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Oxidation of acetylene by photocatalysis coupled with dielectric barrier discharge

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Abstract

Volatile organic compound removal from air requires oxidative processes associating high carbon dioxide selectivity like photocatalysis with fast kinetics like non-thermal plasma. A specially designed coupling reactor has been used to investigate the interaction between photocatalysis and non-thermal plasma. Acetylene has been selected as a model molecule to evaluate oxidation efficiencies. After determining the oxidative efficiency of both techniques used separately, the coupling of plasma with titania photocatalyst has been performed. The influence of UV-irradiation of a photocatalyst placed inside the discharge by external lamps has been investigated. It is reported that photocatalysis leads to a complete mineralization of acetylene, whereas more than 50% of the carbon balance based on CO and CO_2 is missing when plasma alone is performed. The presence of a porous material inside the discharge improves the initial removal rate of acetylene. It tends to favor the formation of adsorbed organic species, indicating that part of plasma reactivity is transferred to the adsorbed phase. Finally, the use of additional external UV-light is reported to improve the formation of carbon dioxide, which means that photocatalysis can be usefully performed in an ionized gas.

Keywords: Dielectric barrier discharge (DBD); Non-thermal plasma; Photocatalysis; Advanced oxidation; VOC

1. Introduction

The removal of volatile organic compounds (VOCs) is one of the main issues of air treatment. Since VOCs are often toxic or irritating, the legislation has become more and more stringent. The first approach consists reducing the emission of VOCs. Nevertheless gaseous wastes cannot be completely avoided. Many organic compounds are strongly odorous or even toxic at concentrations as low as 1 ppmv.

To remove VOCs, various techniques based on adsorption are efficient [1,2], but they consist of pollution transfer from the gaseous phase to a solid surface. Advanced oxidation of VOCs is environmentally safer since VOCs traces are converted to carbon dioxide and water. Catalysis is an efficient mean for organic compound oxidation. Thermally activated catalysts based on noble metal such as Pt and Pd exhibit good perfomances [3–5]. In parallel, photocatalysis has been intensively developed for VOCs elimination during the last 10 years.

Saturated volatile organic compounds advanced oxidation [6–8] as well as unsaturated [9–11] or oxygenated ones [12–14] such as alcohols or ketones, have been intensively studied. Photocatalysis exhibits a high oxidation ability for each type of VOC. Its efficiency has been demonstrated even with a triple carbon–carbon bond, as recently reported by Thevenet et al. [15] for acetylene photocatalytic oxidation.

Dielectric barrier discharge (DBD) has been widely studied first for ozone generation, and nowadays for VOCs oxidation. It is reported as the most hopeful air cleaning technology to remove toxic volatile contaminants in air by Oda et al. [16]. After demonstrating at the end of the 1990s that non-thermal plasma is efficient for organic compounds removal [17,18], the dependance of the VOCs chemical structure on DBD efficiency has been investigated [19]. To improve non-thermal plasma oxidation efficiency, some authors introduced solids into discharges. Porous and high dielectric constant materials were selected. Holzer et al. [20] explored the interaction between plasma and heterogeneous catalysis on Al_2O_3, silica gel and

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quartz. Then, Roland et al. [21] widened the scope to $BaTiO_3$, $PbZrO_3$, $PbTiO_3$, and $LaCoO_3$ materials, leading to the conclusion that interaction is mainly governed by the interaction between the plasma phase and the material porosity. Consequently, several packed reactor were developed and studied, screening the variety of VOCs. Benzene [22], toluene, propane [23], and acetaldehyde [24] are the main VOCs investigated. Many studies selected TiO_2 as a very interesting coupling material [22,25–28]. Nevertheless, only very recent studies tried to understand the interaction between plasma and the UV-activated material with a photocatalytic approach [25].

Acetylene has been selected as model pollutant for several reasons. No other study deals with the mineralization of compounds containing triple carbon/carbon bonding. Moreover, acetylene is the simplest alkyne, making reaction intermediates easier to identify and to quantify; carbon balance and reaction pathways get simpler to investigate.

The reasons for coupling plasma photocatalysis are the following: (i) plasma treatment is expected to improve the kinetics of the oxidation reaction, (ii) photocatalysis is expected to improve significantly the carbon dioxide selectivity and the mineralization carbon balance, and (iii) plasma generated UV, due to excited nitrogen relaxation, as well as activated species, are supposed to activate photocatalytic material.

