

Diphasic process combining a fluidized catalytic bed and a plasma at atmospheric pressure for the degradation of volatile organic compounds

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An experimental study of a diphasic process, combining a fluidized nanostructured catalytic bed and an atmospheric pressure plasma, at room temperature, is presented. The improvement of the removal efficiency of a model pollutant (CH_3CHO) and by-products mineralisation is discussed in terms of pollutant and by-products interaction with the catalytic surface, using adsorption/desorption analysis (breakthrough curves) and reactivity (DRIFT).

1. Introduction

Volatile organic compounds (VOCs) are environmentally damaging chemicals emitted into air from numerous sources including stationary, mobile and natural sources. Thus, growing environmental awareness has led to stringent regulations to control VOC emissions and as a result a disposal process with high efficiency, reliability and cost effectiveness is necessary to convert these compounds. Catalytic oxidation has been proven to be a feasible and cost-effective method in VOC emission abatement. In general, the most popular catalysts are noble metals, but different metal oxides are also used. In addition to catalytic activity, selectivity and durability are also important criteria when selecting the most suitable catalyst. The relatively high temperature required to activate the catalyst, its deactivation, associated to operating conditions that can reduce the catalytic activity from masking, chemical poisoning or thermal sintering, is a significant matter both from the catalyst manufacturers' as well as from the catalyst end users' point of view.

For over ten years, the study of diphasic processes combining a plasma at atmospheric pressure and a catalyst material has emerged and led to an impressive number of scientific articles in the literature. The original idea of the first studies was to combine the advantages of operating non-thermal plasmas at room temperature, their low power consumption, their high chemical reactivity in gas phase (through the production of O, OH, O_3 , N_2^* , ... species), and an additional catalytic effect, by immersing a catalyst in a plasma.

2. Experimental set-up

A TREK 20/20C high voltage amplifier is used to produce a sinusoidal dielectric barrier discharge in cylindrical quartz tube (Fig 1). The gas mixture is composed by CH_3CHO (used as model pollutant and injected at 1000 ppm), oxygen (5-20 %) and nitrogen (balance) at a total flow rate of 100 mL/min.

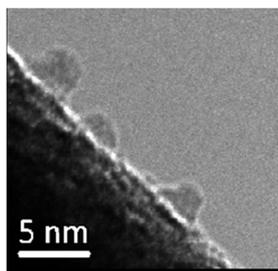


Fig 1: Packed Bed Discharge System Fig 2: cross-section slab TEM (b) images of the monolayer nanoparticulate titania film after UV-A photocatalytic silver reduction

Spherical SiO_2 and Al_2O_3 pellets were packed in the reactor. A new method of nanoparticulate composite films preparation based on photocatalytic activity of nanoparticulate TiO_2 coatings. The nanocoatings were prepared by liquid colloid deposition of chemically active size-selective titanium-oxo-alkoxy nanoparticles on glass substrates. The nanoparticles were generated in a sol-gel reactor with rapid (turbulent) micromixing [1-2]. The silver clusters were grown onto the immobilised monolayer titania nanoparticles by photocatalytic reduction of silver ions under UV light illumination.

High-resolution SEM/TEM and AFM measurements evidence a growth of the silver nanoparticles size and surface number density with the irradiation time (Fig 2).

3. Objectives, results and discussion

The efficiency of the diphasis process is evaluated through the energy consumption, the degradation of the pollutant and mineralisation of by-products using gas chromatography coupled with mass spectrometry, fourier transform infrared spectroscopy, ultra-violet and infrared absorption spectroscopy [3]. The residual acetaldehyde (%) is presented on the Fig. 3, as a function of the specific input energy (SIE in J/L) and the photocatalytic reduction time of silver ions under UV illumination. We do observe a strong influence of the photocatalytic reduction time, leading to a improvement of the removal efficiency of a factor of 10 with a process time fixed at 5 min. This improvement is correlated to an increase in CO₂ production (Fig 4).

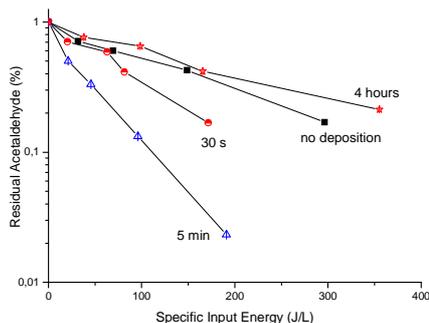


Fig 3: residual acetaldehyde as a function of SIE and photocatalytic reduction time (from 30s to 4 hours)

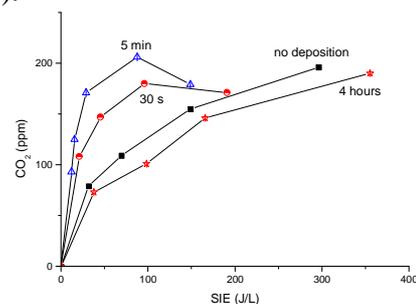


Fig 4: CO₂ as a function of SIE and photocatalytic reduction time (from 30s to 4 hours)

The improvement and decrease of the process efficiency will be discussed in terms of active species and by-products interaction with the glass substrate and silver nanoparticles. Adsorption and desorption processes on the surface will be studied using breakthrough curves, representing the concentration of a single or mixture of molecules as a function of the time (Fig 5). For example, we did observe an heterogeneous reactivity of O₃ and CO on the SiO₂ or Al₂O₃ substrate, leading to CO₂ production. We will show that this reactivity is strongly influenced by the photocatalytic silver reduction time, and may explain the improvement, in term of CO₂ production, observed in the Fig 4. Diffuse Reflectance InfraRed Fourier transform spectroscopy (DRIFT) will also be used to study the reactivity of the single compounds and mixture on the surface.

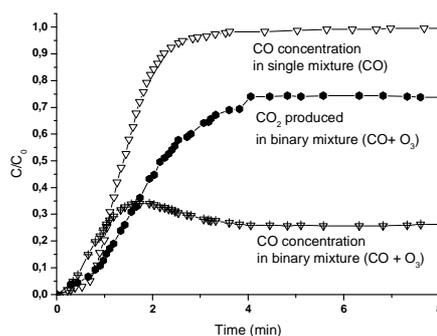


Fig 5: CO₂ heterogeneous production in binary mixture (CO+O₃) on α -Al₂O₃ substrate

References

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