COAL TO CLEAN GASOLINE

Xinjin Zhao, Ron D. McGihon and Samuel A. Tabak, ExxonMobil Research & Engineering Company, USA, discuss ExxonMobil's methanol to gasoline (MTG) technology for the production of clean gasoline from coal.

The recent upsurge in oil prices has spurred renewed worldwide interests in different energy resources. Coal, which has been the subject of much attention because of the many environmental concerns resulting from its sulfur and ash content and significant carbon footprint, is expected to play a key role in the rapidly growing economy in countries such as China, India, and even the US, in the coming decades. It is clear that the world will have to rely on more efficient and clean coal technology. The most probable option is to convert coal into high quality, clean burning transportation fuel.

There are two commercially demonstrated routes for converting coal to transportation fuels through gasification (Figure 1). The widely known Fischer-Tropsch process was first discovered in the 1920s. It has been commercially practiced by Sasol in several different forms to produce fuels from either coal or natural gas. No commercial scale coal-to-liquid (CTL) plants based on Fischer-Tropsch chemistry have been built since the Sasol plants.

Although it is less known, there is another commercially proven alternative for converting coal-to-gasoline, through methanol.[1] ExxonMobil's methanol-to-gasoline (MTG) process efficiently converts crude methanol to high quality clean gasoline. When coupled with commercially proven coal gasification and methanol synthesis technology, MTG offers an effective route to premium transportation fuel from coal. Both coal gasification and methanol synthesis are commercially mature technologies with several commercially available technologies for both steps. Mobil discovered the MTG process in the 1970s and commercialised the technology in New Zealand in the mid 1980s. MTG gasoline is fully compatible with conventional refinery gasoline.[2] Due to its unique properties, methanol has been promoted as the energy carrier for the so-called Methanol Economy®[3] MTG gasoline can be either blended with conventional refinery gasoline or sold separately with minimal further processing. Technically, methanol sources for the MTG process could be from natural gas reforming, coal gasification, biomass conversion, or even purchased methanol in the market place at favourable economic conditions.

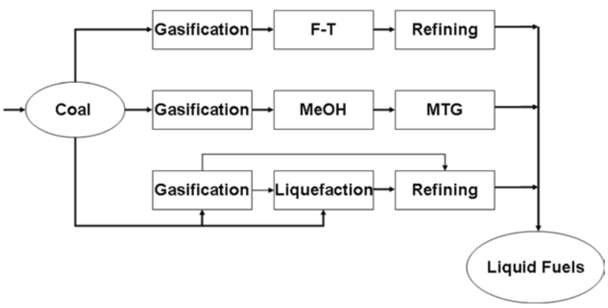

Figure 1. Alternative route for CTL.

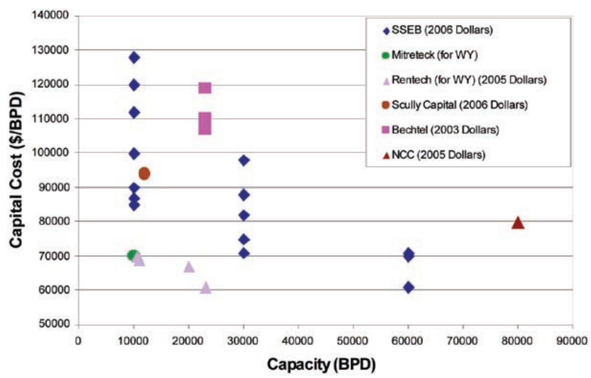

Figure 2. Capital investments for CTL projects.

A third option for coal conversion, direct coal liquefaction, is also attracting renewed attention due to the recent commercial plant being built by Shenhua in Inner Mongolia, a Chinese coal company. Although similar processes were demonstrated in the US at much smaller demonstration scales, no commercial plants were ever built or operated for direct coal liquefaction. It was reported that the four major operating coal liquefaction pilot plants in the US all experienced problems including severe equipment corrosion.[4] The commercial Shenhua plant will be a significant step in determining the viability of the direct liquefaction process route. Different from the two indirect routes, the direct liquefaction route does not go through a syngas step and thus the liquid products have to go through significant upgrading as well as cleanup for sulfur, nitrogen and other impurities.

