Influence of the voltage waveform of a DBD discharge on the conversion of CH_4 and CO_2

N. R. Pinhão [(*)1], A. Janeco [1], L. M. Redondo [2], H. Canacsinh [2], J. Branco [1]

[1] *IST – Instituto Tecnológico e Nuclear, Estrada Nacional 10, 2686-953 Sacavém, Portugal*
[2] *Instituto Superior de Engenharia de Lisboa, R. Conselheiro Emídio Navarro, 1, 1959-007 Lisboa*
[(*)] npinhao@itn.pt

We report the results obtained in rare-gas/CH_4/CO_2 mixtures in a DBD discharge driven by sinusoidal or square voltage waveforms. For the same peak voltage and specific input energy, the values for conversion of CH_4 and CO_2 and the selectivity for H_2 and CO production are significantly higher with the square waveform. In this case the DBD current is characterized by two pulses per period. If the power supply frequency is sufficiently low, the first pulse is broader and with a lower absolute value. The difference between the two pulses is an indication that a memory effect is present on the development of each new pulse.

Introduction

The use of a non-thermal plasma for methane conversion has been studied as an alternative process for the production of *Syngas* and direct production of higher hydrocarbons [1,2]. A large number of these system rely on dielectric barrier discharges (DBD), frequently with a catalyst, where methane is mixed with an oxidant. However the energy efficiency has been relatively low and different alternative mixtures or systems have been studied [3,4] to increase the conversion, selectivity and energy efficiency.

In this work we report a comparison between the results obtained in a DBD discharge maintained by power supplies with a sinusoidal or a rectangular waveform in mixtures with interest for the production of *Syngas*.

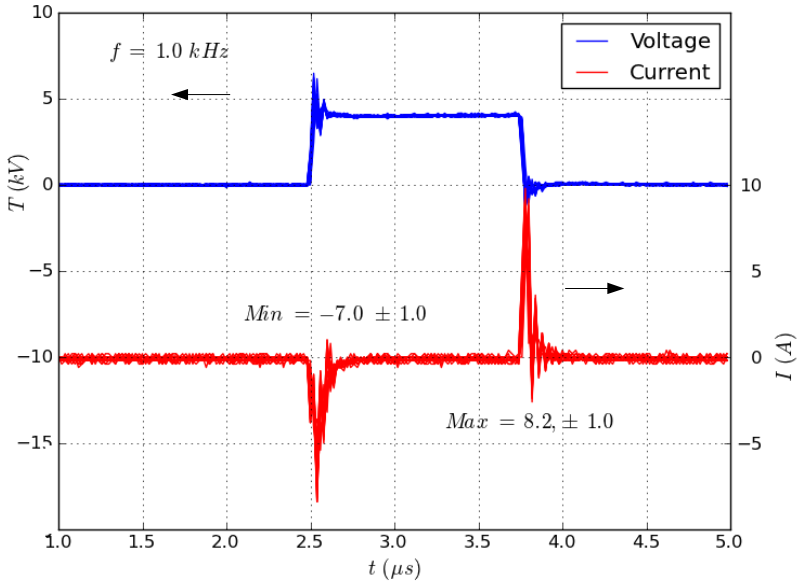

Fig 1: Square voltage pulse and DBD current for a frequency of 1 kHz. The minimum and maximum values are the average of 10 measurements.

Method

The cylindrical DBD reactor consists of a 10 mm I.D. Pyrex tube covered by a (ground) aluminium foil. The inner electrode is a SS rod of 5 mm diameter. The discharge works at atmospheric pressure and the gas mixtures used are rare-gas/CH_4/CO_2 mixtures where [CH_4]:[CO_2] = 1 and the rare gas is

helium or argon with mole fraction between 60% and 85 %. The electrodes are powered with a AC waveform with frequency between 4 kHz and 6 kHz or with positive square pulses of 14 µs duration and frequency between 1 and 12 kHz. The current pulses were measured with a *Pearson* Rogowski coil and a fast digital oscilloscope and the values of power obtained from those signals. The output gas composition was analysed on-line by gas chromatography using a *Restek* ShinCarbon ST column and a *Shimadzu* 9A GC equipped with a thermal conductivity detector (TCD) and a 6-port gas sampling valve with a 0.25 lL loop.

