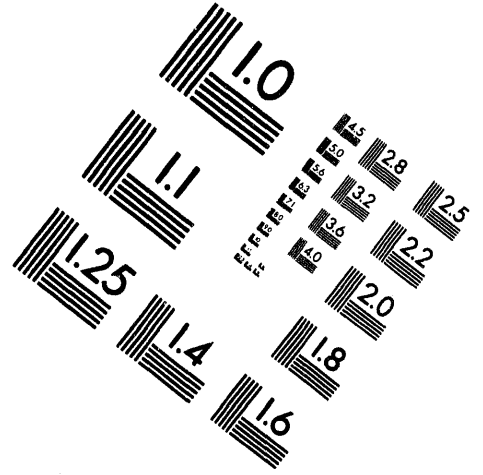
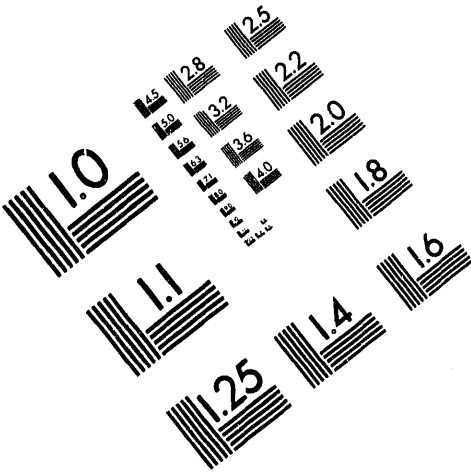




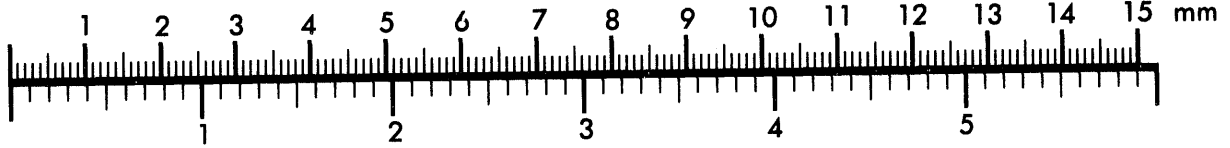
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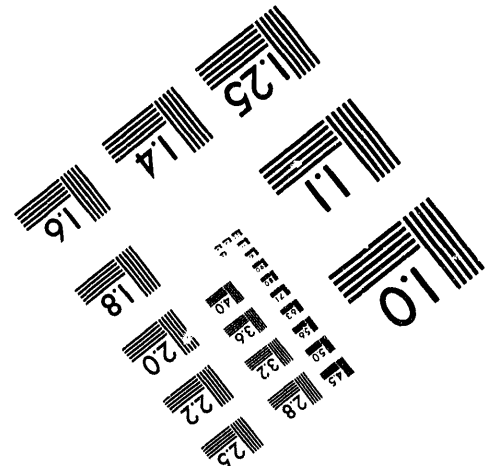
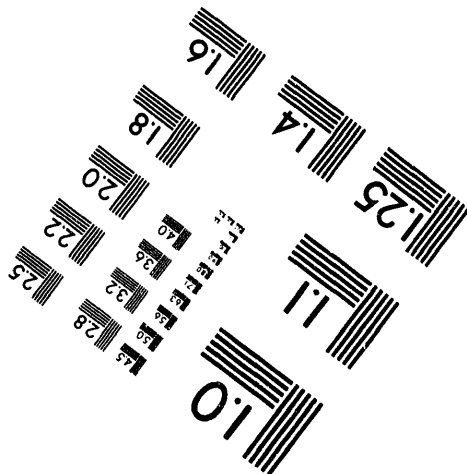
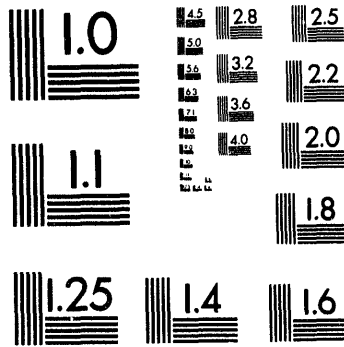
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**Centimeter**



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**ALTERNATE ROUTES FOR THE PRODUCTION OF FUELS FROM COAL  
AND NATURAL GAS**

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RECEIVED  
JUN 03 1994  
OSTI