Both the Fischer-Tropsch and MTG processes convert coal into synthesis gas before converting it to the final liquid products. However, their respective product slates are very different. The Fischer-Tropsch process produces a broad spectrum of straight chain paraffinic hydrocarbons that require upgrading to produce diesel fuel, lube feedstock and paraffinic naphtha for petrochemical applications. In contrast, MTG selectively converts methanol to one simple product: a very low sulfur, low benzene high quality gasoline. Due to the unique low sulfur and low benzene characteristics of the MTG gasoline product, it can be a valuable blending component for meeting environmental regulations specific to sulfur and benzene.

A recent surge in CTL activities has renewed market interest in MTG technology. The current MTG technology represents an advance beyond the technology commercialised in New Zealand in the mid 1980s. The improvements result from programmes undertaken by ExxonMobil in the 1990s that reduce both capital investment and operating expenses.[5] Detailed engineering design and construction of the first coal-to-gasoline process via MTG technology is under construction in China by Jincheng Anthracite Mining Co (JAM). The initial phase of the plant is designed for a capacity of 100 000 tpy, but is expected to expand to 1 million tpy for the second stage of the project. ExxonMobil recently also announced the first US CTL project based on MTG technology. DKRW Advanced Fuels LLC has licensed ExxonMobil's MTG technology through its subsidiary Medicine Bow Fuel and Power LLC for a 15 000 bpd CTL plant in Medicine Bow, Wyoming. Both the JAM and DKRW plants incorporate significant improvements beyond the original New Zealand plant and are based on over ten years of operational experience.

This article will provide an update of the recent development of MTG process and the recent commercial activities for the production of gasoline from coal. When

$$2CH_3OH \rightarrow CH_3OCH_3 + H_2O$$

$$\rightarrow \text{Light Olefin} + H_2O$$

$$\rightarrow \text{Higher Olefins} + \text{n/i Paraffins} + \text{Aromatics} + \text{Naphthenes}$$

Figure 3. Simplified methanol to gasoline chemistry.

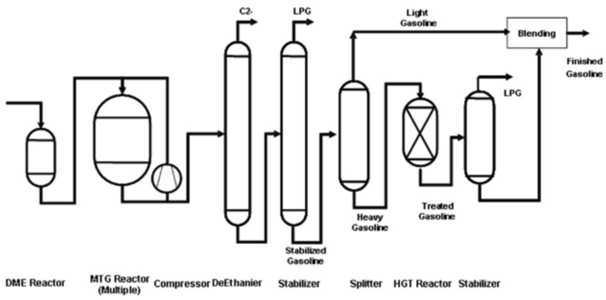

Figure 4. Schematic of MTG process.

appropriate, MTG will be evaluated against the Fischer-Tropsch process for converting coal-to-liquid fuel.

Coal-to-liquid economics

Economics of coal-to-liquid are very complex and difficult to accurately estimate. Part of the reason is that no CTL plants have been built worldwide since the 1980s while construction cost has significantly increased. Moreover, technology improvements to synfuel applications have also occurred in the interim. The capital cost of CTL is also strongly dependent on location, coal type, product outlet, CO_2 capture, and coal supplies. Furthermore, CTL projects in different countries can be affected by government policy and incentives. For example, the surge of CTL or chemicals projects in China is very much driven by energy security to satisfy the increasing energy demand for the growing Chinese economy. On the other hand, the CTL interests in the US and other Western countries tend to be more driven by the market opportunities associated with the surging oil prices. Local governments are providing incentives for job creation and monetisation of stranded local resources. It is difficult to provide summary judgment on the economics of CTL projects as a whole. However, the following general conclusions can be drawn for most CTL projects:

- Recent studies in public domain indicate that the required capital for CTL projects ranges anywhere between US$ 60 000 - 120 000/daily bbl of liquid products. Figure 2 shows a summary of some recently published numbers quoted in a study published by National Petroleum Council CTL/CTG subgroup in 2007.[6] As more CTL projects move forward and more plants are being built, there will be significant opportunities for cost reduction of all technology options.[7]
- Numerous studies conclude that coal gasification, including coal handling and air separation will require 65 - 75% of the overall capital expenditure.[8] As a comparison, syngas generation is reported to be approximately 50% of the investment for Fischer-Tropsch based GTL plants.[9]
- Publicly available information on direct comparison of Fischer-Tropsch route versus methanol route for CTL is limited. In the few public reports available it was shown that required capital for the two routes is similar with at least one report claiming a somewhat lower capital for MTG.[10] Since the overall capital requirement is dominated by syngas generation, technology selection between Fischer-Tropsch route and MTG route is less driven by the difference in capital requirement, but more by factors such as desired product and technical risks.
- Many studies indicate that the CTL will become a competitive option if the oil price remains at approximately US$ 45 - 60 /bbl.[11]
- MTG converts 90% of the hydrocarbon in methanol to a clean gasoline product which is fully compatible with conventional refinery gasoline derived from petroleum. No engine modifications or vehicle modifications are required to use the MTG gasoline. Laboratory and vehicle tests show the performance characteristics of the finished MTG gasoline to compare very favourably in all aspects with commercial premium gasoline.[12] As a comparison, the Fischer-Tropsch process tends to produce multiple slates of products, including potentially more valuable products such as high cetane number distillates and lube products.

