



# Plasma catalysis application of gold nanoparticles for acetaldehyde decomposition



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## HIGHLIGHTS

- Different sizes of gold nanoparticles were obtained by photocatalytic reduction.
- Au/TiO<sub>2</sub> is suitable catalyst for acetaldehyde removal in term of energy yield.
- Plasma treatment doesn't influence the surface Au atoms density.
- The reaction mechanism of acetaldehyde decomposition by Au/TiO<sub>2</sub> was proposed.

## ARTICLE INFO

### Keywords:

Acetaldehyde decomposition  
Non thermal plasma  
Au/TiO<sub>2</sub> nanocatalyst  
Heterogeneous catalysis

## ABSTRACT

The decomposition of acetaldehyde was investigated in a plasma-catalytic reactor. The gold-based catalysts were obtained by growing Au nanoparticles on silica beads or plates previously coated by a titanium oxo-alkoxy monolayer. The characterization of the synthesized catalysts was carried out by SEM, AFM and UV-Visible spectroscopy. Below an exposure time to UV of 20 min, a homogeneous surface coverage by the relatively small gold nanoparticle ( $R < 16$  nm) was achieved. Compared to our previous study with silver catalysts, the same amount of the pollutant, acetaldehyde, can be decomposed with the Au-based nano-structured catalysts, under similar experimental conditions. However, using the gold catalyst enhanced the process selectivity towards CO<sub>2</sub>. Furthermore, we showed that the gold particles size and distribution strongly affect acetaldehyde degradation. Plasma treatment doesn't influence the surface Au atoms density but change the oxidation state of gold. On the basis of the results of the by-products formation, a basic reaction mechanism involved plasma induced chemical reactions in the discharge region and catalytic reactions on the catalyst surface was proposed.

## 1. Introduction

Many studies have been conducted to control volatile organic compounds (VOC) emissions by adsorption [1], thermal and catalytic incineration [2], condensation [3], membrane separation [4] and photocatalysis [5]. However, these conventional technologies have some disadvantages especially for low concentrations VOC treatment. Non thermal plasma is considered as an alternative since it can be operated at atmospheric pressure and room temperature, over a large range of gas flow rates and concentrations [6]. The formation of unwanted by-products, low mineralization and energy efficiency prevent its industrialization.

In the literature, researchers propose a combination of NTP with

catalysts: plasma-catalysis. When the catalyst is introduced in the discharge zone (in plasma catalysis, IPC) [7–10], synergetic effects are related to two aspects: (1) the influence of packing catalysts on the plasma discharge characteristics and (2) the influence of plasma discharge on catalysis. The former can be described as the enhancement of electric field due to the presence of pellets and the latter concerns a shift of the reaction selectivity because of the adsorption of pollutants and intermediates on the catalyst surface. In a previous work, we have reported a new method of preparation of Ag/TiO<sub>2</sub> nanoparticulate coatings with stable and highly reproducible morphology [11] and then studied the degradation of acetaldehyde using a process coupling NTP and this catalyst [12]. But the low CO<sub>2</sub> selectivity motivates us to find another catalyst.

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Gold nanoparticle catalysts on oxide supports have exceptionally high activities for low-temperature CO oxidation [13]. Generally, the catalytic properties of heterogeneous gold catalysts strongly depend on the size and highly homogeneous distribution, and on the extent of the metal–semiconductor contact at the interface [14,15]. For plasma catalysis, atmospheric pressure non-thermal plasma was used to regenerate or to enhance the catalytic activity of TiO<sub>2</sub>-supported Au nanoparticle catalysts for the oxidation of CO [16]. In the presence of gold nanoparticles, for trichloroethylene removal, the concentration of CO and CO<sub>2</sub> increased as compared to those obtained with plasma alone [17]. However, the effect of gold particle size in the plasma catalysis process is still not clear.

The stability of the catalysts under plasma-catalytic process is one of the most important properties governing their application in practice. According to the results published in the literature, discharges may enhance the dispersion of active catalytic components [18] and the oxidation state of the material can be influenced when exposed to plasma discharge [19,20]. The interaction between plasma and catalyst may induce the deactivation of catalysts following by catalyst modifications and sintering. It is accordingly very important to analyze the catalysts before and after the plasma treatment.

In this work, acetaldehyde decomposition was investigated using a plasma-catalyst reactor. In addition, this is the first time that monodisperse gold spherical nanoparticles have been used in plasma catalysis process for acetaldehyde removal. The stability of the catalysts under plasma-catalytic process and the formation of by-products in the plasma region and on catalyst's surface were also investigated.

## 2. Experimental set up

### 2.1. Gold-based nano-structured catalysts preparation

Gold nanoparticles deposition was performed following a very similar method to the one presented in our previous study concerning the preparation of silver nanoclusters [11]. The gold nanoparticles were grown on two types of substrates. The first one, glass plates, is used in order to facilitate catalysts characterization, especially for AFM analysis. The second one, 1 mm glass beads, is used as catalytic packing in the plasma-catalysis process. SEM analysis showed that for a given set of experimental conditions, we can obtain almost the same Au-deposition in terms of size, density and homogeneity, on the glass beads as on the glass plates. Thus, we can consider that there is little change in Au deposition on these two types of substrates. The different supports were prepared for the deposition step following the same procedure as described in reference [11].