**Keywords:** Coal Liquefaction, Gasification, Fischer-Tropsch Synthesis

**ABSTRACT:**

Almost all transportation worldwide is powered by high energy-density liquid hydrocarbon fuels produced from crude oil. Transportation fuels currently use over 50 percent of total world petroleum demand of 66 million barrels per day. Prior MITRE studies indicate that crude oil supply will become severely limited after the year 2030 as increasing world energy demand, driven by population growth and economic development, depletes oil resources. If conventional liquid hydrocarbon fuels that can use existing production and distribution infrastructures are still needed for transportation in the future, then alternate sources of these fuels will have to be utilized. Two such sources are natural gas and coal. Natural gas reserves worldwide are expected to last well into the 21st century, and coal resources are enormous. This paper examines the technologies for producing environmentally superior liquid transportation fuels from coal and natural gas using modern conversion technologies. Estimates of the costs of fuels from these sources are given, and the potential environmental impacts of these fuels are examined.

**INTRODUCTION:**

High energy density liquid fuels are the predominant form of energy used for transportation worldwide. The existing infrastructure for production, refining, distribution, and use of liquid fuels represents an enormous investment worldwide, especially so for the Organization of Economic Cooperation and Development (OECD) countries that are transportation rich. The pressures of potential resource limitations for petroleum and the drive toward a cleaner environment have aroused considerable interest in developing alternative fuels for transportation. These alternatives include non-liquid fuels like natural gas and, perhaps in the future, hydrogen. If it is considered to be economically and technically expedient to continue to use the existing liquid fuels infrastructure rather than to change to a gas-based system, liquid fuels could still be produced from non-petroleum sources. The most abundant fossil fuel source worldwide is coal and this can be used to produce high quality liquid fuels. Natural gas can also be used as a source of liquid transportation fuels. Previous studies at MITRE have examined potential world energy supply and demand scenarios till the year 2100<sup>(1)</sup>. These hypothetical scenarios show that total world energy demand increases from the current annual use of 360 exajoules to about 1,100 exajoules by 2100. This projection assumes that energy conversion and end-use efficiency increase such that after 33 years existing equipment is replaced by new equipment that saves 33 percent of the energy. This 33 cycle continues for another two cycles of continuing efficiency improvements saving an additional 16.6 and 8.3 percent respectively. Recoverable oil and gas resources are assumed to be 10,000 exajoules each<sup>(2)</sup>, and they will be essentially depleted by 2100. The purpose of this look into the future is to demonstrate that after 2030 oil production will be in decline and an alternative to petroleum-based fuels will have to be found.

**Coal as an alternative feedstock for transportation fuels:**

The key to converting solid coal to liquid fuel is hydrogen. Liquid fuels typically contain

50 percent of total world petroleum demand of 66 million barrels per day. Prior MITRE studies indicate that crude oil supply will become severely limited after the year 2030 as increasing world energy demand, driven by population growth and economic development, depletes oil resources. If conventional liquid hydrocarbon fuels that can use existing production and distribution infrastructures are still needed for transportation in the future, then alternate sources of these fuels will have to be utilized. Two such sources are natural gas and coal. Natural gas reserves worldwide are expected to last well into the 21st century, and coal resources are enormous. This paper examines the technologies for producing environmentally superior liquid transportation fuels from coal and natural gas using modern conversion technologies. Estimates of the costs of fuels from these sources are given, and the potential environmental impacts of these fuels are examined.

#### **INTRODUCTION:**

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#### **Coal as an alternative feedstock for transportation fuels:**

The key to converting solid coal to liquid fuel is hydrogen. Liquid fuels typically contain about 14 percent hydrogen whereas coal contains around 5 percent. This hydrogen deficit can be made up by forcing hydrogen into the coal under pressure (so-called direct liquefaction<sup>(3)</sup>), or by gasifying the coal with oxygen and steam to a synthesis gas containing hydrogen and carbon monoxide that is then passed over catalysts to form hydrocarbons (so-called indirect liquefaction<sup>(4)</sup>). For direct liquefaction, coal is slurried with a recycle oil and heated under a high pressure of hydrogen to produce a synthetic crude oil that can be upgraded into specification transport fuels by existing petroleum

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refinery processes. The hydrogen is produced by gasification of coal and residue or by natural gas steam reforming. For indirect liquefaction, the synthesis gas produced is passed over Fischer-Tropsch (F-T) catalysts where a series of hydrocarbons ranging from  $C_1$  to about  $C_{200}$  are produced. These can also be refined to produce specification liquid fuels by using mild refinery operations.

