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Synthesis of nano zinc oxide on granular porous scoria: Application for photocatalytic removal of pharmaceutical and textile pollutants from synthetic and real wastewaters

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ABSTRACT

ZnO nanoparticles were synthesized by sol gel method on granular porous natural scoria. Structure and morphology of scoria, synthesized ZnO nanoparticles, and ZnO/scoria nanocomposite were studied by X-ray diffraction, scanning electron microscopy and energy-dispersive X-ray spectroscopy. Approximately 49 mg of nano ZnO was immobilized per gram of scoria. Photocatalytic performance of ZnO/scoria nanocomposite was examined for phenazopyridine and Acid Blue 113 removal from synthetic aqueous solutions under UV irradiation in a continuous open channel reactor. Removal of Acid Blue 113 in pH of 5.5, flow rate of 15 ml/min and Acid Blue 113 concentration of 20 mg/L was obtained 70.78%. Removal of phenazopyridine in pH of 6.0, flow rate of 15 ml/min and concentration of 15 mg/L was obtained 72.55%. Also, prepared photocatalyst was used for treatment of pharmaceutical and textile pollutants from real wastewaters. COD reductions of these wastewaters were 40.55% and 58.25% respectively in their natural pH, 5 ml/min with initial COD of 800 and 510. Results reveal that ZnO/scoria nanocomposite is efficient for photocatalytic removal of pharmaceutical and textile pollutants with reusability.

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1. Introduction

Photocatalytic oxidation is a strong process for the treatment of a wide variety of organic contaminations in water and wastewater [1–4]. In photocatalytic oxidation, the highly reactive hydroxyl radicals (•OH) which are the main species responsible for the oxidation of organic pollutants, are generated as a result of the interaction between a semiconductor photocatalyst and UV or visible irradiation [5–9].

Zinc oxide (ZnO) due to its excellent chemical and thermal stability [10] has attracted tremendous attention among researchers as a photocatalyst agent. In recent studies it has been shown that ZnO is more effective than TiO_2 as a photocatalyst [11–12]. In this research, ZnO has been selected as photocatalyst because of its high catalytic efficiency, low cost, and non-toxic nature.

Waste drugs are disposed into water through sewer or direct disposal. They are resistant to degradation [13]. The heterogeneous photocatalytic process is one of the ideal techniques to degrade drugs. Phenazopyridine was selected as model drug in this research. The presence of the azo group makes phenazopyridine a stable compound [14]. After a successful test of an artificial drug wastewater, real pharmaceutical wastewater of antibiotic factory was tested in this research. Antibiotics are of specific concern because they can induce microbial aberrations [15].

The dye Acid Blue 113 (AB113) has many applications as a coloring agent in textile industry [16]. It is utilized extensively in various textile dying industries to colorize wool, silk and polyamide fiber in a neutral or acidic bath [16, 17]. World health organization, International Agency for Research on Cancer (IARC), has identified AB113 as a potential carcinogenic agent [18]. Therefore, it was selected as a model dye. Also, wastewater of textile industry was tested in this study.

In this study, scoria stone, a kid of porous lava, was introduced as a new support for ZnO immobilization. ZnO/scoria nanocomposite usage in photocatalytic process was investigated for removal of phenazopyridine and Acid Blue 113 as artificial wastewater for the first time in this research. Finally, pharmaceutical and textile wastewaters as real samples were treated by the prepared photocatalyst.

2. Experimental

2.1. Materials

Whole rock samples of scoria were taken from Qorveh mine in Kordestan, west of Iran. Phenazopyridine was purchased from Shahre

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Daroo Company in Tehran, Iran. Acid Blue 113 was obtained from Sabet Alvand Company in Hamedan, Iran. Zink acetate dihydrate, oxalic acid, ethanol, HNO_3, $NaNO_3$, NaOH, H_2SO_4, $K_2Cr_2O_7$, $HgSO_4$, Ag_2SO_4, and HCl were purchased from Merck.

Pharmaceutical wastewater which contains antibiotics especially cefalexin was obtained from Dana Pharmaceutical Company (Tabriz, Iran) with yellow color, chemical oxygen demand (COD) 800 mg/L, conductivity of 2.67 mS/cm and 4.8 pH. The textile wastewater containing Acid Blue 113 was obtained from Towers Factory in Tabriz, Iran. The physical and chemical specifications of wastewater were: blue color, pH of 8.8 ± 0.2, conductivity of 10.41 mS/cm, and chemical oxygen demand (COD) of 510 mg/L.

2.2. Measurement of pollutants

2.2.1. Measurement of phenazopyridine and Acid Blue 113

The absorbance of phenazopyridine and AB113 aqueous solution were recorded in the range of 200–800 nm and 420 and 566 nm were found as the maximum absorbance wavelength for phenazopyridine and AB113 respectively. Using known aqueous solutions of phenazopyridine and AB113 with different concentrations and their absorbances at maximum wavelength Absorbance = *f*(Concentration) calibration curves were plotted. These curves were used to convert absorbance data to drug and pollutant concentration in the solutions. The removal efficiency of pollutants was defined using Eq. (1):

$$R(\%) = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

where R is pollutant removal efficiency, C_0 (mg/L) is initial concentration of pollutant, and C_t (mg/L) is its concentration at t min.

2.2.2. Measurement of COD of real wastewaters

COD measurement was performed according to the standard method using palintest photometer. Potassium dichromate was used as oxidizing agent. Concentrated sulfuric acid was used for providing the primary digestion catalyst. The secondary catalyst was silver sulfate ($AgSO_4$). After digesting of samples for 2 hours at 150 ± 2 °C, the samples are cooled and the color was measured using a palintest photometer.

