

Plasma-assisted CO₂ reduction in synthetic fuel processing

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Infrared laser absorption spectroscopy (IR-LAS) in conjunction with mass spectrometry was used to characterize the CO₂ reduction in expanding Ar/H₂ plasmas for synthetic fuel production. CO_x was injected into the expansion part where the reaction mechanism could be radical and/or ion driven. The results suggest a separation of dissociation and hydrogenation step. Further work on materials tailored for plasma-catalysis is required to increase the conversion efficiency.

1. Introduction

Plasma processing of carbon dioxide into conventional fuels is considered as an alternative approach. While reducing the green house gas emissions, these fuels can be integrated in the existing transport infrastructure. The formation of CO from CO₂ is thereby considered the rate limiting step [1] in further hydrocarbon or oxygenate forming reactions. This paper discusses conversion efficiencies in plasma-assisted CO₂ reduction, i.e. dissociation and hydrogenation in a low temperature plasma expansion. The potential of plasma catalysis in this context is assessed.

2. Experimental

CO₂ was injected into the background of an expanding thermal plasma created from a cascaded arc in mixtures of H₂ and Ar. The flow of CO₂ was maintained at about 10% of the total flows of 3360 sccm. The total pressure in the magnetized expansion part, where the electron temperature is usually clearly below 1 eV, was kept at 1 mbar.

The plasma composition was characterized at the farther end of the plasma expansion using infrared laser absorption spectroscopy (IR-LAS). Quadrupole mass spectrometry (QMS) was used as a complimentary technique for measuring components that cannot be accessed by IR-LAS. Apart from the native stainless steel, aluminium and copper foils were used in close contact to the vacuum vessel to evaluate the influence on the steady-state gas composition.

3. Results and Discussion

Three distinct regimes of chemistry can be observed depending on the gas composition through the arc (from pure Ar to pure H₂, fig. 1). In the first regime (I) charge exchange between CO₂ and Ar⁺ ions followed by dissociative recombination of CO₂⁺ - as observed for other molecular gases in this type of plasma - may be the dominant process. Ar⁺ is quenched quickly with addition of few percentage of

H₂ to the flow [2] and so are the O₂ production and the CO yield which can be as high as 50 % in pure Ar.

In regime II the CO₂ dissociation levels off and production of methane starts (fig 1). Main stable products apart from CO₂ are CO and H₂O (not shown in fig. 1). In regime III the chemistry is mainly driven by H radicals which leads to an increase in methane production. However, the CH₄ remains at a few %.

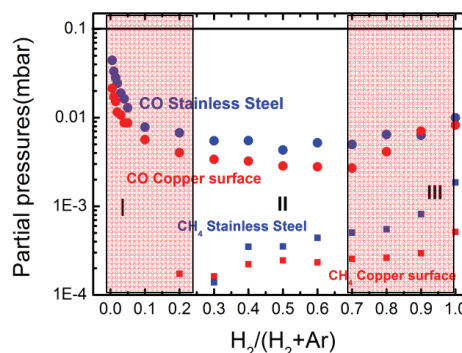


Fig. 1: CO and CH₄ produced during the CO₂ reduction in a plasma expansion of Ar/H₂ using different surface materials. Different regimes of chemistry are indicated (I - III).

Experiments performed with CO in place of CO₂ - where the conversion to methane starts at much lower additions of H₂ and may reach 25 % - confirmed that a syngas step (CO+H₂) is also essential in plasma-catalytic approaches. A difference in the gas composition was observed, when certain surface materials were used (e.g. Cu, fig. 1). However, in contrast to conventional catalysis the CO concentration was reduced and so was the CH₄ formation.

References

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- [2] R.F.G.Meulenbroeks et al *Physical Review E* 49, (1994) 4397-4406.