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Photodegradation of cotrimaxazole by clinoptilolite-supported nickel oxide

Narges Arabpour^{a,b}, Alireza Nezamzadeh-Ejhieh^{a,b,c,*}

^a Department of Chemistry, Shahreza Branch, Islamic Azad University, P.O. Box 311-86145, Shahreza, Isfahan, Iran¹

^b Young Researchers and Elite Club, Shahreza Branch, Islamic Azad University, Shahreza, Iran

^c Razi Chemistry Research Center (RCRC), Shahreza Branch, Islamic Azad University, Isfahan, Iran

ARTICLE INFO

Article history:

Received 27 July 2015

Received in revised form 4 March

2016

Accepted 26 April 2016

Available online 3 May 2016

Keywords:

Nanoparticles

Clinoptilolite

NiO

Cotrimaxazole

Photodegradation

Trimethoprim

Sulfamethoxazole

ABSTRACT

Photocatalytic activity of NiO incorporated into clinoptilolite nanoparticles (NiO-NCP) was studied in the photodegradation of cotrimaxazole pharmaceutical capsule (a mixture of trimethoprim and sulfamethoxazole) in aqueous system under medium Hg-lamp irradiation. Samples were characterized by XRD, FT-IR, UV-Vis-DRS, TEM and BET. The effects of some key experimental parameters affecting the photodegradation extent of the pollutant were studied and the best results were obtained at 13.2% NiO loaded onto NCP, 0.2 g L⁻¹ of the NiO_{13.2%}-NCP catalyst, 50 folds diluted cotrimaxazole solution at pH 3. COD of the pollutant solution was decreased from its initial value of 1930–700 mg L⁻¹ during 4 h photodegradation process (corresponds to 64% degradation of cotrimaxazole). The peak area of the main peak in HPLC chromatogram was also decreased during the irradiation process. HPLC and COD results are in accordance with decrease in UV-vis absorbance during the photodegradation process, confirming the degradation of cotrimaxazole into smaller fragments.

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1. Introduction

The contamination of water, air and soil by different organic and inorganic pollutants is a key concern for environmental and human health (Giri et al., 2010). Antibiotics, extensively used in human and veterinary medicine, as well as in aquaculture for the prevention and treatment of microbial infections are frequently detected in aquatic systems (Dolar et al., 2012; Tong et al., 2011). Cotrimaxazole, as environmentally relevant, is a combination of 1:5 of trimethoprim and sulfamethoxazole (Barnes et al., 2013). Sulfamethoxazole and trimethoprim are two human-use antibacterial compounds that are often prescribed together to treat various bacterial infections. Sulfamethoxazole belongs to the sulfonamide class of antibacterial compounds, while trimethoprim does not belong to any

specific class (Ryan et al., 2011). In general, overuse and misuse of antibiotics cause to entering them into water, seawater, groundwater and drinking water via different ways such as: direct run off and excretion of feces or urine as un-metabolized original drugs or active metabolites and degradation products. The occurrence of the cumulated antibiotics in the natural environments causes irreversible adversity by directly threatening ecosystem function and decreasing the rate of pollutants degradation. This also increases the resistance of bacteria against drugs, spreading the antibiotic resistance genes among bacterial populations. These phenomena threaten the human's health. As a result, there is growing interest in the development of novel technologies to efficiently transform of these compounds to non toxic and pharmaceutically inactive byproducts (Khaleel et al., 2013). Conventional

* Corresponding author at: Department of Chemistry, Shahreza Branch, Islamic Azad University, P.O. Box 311-86145, Shahreza, Isfahan, Iran. Tel.: +98 31 53292515.

E-mail address: arnezamzadeh@iaush.ac.ir (A. Nezamzadeh-Ejhieh).

¹ Tel.: +98 31 53292515; fax: +98 31 53291018.

<http://dx.doi.org/10.1016/j.psep.2016.04.025>

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methods for water disinfection such as chlorination and ozonation inevitably form harmful disinfection by-products which probably provide an unintended health hazard (Gan et al., 2013; Lester et al., 2010). Advanced oxidation processes (AOPs) overcome to these problems by degrading of organic pollutants present in aqueous solution via generation of highly reactive radical species such as hydroxyl radicals (HO^\bullet). These powerful oxidizing radicals capable of attacking a wide variety of organic molecules to convert them into smaller fragments and finally into CO_2 and H_2O (Safari et al., 2015; Giri et al., 2008; Nezamzadeh-Ejhieh and Moazzeni, 2013; Kuo and Hsiao, 2015; Khan et al., 2015; Doocoy and Sharratt, 2004). In heterogeneous photocatalysis, as one AOP method, irradiating a semiconductor by a suitable photon, the generated electron-hole pairs can trap by the oxygen and hydroxyl present in solution to form superoxide ($\text{O}_2^{\bullet-}$) and hydroxyl radicals (OH^\bullet with $E^\circ = 2.80 \text{ eV}$) (Giri et al., 2008; Kumar and Bansal, 2012; Riaz et al., 2012). Recent advances in photocatalysis have been published in a review (Buthiyappan et al., 2016).

