# COAL REFINERIES: A DEFINITION AND EXAMPLE CONCEPTS

## ABSTRACT

A definition of the term "coal refinery" is offered and twenty-seven coal refinery concepts are identified and described. These concepts were selected as being representative of the many possible variations of coal refinery concepts that are possible. The status of development of the different coal refinery descriptions ranges from conceptualized processes in the laboratory stage to integrated processes which are being demonstrated at near commercial scale. The extent of the descriptions ranges from a short summary of the basic processes involved and products produced to more detailed discussions of the processes, coal feeds, environmental aspects, status of development, and research needs. The concepts are grouped into one of four categories and a summary discussion of status and research and development needs for each category is provided. Comments pertinent to the research and development needs for coal refineries in general are also provided.

## FOREWORD

The U.S. Congress has directed the U.S. Department of Energy (DOE) to examine coal refineries and evaluate their potential for meeting new markets and to outline the R&D needs for the potential commercialization of coal refineries. The DOE is to report to Congress on these matters. This report was prepared in response to this directive.

The approach followed in preparing this report was to identify and describe several concepts that illustrate the products from coal refineries and the markets in which these products would compete, and to point out the state of development or commercialization of these concepts. Information provided for these concepts includes process descriptions, coal feedstocks, products, applications, development status, and environmental aspects. The extent of information was not uniform for each of these items nor was it uniform among the various concepts due to several factors including state of development, proprietary and patent concerns, and others. R&D needs for individual concepts are discussed and are summarized for each of four major categories of coal refineries.

## SUMMARY

Because coal is this nation's most abundant fossil fuel, the increasing demand for energy and the potential for coal to supply more markets make it likely and desirable that the use of this resource be expanded. Issues such as greenhouse gases, acid rain, solid wastes, land use, and others make it imperative however that continued and expanded use of coal be done in an efficient and environmentally responsible manner.

Historically, the majority of coal has been used as a fuel serving markets in heating, industrial uses, and in the production of electricity. More recently however, coal is being re-examined by industry as a source of chemical feedstocks, transportation fuels, and other specialty markets in addition to the more traditional markets. These expanded markets stem from a variety of reasons including a dwindling of alternative energy supplies, costs, and national energy security. Several research and development (R&D) activities have been undertaken in both the private and public sectors (along with some joint ventures) to address these potential markets. These activities have included: pre-combustion technologies such as coal beneficiation or cleaning to remove potential pollutants before the coal is burned or further converted; combustion technologies such as fluidized-bed combustors and coal gasification which modify the way the coal is burned; and post-combustion technologies such as those designed to reduce the emissions of sulfur dioxide, oxides of nitrogen, and particulates from the flue gases released from a facility. These R&D activities have led to the commercialization of some processes while others are still being examined.

Many of the potential new markets for coal will necessitate that it be processed in ways other than traditional combustion. For example, using coal as an alternative to petroleum-based fuels in the transportation sector will require the processing of coal into a liquid that can be used with little or no modification to the cars and trucks consuming it or to the delivery system that would provide this fuel to the public. The "coal refinery" concept is a result of the realization that coal must be processed in other than traditional ways in order to meet the needs of potential expanded markets. The term "coal refinery" is defined as a system consisting of one or more individual processes integrated in such a way as to allow coal to be processed into two or more products supplying two or more markets.

There are many proposed concepts that could be called coal refineries. Twenty-seven of these concepts are discussed in this report. For each concept, information is provided on products and markets, environmental aspects, development status, and R&D needs. These concepts have been placed into four categories dependent on the principal coal processing step for the concept.

Ten concepts falling in the Devolatilization category are described. These concepts process coal under non-oxidizing or mild-oxidation conditions at temperatures in the range of 1,000 to 1,500°F and generally low to moderate pressures. A high-carbon char which can be used as a solid fuel or upgraded into a higher-value product is one of the products from these concepts. A liquid is also recovered (approximately one-half to one barrel of liquids per ton of coal) which can be burned as fuel or upgraded for higher-value use.

The Gasification category contains seven concepts. These concepts are based on more severe temperature and pressure conditions where the coal is partially oxidized using oxygen and steam into a gaseous fuel (called "synthesis gas" or "syngas") consisting principally of carbon monoxide, hydrogen, water, and carbon dioxide. This gas can be burned in combustion turbines to produce electricity, upgraded to a substitute natural gas, or converted into chemical products such as methanol, ammonia, or acetic anhydride which can, in turn, be used in several markets.

