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A Review of Coal Liquefaction Using Direct Coal Liquefaction (DCL) and Indirect Coal Liquefaction (ICL) Techniques

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Abstract
Coal liquefaction is a process of converting solid coal fuel into liquid fuel. This process is developed by various countries to overcome the scarcity of liquid fuels; therefore, this coal liquefaction is very useful for countries that have few petroleum reserves yet, have large coal reserves. There are two coal liquefaction techniques, namely direct coal liquefaction (DCL) and indirect coal liquefaction (ICL). In ICL, there is a gasification process that usually develops products, generating Fischer-Tropsch (F-T) liquid, methanol (CH_3OH/MeOH), and dimethyl ether (CH_3OCH_3/DME). On the other hand, DCL involves a hydrogenation reaction to produce synthetic gasoline and diesel, which is the same as LPG, a hydrocarbon fuel product similar to hydrocarbon fuel derived from petroleum. This paper discusses the ICL and DCL coal liquefaction techniques, including the process stages, the type of catalyst used, and the products resulted from these techniques.
Keywords: Coal Liquefaction, DCL, ICL

I. INTRODUCTION

Compared to solid fuel, liquid fuel is the most widely used fuel. Solid fuel derived from coal has been widely converted into liquid fuel through coal liquefaction techniques. Coal liquefaction is used by countries that have abundant coal reserves; however, with minimal petroleum reserves such as China, the United States, and Japan (Erlangga, 2015). Therefore, coal liquefaction is necessary to meet the demand for liquid fuel. Liu *et al.* (2010) said that coal liquefaction is a change in the molecular structure of solid coal into liquid fuel and chemicals with the addition of hydrogen. There are two techniques of Coal liquefaction: the first is the Direct Coal Liquefaction (DCL), which is a technique of liquefying coal at high temperature and pressure, and usually involves a hydrogenation process; the second is coal liquefaction by breaking the coal structure through gasification to produce liquid fuel products, otherwise known as Indirect Coal Liquefaction (ICL).

Coal liquefaction technology was known in the early 20th century in Germany. Direct Coal Liquefaction (DCL) was introduced by Friedrich Bergius in 1913, and Indirect Coal Liquefaction (ICL) was introduced by Franz Fischer and Hans Trophsch, who introduced F-T synthesis or FTS in 1923 (Dadyburjor and Liu, 2004; Schulz, 1999). According to William and Larson (2003), the ICL technique produces Fischer-Tropsch (F-T) liquid, methanol (CH_3OH / MeOH), and dimethyl ether

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(CH_3OCH_3/DME), which have been widely commercialized, and there have also been many modules that discuss this technology. DCL technology includes the manufacture of crude oil from coal, including synthetic gasoline and diesel, which is the same as LPG, a hydrocarbon fuel product similar to petroleum-derived hydrocarbon fuel.

II. LITERATURE REVIEW

II.1. Indirect Coal Liquefaction (ICL)

The ICL technique has been used in South Africa, the United States, and China to produce liquid coal (Larson and Tingjin, 2003). William and Larson (2003) also mention that Indirect Coal Liquefaction (ICL) begins with a synthetic gas manufacturing technique first. ICL is a coal gasification technique to produce synthetic gas (Syngas), which consists of carbon monoxide gas (CO) and hydrogen gas (H_2), and then makes synthetic fuel from this Syngas. ICL also produces hydrocarbon fuel similar to crude oil derivative. Larson and Tingjin (2003) state that the ICL technique can produce methanol (CH_3OH), dimethyl ether (CH_3OCH_3), Fischer-Thropsch (F-T) diesel, and hydrogen (H_2) products. Talla and Taba (2017) state that coal is fragmented into CO, CO_2, H_2, and CH_4, which are then recombined to generate liquid products. The Fischer-Tropsch (F-T) mechanism is known in the coal gasification stage. The main products of the F-T process are paraffin and olefin, according to the following reaction:

$2nH_2 + n\,CO \rightarrow CH_2n + nH_2O$ (a)
$2nH_2 + n\,CO \rightarrow CH_{2n+1} + (n-1)\,H_2O$ (b)

Williams and Larson (2003) and Vasireddy *et al.* (2011) state that the gas of CO and H as the results of the gasification can be reacted into methanol.

