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Article

Kinetic contribution of CO_2/O_2 additive in methane conversion activated by non-equilibrium plasmas[#]

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Abstract A temperature-controlled and pressure-controlled coaxial dielectric barrier discharge (DBD) reactor was developed to decouple the thermal and kinetic effects of Radio Frequency (RF) discharge on methane conversion, and further to compare the kinetic behaviors of the mechanistically similar reactions of methane conversion with O_2 and CO_2 additives. A kinetic mechanism for RF plasma assisted methane conversion was assembled. The formation of products in the RF plasma reactor was measured with Gas Chromatography (GC-TCD) and the data were used to validate the kinetic model. The experimental and computational results showed the different kinetic roles of carbon dioxide and oxygen additives in methane conversion, due to the different dissociation and ionization energy of the two additive gases, as well as the thus produced electron energy distribution function (EEDF). Fuel oxidation by plasma generated O, O (^{1}D), $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g^+)$ and O^+ in partial oxidation of methane was observed essential for methane consumption, which resulted in an increase in methane conversion rate, compared to pure methane pyrolysis and dry reforming of methane with CO_2 additive. It was also found that dry reforming of methane with CO_2 was by far the easier to produce the syngas as well as C_2 hydrocarbon species, due to the weak oxidation ability of CO_2 and also the significant deposition of the electron energy on CH_4 dissociation in a dry reforming discharge mixture. This kinetic study produced comparative data to demonstrate the contribution of CO_2 /O_2 additive in non-equilibrium plasma assisted methane conversion.