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A study of pollution removal in exhaust gases from animal quartering centers by combining photocatalysis with surface discharge plasma: From pilot to industrial scale

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ABSTRACT

In this study, a continuous process combining plasma and photocatalysis, at pilot scale, has been investigated on pollution treatment.

The degradation efficiency and by-products formation has been monitored through the variation of multiple parameters such as inlet concentration, specific energy, and residence time. Results show that process combination leads to an enhancement of pollutant degradation compared to the separate systems even at high flow rate (250 $m^3 h^{-1}$). Synergy factor due to this association ranged from 1.10 to 1.17. Furthermore, CO_2 selectivity has significantly improved when comparing the combined system to that with plasma alone. Moreover, we note that adding a photocatalyst in the discharge zone leads to a reduction of ozone and CO formation compared to plasma process alone.

Based on this investigation, a methodology of scaling up the combined system was proposed. Indeed, as a practical application, an industrial reactor has been developed and tested for treating a real pollution from animal quartering centers. This investigation indicates that the combined system of photocatalysis with plasma is a promising method for removing air pollutants.

1. Introduction

Many studies have shown that Volatile Organic Compounds (VOC) emissions are not negligible and their impacts on the environment and human health are serious and threatening [1,2]. Therefore, there is currently an important deal of interest in developing processes to treat air pollutants, such as photocatalytic (UV/TiO_2) oxidation in the gas phase. UV/TiO_2 process is an attractive technique, as heterogeneous photocatalysis for environmental clean-up. It operates at ambient temperature and the by-products are usually CO_2, H_2O and non-harmful mineral acids [3–5]. Also, non-thermal plasma (NTP) is one of the most frequently used oxidation processes to control air pollution [6]. It consists of generating an electrical discharge that treats gas present between electrodes separated by a dielectric material. However, application of non-thermal plasma leads to low the mineralization [7]. Photocatalysis, due to its adsorption step, tends to keep the by-products in the adsorbed state in order to reach their complete oxidation, achieving, thus, a higher mineralization value [3–5]. Therefore, it is interesting to try enhancing NTP's activity by adding TiO_2 with external UV [8]. Moreover, many researchers found that combining plasma and photocatalysis improved the global performance of the process [9–11]. Indeed, a synergetic effect can be expected between these technologies [9–11]. Although many studies were carried out on the efficiency of associated process at lab scale, the extrapolation of this process at industrial scale has not been extensively explored. Here, our methodology was to extend previous research [12,13] by proposing a new investigation about plasma, photocatalysis and their association *in-situ* at pilot and industrial scales. Firstly, a special attention is given to the comparison of the obtained results at pilot scale with those obtained at lab-scale, which is innovative in comparison to latter studies [14]. The second innovation of this research is about

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the investigation of the real effluent treatment with an industrial reactor.

At pilot scale study, isovaleraldehyde was chosen as a test molecule because it is significantly present in exhaust gases from animal quartering [15]. Furthermore, it is listed as one of the surrogates for the six major classes of indoor VOCs (aromatic, aldehyde, alkane, ketone, alcohol, and chlorocarbon) by the French Environment and Energy Management Agency (ADEME) [15].

In this investigation, a new photocatalysis-surface plasma hybrid system is incorporated in a pilot reactor and the influence of some operating parameters on the removal of exhaust gases from animal quartering centers is investigated. On the other hand, at the same scale, Ochiai and co-workers investigated and developed a photocatalysis-plasma hybrid air-purification reactor using a titanium mesh sheet modified with TiO_2 (TMiP TM) and a surface discharge-induced plasma (SPCP) unit for treat a gas of smoking area [9].

Here, we test the combined system with a novel photocatalyst of glass fiber tissue combined with surface discharge plasma which is established in a continuous reactor to investigate the influence of some operating parameters on the removal in exhaust gases from animal quartering centers.

2. Materials and methods

2.1. Materials and methods at pilot scale

The experimental part will include two major elements: the reactor description and the gas sampling and analysis.

2.1.1. Reactor description

The experimental set-up is an air handling unit manufactured by CIAT (Compagnie Industrielle d'Applications Thermiques-France) with a flow rate capacity up to 5000 $m^3 h^{-1}$ (Fig. 1). It contains a pre-filtration box, a cooling bank, a vapor humidifier and an electrical heater. The second box contains the pollutant injection zone. Two boxes (A & B) permit an upstream and a downstream concentration pollution sampling. Photocatalytic and plasma treatment system is installed between these boxes (A & B), and finally an activated carbon filter is maintained at the exit in order to adsorb the residual pollution. A booster is used to generate the air flow rate. Its quantification is done thanks to a diaphragm by measuring a pressure drop. A "Testo 625" probe is used to measure the temperature and the relative humidity. This pilot was better described in our previous study concerning an investigation with photocatalysis alone [16].

