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Energy recovery in wastewater decontamination: Simultaneous photocatalytic oxidation of an organic substrate and electricity generation

M. Canterino*, I. Di Somma, R. Marotta, R. Andreozzi, V. Caprio

Dipartimento di Ingegneria Chimica, Università di Napoli Federico II, p.le Tecchio, 80, 80125 Napoli, Italy

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ABSTRACT

The contemporary removal of organic pollutants from aqueous solution and electricity generation is studied in the present work by means of an experimental device resulting from the combination of a photocatalytic reactor and an electrochemical cell. The proposed system relies on the capability of Cu^{2+} ions to reduce in the presence of TiO_2 , (solar) UV radiations and a sacrificial organic agent. In the anodic solution of the combined photoreactor-cell, Cu^0 is oxidized to Cu^{2+} and the latter is reduced again to the lowest oxidation state. The use of different sacrificial agents ranging from formic acid (FA) to glycerol (GLY) to glucose (GLU) is investigated along with the adoption of two different cathodes for the cell, the first based again on the couple $\text{Cu}^{2+}/\text{Cu}^0$ and the second on the couple $\text{O}_2/\text{H}_2\text{O}$.

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1. Introduction

During the last decade increasing concerns for CO_2 emissions in atmosphere from fossil fuel combustion stimulated the search for a new way of producing energy from renewable substrates (biomasses) and to recover it from the treatment of wastes and wastewaters (Venkata Mohan et al., 2008; Tilche and Galatola, 2008; Alzate-Gaviria et al., 2007). Among the others, microbial fuel cells gained a great attention in the Scientific Community due to their capability to use the catalytic activities of microorganisms to oxidize organic and inorganic matter and generate current (Feng et al., 2008; Kim et al., 2007; Logan, 2005). The same aim may be reached through an alternative non-biological system in which the oxidation of an organic species is exploited to generate electricity. This is made possible by the use of an electrochemical cell combined with a photocatalytic reactor. This process

enables, in the presence of TiO_2 particles and a solar UV radiation (and in the absence of oxygen), the contemporary oxidation of an organic substrate, which is the sacrificial agent, (r2) and the reduction of a metal ion in the solution (r3):



The most simple way of assembling this type of device is that of considering an electrochemical cell in which the same couple ($\text{M}^{(n+1)+}/\text{M}^{(n)+}$) is used in both the half-elements but at different concentrations. It is well known that this device is capable of generating an electrical output until the concentrations of active species in both the compartments become equal.

* Corresponding author. Tel.: +39 0 817682253; fax: +39 0 815936936.

E-mail addresses: canterin@unina.it (M. Canterino), idisomma@unina.it (I. Di Somma), rmarotta@unina.it (R. Marotta), roberto.andreozzi@unina.it (R. Andreozzi), caprio@unina.it (V. Caprio).
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Reactions occurring during cell working are:



When the system achieves equilibrium the current in the external circuit ceases unless some kind of replenishment of metal ions in the low and high state of oxidation is ensured to anode and cathode, respectively.

Once the cell was discharged, in the anodic solution the concentration of the species $M^{(n)+}$ is increased through the reduction of $M^{(n+1)+}$ ions (r3) and the contemporary oxidation of sacrificial agent (r2) by submitting it to a photocatalytic process. Reoxidation of $M^{(n)+}$ ions in the cathodic compartment may be simply obtained through a continuous feeding to it of an air stream:



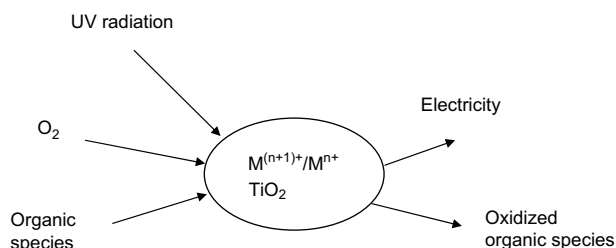
The overall system may be thus depicted as in Scheme 1.

The adopted device as a whole can be seen as a system to which oxygen, an organic species and an UV radiation are fed whose outputs are electricity and the oxidized organic species (or a mixture of oxidation products). That is, the proposed device allows the oxidation of an organic species (indirectly) by oxygen while producing electricity. In this sense it can be classified as a fuel cell. However, since its work is based also on a photocatalytic process (necessary for the replenishment of $M^{(n)+}$ in the anode of the cell) it can be more precisely indicated as a “TiO₂-based photocatalytic (solar) fuel cell”.

