Experimental investigation of the complex chemistry in dry reforming microwave discharges

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In the endeavour to electrify the chemical industry, plasma based Dry Reforming of Methane (DRM) has been identified as a carbon neutral technology for syngas production. Traditionally syngas production is carbon intensive, as it is generated using steam methane reforming combined with the water gas shift reaction. In DRM it is attempted to couple carbon dioxide and methane directly into syngas, without the need for a subsequent gas separation step:

$$CO_2 + CH_4 \rightarrow 2\,CO + 2\,H_2\ \Delta H_r^{\ominus} = +247 \text{ kJ/mol} \quad (1)$$

Precisely the high endothermicity of this reaction makes plasma technology an ideal candidate to drive the process. While DRM in plasma is less well studied when compared to methane pyrolysis or carbon dioxide dissociation, especially warm plasma (i.e. microwave or arc discharges) have given promising results.

In this work we present an experimental investigation of DRM plasma in a vortex stabilised 2.45 GHz microwave discharge operating at 150 mbar and 1 kW power. Several diagnostics are set up, firstly effluent gas composition is measured using gas chromatography. Secondly, the plasma is characterised using optical emission spectroscopy that is both spatially and spectrally resolved. This is combined with axisymmetric tomography (the Abel inversion) to resolve local emissions. Lastly the core gas temperature is probed using a high resolution spectrometer to resolve the Doppler broadening of the atomic oxygen triplet at 777 nm.

The methane fraction is varied and the change in gas composition is seen to greatly influence the discharge. Upon increasing the methane fraction the emission profile is found to change: The discharge increases in size, atomic and chemiluminescent emissions shift in relative intensity, and Planckian radiation from solid carbon particles in the gas is seen for higher methane fraction.

The Spectral Soot Emission (SSE) technique [1] is applied on the 700-900 nm range to probe the carbon particle temperature and volume fraction based on the Planckian radiation and the assumption of Rayleigh-Debye-Gans scattering for fractal aggregates. The density of solid carbon particles in the discharge as determined from SSE is seen to increase with increasing methane fraction, eventually leading to carbon deposition on reactor walls and discharge instability.

The coupling of temperature profiles with local optical emission profiles greatly enhances our understanding of chemical and plasma processes in the discharge. It allows for a qualitative analysis of the highly complex chemistry found in DRM discharges.

[1] R. R. Snelling, *AIAA Journal* Vol. 40 No. 9 (2002) 1789–1795.