



HAL
open science

Conversion of propane by pulsed dielectric barrier discharges in air

Olivier Aubry, Ahmed Khacef, Jean Marie Cormier

► **To cite this version:**

Olivier Aubry, Ahmed Khacef, Jean Marie Cormier. Conversion of propane by pulsed dielectric barrier discharges in air. 10th International Symposium on High Pressure Low Temperature Plasma Chemistry, 2006, Saga, Japan. pp.7P-07. hal-00817903

HAL Id: hal-00817903

<https://hal.science/hal-00817903>

Submitted on 25 Apr 2013

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

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

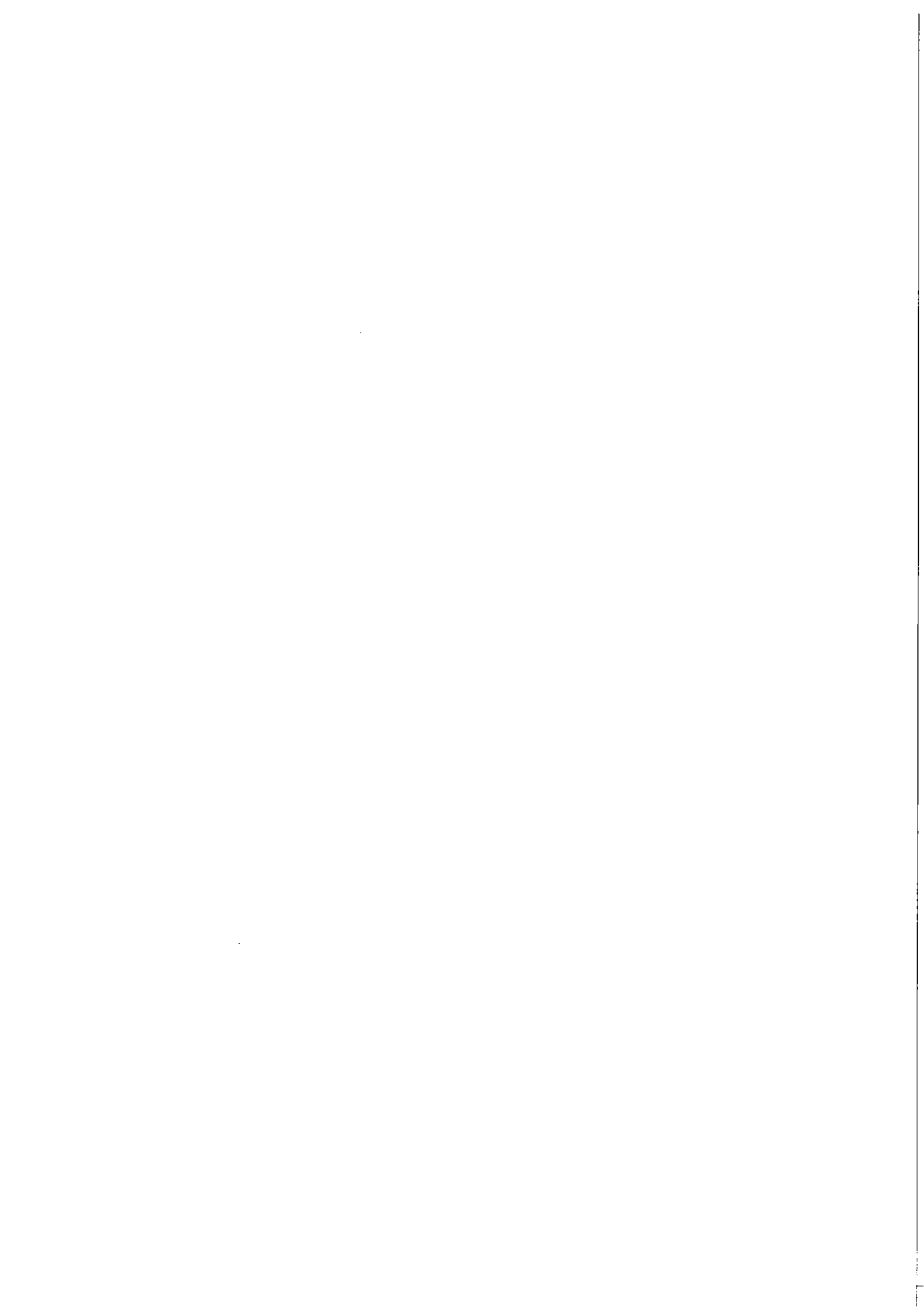
Non-thermal atmospheric pressure plasma processing is being actively studied for the effective removal of volatile organic compounds (VOCs) diluted in air [1-4]. Despite well-established implementations, fundamental knowledge of chemical processes need to be improved: products analysis, mass balance. 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 C₃H₈, CO₂ and CO. In addition of these main



products, by-products such as formaldehyde, acetaldehyde, formic acid, NO_x and $\text{C}_x\text{H}_y\text{NO}_z$ 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.