Eight concepts are described in the Liquefaction category. They are generally configured to yield a greater fraction of liquid products than most of the concepts in the other categories. Products from these concepts can be similar to those of a conventional petroleum refinery which produce a wide variety of products ranging from light-hydrocarbon gases to light, medium, and heavy distillates, up to high-molecular-weight waxes. Solids from some of these concepts, along with associated heavy oils, could be used as fuel for electricity production.

The two concepts described in the Bioprocessing category represent the growing field of using biological agents in materials production. Concepts are based on the use of micro-organisms in the production of gaseous or liquid fuels. As compared with most coal refinery concepts, the bioprocessing concepts are generally at an earlier state of development.

Summary information on the coal refinery concepts described in this report is presented in Tables S.1-S.4. These tables indicate the sponsor or proponent of the concept, the major products, and the development status for each concept. With many concepts, there is flexibility in the products which can be exercised with some design and/or operational changes in the concept. The major products listed in these tables are for the specific concepts as described in this report.

Development status is generally noted as bench scale, process development unit (PDU), pilot plant, demonstration, or already commercial. While there is overlap in the definitions of these terms (particularly with respect to the quantities of coal processed per unit time), the distinctions used in these tables are roughly as follows:

Bench scale: Laboratory batch experiments designed to establish initial technical feasibility for the main process concepts and to produce research samples of the final product, usually at a scale of a few pounds of coal per hour.

PDU: Continuous, larger scale test of individual processes that make up a concept often done to determine impact of design changes, feedstock, operating conditions, recycle streams, etc. on the product yields.

Pilot plant: Major process components are linked into a system but the degree of system integration is not as complete as would be anticipated in a demonstration or commercial facility. Typical sizes range from several tons of coal per day to about 100 tons per day.

Demo: A larger, more fully integrated facility in which most of the individual components are linked in a way anticipated for commercial facilities. Some components for which there is little or no uncertainty in its performance are often deleted from demonstration plants in order to reduce costs while still demonstrating that the overall concept is feasible and practical. A demonstration facility may be one train of a commercial-scale plant or at a scale at which minimal risk exists for scale-up to commercial scale. The purpose is to obtain operating, environmental, and economic data for a commercial-scale design.

Commercial: Fully developed integrated facility operating for the purpose of generating revenue.

Research and development needs for coal refinery concepts range from basic research of chemical and physical phenomena to the demonstration of an integrated system of all the component parts of a particular coal refinery. In some instances, the major components in the coal refinery concept are being utilized commercially. R&D programs within the Department of Energy and in the private sector are directed toward the ultimate commercialization of coal refinery concepts and the introduction of coal refinery products into new and expanded markets. One example of such R&D activities is the ENCOAL concept in which devolatilization techniques are used to yield liquid fuels and a premium solid fuel. A Department of Energy/private sector joint venture to construct and operate a plant using this concept is underway as part of the Clean Coal Technology Demonstration Program. Another example of ongoing R&D activities is a project demonstrating the ability to more efficiently produce methanol from coal-derived synthesis gas. Several examples of ongoing R&D efforts are cited in the descriptions of the individual concepts. Thus, coal refinery concepts are being examined within the context of both the R&D and large-scale demonstration levels, and that these activities are taking place within the existing structure and programs of the Department of Energy.

