**Plasma catalysis for CO2 conversion:**

**A better understanding of the underlying mechanisms**

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Plasma catalysis is gaining increasing interest for CO2 conversion. To improve this application in terms of conversion, energy efficiency and product formation, a good insight in the underlying mechanisms is desirable. We try to obtain this by computer modelling and experiments.

Experimentally, we study two types of plasma reactors, i.e., dielectric barrier discharges (DBDs) in which we insert packing materials to investigate plasma catalysis, as well as a gliding arc (GA) discharge. In this talk, I will show some results for both, illustrating the superior energy efficiency of a GA reactor.

In addition, we also simulate the plasma chemistry as well as the optimum reactor design, in the three types of plasma reactors most commonly used for CO2 conversion, i.e., DBDs, GA discharges and microwave (MW) plasmas. For the plasma reactor design, we use 2D or 3D computational fluid dynamics modelling. For the plasma chemistry, we make use of zero-dimensional chemical kinetics modeling, which solves continuity equations for the various plasma species, based on production and loss terms, as defined by the chemical reactions. Typically, up to 100 different species are considered, which react among each other in up to 1000 different chemical reactions.

When studying the plasma chemistry in pure CO2, we focus especially on the the role of vibrationally excited CO2 levels, which are the key species for enhanced energy efficiency of the CO2 conversion. Our model reveals the relative importance of various processes, responsible for the CO2 conversion, and this is linked to the energy efficiency in the various types of plasma reactors.

We have also studied the plasma chemistry in CO2/CH4 and in CO2/H2O mixtures, to produce value-added chemicals. The main products formed are a mixture of H2 and CO, or syngas, with a tuneable H2/CO ratio depending on the gas mixing ratio. The production of oxygenated compounds, such as methanol, formaldehyde, etc, is very limited, showing the need for combining with a catalyst. A detailed chemical kinetics analysis allows to elucidate the different pathways leading to the observed results, and to propose solutions on how to further improve the formation of value-added products.

Likewise, we also studied the plasma chemistry in a CO2/N2 mixture, to investigate the effect of this important impurity in effluent gases on the CO2 conversion, energy efficiency and product formation. Several harmful compounds, i.e., N2O and NOx compounds, are produced in the range of several 100 ppm. The reaction pathways for the formation of these compounds are again explained based on a kinetic analysis, which allows proposing solutions on how to prevent the formation of these harmful compounds.

Finally, we also study plasma-catalyst interactions, by experiments, atomic scale simulations (density functional theory), as well as fluid and particle-in-cell – Monte Carlo simulations. The latter is used to investigate whether plasma can be formed inside catalyst pores.