As for silver, the first step is to prepare of a monolayer of size-selected titanium oxo-alkoxy (TOA) nanoparticles [21]. Then, by the photocatalytic reduction of Au<sup>3+</sup> ions, gold nanoparticles can be deposited on its surface. The deposition of TOA nanoparticles on the glass supports is realized by dip-coating deposition method described elsewhere [21]. The deposition of gold nanoparticles was realized by

photocatalytic reduction of Au<sup>3+</sup> ions in an aqueous solution of the gold precursor (HAuCl<sub>4</sub>, 99.99%, Sigma Aldrich). Ethanol as an efficient hole scavenger has been introduced to the aqueous solution (in ratio 5:1). The initial pH value (pH 2.8) was adjusted at pH 5 with 0.1 M NaOH solution, which is below the isoelectric point of titania (IEP of anatase = 6), in order to obtain a higher Au content [21]. The irradiation was carried out by an 8 W power UV-A lamp (Philips) operating at 364 ± 10 nm on the support contained of TOA nanoparticles in the solution previously prepared. The irradiation time of photo-deposition (t<sub>ph</sub>) is a key parameter to control the gold nanoparticle morphology which was varied from 1 to 300 min. After the UV irradiation, the samples were removed from the solution, washed and dried during 4 h at 80 °C.

### 2.2. Catalysts characterization

Scanning electron microscopy (SEM) (Zeiss Supra 40 VP SEM-FEG) was used to observe the morphology and growth of gold nanoparticles. AFM analysis is carried out to provide the structural and morphological surface detail of the as prepared samples. The measurements were carried out in the tapping mode, with a radius probe below 10 nm, in a Veeco Nanoscope dim3. UV–vis absorption spectra were obtained by using a monochromator (Spex) (grating 150 L/mm, f = 30 cm, slit 20 μm) coupled to a Peltier-cooled CCD detector (Princeton) and cw-spectroscopic UV/visible/IR light continuum source (Oriel). UV–Vis absorption analyses were used in order to identify the characteristic signal of the plasmon band attributed to the presence of metallic gold in the as-prepared samples. The nanoscale morphology of the prepared nanoparticulate materials (size, shape) was characterized using JEOL2011 transmission electron microscopy (TEM) operated at 200 keV with LaB<sub>6</sub> filament as the electron-beam source. X-ray photoelectron spectra (XPS) were gathered using a VG Scientific ESCALAB 250 system equipped with a micro-focused, monochromated Al Kα X-ray source. The spectra were acquired with 160 eV pass energy for full range scan and 40 eV pass energy for detailed scan.

### 2.3. Plasma catalysis process

Fig. 1a presents the dielectric barrier discharge reactor for acetaldehyde removal study. It is composed by an external quartz cylinder (2 cm wide and 10 cm long), externally covered by a silver plate and grounded. The high voltage sinusoidal signal is applied to a stainless steel cylindrical center electrode (1 cm in diameter) through a 5 mm gap. The experiment setup for the catalyst surface modification study is shown in Fig. 1b. Both of the two plane-parallel stainless-steel electrodes (100 mm × 100 mm) are covered by 1 mm thick quartz glasses. The gas gap can be adjusted in the range of 0–20 mm. The upper electrode can be connected to an AC power supply, and the DBD plasma is generated between the two parallel plate electrodes. In order to easily access the XPS and SEM analysis, Au/TiO<sub>2</sub> catalyst were prepared using the same method (Section 2.1) on a glass plate. The sample is put in the

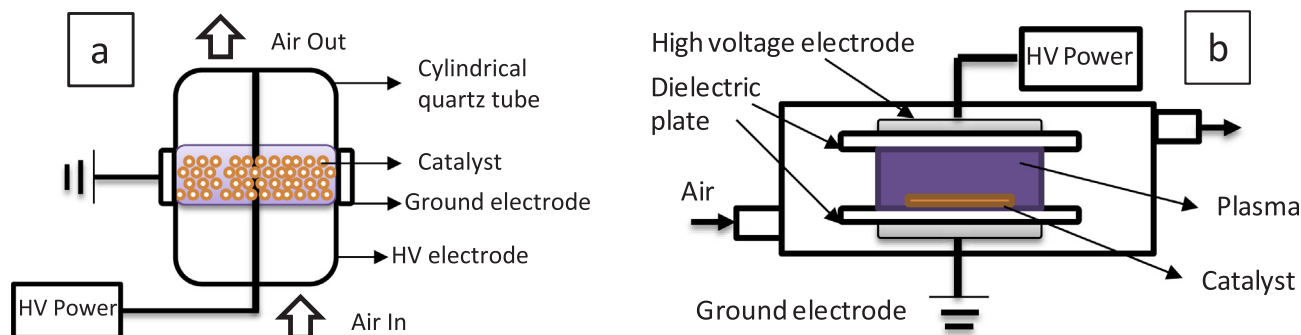


Fig. 1. Dielectric barrier discharge reactor for acetaldehyde removal study (a) and catalyst surface modification study (b).

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