

1.0 INTRODUCTION

The future use of coal as a source of conventional transportation fuel will depend on the development of an economical and energy efficient liquefaction process. Technologies that have been commercially proven or that are close to commercialization include the fixed- and fluidized-bed FT synthesis, methanol synthesis (fixed-bed and slurry-phase) and the Mobil methanol-to-gasoline process. Of these technologies, the Fischer-Tropsch hydrocarbon synthesis produces the widest slate of products and has been in operation for the longest period.

The FT reaction, in which carbon monoxide is reduced by hydrogen and polymerized, produces hydrocarbons with a broad range of molecular weights, from methane to paraffin waxes. The distribution of products can be accounted for by using general polymerization kinetics in which there is a constant ratio of propagation and termination rates. These kinetics were derived independently by Schulz and Flory in studies unrelated to the FT reaction. Nearly all FT catalysts give product distributions that adhere to these polymerization kinetics and are thus called Schulz-Flory distributions.

This broad product distribution significantly limits the maximum yield of transportation fuel fraction and creates the need for further downstream processing such as hydrocracking and light olefin oligomerization to increase such yields. Research has shown that

selectivity is controlled mainly by catalyst composition and process conditions. In order to improve the economics of the FT process, a catalyst should display high activity and minimize the formation of both light hydrocarbons (C_1-C_4) and waxes (C_{24+}), while producing the bulk of the product fraction in either the gasoline (C_5-C_{11}) or diesel ($C_{12}-C_{18}$) ranges.

Product selectivity, however, has been more successfully controlled by manipulating process conditions. Since the FT reaction is exothermic, control of the reaction heat plays a major role in controlling product selectivity. The slurry-phase process offers the best means of heat transfer and temperature control and has been shown to improve liquid product selectivity mainly by lowering the light gas yield (1-4).

Under an earlier contract with the Department of Energy, Air Products and Chemicals, Inc. developed several new slurry-phase FT catalysts that enhanced selectivity to liquid fuel products (5a-c). One group of catalysts included supported metal carbonyl clusters modified by promoters. To further develop and improve these catalyst systems, Air Products, under the current DOE contract, conducted another program to thoroughly investigate metal carbonyl cluster-based catalysts for use in slurry phase FT technology. This final report covers the programs' accomplishments for DOE contract no. DE-AC22-84PC70030.