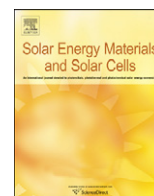




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Fe^{III} supported on ceria as effective catalyst for the heterogeneous photo-oxidation of basic orange 2 in aqueous solution with sunlight

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ARTICLE INFO

Article history:

Received 26 August 2009

Received in revised form

18 June 2010

Accepted 27 June 2010

Available online 10 July 2010

Keywords:

Fe^{III}-ceria catalyst

Iron-cerium oxide

Heterogeneous Fenton-like reaction

ABSTRACT

Fe^{III} supported on ceria as an effective catalyst for oxidation was prepared and used for the degradation of basic orange 2 azo textile dye (BO2). BO2 was chosen as a model pollutant and the catalytic oxidation was carried out in a batch reactor using hydrogen peroxide as the oxidant at pH 3. The influent factors on BO2 oxidation, such as catalyst dosage, H₂O₂ concentration, and BO2 concentration were studied by considering the BO2 conversion and chemical oxygen demand (COD) removal. The Fe^{III}-ceria catalyst showed a high catalytic activity for the oxidation of BO2 in aqueous solution. It was observed that the solution became colorless after 5 h of oxidation and over 90% COD removal was achieved with all the Fe^{III}-ceria catalysts used under dark conditions in the catalytic oxidation system. The catalytic removal of BO2 during BO2 oxidation was improved under solar radiation, which notably increased the BO2 degradation rate. Consecutive BO2 oxidation cycles carried out with the same Fe^{III}-ceria catalyst and untreated fresh dyestuff solution showed that the catalyst had good stability and good degradation performance, thus evidencing the possibility of being used in continuous processes. This study showed that the Fe^{III}-ceria catalytic oxidation process is an efficient method for the treatment of BO2 aqueous solutions.

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1. Introduction

The development of clean technologies compatible with the environment for wastewater treatment and water recycling is imperative nowadays. There are several advanced oxidation processes that represent an alternative when common wastewater treatment technologies are insufficiently effective. Amongst these technologies, Fenton's reagent (H₂O₂ in the presence of added Fe²⁺) offers a cost effective source of highly reactive oxidizing species that are responsible for the degradation of organic pollutants present in aqueous effluents. However, the iron used in the homogeneous catalytic oxidation of the Fenton process needs to be removed to prevent it from forming sludge at the end of the wastewater treatment. This can be avoided by employing the heterogeneous catalytic oxidation in the Fenton-like process, which also offers the possibility of recovering the heterogeneous iron catalyst by filtration after treatment. The catalyst can be recovered also by other processes (e.g. simple

sedimentation), if a slurry reactor is used; otherwise, a simple packed bed reactor can be employed. Several studies have reported the development of heterogeneous catalysts (prepared by the incorporation of iron ions or iron oxide into diverse support materials) applied for the decomposition of H₂O₂ and for the degradation of organics and dyestuff. The iron catalyst used in heterogeneous Fenton process are supported on metal oxide surface such as TiO₂, Al₂O₃ and CeO₂ [1–4], synthetic zeolites [5], pillared clays [6], laponite and bentonite clay-based Fe-nano-composites [7,8], ceramic materials [1] activated carbon [9], resin [10], etc. Also, the heterogeneous photo-Fenton enhances Fenton's reaction destroying completely pollutants in liquid and gaseous phase, using solar or artificial light illumination [11]. Different iron compounds, such as Fe⁰, Fe₃O₄, Fe₂O₃, and FeOOH, have been reported to promote the Fenton reaction for the oxidation of organic compounds [12,13].

The objective of this work was to study the oxidation of the BO2 textile dye in mildly acidic aqueous media by heterogeneous Fenton and photo-Fenton-like processes employing an iron oxide catalyst supported on cerium oxide (Fe^{III}-ceria catalyst). The effects of iron leaching, catalyst dosage, dye concentration, catalytic activation of H₂O₂, H₂O₂ concentration and catalyst

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recycling were studied as influencing factors on the BO2 conversion.

2. Experimental

2.1. Material and characterization

BO2 was provided by the local textile industry (93% purity). $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ and $\text{Ce}(\text{NO}_3)_4 \cdot 6\text{H}_2\text{O}$ (99%, purity) were purchased from Aldrich. Fe^{III} -ceria catalysts were prepared with various molar concentrations of iron nitrate as precursor salts mixed in different molar ratios of $\text{Fe}(\text{III}):\text{Ce}(\text{IV})$ of 0.25:0.75, 0.5:0.5 and 0.75:0.25. Each molar ratio was mixed in aqueous solution at pH 12 (using sodium hydroxide solution) to form the mix of iron and cerium hydroxide (catalyst). The catalyst was precipitated and kept under vigorous mechanical stirring during 4 h. After filtering and washing three times with distilled water, the catalyst was dried (at 110 °C for 16 h) and finely ground followed by thermal treatment. The calcinations were carried out at 500 °C for 4 h. Three more catalysts were prepared using an ultrasonic bath following the same procedure described above. The catalysts were subjected to ultrasonic stirring for 20 min alternating the agitation with mechanical stirring for the same time during 4 h. The iron loading on the catalyst was measured using the 1, 10 phenanthroline method after dissolution in *aqua regia* (a mixture of hydrochloric acid and nitric acid) in the absence of BO2.

