Catalysis Today xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Catalysis Today

journal homepage: www.elsevier.com/locate/cattod

Irradiation of liquid phase plasma on photocatalytic decomposition of acetic acid-containing wastewater over metal oxide photocatalysts

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ARTICLE INFO

Keywords:
Liquid phase plasma
Photocatalytic decomposition
Hydrogen evolution
Wastewater
Acetic acid

ABSTRACT

The photocatalytic decomposition of acetic acid-containing wastewater was assessed for the removal of pollutants and the production of hydrogen. The effects of irradiation of the liquid phase plasma were evaluated in the photocatalytic decomposition reaction. The evolution of hydrogen was also characterized by the photocatalytic decomposition of wastewater. Ni-loaded TiO_2 photocatalysts were introduced to the photocatalytic reaction. With the Ni loading to the TiO_2, the adsorption wavelength was enlarged to the visible range. The photochemical decomposition by irradiation of the liquid phase plasma without photocatalysts produced some hydrogen evolution with the degradation of acetic acid, which was attributed to the decomposition of the reactant by active species generated by the irradiation of liquid phase plasma. The rate of hydrogen evolution from water on the photocatalyst was ca. 1.6 mmol/min at 1 h of process time. The photocatalytic decomposition of acetic acid was improved by the addition of Ni-loaded TiO_2 photocatalysts by the irradiation of liquid phase plasma. The Ni loading on TiO_2 brought out an enhancement of acetic acid degradation and hydrogen evolution. Hydrogen evolution was accelerated significantly due to the additional hydrogen production by the photocatalytic decomposition of acetic acid.

1. Introduction

Hazardous and toxic wastewater pollutants have caused serious environmental problems [1–4]. Among them, a large amount of wastewater containing acetic acid has been discharged from the production of chemicals, such as terephthalic acid, dimethyl terephthalate, isophthalic acid, cellulose acetate, and esters [5]. In the various techniques for the treatment of wastewater, heterogeneous photocatalysis has been considered an effective technology for water remediation [6–8]. In the photocatalysis reaction, the light sources and photocatalysts are important elements affecting the photocatalysis. Although a range of light sources have been employed in photocatalysis, there are few reports on photocatalysis using liquid phase plasma (LPP) by irradiation into water directly [9].

LPP is generated by high-voltage discharging in liquid. The high density plasma by discharging emits a strong UV and visible light with a large active species. High-voltage pulse discharge plasma has been considered to be promising alternatives for water treatment [10,11]. High-voltage discharge plasma for the degradation of organic pollutants in water has characteristics such as a non-selective high degradation rate for organic pollutants, no secondary pollution, normal temperature and pressure, and low power consumption. Discharging in liquid can generate a higher density of plasma and larger spatial distribution compared to UV lamp irradiation [12]. Therefore, these advantages can lead a decomposition of raw materials and an enhancement of hydrogen production.

The development of an efficient purification technology for organic pollutants is becoming necessary. The electrical discharge is a non-equilibrium discharging process with a dielectric layer being inserted in the discharging space. The purification of organic compounds in wastewater has attracted significant attention because it has high purification efficiency without secondary pollution [13,14]. Recently, an electrical discharge in the liquid phase has been used to degrade organic pollutants in water because it can produce both hydroxyl

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http://dx.doi.org/10.1016/j.cattod.2017.04.039
Received 26 September 2016; Received in revised form 28 March 2017; Accepted 17 April 2017

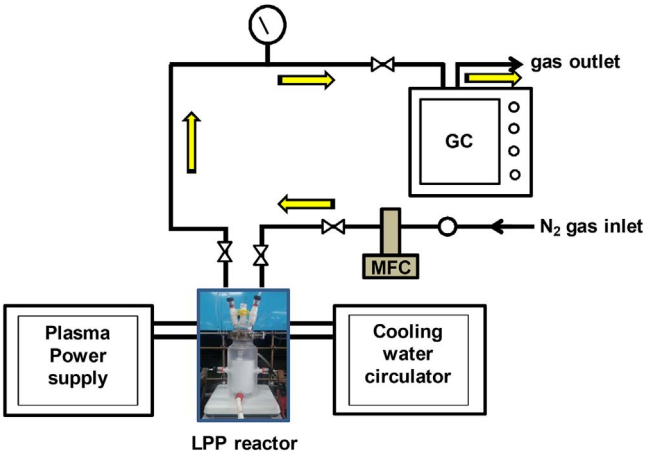

Fig. 1. Schematic diagram of photocatalytic reaction apparatus using LPP system.

radicals and atomic oxygen, which have a high oxidation potential [15–17]. The purification of organic waste water using LPP can promote simultaneously the remediation of liquid and hydrogen evolution in the gas phase. Pulsed discharge can produce a higher peak current and effective electrons and radicals. In addition, it can produce wide non-equilibrium plasma easily [18–20]. LPP can generate a higher plasma density and larger spatial distribution [21,22]. These properties bring out the decomposition of organic pollutants and hydrogen production. The LPP has longer discharge channels and a higher partial discharge intensity compared to arc discharge. Therefore, it can induce energy consumption. LPP has been reported to improve energy efficiency and prevent carbon deposition during discharge [23,24]. Therefore, hydrogen production by the LPP is a promising clean technology.