It should be emphasised again that the full scope of the methodologies used in these analyses is not necessarily known. The quoted cost estimates are likely lower than would be generated today due to today's higher cost construction environment. However, the key message in all studies is that gasification and gas clean-up dominates the capital investment and key issues in technology comparison are the yields and disposition of liquid transportation fuel.

In addition, the following factors will likely continue to push the strong interests in CTL activities for the near future:

- Although the GTL activities have slowed somewhat because of the resurgence of a liquefied natural gas (LNG) option over the past few years, there is no similar established alternative to convert coal to clean transportation fuels.
- As more countries move towards increasingly stringent fuel and environmental regulations, the premium for clean transportation fuel products will likely increase.
- As the technologies mature, the associated risks will be significantly reduced with a corresponding reduction in capital investment if the price of oil remains at a relatively high level, financing for future CTL projects will become easier, especially if it is associated with lower overall technical risks.

MTG technology description

Both coal gasification and methanol synthesis are commercially mature technologies with several

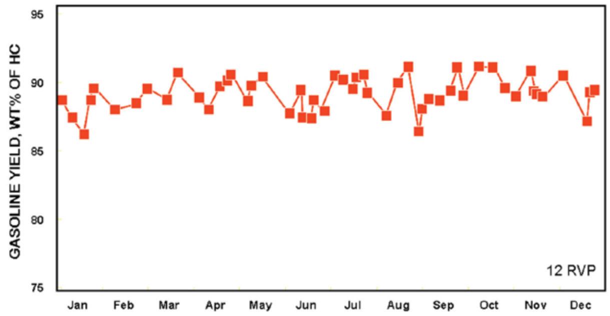

Figure 5. New Zealand commercial MTG gasoline yield.

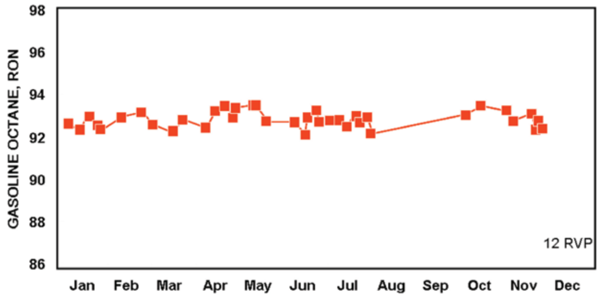

Figure 6. New Zealand commercial MTG gasoline octane.

commercially established routes for both steps. This discussion will focus on MTG, the last step of the process.

MTG chemistry

MTG chemistry was discovered by Mobil scientists in the 1970s.[13] However, it took many years of extensive studies to fully understand the detailed chemistry behind the reaction. A very simplified view of the MTG chemistry is shown in Figure 3. Methanol is first dehydrated to dimethylether (DME). The equilibrium mixture of methanol, DME and water is then converted to light olefins (C2-C4). A final reaction step leads to a mixture of higher olefins, n/iso-paraffins, aromatics and naphthenes. Interrupting the reaction would lead to a production of light olefins instead of gasoline.

Methanol-to-gasoline process[2]

In the MTG process, the conversion of methanol to hydrocarbons and water is virtually complete and essentially stoichiometric. The reaction is exothermic with a heat of reaction of approximately 1.74 MJ/kg of methanol with an adiabatic temperature rise of approximately 600 ˚C. In the fixed bed process commercialised in the New Zealand plant, the reaction is managed by splitting the conversion into two parts. A schematic of the process is shown in Figure 4. In the first part, methanol is converted to an equilibrium mixture of methanol, dimethylether, and water. This step releases 15 - 20% of the overall heat of reaction and is controlled by chemical equilibrium. As such, it is inherently stable.

In the second step, the equilibrium mixture is mixed with recycle gas and passed over specially designed ZSM-5 catalyst to produce hydrocarbons and water. Most of the hydrocarbon products are in the gasoline range. Most of the gas is recycled to the ZSM-5 reactor. The water phase contains 0.1 - 0.2 wt% oxygenates and is treated by conventional biological means to give an acceptable effluent for discharge.

