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Catalytic bubble-free hydrogenation reduction of azo dye by porous membranes loaded with palladium nanoparticles

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ABSTRACT

Catalytic bubble-free hydrogenation reduction of azo dye by porous membranes loaded with palladium (Pd) nanoparticles was studied for the first time. The effects of Pd loading, dye concentration and reuse repetitions of membranes were investigated. In reduction, the dye concentration decreased whereas the pH rose gradually. An optimal Pd loading was found. The catalytic membranes were able to be reused more than 3 times.

Introduction

Azo dyes, containing the azo chromophoric group in addition to aromatic systems and auxochromes, are the largest class of commercial dyes used in the industries of textiles, food, pharmaceuticals, cosmetics, paper, etc. (Méndez-Paz et al., 2005). Their environmental impact either as pollutants or carcinogens is of major concern (Pinheiro et al., 2004). Most azo dyes with complex aromatic structures are highly recalcitrant to conventional biological wastewater treatment processes (Greaves et al., 2001). Therefore, pre-treatment processes are required to decolorize the dyes efficiently (Hong et al., 2012). Nevertheless, traditional flocculation (Supaporn et al., 2010) and adsorption (Laszlo et al., 1997) simply transfer the pollutants from aqueous solution to another phase rather than destroying them. Oxidation by ozone, H₂O₂ or chlorine dioxide (Nadupalli

et al., 2011) is expensive. Fenton, photo-Fenton (Elmorsi et al., 2010) or electrochemical advanced Fenton oxidation processes (El-Desoky et al., 2010; Wang et al., 2012) have a harsh working pH range (pH 2–4) and may generate large volumes of iron sludge for further disposal. The quantum yields of photo-catalytic degradation using TiO₂ are low due to hole-electron recombination (Li et al., 2009). By reduction with sulphide (Pereira et al., 2010), zero-valent iron (Fan et al., 2009; Zhang et al., 2010) or Fe-Ni (Bokare et al., 2007) nanoparticles, the azo bonds can be cleaved to give aromatic amines, resulting in decolorization and enhancement of biodegradability (Chang et al., 2006). However, acidic conditions are usually needed in reduction by zero-valent iron (Lin et al., 2008).

Hydrogen is the cleanest reducing agent and has received a great deal of attention. Traditionally, catalytic reduction by hydrogen is carried out in a stirred tank reactor, venturi, fluidized-bed, loop, trickle-bed, compact multichannel or monolithic reactor, etc. (Bradley et al., 1992; Henglein, 2000). In these devices, the limited

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gas-liquid interfacial area changes with hydrodynamic conditions, resulting in difficulty in process control and scale-up. The dispersive contacting mode may also cause problems such as flooding, hazard, emulsion and foams.

In this article, the catalytic bubble-free hydrogenation reduction of azo-dye solution by porous membranes loaded with palladium (Pd) nanoparticles is reported for the first time. Congo Red (CI 28, 22120) was employed as a model dye. Pd nanoparticles were immobilized on the membranes' inner surfaces by the gas-liquid membrane contacting method (Jia et al., 2011). In the reduction, the azo dye aqueous solution and hydrogen, being on opposite sides of the hydrophobic polypropylene hollow fiber membrane, come in contact and react at the mouth of the membrane pores. The membranes provide substantially large and stable gas-liquid interfacial area, which is favorable for mass transfer, process design and scale-up. All the hydrogen can be absorbed in this non-dispersive contact-reaction process, and the problems of emulsion and foaming can be avoided. Therefore, this method provides a novel candidate for catalytic reduction of azo dyes and various other unsaturated organic compounds. The effects of Pd loading, dye concentration and number of reuses of membranes were investigated.

1 Experimental

1.1 Materials

Congo red was provided by Beijing Xudong Chem. Co. Ltd., China. The chemical reagents, including PdCl₂, hydrochloric acid and sodium chloride, were analytical grade. Ultrapure water was employed. Five hundreds mL of 2.0 × 10⁻⁴ mol/L PdCl₄²⁻ aqueous solution containing 0.0170 g of PdCl₂, 8.0 mL of 0.1% (V/V) hydrochloric acid solution and 0.2240 g of NaCl was prepared.

