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Selection, Design and Scale Up of the Fischer-Tropsch Reactor

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1. INTRODUCTION

It is expected that the demand for gasoil and kerosene (middle distillates) will grow in the near future, especially in the Asian region [1]. Middle distillates can be distilled directly from crude oil, but can also be produced by converting coal or natural gas using the Fischer-Tropsch reaction. An advantage of using natural gas is that it is relative abundant and can be converted in excellent quality (clean) middle distillates. From an economic point of view, potential processes need to be operated on a large scale [2]. Besides this, most of the large natural gas reserves are located in remote areas. These considerations make development and scale up of potential processes both a difficult and challenging task. Shell (SMDS process [3]) and Sasol [4] are currently the only companies having a Fischer-Tropsch natural gas to middle distillates process in commercial operation. Several other major enterprises such as Exxon [2] are currently developing improved processes for conversion of natural gas to liquid products.

A schematic flow diagram of these processes is shown in Figure 1. Natural gas is converted into synthesis gas in the first process step. In the second step syngas is converted into long chain hydrocarbons. Selective hydrocracking and hydroisomerisation of these long chain hydrocarbons yields marketable middle distillates.

One of the most important subjects in the development of the Fischer-Tropsch process is the selection, design and scale-up of the reactor for the heavy paraffin synthesis. The synthesis can be carried out in both trickle bed reactors and slurry bubble columns (Figure 1). Slurry bubble columns can be operated in two flow regimes: the homogeneous flow regime and the churn turbulent flow regime. In this study the Fischer-Tropsch slurry reactor is simulated and optimised. A comparison is made between operation in the homogeneous flow regime and churn turbulent operation. The performance of the slurry reactor is compared with simulated performance of the Fischer-Tropsch trickle bed reactor.

This study differs from others on reactor selection for the Fischer-Tropsch process [5], [6] in that recently obtained hydrodynamic details of the slurry reactor [7] are incorporated into the simulations, which are used to arrive at the optimal reactor configuration.

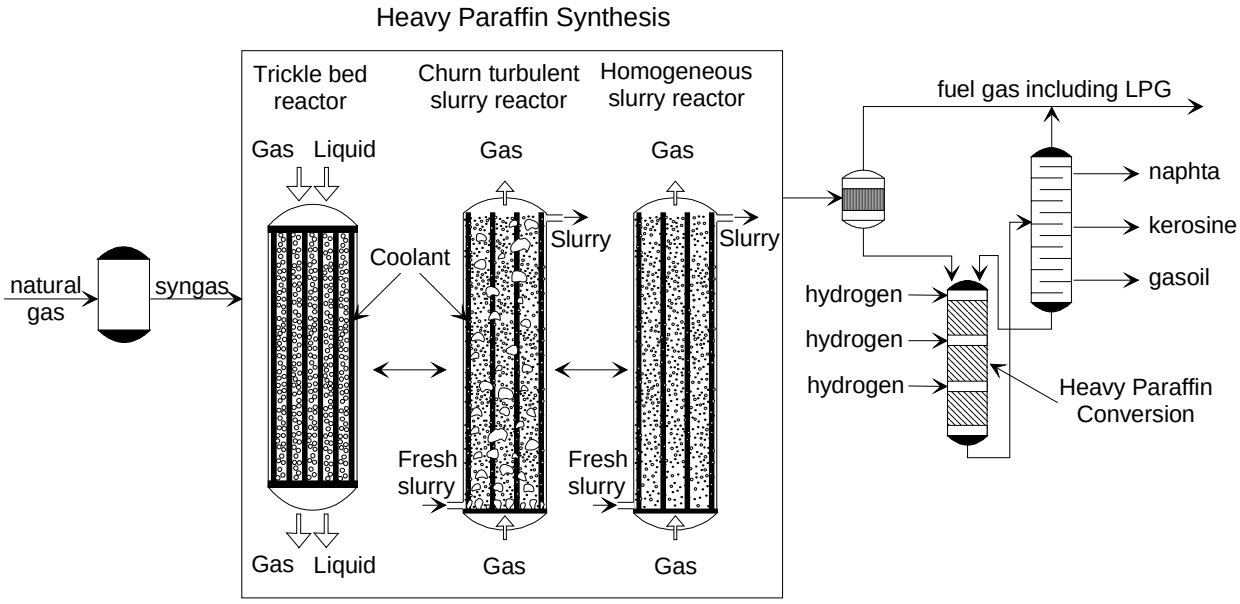

Figure 1. Schematic representation of the Fischer-Tropsch process for converting natural gas into middle distillates

