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New Wave of Coal-to-liquids

An Opportunity to Decrease Dependency on Oil and Gas Imports and an Appropriate Approach to a Partial Revival of Domestic Coal Industries

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Neue Welle der Kohleverflüssigung – Eine Gelegenheit zur Minderung der Abhängigkeit von Öl- und Gasimporten und ein geeigneter Zugang zur teilweisen Wiederbelebung der heimischen Kohleindustrie –

Derzeit erlangt weltweit die heimische Kohle sowohl wirtschaftlich als auch politisch wieder an Bedeutung. Beispiele aus den USA und aus China belegen dies zweifellos. Ausgehend von der Kohle als fossilem Energieträger, insbesondere Kohle geringerer Qualität, also mit hohem Asche- oder Schwefelgehalt, werden über mehrere Prozeßschritte unterschiedliche Endprodukte wie Treibstoffe, Chemikalien oder auch Strom hergestellt. Die Auswahl der herzustellenden Endprodukte wird hauptsächlich durch den zu beliefernden Absatzmark bestimmt und ist daher je Anlage zielgerichtet festzulegen. Eine Besonderheit bilden hierbei die IGCC's, die zur reinen Verstromung von Kohle dienen und keine weiteren Endprodukte generieren. Sowohl Koppelproduktionsanlagen zur Herstellung der oben genannten Ko-Produkte als auch IGCC's haben jedoch als ersten wesentlichen Prozeßschritt stets die Feststoffvergasung, bei der die Kohle in ein synthetisches Gas, dem Syngas, konvertiert wird. An dieser Stelle sei bereits darauf hingewiesen, dass durch Beimischung der Kohle mit nicht unerheblichen Mengen an Ersatzbrennstoffen, wie z. B. Biomasse oder Petrolkoks, die Betriebskosten von Anlagen basierend auf Feststoffvergasung zusätzlich gesenkt werden können. Im Anschluß an die Feststoffvergasung erfolgt in weiteren Prozeßschritten, auf Basis kommerziell erprobter Technologien, die in dem vorliegendem Beitrag noch detaillierter vorgestellt werden, die Umwandlung in hochwertige Produkte.

Aktuelle Kostenschätzungen für die zur Zeit insbesondere in den USA diskutierten Projekte belegen, dass die erwarteten Investitionskosten für IGCC-Anlagen sich im Bereich von 1 bis 2 Milliarden Euro belaufen werden, bei Koppelproduktionsanlagen hingegen mit 2 bis 3 Milliarden Euro gerechnet wird. Trotz dieser hohen Summen handelt es sich in beiden Fällen, unter Berücksichtigung aller projektspezifischen Gesichtspunkte, um profitable und wettbewerbsfähige Anlagentypen.

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Introduction

Soaring prices for the primary fuels oil and gas are currently driving demand for alternative energy sources with the fundamental aim of limiting dependency on expensive energy imports. At the same time, thought is being given on a national level as to how far the utilisation of domestically available energy resources, which has in the past lost appeal, can now be re-intensified.

Throughout the world domestic coal is now once again gaining in importance both economically and politically. Examples in the USA and China prove this without a doubt. Starting with the fossil fuel, coal – especially coal of a low quality, i.e. with a high ash or sulphur content – can be subjected to a number of process steps to obtain a variety of end products, such as fuels, chemicals and electricity. The end products selected for production largely depend on the market to be supplied and should thus be considered separately for each plant. In this respect, IGCCs, which are purely coal-fired power generation plants and do not generate any other end products, are a special case. However, the first main process step in both co-production plants for the production of the co-products mentioned and IGCCs is always solids gasification, in which coal is converted into a synthetic gas – syngas. Here, it should perhaps already be mentioned that the operating costs of plants based on solids gasification can be further reduced by adding significant quantities of substitute fuels, such as biomass or petroleum coke, to the coal. Once the solids have been gasified, their conversion into high-quality products takes place in further process steps based on commercially-proven technologies, which will be described in greater detail in this article.

Current cost estimates for projects currently being discussed especially in the U.S.A. show that investment costs for IGCC plants are expected to be in the range of EUR 1 to 2 billion, whereas those for co-production plants are estimated to be EUR 2 to 3 billion. When all project-specific aspects are taken into account, both alternatives are, despite these high sums, profitable, competitive plant types.

After briefly reviewing the history of coal liquefaction, this article presents the most active markets today.

In addition, some of the market factors impacting on gasification, coal-to-liquids (CTL) and coal-to-gasoline (CTG) are examined.

Subsequently, the article describes and compares a number of CTL/CTG technologies and their main process stages and outlines the current situation from the licensors' side.

CTL/CTG Roots and Current Demand

Coal-to-liquids (CTL)

In 1925, the German scientists *Fischer* and *Tropsch* developed and patented the F-T synthesis process for the conversion of synthesis gas to intermediate wax products, which are in turn converted into high-value products, such as diesel, naphtha and kerosene, by means of hydro-cracking in the subsequent product work-up unit.

In the 1940s the F-T synthesis process already supplied about 50 % of Germany's fuels based on coal gasification and has since then been further commercialised in South Africa, where Sasol now generates 175,000 barrels per day of F-T fuels in their CTL facilities based on coal gasification. This technology was also developed in and supplied from Germany.

