Electrical characterisation of an atmospheric plasma DBD reactor for non-oxidative methane to hydrogen conversion

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Abstract: The electrical characterisation of an atmospheric plasma DBD reactor used for non-oxidative methane conversion to hydrogen was carried out. Applied power, voltage, current across the discharge were assessed, and the influence of different natures of dielectrics were investigated.

Keywords: Atmospheric plasma, electrical characterisation, non-oxidative CH_4 conversion

1. Introduction

In the urge to move away from fossil fuels a growing dynamic aims to replace them with renewables. The expansion of this renewable energy as a bigger share of energy consumed worldwide suffers from the absence of effective energy storage as this forms an important limitation of intermittent energies such as wind and solar energy. Using chemical reactions allows to store that electrical energy when it cannot be used on the energy grid and therefore allows a broader use of clean energy sources. In the present work, we use an atmospheric plasma DBD cylindrical reactor for non-oxidative conversion of methane to hydrogen. An electrical characterisation is carried out. This work is mainly based on a study carried out by A. Ozkan on CO_2 conversion in which the importance on conversion of factors such as power and the nature of the dielectric material was assessed.

2. Experimental

The setup consisted of a cylindrical DBD reactor, as shown in Fig. 1 that was also used in previously published papers [1,2,3]. It consists of a central copper electrode of 22 mm diameter and a steel mesh (10 cm in length) wrapped around the walls of the reactor. The reactor walls are made out of a dielectric barrier of variable outer diameter and with an inner diameter of 26 mm, giving a 2 mm gap to the discharge.

Methane is fed in the reactor at 100 sccm and powers of 70 to 100 W were used. The dielectrics chosen for comparison were of different natures: borosilicate, quartz, alumina and mullite.

Table 1. Physical properties of the dielectric barriers

	Relative permittivity (ε_r)	R_{rms} (nm) [3]	Thermal conductivity ($W.m^{-1}.K^{-1}$) at 20 °C [3]
Alumina	9.6	6800	29
Mullite	6.0	3100	2
Borosilicate	4.6	780	1.1
Quartz	3.8	89	1.4

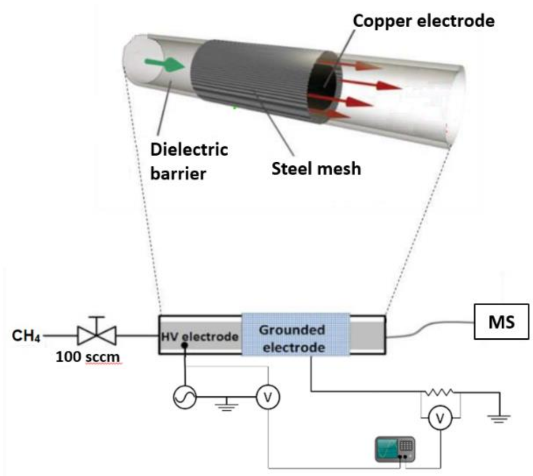

Fig. 1. Illustration of reactor and setup. Adapted from Ozkan et al (2017).

The electrical system applied an AC voltage to the reactor at a fixed frequency of 22600 Hz. Power measurements across the reactor were done using a high voltage probe (Tektronix P6015A) and additional voltage probe connected to a resistor. All electrical measurements were averaged over 3 repetitions. Lissajous plots allow to assess capacitance of the system and voltage applied to the reactor. As our discharge operates in a filamentary mode, we also investigate the microdischarges present during operation, also as Ozkan et al. suggest their importance relative to conversion in following article [3]. As the discharge current can be separated in two components: dielectric current (displacement current) and plasma current (resistive current), time-resolved plotting of the current curves gives us insights on these microdischarges: we hereby estimate their number for each period. Product characterisation in itself in the form of conversion and selectivity was done using a quadrupolar mass spectrometer (Hiden QGA).

3. Results

Using arbitrary reactor dimensions and gas flow (100 sccm) conversion of CH_4 was obtained in the range of 10-16 % depending on the chosen dielectric material, wall thickness and applied power (Fig. 2). Selectivity remained constant for these factors in the studied ranges.

