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Original Article

Green Processing of Coal to Liquid Fuels: Pakistan's Perspective

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Abstract: The role of energy supplies in economic prosperity and environmental quality is one of the most important challenges in Pakistan. The continuing upward trend in crude oil prices in the world and heavy reliance on petroleum and its derivatives in daily life makes the economy vulnerable to critical stress. Therefore, the energy security has gained increasing importance. In Pakistan, only the transportation sector consumes one-third of petroleum derived fuels while other consumers include electric power generation and a number of petrochemical products. The use of oxygenates (fuel additives), olefins (petrochemicals), etc. are becoming popular. The recent discoveries of coal reserves and its pronounced scope in the energy sector in the wake of new technologies have led to its green processing and effective utilization. The challenge of efficient utilization and green processing of coal at manageable cost is of interest to researchers. It is through the Coal to Liquid (CTL) technology that coal is converted to valuable liquid hydrocarbons. The two step process, i.e., gasification, followed by its conversion to liquid fuel by Gas-to-Liquid (GTL) technology is a proven strategy, commonly known as Fischer–Tropsch synthesis (FTS) process. Significant improvement of scope in this technology through improved catalysts and process conditions is of interest. Underground Coal Gasification (UCG) is an attractive option for GTL technology for economic gains. Preliminary studies have already been conducted in the country. The prospective use of CTL and GTL fuels technologies in Pakistan has been reviewed in this paper.

Keywords: Coal, green processing, GTL, CTL, Fischer–Tropsch synthesis, gasification

1. INTRODUCTION

The world fuel reserves are estimated as: crude oil, 5775 Quads (Q); gas 5137 Q; and coal 30100 Q [1]. The overwhelming energy security as coal is appreciated by industry and it is believed that this status may not change in the forth coming era. The possibility of using coal as a source of syngas production for liquid fuels and petrochemicals is obvious [2, 3]. Pakistan is amongst the country having significantly high coal reserves, but unfortunately the coal has not been used extensively as energy source. Primarily this is due to lack of infrastructure, investment in modern coal mining and processing technology. Pakistan's total coal reserves are approximately 185 billion tones, while the economic coal deposit is restricted to Paleocene and Eocene rock sequences [4]. At present the country faces serious energy crises and its future demand is growing at a rate of 7.5 % per annum. To the future requirements of the country with indigenous resources, domestic exploration is expected to be intensified. Currently, the attention is focused for development and utilization of Thar coalfield, one of the world's largest lignite deposits (approximately 175 billion tones) spread over more than 9,000 sq km [4, 5].

A feasibility study on coal gasification has been undertaken and the gasification of coal was found feasible. The importance of coal as an industrial fuel and its role in a wide range of industrial applications are well known in the industry. Coal is used as boiler fuel for the supply of steam the to process plants in paper, chemical, electrical and food processing industries. It is also used for direct

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firing in the manufacturing of cement, bricks, pipes, glass tanks, and metal smelting. Another effective technology for power generation from lignite coal is Circulating Fluidized Bed (CFB), where coal mixed with limestone is burned in a fluidized bed [6]. The sulfur in the coal is absorbed by the calcium carbonate, and the emissions are free from sulfur oxides [7].

Continuous increase in the prices of gas and oil have severely affected the energy prices. Underground coal gasification, a means to generate coal gas economically, is known to have the potential for power generation and production of other high value chemical products such as diesel, gasoline, olefins, methanol, and ammonia. The technology of UGC is available for both horizontal and inclined coal beds. The mathenated synthesis gas (known as SNG) can be blended with natural gas and transported through pipelines.

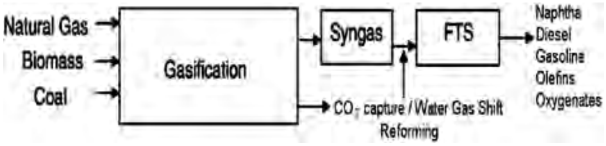

Fig. 1. CTL/GTL process.