$CO + 2H_2 \rightarrow CH_3OH$ (c)

Meanwhile, CO_2 gas will be reacted according to the following reaction:

$CO_2 + 3H_2 \rightarrow CH_3OH + H_2O$ (d)

Or else

$CO + 2H_2 \rightarrow CH_3OH$ (e)

The derivative of synthetic intermediate distillation via the Fischer-Tropsch (F-T) process which can be used directly as diesel or mixed with diesel oil derivative. Another possibility is the gasoline from the first process of making MeOH from Syngas, which is then converted into gasoline from the Mobil process. MeOH can also be used directly as fuel, and other oxygenates which are contained in the fuel, for example, DME, which can be used as a direct fuel.

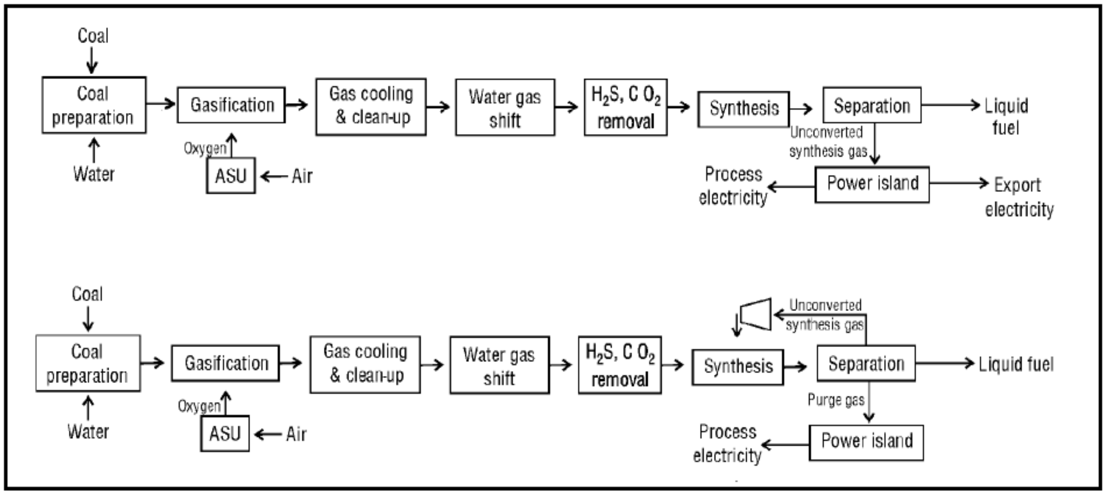


Figure 1. Coal liquefaction process configuration using ICL technique (Larson and Tingjin, 2003)

Figure 1 is the coal liquefaction process configuration using the ICL technique to generate MeOH, DME, and Fischer-Tropsch oil products. The difference between the two processes above is in the first figure in which electricity is a by-product—starting with pre-treatment, which is to put the coal in a certain size and add water into the inner tank so that it forms a slurry. The addition of water functions to increase the H_2 fraction in the coal. The tank operates at a pressure of 75 bar. Furthermore, the gasification process is carried out by converting the coal into Syngas, whose main content is hydrogen gas (H_2) and carbon monoxide (CO). Oxygen is obtained by separating the oxygen from the air, as shown in the process figure above. The heat from the partial oxidation of the coal is used to drive the gasification reaction. After the gasification process, the Syngas is then cooled and cleaned of contaminants. The cooling can be directly done using sudden water (direct water quench), or the hot Syngas is passed through the cooler. A water – gas shift reactor is installed after the initial cooling to regulate the ratio of H_2 and CO in the Syngas. Furthermore, removing sulfur is an important step in order to limit the SO_2 emission and protect the catalyst used in the synthesis stage [Turk *et al*., 2001]. The way to limit the sulfur at Level 1 ppmv is to absorb it in organic fluid and to react the amine with the sulfur in gas (Moore, 2003). Selexol or Rectisol solvents can be used to absorb CO_2 and H_2. The clean Syngas leaves the sulfur removal unit, then goes into the synthetic unit. The synthetic reactor operates at a temperature of 260°C. In the reactor, the H_2 and CO are converted into fuel oil products, after which they go into the purification unit, which is the flash tank or distillation to get the desired final product.