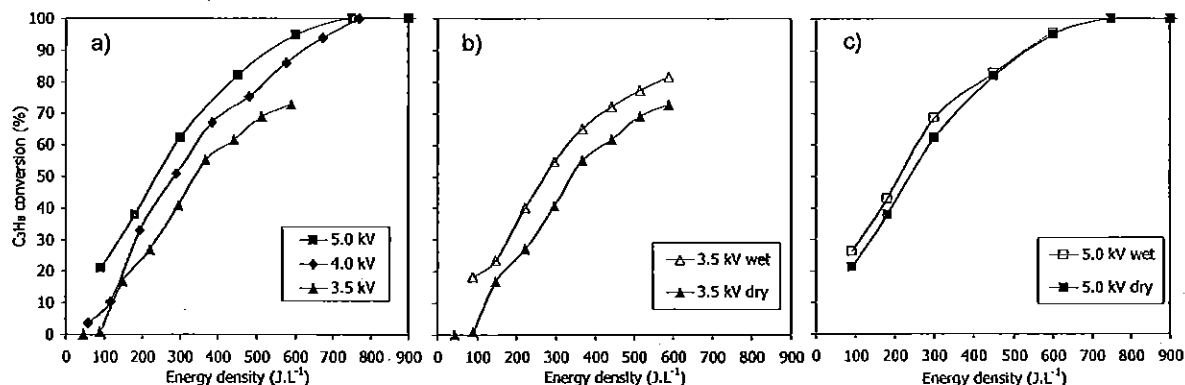
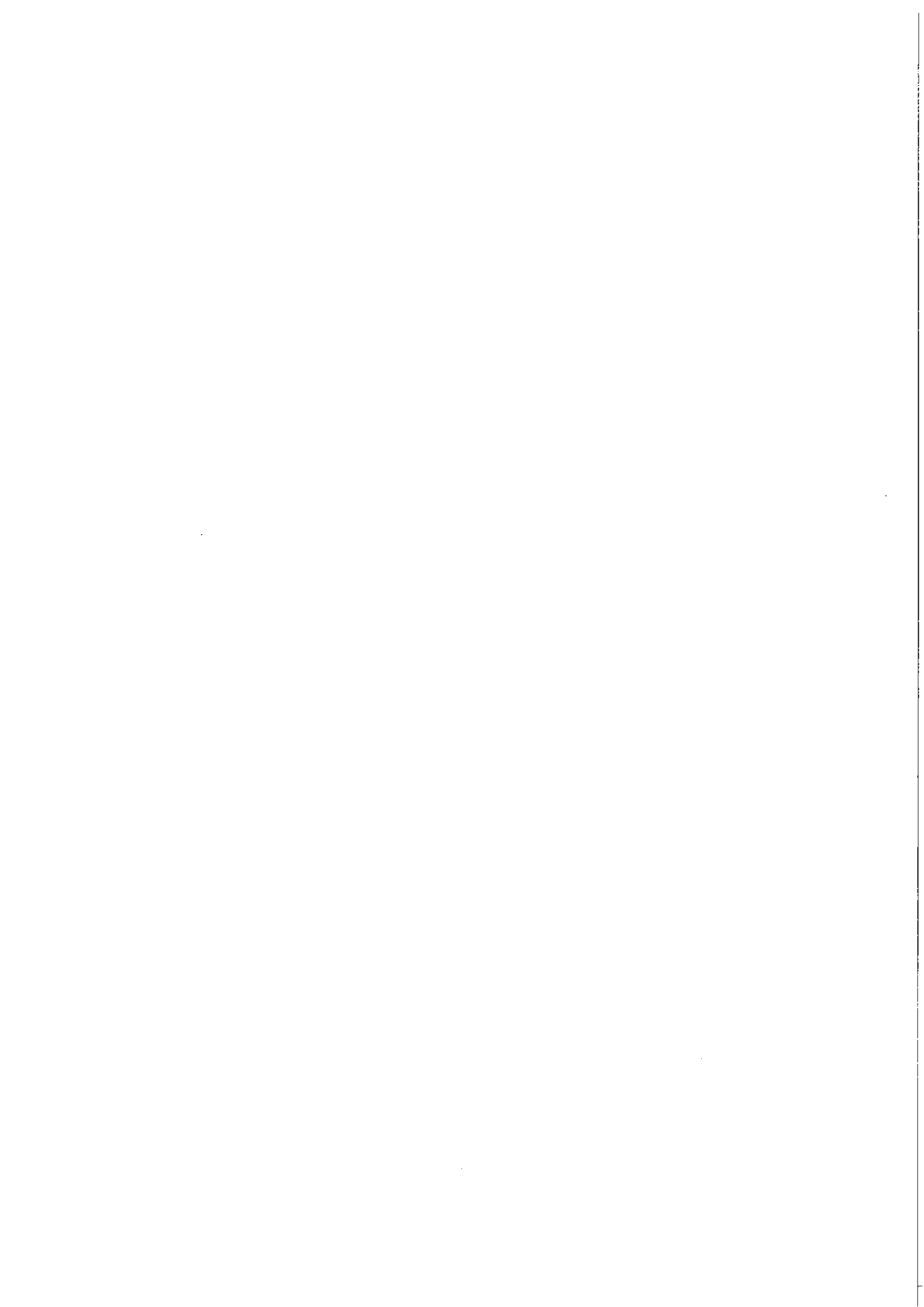


