The Application of Dielectric Barrier Discharge Plasma on Fischer-Tropsch Synthesis: A Review

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Abstract:

Fischer-Tropsch (FT) Synthesis has been widely known for centuries as the process of converting syngas to liquid fuels. Several reactors including Slurry bubble column, fluidized-bed, and fixed bed reactors have been used for FTS on an industrial scale. Although science has seen remarkable development in technology for FT synthesis, the industry still faces challenges in optimizations of process parameters and achieved desired selectivity. Extensive research has been continuously conducted to seek the best FT reactor offering heat uniformity and efficient heat transfer across the reactor to increase the catalytic activity and its lifetime. Dielectric Barrier Discharge (DBD) plasma has become one of the options to deal with these issues. This reactor work under low temperature delivers a synergistic effect between plasma and catalyst to break H₂ and CO bond. DBD plasma is also suitable for feedstock with high H₂/CO molar ratios. It is also found that FT catalyst such as cobalt catalyst used in DBD plasma was well dispersed on the support which in turn favour the selectivity toward liquid hydrocarbon.

Keywords: *syngas, GTL, plasma reactor, cobalt catalyst, non-thermal plasma*

Abstrak:

Fischer-Tropsch Sintesis (FTS) telah dikenal selama berabad lamanya sebagai proses untuk mengubah syngas menjadi bahan bakar cair. Beberapa reaktor seperti Slurry bubble column, fluidized-bed, dan fixed bed reactors telah digunakan untuk FTS dalam skala industri. Meskipun ilmu pengetahuan telah mengalami perkembangan yang luar biasa dalam teknologi FTS, tetapi dunia industri masih menghadapi banyak tantangan dalam mengoptimasi parameter proses dan mencapai selektivitas yang diinginkan. Riset ekstensif secara kontinu dilakukan untuk mencari reaktor terbaik yang memiliki keseragaman panas dan perpindahan panas yang efisien di keseluruhan reaktor sehingga dapat meningkatkan aktivitas katalis dan umur katalis. Dielectric Barrier Discharge (DBD) plasma telah menjadi salah satu opsi untuk mengatasi isu-isu tersebut. Reaktor ini bekerja pada temperature rendah menghasilkan efek sinergitas antara plasma dan katalis untuk memutus ikatan H₂ dan CO. DBD plasma juga cocok untuk bahan baku dengan H₂/CO molar ratios yang tinggi. Ditemukan juga bahwa katalis FT seperti cobalt yang digunakan dalam reaktor ini terdispersi dengan baik diatas katalis support sehingga meningkatkan selektivitas terhadap hydrocarbon cair.

Keywords: *syngas, GTL, reaktor plasma, katalis cobalt, non-thermal plasma*

I. Introduction

Fischer-Tropsch (FT) Technology Development

The utilization of syngas for liquid fuels production via the renowned process called Fischer-Tropsch (FT) synthesis has been widely known. FT synthesis is a process which converts synthesis gas (syngas) including gas hydrogen and carbon monoxide derived from various feedstock to hydrocarbon products. Most of the process employs catalyst cobalt and iron. Interest in FT technology is due to recent development in technology to use syngas from stranded natural gas and remotely located natural gas. This source of syngas is still able to produce liquid hydrocarbon products for commercial purpose. This is often referred to the Gas-to-Liquids (GtL) industry [1]

The first feedstock used in FT synthesis was coal in 1923. The high-pressure process employed cobalt, iron and ruthenium catalyst. In 1936, Fischer and Pilcher were able to run the process at medium pressure. Much improved GtL technology came in the 1970s as the world saw a significant increase in oil price. Sasol built two plants in 1980 and 1982 at Secunda named Sasol II and III which had a total capacity of 150,000 barrels per day [1]. Stranded gas reserves had help promoting GtL technologies in this era. The Moss gas project then established the Shell plant at Bintulu, Malaysia in 1993.

CtL and GtL have recognized as established technology throughout the world. Later, BtL processes begun to grow which is initiated by environmental awareness on emissions from the transport sector.

Germany is the front lead in the development of FT technology. Choren Industries has built several BtL plant in Germany based on Shell technology [2]

Figure 1. Source of synthesis gas [3]

The need for FT products may remain since the price of oil and gas fluctuate, **Figure 1** describes that the FT process can be derived from the various feedstock. To date, [3] state that the use of FT reactors commercially is limited to slurry reactors and fixed bed. Several technologies have been considered to come into practise such as plasma technology. The author will discuss this technology in the later section.