Direct liquefaction, invented in the early 20th century by Bergius, was used extensively by the Germans in World War II to produce high octane aviation fuel, and since that time research and development have completely transformed the technology. Research sponsored by the United States Department of Energy over the last fifteen years has led to the development of a catalytic two-stage liquefaction process<sup>(5)</sup> (CTSL) that uses two high pressure ebullating bed reactors in series to solubilize coal and upgrade it to a distillate raw product containing about 13 percent hydrogen at an overall thermal efficiency of about 66 percent. Liquid distillate yields of over 70 percent on a moisture ash-free (MAF) coal basis are regularly obtained with bituminous coals, and yields of 60 to 65 percent are usually obtained with low rank coals as feedstock. This translates into oil yields of over 3.5 barrels (557 litres) per tonne of MAF coal.

Indirect liquefaction technology is commercialized in South Africa and produces about a third of that country's gasoline and diesel fuel. The South African Synthetic Oil Company (SASOL)<sup>(6)</sup> plants produce together over 100,000 barrels per day of fuels. When SASOL made the decision to build these plants the only commercially available coal gasifier that met the requirement of processing high ash South African coals was the Lurgi dry-ash gasifier. Today, however, research and development in coal gasification has resulted in the commercialization of highly efficient entrained gasifiers such as Shell and Texaco. These systems that gasify coal at high temperatures and pressures can process all coals to produce a synthesis gas containing only carbon monoxide and hydrogen. These entrained gasifiers that have net efficiencies for synthesis gas production of about 80 percent greatly improve the overall efficiency, hence the economics, of indirect liquefaction. The other area that has led to significant improvements in the efficiency and economics of indirect liquefaction is the development of advanced F-T synthesis technology. Shell has developed advanced fixed-bed reactor technology for F-T synthesis and is currently operating a plant in Malaysia for the production of diesel fuel and waxes from off-shore natural gas. SASOL has developed an advanced Synthol reactor that uses a fixed fluid bed concept as opposed to the circulating fluid bed currently at use in the SASOL plants. SASOL has also developed a slurry F-T reactor that promises to be even more cost effective. The United States Department of Energy is also funding research aimed at developing both an advanced slurry F-T reactor system and an effective F-T catalyst to use in the advanced reactor. This system being studied both at the bench-scale and at the Alternate Fuels Development Unit at Laporte Texas will be compatible with the synthesis gas produced in the advanced entrained coal gasifiers mentioned above.

In a commercial indirect coal liquefaction facility, the synthesis gas produced in the coal gasifiers is purified and shifted then sent to the F-T reactors where a whole range of hydrocarbons are produced. The unconverted synthesis gas together with methane and ethane is sent to an autothermal reformer where the methane and ethane is converted to hydrogen and carbon monoxide. This is then recycled to the F-T reactors. The hydrocarbon products are separated and the gasoline and diesel boiling range fractions are sent to the downstream refinery to be upgraded to specification fuels. The heavy wax hydrocarbons are hydrocracked to additional gasoline and diesel and also sent to the refinery.

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#### **Natural gas as feedstock for production of liquid transportation fuels:**

As an alternative to using natural gas in its gaseous state as a transportation fuel, it can be converted to specification gasoline and diesel fuel so that the existing liquid fuels infrastructure can be utilized. This is commercialized in New Zealand where natural gas is converted into methanol and the methanol is converted into gasoline using the Mobil

**Methanol-to-Gasoline (MTG) technology<sup>(9)</sup>.** Shell is converting remote natural gas in Malaysia into liquids that can be transported by tanker to market.

For indirect liquefaction, natural gas can be steam reformed or partially oxidized to synthesis gas. This gas is then processed in the same manner as the coal-derived synthesis gas described above. Thus improvements in F-T technology are applicable to natural gas processing. In addition, there have been recent advances in catalytic partial oxidation that can produce lower hydrogen to carbon monoxide synthesis gas at lower cost. For direct liquefaction, natural gas can be used as the source of hydrogen instead of coal, so that coal is only sent to the liquefaction reactors. This results in elimination of the coal gasification plant from the direct plant. In addition, the coal handling units can be reduced in size and the plant electric power required is reduced. Thus, using natural gas for this application lowers capital investment of the direct coal liquefaction plant and, depending on the cost of the natural gas, can result in a lower required selling price of the coal derived transportation fuels. A sensitivity analysis of the equivalent crude price versus natural gas price for this case, where gas is used to produce hydrogen in the direct coal liquefaction plant, shows that using natural gas can result in lower costs for coal liquids up to a natural gas price of about \$4 per million Btu (\$4.22 per GigaJoule). Using natural gas for hydrogen production also has a significant positive impact on the carbon dioxide produced per product barrel. This quantity can be reduced from about 0.42 tonnes per product barrel when coal is used for hydrogen to 0.21 tonnes in the natural gas case.