2.3. Scoria preparation

Scoria samples were washed with tap water until a clear washing effluent was obtained. Then they were washed with distilled water and were retained 10 h in distilled water to remove any dust and other waste-soluble impurities. Then, the samples were treated by 0.5 M acid (HCl) for 10 h for further purification. After that, stones were washed with distilled water and were dried at 80 °C in an oven (GRIFFIN, United Kingdom).

2.4. ZnO/scoria synthesis and analysis

Solutions of zinc acetate with different concentrations (0.1, 0.2, and 0.3 M) were prepared by dissolving zinc acetate dihydrate [$Zn(CH_3COO)_2 \cdot 2H_2O$] in 100 ml of ethanol and heating the mixture in a water bath at 60 ± 5 °C for 30 min to obtain a transparent sol (solution A). The oxalic acid solution was prepared by dissolving oxalic acid (1.575 g) in 50 ml of ethanol while stirring with magnetic stirrer in a water bath at 50 ± 5 °C for 30 min (solution B). Solution (B) was slowly added drop-wise to solution (A) under vigorous magnetic stirring until a gel was formed in a water bath at 50 ± 5 °C [19].

ZnO thin films were deposited on scoria stones (approximately 1.0 cm in diameter and 1.5–2.0 g weight) by the dip-coating method for 5 min. After coating each layer, the film was dried at 80 °C in an oven for 30 min to evaporate the solvent from it. Dip-coating and drying were repeated 1–5 times. The coated films were subsequently calcinated by heating at 400 °C for 2 h to remove undesired species and to achieve ZnO thin films. The amount of the mounted ZnO over scoria was estimated by the change of its weight before and after ZnO immobilization using a digital scale (Shimadzu AEL-200, Japan) [20].

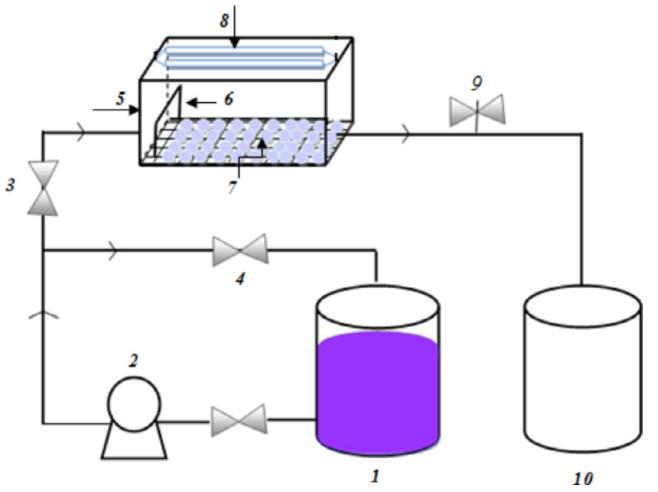

Fig. 1. Experimental set up. (1) Reservoir, (2) pump, (3) inlet valve, (4) by pass valve, (5) photoreactor, (6) vertical wall, (7) granular ZnO/scoria nanocomposite, (8) UV-C lamps, (9) sampling valve and (10) treated water reservoir.

The structural characterization of scoria, synthesized ZnO nanoparticles, and ZnO/scoria nanocomposite were carried out by X-ray diffraction (XRD) pattern using Siemens D-500 (Germany) X-ray diffractometer with Cu K_a radiation at a wavelength of 0.15406 nm. Morphology of scoria, ZnO nanoparticles and ZnO/scoria nanocomposite were studied by scanning electron microscope using a MIRA3 FEG-SEM Tescan (Czech). To determine the particle size distribution, the obtained SEM micrograph was analyzed using Manual Microstructure Distance Measurement Software (Nahamin Pardazan Asia Company, Ian). Energy dispersive X-ray spectroscopy (EDS) was used to confirm the syntheses of ZnO/scoria nanocomposites and its stability after multiple uses using a MIRA3 Tescan (Czech). Also, the chemical composition of the scoria was studied by *XRF* analysis *using X-Ray fluorescence diffractometer (Philips PW 2404, Netherlands).* Energy dispersive X-ray spectroscopy (EDS) was used to confirm the syntheses of ZnO/Scoria nanocomposites and its stability after multiple uses using a MIRA3 Tescan (Czech). Surface area of the prepared nanocomposite was obtained through the Brunauer–Emmett–Teller (BET) with micromeritics Gemini 2375 (USA) analyzer.

2.5. Reactor structure and decolorization process

The continuous photoreactor, used in this study, has been depicted in Fig. 1. The photoreactor consisted of a rectangular container with the dimensions of 20 cm × 8 cm × 8 cm with the effective volume of 300 ml. In the bottom of the photoreactor, a reticulated sheet was placed and it was covered by 160 g ZnO/scoria nanocomposite. Two 6 W UV-C mercury lamps (Taiwan) were placed 7 cm above of the dye solution surface. Also, in the inlet of reactor a vertical wall was used to avoid from canalization of the feed solution. Pollutant solution was fed into the reactor by a pump and UV radiation was started and treated solution was collected. Samples were withdrawn every 15 min and its removal efficiency was calculated according to Eq. (1). Effect of different variables including pH, feed flow rate, and pollutant concentration were investigated. Real wastewaters were treated in the same photoreactor but effluent flow rate was 5 ml/min and samples withdrawn after 1 h treatment and were analyzed by COD measurement.

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