Table S.1: Summary of Devolatilization Coal Refinery Concepts

| COAL REFINERY CONCEPT | SPONSOR / PROPONENT | MAJOR PRODUCTS | DEVELOPMENT STATUS |
|---|---|---|---|
| Calderon | Calderon Energy Co.; Bowling Green, OH | Electricity and methanol | 24 ton/day PDU |
| Charfuel | Carbon Fuels Corp.; Englewood, CO | Char-liquid-slurry fuel, methanol, ether, naphtha, and BTX | 1 ton/hour PDU (using Rockwell hydropyrolysis reactor) |
| Coal Liquid and Coke Mild Gasification | CLC Associates; Bristol, VA | Form coke, coal-derived liquids | 1,000 lb/hr PDU |
| ENCOAL Mild Gasification | Shell Mining Co.; Houston, TX | Solid and liquid boiler fuels | 200 lb/hr PDU; Demonstration plant construction underway |
| Hydrocarb | Brookhaven National Lab.; Upton, NY | Coal-derived slurry fuel, utility fuel, and (possibly) methanol | Bench scale |
| Institute of Gas Technology Mild Gasification | Institute of Gas Technology; Chicago, IL | Form coke and coal-derived liquids | 100 lb/hr PDU |
| Marshall Owen | Marshall Owen Ent.; Reno, NV | Smokeless solid fuel, crude tar, crude light-oil, and high-Btu gas | Concept based on commercial process in England |
| SFuel | Institute of Chemical Processing of Coal; Poland | Smokeless solid fuel and electricity | 7.2 ton/day pilot plant |
| UNDEERC Mild Gasification | Univ. of N. Dakota Energy and Environ. Research Center; Grand Forks, ND | Form coke and coal-derived liquids | 100 lb/hr PDU |
| WRI/AMAX Mild Gasification | WRI and AMAX; Laramie, WY | Carbon black, pitches, and diesel-blend fuel | 100 lb/hr PDU |

Table S.2: Summary of Gasification Coal Refinery Concepts

| COAL REFINERY CONCEPT | SPONSOR / PROPONENT | MAJOR PRODUCTS | DEVELOPMENT STATUS |
|---|---|---|---|
| Coproduction of Acetic Anhydride, Acetic Acid, and Methanol | Tennessee Eastman Co.; Kingsport, TN | Acetic anhydride, acetic acid, and methanol | 900 ton/day commercial plant |
| Coproduction of Methanol and Electricity | Air Products and Chemicals, Inc.; Allentown, PA | Methanol and electricity | Commercial components; Conceptual design for system; 10 ton/day PDU for methanol |
| Coproduction of SNG, Electricity, Methanol, and Chemical Intermediates | Great Plains Coal Gasification Plant; Beulah, ND | SNG, methanol, and electricity | 17,000 ton/day commercial SNG plant |
| Coproduction of Urea and Electricity | TVA; Chattanooga, TN | Urea and electricity | Commercial components; Conceptual design for proposed system |
| Dual Production of Ammonia and Electricity | Adaptation of proposed modification to Ube Industries; Japan | Ammonia and electricity | Commercial components; Conceptual design for system |
| Once-Through Fischer-Tropsch with Power Generation | Concept examined for DOE by MITRE Corp.; McLean, VA | Chemical feedstocks, liquid fuels, and electricity | Commercial components; Conceptual design for system |
| Shell Middle Distillate Synthesis | Royal Dutch Shell; Netherlands | Naphtha, kerosene, and gas oil | Commercial with natural gas feed |

Table S.3: Summary of Liquefaction Coal Refinery Concepts

| COAL REFINERY CONCEPT | SPONSOR / PROPONENT | MAJOR PRODUCTS | DEVELOPMENT STATUS |
|---|---|---|---|
| Agflotherm | Alberta Research Council; Canada | Processed oil and solid fuel | Bench scale |
| Catalytic Two-Stage Liquefaction | Hydrocarbon Research Inc.; Princeton, NJ | Transportation fuels, fuel oil, and utility fuel | Similar to 6 ton/day PDU |
| Coal Deploymerization-Liquefaction | Department of Fuels Eng.; University of Utah | Coal-derived liquids, kerosene, and oils | Bench scale (Exploratory) |
| Coal/Oil Coprocessing | Hydrocarbon Research Inc.; Princeton, NJ | Naphtha, middle distillates, vacuum gas oil, and liquefied petroleum gases | 3 ton/day PDU |
| Enhanced Coal Liquefaction by Rapid Heating | Department of Fuels Eng.; University of Utah | Liquid hydrocarbons | Bench scale (Exploratory) |
| Liquid Solvent Extraction | British Coal Corp.; Great Britain | Liquefied petroleum gases, naphtha, and middle distillate | 2.5 ton/day pilot plant |
| NEDOL | New Energy Development Organization; Japan | Naphtha, middle distillates, and (potentially) liquified petroleum gases | 1 ton/day PDU; 150 ton/day pilot plant under construction |
| Nippon Brown Coal Liquefaction | Nippon Brown Coal Liquefaction Co. Ltd.; Japan | Naphtha and middle distillates | 50 ton/day pilot plant |