Methanol-to-gasoline (MTG)/ Coal-to-gasoline (CTG)

The mechanism of methanol to gasoline synthesis by shape selective zeolite catalysis was discovered in the early 1970s. Partly as a result of the oil price crisis and subsequent search for alternative gasoline production routes, the process was further developed to the pilot plant stage (capacity < 4 bbl/day) during this decade. In the late 1970s and early 1980s a 100 bpd demonstration plant was developed and built at UK Wesseling/Germany. It was built and operated by Mobil (now ExxonMobil) Research & Engineering, Uhde GmbH and Union Rheinische Braunkohlen Kraftstoff AG in a joint project under sponsorship of the DOE and the German BMFT. This pilot plant successfully demonstrated the MTG as well as the Methanol to

Olefins (MTO) applications. A commercial-scale CTG study was carried out based on the results of that plant; however no plants of this type have yet been built. Instead, a commercial-scale MTG plant with a capacity of 14,500 bbl/day based on natural gas was built in New Zealand in the early 1980s as part of the New Zealand government's strategic energy supply plan. This plant, which was 75 % owned by the NZ government and 25 % by Mobil, was started up in 1985 and operated successfully for approximately ten years before it was sold to Methanex and converted to the production of technical-grade methanol. Throughout the 1990s, the spotlight turned away from MTG processing due to the prevailing low price for crude oil. However, events in the early years of the new millennium have led to a sharp and apparently long-term increase in crude oil prices and attention has once again turned to MTG/ CTG.

New Wave of Gas-to-liquids (GTL)

Currently there is a large number of GTL projects in the Middle East, where a multitude of licensors are at work. The Qatar Petroleum and Sasol project with a capacity of 33,700 bbl/d is scheduled to start operation this year. The products will be fuels and naphtha. ExxonMobil is currently planning a 100,000 bbl/d plant for the production of specialties and lubes in Qatar. Shell is already realising a 2 · 70,000 bbl/d GTL plant in Qatar. Furthermore, Invanhoe Energy as well as Qatar Petroleum along with Conoco Phillips are aiming to build plants for fuels in Qatar with respective capacities of 185,000 bbl/d and 120,000 bbl/d. Others, mainly with capacities below 75,000 bbl/d, will also be built.

New Wave of Coal-to-liquids (CTL)

The most lucrative and prosperous market is to be found in the U.S.A., where political will and industrial strength are jointly moving towards clean coal technologies. The Energy Policy Act of 2005, (EP Act 2005) signed by *President Bush*, authorises a $ 1.6 billion allocation for research and development funding of clean-coal initiatives, including the production of ultra-clean fuels. In addition, the legislation provides for loan guarantees associated with the construction of commercial-scale CTL plants.

The second piece of legislation, entitled the Safe, Accountable, Flexible, Efficient Transportation Equity Act of 2005 (SAFETEA), also signed by *President Bush*, provides a 50 cent-per-gallon excise tax credit for alternative fuels. This includes ultra-clean FT fuels made from coal.

It is expected that the first CTL plant in the USA to benefit from the new legislation will be the WMPI “Waste Coal to Clean Liquids Fuels Project”, a demonstration plant in Gilberton, Pennsylvania. The WMPI project utilises waste coal containing 40 % ash. This high-ash-containing feedstock requires a gasification process with high operating flexibility, a high conversion rate and a versatile coal feeding and slag handling system. The utilisation of low-value coal wastes is economically advantageous compared with traditional commercially-available but higher-priced coal. The plant will convert the abundant waste coal resources scattered across the north-eastern part of the U.S.A. Anthracite waste (culm) and bituminous waste (gob) have been accumulating in the State of Pennsylvania, U.S.A., for centuries. These materials are rock and coal that contain various amounts of carbon material once the chunks of saleable coal have been separated out. Over one billion tonnes of waste coal has been piled up. The plant is expected to produce 5,038 barrels per day of ultra-clean fuels and approximately 50 MW_e of power. The net efficiency calculation has to take into account the multiple products, but based on total energy input divided by the total usable energy output, the estimated net efficiency is about 42 %. The project is sponsored by the US Department of Energy (DOE) and National Energy Technology Laboratory under the Clean Coal Power Initiative (CCPI). A potential off-taker of CTL fuels is the US Department of Defence.

According to Energy Information Agency data, the U.S.A. is the largest coal reserve holder in the world. If only 5 % were utilised to produce ultra-clean transportation fuels, it would equal to the country's oil reserves.

While the U.S. market depends largely on diesel, China depends on gasoline. Taking into account the soaring demand for energy in China during the past few years, it can be foreseen that synthetic fuels derived from coal will very soon play a major role in China's energy policy.

Key Market Factors for CTL/CTG

Oil and Gas Prices

Soaring prices for oil and gas, currently above $ 75.00 per barrel for crude oil and more than $ 8.00 per million BTUs with peaks close to $ 15.00 per million BTUs, are urging policy makers and industry to seriously look for more stable alternatives and to minimise dependency on unpredictable primary energy supplies.

China's Economic Growth

A big market factor with respect to the rising interest in CTL/CTG is the continuing economic boom in China, which reached an annual growth of 9.4 % in 2004. This growth is supported by an ever-increasing acceleration of industrialisation and urbanisation and has sparked a huge and growing thirst for primary energy sources. The civil transport sector alone has grown by more than 11 % per year in the period from 1995 to 2004 and reached the mark of 27.4 million privately-registered cars in 2004. In addition, approximately 9.3 million commercial vehicles were registered in China in 2003, representing an average annual growth of 9.6 % over the first three years of the new millennium.

There are no indications that these enormous growth rates will slow down, and this will continue to increase energy prices and therefore make alternative fuel production like CTL and CTG increasingly attractive.