The importance of coal as an industrial fuel and its role in a wide range of industrial applications are well known. In this paper we will highlight green processing of coal to energy and explore the space for more valuable products. Indirect use of coal processing to liquid fuels proceeds in two steps: (i) coal gasification; and (ii) conversion of the gas (called syngas) to liquid fuels (Fig. 1). A variety of CTL/GTL technologies for converting coal feedstock into liquid fuels exists, the most popular being Fisher Tropsch process. Generally the steps involved are: (i) feedstock preparation; (ii) gasification; (iii) syngas clean-up; (iv) compression; (v) mathenation; and (vi) conversion into liquid fuel in a reactor. Germany in the Second World War and South Africa at present have used this technology extensively. Recently, interest has aroused to make use of this technology all over the world. Qatar is about to produce about 394,000 barrels of GTL products per day and will become prominent figure in the world in GTL. A list of Qatar's GTL ventures is shown in Table 1 [8]. It is expected that the total GTL production in the world shall reach 1–2 million bpd by 2015 [9].

Table 1. GTL joint venture projects in Qatar.

Project Installer	Full Capacity (bpd)
ConocoPhillips	160,000
Pearl(Shell)	140,000
ExxonMobil	154,000
QP/Sasol Chevron	130,000
Marathon	120,000
Oryx	100,000

Source: Culligan [8].

2. COAL GASIFICATION

Coal and natural gas as source of syngas have been extensively studied [10]. Coal gasification is the most capital intensive part (~40%) of a CTL process. Therefore, selection of gasification method and its design has considerable impact on overall utilization of coal. Partial oxidation (catalytic/non-catalytic), steam-reforming, auto-thermal reforming, compound reforming, underground gasification and ceramic membrane reforming are known gasification techniques. A simplified coal gasification process is shown in Fig. 2. Steam reforming, partial oxidation or a combination of both oxygen blown Auto-Thermal Reforming (ATR) were the potential technologies for CTL/GTL [11–14]. Coal is prepared by milling, grinding and drying operations and then fed to the gasifier where it reacts with steam and an oxidant agent, in this case, pure oxygen, to generate a mixture of gases (mainly CO, H_2 and, CO_2). In steam reforming, a multi-tubular fixed bed catalytic reactors produce high H_2/CO ratio of syngas ranging from 3 to 5. The main reforming reactions are

$$CH_4 + H_2O = CO + 3H_2; \Delta H^0_{298K} = 205.92 \mathrm{kJ/mol}$$

$$CH_4 + CO_2 = 2CO + 2H_2; \Delta H^0_{298K} = 247.32 \mathrm{kJ/mol}$$

Partial oxidation is simple but suffers from soot formation and high outlet temperature, i.e., 1500^{o}C.

$$CH_4 + 1/2O_2 = CO + 2H_2; \Delta H^0_{298K}$$
$$= -35.25 \mathrm{kJ/mol}$$

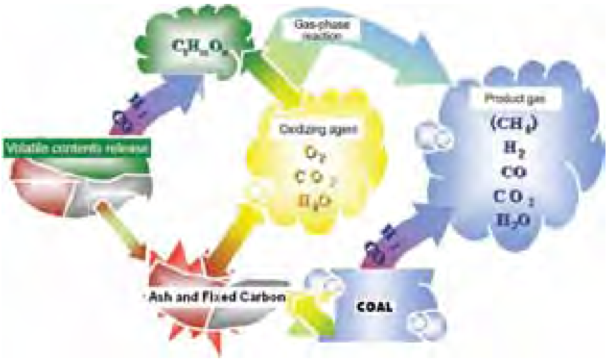

Fig. 2. Overview of coal gasification process.

In the auto-thermal reforming process, both partial oxidation and steam reforming are integrated in reactor and higher pressure syngas is obtained. In ATR process large volume of CO_2 is produced. In compound reforming both early-stage steam reforming reaction and the late-stage automatic-thermal reforming reaction take place in separate reactors and finally high-pressure gas is obtained. Catalytic Partial Oxidation (CPOX) on membranes is known to be the most economical technology as combustion is employed.