2.2. Catalyst preparation

Fe^{III} -ceria catalysts (iron oxide supported on cerium oxide catalysts), were prepared with different amounts of Fe loading using iron nitrate and cerium nitrate as precursor salts mixed in different molar ratios of $\text{Fe}(\text{III}):\text{Ce}(\text{IV})$ of 0.25:0.75, 0.5:0.5 and 0.75:0.25. Each molar ratio was mixed in aqueous solution at pH 12 (using sodium hydroxide solution) to form the mix of iron and cerium hydroxide (catalyst). The catalyst was precipitated and kept under vigorous mechanical stirring during 4 h. After filtering and washing three times with distilled water, the catalyst was dried (at 110 °C for 16 h) and finely ground followed by thermal treatment. The calcinations were carried out at 500 °C for 4 h. Three more catalysts were prepared using an ultrasonic bath following the same procedure described above. The catalysts were subjected to ultrasonic stirring for 20 min alternating the agitation with mechanical stirring for the same time during 4 h. The iron loading on the catalyst was measured using the 1, 10 phenanthroline method after dissolution in *aqua regia* (a mixture of hydrochloric acid and nitric acid) in the absence of BO2.

2.3. BO2 oxidation

The degradation experiments were carried out employing 0.1 L of solution using several batch reactors (Duran™ with capacity of 0.25 L) set on a planar surface under solar radiation. The reactors were withdrawn at different time periods during dye degradation. The dye solution was fed into the reactor followed by the iron oxide catalysts at a suitable dosage. Then the amount of hydrogen peroxide was introduced into the reactor. At the desired time interval thereafter, liquid sample was taken from the reactor and chemical oxygen demand (COD) was analyzed using the standard methods [14]. COD test interferences due to presence of hydrogen peroxide were avoided by the addition of sodium sulfite (0.01 M Na_2SO_3) to the sample prior COD analysis. The concentration of H_2O_2 in solution was measured by a self-indicating potassium permanganate titration; this was performed in the absence of dye to avoid interferences. Solutions were titrated using standard potassium permanganate solution until the first permanent pink color appeared [15]. The BO2 concentration during oxidation was determined using a calibration curve built at the wavelength of maximum absorbance (456 nm). Also the absorbance spectra were recorded using a spectrophotometer HACH (DR/4000U) at time intervals during degradation time. The reaction was also carried under dark at room temperature (25 ± 5 °C). The experiments, under the sunlight, were carried out between 10:00 and 16:00 h. All the experiments were carried out under similar conditions of solar radiation (sunny and relatively cloudless days)

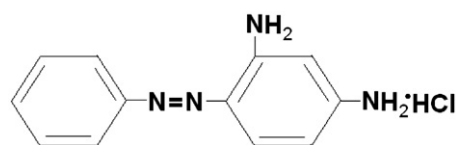


Fig. 1. Molecular structure of basic orange 2.

to avoid differences in the kinetics of degradation due to widely different experimental conditions of photon flux that change during the day. The global sun irradiance averaged during exposure time was measured with a pyranometer (CS300 Campbell Scientific Inc). The accumulated energy, integrated over the time, was calculated using the following equation [16]:

$$Q_{UV,n} = Q_{UV,n-1} + (t_n - t_{n-1}) \overline{UV}_{G,n} \frac{A}{V} \quad (1)$$

where $Q_{UV,n}$ is the accumulated energy per unit of volume (V) incoming the reactor surface (A) for a period t_n (J L^{-1}), t_n the exposure time (s), $\overline{UV}_{G,n}$ the average UV_G during exposure time (W m^{-2}).

Catalyst loading range of 0.1–1.0 g L^{-1} , BO2 initial concentration range of 0.05×10^{-3} – 0.5×10^{-3} mol L^{-1} and hydrogen peroxide range of 2×10^{-3} – 12×10^{-3} mol L^{-1} were employed. Adsorption tests were performed in the absence of H_2O_2 using 0.1 L of BO2 solutions at pH 3 with stirring during 3 h. Blank experiments were carried out only using hydrogen peroxide and BO2 in solution.

3. Results and discussion

The chemical oxidation of the basic orange 2 (BO2), might involve, a complete conversion to CO_2 and H_2O . The BO2 contains an azo group, $-\text{N}=\text{N}-$, as part of its molecular structure connecting two aromatic ring compounds, as depicted by Fig. 1. The chemical name of the BO2 is 1,3-benzenediamine, 4-(phenylazo) with a molecular weight of 248.72 g mol^{-1} ($\text{C}_{12}\text{H}_{12}\text{N}_4\text{HCl}$). This dye is also known as orcozine chrysoidine 4.

3.1. Dye adsorption on catalyst surface

The results show that, in most of the experimental conditions, the adsorption of dye on all the Fe^{III} -ceria catalysts tested is approximately 10% when 0.1 g/L of catalysts was employed at pH 3. It was observed that color removal due to adsorption slightly increased (up to 11%) by increasing dye concentration within the range of 0.1×10^{-3} to 0.5×10^{-3} mol/L BO2 using 0.1 g/L of C2-Us/M catalyst load. Similar adsorption variation was observed when the catalyst load varied between 0.1 and 1.0 g/L of C2-Us/M in a dye solution of 0.1×10^{-3} mol/L BO2 at pH 3 as compared to dye concentration variation. It was also observed that the adsorption equilibrium was achieved after 5 min of contact with 0.1 g/L of all the catalysts tested inside the dye concentration range of 0.1×10^{-3} – 0.5×10^{-3} mol/L BO2; whereas, when the catalyst load was over 0.1 g/L, the adsorption equilibrium was acquired after 2.7 h.

3.2. Influence of iron content and catalyst dosage

The contents of iron (determined by photometry after catalyst digestion with *aqua regia*) present in the final catalysts are shown in Table 1, together with iron leached after 340 min at pH 3. Iron content in the catalyst and iron leaching analyses were carried out in the absence of BO2 dye. XRD were not performed to characterize the catalysts; however, Sahoo et al. [17] reported

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