Basic Characteristic of FT Synthesis

The basic FT synthesis reaction produces a mixture of hydrocarbons from hydrogen and carbon monoxide as follow:

The product mixture including hydrocarbon and alcohol is affected by process conditions (pressure and temperature), syngas composition and catalyst, [4]. Based on operating temperature, FT synthesis can be categorized into two groups, namely high-temperature FT (HTFT) and low-temperature FT (LTFT). HTFT operates at a temperature range of 300 – 350°C and pressure of 20-40 bars whereas LTFT runs at a temperature range of 200–250°C and a pressure range of 20-45 bars. LTFT favours the formation of long chain hydrocarbon whilst the HTFT process is used for producing light olefin products [1].

Plasma Technology

Plasma can be defined as ionized gases which consist of positive (and negative) ions and electrons, as well as neutral species. Plasma is produced by applying energy to a gas to organize the electronic structure of the species (atoms, molecules) and to produce excited species and ions. This energy can be thermal or carried by either an electric current or electromagnetic radiation. The ionization degree can vary from 100% (fully ionized gases) to very low values e.g. 10^{-4} – 10^{-6} (partially ionized gases) [5]. The atmospheric plasma can be categorised into two types: thermal plasma and non-thermal plasma according to gas temperatures. For thermal plasma, of which gas temperature varies from 1,000 to 10,000 K e.g.

For non-thermal plasma, such as dielectric barrier discharge (DBD), all discharged species are in a thermally non-equilibrium state. The electron temperature is in a range of 10,000–100,000 K, equivalent to 1–10 eV, which is much higher than the gas temperature at ~100 K [6]. Instead of providing energy to the system, the non-thermal plasma generates radicals and excited species, which enable to initiate and enhance the chemical reactions [7]. The benefit of the non-thermal plasma is lower operation temperatures, leading to

lower energy consumption and lower electrode erosion because cooling process of the electrodes is no longer required [8].

This paper will discuss the challenge of the existing FT Synthesis process and how application of DBD plasma may overcome the limitation. The review will also outline parameter affecting FTS including H_2/CO , temperature, pressure.

II. Discussion

Process Condition Affecting FT Synthesis

Table 1. Selectivity control in FT synthesis by process conditions [9]

Parameter	Chain length	Olefin selectivity	Alcohol selectivity	Carbon deposition	Methane selectivity
Temperature	↓	*	↓	↑	↑
Pressure	↑	*	↑	*	↓
H_2/CO	↓	↓	↓	↓	↑
Conversion	*	↓	↓	↑	↑
Residence time	*	↑	↑	*	↓

Note: Increase with increasing parameter: ↑. Decrease with increasing parameter: ↓. Complex relation: *

Table 1 describes how process conditions affecting the selectivity in conventional FT synthesis. For instance, to increase the selectivity towards C_{5+} , temperature and H_2/CO ratio should be set at low conditions while pressure is set at high levels recommended H_2/CO ratio and pressure in industrial practice is 1.7-2.54 and 23-25 bar, respectively [4]. However, this does not necessarily mean high conversion is to be achieved. Optimum conditions should be studied to achieve high conversion without compromising selectivity towards longchain hydrocarbon. Plasma catalysis for FT synthesis was reported by [10] using DBD plasma with Cu/Co-based catalyst which was summarized in **Table 2**.

Table 2. The effect of Pressure, H_2/CO molar ratios and plasma power on selectivity and CO conversion in DBD plasma catalysis [10]

Parameter	C_{5+} selectivity	CO_2 selectivity	Methan selectivity	CO Conversion
Pressure	↑	↓	-	↓
H_2/CO	↑	↓	↓	↑
Plasma power	↓	↑	-	↑

Note: Increase with increasing parameter: ↑. Decrease with increasing parameter: ↓. No influence: -

It is interesting to note that increasing H_2/CO molar ratios can increase selectivity towards long chain hydrocarbons (C_{5+}) in plasma catalysis which is not the case for a conventional FT synthesis. For this reason, DBD plasma was suitable for high feedstock with high H_2/CO molar ratios.

Reaction Mechanism and Catalysis

The most credible mechanism of FT reaction for the hydrocarbon product is the carbide mechanism by CH_2 insertion. the route monomer formation via hydrogenation of dissociated or undissociated CO is still indeterminate. **Table 3** below describe the series of basic reactions proposed for the formation of linear hydrocarbons.