Gasifier designs are characterized as: wet or dry feed, air or oxygen blown, reactor flow direction and the gas cooling process. High temperature, entrained flow design gasifiers produce by-products like slag while lower temperature design produces ash. The majority of successful coal gasification processes have been achieved using pressure at 20–70 bar, entrained flow, and slagging gasifiers operating temperatures is about 1400^{o}C [13]. Most advanced gasifiers with lower methane and CO_2 content are the Shell Gasifier, PRENFLO, and E-Gasifier, etc. Benefits of entrained flow designs are clean, tar-free syngas, high operation temperatures, inert slag separation, high oxygen consumption, etc.

Nitrogen is especially undesirable when the syngas produced is intended for FT synthesis as it increases the volume of gas to be compressed; hence, Air Separation Unit (ASU) is critical. At present cryogenic distillation for oxygen-nitrogen separation is the only commercially proven technology for large scale systems. Apart from the reduced size of the gasifier and downstream equipment, other advantages associated to an oxygen-blown gasifier are: (a) the volume of gas produced is reduced; correspondingly, the sensible heat loss from the gasifier is reduced; (b) the gasifier can be operated economically at higher pressures; and (c) the heat-exchangers for the recovery of the sensible heat from the syngas are thus smaller.

During gasification, the sulfur is converted to H_2S or carbonyl sulfide (COS). One of the main concerns in this system is the removal of hydrogen sulfide, carbonyl sulfide, particulate matter, carbon dioxide, and hydrogen chloride. For most syngas contaminants the critical aspect of filtering out these undesirables is the volatility of various components. The maintenance of temperature is of concern. The catalyst poisoning concerns have led to limit the concentration limits of FT reactors to 1 ppm for particulates and 10 ppb for sulfur compounds. Bio-desulphurization of coal is also an option to avoid the sulfur contaminations [15]. The necessary step in acid gas clean-up is to convert the COS into H_2S and CO_2 by COS hydrolysis. The H_2S and CO_2 get absorbed from gas stream through an absorber where the Selexol solvent before the FT reactor.

The coal production, transportation, and utilization processes have impact on the environment in terms of dust, ash, CO_2, NO_x, SO_x, etc. Therefore, green processing to minimize the harmful impact coal utilization on the environment is attractive for rapid commercialization. Numerical simulation of gasification processes is a very effective tool to predict the characteristics of pilot to full scale production unit and allows optimization. There is, however, a significant issue of variation in coal characteristics, even if the coal has come from the same coal mine.

3. SYNGAS TO LIQUID FUELS

The syngas generated from gasifier is sent to a Fischer Tropsch (FT) synthesis reactor after cleaning. The reactor having various metal based patented catalyst (e.g. Fe, Ga, Ni, Cr, Co, Mo, Mg, Pt, Ru, Re, etc.), produces liquid fuel through water-gas shift reaction. The conversion of hydrocarbons into liquid fuels in FT reactors proceeds via surface polymerization reactions over a catalyst. Syngas components get adsorbed on the catalyst surface to form monomer and water. The FT reactions that lead to the formation of synthetic liquid fluids are given below, where the H_2/CO ratio is reaction dependent and is between 2–3.

Main Reactions:

Paraffins $(2n+1)H_2 + nCO \rightarrow C_nH_{2n+2} + nH_2O$
Olefins $2nH_2 + nCO \rightarrow C_nH_{2n} + nH_2O$
Water gas shift reaction $CO + H_2O \rightleftarrows CO_2 + H_2$

Side reactions:

Alcohols $2nH_2 + nCO \rightarrow C_nH_{2n+2}O + (n-1)H_2O$
Boudouard reaction $2CO \rightarrow C + CO_2$

3.1. Fischer-Tropsch (FT) Reactors

The selection of reactor and their design for FT synthesis is based on temperature and hydrodynamics of catalyst [16]. In this exothermic reaction large amount of heat is generated and effective heat removal manages carbon deposition and subsequent deactivation of the catalyst. The temperature, pressure at the FT reactors and type of active metals in catalyst will determine the final product distribution. Higher temperatures generally favor the formation of smaller chains hydrocarbons and lower temperature favors the formation of higher molecular weight products. High performance for high temperature FT reactions for the production of gasoline are entrained fluidized bed reactor with riser coolers {such as Sasol's Synthol reactor [17]}, and the fixed fluidized-bed reactor with internal cooling coils (used at Carthage-Hydrocol plant, Texas [18]). In order to produce middle distillates or olefins the most feasible options are fixed bed tubular reactors (Sasol fixed bed tubular design is known as ARGE), dual bed and slurry bubble columns. The alternative to the fixed bed is a slurry bubble column which has been introduced in market by Sasol, Exxon and Rentech [19]. In these three phase reactors, solid catalyst is suspended in a liquid phase, often well mixed FT wax with syngas is in slurry phase [19]. The slurry phase reactor has therefore better catalyst dispersion and results in a higher single pass conversion compared to the ARGE reactor. Both types of reactors have some limitations; in fixed bed the catalyst is poisoned near the gas inlet while in slurry column attrition and continuous separation between the catalysts-liquid is a problem [19].

