



Photoelectrocatalytic degradation of acid dye using Ni–TiO₂ with the energy supplied by solar cell: Mechanism and economical studies

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ABSTRACT

This paper reports an investigation into the effect of a number of operating factors on the removal of Acid Red 88 from an aqueous solution through photoelectrocatalysis: photocatalyst dose, dye concentration, pH, bias potential, and electrolyte concentration. The photocatalyst was Ni–TiO₂ applied in suspension to the solution to achieve a larger catalyst surface area. The optimum values for photocatalyst dose, dye concentration, and electrolyte concentration turned out to be 0.6 mg L⁻¹, 50 mg L⁻¹, and 5 mg L⁻¹, respectively. Also, the best pH was found to be 7, and bias potential proved to be best at 1.6 V. The aqueous solution was characterized for its COD and TOC. Photocatalyst efficiency was evaluated using SEM and XRD techniques. The characterization of the post-treatment product using FT-IR, HPLC, and GC-MS studies revealed intermediate compounds. A pathway was proposed for the degradation of the dye. The energy required by the experiment was supplied by solar cells, meaning the money that would have otherwise been spent on electricity was saved. Cost analysis was also done for the treatment process.

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1. Introduction

The dye stuff lost in the textile industry poses a major problem to wastewater sources (Tsai et al., 2001; Bhattacharyya and Sharma, 2005; Chen and Lu, 2007; Aksu et al., 2008; Mu et al., 2009). Over the years many techniques have been developed to treat colored water. These include adsorption, coagulation-flocculation, oxidation, photodegradation, sedimentation, and chemicoagulation (Kobya et al., 2007; Mohan et al., 2007; Zuo et al., 2008). However, these methods are not without their disadvantages. For instance, biological approaches are time-consuming (Can et al., 2006). Absorbents are generally not reusable (Can et al., 2006). Chemicoagulation causes pollution due to inappropriate reactions in wastewater and produced colloids (Kobya et al., 2007). Chemical oxidation is slow and needs reactive materials which are hazardous (Daneshvar et al., 2004). Oxidation methods such as ozonation, photo catalyst, and photo fenton are expensive and uneconomical (Can et al., 2006). For these reasons, researchers have turned to another technique by the name of photoelectrocatalysis (PEC) (Su et al., 2008; Wang et al., 2009). This process generally involves

subjecting a semiconductor used as the photocatalyst to ultraviolet (UV) irradiation. As a result of this, the electrons in the semiconductor are excited from the valence band to the conduction band, thus generating the electron–hole (H⁺/e⁻) pairs (Chandrasekaran and Kamat, 2000; Dong et al., 2010; Yae et al., 2001; Okato et al., 2005; Tachikawa et al., 2007). These photo-generated pairs produce hydroxyl radicals (laqing Li et al., 2006), which convert organic pollutants adsorbed on the surface of the photocatalyst to less harmful products (Manivel et al., 2010).

Nano-sized titanium dioxide (TiO₂) has been widely utilized as a photocatalyst for the degradation of recalcitrant contaminants such as dyes, organic materials, and oils (Zhao and Liu, 2008; Santosa et al., 2005). This is because TiO₂ is comparatively inexpensive, non-toxic, highly stable, and environmentally friendly (Tanaka and Reddy, 2002; Zhang et al., 2004; Yu et al., 2007; Kaur and Gupta, 2009; Manivel et al., 2010), and it has appropriate band-gap (3.0 eV) to adsorb UV-A light at 365 nm (Wang et al., 2010). Nonetheless, the photoelectrocatalytic capability of TiO₂ can be enhanced by doping it with metals such as gold, silver, copper, and Nickel (Esquivel et al., 2011). Doping enhances degradation by forming more electron–hole pairs, keeping the electrons and holes separate (Liu et al., 2000), and extending the light absorption spectrum into the visible region (Kumar et al., 2011). TiO₂ powder is usually applied as a thin film on the anode electrode (Jiao et al.,

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2008; Liu et al., 2009) or on carriers such as glass, ceramics, and zeolite (Lee and Cho, 2001; Marcos and Marto, 2008), which are then placed in the photocatalytic microenvironment.

The present study explores the impact of a number of operating parameters on the efficiency of removing Acid Red 88 from an aqueous solution through PEC. The parameters were photocatalyst quantity, dye concentration, pH, bias potential, and NaCl (supporting electrolyte). The photocatalyst was TiO₂ doped with Nickel (Ni–TiO₂) applied to the solution in suspension. This suspension mode was chosen because it results in a larger photocatalyst surface area, meaning that it increases the contact between the photocatalyst and the pollutant molecules and improves the efficiency of photoelectrocatalytic degradation (Pourmanda and Taghavinia, 2008). However, there was the problem of separating Ni–TiO₂ powder from the treated solution (Sonawane et al., 2004; Zhang et al., 2006). This problem was dealt with by subjecting the post-treatment solution to centrifugation. To judge the efficiency of the PEC process, the COD (Chemical Oxygen Demand) and TOC (Total Organic Carbon) tests were used. The efficiency of the photocatalyst was studied using SEM and XRD techniques. The post-treatment product was also characterized using FT-IR, HPLC, and GC-MS techniques. Cost analysis was also performed, considering the costs of UV irradiation, electrical energy, electrolyte, and photocatalyst material as major cost items. The electrical energy of the treatment was supplied by solar cells. This was done knowing that in our review of previous research we found no instance of solar energy being used in PEC.