Oil/Coal Reserves

Global oil consumption is roughly 85 million bbl/day, and with a projected annual growth of more than 3 %, this will lead to a projected consumption of approximately 118.6 million bbl/day in 2020 (Source US Gov't DOE – International Energy Outlook). 48 % of the world's oil is supplied from 116 fields, of which 87 % were discovered prior to 1980. Only approximately 16 % of the oil fields discovered since the 1980s are fields which supply in excess of 200,000 bbl/day. Big oil-consuming countries like the U.S.A., India or China are developing increasingly large deficits in crude oil supply. The U.S.A. alone is projected to develop an oil deficit of 15.7 million bbl/day in 2020.

There are many indications that oil supply will soon be unable to cover the world's energy demands. However, the world has proven coal reserves in the order of 1,100 billion tonnes. Countries like China, the U.S.A. and also European countries rely heavily on coal for energy production. Global coal consumption rates are currently in the range of only 5 billion tonnes per year (of which 1.96 alone is in China), meaning that the proven coal reserves will by far outlast crude oil reserves. Therefore, CTL and CTG are also safe options in providing sustainable routes for future fuel supply.

Technological Advances

Growing commercial experience has led to many technological advances in the field of coal-to-products processing, which have on the one hand significantly strengthened the reputation of coal liquefaction technologies and on the other led to continuously decreasing investment cost with continuously increasing capacities, and thus to increasingly feasible plant concepts.

The development and commercial demonstration of modern entrained-flow gasification technologies allow single gasifier capacities of up to more than 5,000 tonnes per day

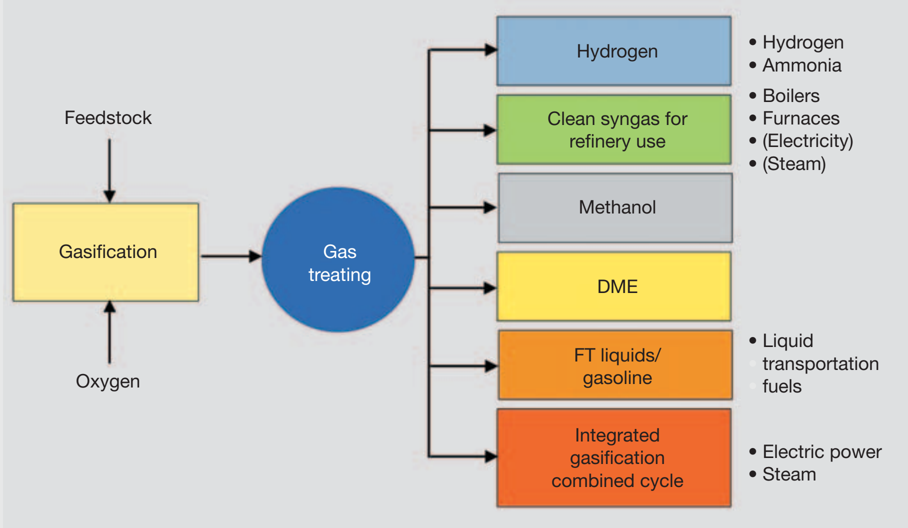

Figure 1. Gasification Applications.

of coal feed. Experience and evaluations of lessons learnt from the commercial plants have led to modern gasifier designs that are both highly efficient and robust and at the same time display high availability.

In Fischer-Tropsch synthesis, the development of slurry-phase reactors as well as the development of efficient catalyst systems have also allowed a continuous increase in single-train capacities of up to more than 15,000 bbl/day and therefore a continuous decrease in specific investment. Further development now focuses on narrowing the product spectrum produced by F-T synthesis to reduce the cost for required product work-up. In methanol synthesis units, single-train capacities of more than 5,000 tonnes per day are now possible.

As a result of all these developments, it is now possible to build modern CTL/CTG plants with large single-train capacities and highly-efficient, robust designs, thereby exploiting economies of scale to the full.

Description of CTL/CTG

Technical Background to Indirect Coal Liquefaction

In general, coal can be used directly or indirectly for the production of chemical products. However, direct use (or direct liquefaction), which is the hydrogenation of coal under very high pressures, now only attracts interest in China due to the fact that products resulting from direct coal liquefaction are highly aromatic and therefore difficult to use as high-quality synthetic fuel products.

The indirect use of coal for the production of chemicals (coal to products) involves as a first step the gasification of coal to produce a raw synthesis gas (syngas) stream containing mainly carbon monoxide (CO) and hydrogen (H_2). This syngas is then further treated to produce the syngas quality required for the downstream synthesis step. Such gas treatment generally involves absorption processes to remove unwanted components such as sulphur components (H_2S, COS) and also a CO shift step to adjust the molar H_2:CO ratio required for the synthesis.

With this alignment, the indirect use of coal is very flexible and can be used to produce a wide range of products, such as electrical power, clean hydrogen, chemical products like ammonia, methanol, dimethylether (DME) or ultra-clean synthetic fuel products (Figure 1). Plant design depends on the desired product application in the specific market. Plants can be designed for the co-production of several products to allow operating modes to be adjusted according to market requirements and thus achieve maximum return under the specific market conditions.

There are two major routes in the production of ultra-clean, high-quality synthetic fuel products, as shown in Figure 2. The so-called CTL route describes the gasification of coal and subsequent gas treatment, followed by Fischer-Tropsch synthesis and further product-work up of the raw Fischer-Tropsch product. This route is mainly used to produce ultra-clean diesel. The CTG route describes the use of the clean syngas for the production of methanol and the further conversion of methanol to produce gasoline in a MTG conversion step. This route is mainly used to produce a high-quality synthetic gasoline product.