Table S.4: Summary of Bioprocessing Coal Refinery Concepts

| COAL REFINERY CONCEPT | SPONSOR / PROPONENT | MAJOR PRODUCTS | DEVELOPMENT STATUS |
|---|---|---|---|
| Alcohol Fuels from Bioprocessing | University of Arkansas; Fayetteville, AR | Chemical feedstocks and alcohol fuels | Bench scale |
| Lignite Refinery | Houston Lighting & Power; Houston, TX | Chemical feedstocks, carbon dioxide, and methane | Concept |

# 1. INTRODUCTION

## 1.1 BACKGROUND

Coal is the nation's most abundant fossil fuel and has long been a mainstay in the use of energy throughout the United States and the rest of the world. Coal has provided an energy source for many sectors including residential and commercial space heating, industrial process heating, transportation, and electricity production. Another major use has been the production of coke for the metallurgical industry. The aromatic hydrocarbons released when producing coke were the basis of the modern chemical industry. It is important that continued widespread and expanded use of this resource be done in an environmentally responsible and economically competitive manner.

Coal is a very complex and diverse material containing principally carbon, hydrogen, and oxygen, smaller amounts of nitrogen and sulfur, trace elements, and ash - a combustion residue. The characteristics of coal, e.g., its carbon content, sulfur content, calorific value, hardness, swelling, caking, and coking, are important parameters in determining the processes wherein the coal is used, the kinds and quantities of potential products yielded, the environmental effluents, and other factors. There are detailed classification or ranking systems that have been developed to characterize the various types of coal and which are used in the design and operation of coal-utilization facilities. The simplest level of classification is by rank where the coal is characterized as lignite, subbituminous, bituminous, or anthracite depending on its fixed carbon content and its calorific value. In this report, the term "coal" is used to describe materials falling into these ranks and where it is necessary to specify the type of coal, this simple ranking system will be used.

Extensive research and development (R&D) activities have been undertaken to enable coal to be used in various applications with concern for environmental impacts. Such R&D has been in the pre-combustion area such as coal beneficiation or cleaning to remove potential pollutants before the coal is burned or processed further; in the combustion area such as more efficient burners, fluidized-bed combustors; and coal gasification processes which modify the way the coal is combusted; and in the post-combustion area where flue-gas desulfurization, nitrogen-oxide control, and ash removal technologies have been developed to remove pollutants from the flue gas after the coal has been burned. Advances in coal liquefaction, pyrolysis, and gasification have also been made through R&D programs in both the public and private sectors.

The advancement of the coal conversion technologies of devolatilization, gasification, and liquefaction offers the potential for using coal to meet demands in many applications. Some of the applications and technologies for using coal in markets other than the electric utility sector are noted in Ref. 1. Such applications include coal's expanded use as a boiler fuel, its use as a transportation fuel (in ways other than the coal-fired locomotives of years gone past), and its use in non-fuel applications such as chemical feedstocks. It has long been, and continues to be, one of the goals of the U.S. Department of Energy (DOE), working with the private sector, to explore and develop coal-based technologies that will allow this vast resource to be used in a more efficient and environmentally responsible manner. The status of selected coal-based technologies in addressing some of these potential new and expanded applications is one of the focal points to which this report is directed.

## 1.2 CONGRESSIONAL DIRECTIVE

Recognizing the potential for coal to meet expanded markets in a cost effective and environmentally responsible manner, the U.S. Congress has directed DOE to "...prepare an analysis of the coal 'refinery' concept in which coal is processed to provide a suite of marketable products such as transportation fuels, electricity, byproducts such as sulfur, and carbon-based fuels" [2]. This report is in response to the Congressional directive.

## 1.3 COAL REFINERY PRODUCTS AND MARKETS

Products from coal refineries could potentially be used in many markets. Electric utility fuels, industrial fuels, transportation fuels, chemical feedstocks, and specialty chemicals such as carbon black are among the many possible areas where coal refinery products could be used. The likely builders and operators of coal refineries encompass an equally diverse range. Some refineries may be built with the principal objective of generating electricity with other products being used as alternative fuels or to supply a chemical feedstock need. Other refineries may have the objective of upgrading coal into higher value fuels for the electric utility and industrial heating sectors. Still other refineries may be designed to yield coal-derived liquids that can be upgraded into transportation fuels.