The purpose of this work was to explore and to understand the interaction between non-thermal plasma and titanium dioxide nanoparticles supported on glass fibers. The oxidation capacity for each oxidative technique was separately evaluated. Subsequently, their coupling was explored, with the joint influence of plasma input power and additional external UV-light.

2. Experimental

Coupling of plasma-photocatalysis has been studied in a specially designed Pyrex-glass reactor allowing a plan-to-plan geometry (Fig. 1). The active part of the system consists of rectangular section channel. Two 16 cm^2 copper electrodes are

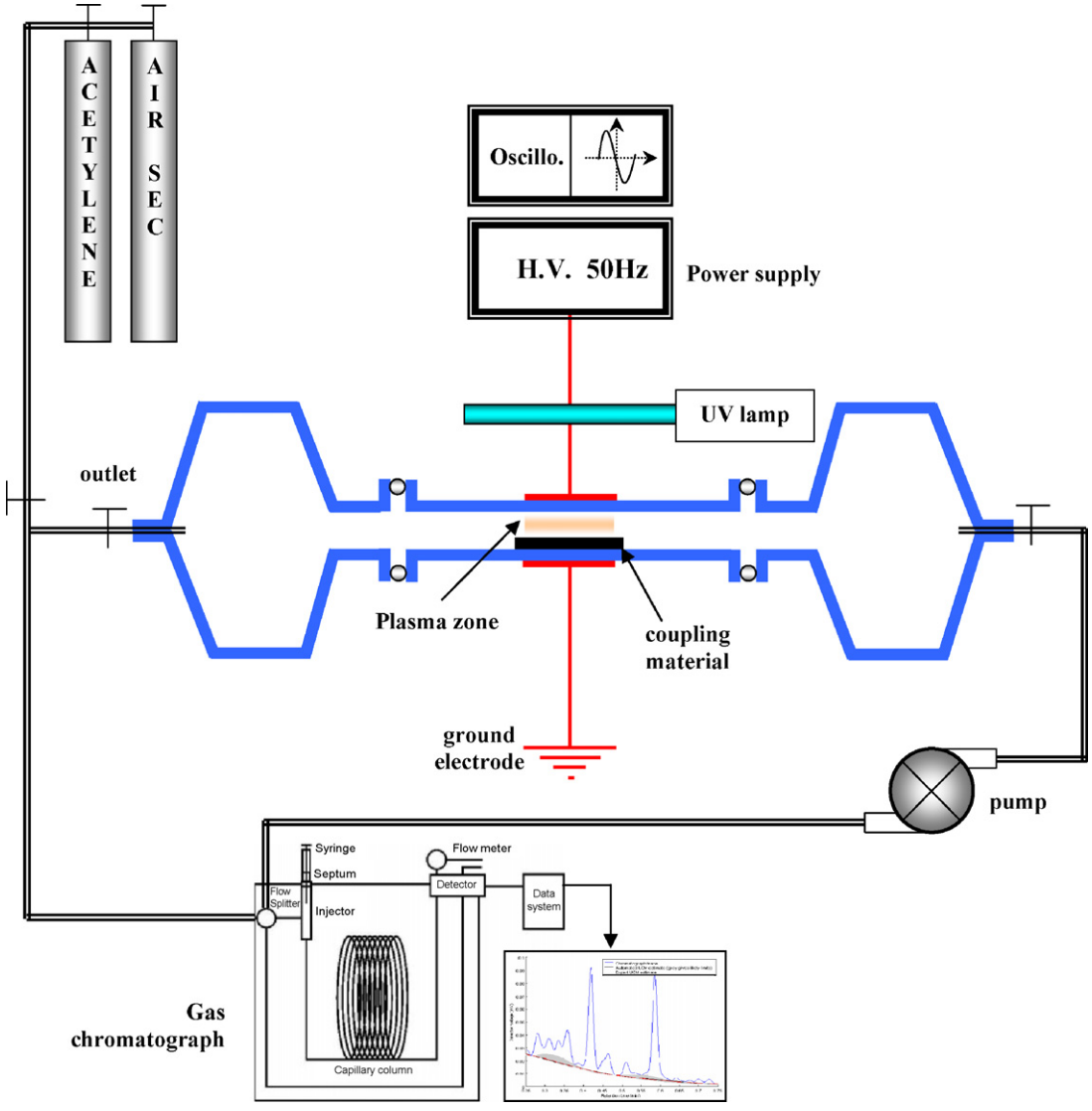

Fig. 1. Scheme of the experimental set up for coupling non-thermal plasma and photocatalysis.