Hydrogen is a promising alternative form of storable and clean energy for the future, and has attracted considerable interest in the development of new methods to produce hydrogen from renewable and sustainable resources. Photocatalytic water splitting using solar energy provides a promising option to produce hydrogen [25,26]. In general, hydrogen production from solar water-splitting processes can be classified as photobiological water splitting, thermochemical water splitting and photocatalytic water splitting [27]. Among them, photocatalytic water splitting has attracted interest as an effective clean technology [28–30]. The photocatalytic decomposition of organic wastewater using LPP is an effective clean technology that offers hydrogen production and water purification.

This paper proposes photocatalysis using LPP as a light source to the wastewater containing acetic acid over nanocrystalline metal oxide photocatalysts to decompose the pollutant and hydrogen evolution. The nickel loaded TiO_2 nanocrystallines were introduced as the photocatalysts. The effects of LPP irradiation on the photocatalytic decomposition of acetic acid-containing wastewater were evaluated for hydrogen evolution. The hydrogen evolution and decomposition of acetic acid were also characterized according to LPP irradiation and the photocatalysts.

2. Experimental

2.1. Materials

Titania (P25, Degussa), consisting of anatase (ca. 80%) and rutile was used as a parent photocatalyst. Nickel(II) nitrate hexahydrate (Daejung, 98%) was introduced as a precursor. Ni-loaded TiO_2 photocatalysts were prepared using the typical incipient wetness method with various concentrations of Ni precursor. An acetic acid solution (10 vol %) was employed as the reactant. The TiO_2 and Ni-loaded TiO_2 were introduced as the photocatalysts in the photocatalytic reaction. NiO/TiO_2 photocatalysts were prepared from Ni/TiO_2 through reduction and oxidation. The Ni/TiO_2 was reduced at 300 °C in a flowing 10% H_2 and 90% N_2 gas mixture for 3 h to prepare the NiO/TiO_2 photocatalysts. Thereafter, the photocatalysts were oxidized at 500 °C in an air flow for 5 h. The wt% Ni loaded in the preparation process is written in the parentheses of Ni-loaded TiO_2 names.

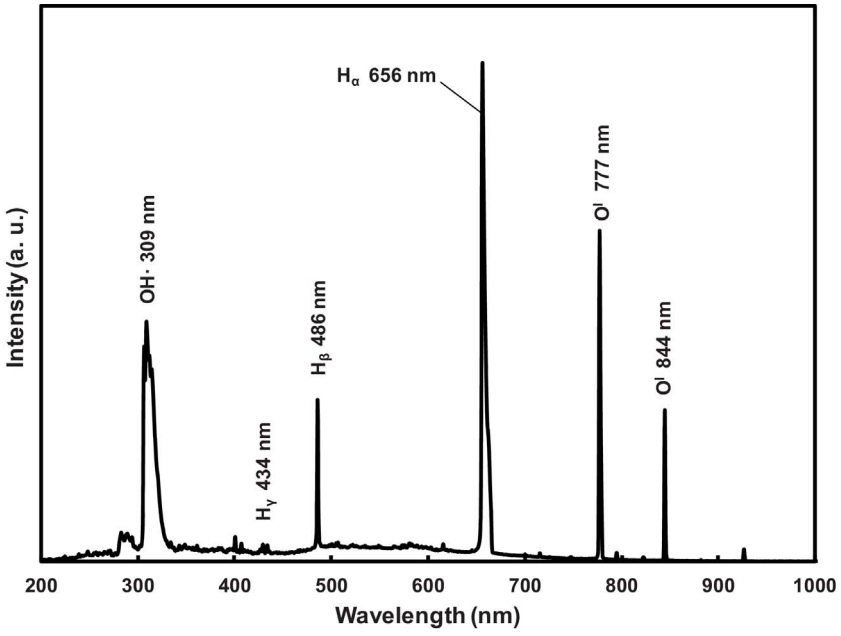

Fig. 2. Optical emission spectrum of LPP measured in the reactant.