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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 CH_3OH , 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 CH_4/O_2 and CH_4/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 CH_3 gives the peroxy radical CH_3OO , the precursor to the methoxy radical CH_3O and methanol.

Observed methane conversion levels are generally 6-20% at an efficiency of approximately 100-200 nmol/J. Observed products are mainly ethane C_2H_6 (<17800 ppm), ethylene C_2H_4 (<523 ppm) and acetylene C_2H_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 CH_4/O_2 mixtures. Higher alkanes up to C_6 have been especially observed with CH_4/CO_2 mixtures: propane, butane, isobutene, pentane and 3-methylpentane. The same is true for the detected oxygen-functional hydrocarbons: C_2 - C_4 aldehydes, acetone, 2-butanone, dimethylether and methylformate. Maximum observed selectivities are C_2H_6 : 30.1%, C_2H_4 : 1.1%, C_2H_2 : 3.3%, CH_3OH : 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.

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