In the present work, a device as that described above is adopted with an anode based on the system Cu^{2+}/Cu^0 . Sacrificial photocatalytic processes in the presence of Cu^{2+} ions have been extensively investigated in the past (Vinu and Madras, 2008; Krysa et al., 2006; Kyung et al., 2005; Kanki et al., 2004; Foster et al., 1995) and recently also by the authors (Canterino et al., 2008). Two different cathodic systems and various sacrificial agents are investigated in the present study.

2. Experimental

Experiments were carried out using a photocatalytic reactor and a modified electrochemical H-shaped cell (Fig. 1). The latter consists of two compartments with the anode made by an electrode of metallic copper, with a surface area of 40 cm², and a solution containing cupric sulphate. Two different cathodes were used during the investigation. The first was similar to the anode; the second is an O₂ electrode that had an



Scheme 1

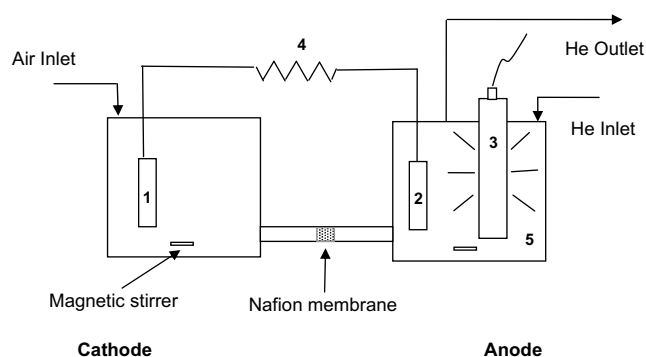


Fig. 1 – Photoelectrochemical cell. 1: Copper or platinum black electrode; 2: Copper electrode; 3: UV lamp; 4: Resistance; 5: TiO₂ suspension.

inert wire coated with a finely divided layer of platinum (platinum black) inserted in an aqueous solution in which an air stream was bubbled. The anode and cathode were connected through a Nafion117 cation exchange membrane. An UV high-pressure lamp (125 W power input by Helios Italquartz), mainly emitting at 305, 313 and 366 nm (manufacturer's data), enclosed in a glass sleeve, was used in a photocatalytic reactor or in the anode of the photoelectrochemical cell. In the last case the lamp and the anodic compartment were wrapped with an aluminium foil to shield the radiation and protect the operator. The power of the lamp at 305 nm $I^{\circ}_{(305)}$ was $5.70 \times 10^{-7} \text{ E s}^{-1}$ measured, according to Kuhn et al. (2004), by hydrogen peroxide photolytic experiments, at 313 nm $I^{\circ}_{(313)}$ was $9.83 \times 10^{-7} \text{ E s}^{-1}$ determined according to Zepp et al. (1998) by valerophenone actinometry and at 366 nm $I^{\circ}_{(366)}$ was $2.98 \times 10^{-6} \text{ E s}^{-1}$ measured using an UV radiometer Delta Ohm HD 9021.

A helium stream was continuously fed to the reacting solutions – to prevent the dissolution of atmospheric oxygen – during the photocatalytic reduction of dissolved Cu^{2+} ions in the presence of an organic compound as sacrificial agent.

The operating conditions (pH, catalyst load, etc.) under which Cu^{2+} ions were allowed to reduce were the same as those adopted in a previous work (Canterino et al., 2008) at which a good understanding of the system behaviour had been achieved.

The concentration of Cu^{2+} ions during the experiments was measured by means of a photometric method using an analytical kit based on oxalic acid bis-cyclohexylidene hydrazide (cuprizone), purchased by Macherey–Nagel, that allows only the determination of Cu(II) according to supplier's indications. An UV/vis diode array spectrophotometer (Unicam) was used for the measurements at a wavelength of 585 nm. Formic acid (FA), glycerol (GLY) and glucose (GLU) were used as sacrificial agents. The concentrations of FA and GLY at different reaction times were evaluated by HPLC analysis. For this purpose, the HPLC apparatus (Agilent 1090) was equipped with an UV–VIS detector ($\lambda = 215 \text{ nm}$) and a Phenomenex Rezex column (ion-exclusion column), using a mobile phase of sulphuric acid $2.5 \times 10^{-3} \text{ mol L}^{-1}$, flowing at 0.6 mL min^{-1} . The Open Circuit Voltage (OCV) and cell voltage (E_{cell}) were measured by a digital multimeter (DMM) ISO-tech IDM101.

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