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An experimental and thermodynamic study of iron catalyst activation and deactivation during Fischer Tropsch synthesis

Gorimbo, Joshua

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ABSTRACT

One gram amounts of a commercial iron based catalyst were loaded into three reactors and reduced with syngas, hydrogen and carbon monoxide respectively. Fischer Tropsch experiments on the three reactors in parallel with the same operating conditions, namely 60 mL(NTP)/min, 1 bar gauge and 250 °C, were then conducted for extended periods and the gaseous products analysed.

Initially (for about 150 hours) the three catalysts had quite different carbon monoxide conversions. After this until about 1000 hours the conversions were similar. However the distribution of products for the differently reduced catalyst was significantly different. This suggested that permanent changes had been done to the catalysts by the different reducing conditions.

To try to understand what the differences during the reduction process might be, a thermodynamic analysis of the solid phases after reduction was done. Unfortunately because all the thermodynamic data for the possible carbides was not available this analysis was of limited value. However it did suggest that hydrogen reduced catalyst might contain more oxides and the carbon monoxide reduced catalyst might contain more carbides. Some electron microscope and XRD experiments supported these ideas and might account for the different selectivities of the differently reduced catalysts.

Runs after about 5000 hours were done at different flowrates (60, 30 and 15 mL(NTP)/min) of syngas and again the big effects were on differences between the selectivities, the big effects being when going to the lowest flowrate.

After about 12000 hours regeneration of the catalysts was then done by oxidation and then the same syngas reduction on all the catalysts. Runs were then done at different pressures (1, 10 and 20 bar gauge) and again selectivities were the biggest effects that remained, clearly showing the initial reduction had made permanent changes.

In the final section some novel plots were used to try to make more sense of the results. It was shown that for all the catalysts the Olefin to Paraffin ratios were tied to each other under all conditions and that they were mainly a function of the conversions with much higher values at low conversions.