NiO, with excellent durability and electrochemical stability, has band gap energy value in the range of 3.4–3.8 eV (average 3.5 eV; valence band [3.1 eV] and conduction band [−0.5 V]) that make it suitable for photocatalytic processes. On the other hand, the optical transition in NiO takes place through direct inter-band transition (Patil and Kadam, 2002). In addition, NiO acts as a promoter for the generation of OH radicals (Zhang et al., 2009). In this work, for increasing the photoactivity of NiO it was supported on clinoptilolite nanoparticles. The obtained catalyst was used in the photodegradation of an aqueous solution containing cotrimaxazole drug and the effects of some key operating experimental parameters were studied. Clinoptilolite is a very attractive material for environmental applications, especially for removing heavy metal cations from wastewater, because of its high cation exchange capacity (CEC). It is also an abundant zeolite with low extraction cost and high chemical stability, caused to increasing attention for using it by researchers (Akgul and Karabakan, 2010; Garcia-Basabe et al., 2010).

2. Experimental

2.1. Reagents

An Iranian natural clinoptilolite tuff that originates from the Semnan province in north-east of Iran was purchased from Afrand Touska Company (Isfahan, Iran). Cotrimaxazole (trimethoprim/sulfamethoxazole $\text{C}_{14}\text{H}_{18}\text{N}_4\text{O}_3/\text{C}_{10}\text{H}_{11}\text{N}_3\text{O}_3\text{S}$) pharmaceutical capsule (500 mg) was obtained from Chemidarou Company in Iran. Nickel(II) chloride six hydrate and other reagents used in the present study from Merck were purchased from Normal Labo Co. (Isfahan, Iran). Sodium hydroxide or hydrochloric acid solution was used for adjusting pH. Distilled water was used throughout the experiments.

2.2. Preparations

Natural clinoptilolite zeolite was mechanically pretreated, crushed in an agate mortar and sieved in analytical sieves, for separation of the particles with mesh (<400). This powder will refer 'micronized zeolite/particles' in next sections. Nanoparticles of clinoptilolite were prepared using a planetary ball mill (PM100; Retsch Corporation, Steel balls) in a dry milling method at the milling duration of 6 h, speed of 600 rpm

and balls-to-powder ratio of 4. The obtained NCP powder was added to a 250 mL beaker containing 150 mL water. A magnet was then added and the resulted suspension was heated at 70°C on a magnetic stirrer for 8 h ($n=3$) to eliminate water soluble and magnetic impurities that entered in NCP powder during ball mill process. At definite time intervals, magnet was separated out of suspension and the adsorbed impurities were removed. The suspension was finally centrifuged and air dried. For ion exchange experiments, 4 g of NCP was added to 100 mL Ni^{2+} solutions (as chloride salt) with concentrations of 0.1, 0.2, 0.3 and 0.5 M. The prepared suspensions were shaken for 24 h. After centrifuging, the solid materials (Ni-exchanged NCPs) were air dried for 6 h and calcinated in a furnace at 450°C for 12 h to prepare NiO-nanoclinoptilolite catalysts (NiO-NCP) with different NiO contents. Briefly, 'micronized clinoptilolite', 'clinoptilolite nanoparticles', 'NiO-contained micronized clinoptilolite' and 'NiO-contained clinoptilolite nanoparticles' will show as 'CP', 'NCP', 'NiO-CP' and 'NiO-NCP', respectively.

2.3. Characterizations

Following methods/instruments were used for preparation/characterization of samples: Planetary ball mill (SP4, Farapajohesh, Iran); atomic absorption spectrometer (Analyst 300, Perkin Elmer, USA), FT-IR spectrophotometer (Spectrum 65, Perkin Elmer, USA), UV-Vis DRS spectrophotometer (V-670, JASCO, Japan), XRD diffractometer (D8ADVANCE, Bruker, Germany) with $\text{Cu K}\alpha$ radiation: 1.5406 \AA source; filter: Ni; precision: $\pm 0.01^\circ$ and step size 0.05° , transmission electron microscope (TEM) (CM10, Philips, Netherlands) (40–100 kV accelerating voltage adjustable in 20 kV increments: $\pm 60^\circ$ specimen tilt; magnification range: $20\times$ – $510,000\times$, a double beam UV-Vis spectrophotometer (Cary100, Australia), BET instrument (Nova 1200, Spain), determination of the pH of point of zero charge, pH_{pzc} , for the catalyst by the reported procedure in literature (Nezamzadeh-Ejhieh and Zabihi-Mobarakeh)

2.4. The photocatalytic activity

To prepare cotrimaxazole solution, the content of a 400 mg tablet was dissolved in water and it was shaken for 30 min, filtered in 100 mL volumetric flask and diluted to the mark. The 10, 50 and 100 times diluted solutions were prepared via serial dilution method. Photodegradation experiments were performed with a photocatalytic reactor system equipped with a 50 mL Pyrex-glass cell. During the photocatalytic degradation experiments, 20 mL of 50 times diluted solution (as the optimized concentration which assigned cotrimaxazole solution later) containing 0.2 gL^{-1} of the NiO-NCP at initial pH 5.5 was irradiated with a medium Hg lamp (30 W). The lamp was located in 30 cm length with respect to the cell. In specific time intervals, an aliquot suspension was withdrawn and centrifuged at 5000 rpm to remove solid particles. The degradation extent was calculated according to the following formula based on the absorbance of the solutions (at $\lambda_{\text{max}} = 264 \text{ nm}$) before and after irradiation:

$$\text{Degradation\%} = \left[\frac{C_0 - C_t}{C_t} \right] \times 100 = \left[\frac{A_0 - A_t}{A_t} \right] \times 100 \quad (1)$$

In this equation, C_0 and C_t are the initial and final concentrations of cotrimaxazole at any time 't', respectively. A_0

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