The conversion reactor inlet temperatures are controlled individually by adjusting the flow of reactor effluent to the recycle gas/reactor effluent heat exchangers and by adjusting the temperature difference across exchangers. Excess reactor effluent, superfluous to that required to heat recycle gas in the recycle gas/reactor effluent exchangers, is used to preheat, vaporise and superheat the methanol feed to the DME reactor. The heat flexibility in the excess reactor effluent system is retained by utilising some of the reactor effluent to generate moderate pressure steam in a boiler. Steam generation is adjusted to balance process heat requirements.

Excess reactor effluent from the feed preheat system together with reactor effluent from the recycle gas heat exchangers is then further cooled to 25 - 35 ˚C and passed to the product separator where gas, liquid hydrocarbon and water separate. The water phase, which contains trace quantities of oxygenated organic compounds, is sent to effluent treatment. The gas phase (mostly light hydrocarbons, hydrogen, CO and CO_2) is returned to the recycle gas compressor.

The liquid hydrocarbon product (raw gasoline) contains mainly gasoline boiling range material as well as dissolved hydrogen, carbon dioxide and light hydrocarbons (C1-C4). Essentially all of the non-hydrocarbons and light hydrocarbons are removed by distillation to produce gasoline meeting the required volatility specifications. Methane, ethane and some propane are removed in a de-ethaniser. The liquid product from the de-ethaniser is then sent to a stabiliser where propane and part of the butane components are removed overhead (to fuel gas). Stabilised gasoline is then passed to a gasoline splitter where it is separated into light and heavy gasoline fractions. Each stream is cooled and sent to storage.

MTG gasoline contains 1,2,4,5-tetramethyl benzene (durene), which, though present in commercial gasoline, is at a higher level in MTG product. This lower specification is established to improve drivability performance durene is concentrated in the heavy gasoline fraction of a gasoline splitter and then subjected to a mild hydrofinishing process over a proprietary ExxonMobil catalyst in the heavy gasoline treater. Here durene undergoes isomerisation, disproportionation and demethylation in the presence of hydrogen. The product is recovered in nearly quantitative yield with virtually unaltered RON but with greatly reduced durene content.

Commercial success of New Zealand MTG operation[14]

By all accounts, the startup of the New Zealand operation was a complete success for a world scale, first of its kind plant.[15]

The first methanol unit was brought onstream on October 12th 1985 and achieved design rate within two days of initial production. The first gasoline was produced on October 17th . The second methanol unit was commissioned on December 12th . Subsequently

Table 1. MTG product properties		
	Average	Range
Octane number, RON	92.2	92.0 - 92.5
Octane number, MON	82.6	82.2 - 83.0
Reid vapour pressure, kPa	85	82 - 90
Density, kg/m^3	730	728 - 733
Induction period, min.	325	260 - 370
Durene content, wt%	2	1.74 - 2.29
Distillation		
% Evaporation at 70 ˚C	31.5	29.5 - 34.5
% Evaporation at 100 ˚C	53.2	51.5 - 55.5
% Evaporation at 180 ˚C	94.9	94 - 96.5
End point, ˚C	204.5	196 - 209

Table 2. MTG gasoline versus US conventional refinery gasoline					
	Summer		Winter		MTG
	2004	2005	2004	2005	
Oxygen (wt%)	0.97	0.95	1.07	1.08	
API gravity	58.1	58.4	61.8	61.9	61.8
Aromatics (%vol)	28	27.7	24.6	24.7	26.5
Olefins (%vol)	11.2	12	11.4	11.6	12.6
RVP (psi)	8.31	8.3	12.21	12.12	9
T50 (˚F)	212.7	211.1	199.8	199.9	201
T90 (˚F)	334.7	330.7	326.5	324.1	320
Sulfur (ppm)	118	106	120	97	0
Benzene (%vol)	1.15	1.21	1.08	1.15	0.3

two additional MTG reactors were streamed and the complex was operated at 100% of design capacity by December 27th,1985.

The MTG plant was an excellent example of the ability of engineers to successfully scale up a plant from a small pilot plant (500 kg/d to 1700 tpd). Production, yields, product qualities and catalyst performance were consistent with the estimates developed from the pilot plant data (Figures 5 and 6).