Results

We have obtained the voltage and current signals with both voltage waveforms. With a square voltage waveform only two current pulses are obtained, corresponding to the square voltage transitions. However for the same peak voltage the absolute value of amplitude of the pulses are much higher than the pulses obtained with a sinusoidal voltage. For a square voltage waveform the shape of the first current pulse depends on the power supply frequency: for low frequency values, the pulse is broader and with a lower absolute value of amplitude, as indicated in Fig 1.

The results for conversion of CH_4 and CO_2 and the selectivity for H_2 and CO production selectivity show a marked increase and shift to lower specific input energy (ratio power/volume flow rate) as shown in figure 2. The energy efficiency of the discharge, measured from the ratio of the difference of total enthalpy of formation between the exhaust and input gases, and the energy supplied to the reactor, has also increased from 2.5% to 5%. This value, however, is still too small for industrial applications.

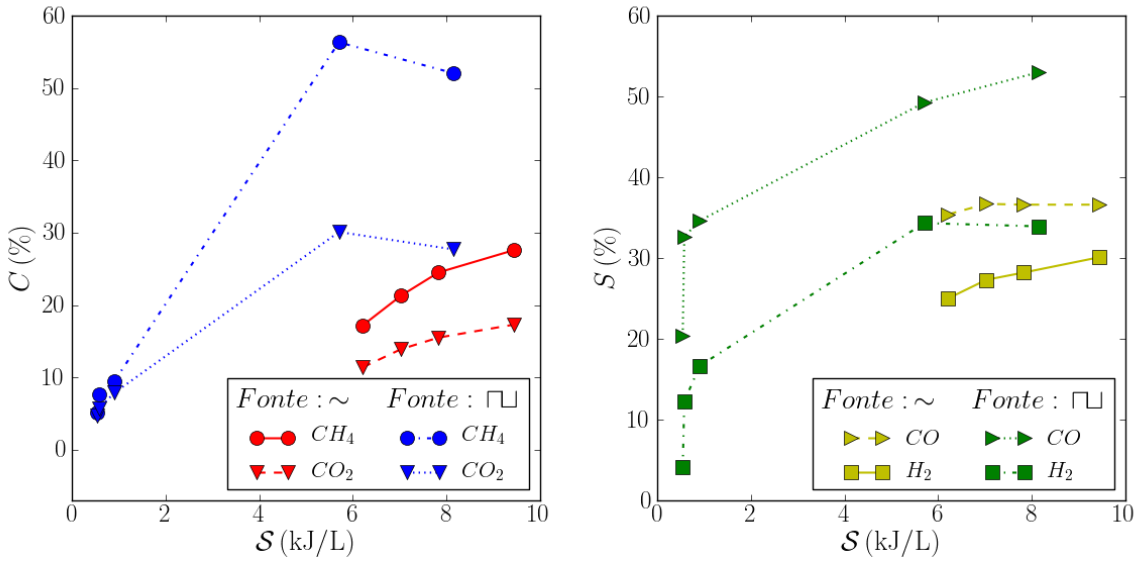

Fig 2: Values of conversion (C) and selectivity (S) as a function of the specific input energy ($\mathcal{S}$) in a mixture of 70% He with CH_4/CO_2, obtained with a sinusoidal or square power supply.

Acknowledgement

We acknowledge the financial support by FCT under the contract number PTDC/EQU-EQU/65126/2006.

References

[1] A. Indarto,J-W Choi, H. Lee, H.K. Song (2006) *J Nat Gas Chem* 15:87–92
[2] S-S. Kim, H. Lee, J-W Choi, B-K Na, HK Song (2007) Catal Commun 8:1438–1442
[3] A. Mfopara, MJ. Kirkpatrick, E. Odic (2009) *Plasma Chem Plasma Process* 29:91–102
[4] N.R. Pinhão, A. Janeco, J. B. Branco (2011) *Plasma Chem Plasma Process* 31:427–439