The Syngas passes through the synthetic reactor once only, as shown in the process configuration figure above. Unconverted gas into a fuel can be used for a gas turbine. The hot gas exiting the turbine is used together with the heat resulted from various processes recovery. The power generated by the turbine combined cycle is sufficient to provide power to operate the factory. As shown in the configuration figure above, the bottom one, the unconverted Syngas from the product in the recovery area, is returned to the synthetic reactor to produce additional liquid fuel. The unconverted gas-only meets the process needs without additional electricity from outside.

Liquid fuel from figure 1 can be synthesized into methanol, dimethyl ether, and ethylene glycol. Methanol from Syngas is carried out on traditional CuO-ZnO-Al_2O_3 (CZA) catalysts. Methanol synthesis has been explored as a reaction to CO_2 abatement. Methanol can be synthesized directly from CO_2 hydrogenation, while H_2 could be obtained from renewable sources. CO_2 reformed from coke oven gas to decrease CO_2 emission and save energy. By using this method, not only can partial recycle of CO_2, but also a syngas with H_2/CO ratio close 2. This ratio is the best ratio for methanol synthesis, which can be provided under stoichiometric conditions of CH_4 and CO_2 (Jin et al., 2014).

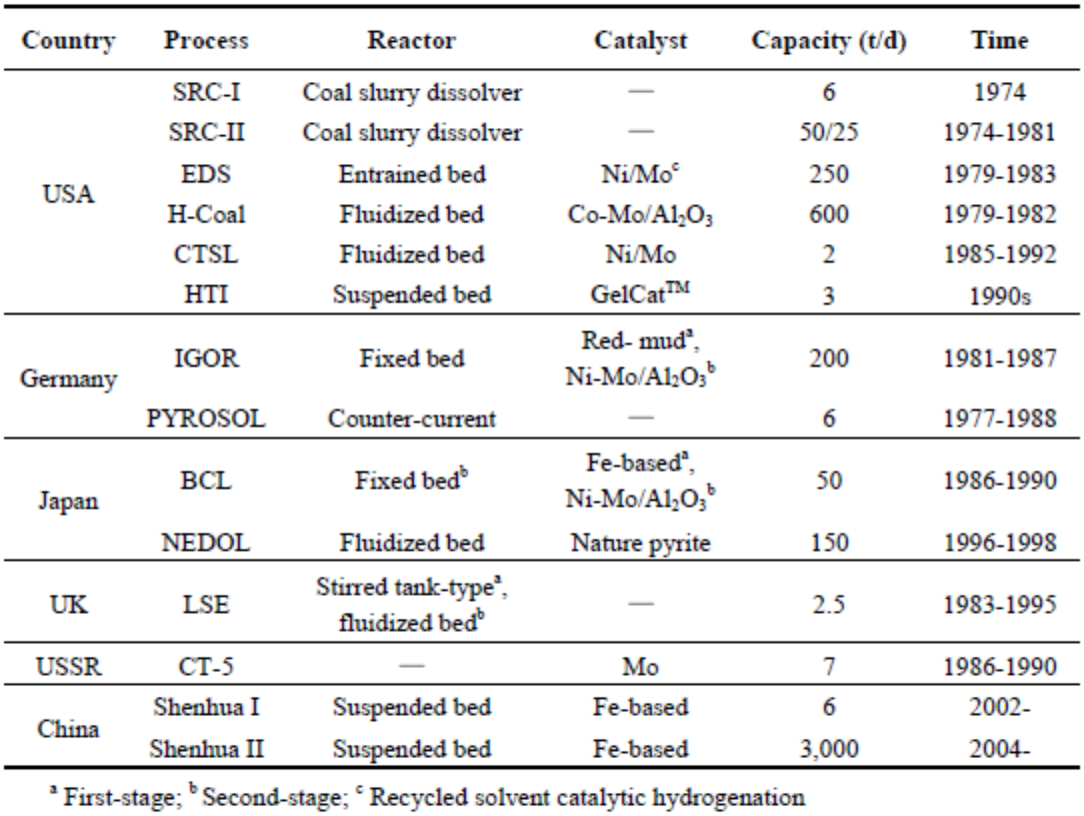
If the efficiency is taken into account in the final product, the ICL technology can be as efficient as DCL technology. DME, which is a product of ICL, is superior to products from DCL in terms of impacts on the air quality. Compared to DCL, ICL requires stronger infrastructure and technology support (William and Larson (2003).