FIGURE 1. C_3H_8 conversion rates as functions of Ed and charging voltages in: a) dry air and b) wet air.

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.

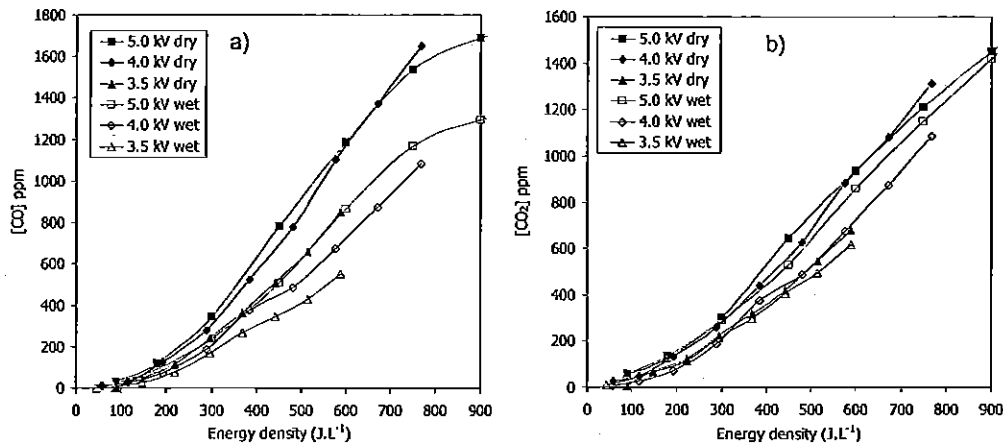


FIGURE 2. Effect of energy density on: a) CO, and b) CO₂ concentrations in dry and wet conditions.

In wet air conditions, CO and CO₂ 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 CO₂ produced, humid air plays a minor role on [CO₂], a light decrease of CO₂ produced appears in wet air conditions (up to 200 ppm for U_{ch} =4 kV).

From non-consumed C₃H₈, CO and CO₂ concentrations, we calculated CO+CO₂ selectivity, S_{CO+CO_2} , and measured carbon concentration, C_M , expressed by:
 $S_{CO+CO_2} = ([CO] + [CO_2]) / (\text{inlet total carbon})$ and $[C_M] = 3 \cdot [C_3H_8] + [CO] + [CO_2]$, respectively.

Effects of Ed on S_{CO+CO_2} 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 CO₂ are only the carbon species produced in dry air: S_{CO+CO_2} is close to 100% and C_M is equal to the inlet carbon injected (3150 ppm). In wet air, maximum S_{CO+CO_2} is only 80% at 900 J/L. C_M curves show effects of Ed on the species produced and influences of charging voltage.