A comparison of the average gasoline properties and the range during the first year of MTG operation is provided in Table 1. It is clear that the operation is very predictable and stable with little variation in the product. It is also interesting to compare the MTG gasoline properties with today's refinery gasoline. Table 2 compares the MTG gasoline properties with the average properties of the conventional gasoline sold in the US markets during 2004 and 2005.[16] The two are virtually identical with only noticeable difference being MTG gasoline's lower benzene content and essentially zero sulfur.

Second generation MTG technology

The current MTG technology is based on the original MTG process developed by ExxonMobil in the 1980s. However, it also reflects improvements made by ExxonMobil in the late 1990s that led to a second generation technology. Detailed engineering design and construction of the first coal-to-gasoline MTG plant, utilising this improved technology, is under construction in China by Jincheng Anthracite Mining Co (JAM). The MTG plant is part of a demonstration scale complex, which also includes a fluidised bed hard coal gasification plant and a methanol plant. The initial phase of the plant is designed for a capacity of 100 000 tpy, but it is expected to expand to 1 million tpy for the second stage of the project.[17] ExxonMobil recently also announced the first US CTL license based on MTG technology. DKRW Advanced Fuels LLC, through its subsidiary Medicine Bow Fuel and Power LLC, has licensed ExxonMobil's MTG technology for its 15 000 bpd CTL plant in Medicine Bow, Wyoming.[18]

Although it is well documented that the original MTG chemistry was developed based on ZSM-5 zeolite, it is worth mentioning that MTG chemistry is very specific to certain aspects of ZSM-5 properties. In fact, over 100 different zeolites were tested during the original MTG technology development. Since the commercialisation of the MTG process over 20 years ago, ExxonMobil has continued R&D efforts and made significant improvements in zeolite applications and manufacturing capabilities. Many of the new learning's are readily applicable to the MTG process and will significantly improve MTG catalyst performance.

The second generation technology incorporates improvements in design that are derived from the operation of the New Zealand plant. The newer design significantly reduces the number of heaters required in the original plants by better heat integration and process optimisation. In addition, the newer design also reduces the size of the heat exchangers and compressor requirements. The combination of the improvements translates into a prospective capital reduction of 15 - 20% versus the original design.

Advantages of the methanol to gasoline option

Project development for CTL is a highly complex process that requires companies to consider many diverse factors when making the technology decision. In the absence of a commercially proven technology, companies have to go through an extensive feasibility study to assess monetary risk to improve the project economics. MTG, as a commercially proven technology, offers unique option which improves the attractiveness for many CTL projects.

Product simplicity

As discussed previously, both the MTG and Fischer-Tropsch processes convert coal into synthesis gas as an intermediary before producing the final products. However, their respective product slates are very different.

- Fischer-Tropsch process produces a broad spectrum of straight chain paraffinic hydrocarbons that requires upgrading to produce gasoline, diesel fuel and lube feedstock. Due to the complexity of the product distribution, the economic justification for further upgrading/processing of all the products improves for large scale projects (e.g. 50 000 - 80 000 bpd). In addition, large projects require large coal reserves (e.g. 2 - 4 billion t), which could require more than one typical mine, thus increasing rail transportation expenses.

- MTG, in contrast, selectively converts methanol to high quality gasoline with virtually no sulfur and low benzene which can be either blended with refinery gasoline pool or sold separately. Approximately 90% of the hydrocarbon in methanol is converted to gasoline as the single liquid product. It is also easier to scale up and down the reactor due to the simple fixed bed process design.

Table 3 shows a comparison of MTG products versus reported product distribution from both the low temperature and high temperature Fischer-Tropsch process reported by Sasol[19] and coal liquefaction yield from the H-Coal process reported by HRI.[20] In both cases, the liquid products require hydrocracking/ hydrotreating and other reforming processes before the liquid products can be used as transportation fuels.

In the case of MTG, the gasoline products can be used with minimal further upgrading. There is also a significant difference in the oxygenate levels in the products. MTG product contains significantly less oxygenates (e.g., approximately 0.1 wt% versus several percentage in the Fischer-Tropsch products). In the case of the Fischer-Tropsch products, the oxygenates have to be further separated and processed.

Technical risk

MTG, as a commercially proven process with nearly a decade of operational experience, provides a low technical risk option for the production of clean gasoline. By comparison, commercially proven Fischer-Tropsch technology options are not always readily available in the market place. The major oil and chemical companies with commercially proven Fischer-Tropsch technology tend to be restrictive in licensing their commercially proven Fischer-Tropsch technologies.