3.2. FT Catalysts

Most of Group VIII transition metal oxide catalysts are generally capable of CO hydrogenation; for example Ruthenium based catalyst has highest activity and selectivity for producing high molecular weight products [20], but is expensive. Nickel has a good activity but promote methane formation and its stability is also an issue. Iron is the most commonly used catalyst for FT process, although produces unwanted CO_2 [21, 22]. Generally, Fe catalysts are good for water gas shift reaction, therefore, needs a separate WGS reactor. Cobalt-based catalysts shows low selectivity for WGS reaction, therefore, it is only suitable for a syngas of high H_2/CO ratio and used as separate WGS reactor prior to the FT synthesis reactor. Active metal supports in the catalysts also plays vital role in controlling FT reactions and their important features are higher surface area, hydrothermal stability, active metal dispersion, shape selectivity effect, etc.

The product distribution in F-T synthesis reaction proceeds as explained by Anderson-Schulz-Flory polymerization model [23].

$$\log \frac{m_p}{P} = \log(\ln^2 \alpha) + P \log \alpha$$

m_p is weight fraction of each carbon number fraction

P is carbon number

α is the probability of chain growth

According to above model the maximum production of C_2-C_4 hydrocarbons is about 56% of the total yield. Most of the syngas to olefins conversion catalysts are either mixed oxides or carbonyls derives on different supports. Nickel-palladium, Zn/Cr, Fe/Co, and cobalt-cerium oxides with number of supports and promoters, prepared using a co-precipitation procedure were studied as catalysts for the direct conversion of syngas to light olefins. The higher Co percentage (around 80%) relative to Ce (i.e. around 20%) in the catalyst's proposed optimum in activity and selectivity with H_2/CO molar feed ratios 2/1. The yield and selectivity of lower olefins in direct conversion route is still far from optimum and has scope for R&D in the development of robust catalyst.

4. SYNGAS TO LIQUID FUEL TECHNOLOGIES

The technologies for converting coal to gaseous and liquid fuels are in commercial use as well as improvement through R&D activity. These technologies provide an opportunity to reduce dependence on crude based feedstock. Various syngas production technologies include partial oxidation (catalytic/non-catalytic), steam reforming, auto-thermal reforming, compound reforming ceramic membrane reforming, etc. There is significant room for development and optimization. One of the most economical technology in this reference is considered to be oxygen blown auto-thermal reforming (ATR) process. It has been commercialized by Haldor Topsøe [21]. In this case the process is operated at 0.6 steam to carbon (S/C) ratio. Recently, syngas to light olefins concept was introduced and in future, there is a huge potential for syngas to spatiality chemicals. Extensive experimentally study, construction of pilot/demonstration scale plants and operation of commercial plants makes GTL a mature technology. Many well-known companies have large-scale plant operating experience like ExxonMobil in GTL, Sasol, Shell, IFP, BP, Syntroleum, Rentech and Conoco [22]. A brief of the new developments are summarized here.

