Investigating the catalytic effect of the carbon product on enhancing hydrogen selectivity in molten salt methane pyrolysis

Rukshan Madhawa Jayathilake[1], Schalk Cloete[1], Stein Tore Johansen[1], Abdelghafour Zaabout[2], Jan Hendrik Cloete[1]

[1] *Department of Process Technology, SINTEF Industry, Trondheim, Norway*

[2] *Applied Chemistry and Engineering Research Centre of Excellence (ACER CoE), Mohammed VI Polytechnic University, Benguerir, Morocco*

*Corresponding author: henri.cloete@sintef.no (J.H. Cloete)

Methane pyrolysis is receiving widespread attention as a pathway for producing low-carbon hydrogen while simultaneously producing solid carbon as a valuable by-product. However, carbon separation is a major challenge when using conventional solid catalysts, motivating the development of processes in molten media, such as metals or salts. In this approach, the molten medium allows separation of the solid product due to the density difference between the carbon and the melt while acting as a heat transfer medium and catalyst.

The catalytic effect of molten $MnCl_2$ mixtures on methane pyrolysis at high temperatures is well-known. Nevertheless, the possible catalytic potential of carbon particles formed during the molten salt methane pyrolysis process to enhance methane conversion and hydrogen (H_2) selectivity has not been studied in detail. This study therefore investigates the catalytic effect of carbon particles on H_2 production at 900°C and lower during molten salt methane pyrolysis. Operating at these relatively low temperatures would help reduce challenges with salt evaporation and corrosivity, but, from literature, would also reduce methane conversion and hydrogen selectivity. It is therefore important to investigate strategies that can improve hydrogen production in this temperature range.

Experiments conducted by bubbling 0.1 Nl/min of methane through a bath containing 240 g of a 20/80 $MnCl_2$-NaCl salt mix indicate that initial hydrogen selectivity is relatively low, with mostly propane formed as a by-product. After a period of 1.5 hours, however, the H_2 selectivity reaches close to 100%, where it remains stable for the remainder of the experiment while other hydrocarbon byproducts remain at very low concentrations. This leads to the hypothesis that carbon formed during the methane pyrolysis process has an improved catalytic activity for methane conversion to hydrogen, and that the addition of carbon particles to the molten salt mix at the start of the experiments can enable high H_2 selectivity within a temperature range of 800-900°C. Literature supports the hypothesis that carbon particle formation is an important catalytic contributor in molten salt methane pyrolysis, but its effect on H_2 selectivity is yet to be quantified. The catalytic effect could also reduce the activation energy for methane decomposition, enhancing methane conversion at lower temperatures.

Ongoing experiments with carbon particle seeding will provide a better understanding of how to optimize the molten salt pyrolysis process, targeting high methane conversion and hydrogen selectivity at lower operational temperatures. This will reduce the technical barriers for further development and scale up of this technology, paving its way to faster commercialization.