Both routes described are essentially three-step processes. The chosen process route again depends mainly on the product application in the specific market.

Due to the fact that very high investment costs are involved in the construction of these large complexes and the main feed (coal) is relatively cheap, their feasibility is mainly dependent on exploiting the economies of scale. Therefore, world-scale CTL plants today have capacities of 30,000 bbl/day and more, while world-scale CTG plants have capacities of 1,000,000 mtpa and more. In both cases, multiple parallel gasifiers are applied and also multiple parallel synthesis trains (for F-T synthesis and methanol respectively).

Both routes are now briefly described below. Details of the main processing steps are described in later chapters.

Coal-to-liquids (CTL)

In a CTL complex with F-T, as shown in simplified form in Figure 3, coal feed is prepared in the milling and drying section and then sent to the gasifier, where it is fully converted to raw synthesis gas. Oxygen for gasification is supplied by an air separation unit (ASU). A part of the raw synthesis gas is treated in the CO shift to convert the steam and CO to hydrogen and CO_2, after which this treated part is re-mixed with the untreated portion. Split between the portions is controlled such that the required H_2:CO ratio for the F-T synthesis is always met. The shifted syngas is then submitted to an acid gas removal step, where COS, H_2S and CO_2 are removed to produce clean syngas feed for the F-T synthesis unit. To sufficiently protect the F-T catalyst from contaminants, a Rectisol Absorption process is typically applied for the acid gas removal as this process is commercially proven in CTL applications.

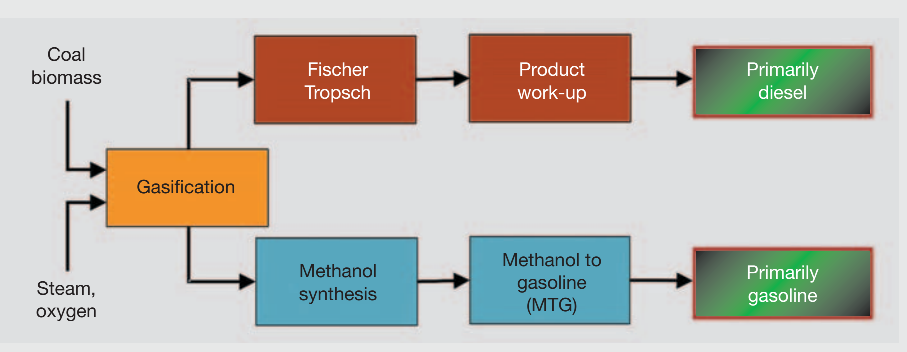

Figure 2. Block Flow Diagram – Routes to Synthetic Transportation Fuels.

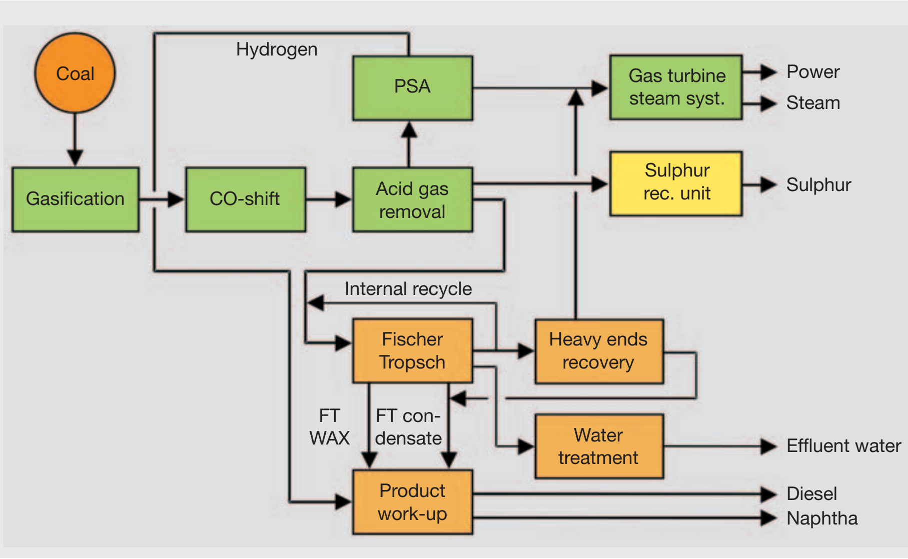

Figure 3. Block Flow Diagram – Coal-To-Liquids.

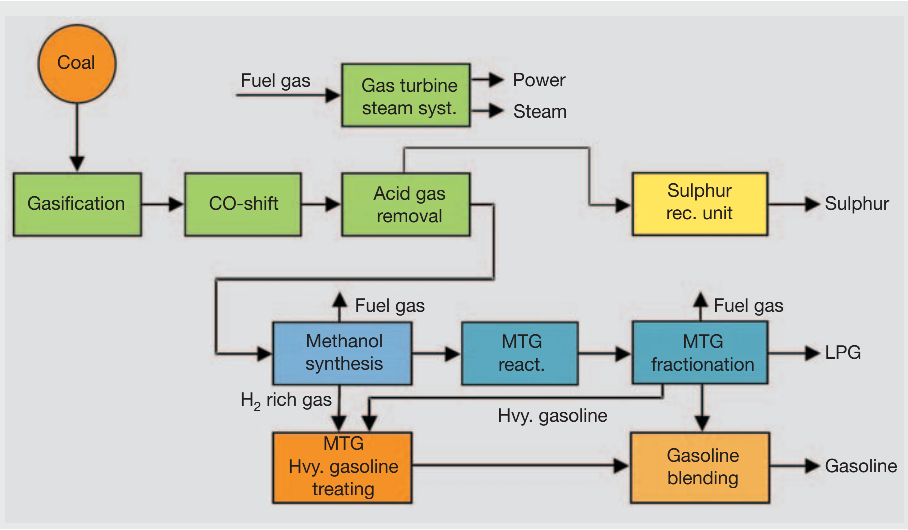

Figure 4. Block Flow Diagram – Coal-To-Gasoline.