Sasol a South African company founded in 1950 is well known due to coal driven syngas to liquid fuels. In 1951, Sasol construction first production facility 'Sasol-I' on German Fischer-Tropsch technology began in Sasolburg, and started production in 1955. Coal based Sasol/Lurgi fixed-bed dry bottom gasifiers at Sasolburg and Secunda. This syngas is fed to Sasol FTS plant having designed capacity around 135,000 bpd. Recently, Sasol start integrated their FT technology with Haldor Topsoe auto-thermal reforming technology. Sasol has developed several types of Fischer-Tropsch technologies as listed below:

(a) High Temperature Fischer-Tropsch (HTFT) reactors:
   1) Synthol-Circulating Fluidized Bed (SCFB) reactor (Synthol)
   2) The Sasol Advanced Synthol (SAS) reactor

(b) Low Temperature Fischer-Tropsch (LTFT) Reactors:
   1) Multi-Tubular Fixed Bed (MTFB) reactor
   2) Slurry Phase (SP) reactor

Mossgas (Pvt) Ltd., is a South African government-owned company, introduced three step production process to synthetic diesels: (i) syngas by steam reforming of natural gas; (ii) high temperature FTS (Sasol technology) to form an olefinic synthetic distillate, synthol light oil (SLO); and (iii) Mossgas process convert lighter olefinic gasses to distillate (COD). In this process olefins are oligomerized over COD-catalyst to form high quality diesel fuel, kerosene, gasoline components, liquid petroleum gas (LPG) and a range of anhydrous alcohols. The 22,500 bpd sulfur-free and eco-friendly fuel is produced by Mossgas.

Royal Dutch Shell introduced a state-of-the-art proprietary GTL process - Shell Middle Distillate Synthesis (SMDS). In 1973, it started research on a modified low-temperature Fischer-Tropsch (F-T) process, leading to the development of the Shell Middle Distillate Synthesis (SMDS) route and first commercialized in Shell's Bintulu plant in Malaysia in 1993. This plant has the capacity to convert 100 million standard cubic feet per day (MMSCFD) of natural gas into 12,500 bpd of middle distillates (gasoil, kerosene, naphtha) and specialty products. In 1997, an explosion took place in the air separation unit (ASU) and damaged the GTL facility. The facility was rebuilt and started production again in 2000. Shell had a good experience in development of low temperature FTS catalyst and its use in the Shell proprietary multi-tubular reactor. It has been claimed that the catalyst has higher yield and selectivity of 90% for desirable middle distillate products. After getting operational and scale-up experience at Bintulu and breakthrough in the low-temperature FTS catalyst development, Shell signed agreements with Qatar Petroleum in 2003 to build the world's largest GTL plant in Ras Laffan, Qatar is likely to produce 140,000 bpd of products primarily naphtha and transport fuel.

Syntroleum, an Australia GTL process design company is working since 1980s. A highly active cobalt-based FTS catalysts for air fed auto-thermal reactor syngas conversion in a multi-tubular fixed-bed reactor or/and slurry reactor has been developed. An 11,500 bpd plant to convert natural gas into ultra-clean specialty products, such as lubricants, industrial fluids and paraffin's, as well as synthetic transportation fuels was installed at Sweetwater Australia. Syntroleum has grown significantly and now has nine commercial GTL projects worldwide with six in Qatar as joint ventures with major international oil companies [24].

ExxonMobil was created by Exxon's 1999 after acquisition of Mobil. Exxon has invested heavily in research to develop its "Advanced Gas Conversion technologies" and build 200 bbl/day GTL three step pilot plant in Baton-Rouge, USA, in 1995-96. Catalytic partial oxidation using fluidized bed reactor for conversion of synthesis gas by slurry phase (F-T) reactor and fixed bed hydro-isomerization. Exxon claims its proprietary process has high productivity and economic benefits. The products of Exxon's GTL process are clear, colorless, biodegradable, very-clean burning liquids with low odor, free of Sulphur, Nitrogen, Aromatics and other impurities; they are ideal feeds for petrochemical and refining applications. Recently, Syntroleum executed an agreement with ExxonMobil that grants it a worldwide license under "ExxonMobil's GTL" patents to produce fuels from natural gas and coal [25].

Chevron is engaged in design and engineering for Nigeria GTL facility, which is likely to convert natural gas into synthetic crude oil. Chevron also signed 50/50 joint venture with Sasol for Sasol's F/T technology and Chevron's Iso-cracking technology offers a unique combination of world class technologies to establish GTL [op.cit.]. Rentech GTL Technology, Colorado, USA has developed F-T process in 1985, using slurry reactor and precipitated iron catalyst to convert synthesis gas produced from range of feed stocks into clean, sulfur-free, and aromatic-free alternative fuels [op. cit.]. Rentech GTL Technology has unique features in its technology in terms of formulation of catalyst and reactor configuration.