II.2. Direct Coal Liquefaction (DCL)

Direct coal liquefaction has also been performed in various countries such as the United States, Europe, Japan, and China. Table 1 below shows that the DCL technique has been implemented in various countries. SRC-I and SRC-II (Solvent Refined Coal) have been developed in the United States. China has commercially built a DCL factory in Shenhua. Thus, the process is called the Shenhua DCL process (Comolli *et al*., 1999).

Talla and Taba (2017) said that the direct coal liquefaction process that has the potential to be developed in Indonesia is a catalytic method. The direct coal liquefaction (DCL) technique, which uses a catalyst, is carried out with solvent rock that is given high pressure and high temperature. This aims to break the coal hydrocarbon structure into reactive fragments (free radicals). The assistance of solvents, H_2 gas, and catalyst are needed to stabilize free radical fragments to form oil fraction. The DCL technique, which uses a catalyst, has the potential to be applied to low-rank coal and natural catalysts, which are abundant in Indonesia. The role of a catalyst in coal liquefaction is to help insert hydrogen atoms from the dissociation of hydrogen molecules into coal or a mixture of coal with a solvent, thereby increasing the availability of active hydrogen. This hydrogen functions to hydrogenate aromatic compounds, promote bond-breaking reactions, and stabilize free radicals. By using a catalyst, the hydrogenation reaction between hydrogen gas and coal can take place very quickly.

Table 1. Summary of major DCL processes developed around the world (Shui *et al*., 2010)



Country	Process	Reactor	Catalyst	Capacity (t/d)	Time
USA	SRC-I	Coal slurry dissolver	—	6	1974
	SRC-II	Coal slurry dissolver	—	50/25	1974-1981
	EDS	Entrained bed	Ni/Mo^c	250	1979-1983
	H-Coal	Fluidized bed	Co-Mo/Al_2O_3	600	1979-1982
	CTSL	Fluidized bed	Ni/Mo	2	1985-1992
	HTI	Suspended bed	GelCat^{TM}	3	1990s
Germany	IGOR	Fixed bed	Red- mud^a, Ni-Mo/Al_2O_3^{b}	200	1981-1987
	PYROSOL	Counter-current	—	6	1977-1988
Japan	BCL	Fixed bed^b	Fe-based^a, Ni-Mo/Al_2O_3^{b}	50	1986-1990
	NEDOL	Fluidized bed	Nature pyrite	150	1996-1998
UK	LSE	Stirred tank-type^a, fluidized bed^b	—	2.5	1983-1995
USSR	CT-5	—	Mo	7	1986-1990
China	Shenhua I	Suspended bed	Fe-based	6	2002-
	Shenhua II	Suspended bed	Fe-based	3,000	2004-

^{a} First-stage; ^{b} Second-stage; ^{c} Recycled solvent catalytic hydrogenation

For example, Talla *et al.* (2013) conducted coal liquefaction with Sorong (lignite) coal and noble (sub-bituminous) coal. This experiment was carried out in a 5-liter autoclave with anthracene solvent and iron ore catalyst at a temperature of 375 to 450°C and a pressure of 100 bar. As a result, the Sorong coal was converted as much as 89.94% at 400°C, and the noble coal was converted at 87.28% at 450°C. According to Talla *et al.* (2013), the problem that occurs in low-level coal is its high water content. The high water content will affect the pressure in the autoclave. Therefore, the coal needs to be dried first.

Marco *et al.* (1990) conducted coal liquefaction with coal rank variables and concluded that the low-rank coal produced higher liquid product than the high-rank coal. This is supported by Talla *et al.* (2013), that the low-rank coal is easy to liquefy because of the higher H/C ratio, and the structure is composed of weak bonds that break easily. The high hydrogen content in coal affects the ratio of hydrogen (H) and carbon (C), which is also getting higher. The high H/C ratio in coal causes the coal to be more reactive or easy to convert at low temperatures.