A small fraction of the clean syngas is sent to a pressure swing adsorption unit (PSA) for the production of pure hydrogen, which is required in the product work-up section and the major part of the syngas is routed to the F-T plant.

In the F-T synthesis step the syngas is converted to the raw F-T product. The light ends of the raw F-T product are partially recycled internally and sent to the heavy ends recovery unit. The tail gas from the heavy ends recovery unit is sent to a combined cycle plant to produce sufficient power for the entire CTL complex.

The F-T products, namely F-T waxes and F-T condensate, are sent to the product section, where they are hydro-treated with hydrogen from the PSA to produce primarily diesel product.

Not only hydrocarbons but also a significant amount of water is produced during F-T synthesis. This water fraction contains impurities such as oxygenates, which have to be removed in a water treatment step.

Sour gas (H_2S, COS) removed in the acid gas removal unit is sent to a sulphur recovery unit (Claus plant), where it is converted into elementary sulphur. The tail gas from the Claus plant is hydrogenated and afterwards recycled to the acid gas removal unit.

The yield of F-T products in a fully self-sufficient CTL plant based on hard coal gasification is approximately 0.25 tonnes of F-T product/ton of coal. The sulphur produced depends on the sulphur concentration in the coal.

Coal-to-gasoline (CTG)

In a CTG complex with MTG, as shown in simplified form in Figure 4, the clean syngas from the acid gas removal unit is sent to a methanol synthesis loop, where it is converted to raw methanol. The purge gas stream from the methanol synthesis loop is sent to a hydrogen recovery unit to produce the clean hydrogen required in the heavy gasoline treatment step of the MTG plant.

The raw methanol is sent to the MTG plant. Here, it is reacted to gasoline in the reaction section and the raw product sent to the fractionation section, where it is fractionated into a minor amount of fuel gas, an LPG by-product and gasoline. The gasoline product is separated into a light gasoline and a heavy gasoline fraction. The heavy gasoline is sent to a heavy gasoline treatment step, where it is hydro-treated and stabilised. It is then blended with the light gasoline to obtain the final gasoline product.

In addition to the hydrocarbons, a large amount of water is produced in the MTG, which however can be reused inside the CTG complex to produce the hydrogen required for the methanol synthesis.

As in the CTL plant, the sour gas (H_2S, COS) removed in the acid gas removal unit is sent to a sulphur recovery unit (Claus plant), where it is converted to elementary sulphur. The tail gas from the Claus plant is hydrogenated and afterwards recycled to the acid gas removal unit.

The yield of gasoline in a fully self-sufficient CTG plant based on hard coal gasification is approximately 0.30 tonnes of gasoline/ton of coal. The sulphur produced depends on the sulphur content in the coal.

Major Process Steps

Gasification

Gasification is the partial combustion of carbon products, such as coal, in the presence of a moderator (steam, carbon dioxide (CO_2)) to produce raw syngas.

$$2\,(CH_2) + O_2 \Rightarrow 2\,CO + H_2$$

Contrary to complete combustion, gasification takes place in a reducing atmosphere. It can be performed with a wide range of feedstocks, ranging from gaseous feeds such as refinery gases, natural gas or LPG, and liquid feeds, such as various oil fractions, heavy residues or orimulsions, to solid feeds, such as coal, petroleum coke, biomass or even waste products.

Different coal gasification technologies have been developed from the initial laboratory test stage to full commercial scale in the past century and a number of state-of-the-art technologies which have been fully proven on a commercial scale are now available for the execution of CTL and CTG projects. Some examples of commercial-scale coal gasification

Table 1. Gasification systems.

	Fixed Bed	Fluidized Bed	Entrained Flow
Gasification of all types of coal	NO	YES	YES
Raw gas temperature (outlet gasifier)	~450 °C	~800 °C	~1400 °C
By-product formation	YES	YES	NO
Carbon conversion	>99 %	~94 %	>99 %
Licensors/technologies (selection)	British Gas Lurgi (BGL) Sasol Lurgi	RWE (HTW)	Shell (SCGP) GE (Texaco) Uhde (PRENFLO) Sustech (GSP) ConocoPhillips (E-Gas)

Table 2. Fischer-Tropsch and MTG products.

Fischer-Tropsch Products	MTG Products
Ultra clean fuels (Naphtha, Diesel, Jet) – high cetane (> 70) – free of sulphur (< 1 ppm) – free of aromatics (< 1 ppm) and heavy metals – paraffin content > 99 %	**Gasoline (main product, typical specification)** – according to EURO III regular – RON 92, MON 82 – free of sulphur and nitrogen impurities – RVP approx. 8 psi – Density approx. 730 kg/m^3 – Total Aromatics < 35 wt. %
Specialties – lube oils – paraffins (e.g. for detergents, solvents, etc.) – paraffin waxes (e.g. for candles, colours, pharmaceuticals, etc)	

plants are the Puertollano IGCC (318 MW$_e$), which is based on the PRENFLO gasification process developed and owned by Uhde, and the Buggenum IGCC operated by NUON (253 MW$_e$), which is based on the Shell Coal Gasification Process (SCGP).