The location of refineries may also be dictated, at least in part, by the proximity of existing, compatible components of the refinery or by the location of the users. For example, a coal/oil coprocessing refinery would likely (at least for the initial plants) be located near a conventional oil refinery so that advantage could be taken of existing equipment and infrastructure. Refineries producing small quantities and/or low-grade liquids would likely have to be near an existing petroleum refinery with the ability to upgrade the liquids or a buyer who would burn the liquids as they are produced. Other coal refineries would likely be located near the mine supplying the coal to minimize transportation costs and to take advantage of the synergy that would exist between these operations.

The multiplicity of potential products from coal refineries allows several markets to be addressed. The ultimate commercialization of any concept will depend not only on the value of the products and their production costs, but also on the extent of the market for these products. If the market is very specialized and small so that only a very small number of coal refineries would overwhelm the market, the value of these products could fall and the incentive to operate and build coal refineries might not exist. In some cases, the refineries could switch their processes somewhat to yield different, more valuable, products (similar to a petroleum refinery) but the flexibility in this regard is probably limited. On the other hand, if the demand for the products is large and reasonably constant or likely to increase, the product value can be expected to be more consistent and predictable so that investors may be more likely to utilize these concepts.

Several coal refinery concepts yield a solid fuel as one of their products. Many of these solid fuels have characteristics that make them more favorable than coal. Such characteristics may include some combination of higher energy content per pound, lower sulfur content, or lower ash content. If economically competitive, the market for such fuels is very large, particularly in the electric utility sector as environmental requirements become more stringent, the age of existing combustors increases, and the demand for electrical energy continues to rise.

Liquid fuels from coal refinery concepts can be expected to see large and possible growing markets. Depending on the extent of upgrading, these liquid fuels can be directly burned in industrial or electric-utility boilers or they can be used as transportation fuels thereby helping to displace potentially unreliable oil imports with domestic resources. Assuming that the coal liquids can meet the performance requirements in these markets, economics will continue to be the most important factor in determining their role in the marketplace.

Some of the other potential products from coal refineries will compete in markets where the demand is not as obvious as those of liquid and solid fuels. The yields from the coal refineries should be compared with the demand to be certain that supply will not overwhelm demand and drive down the value of these products. For example, the quantity of methanol produced annually in the electricity/methanol coal refinery is estimated at approximately 55,000 tons for a facility processing 1,000 tons of coal per day. When comparing this production rate with the 3 million tons of methanol currently produced each year, it becomes apparent that a significant number of coal refineries producing methanol in this way could operate without changing the supply/demand balance. It also should be noted that changes in the demand could play an important role in determining the number of refineries that would be economically viable. For example, continuing the case of the electricity/methanol refinery, increased use of methanol as a fuel (e.g., as an electric-utility fuel or as a transportation fuel) could drive demand up significantly and allow more such facilities to be economically operated.

The coal refineries based on devolatilization all yield a solid product or solid/liquid slurry. As noted above, there is a large market for this product as a fuel in industrial or electric-utility combustors. The value of this product may, however, be only slightly greater than run-of-mine coal. In order to increase the product value, some coal refinery concepts include an upgrading stage for the solids in the expectation of meeting more specialized markets. In most cases, these special markets are more limited than the conventional markets. For example, this solid product can be upgraded to carbon black where approximately 1.5 million tons are used annually in the rubber tire industry with prices generally in the range of $400-$600/ton. As the output from a 1,000 ton/day refinery is estimated at approximately 95,000 tons/yr, it can be seen that there is a place for such coal refineries but that the market is limited.

The examples discussed above indicate that market constraints are factors in the economic evaluation of coal refineries. While the products from a given facility can potentially be sold at a high price, there may be limitations on the number of such facilities that can be economically viable.

## 1.4 DEFINITION OF COAL REFINERY

There are many points of view as to just what a coal refinery is or should be. Various concepts incorporating many different technologies or processes have been proposed and evaluated under this heading. To some people, there must be a close correlation to a petroleum refinery in that a full slate of liquid products must be possible and the system must be capable of changing product yields with temporal changes in market conditions. While any coal-processing facility having these traits would be a "coal refinery," that definition of the term was considered to be too limiting and was therefore expanded to include facilities that yield products other than those typically produced in a conventional petroleum refinery. By expanding the definition, the term "coal refinery" encompasses more processes and thereby reflects the wide range of products that can be produced from coal.

