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Fischer Tropsch Synthesis in Compressed Carbon Dioxide with Integrated Recycle

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Abstract : Fischer-Tropsch (FT) synthesis is a complex series of heterogeneous reactions between CO and H2 molecules (present in the syngas) on the surface of an active catalyst (Co, Fe, Ru, Ni, etc.) to produce gaseous, liquid, and waxy hydrocarbons. This product is composed of paraffins, olefins, and oxygenated compounds. The key challenge in applying the Fischer-Tropsch process to produce transportation fuels is to make the capital and production costs economically feasible relative to the comparative cost of existing petroleum resources. To meet this challenge, it is imperative to enhance the CO conversion while maximizing carbon selectivity towards the desired liquid hydrocarbon ranges (i.e. reduction in CH4 and CO2 selectivities) at high throughputs. At the same time, it is equally essential to increase the catalyst robustness and longevity without sacrificing catalyst activity. This paper focuses on process development to achieve the above. The paper describes the influence of operating parameters on Fischer Tropsch synthesis (FTS) from coal derived syngas in supercritical carbon dioxide (ScCO2). In addition, the unreacted gas and solvent recycle was incorporated and the effect of unreacted feed recycle was evaluated. It was expected that with the recycle, the feed rate can be increased. The increase in conversion and liquid selectivity accompanied by the production of narrower carbon number distribution in the product suggest that higher flow rates can and should be used when incorporating exit gas recycle. It was observed that this process was capable of enhancing the hydrocarbon selectivity (nearly 98 % CO conversion), reducing improving the carbon efficiency from 17 % to 51 % in a once through process and further converting 16 % CO2 to liquid with integrated recycle of the product gas stream and increasing the life of the catalyst. Catalyst robustness enhancement has been attributed to the absorption of heat of reaction by the compressed CO2 which reduced the formation of hotspots and the dissolution of waxes by the CO2 solvent which reduced the blinding of active sites. In addition, the recycling the product gas stream reduced the reactor footprint to one-fourth of the once through size and product fractionation utilizing the solvent effects of supercritical CO2 were realized. In addition to the negative CO2 selectivities, methane production was also inhibited and was limited to less than 1.5%. The effect of the process conditions on the life of the catalysts will also be presented. Fe based catalysts are known to have a high proclivity for producing CO2 during FTS. The data of the product spectrum and selectivity on Co and Fe-Co based catalysts as well as those obtained from commercial sources will also be presented. The measurable decision criteria were the increase in CO conversion at H2:CO ratio of 1:1 (as commonly found in coal gasification product stream) in supercritical phase as compared to gas phase reaction, decrease in CO2 and CH4 selectivity, overall liquid product distribution, and finally an increase in the life of the catalysts.

Keywords : carbon efficiency, Fischer Tropsch synthesis, low GHG, pressure tunable fractionation
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