Conoco Philips initiated a GTL research and development program in 1997 and began operation of a 400 bbl/day GTL in a demonstration plant in Ponca City, Oklahoma in 2003. They design a number of catalysts for said process. Conoco's proprietary catalysts to be used in synthesis and Fischer-Tropsch processes are known to be unique. Lurgi of Germany founded in 1897, recently builds the largest three-train coal gasification to olefins (syngas to methanol to propylene to polypropylene) plant in China. Previously they have wide experience in Methanol-to-Olefins (MTO) and/or Methanol-to-Propylene (MTP). IFP France is also in the process of piloting a GTL plant.

5. GTL CHALLENGES IN PAKISTAN

The design and development of CTL/GTL plants of commercial scale is very complex and challenging especially as R&D is lacking along with engineering fabrication and related potential in Pakistan. At the moment there is no piloting experience in these technologies and their catalyst development. Recently the Government of Pakistan has shown keen interest in developing these technologies in the wake of acute energy short supply and shortage indigenizes the resources. In this regards, Department of Chemical Engineering, University of Engineering & Technology (UET), Lahore and Center of Energy Systems, National University of Science and Technology (NUST), Islamabad have joined hands to promote these technologies. Initially, UET-built pilot scale gasification facilities, including down draft, cross draft, circulating fluidized bed gasification and underground coal gasification. Simulation and modeling studies have also been undertaken. On the other hand, NUST is pursuing piloting GTL process and integrated catalyst development. In order to address the challenges, international collaboration has been established in South African, UK and Germany. The other major challenge is to integrate internal utilities, like high grade energy users in reformer, oxygen/steam generator, product-workup fired heaters and recycle compressors. Increasing capacity of single-train brings more issues as ASU capacity, compressors, reduction of steam to carbon (S:C) ratio leads to further challenges in burner and reactor design, controlled catalytic partial oxidation, etc.

The art of process design and optimization of coal technologies should be addressed with the state of minimum entropy production in several process units. These studies gave insight into the design with more or less fixed boundary conditions. One of the important questions in such chemical processes is: how does the yield in the reactor affect the downstream units and energy efficiency? (e.g., composition, flow rate of the recycle streams, separation equipments, compressors, etc.) Optimum synthesis of a GTL technology is complicated due to many degrees of freedoms in a highly constrained design space. In a confined design space of equipments and operation, the selection of alternative syngas technologies, different types of Fisher-Tropsch catalysts and reactors, choice of air separation units, compressors, sulfur removal, heat integration options and a range of operational conditions. The state of art computational modeling expertise is being developed to enable the design of sophisticated GTL process design where economic performance should be aligned with carbon and energy efficiencies. Coal-to liquid process is a promising choice to convert coal to syngas and then to synthetic liquid fuels. In the second step, the green GTL synthetic fuel produced from synthesis gas ($CO+H_2$) through FTS (Fischer–Tropsch synthesis) retains extremely low sulfur and aromatic compounds using Fe- or Co-based catalysts, and reduces emissions of carbon monoxide, nitrogen oxides, etc.

6. CONCLUSIONS

To ease out the energy crisis situation in Pakistan, it is inevitable to focus on the development of synthetic fuel from coal in a manner that it does not deteriorate the environment. Rapidly expanding population and infrastructure in the country will inevitably lead to increase the fuel consumption for transportation, energy generation and petrochemical products. To meet future energy requirements of the country, Pakistan has to explore the unused coal reserves through CTL/GTL technologies. Both of these are well developed and proven technologies and offer an important option for producing FTS liquids, oxygenates, fuel additives and chemicals. Different strategies to convert coal to liquid fuels and commercial GTL activities have been discussed. Mega GTL plants with large capacities can be commercialized with ASU and slurry bed reactors having cobalt-based catalyst. Moreover, utilities requirement for CTL/GTL depends upon the train capacity of the unit, like heat integration, heat removal from the syngas and FTS units. Thus, in today's coa- rich Pakistan, GTL technology may be favored and catalyst R&D must be focused.

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