2. REACTOR MODELLING

A complete 5000 tonne middle distillates/day Fischer-Tropsch plant is designed to arrive at the 'best' reactor configuration (middle distillates are defined as molecules with carbon number between 11 and 30). The heavy paraffin synthesis step is modelled using a Cobalt catalyst (probability for chain growth α equals 0.92). The rate of the water gas shift reaction is assumed to be negligible. The reaction is modelled as first order in hydrogen and kinetics are taken from [8]. The reactors operate at a pressure of 40 bar and a temperature of 510 K. The usage ratio of hydrogen and carbon monoxide is assumed to be equal to two. Gas phase contraction is taken into account by application of a gas phase contraction factor [9] which is assumed to be equal to -0.5. A particle diameter of 50 μm is used in the slurry reactor at concentrations up to 25 vol% (volume % catalyst in gas free slurry). In the trickle bed reactor bigger particles are employed to avoid excessive pressure drops, d_p = 2 mm, and the bed porosity equals 0.45. To avoid an (expensive) recycle of unconverted syngas, reactor configurations are designed to achieve an overall syngas conversion higher than 90%.

As already mentioned in the introduction, economy of scale is important and reactor sizes must be as big as possible. The maximum size of the reactors is assumed to be determined by the possibility to transport the reactors and install them at the remote locations where the plant is to be erected. This indicates that reactor weight is a limiting factor in scaling up the reactors. In this study a maximum reactor weight of 900 tonnes is adopted for both the slurry reactors and the trickle bed reactors.

Simulation of multiphase Fischer-Tropsch reactors involves a lot of physical properties, parameters and relations. Due to the limited space, they can not all be published in this paper and reference is made to [10].

3. HYDRODYNAMICS

3.1 Slurry reactor operating in the homogeneous bubbly flow regime

The hydrodynamic model used for simulation of a slurry bubble column operating in the homogeneous flow regime is pictured in Figure 2a. In a slurry bubble column, the homogeneous flow regime prevails at relative low gas velocities. Synthesis gas is distributed into the reactor at the bottom. As the gas rises through the suspension of catalyst and liquid products, hydrogen and carbon monoxide are dissolving into the liquid phase followed by reaction on the catalyst surface. In the homogeneous flow regime the gas phase consists of small bubbles, typically smaller than 5 mm. These small bubbles all have approximately the same diameter and rise velocity and are modelled with a plug flow equation. The suspension of liquid products and catalyst is modelled as being completely mixed.

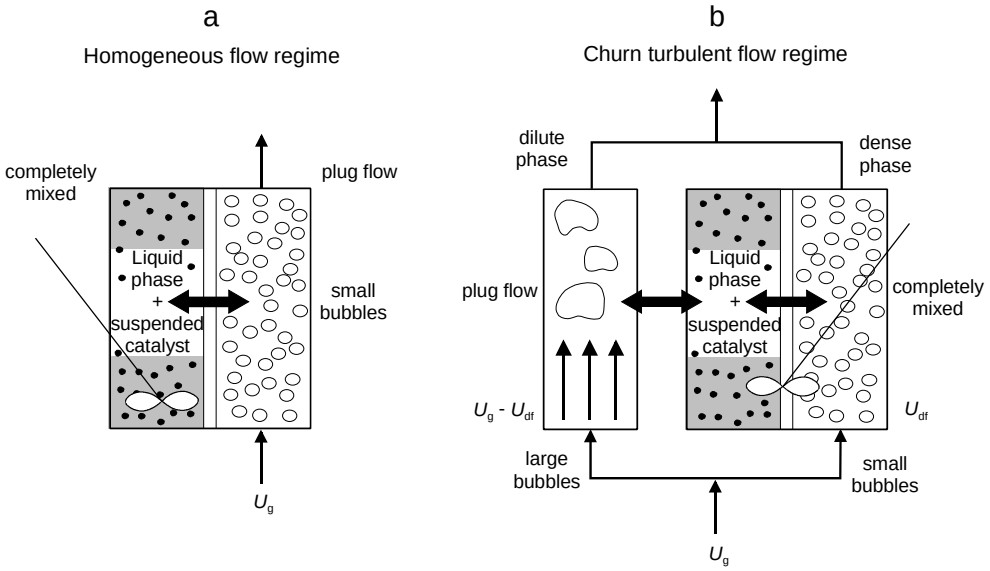

Figure 2. Hydrodynamic models for slurry bubble columns operating in the homogeneous flow regime (a) and churn turbulent flow regime (b)

Liquid products and catalyst are withdrawn from the reactor at the top. Concentrated fresh catalyst enters the reactor at the bottom, so the reactor operates in a co-current mode with respect to slurry and gas. Due to the high degree of backmixing in the slurry phase countercurrent operation does not offers much advantage. Co-current operation however avoids catalyst settling tendencies, which could result in serious heat transfer problems (hot spots). Heat removal is by means of cooling tubes installed in the reactor.

