Hydrogen production through chemical looping using NiO/NiAl$_2$O$_4$ as oxygen carrier

A new autothermal route to produce hydrogen from natural gas via chemical looping technology was investigated. Nowadays, 90% of the worldwide produced hydrogen comes from fossil fuels, the most common process being Steam Methane Reforming (SMR), which produces several kg of CO$_2$ equivalent per kg of hydrogen. In classic SMR technology, most of the greenhouse gases are vented to the atmosphere. Chemical Looping Combustion (CLC) is a promising concept based on circulating fluidized beds to combust solid, liquid and gaseous fuels with inherent CO$_2$ capture and minimal NOx formation. Chemical Looping Reforming (CLR) utilizes similar principles as CLC but the objective of CLR is hydrogen, as opposed to energy for CLC.

Tests were conducted in a micro-fixed bed reactor loaded with 200mg of NiO/NiAl$_2$O$_4$ as oxygen carrier. A mass spectrometer (Hiden Analytical) was coupled directly to the reactor exhaust. It was set to scan molecular masses of 2, 16, 18, 28, 32, 40 and 44 for hydrogen, methane, water, carbon monoxide, oxygen, argon and carbon dioxide respectively. The high sampling frequency of the outlet gases by the MS makes the data analysis very precise compared to classic laboratory tools typically used (i.e. reactor + GC or TGA). Methane reacts with a nickel oxide in the absence of molecular oxygen at 800°C for a period of time as high as ten minutes. The NiO is subsequently contacted with a synthetic air stream (21% O$_2$ in argon) to reconstitute the surface and combust carbon deposited on the surface. Methane conversion nears completion but to minimize combustion of the hydrogen produced, the oxidation state of the carrier was maintained below 30% (where 100% represents a fully oxidized surface). Co-feeding water together with methane resulted in stable hydrogen production. Although the carbon deposition increased with time during the reduction cycle, the production rate of hydrogen remained virtually constant. A new concept is also presented where hydrogen is obtained from methane with inherent CO$_2$ capture in an energy neutral 3-reactors CFB process. This process combines a methane combustion step where oxygen is provided via an oxygen carrier, a steam methane reforming step catalyzed by the reduced oxygen carrier and an oxidizing step where the O-carrier is reconstituted to its original state.
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