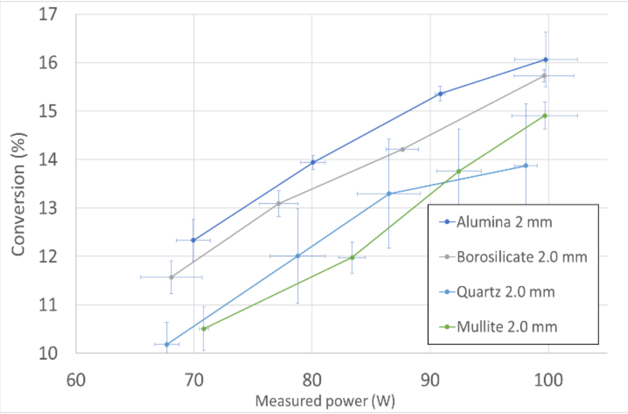

Fig. 2. Methane conversion with regards to power for Alumina, Borosilicate, Quartz and Mullite dielectrics (2 mm thickness).

A linear rise in conversion with rising measured power is illustrated by Fig. 2. This is expected as the injected charge increases with power. Also a slight increase in voltage is observed (as illustrated by Fig. 3). This means that more electrons with slightly higher energy are able to contribute to the splitting of CH_4. We note a more important contribution of the charges than of the voltage to the total area of the Lissajous plot, i.e. the total measured power, when comparing for any dielectric material at different powers (Fig. 3). A capacitor acts as a passive electrical component by storing electrical energy. As with the dielectric nature relative permittivity (ε_r) and therefore capacitance change, the ability to store that electrical energy as charges varies with the chosen dielectric. This is illustrated by Fig. 4. A higher permittivity ($\varepsilon = \varepsilon_0\varepsilon_r$) (see Table 1.) allows to store more charges with lower voltage while a lower permittivity means a higher voltage for any given power (illustrated by Fig. 4).

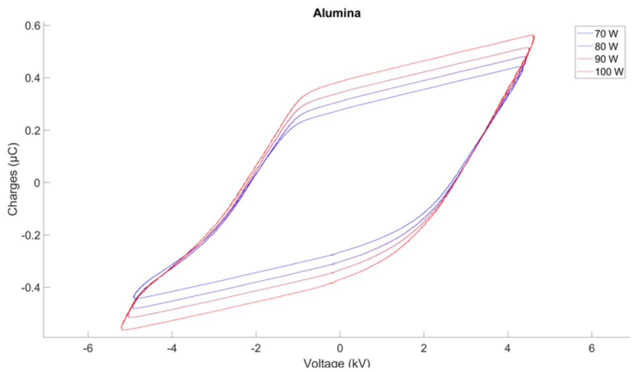

Fig. 3. Lissajous plot of DBD reactor with alumina dielectric of 2 mm wall thickness at different powers.

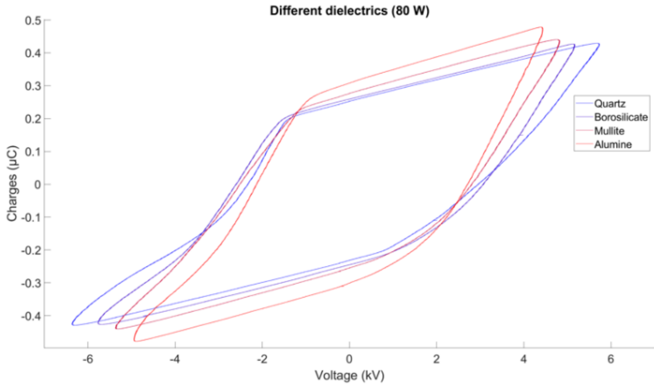

Fig. 4. Lissajous plot for dielectrics of different nature

$$Capacitance = \frac{2\pi\varepsilon_0\varepsilon_r l}{\ln\left(\frac{R_{outer}}{R_{inner}}\right)} \qquad (1)$$

The same logic as with the power (being higher number of charges is equivalent to higher conversion) is not applicable here with different dielectrics at same power. Evidently, the variation of voltage is also quite important. For the different experiments to have the same power, the Lissajous plots have to have the same total area. Therefore, a higher number of charges means lower voltage.

The same experiments (varying the capacitance) were also carried out using different wall thicknesses of the same dielectric, but no significant variation of conversion was found on the studied range (1.5 mm, 2.0 mm, 2.8 mm).

As suggested by literature [3,4,5], more factors might be at play, especially in a filamentary discharge. We investigated the microdischarges, as a filamentary plasma discharge ignites through a variable number of these microdischarges. The corresponding plasma current adds in the form of current peaks to the sine current characteristic of an RC system. We estimated the amount present in our discharge by counting the number of peaks.

