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# CONVERSION OF PROPANE BY PULSED DIELECTRIC BARRIER DISCHARGES IN AIR

O. Aubry, A. Khacef and J.M. Cormier

*GREMI, Polytech'Orléans, 14 rue d'Issoudun, 45067 Orléans cedex 2, FRANCE*

**ABSTRACT.** Dielectric barrier discharge reactor has been used to study the effect of voltage, pulse repetition rate and inlet mixture composition on propane conversion. The reactor is powered by a pulsed high voltage supply with amplitudes up to 30 kV and pulses repetition rate varied up to 200 Hz. Injected energy density and dry or wet air conditions play a major role on propane removal efficiency and the species produced (nature and concentrations).

## 1. INTRODUCTION

Non-thermal atmospheric pressure plasma processing is being actively studied for the effective removal of volatile organic compounds (VOCs') diluted in air [1-4]. Despite well-established implementations, fundamental knowledge of chemical processes need to be improved: products analysis, mass balance. Propane conversion results in a pulsed dielectric barrier discharge reactor (DBD) are reported in this paper. The aim is to characterize experimentally removal efficiency and the products by varying of voltages and pulse repetition rates.

## 2. EXPERIMENTAL

The experimental set-up used in this work was described previously [1,5-7]. The composition of the inlet gas mixture was controlled by mass flow controllers and the total flow rate was fixed at 1 $L.min^{-1}$. The inlet mixtures composition was dry or water saturated air with 1050 ppm of propane at room temperature and atmospheric pressure. The geometry of the DBD reactor was a wire to cylinder type.

The high-voltage generator is composed of two parts: the first one is a bank of ceramic knob storage and transfer capacitors of 4 nF. Each capacitor was connected in a Blumlein-type configuration. The second one was the cable transformer system. The storage capacitors were charged at a voltage ($U_{ch}$), and commuted with a fast switching element. Then, this high voltage fed the DBD through the cable transformer. A maximum open circuit voltage of six times the charging voltage of each cable was achieved. This pulse generator was able to deliver open circuit voltage up to 150 kV output into 80 ns (FWHM) pulses and short rise time (40 ns). In ours experiments, $U_{ch}$ was between 3.5 and 5 kV and applied high voltage in the plasma reactor reached up to 30 kV ($U_{appl}$). The pulse repetition rate was adjustable, up to 200 Hz. Applied voltages and current measurements were displayed with an oscilloscope through high voltage and current probes.

Chemical analyses were performed by Fourier Transform Infra Red (FTIR, Nicolet Magna-IR 550 series II). Species quantified are only $C_3H_8$, $CO_2$ and CO. In addition of these main

products, by-products such as formaldehyde, acetaldehyde, formic acid, $NO_x$ and $C_xH_yNO_z$ were observed.

## 3. RESULTS AND DISCUSSION

### 3.1. Propane conversion

The discharge pulse energy into the plasma was determined with a capacitive circuit and was given by: $Ed = \frac{(0.5\,C\,U_{ch}^2)\,f}{Q}$ where C is the transfer capacitance of the capacitors bank, $U_{ch}$ the charging voltage, f the pulse repetition rate and Q the total flow rate. Charging voltages and pulse repetition rates range studied were the following: 3.5 kV, 15-200 Hz; 4.5 kV, 100-200 Hz; and 5 kV, 15-150 Hz. From these values, we reported species concentrations as functions of energy density (Ed). Ed range studied in this work is between 0 and 900 J/L.

In figure 1a, the effect of Ed on propane concentration in dry air is reported. Propane conversion rate increases significantly with increasing Ed. For a given charging voltage, a rise of the frequency implies an enhancement of the $C_3H_8$ consumption. This is observed too for a given f when $U_{ch}$ increases. $C_3H_8$ is totally consumed when Ed is higher than 700 J/L. Ed is not the main parameter on the propane removal efficiency: for a given Ed, propane consumption depends on $U_{ch}$ and f. For Ed=700 J/L, we have same conversion rates for $U_{ch}$ equals to 4.5 and 5.0 kV and for pulse repetition rates of 200 and 125 Hz respectively.

Figures 1b and 1c show the influences of Ed and charging voltages on propane conversion rates in wet and dry air conditions. We can observe a similar effect of Ed on propane conversion rate in humid air and dry air: an increase of Ed leads to a rise of the conversion rate. Humid air conditions leads to a beneficial effect on the propane consumption with an increase of the propane conversion rate for the lowest $U_{ch}$. When the voltage rises, the effect decrease strongly: for 5 kV, a minor effect is observed when F<75 Hz.

