto natural red clay of Tejera-Esgkira followed the pseudo-second-order kinetic model. Langmuir model was found to describe the equilibrium data with the calculated maximum adsorption capacity of 23.59 mg g⁻¹ for acid-activated clay. Photo-Fenton experiments proved high activity of the natural clay catalyst, which was able to completely degrade the phenol present in the treated solution after 30 min and in the presence of ultraviolet light C (UV-C). Total organic carbon and gas chromatography analysis confirmed a 2-CP degradation mechanism toward an almost complete mineralization of the organic compound.

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1. Introduction

Wastewater produced from process industry generally contains inorganic pollutants and other organic contaminants [1–4]. Heavy metals such as cadmium (Cd), copper, lead, chromium, and so forth along with phenol and its derivatives are hazardous pollutants generated in petroleum refinery [3,5]. The presence of these pollutants in the aquatic environment causes deleterious effects (i.e., chemical, physical, or biological threats). Moreover, the

consumption of polluted water can cause detrimental effects on living organisms [3,7]. Cd and 2-chlorophenol (2-CP) are among the most harmful pollutants because of their detrimental effects to living organisms even at trace levels. The US Environmental Protection Agency imposes very severe regulations for Cd and 2-CP contents in wastewater; the maximum concentration should be less than 5 ppb for Cd(II) ions and 1 mg/L for 2-CP. Therefore, the removal of these pollutants from wastewater is needed to reduce the concentration of pollutants.

Several techniques used in the removal of Cd(II) from the wastewater include chemical precipitation, coagulation/flocculation, ion exchange, solvent extraction, cementation, complexation, electrochemical operations, biological operations, adsorption, evaporation, filtration,

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and membrane processes [8]. Similarly, various methods such as distillation, liquid–liquid extraction, adsorption, pervaporation, and membrane extraction, thermal oxidation, catalytic oxidation, and photocatalytic degradation were used for the removal of 2-CP from aqueous solutions [9,10].

The choice of the treatment technique depends on various factors including efficiency of the process, operation, and costs. On the basis of these conditions, adsorption and photo-Fenton process are expected to have the required specifications for the removal of Cd and 2-CP in aqueous conditions.

Adsorption is one of the most commonly used processes because of its simple and convenient unit operation and for its low cost compared with other treatment processes. The use of adsorption contacting system for industrial and municipal wastewater treatment has become prevalent during recent years [11]. Adsorption is often used at the end of a treatment sequence for pollution control because of the high efficient purification. Activated carbon is recognized as an effective adsorbent because of its large surface area and high adsorption capacity. However, its high cost and difficult recovery from treated water limit its use as an adsorbent [12]. Numerous researchers including Sdiri et al. [13], Li et al. [14], Heidari et al. [15], and Eloussaief et al. [11] referred to several potential low cost adsorbents such as limestone, silicas, resins, clays, zeolites, and so forth.

Textural properties (e.g., low cost, high mechanical and chemical stability, availability, and ion exchange capability) confer good adsorption efficiencies to natural clays [11,16]. Tunisian Illitic, kaolinitic, and smectitic clays were used in the adsorption of volatile organic compound [17], heavy metals [11,16], and in the purification of industrial phosphoric acid [18].

Photo-Fenton or photo-assisted Fenton process has been reported as one of the fastest and most economical treatment processes. Using safe and environmentally benign reagents such as Fe^{2+} and H_2O_2 , it produces $\cdot\text{OH}$ groups in the presence of irradiation with sunlight or an artificial light source [19]. The shortcomings of the homogeneous Fenton process reside in its requirement for high Fe concentration (between 50 and 80 ppm). This is well above the limits set by European Union directives, which allow a maximum of 2 ppm Fe in treated water to be discharged directly into the environment [19,20]. Therefore, the replacement of such a homogeneous catalyst by a heterogeneous system containing the active phase is needed. Different porous materials, including zeolites (synthetic and natural), clays, polymers, silica, carbon, or resins are among the best supports to disperse active phases [9,21,22]. Among these materials, clay minerals containing iron (i.e., pillared interlayered clays) are proposed as good candidates (as heterogeneous catalysts) for the oxidative degradation of organic compounds through the Fenton reaction [21,23]. The application of clay minerals as heterogeneous photo-Fenton catalysts is a very promising technology for the treatment of water containing organic pollutants because of the high surface areas, easy control, and high porosity [19]. In our previous works, iron-pillared Tunisian clays showed promising adsorption capacity and excellent photocatalytic ability for the removal

of organic pollutants such as Congo red, Malachite green, and phenol in aqueous solution [19,20]. Natural Fe-rich clays could also be used as heterogeneous catalysts in the Fenton-like reaction. In fact, Tejera-Esghira Fe-rich clay is an ideal candidate that can be used as a Fenton catalyst because of the distribution of iron oxide species on the clay surface.

The application of natural clay as an adsorbent or as a Fenton catalyst is an attractive and innovative alternative because of the low cost and widely available material with high iron content, a key in the Fenton reaction mechanism. Those characteristics agree well with some of the “green chemistry” principles, such as the use of alternative feedstocks that are more innocuous and the design of eco-compatible chemicals [24]. In this context, this study has been undertaken to evaluate the potential use of natural Tunisian clay (RC) as natural adsorbent for Cd(II) and 2-CP removal in aqueous systems. Acid-activated natural red clay (AC) was also tested for the adsorption of Cd(II) ions in batch mode. The effect of metal ion concentration was investigated. Langmuir and Freundlich isotherms were used to analyze the equilibrium data. The natural red clay (i.e., RC) was used as heterogeneous photo-Fenton for the degradation of 2-CP from aqueous solution. The kinetics and mechanism of the degradation of 2-CP were considered as well.

2. Experimental section

2.1. Materials and chemicals

The RC used in this study was sampled in Jebel Tejera-Esghira deposits located in the Medenine district, South-eastern Tunisia (Fig. 1). From a geological point of view, those clays were attributed to the Lower Triassic, a very thick series with dominant sandstone alternating with red clays and some silty intercalations. The outcrops extended from the Beni Kheddache cliff to the J. Tebaga of Medenine; it occupies the anticline of the J. Tejera site. The outcropping feature of the Jebel Tejera-Esghira was estimated to be 80 m-thick deposits of ferruginous clays (Fig. 1).

The natural clay was first purified by dispersion in water, decantation, and extraction of the desired fraction ($\leq 2 \mu\text{m}$ -sized fraction) [24,25]. The cation exchange capacity of this material was 18.66 meq/100 g [6].

To enhance its textural properties (i.e., porosity, surface area, and so on), natural clay was also acid activated as follows: 10 g of powdered clay was suspended in 40 mL of distilled water. A solution of 6 M of H_2SO_4 was added dropwise at 7 °C under continuous stirring at 200 rpm for 4 h [2,17,26]. Then, the acid-treated clay was repeatedly washed to obtain a pH = 6. Finally, the obtained samples were dried at 60 °C and stored for further analyses.

2.2. Physicochemical characterization

Chemical composition and textural properties of both natural and activated clay were analyzed using X-ray fluorescence (ARL1 9800 XP spectrometer), powder X-ray diffraction (Philips1 PW 1710 diffractometer, $\text{K}\alpha$, 40 kV/40 mA, scanning rate of 2θ per minute). Specific surface

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