In addition, the effect of volatile matter in the low-rank coals is dominated by the aliphatic structure and the ether (-O-) functional groups, which are weak and break easily when heated. This is similar to the opinion of Karaca *et al.* (2006), that the low-rank coal produces higher conversion in liquefaction since it is composed of a weak structure and breaks easily at high temperatures and high pressures. Another factor is the maceral composition. Vitrinite and liptinite macerals are the groups that are easy to liquefy and produce high conversions because the H/C ratio of maceral and liptinite is close to the H/C ratio of oil.

Hirano and Kanda (2001) state that in the coal liquefaction process, catalysts play a role in increasing the total conversion and expected oil products, as well as influencing the use of hydrogen and accelerating the process of the chemical reactions taking place. In addition, the catalysts play a role in the re-hydrogenation reaction of the hydrogen donor solvent so that it can increase the amount of hydrogen removed and reduce the coal product that becomes gas (Derbyshire, 1988). Some of the iron-based catalysts that have been developed are Limonite, Pyrite, and Laterite (Kaneko *et al.*, 2000). The results of Kaneko's research (2002) show that limonite looks very good as a catalyst in the coal liquefaction process. Natural limonite contains various metal elements, including Fe, Ni, Cr, Co, Mg, Al, and other elements.

Ningrum *et al.* (2009) tested several iron-based catalyst materials and the effect of Ni metal elements and chromium oxide (CrO_3) on the formation of the active compound $Fe_{1-x}S$ pyrrhotite. The catalysts in coal liquefaction are classified into three groups, namely artificial/synthetic catalysts, mineral catalysts, and catalyst derived from industrial waste. The examples of artificial/synthetic catalysts are $CoMo/Al_2O_3$, $ZnCl_2$, and SnS. The examples of mineral catalysts are Pyrit (FeS), Cassiterite (SnO_2), Limonite (FeOOH), and Fe_2O_3. These mineral catalysts are found in rocks that contain oxides such as laterite, iron sand, and others. Mineral catalysts have a lower price, and their reactivity is almost the same as synthetic catalysts. Meanwhile, red mud is an example of catalysts that come from industrial waste.

According to Silalahi (2006), the catalyst from transition metal is expensive and cannot be used repeatedly, and the catalyst activeness after the reaction is greatly reduced so that the use of this catalyst cannot be used on an industrial scale. The iron-based catalyst is cheap, easy to obtain, and can be reclaimed and activated before using it. The iron-based catalyst needs to be added with sulfur to increase the active catalyst power. This catalyst is suitable for obtaining high process efficiency to be commercialized.

Silalahi (2006) conducted lignite-type coal liquefaction using a limonite iron-based catalyst (α-FeOOH). The experiment was carried out on a 5-liter autoclave equipped with a stirrer, reactant gas pipeline, and equipped with a heating jacket system. The autoclave was run at a temperature of 450°C, at the initial pressure of 90 kg/cm^2G, and 1-hour reaction time. The solvent used was creosote oil. The results showed that the limonite catalyst was able to produce distillate yield up to 51% of ash-free dry coal.

III. CONCLUSION AND FURTHER RESEARCH

There are two coal liquefaction techniques: ICL and DCL. The ICL technique goes through a gasification process, where generally coal liquefaction begins with the making of Syngas, then the Syngas is synthesized into fuel and other chemicals, for example, Fischer-Tropsch (FT) liquid, methanol (CH_3OH/MeOH), and dimethyl ether (CH_3OCH_3/DME). On the other hand, DCL involves a hydrogenation reaction to produce synthetic gasoline and diesel, which is the same as LPG, a hydrocarbon fuel product similar to hydrocarbon fuel derived from petroleum. The coal liquefaction using the DCL technique can either involve catalysts or not. Some of the catalysts that can be used in the DCL technique are artificial/synthetic catalysts, mineral catalysts, and industrial waste catalysts. Examples of synthetic catalysts are $CoMo/Al_2O_3$, $ZnCl_2$, and SnS. Examples of mineral catalysts are Pyrit (FeS), Cassiterite (SnO_2), Limonite (FeOOH), and Fe_2O_3. Iron-based catalysts such as Limonite, Pyrite, and Laterite. Meanwhile, an example of an industrial catalyst is red mud. ICL technique requires stronger infrastructure and technology support than DCL.

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