The high voltage electrode was an array of grid wires placed at approximately 4 mm above the ground electrode. The grid had a thickness of 2 mm and the distance between adjacent wires was 20 mm. The ground electrode was a copper tape placed into a plat of glass. The photocatalytic zone had a cross-section area (0.30 m × 0.30 m). It contains the pleated photocatalytic media, which is maintained by grid wire into glass plate, offering a surface of 2.16 m^2. This area was irradiated by 36 Philips UVA lamps (PL-L 24W/827/4P). The used material was a coated Glass Fiber Tissue (GFT) with 6.5 g m^{-2} of colloidal silica to ensure the fixation of 6.5 g m^{-2} of titanium dioxide nanoparticles. It is supplied by Ahlstrom Research and Services. This photocatalytic material had been described in detail in our previous works [3,12–14].

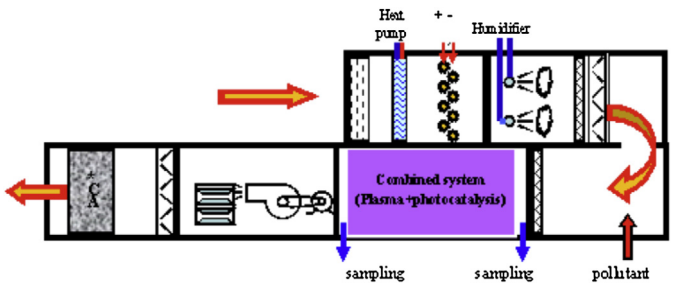

Fig. 1. A functional diagram of pilot unit supplied by CIAT.

Plasma was obtained by submitting the electrodes to a sinusoidal high voltage ranging from 0 to 30 kV at a 50 Hz frequency. The outer electrode was connected to the ground through a 50 nF for collecting the charges transferred from the reactor. The applied voltage (U_a) and high capacitance voltage (U_m) were measured by LeCroy high voltage probes and recorded by a Lecroy oscilloscope (Wave Surfer 24 Xs, 200 MHz).

2.1.2. Gas sampling and analysis

The preparation of synthetic polluted air stream was largely described in previous studies [3,12–14]. Briefly, the air-isovaleraldehyde gas mixture was prepared by passing synthetic air (Air Liquide) through liquid isovaleraldehyde (Sigma-Aldrich, 97%). Indeed, pollutant liquid was heated, vaporized and mixed with a zero-air flow in an especially designed Bronkhorst vaporization/mixing chamber (CEM). In these conditions, the tested concentrations ranged from 2 to 10 mg m^{-3}. Gas samples were analyzed by gas chromatography with flame ionization detection (Chrompact FFAP-CB column). A standard iodometric titration method was used to estimate the formation of the downstream ozone.

CO concentrations were measured by CO ZRE gas analyzer. To monitor the progression of pollutant mineralization, CO_2 was analyzed by a Cosma Beryl analyzer reference 100 equipped by spectrophotometer Fourier Transform Infrared (FTIR) brand Environnement SA.

2.2. Materials and methods at industrial application

Several materials were used in order to better investigate and analyze the performance of combined system.

2.2.1. Analysis system

The industrial emissions were measured by a FISONS Gas-phase Chromatograph (GC) using a Flame Ionisation Detector (FID). Indeed, different Carbotrap cartridges have been used in order to concentrate the pollution: Carbopack Y, allows the selective retention of heavy compounds from C_{12} to C_{20}. Another Carbopack B traps the compounds C_5 to C_{12}, while the light compounds from C_2 to C_5were adsorbed with Carboxen 1003. Thus, pollution was firstly concentrated on different Carbotrap (Supelco); the pumping was done by a Gillian LFS-113 pump under a flow rate of 50 mL min^{-1}. Secondly, Turbomatrix Perkin Elmer unit was used to desorb thermally the concentrated compounds, which will be analyzed through a transfer line to the GC/MS Perkin Elmer Clarus 500. A detailed description of the experimental protocol of analysis was published elsewhere [13]. In addition, TRS MEDOR analyzer (Chromatotec, France) was used to analyze the sulfur compounds concentrations. In fact, the sample under a 100 mL min^{-1} flowrate was continuously swept to an analyzer which was equipped by a capillary column and an electrochemical detection [14].

Along with the concentration on Carbotrap, a series of analysis were done *in-situ* using the equipment provided by Explorair Company (France). A thermal desorption unit coupled to Fast Gas Chromatograph-Mass spectrometer (TD/FASTGC/MS) was used to analyze concentration at different sampling points A, B, C and D (Fig. 2). This TD/FAST GC/MS gives new possibilities for continuous analysis by offering excellent performances represented by the speed, flexibility, sensitivity and repeatability of the analysis. An RTX column (20 m length 0.18 mm external diameter and 1 μm thickness), which is especially adapted for volatile fatty acids, was