Another approach considered in arriving at the definition of coal refinery was to include all processes that yield more than one marketable product and, as a result, to define a coal refinery as an "integrated energy facility." This approach, however, was felt to be too general in that too many technologies would qualify as coal refineries. For example, an electric power plant producing gypsum from its flue-gas-desulfurization system and a coal beneficiation plant with two solid product streams would be coal refineries under this definition, as would any electricity generating concept that made beneficial use of waste heat, such as for water desalination.

Other approaches in arriving at a definition looked at the number and types of markets, processes, the degree of integration among individual systems, or any combination of these or other ideas. A difficulty with this type of definition was that it tended to become very cumbersome due to the multiplicity of potential conditions and traits.

The following definition of "coal refinery" is put forth and has been used in the selection and discussion of the concepts considered in this report. The definition is:

> COAL REFINERY: A system consisting of one or more individual processes integrated in such a way as to allow coal to be processed into two or more products supplying at least two different markets.

This definition insures that concepts producing a range of refined products from coal for multiple markets are included while excluding processes that produce only one major product supplying a single market, e.g., conventional coal beneficiation or cleaning facilities. It also allows the many existing and developing coal-based technologies, e.g., gasification, liquefaction, advanced beneficiation, to be used singularly or in combination with others in refining coal into a wide range of products for many markets.

## 1.5 FOCUS OF REPORT

The principal objective of this Report to Congress is to identify and describe several concepts that illustrate the products that can be produced in coal refineries, identify potential markets, point out the state of development or commercialization of these concepts, and identify (where appropriate) major R&D needs. Information provided for these concepts includes process descriptions, coal feedstocks, products, applications, development status, and environmental aspects. R&D needs for individual concepts are discussed and are summarized for each of the major categories of coal refineries.

Most of the information presented in this report has been taken from published documents. Contacts with concept advocates and/or other experts were used to supplement or verify published information for almost all of the concepts described in this report. The available information was then summarized for presentation in this report. Some of the information is based on assumptions made by concept advocates which may be optimistic or based on limited experimental data. It has not been possible for the authors of this report to verify all of the assumptions and claims associated with each concept. One of the objectives of any further R&D on coal refinery concepts would be the verification of the important assumptions and operational results that influence product yields, costs, environmental effluents, and other parameters of interest.

There are many concepts that could potentially be classified as "coal refineries." Of these many concepts, twenty-seven are identified and presented in this report. These concepts represent the breadth of products, markets, and development status that exists among coal refinery concepts. For the purpose of better organizing this report, these concepts are somewhat arbitrarily grouped into four general categories according to the principal processing steps used in the concept. Each of these four categories includes concepts that can convert coal into some or all of the products noted in the Congressional directive to DOE. The coal refinery categories are:

1) Devolatilization
2) Gasification
3) Liquefaction
4) Bioprocessing

Chapter Two of this report contains a brief history of some early coal refinery concepts while Chapter Three contains information on the specific twenty-seven coal refinery concepts identified as part of this effort.

Chapter Three also contains further information on the categories used to characterize the coal refinery concepts. A list of the concepts considered in this study is provided. This list is not intended to be all-inclusive of potential coal refinery concepts nor has there been an attempt to make it so. Instead, it is intended to include concepts representative of the wide range of processes, products, applications, and state of development that is exhibited by the numerous coal refinery concepts extant. Brief summaries of each of the concepts are provided with the list of concepts. More information on the twenty-seven concepts is presented in the appendices.

In addition to the list of concepts, Chapter Three contains a summary for each of the four refinery categories. These summaries include generic information on the products, markets, status, and R&D needs for the concepts within each category. The objective of these summaries is to point out the general characteristics and development status for each category of coal refinery concepts.

The final portion of this report consists of four appendices, one for each of the coal refinery categories. More extensive descriptions are provided for each of the concepts. The level of detail, however, varies considerably among the concepts. This variation is due to a number of factors including the amount of published information, the willingness of private sponsors to provide information because of proprietary or patent considerations, and the state of development of the concepts. Therefore, the length of the description for any given concept should not be construed as a measure of its development status or its potential role in the marketplace.

## 2. BRIEF HISTORY OF COAL REFINERIES

Coal consists principally of two of the elements, hydrogen and carbon, that have been key sources of energy for many decades. These same elements are also building blocks for chemicals, feedstocks, and synthetic materials for which markets have rapidly grown in the past 30 to 40 years and for which new markets are being found every day. Much of the demand in these markets is currently met by the other main fossil fuels, natural gas and oil. Reasons for using these other resources include economics, ease of distribution, relative ease in processing oil and gas into the desired chemical structure, and the relative purity of natural gas and most oils as compared to most coals with respect to ash, sulfur, and trace elements. Furthermore, oil and natural gas have been generally available (with some notable and important exceptions) in virtually unlimited quantities and at reasonable prices.