3.2 Slurry reactor operating in the churn-turbulent flow regime

The churn turbulent flow regime prevails in slurry bubble columns operating at higher gas velocities. The model used for simulation of a slurry bubble column operating in the churn turbulent flow regime is based on [11] and is schematically represented in Figure 2b.

The gas entering the reactor at the bottom is subdivided in two parts: part of the gas rises through the reactor in the form of large bubbles; the rest rises through the reactor in the form of small bubbles. The large bubbles rise with a high velocity through the slurry (their rise

velocity can easily exceed 1 m/s) and are modelled with a plug flow equation. Due to the high rise velocity, large bubbles induce circulation patterns in the suspension of liquid and catalyst. These circulation patterns severely mix up the small bubble population. Due to their high degree of mixing, the small bubbles and (liquid + catalyst) are modelled as completely mixed. Catalyst feed, withdrawal of liquid products and reactor cooling are the same as in the homogeneous case.

3.3 Trickle bed reactor

The trickle bed reactor is operated in co-current mode with respect to gas and liquid. Synthesis gas and recycled liquid products are fed at the top of the reactor and flow downwards over the catalyst. Plug flow is assumed for the gas phase. Inter- and intra-particle diffusion resistances along with the intrinsic reaction kinetics are used to calculate the overall reaction rates.

4. RESULTS AND DISCUSSION

4.1 Homogeneous slurry reactor

The optimal homogeneous slurry reactor configuration for a Fischer-Tropsch plant producing 5000 tonnes middle distillates per day operates at a superficial gas velocity of 0.01 m/s, just below the regime transition velocity. The superficial gas velocity at which regime transition to the churn turbulent flow regime occurs decreases strongly with increasing catalyst concentrations in the slurry [11]. For the Fischer-Tropsch process, where high catalyst concentration favours reactor performance, this means that the operating window for operation in the homogeneous flow regime will be very narrow. Using 25 vol % slurry concentration a conversion of 91% is achieved with a 7 m dispersion height. The maximum allowable diameter for a reactor to weigh 900 tonnes was found to be 11 m. Seventeen 900 tonnes homogeneous slurry bubble columns are needed to produce 5000 tonnes middle distillates per day; see Figure 3.

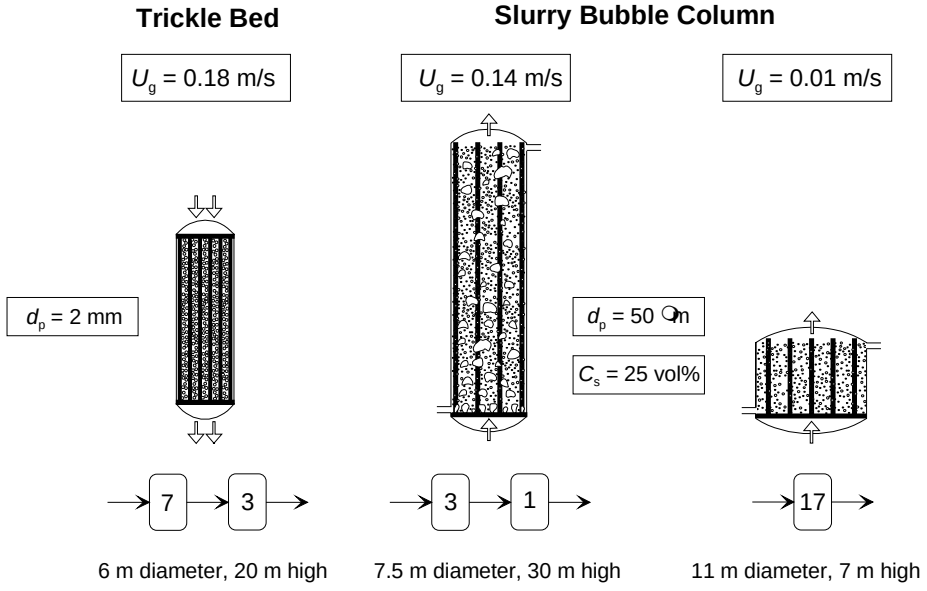

Figure 3. Representation of the results of the simulations

4.2 Churn turbulent slurry bubble column reactor

If a slurry bubble column is operated at a superficial gas velocity higher than the transition velocity the churn turbulent flow regime prevails. To obtain a conversion of 91% it was necessary to employ 30 m tall reactors and place them in two stages. The optimum superficial gas velocity for the Fischer-Tropsch slurry reactor operating in the churn turbulent regime was found to be 0.14 m/s for the first stage. The maximum allowable diameter for the reactors to weigh 900 tonnes was found to be 7.8 m. To produce 5000 tonne middle distillates per day three reactors are placed in parallel followed by one in series; see Figure 3 and Table 1.

