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## METHANE PARTIAL OXIDATION FOR FUTURE FUELS

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Partial oxidation of methane yields a very interesting spectrum of hydrocarbons. By converting methane into the most simple liquid oxygen-functional fuel i.e. methanol  $\text{CH}_3\text{OH}$ , the fuel energy density is already increased from 0.04 MJ/L to 18 MJ/L. Higher values are observed with longer chain hydrocarbons including oxygen functionality, also products of partial oxidation of methane.

A pulsed corona discharge reactor has been applied for studying the conversion of  $\text{CH}_4/\text{O}_2$  and  $\text{CH}_4/\text{CO}_2$  mixtures. Gas discharges are applied in a wire-to-water film capacitive discharge reactor with typical energy densities of 15-45 kJ/L, using a drift-step-recovery-diode based high voltage circuit [1]. Typical pulse parameters are peak voltage 65 kV, rise time 10 ns and repetition rates up to 65 Hz. The mainly applied diagnostic tool is a gas chromatograph-mass spectrometer with parallel flame ionization detector and oxygen-functional hydrocarbon separation column.

The chemistry of methane partial oxidation is explained via methyl and alkyl radical formation via reactive oxygen species, electron impact or vibrational excitation [2,3,4]. Polymerization of alkyl radicals is a major process. Molecular oxygen attachment to the methyl radical  $\text{CH}_3$  gives the peroxy radical  $\text{CH}_3\text{OO}$ , the precursor to the methoxy radical  $\text{CH}_3\text{O}$  and methanol.

Observed methane conversion levels are generally 6-20% at an efficiency of approximately 100-200 nmol/J. Observed products are mainly ethane  $\text{C}_2\text{H}_6$  (<17800 ppm), ethylene  $\text{C}_2\text{H}_4$  (<523 ppm) and acetylene  $\text{C}_2\text{H}_2$  (<1640 ppm). Methanol was detected at maximum 234 ppm level, while ethanol  $\text{C}_2\text{H}_5\text{OH}$  occurred at levels up to 57 ppm. Methanol seems exclusively produced with  $\text{CH}_4/\text{O}_2$  mixtures. Higher alkanes up to  $\text{C}_6$  have been especially observed with  $\text{CH}_4/\text{CO}_2$  mixtures: propane, butane, isobutene, pentane and 3-methylpentane. The same is true for the detected oxygen-functional hydrocarbons:  $\text{C}_2$ - $\text{C}_4$  aldehydes, acetone, 2-butanone, dimethylether and methylformate. Maximum observed selectivities are  $\text{C}_2\text{H}_6$ : 30.1%,  $\text{C}_2\text{H}_4$ : 1.1%,  $\text{C}_2\text{H}_2$ : 3.3%,  $\text{CH}_3\text{OH}$ : 0.2%,  $\text{C}_2\text{H}_5\text{OH}$ : 0.1%. Analysis of the aqueous phase has revealed that, together with a significant pH decrease, high molecular weight species may be formed.

- [1] A.J.M. Pemen, I.V. Grekhov, E.J.M. van Heesch, K. Yan, S.A. Nair, S.V. Korotkov, *Rev. Sci. Instr.*, 74(10): 4361-4365, 2003
- [2] K.V. Kozlov, P. Michel, H.-E. Wagner, *Plasma Polym.*, 5(3/4): 129-150, 2000
- [3] T. Nozaki, A. Hattori, K. Okazaki, *Cat. Today*, 98: 607-616, 2004
- [4] A. Fridman, *Plasma Chemistry*, Cambridge University Press, 2008