The membrane contactor was made up of twenty symmetric polypropylene hollow fiber membranes with average pore diameter of 0.05 μm, porosity of 60%, O. D. 0.5 mm, I. D. 0.4 mm and effective length of 45.0 mm (Tianjin Blue Cross Membrane Technology Co. Ltd., China). The fibers were well-separated by two porous stainless steel sheets mounted at the two ends of the membrane module.

1.2 Preparation of calibration curve of Congo Red solution

Congo Red aqueous solutions with concentrations of 1.25 × 10⁻⁶, 2.50 × 10⁻⁶, 5.00 × 10⁻⁶, 1.00 × 10⁻⁵ and 2.50 × 10⁻⁵ mol/L were prepared. The UV-Vis absorption of the solutions was analyzed by a UV-Vis spectrophotometer (Cintra 10e, Australia). The calibration curve was obtained by plotting the absorbance at 498 nm vs. the dye concentration.

1.3 Loading of Pd nanoparticles on membrane inner surface

The experimental set-up was schematically shown in our previous paper (Jia et al., 2011). Pure hydrogen with gauge pressure of 1 kPa entered into the shell side of the membrane reactor. The PdCl₄²⁻ solution (2.0 × 10⁻⁴ mol/L, 80.0 mL) in the stirred tank flowed through the membrane lumens at a flow rate of 8 mL/min at 20°C, contacted with the hydrogen permeating through the membrane pores, reacted and then flowed back to the stirred tank for circulation. After reaction, the membranes were rinsed with pure water 5 times, the dried in vacuum to constant weight. The Pd loading, *L* (%), on the membranes' inner surface is expressed as:

$$L = \frac{W_1 - W_0}{W_0} \times 100\% \quad (1)$$

where, *W*₀ (g) is the weight of the pristine membranes, and *W*₁ (g) is the weight of the catalytic membranes. After gold sputtering, the membrane inner surface was observed with cold-field scanning electron microscopy (SEM S-4800).

1.4 Catalytic bubble-free reduction of dye solution

The schematic diagram of the membrane contactor is shown in Fig. 1. The dye solution of 100.0 mL in the stirred tank flowed through the catalytic membranes lumens and then flowed back to the stirred tank for circulation until the absorbance of the solution at 498 nm attained a constant value, indicating that adsorption equilibrium was reached. Then, pure hydrogen (pressure gauge of 1.0 kPa) was introduced into the shell side and then the catalytic membrane pores, contacted and reacted with the dye solution. The pH of the dye solution was monitored. An aliquot (2.0 mL) sample was withdrawn at set intervals during reaction, and the absorbance at 498 nm was measured by a UV-Vis spectrophotometer to determine the dye concentration. The decolorization efficiency, *D*, is expressed as:

$$D = \frac{C_0 - C_t}{C_0} \times 100\% \quad (2)$$

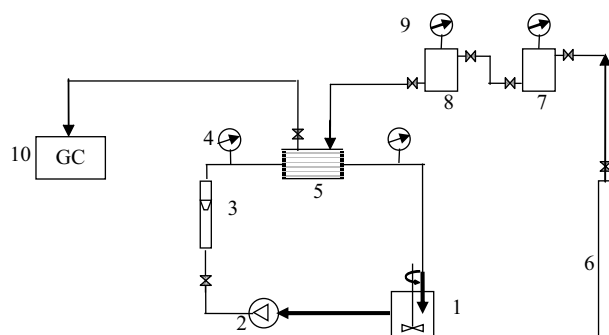


Fig. 1 Schematic diagram of bubble-free hydrogenation. (1) stirred tank; (2) peristaltic pump; (3) liquid rotameter; (4) pressure gauge; (5) membrane contactor; (6) H₂ generator; (7) gas supply tank; (8) buffer tank; (9) barometer; (10) gas chromatometry.

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