2. Materials and methods

2.1. Reagents

The chemicals used in this study were titanium tetraisopropoxide (TTIP), Nickel nitrate hexahydrate, diethanolamine, glacial acetic acid, absolute alcohol, and deionized water. Deionized water was produced by the researchers. All the other chemical reagents (purchased and used as received from Merck chemical company in Germany) were of analytical grade or better quality.

2.2. Photocatalyst preparation

The photocatalyst used in this research was TiO₂ doped with Nickel (Ni–TiO₂). The components of the nano-sized powder were synthesized separately before they were mixed via a sol–gel process. The procedures are described below.

To prepare TiO₂, first hydroxyl propyl cellulose (HPC) was dissolved in ethanol under fast stirring for 5 min. Then, TTIP was added to the mixture. This was followed by 15 min of stirring. Subsequently, a mixture of glacial acetic acid, absolute alcohol, and deionized water was added. Fifteen minutes of stirring ensued to ensure a yellow transparent acidic TiO₂ sol. The sol was allowed to stand for 30 min at room temperature. It is worth noting at this point that glacial acetic acid was used as an inhibitor to slow down the TTIP fast hydrolysis.

The Nickel sol was prepared as follows. First, Nickel nitrate hexahydrate was dissolved in absolute alcohol and was stirred for 5 min. Then, a mixture of diethanolamine, absolute alcohol, and deionized water was poured into the solution under fast stirring. The resulting solution was continuously stirred for 15 min to achieve a transparent alkaline Nickel sol.

For the final Ni–TiO₂ photocatalyst, the Ni sol was directly incorporated into the TiO₂ sol. The resultant nanocomposite was allowed to dry at room temperature. Then, it was calcined at 350 °C for 10 min and subsequently at 500 °C for 5 h (The temperature was

increased at a speed of 5 °C/s). Finally, the specimen was cooled before use.

2.3. Materials and equipment

The azo dye used to color the aqueous solution was Acid Red 88 obtained from the German company of Merck. NaOH and HCl Merck were used to adjust the pH of the solution. The electrolyte was NaCl, which was also purchased from Merck. This salt has negligible effect on pH and is non-toxic, inexpensive, soluble, and highly conductive. The solar cell was purchased from the German company of PTL Solar. In addition, the solar cell battery was obtained from Faran Electronic Industries, an Iranian manufacturer. The electrical current was controlled by a rheostat. The photoelectrocatalytic (PEC) reactor was a glass container (80 × 70 × 290 mm) whose walls were covered with aluminum foil. A pair of parallel electrodes (200 × 20 × 2 mm) made of steel were placed in the reactor at a space of 10 mm from each other. The excitation source was a 6W UV lamp (irradiation at 365 nm) purchased from the Dutch supplier of Philips. The lamp was put inside a single-walled quartz tube placed in the middle of the reactor. The efficient volume of the PEC reactor was 1000 mL after subtracting the volume occupied by the quartz tube. The sketch diagram of the reactor used in the experiment is shown in Fig. 1. To analyze the aqueous solution, TOC was measured using a Shimadzu TOC-V Analyzer, and COD levels were determined through a Hach spectrophotometer (Diar 5000). Weight measurements were performed using a digital analytical balance with a readability of 4 decimal places (purchased from Cole-Parmer, USA).

2.4. Experimental procedure

The aqueous solution was prepared by dissolving dye into pure water. Then, NaCl and Ni–TiO₂ were added to the solution. The pH of the solution was adjusted with NaOH and HCl. The solution was poured into the reactor. Then, the solution was magnetically stirred at 200 rpm to ensure homogeneity. Then, the electrodes were immersed in the solution. Before and after each run of the experiment, the electrodes were thoroughly washed with water, placed in HCl solution for at least 15 min, and then washed with water

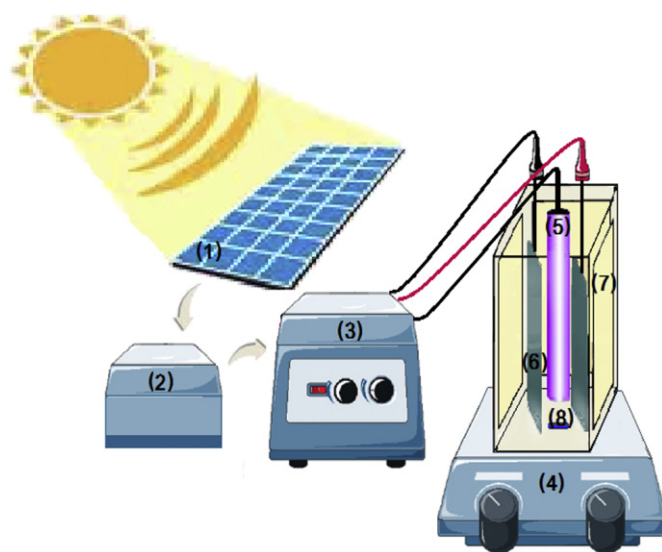


Fig. 1. Schematic representation of the photoelectrochemical system used (1: solar cell; 2: charge controller; 3: DC power; 4: stirrer; 5: UV lamp; 6: electrodes; 7: aluminum foil; 8: magnet).

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