Table 3. Proposed Mechanism of the Hydrocarbon Synthesis from CO and H_2 [9]

Adsorption	
1	$CO + s \leftrightarrow CO_s$
2	$CO + s \leftrightarrow C_s + O_s$
3	$H_2 + 2s \leftrightarrow 2H_s$
Surface Reaction	
Water formation	
4	$O_s + H_s \leftrightarrow HO_s + s$
5	$HO_s + H_s \leftrightarrow H_2O + 2s$
Or	$O_s + H_2 \leftrightarrow H_2O + s$
Chain initiation	
6	$C_s + H_s \leftrightarrow CH_s + s$
7	$CH_s + s \leftrightarrow CH_2s + s$
8	$CH_2s + s \leftrightarrow CH_3s + s$
or	$CO_s + H_2 \leftrightarrow CHOH_s$
	$CO_s + H_2 \leftrightarrow CH_2s + H_2O$
Methanation	

9	$\text{CH}_3\text{s} + \text{Hs} \leftrightarrow \text{CH}_4 + \text{s}$
Chain growth	
10	$\text{C}_n\text{H}_{2n+1}\text{s} + \text{CH}_2\text{s} \leftrightarrow \text{C}_{n+1}\text{H}_{2n+3}\text{s} + \text{s}$
Hydrogenation to paraffins	
11	$\text{C}_n\text{H}_{2n+1}\text{s} + \text{Hs} \leftrightarrow \text{C}_n\text{H}_{2n+2} + 2\text{s}$
β -Dehydrogenation to olefins	
12	$\text{C}_n\text{H}_{2n+1}\text{s} \leftrightarrow \text{C}_n\text{H}_{2n} + \text{Hs}$

*s is catalyst site

The initial reactions begin with the adsorption of CO and H₂ on the catalyst sites. The catalyst plays an important role in the dissociative adsorption of the reactant on catalyst sites. The chain initiation will follow, and this could be either continue to chain growth to produce longer hydrocarbon chains or stop at methane. Paraffins and olefins are the main and favourable products in the FT synthesis.

Unlike conventional FTS, the reaction is performed via different routes in Plasma FTS. High energy electrons in DBD Plasma activate CO on the catalyst surface as well as activate hydrogen under ambient pressure and low temperature. The synergistic effect between DBD Plasma and catalyst will help to break the CO bond (11.16 eV). H₂ dissociation was not a problem in the DBD reactor as it has low dissociative energy at 4.25 eV. Hydrogen species activated by plasma will react with CO to generate CH_x-M, which will promote the initiation, propagation, and chain-growth termination steps of the reaction. The typical problem of carbon formation on the catalyst can be avoided by the formation of plasma-activated hydrogen. Oxygen species on the catalyst surface will then react with CO to form CO₂, while hydrogen follows the same route to produce water. The reaction mechanism of FTS was proposed as follows [10]

- a. H₂ dissociation and CO dissociative adsorption on metal atom was induced by plasma

- b. Insertion of plasma dissociated hydrogen

- c. Propagation

The developments of FT catalysts to increase the selectivity toward desired products, contribute to the most interesting research in the FT synthesis [11][12]. The mechanism of reaction was one of the topics to explore and to understand this problem [13][14]. Catalysts from group VIII metals are commonly used for FT synthesis. To ensure reactions taking place efficiently, the active phase on the catalyst surface must be vastly dispersed over a large specific surface area of the catalyst. A highly porous material (i.e silica) is used as a catalyst support to help high thermostability on the high surface area as well as provide better mechanical

strength. The preparation of these catalysts usually involves (i) introduction of a metal precursor on the support using ion-exchange, impregnation, co-precipitation, and deposition, (ii) drying, (iii) calcination and (iv) reduction if necessary [15][16].

Catalysts from group VIII metal may be used for FT synthesis. However, cobalt and iron have been used commercially because of their price and high catalytic activity. Cobalt is much more expensive than iron but offers better resistance to deactivation [9][17]. Although both catalysts can be used at low temperatures (200-250°C), cobalt is an ideal choice at low-temperature FT synthesis due to its high hydrocarbon productivity, stability and conversion [18]. The major factors contributing to the activity and product selectivity of the FT synthesis catalyst are illustrated in **Table 4**.

