Abstract

The use of natural gas hydrocarbons has been explored in the steam reforming of methane and ethane for the production of syngas and hydrogen via a plug-flow, diffusion-free model based on nickel-oxide catalysts supported over alumina (NiO/Al_2O_3). An industry-scale circulating fast fluidized bed membrane reactor (CFFBMR) was designed for the production of syngas and hydrogen while considering the reaction-controlled intrinsic kinetics of steam reforming of a feed mixture of 90 mol% methane and 10 mol% ethane. The CFFBMR design consisted of Ni-based catalytic particles, hydrogen-permselective membrane tubes, cyclones (G/S separator), and catalyst regenerators to produce a high yield of hydrogen and a low yield of carbon dioxide at a full-scale feed capacity of 32.5 MMcfd of methane and ethane. In the study of the fluidized reactor for the production of hydrogen and syngas, improvements in the process design of catalyst recirculation, gas-solid separation, and hydrogen permeation were also examined.

Although the carbon dioxide yield decreased steeply after 20% of the reactor length, 32.8 m, the hydrogen yield stabilized within 10% of the reactor length. Thus with the use of both hydrogen permeation and carbon dioxide recycle, the reactor length should not be designed too short because otherwise too much carbon dioxide would be formed, or too small of a quantity of CO_2 would be recycled. Likewise, the reactor length should not be designed with too long of a length because the hydrogen yield did not significantly improve after 20% of the reactor length. Therefore, 50% of the original reactor length, or 16.4 meters, would be an optimal length to achieve a balanced outcome between a low formation of CO_2 and a reasonably high yield of H_2. Additionally, 50% of the reactor length would produce significant savings in the capital costs of the reactor

system. Between methane-ethane steam reforming (MESR) and heptane-methane steam reforming (HMSR), higher values in the hydrogen-to-carbon monoxide ratio were found in MESR. In MESR, the H_2/CO ratio was stabilized at a value near 2.5 for the two MESR membrane reactors, CFFBMR without CO_2 recycle and CFFBMR with CO_2 recycle, while the H_2/CO ratio was stabilized at a value near 4.5 for MESR circulating fast-fluidized bed reactor without hydrogen permeation (CFFBR). On the other hand in HMSR, the H_2/CO ratio was stabilized at a value near 1.25 for the two HMSR membrane reactors and at a value near 2.25 for HMSR CFFBR. Since syngas can be used for gas-to-liquids (GTL) processes when the ratio is between 1.0 and 3.0, hydrogen permeation would be ultimately necessary for methane and ethane steam reforming to avoid a H_2/CO ratio higher than 3.0.