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Methane as a feedstock in plasma-liquid processes

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Natural gas is one of the abundant fossil fuels, and probably the most flexible considering its relatively simple composition. The primary constituent is methane, and methane is used in several industrially crucial processes, such as steam-methane reforming and partial oxidation for the production of hydrogen and syngas, respectively. However, there are still plenty of other reactions that involve methane, which have not yet been established in any scale. The most important examples are dry reforming, oligomerization of methane, organic synthesis of cyanides and methanol synthesis. The common problem in these reactions is either the stability of the catalyst or low conversion levels.

While all the reactions mentioned above are generally studied in gas phase utilizing heterogeneous catalysts, methanol synthesis can be conducted catalyst-free in liquid phase, if a plasma is utilized. Since methane can easily be broken into methyl and hydrogen radicals by the free electrons of the plasma, both oligomerization and methane pyrolysis are feasible. In other words, the degree of methane dehydrogenation is expected to dictate the product spectrum in a methane plasma. In the presence of water and its vapor, methanol synthesis is expected to occur only if dehydrogenation is limited, which requires control over the electron population, electron energy distribution and residence time. Increasing the extent of dehydrogenation would favor carbon formation, whereas decreasing it is hypothesized to improve the yield of larger alcohols. Therefore, a combined approach of methane plasma kinetics and chemical reaction engineering is needed to tune the product spectrum.

This talk will focus on the use of plasma-liquid interaction for direct methane conversion into alcohols. We will first critically review previous work on the combination of water and methane in a plasma. Following that, a reaction kinetics model and computational fluid dynamics calculations will be presented with the aim of assessing the impact of plasma operation and residence time on the products. Results of the preliminary experiments obtained in a pin-to-plane setup consisting of pure methane and water will also be discussed.

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