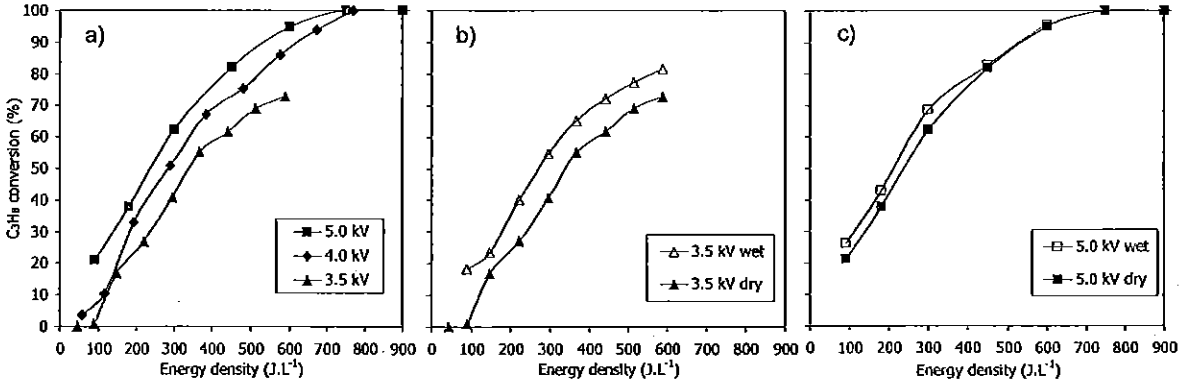


**FIGURE 1.** $C_3H_8$ conversion rates as functions of Ed and charging voltages in: a) dry air and b) wet air.

### 3.2. Species produced

Figures 2 display CO and $CO_2$ concentrations as functions of Ed for various $U_{ch}$ and f values for propane diluted in dry air (full symbols). $CO_x$ are the main carbon species produced when propane is consumed. An increase of Ed leads to a rise of CO and $CO_2$ concentrations. For a given Ed, there is a voltage effect between 3.5 and 4.5 kV, species concentrations increase. Between the highest voltages (4.5 and 5.0 kV), none effect of $U_{ch}$ is observed at given Ed. In the lowest Ed range, $CO_2$ concentrations are higher than CO ones and this range

is varying with $U_{ch}$: 0-300 J/L for 3.5 kV and 0-180 J/L for 5 kV.

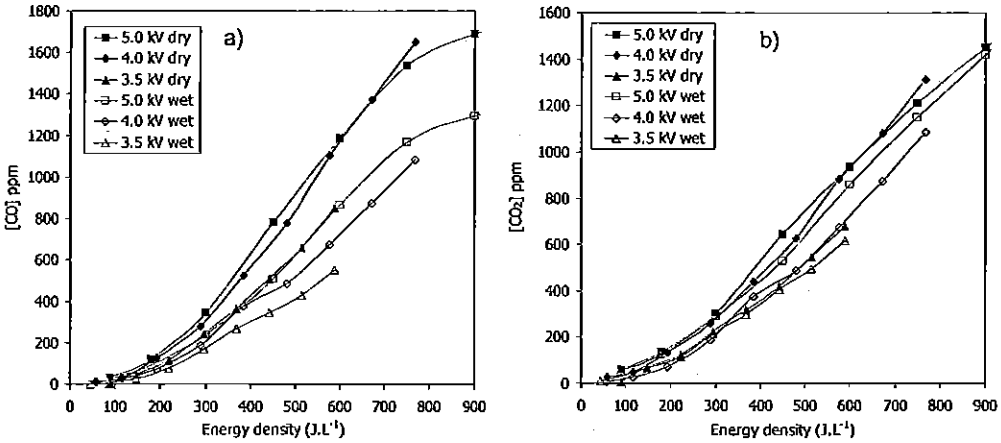


**FIGURE 2.** Effect of energy density on: a) CO, and b) $CO_2$ concentrations in dry and wet conditions.

In wet air conditions, CO and $CO_2$ concentrations increase when Ed rises (figures 2, open symbols). A comparison between results in dry and wet air shows that CO concentration strongly depends on the inlet mixture composition (up to 400 ppm for Ed=700 J/L). An increase of $U_{ch}$ implies same effects on [CO] in dry or wet air conditions. Concerning $CO_2$ produced, humid air plays a minor role on $[CO_2]$, a light decrease of $CO_2$ produced appears in wet air conditions (up to 200 ppm for $U_{ch}$=4 kV).

From non-consumed $C_3H_8$, CO and $CO_2$ concentrations, we calculated $CO+CO_2$ selectivity, $S_{CO+CO2}$, and measured carbon concentration, $C_M$, expressed by:
$S_{CO+CO2}$=([CO]+[$CO_2$])/(inlet total carbon) and $[C_M]=3.[C_3H_8]+[CO]+[CO_2]$, respectively.