There are several other technology providers in the marketplace for Fischer-Tropsch technologies, but these technology options are generally either in the pilot plant or demonstration stage. The risk associated with financing for CTL projects without commercial references is often too high to overcome. As demonstrated by the recent commercial experience in Oryx, successful design and economical operation of a commercial Fischer-Tropsch plant, even for companies such as Sasol Chevron, remains a challenge.

Coal gasification, methanol synthesis and MTG are commercially proven as the three processes in the CTL technology. Coal gasification is generally considered a mature technology, although there are still many new licensors entering the marketplace to meet the technology needs. Methanol synthesis technology is commercially practiced worldwide, and there continues to be significant advances in the technology.

Process simplicity

The MTG process uses a conventional gas phase fixed bed reactor, which can be scaled up very readily. In the first commercial application in New Zealand the process was successfully scaled up from 500 kg/d to 1.7 million kg/d. On the other hand, most of the technology advancement for the new Fischer-Tropsch technology options relies on slurry phase reactors that are inherently more complex. Scale-up of slurry phase reactor requires a significantly more sophisticated demonstration and modelling in the absence of direct commercial operational experience.

Unlike world scale GTL projects that are generally located near oceans, many of the coal-to-liquids projects are likely situated in locations that do not have easy access to barge or ship transportation. The sizes and weights of the equipment could potentially dictate the choice of technology or could limit where equipment must be fabricated and transported. For example, the Sasol Oryx Fischer-Tropsch reactor is reported to be approximately 2200 t and the Shenhua direct coal liquefaction reactor weighs 2250 t, which is the worlds largest reactor. In contrast, an individual reactor for the New Zealand MTG plant weighed only approximately 80 t (Table 4). These considerations may not always be factored into conceptual studies, but the logistics of fabrication and transportation can be a significant barrier for project development and implementation when accessing the project site via bridges and tunnels

Table 3. MTG gasoline versus Fischer-Tropsch products

	Low temp FT*	High temp FT*	H coal™**	MTG***
	Co catalyst @428 ˚F	Fe catalyst @644 ˚F	Direct liquefaction	
Methane	5	8	No C1 - C4 yields reported	0.7
Ethylene	0	4		-
Ethane	1	3		0.4
Propylene	2	11		0.2
Propane	1	2		4.3
Butylenes	2	9		1.1
Butane	1	1		10.9
C5 - 160C	19	36	36.5	82.3
Distillate	22	16	43.2	-
Heavy oil/wax	46	5	20	-
Water sol. oxygenates	1	5	0.3	0.1
Total	100	100	100	100

** STEYBERG & DRY, 'Fischer Tropsch technology', Elsevier, 2004 (All FT yields are prior to refining for gasoline octane, and diesel pour improvement).*

*** H-coal data from HRI1982 publication*

**** Final plant product with gasoline octane 92 R+O*

Flexibility and process reliability

There have been significant increases in methanol capacity in China in the last few years and it is expected that additional planned capacity will be coming onstream in the next decade. MTG offers a natural extension to companies that want to move to the clean gasoline market that is less affected by the fluctuations of local supply and demand variations of commodity methanol. The methanol route for converting coal-to-gasoline also provides a potential flexibility for producing either methanol or gasoline as market conditions change. In fact, the New Zealand MTG plant was converted to a chemical grade methanol production facility when the oil price plunged to approximately US$ 15/bbl in 1990s. Furthermore, there has been a significant amount of development work dating from the 1980s demonstrating the production of both gasoline and diesel from methanol.

Another process benefit from the methanol route is the fact that the methanol synthesis process and MTG process are linked by liquid methanol that can be easily inventoried in a methanol storage tank. In case of operational issues in either plant, the two plants can be independently operated without a complete shutdown of the whole plant. In contrast, a Fischer-Tropsch plant is linked by syngas to the coal gasification process. It would be difficult to maintain operations if a problem occurred in either plant.

Conclusion

Interests in coal to clean transportation fuel will continue as long as the pressure on oil price remains high. ExxonMobil's commercially proven methanol-to-gasoline (MTG) technology, coupled with established commercial coal gasification and methanol technologies, provides an economically competitive and low risk option for the production of clean gasoline from coal. The MTG route for coal conversion also provides the additional flexibility for directly applying the technology to extend the product slate and flexibility of existing methanol plants.

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Table 4. Reactor weights for different technology

Sasol/Chevron Oryx F-T reactor	2200 t
Shenhua Direct Liquefaction reactor	2250 t
New Zealand MTG individual reactor	80 t

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