



Simultaneous sorption of 4-nitrophenol and 2-nitrophenol on a hybrid geocomposite based on surfactant-modified pillared-clay and activated carbon



Sabiha Hamidouche^a, Omar Bouras^a, Faiza Zermane^a, Benamar Cheknane^a, Mohamed Houari^a, Jean Debord^b, Michel Harel^{c,d}, Jean-Claude Bollinger^{e,*}, Michel Baudu^e

^aLaboratoire Eau Environnement et Développement Durable, Faculté de Technologie, Université Blida 1, BP 270, 09000 Blida, Algeria

^bService de Pharmacologie-Toxicologie, Hôpital Dupuytren, 2 Avenue Martin Luther King, 87042 Limoges, France

^cInstitut de Mathématiques de Toulouse (UMR CNRS 5219), 31062 Toulouse, France

^dESPE, Université de Limoges, France

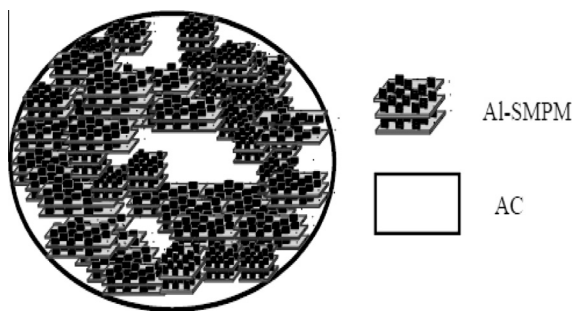
^eUniversité de Limoges, Groupement de Recherche Eau Sol Environnement (GRESE), Faculté des Sciences & Techniques, 123 Avenue Albert Thomas, 87060 Limoges, France

HIGHLIGHTS

- Solids are produced from surfactant-modified pillared clay and activated carbon.
- These mesoporous sorbents prove to be efficient for both 2- and 4-nitrophenol.
- According to the Freundlich model, sorption capacities increase as pH decreases.
- Binary mixture sorption is modeled using an extended Freundlich model.
- Competing sorption effects are displayed in binary mixtures of 2NP and 4NP.

GRAPHICAL ABSTRACT

These mesoporous hybrid sorbents prove to be efficient for both 2- and 4-nitrophenol; competing sorption effects are displayed in binary mixtures, they are modeled using an extended Freundlich model.



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ABSTRACT

New generations of hydrophobic composite geomaterials intended to sorb certain hazardous wastes are assembled by mixing surfactant-modified aluminum-pillared montmorillonite (Al-SMPM) with activated carbon (AC). The sorption of 4-nitrophenol (4NP) and 2-nitrophenol (2NP) on these sorbents was studied in both single and binary component systems from aqueous solutions using batch tests. Results showed the efficiency of these composite sorbents relative to the two phenolic compounds, with a dominant contribution from Al-SMPM. In single-component systems, sorption isotherms of 4NP and 2NP as a function of pH were analyzed with the Freundlich equation, whose statistical interpretation was also developed. Sorption capacities sharply decreased when the solution pH value was raised from 3 to 9. In binary-component systems, the fit between measured and predicted simultaneous sorption capacities of both 4NP and 2NP indicated that the Sheindorf–Rebhun–Sheintuch model, an extended Freundlich model, is fully applicable. The 2NP favorably influences the sorption of 4NP. The synergistic effect between these two nitrophenol compounds has been confirmed by the higher competition coefficients, which increase with increasing pH. A number of hypotheses, based on the Abraham's solvation parameters model, have been advanced to discuss this mechanism.

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* Corresponding author. Tel.: +33 624 307 201; fax: +33 555 457 203.

E-mail address: jean-claude.bollinger@unilim.fr (J.-C. Bollinger).

1. Introduction

Phenol and its derivatives, such as mononitrophenols, constitute hazardous wastes and high-priority pollutants given their harmful effects on organisms at low-concentration exposure. They are mainly used in a range of applications, including chemicals, pharmaceuticals, petroleum, paper, wood, rubber, dyes, pesticides, explosives and insect control products. Due to their high solubility, low biodegradability, weak ionization capacity and low vaporization pressure, phenol and its derivatives are commonly detected in surface water, and both agricultural and industrial wastewater.

According to a World Health Organization recommendation, the permissible concentration of phenolic content in tap water is $1 \mu\text{g dm}^{-3}$. More specifically, nitrophenolic compounds are highly toxic components of water and soil pollutants and moreover have been classified as priority hazardous pollutants due to their toxicity to organisms even at low concentrations [1]. In particular, 2-nitrophenol (2NP) and 4-nitrophenol (4NP) have been listed by United States Environmental Protection Agency [2] within the 126 priority organic pollutants under the Clean Water Act (CWA), with a maximum allowable concentration of 20 ppb in the environmental matrix. For this reason, 4NP and 2NP have been selected herein as the target organic contaminants.