In general, three different gasification technologies can be distinguished and are commercially available (refer to Table 1), namely:

- fixed-bed gasification,
- fluidised-bed gasification,
- entrained-flow gasification.

The most effective technology is the entrained-flow gasification process as it converts almost all the carbon contained in the feed and there are no by-products, such as methane, tars or heavy aromatics. Also, it is operationally flexible, i.e. different feedstocks and almost all types of coal can be used. The raw syngas produced has a constant quality irrespective of the feedstock used and contains very little CO$_2$.

Fixed-bed and fluidised-bed gasification technologies are more restricted in their choice of feedstock and also produce by-products such as methane, aromatics and tars, which reduce process efficiencies and also require additional cost-intensive treatment to remove the side products produced.

A brief description of a dry-feed entrained-flow gasification process, such as the SCGP, PRENFLO or Sustech (GSP gasifier) processes, now follows.

Coal feed is milled and dried to produce coal powder with particle sizes in the range of approximately 10 to 100 microns. This powder is then pressurised with nitrogen gas in a sluice system and dry-fed together with high-pressure oxygen supplied from the ASU and steam to the gasifier burners, where it is gasified to syngas at temperatures of around 1,600 °C. The gasifier vessel itself has a cylindrical (membrane wall) reactor cage. High-pressure boiler feedwater is directed through the tubes of the membrane wall, thus cooling it through steam production. Inside the gasifier, a solid slag layer forms which protects the membrane wall. The molten slag on the inner side of the slag layer flows downwards through a slag tap in the conical bottom of the reactor cage into a water bath, where it solidifies and scatters into small granules. This arrangement ensures simple, effective separation of the hot syngas and the slag and ash.

The slag layer has a self-repairing effect as cold spots form at any point where solid slag breaks out of the layer and slag immediately attaches thereto, solidifying and repairing the damage.

The syngas produced is withdrawn either at the top (SCGP, PRENFLO) or at the bottom (Sustech) of the gasifier and cooled by quench gas and subsequent steam production or liquid quench. The raw syngas is directed to a hot filter, where fly ash is removed. Subsequently, the gas is sent to a wet scrubbing unit to remove hydrochloric and hydrofluoric acid as well as trace solids. The water saturated syngas then leaves the wet scrubbing unit and is sent to the gas treatment unit.

Waste water produced in the various sections of the gasification process is treated in the sour water stripper to remove gases dissolved therein such as H$_2$S, NH$_3$, CO$_2$ and HCN. The water is further treated in a water clarification section to remove solids, which are subsequently thickened in a vacuum belt filter press and recycled back to the gasifier.

Fischer-Tropsch Synthesis (F-T)

F-T synthesis can be divided into two major technologies: high-temperature Fischer-Tropsch (HTFS) and low-temperature Fischer-Tropsch (LTFS).

HTFS takes place at temperatures of approximately 300 to 330 °C and produces predominantly naphtha and olefins. It is used commercially by Sasol in its so-called Sasol Advanced Synthol (SAS) process in Secunda, South Africa, where a significant portion of chemical products such as ethylene, alpha olefins, solvents, ammonia, phenolics, etc., are produced.

Today, however, LTFS is the major focus of attention in all GTL and CTL projects in the world. LTFS takes place at temperatures of approximately 200 to 230 °C. The raw product produced contains a wide spectrum of mainly paraffinic and waxy products and is then further hydro-treated in the product work-up section to produce an ultra-clean synthetic diesel product, as shown in Table 2. This product has a very high cetane number in excess of 70 and is virtually free of sulphur, aromatics and nitrogen. Engine tests have shown that use of this diesel product produces significantly less particulate, NO$_x$, CO and hydrocarbons than conventional diesel.

Blends of conventional diesel with minor amounts of ultra-clean synthetic diesel are successfully marketed, e. g. by Shell.

Due to a considerable rise in market interest in F-T technology, many companies have developed proprietary Fischer-Tropsch technology (refer to Table 3). These companies can be split into two major groups:

- major oil and chemical companies, such as Sasol, Shell, ExxonMobil, BP, ConocoPhillips etc., who are mainly interested in using their technology for their own projects and who are, to say the least, very restrictive in licensing their technology,
- technology licensors, such as Syntroleum and Rentech, who actively pursue the licensing of their technology worldwide to any potential client.

All F-T technologies can be classified into three different groups depending on the type of reactor and catalyst applied:

Table 3. Potential Fischer-Tropsch licensors.

Licensor	Commercial References – gas based – coal based	Reactor System	Catalyst
SASOL	Yes (67,000 b/d)[1] Yes (150,000 b/d)	fixed bed/slurry phase fluidised bed	iron/cobalt
SHELL	Yes (12,500 b/d)[2] No	fixed bed	cobalt
CONOCO PHILLIPS	No No	slurry phase	cobalt
RENTECH	No No	slurry phase	iron
SYNTROLEUM	No No	fixed bed	cobalt
BP	No No	fixed bed	cobalt
STATOIL	No No	slurry phase	cobalt
EXXON MOBIL	No No	slurry phase	cobalt
ENI	No No	slurry phase	cobalt

[1] includes the ORYX I GTL plant in Qatar (34,000 b/d)
[2] does not include the Pearl GTL project in Qatar (2 x 70,000 b/d)

- slurry-phase: in this technology the syngas bubbles through a slurry phase consisting of catalyst and molten wax produced by the process itself,
- fixed-bed: here the syngas flows through tubes containing the catalyst,
- fluidised-bed: here the syngas passes through the catalyst, thereby fluidising the catalyst bed and forming an isothermal reaction zone.