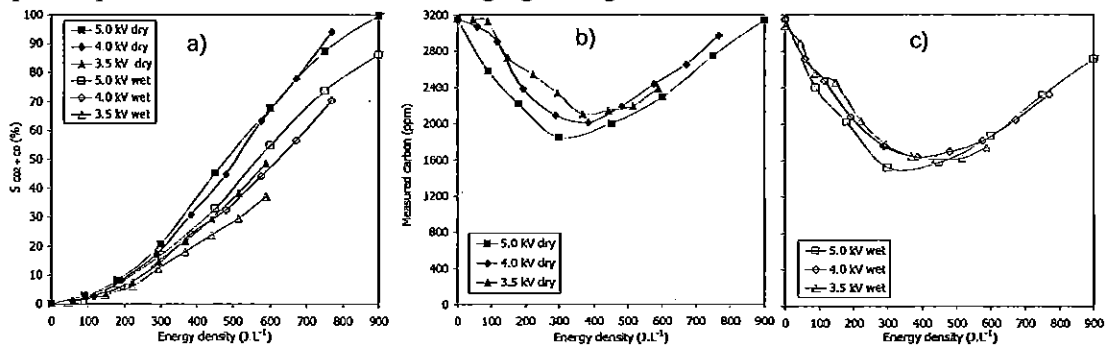
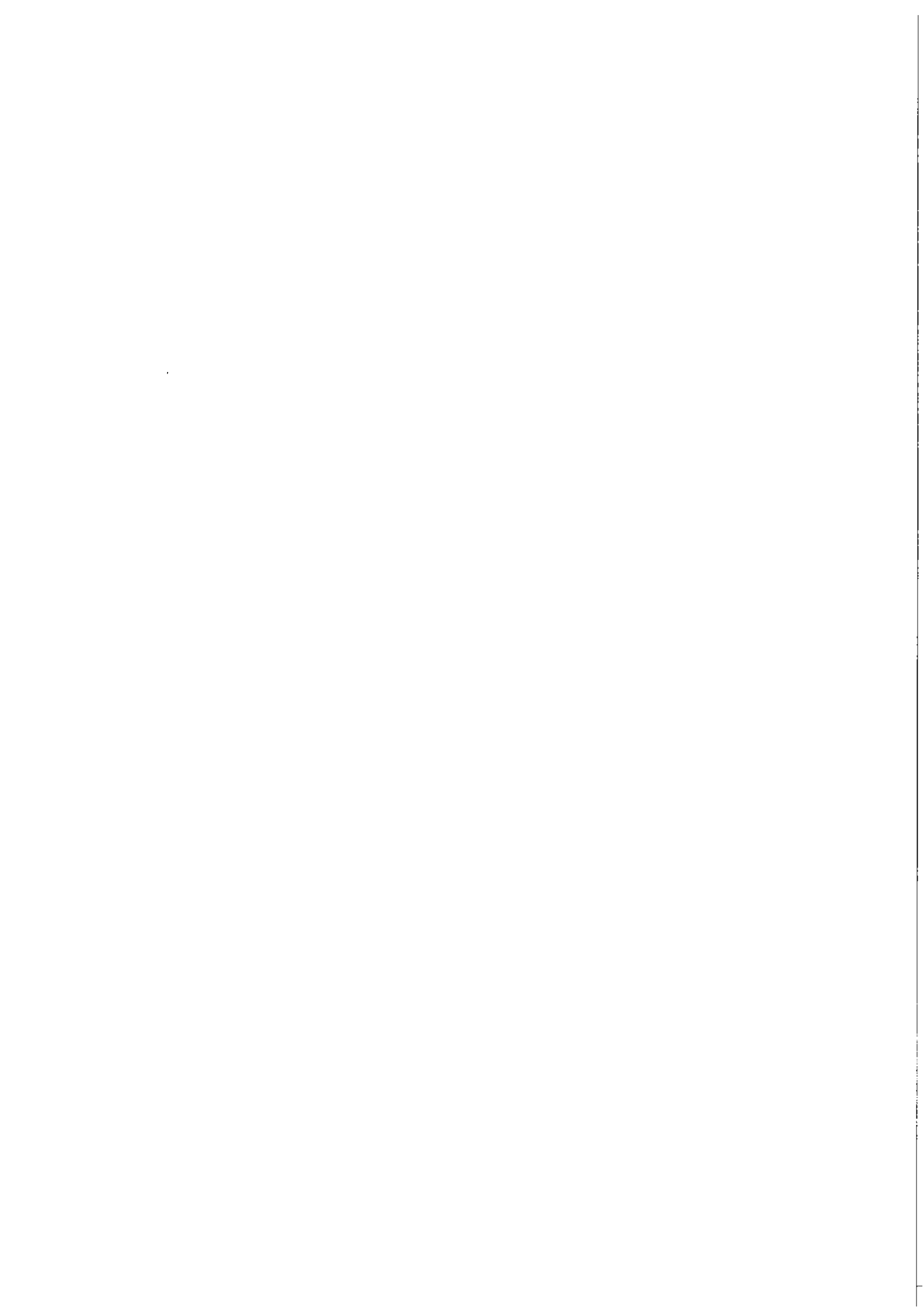