The vast quantities of coal extant in the U.S. and throughout the world, combined with the fact that hydrogen and carbon remain the basic building blocks in the energy and chemical industries, have long spurred interest in processing coal to allow it to be used in more ways than direct combustion. For example, carbonization, or coking, of coal had its beginning near the end of the 16th century in England. Dudley discovered in 1619 that blast-furnace performance was improved when a "coak" produced after certain coals had been "charked" was substituted for the more traditional wood charcoal. The first commercially successful carbonization process was developed by Darby in 1709 [3]. In 1791, William Murdoch patented a process for distilling coal for the production of illuminating gas which he used to light his office and the cottages of coal miners. In 1807, Westminster Bridge and Pall Mall in England became the first public places to be lighted by coal gas and in 1817, Baltimore became the first American city to use coal gas for lighting streets. The first use of coal gas for heating purposes was in 1855 when the Bunsen burner was developed [4].

Friedrich Bergius discovered in 1913 that if coal is treated with hydrogen at high temperature and pressure and in the presence of a catalyst, an oil similar to crude petroleum is produced [5]. There were considerable activities in the development of this process in the 1930s and 1940s. During this time, work was also being done on the Fischer-Tropsch process for indirect liquefaction of coal. Together, these two processes led to the well documented use of coal liquids as transportation fuels for the German war machine during World War II. The SASOL facilities in South Africa for making liquid and gaseous fuels and chemicals from coal also grew out of this early work on the Fischer-Tropsch process.

More recent history of coal refineries begins with the oil embargo and natural gas shortages of the early 1970s. R&D efforts on many alternative energy sources, including new coal-based applications, were initiated in part to relieve the U.S. dependence on energy imports, particularly oil. Concurrent with the desire to develop alternative energy sources was a growing awareness and concern for environmental issues including air and water pollution, leaching from landfills, and coal-mine runoff. Thus, the development of alternative energy sources had to be done in the context of environmental consciousness as well as economics.

Some of the concepts examined in the 1970s were intended to yield a multiplicity of products that could meet demands in several markets. One such concept was referred to as "Project POGO" where the acronym stands for Power-Oil-Gas-Other [6]. This concept, developed by the Ralph M. Parsons Company for DOE and its predecessor agencies, combined the hydroliquefaction, pyrolysis, and gasification conversion processes with combustion turbines, heat-recovery steam generators, and steam turbines to yield a broad slate of synfuels plus electricity. Several options were examined as part of Project POGO. The specific concept described in Ref. 4 had a feed rate of 45,000 tons per day of clean bituminous coal and produced approximately 150 million standard cubic feet of Substitute Natural Gas, 13,000 barrels of $C_3$-Liquefied Propane Gas, 2,000 barrels of $C_4$-Liquefied Propane Gas, 35,000 barrels of gasoline, 27,000 barrels of distillate fuel oil, 1,600 tons of a premium grade coke, and 1,000 MW of electricity. Due to the large number of products from this facility, it would be very complex and would require a very large capital investment to build and operate.

Other concepts focussed on a more limited product slate. A great deal of R&D has been done by both the private sector and DOE in the area of coal liquefaction. Several processes including the Solvent Refined Coal (SRC) I and II, H-Coal, Exxon Donor Solvent, and Two-Stage Liquefaction have been developed through these R&D activities. The 17,000 ton/day Great Plains Gasification Facility in North Dakota started operation in 1984 with the objective of producing pipeline-quality gas from coal [7]. This facility grew out of work supported by DOE.

Several concepts investigated by the Electric Power Research Institute (EPRI) and others have electricity as a major product. Byproducts from these concepts include carbon dioxide, trace elements, and ash which are normally considered to be wastes in conventional power plants. Other products might include liquid or gaseous fuels, chemical feedstocks, or chemicals. Two concepts that demonstrate this philosophy center around the use of fluidized-bed combustion or the combustion turbine/combined cycle for power generation coupled with carbon dioxide recovery and resource recovery from the solid waste streams [8, 9]. Although the specific design details differed, both combustion options were integrated with carbon dioxide recovery and byproduct metal recovery from ash.

