Investigation of the dry reforming reaction by plasma-catalysis

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Methane emissions have increased in the past decade, with a substantial contribution from the agriculture, waste, and fossil fuel sectors. The dry reforming of methane (DRM) reaction converts one molecule of CH_4 and one of CO_2 into syngas, a value-added gaseous mixture of carbon monoxide and hydrogen [1]. Although the benefits of DRM are prominent and this process has been studied for decades, it still struggles to become a mature industrial technology, mainly because of its low energy efficiency and catalyst deactivation at high operating temperatures. Among non-thermal plasmas, the pulsed nanosecond discharge is gaining growing attention as one of the most energy-efficient ones to promote chemical reactions, taking advantage of the high electron densities and electron energies that can be reached out of thermal equilibrium.

In recent years, a developing field of interest has been the study of the effects caused by changes in the discharge's pulsing scheme. In the plasma-driven CO2 splitting, Montesano et al. showed that conversion and efficiency increased by shortening the time between successive discharges for the same total energy [2]. Below 100 µs, subsequent pulses do not act independently but occur in an environment perturbed by the initial pulse. The same phenomenon was then investigated for the DRM reaction [3]. At the maximum explored SEI (Specific Energy Input), around 6 kJ dm^{-3}, the CO_2 conversion doubled, and that of CH_4 increased by almost 50%, by shortening the pulse interval (Tp) from Tp > 833 µs to Tp < 40 µs. We suggest that the increased performance observed shortening the pulse separation is due to the progressive modification of the discharge conditions (gas composition, temperature, and load impedance). A "memory effect" is present when closer pulses are coupled in a post-discharge medium not fully relaxed to the initial conditions. The imaging of the discharge supported this finding. When the camera exposure time is large enough to catch three subsequent pulses, a difference in the discharge path is observed at varying inter-pulse times. They are spatially independent for inter-pulse times greater than 100 µs, while tending to 40 µs they converge to follow the same path.

An upgrade of the reactor configuration can be achieved by integrating a catalyst in the discharge reactor. Early results show an improvement in the DRM performance after introducing an alumina monolith cylinder coaxial to the discharge (Figure 1). Further investigations are ongoing.

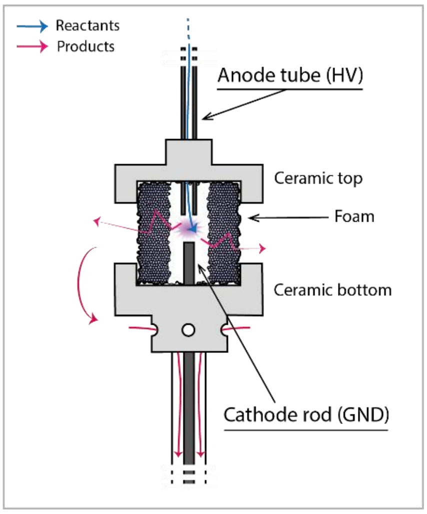

Fig. 1: *Configuration of the NRP discharge and monolith foam catalyst.*

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References

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