FIGURE 3. Effects of Ed on: a) CO+CO₂ selectivity, b) C_M in dry air and c) C_M in wet air.

ΔC 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, ΔC , 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 ΔC is 1300, 1160 and 1050 ppm, respectively. In wet air, ΔC increases



up to 1600 ppm for 5 kV.

CH₂O 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, CH₂O is not promoted excepted for the higher Ed values where S_{CO+CO₂} is lower. The trends of CH₂O correspond to the carbon deficit observed in figure 4b. Maximum CH₂O 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.

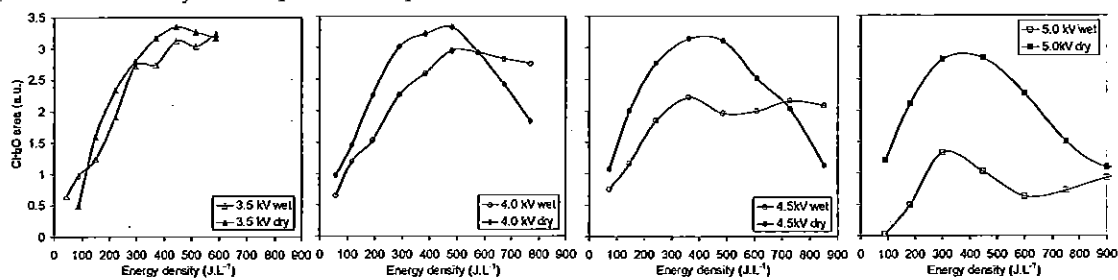


FIGURE 4. Effects of Ed, charging voltages and air conditions on CH₂O areas measured by FTIR.

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 CO₂. CO+CO₂ 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+CO₂ selectivity decreases in favor of C_xH_yO_z species with x > 2.

REFERENCES

- [1] Motret O., Aubry O., Thuillier C., Lascaud M., Met C., Cormier J.M. (2005). Conversion of Ethylene by Pulsed Dielectric Barrier Discharge in Dry and Wet - Parametrical Study. *Proc. 17th Int. Symp. Plasma Chemistry*, Toronto (Canada).
- [2] McAdams R. (2001). Prospects for non-thermal atmospheric plasmas for pollution abatement. *J. Phys. D: Appl. Phys.*, 34, 2810-2821.
- [3] Guo Y.F., Ye D.Q., Chen K.F., Tian Y.F. (2006). Humidity effect on Toluene Decomposition in a Wire-plat dielectric barrier discharge reactor. *Plasma Chem. Plasma Process*, 26, 237-249.
- [4] Blin-Simiand N., Jorand J., Belhadj-Miled Z., Pasquiers S., Postel C. (2006). Influence of temperature on the removal of toluene by dielectric barrier discharge. *Proc. ISNTPT 5*, Oleron Island (France).
- [5] Khacef, A., Viladrosa C., Cachoncinlle C., Robert E., Pouvesle J.M. (1997). High repetition rate compact source of nanosecond pulses of 5–100 keV x-ray photons, *Rev. Sci. Instrum.*, 68, 2292-2297.
- [6] Khacef A., Cormier J.M., Pouvesle J.M. (2002). NO_x remediation in oxygen-rich exhaust gas using atmospheric pressure non-thermal plasma generated by a pulsed nanosecond dielectric barrier discharge, *J. Phys. D: Appl. Phys.*, 35, 1491-1498.
- [7] Aubry O., Khacef A., Cormier J.M. (2006). CO conversion by a dielectric barrier discharges using the water gas shift reaction. *Proc. ISNTPT 5*, Oleron Island (France).