Effects of Ed on $S_{CO+CO2}$ are reported in figure 3a and ones on $[C_M]$ are shown in figures 3b and 3c. For the highest Ed (>700 J/L), CO and $CO_2$ are only the carbon species produced in dry air: $S_{CO+CO2}$ is close to 100% and $C_M$ is equal to the inlet carbon injected (3150 ppm). In wet air, maximum $S_{CO+CO2}$ is only 80% at 900 J/L. $C_M$ curves show effects of Ed on the species produced and influences of charging voltage.

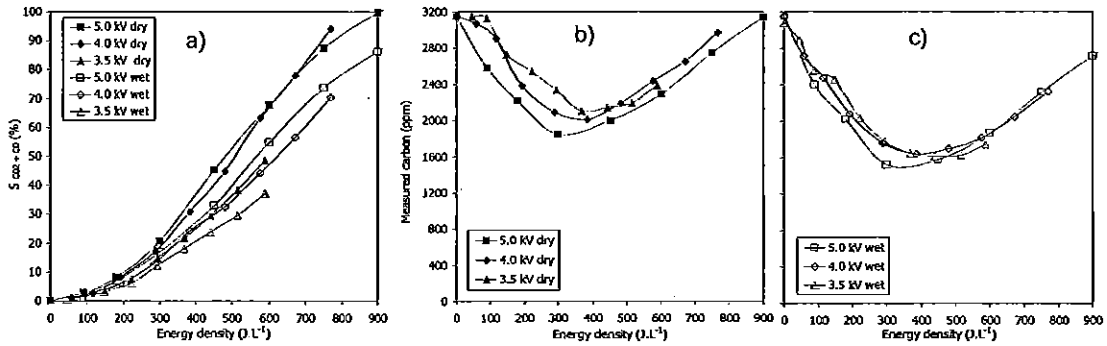


**FIGURE 3.** Effects of Ed on: a) $CO+CO_2$ selectivity, b) $C_M$ in dry air and c) $C_M$ in wet air.

$\Delta C$ is defined as the difference between $C_M$ and the initial carbon injected. $\Delta C \neq 0$ means that others carbon species are produced ($C_xH_yO_z$) and mainly between 150 and 800 J/L. The $C_xH_yO_z$ production is linked to the humidity in air and to $U_{ch}$ for the lowest Ed values. Maximum carbon deficit, $\Delta C$, in dry air is reached at 300 J/L for 5 kV, 350 J/L for 4.0 kV and 375 J/L for 3.5 kV and $\Delta C$ is 1300, 1160 and 1050 ppm, respectively. In wet air, $\Delta C$ increases

up to 1600 ppm for 5 kV.

$CH_2O$ areas measured from experimental FTIR spectrum are reported in figures 4. Concentrations of formaldehyde are higher for dry conditions excepted for the lowest charging voltage studied where none influence of the air conditions are observed. In humid air, $CH_2O$ is not promoted excepted for the higher Ed values where $S_{CO+CO2}$ is lower. The trends of $CH_2O$ correspond to the carbon deficit observed in figure 4b. Maximum $CH_2O$ concentrations are reached between 300 and 400 J/L in function of $U_{ch}$. FTIR spectra show that others $C_xH_yN_wO_z$ species are promoted in wet air conditions.

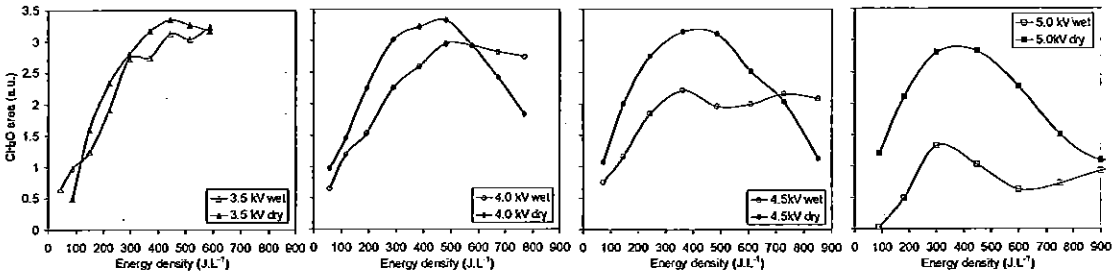


**FIGURE 4.** Effects of Ed, charging voltages and air conditions on $CH_2O$ areas measured by FTIR.

## CONCLUSION

This work shows that high propane conversion efficiency can be reached with an energy density equals to 700 J/L and main species produced are CO and $CO_2$. $CO+CO_2$ Selectivity reaches 100 % when propane is entirely consumed. Propane diluted in humid air implies a increase of the propane conversion for the lowest charging voltage studied and $CO+CO_2$ selectivity decreases in favor of $C_xH_yO_z$ species with $x > 2$.

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