Abstract

CH_4 and CO_2 are the most impactful greenhouse gases. Their atmospheric concentration could be lowered via conversion of the gases collected at the emission site or captured from the atmosphere. The development of new technologies for such conversion with high energy efficiency and useful products is crucial. The PhD thesis is divided into three parts, each dedicated to the study of one such promising process: reverse water-gas shift reaction, dry reforming of methane and partial oxidation of methane.

In the reverse water-gas shift section, synthesis of five different copper-based catalysts with different supports was carried out. The catalysts were characterised with different analytical techniques and had their activity measured at different operating temperatures, pressure, gas flow rates and reagent ratios. Three different reactor mathematical models were developed, taking into account the mass transport as well as the catalytic surface reactions. New insights regarding mass transport were obtained, and numerical regression of the reaction constants was performed in order to increase the model accuracy. Methane dry reforming reaction was studied in a spark plasma reactor. Reaction kinetics were studied both in pure plasma and plasma-catalytic modes of operation, the latter utilising Ni-based structured alumina foam catalysts, under different operating conditions (temperatures, plasma powers, gas flow rates, reagent ratios) and special attention was paid to stability under coke-deposition conditions. A three-dimensional mathematical model was developed for this process using fluid dynamics. Methane partial oxidation was carried out with different reagent ratios and total gas flow rates. In addition, a multitude of different catalytic materials were coupled with plasma, such as pure zeolites and metal-based catalysts (Pd, Fe and Mo).

Keywords: dry reforming of methane, reverse water-gas shift reaction, partial oxidation of methane, plasma catalysis, mathematical modelling, reaction kinetics