All three types have been commercially applied in the past. Nowadays, most companies which supply LTFS technology apply slurry-phase reactors as this technology enables very large single-reactor capacities to be achieved. Catalyst regeneration is continuous and product distribution is more uniform than in fixed-bed reactors, which have discontinuous catalyst regeneration and allow less throughput. The major advantage of the fixed-bed system as compared to the slurry-phase system is the easier scale-up of the reactors.

With respect to catalysts, there are now two major types used for F-T: iron and cobalt catalysts (Table 3). The iron catalyst is significantly cheaper, and is more flexible and robust with respect to the quality of the syngas feed, as it can use syngas H_2:CO ratios of 0.7:1 up to 2:1, whereas the cobalt catalyst uses a H_2:CO ratio of 2:1. However, the iron catalyst has a much shorter lifetime and produces heavier molecular weights products and more olefinic products compared to the cobalt catalyst.

Therefore, cobalt catalysts are used in most of today's GTL plants as the cobalt catalyst is much more efficient. For applications based on coal-derived syngas, however, only the iron catalyst has so far been commercially applied. With this system an overall CTL capacity of more than 150,000 bbl/day of F-T-products is in commercial operation. From that perspective, it is currently only commercially viable to consider technologies using iron catalysts for CTL projects.

However, companies such as Syntroleum and others are pushing to establish their cobalt catalyst-based technology for application in CTL. As announced in November, 2005, Syntroleum started a testing programme for this purpose in January 2006 with the aim of demonstrating beyond doubt the applicability of their GTL cobalt catalyst for CTL application. This programme is supposed to go on for six months.

A brief description of LTFS based on the slurry-phase process (e.g. Sasol SSPD or Rentech) now follows.

The clean syngas is sent to the slurry-phase reactor, where it is bubbled through a slurry phase consisting of catalyst and molten wax product, which is produced in the reactor itself. The gases are withdrawn at the top of the reactor and partially recycled to the feed side after separation from the water produced in the process. The remaining gas stream is sent to the heavy ends recovery section, which is a low-temperature separation section to recover condensate from the tail gas, which is used as fuel gas. Recovered condensates are sent to the product work-up section.

The waxy product is withdrawn at the side of the reactor and the wax is continuously separated from the catalyst. The waxy product is then sent to the product work-up section, which consists of a hydro-cracker, a hydro-treater and a distillation section. The wax product is sent to a hydro-cracker to produce the diesel product, while olefins in the condensate are hydro-genated to paraffins in the hydro-treater. The hydro-cracking step can be adjusted to maximise the production of ultra-clean diesel product. The products from the hydro-treater and hydro-cracker are then fractionated into diesel and naphtha products. Approximately 80 % of the product is diesel and the remaining portion is naphtha.

Methanol-to-gasoline (MTG)

MTG is the conversion of raw methanol over a zeolite catalyst system for the production of a gasoline product. The gasoline produced complies with the EURO 3 unleaded regular gasoline quality standard without the need for additives such as MTBE or ETBE and is therefore of significantly higher quality than standard refinery naphtha products (raw gasoline). The product is virtually free of sulphur, and also in full compliance with current and forthcoming stringent European regulations for gasoline with respect to olefins, benzene and total aromatics.

Two different MTG process options have been developed: fluidised-bed and fixed-bed technology.

The fixed-bed MTG technology licensed by ExxonMobil has been launched commercially and successfully demonstrated in a 14,500 bbl/day MTG plant in New Zealand. The direct gasoline yield from the MTG reaction is higher and scale-up is significantly easier in the fixed-bed technology compared to the fluidised-bed technology. An LPG by-product is produced. The gasoline yield from the methanol feed in fixed-bed processing is approximately 37 to 38 wt.% and the yield of LPG by-product approximately 3.7 wt.%. Water production is approximately 58 wt.% with the remaining fraction being fuel gas.

In the fluidised-bed technology a product containing a higher amount of olefins and less aromatics is produced than with the fixed-bed system. The fluidised-bed technology was demonstrated in a 100 bpd demo plant at UK Wesseling, Germany in the early 1980s as described above. An alkylation unit is required for this technology in order to boost the gasoline yield by converting light olefin products to high RON. The combined gasoline yield from fluidised-bed MTG and alklyation is slightly higher than the gasoline yield from fixed-bed MTG and a slightly higher average RON is also achieved.

MTG technology is in principle flexible and can be adjusted to MTO operation, where the olefins can be further used for the production of synthetic diesel or jet fuel by oligomerisation over a ZSM-5 catalyst. This process chain has also been developed by Mobil as the Mobil Olefin to Gasoline/Distillate (MOGD) process.

Besides ExxonMobil, several other companies have worked on MTG processing. Haldor Topsoe has developed the so-called TIGAS (Topsoe integrated gasoline synthesis) process, which integrates the MTG synthesis with methanol synthesis in a single process loop. Lurgi AG, Germany, has developed the Methanol to Synfuels (MTS) process, which is in principle similar to the MOGD process. This process produces gasoline (RON 80) and diesel (Cetane ~ 55) in the ratio of approximately 1:4 and is therefore, with regard to the product spectrum, more comparable to F-T than to MTG.