As shown by Fig. 5 and Fig. 6 the amount of microdischarge peaks (present on oscillograms) does not vary significantly with regards to power or with regards to dielectric thickness. This suggests no change in the

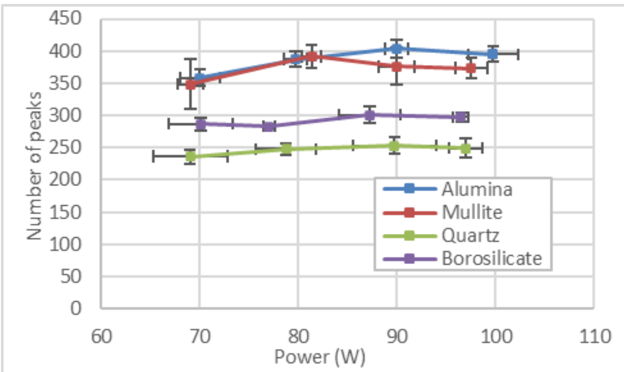

Fig. 5. Number of peaks with regards to measured power for the 4 different dielectrics.

filamentary behaviour in the studied power range but also no dependance on capacitance, as this changes with wall thickness of the dielectric.

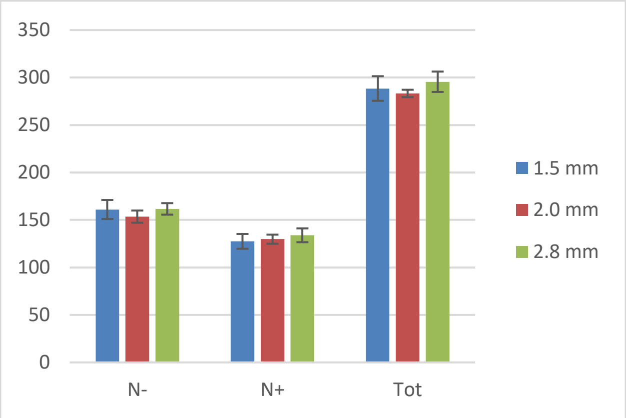

Fig. 6. Number of microdischarge peaks with regards to borosilicate wall thickness.

We did find a significant difference in the number of peaks when comparing the several dielectrics of same wall thickness at the same power (80 W) (Fig. 7.). Here, both alumina and mullite give a significantly higher number of peaks than quartz and borosilicate. This might have to do, at least partially, with the roughness of the ceramics, as discussed in following work [3]. A higher surface roughness can indeed ease the creation of filaments by reducing the gas gap locally and by the higher electric field around pins / sharp edges (pin effect). As also discussed in the mentioned works [3,4,5], thermal conductivity of the different materials might also play a role in the heating of the gas, playing a role in the amount of filaments and/or the conversion.

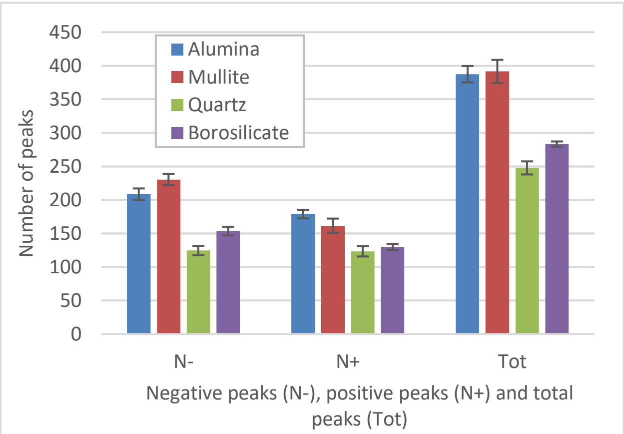

Fig. 7. Number of peaks (negative, positive and total) for alumina, mullite, quartz and borosilicate dielectrics.

4. Conclusion

An electrical characterisation of our non-oxidative methane to hydrogen reactor was carried out. We managed to understand the workings of some of the main electrical parameters on our system. We found that the added conversion with power comes mainly from the added charges than from the voltage, as the areas on the Lissajous plots suggest. We varied the capacitance of the system through the nature of the dielectric material and through the wall thickness of the material. No significant variation of the conversion could be assessed on the studied dielectric thicknesses, while changing the nature did affect the conversion, alumina showing the highest. This difference illustrates that there are more factors at play than the sole capacitance. For instance, we found a variation in the number of microdischarges, the dielectrics of highest roughnesses having more. Nevertheless, alumina and mullite showing the highest and lowest conversion of methane respectively, a study of their influence on reactor temperature is still needed, as Ozkan et al. have shown this important influence on their CO_2 conversion system. This is only a preliminary work but more experiments will be pursued in the near future to complete the study.

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