Table 4. Key factors affecting FT synthesis [19]

Engineering Factors	Catalyst Factors
1) Reactor design	1. Identity of active metal (Ru, Co, Fe, Ni)
2. Operation condition	2. Chemical state of active phase (metal oxide or carbide)
	3. Support (identity, pore structure, physicochemical properties)
	4. Promoter (noble metals, oxides of Mn, Zr or rare earth metals, alkali metal ions)
	5. Size of an active phase
	6. Microenvironment of active phase

An increase in CO conversion may affect the decrease of selectivity towards C5+ as the relationship between the two is complicated [9] [20]. Fe and Co have commonly been used for industrial applications of FTS and they offer different active phases during FTS which affects the selectivity of catalysts [21][22].

The design of catalyst for FT synthesis should satisfy higher surface area ($> 100 \text{ m}^2/\text{g}$), porosity, stability and optimum metal-support interaction. For cobalt case, weak metal-support interaction helps high cobalt reducibility but provides agglomeration of supported cobalt particles. In contrast, strong interaction between cobalt and metal-support could lead to the formation of cobalt silicate which is not favourite phase to catalyse FT synthesis [23]. Once the new catalyst is has been made, it is important to test the performance and compare with that of conventional and commercial catalysts [18].

Fischer-Tropsch Synthesis Performance

DBD has been a subject of extensive research in plasma including ozone production [24] [25], ammonia decomposition [26], methane dry reforming [27], degradation of toluene [28]. The research on DBD plasma assisted FTS, however, is still limited.

Figure 2. DBD Plasma Assisted FT Synthesis with Co/SiO₂ catalyst [29] and Cu/Co Catalyst [10]

DBD application on FTS is shown in figure 2. Catalyst Co/SiO₂ has better CO conversion at 42% compared to Cu/Co at 18%, the latter deliver better selectivity toward C₅+ hydrocarbon at 88%. These results are lower than those of the existing industrial FT process including, fluidized-bed reactors, Slurry

bubble column (SBCR), and fixed bed reactor. The fluidized bed reactor which is designed mainly for HTFT application has several drawbacks including high temperature operation (up to 350 °C), pressure drops in the catalyst beds, and selectivity toward short chain hydrocarbon [22]. Slurry reactors offer better mixing of the phase as syngas continuously contacted with a mixture of liquid product and catalyst. The problem of this type of reactor lay in the fact that sulphur could poison the catalyst resulted from continuous circulation. Fixed Bed Reactor is more common in FTS due to the ability to reach the highest productivity under uniform temperature. This reactor is also able to produce a wide range of hydrocarbon chain. The drawbacks of this reactor involve heat removal and pressure drops which in turn will limit catalyst utilization [22].

Table 5. Characteristic of Conventional Reactor [30] and DBD Plasma Reactor [29]

Requirements		Fluidized Bed	Slurry	Fixed Bed	DBD Plasma
Process	- High activity	~	~	~	±
Parameter	- Temperature mixed	+	+	-	+
	- Water vapor removal	-	-	-	NA
	- Heat removal	+	+	-	+
	- Good heat-transfer characteristics	+	+	-	+
Catalyst	- Catalyst attrition/separation	-	-	+	+
	- Particle size	< 100 µm	< 100	> 1 mm	8-10 nm

Note: +: sufficient/positive; -: negative; ±: moderate; ~: in some independent reactor type; NA: Not Available

It can be seen from **Table 5** that conventional reactor shows at least two disadvantages although recent development has improved on this issue [22]. Heat removal or catalyst attrition is still the major problem with the existing FT reactor. DBD plasma has a benefit over the conventional reactor in this matter.

Despite lower conversion, DBD plasma application on FTS shows potential for further exploration. The synergistic effect between plasma and catalyst enhances catalytic performance [29]. Besides, the characteristic of DBD which operate at low-temperature would prolong the catalyst lifetime as well as lower the energy used. The synergistic effect contributes to a substantial improvement in the energy efficiency of CO conversion. The advantages from the combination of low temperature plasma and solid catalyst can be attributed to both solid plasma–catalyst interactions and improved activity of the catalyst calcined at low temperatures [31][32].

III. Conclusion

Understanding reactor characteristic and process parameters are crucial to optimize efficiency and attain desired product selectivity in FT synthesis. Conventional reactor including Slurry bubble column, fluidized bed and fixed bed have been widely used in FT synthesis. As an exothermic process, the FT reactor must be designed to deal with the heat. DBD plasma can perform FT synthesis at lower operation temperatures, leading to better heat removal. The synergistic effect between plasma and catalyst also enhances catalytic activity and prolong catalyst lifetime. Further research in DBD Plasma Assisted FT synthesis is required to improve CO conversion and understand tune FT process parameter toward desired hydrocarbon selectivity.

IV. References

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