4.3 Trickle bed reactor

For the Fischer-Tropsch trickle bed reactor an extra constraint had to be incorporated into the design. Because of the much lower heat transfer coefficient (see Table 1), higher catalyst concentration and the plug flow nature of gas and liquid, heat removal in a trickle bed is much more of a problem than in a slurry bubble column. Non-isothermal operation causes a temperature peak (hot spot) near the entrance of the reactor. Selectivity of the reaction is decreased and catalyst activity can be destroyed. A maximal temperature rise of 15 K was allowed in the design. It was found that the optimal trickle bed configuration again consists of 2 stages. Seven 800 tonnes (diameter = 6.2 m, height = 20 m) reactors are placed in the first stage, followed by three in series; see Figure 3 and Table 1.

5. CONCLUSIONS

In Table 1 the main results are summarised. First the total reactor volumes required to produce 5000 tonne middle distillates per day are compared. Churn turbulent slurry operation is much more attractive than homogeneous operation (less reactor volume is required). Reactor volumes for trickle bed and churn turbulent slurry operation are almost equal: approximately 5700 m^3. The active reactor volume (total volume minus volume of cooling tubes) is smaller for the trickle beds, due to a higher catalyst concentration. However, the volume occupied by the cooling system is larger for the trickle beds. A lower heat transfer coefficient and the plug flow nature of gas and liquid phase result in more cooling requirements. Overall, in the light of total reactor volume, the lower catalyst concentration in the slurry reactors cancel against the higher cooling requirements of the trickle beds.

With the assumption of reactors weighing 900 tonnes, slurry reactor operation in the churn turbulent flow regime is the most attractive option. Four reactors are required against 10 trickle beds and 17 homogeneous slurry reactors. Although the total reactor volumes of churn turbulent slurry and trickle bed are almost equal, trickle bed operation requires 6 more 900 tonnes reactors. This is due to the higher cooling requirements of the trickle beds, which results in much more (heavy) cooling tubes.

As said before, the catalyst concentration is an important parameter in the performance of Fischer-Tropsch reactors. The slurry reactors are designed with a 25 vol% concentration catalyst in the liquid phase. The maximal allowable concentration will be dictated by the handlability of the slurry phase. This will, amongst other things, depend on catalyst morphology (particle size, particle size distribution, pore volume). If catalysts are developed which can be allowed to have a higher concentration than 25 vol%, the slurry reactor will perform even better. In the future, new catalyst developments can result in more active catalysts. Due to their heat transfer limitations, trickle bed reactor will not profit much of this. If catalyst stability needs to be considered [6], the slurry reactor offers greater flexibility. Catalyst can be replaced during operation, whereas trickle beds need a considerable downtime

to replace the catalyst. Product selectivity is very important for the Fischer-Tropsch process. Since non-isothermal operation will decrease product selectivity, the isothermal slurry reactor will be more attractive than the non-isothermal trickle bed.

	Trickle Bed		**Slurry Bubble Column**		
			Homogeneous	**Churn Turbulent**	**Churn Turbulent**
	parallel	serial		parallel	serial
Number	7	3	17	3	1
D_t /[m]	6.2	5.8	11	7.8	7.6
***H*/[m]**	20	20	7	30	30
N_{tubes} per reactor	8000	4900	2600	2000	1000
Heat transfer coefficient/ [W/m^2/K]	262	191	853	1435	1435
U_g /[m/s]	0.18	0.23	0.01	0.14	0.27
Hydrogen Conversion	0.73	0.66	0.92	0.78	0.61
Weight/[tonne]	865	766	833	896	756
Productivity/[tonne middle distillates /day]	570	332	293	1415	747
Overall Conversion	0.91		0.92	0.91	
Total volume/[m^3]	5717		10912	5657	
Total active volume/[m^3]	2941		10304	5302	
Overall productivity/[tonne middle distillates /day]	4983		4983	4992	

Table 1. Results of the reactor simulations

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