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Computational and experimental analysis of H-atom-assisted non-thermal conversion of methane-hydrogen plasma to acetylene

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Methane (CH_4) is available in large quantities from natural gas. It is of practical importance for the synthesis of hydrocarbons such as acetylene (C_2H_2) and ethylene (C_2H_4) [1]. Traditional oxidative methods for methane conversion often result in undesired CO_2 formation [2]. Plasma pyrolysis, though effective in producing C_2H_2 at high temperatures (~3000 °C) [3], suffers from low energy efficiency and carbon deposition. Alternatively, non-equilibrium plasma reactors, utilizing microwave or RF discharges, enable vibrational excitation of CH_4, which promotes dissociation at lower temperatures and improved energy efficiency [1].

In this study [4], we explore carbon-neutral pathways for CH_4 to C_2H_2 conversion using a chemical dynamics approach. Reaction mechanisms were analyzed via quantum chemistry, comparing two distinct pathways - one in the presence of abundant H atoms (Fig. 1) and another under H-deficient conditions. DFT (B3LYP) calculations were employed to identify local minima and transition states. The pathway involving abundant H atoms was found to exhibit a more stable potential energy surface (PES) for C_2H_2 formation.

Furthermore, relative species concentrations were estimated using the Boltzmann distribution function [5], revealing that spontaneous CH_4 conversion and selective C_2H_2 production occur efficiently at lower energy ranges (< 14 eV) when H atoms are abundant (Fig 2). Surface wave plasma experiments with CH_4-H_2 mixed-gas were performed, where high specific energy input (SEI) is applied for enhanced C_2H_2 yields.

The computational findings qualitatively support the experimental results. The transient nature of plasma intermediates (10^{-9} – 10^{-6} s) poses challenges for direct validation. Further studies are ongoing to incorporate advanced kinetic modeling. Additionally, experimental determination of critical plasma parameters such as electron and gas temperatures would further strengthen the correlation between simulation and observation. This study contributes to the development of sustainable, low-energy routes for hydrocarbon synthesis via non-equilibrium plasma processes.

References

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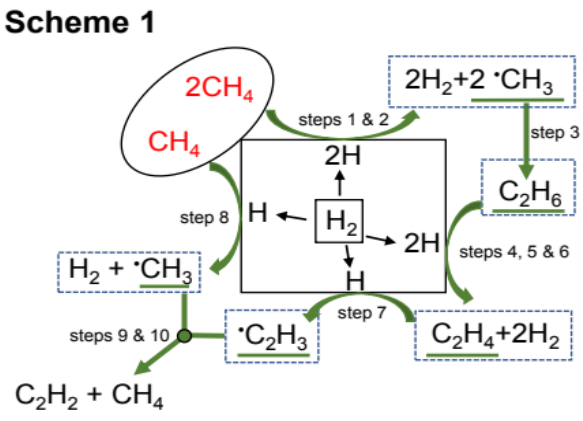

Figure 1. Scheme 1: CH_4 activation by an H atom

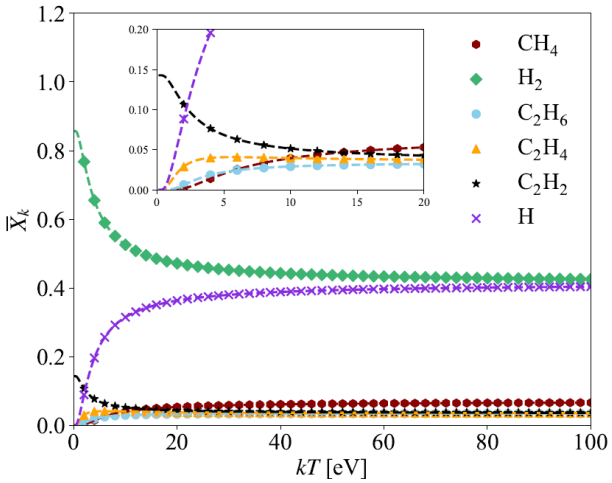

Figure 2. Thermodynamic energy dependency of the modified relative concentration of hydrocarbons formed in Scheme 1