Various processes have been employed to remove phenolic compounds from aqueous media; these include advanced oxidation [3], membrane filtration [4], biological degradation [5], electrochemical oxidation [6], photocatalytic degradation [7] and sorption onto activated carbon (AC) [8]. As a conventional method of pollutant removal, sorption is considered a promising technology; it has also been developed as an efficient and economical method for removing pollutants from wastewater and water supply [9].

Due to their hydrophilic nature, natural clays such as montmorillonites have very low sorption capacities, towards non-polar organic molecules or anionic molecules. Montmorillonite minerals carry negative surface charges that are compensated by exchangeable cations, particularly Na^+ and Ca^{2+} , that are mainly present within the interlayer space. The use of raw bentonites in the removal of organic compounds such as phenols [10] has been previously reported in the literature.

Over the past 20 years, many reports have shown that both clays and modified clays are cost-effective in the sorption of several contaminants as a result of being hydrophobic and organophilic, readily available and environmentally stable. The structural diversity of organic-inorganic modifications in surfactant-modified pillared-montmorillonites (SMPM) offers the potential to increase sorption capacities. Many studies have focused on using SMPM to eliminate organic pollutants in a wide variety of environmental aqueous solutions, including: lead and chlorobenzene [11]; humic acids and pentachlorophenol [12]; phenolic compounds [13–22].

It was recently demonstrated that modified powdered clays could be shaped as gel-like beads [23,24] or grains [25,26] by means of encapsulation or wet granulation, respectively. These solids have in fact been used in the dynamic sorption of pentachlorophenol and some organic dyes.

In binary component systems, the selective sorption of solutes or competition between them for sorption sites is observed. Until now, only a few studies have been conducted in binary systems on the simultaneous sorption of micropollutants from their solutions by inorganic, organic and/or organic-inorganic clays [12,25–28]. As such, other conventional materials like activated carbons (AC) may also be considered as potential sorbents thanks to their large specific surface areas and strong affinity towards organic pollutants [8].

The preparation of solid geocomposites by combining several types of materials possibly creates properties that cannot be

obtained using a single type of solid material. Such geocomposites could thus be considered as powerful competitors to conventional AC in the water-based treatment of industrial effluents and moreover would offer relevant functionalities.

Previous investigations on reinforced geocomposites have focused on the hybridization of clay with AC and cement for reinforcing final solids [29–32]. These materials have been chosen herein in order to associate the following properties:

- the mechanical characteristics supported by both AC and the surfactant-modified, aluminum-pillared montmorillonite (Al-SMPM);
- Al-SMPM is able to reduce the mobility of phenolic compounds, since these modified clays present hydrophobic surfactant alkyl chains;
- AC is characterized by a strong affinity towards non-polar molecules.

In order to understand the behavior of these new generations of hydrophobic geocomposite sorbents with respect to organic pollutants under environmental conditions, we investigated the sorption of 4NP and 2NP in both single and binary-component systems from aqueous solutions. The kinetic and sorption isotherms have been studied at various 4NP/2NP ratios and pH values.

2. Materials and methods

2.1. Solid sorbents

The starting material was a bentonite extracted from the Roussel site in Maghnia (western Algeria) supplied by ENOF Company (Algeria); its characteristics were described previously [33].

The aluminum-pillared montmorillonite was synthesized from this raw bentonite according to previously published methods [33–35].

A 0.2% (w/v) aqueous solution of the cationic surfactant was prepared by dissolution of the appropriate amount of cetyltrimethylammonium bromide (CTAB) in distilled water. The purity of CTAB was >99% and it was used without further purification. In order to increase its hydrophobicity by coadsorption with a surfactant, a suspension of aluminum-pillared montmorillonite was treated with CTAB (0.5%, w/w) at room temperature. The resulting solid, named Al-SMPM, was separated by vacuum filtration, washed several times with distilled water, dried at 40 °C for at least 72 h, and was finally ground and protected from light.

The activated carbon (AC) used in the production of the geomaterial composites is a microporous powder of a very fine granular structure, prepared by the activation of wood coal and purchased from Aldrich. Its main characteristics are presented in Table 1.

The hybrid geomaterials were prepared by mixing Al-SMPM and AC as a 1% (w/v) aqueous suspension with different weight ratios ($F = 10\%$, 50% and 90%).

These materials have all been chosen to best associate the mechanical characteristics (as supported by modified clay) with sorption properties (as supported by both AC and Al-SMPM). After mixing at moderate speed with a magnetic stirrer (IKA model RH-KT/C) for 48 h, all solid sorbents were separated by vacuum filtration, washed several times with distilled water, dried at 40 °C for at least 72 h, and ultimately ground, homogenized and protected from light before use.

We have initially prepared three formulations ($F = 10\%$, 50% and 90%) in order to examine the effect of adding Al-SMPM into the matrix. From the results (detailed data not shown) it is clear that the adsorption capacity increased with the amount of Al-SMPM

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