A brief description of a fixed-bed MTG process follows.

Raw methanol from the methanol plant is vaporised and preheated before being sent to a DME reactor, where it is partially dehydrated to DME. The partially converted gas is then sent to the MTG reactors. Multiple parallel reactors are installed to allow for regeneration of the MTG zeolite catalyst while maintaining a constant product flow. The raw product is cooled in a heat recovery step and separated into three phases, namely an unconverted gas phase which is recycled to the reactors, a water phase which is sent for reuse within the CTG plant and a hydrocarbon product phase, which is sent to the distillation section. In the distillation section the C2 fraction, which is to be used as fuel gas, is separated in a de-ethaniser column followed by separation of LPG-by-product and gasoline product in a stabiliser. The gasoline is then sent to a gasoline splitter column, where it is separated into a light and a heavy gasoline fraction. The heavy gasoline fraction is sent to a hydro-isomerisation reactor and subsequently further stabilised. After treatment, the heavy gasoline and light gasoline are blended into the final gasoline product.

Comparison of CTL and CTG

CTL and CTG are the two major routes for indirect coal liquefaction for the production of high quality synthetic fuels. Both are essentially three-step processes, with each step having been fully demonstrated on a commercial scale and the design advocated by experienced process licensors.

The application of one or the other route depends mainly on the market application of the product. CTL is in the spotlight due to significant growth in the diesel market and the increasing stringency of environmental regulations for fuel products, which has opened up a large market potential for ultra-clean synthetic F-T diesel. CTG, on the other hand, is of major interest in gasoline-dominated markets such as China, especially in remote locations, where CTG can supply high-quality gasoline direct to the local market. Another very interesting option in CTG is the possibility for flexible production (co-production) of methanol or gasoline, thus allowing optimised adjustment of plant operation to the prevailing market environment.

Therefore, both technologies are viable alternatives in efficiently utilising coal reserves, especially low-quality or waste coals, for the production of high-quality products. The large amount of projects for indirect coal liquefaction in the U.S.A., China, Australia and potentially also in Europe, will eventually lead to large and modern production plants and a new era of efficient coal utilisation.

Outlook

The driving forces behind the desire to implement coal-based technologies to substitute the primary energy carriers oil and gas are currently:

- high prices for oil and gas,
- stringent environmental requirements, especially in the USA,
- continuing economic boom in China,
- long-term coal reserves,
- availability of proven technology for coal gasification,
- increased demand from potential plant owners and operators,
- increasing interest from the investment community.

The following assets can be considered as an added value for any future IGCC plant:

- vast operational experience from commercial IGCC plants, i.e. elimination of main problems through lessons learnt, plant availability of 83 %+ has been achieved,
- IGCC is very attractive due to environmental issues, fuel flexibility (biomass, hard coal, lignite) and co-production,
- development of the gasifier capacity is a key factor in the profitability of IGCC (economy of scale), which leads to commercial credibility.

Finally, the commercialisation of gasification/CTL projects will bring substantial socio-economic and environmental benefits, especially to coal regions, including

- economic benefits through re-energisation of the coal production industry, the creation of high-quality jobs, an improvement in job security and productivity, the revitalisation of communities in coal-producing regions and the diversification of domestic sources of energy,
- environmental benefits through the production of e.g. F-T liquids, which are clean-burning fuels superior in quality to their petroleum-based counterparts. They are essentially free of sulphur and nitrogen, and their use as transportation fuels or as feedstock in the production of chemicals (naphtha steam reforming for olefin production) would help in reducing overall greenhouse gas emissions. All plants can be optionally designed to capture CO_2.

Uhde is currently being involved in several CTL projects in the USA and is performing feasibility studies for CTG plants in China. Based on the experience gained on these two particular markets, Uhde is, together with key partners from various industrial fields, such as coal, oil, power and chemicals, at present developing a major CTL plant in Europe based on the solids gasification.

Abbreviations

ASU	Air Separation Unit
bbl	barrels
bpd	barrels per day
BMFT	Bundesministerium für Forschung und Technologie (Federal Ministry of Research and Technology)
BTL	Biomass-to-liquids
BTU	British Thermal Unit
CCPI	Clean Coal Power Initiative
CTG	Coal-to-gasoline
CTL	Coal-to-liquids
DME	Dimethylether
DOD	US Department of Defense
DOE	US Department of Energy
ETBE	Ethyl tert butyl ether
F-T	Fischer-Tropsch Process
GSP	Gasifier according to Gaskombinat Schwarze Pumpe
GTL	Gas-to-liquids
HTFT	High-temperature Fischer-Tropsch
IGCC	Integrated Gasification Combined Cycle Power Plant
LPG	Liquefied Petroleum Gas (mostly commercial propane and commercial butane)
LTFT	Low Temperature Fischer-Tropsch
MOGD	Mobil Olefin to Gasoline/Distillate
MTBE	Methyl tert-butyl ether
MTG	Methanol-to-gasoline
MTO	Methanol-to-olefins
MTPA	Metric tons per year
MTS	Methanol-to-synfuels
PRENFLO	Pressurised Entrained-flow Gasification Process (Uhde)
PSA	Pressure Swing Adsorption
RON	Research Octane Number
SAS	Sasol Advanced Synthol
SCGS	Shell Coal Gasification Process
SGP	Shell Gasification Process
SSPD	Sasol's Proprietary Slurry-phase Distillate
stpd	Short tonnes per day
TIGAS	Topsoe Integrated Gasoline Synthesis ☐