#### **Economics of fuels production from coal and natural gas:**

At the MITRE Corporation, computerized simulation models of coal and natural gas based liquefaction technologies have been developed as part of our funding support from Sandia National Laboratories and the United States Department of Energy<sup>(7,8)</sup>. In these models, test data from ongoing research and development is used to develop conceptual commercial plants for direct and indirect coal liquefaction, and natural gas based plants. Construction and capital costs of the plants are estimated together with operating costs. Using a constant set of economic parameters the required selling price of liquid fuels can be calculated. This price is then adjusted to an equivalent crude price.

#### **Quality and environmental impact of coal-derived transportation fuels:**

Direct coal liquefaction produces an all distillate product that can be refined using conventional hydrotreating, hydrocracking, fluid catalytic cracking, and reforming to yield high octane gasoline, high density jet fuel and 45 cetane diesel. Indirect liquefaction produces a paraffinic gasoline whose octane can be adjusted by reforming or by adding octane enhancers like alcohols or ethers. The diesel fraction is excellent, has a cetane of over 70 and zero aromatics and sulfur. These refined products can exceed current transportation fuel specifications and their use will have a positive effect on air quality. The paraffinic indirect naphtha can be blended with the aromatic direct naphtha to minimize the amount of refining required. Similarly, the aromatic diesel from direct liquefaction can be blended with the paraffinic diesel from indirect. Thus a hybrid plant concept where both direct and indirect technologies are sited at the same location may have considerable merit.

#### **CONCLUSION:**

Coal and natural gas can be used as resources to produce specification liquid transportation fuels that make use of the existing liquid fuels refining, distribution and end-use infrastructure. Although the costs of these fuels are higher than current crude

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#### **CONCLUSION:**

Coal and natural gas can be used as resources to produce specification liquid transportation fuels that make use of the existing liquid fuels refining, distribution and end-use infrastructure. Although the costs of these fuels are higher than current crude prices, they can be competitive with crude oil at about \$30 to \$35 per barrel. The United States Energy Information Agency (EIA)<sup>(10)</sup> has just published its latest World oil price (WOP) projections. In their reference scenario, the WOP is expected to reach \$35 per barrel by the year 2015.

(This work performed at The MITRE Corporation was supported by Sandia National Laboratories, which is funded by the U.S. Department of Energy under contract DE-AC04-94AL85000.)

## **REFERENCES:**

- 1) Gouse, S. W., D. Gray, D. L. Morrison, and G. C. Tomlinson, *Potential World Development through 2100: The Impact on Energy Demand, Resources and the Environment*, Proceedings of the 15th Congress of the World Energy Council, Madrid, Spain, 20-25 September 1992, pp. 18-33, *World Energy Council Journal*, December 1992.
- 2) Masters, C. D., E. D. Altanasi, W. D. Dietzman, R. F. Meyer, R. W. Mitchell, and D. H. Root, *World Resources of Crude Oil, Natural Gas, Natural Bitumen, and Shale Oil*, Proceedings of the 12th World Petroleum Congress, Vol 5, pp. 3-27, published by John Wiley.
- 3) *Liquid Transportation Fuels from Coal Part 1: Direct Liquefaction*, PETC Review, Issue 3, pp.4-13, March 1991.
- 4) Stiegel, Gary J., *Transportation Fuels from Coal Part 2: Indirect Liquefaction*, PETC Review, Issue 4, pp.14-23, Fall 1991.
- 5) Advanced Coal Liquefaction Research and Development Facility, Wilsonville, Alabama, *Technical Progress Report Run 258 Subbituminous Coal*, DOE/PC/50041-130, May 1991.
- 6) Dry, Mark, E., *The Sasol Fischer-Tropsch Processes*, Applied Industrial Catalysis Volume 2, Ed. Bruce Leach, Academic Press 1983.
- 7) Gray, David, Abdel ElSawy, and Glen Tomlinson, *Quantification of Progress in Indirect Coal Liquefaction*, Proceedings of Liquefaction Contractors' Review Meeting, Pittsburgh, Pa. pp.344-356, September 1991.
- 8) Gray, David, and Glen Tomlinson, *Assessing the Economic Impact of Two-Stage Liquefaction Process Improvements*, Sandia Contractor Report SAND87-7147, August 1988.
- 9) Haggin, Joseph, *Methane-to-Gasoline Plant Adds to New Zealand Liquid Fuel Resources*, Chemical and Engineering News, pp.22-25, 22 June 1987.
- 10) *Annual Energy Outlook 1993*, published by Energy Information Administration, January 1993.

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