Reference 10 describes an "integrated energy facility" which is, in essence, a coal refinery consistent with the definition given in Section 1.4 of this report. This general concept uses coal gasification to produce chemicals, feedstocks, and fuels in addition to electricity. One of the principal driving forces in developing multiproduct concepts is the belief that "Coproducing electricity and chemical products in a fully integrated energy facility could potentially reduce the cost of generating electricity by approximately 40 percent, while minimizing air emissions and solid waste production" [11]. In this concept, coal is gasified to produce a low- or medium-Btu fuel gas. In the course of this gasification process, elemental sulfur and carbon dioxide can be recovered, steam can be produced, and the slag from the gasification process can be used in road construction or as a building material. The fuel gas can be used in advanced combined-cycle units for the production of electricity or in a chemical production facility to yield specific fuels such as methanol, gasoline octane enhancers, hydrogen, or substitute natural gas. It could also be used in the production of ammonia or other industrial chemicals or feedstocks. This "integrated energy facility" serves as the foundation for several of the coal refinery concepts discussed in Sec. 3.2 and the Appendices of this report.

# 3. COAL REFINERY CONCEPTS

## 3.1 REFINERY CATEGORIES

As noted previously, the coal refinery concepts considered in this report are divided into four major categories. The basis for these categories is the principal processing step(s) used in refining the coal. Rudimentary descriptions of the refinery categories are as follows.

### 3.1.1 Devolatilization

The devolatilization category contains concepts that process coal under non-oxidizing or mild-oxidation conditions. The immediate products from concepts in this category usually include a gas that can be used as a fuel or as a feedstock for chemical production, some liquids, and a high-carbon char. These immediate products however, are sometimes processed further and may not always correspond with the final products. In general, the degree of coal conversion is usually more limited than that associated with the gasification or liquefaction concepts.

These concepts generally use low temperature and pressure (approximately 1,000-1,500°F and near atmospheric pressure) conditions to drive off the volatiles in the coal and leaving a devolatilized, solid char behind. The volatiles are then condensed into a variety of liquid products while the char can be used as a solid fuel, mixed into a slurry, or upgraded into a higher-value product such as form coke or carbon black. The liquid yield from these concepts is generally quite low, at less than one barrel of liquids per ton of coal.

### 3.1.2 Gasification

Gasification concepts rely on more severe conditions to convert the coal into a gaseous product consisting of large fractions of carbon monoxide, hydrogen, carbon dioxide, and water. This gaseous product can than be used in one or more ways. It can, after removal of potential pollutants, be burned in a combustion turbine for the production of electricity. It can be used as a chemical feedstock, or its composition can be adjusted for conversion into products such as substitute natural gas, pure hydrogen, methanol, ammonia, acetic anhydride, and various hydrocarbon fuels which can then be used in various markets.

### 3.1.3 Liquefaction

Concepts in the liquefaction category are generally configured to yield a greater fraction of liquid products than concepts in other categories. These liquids can potentially be used as fuels or as feedstocks for the full slate of petrochemical products.

Energy and material inputs to these refineries, in addition to the coal, range from natural gas to very heavy crude oils. Bitumen and heavy residual oils are also possibilities for use in some liquefaction concepts.

The output products can be similar to those of a large petroleum refinery or they can be more narrow, such a synthetic crude oil.

### 3.1.4 Bioprocessing

The development of micro-organisms, both natural and tailored, for use in commercial processes is increasing. Research on biotechnology for coal processing is a growing area and some of the potential applications may be considered to be coal refineries. For this reason, a bioprocessing category of coal refineries has been included that covers solubilizing of coal and conversion to gaseous and liquid products as well as the use of bio-organisms in the conversion of synthesis gas generated by conventional coal gasification step into liquid and gaseous fuels.

In most cases, the bioprocessing effort has been in the research laboratory stage with some work at the bench-scale level. Engineering work has been done for some processes that may evolve into commercial activities. As compared to the other general categories of coal refineries, the bioprocessing concepts are generally at a lesser state of development.

## 3.2 POTENTIAL COAL REFINERY CONCEPTS

As noted earlier, there are many concepts that could be classified as coal refineries. Even though the definition given above restricts the number of refineries, a great many concepts may still meet the definition. For this reason, several general classes of concepts that may meet the strict definition of a coal refinery have been excluded from consideration in this report. For example, the Integrated Coal-Gasification Combined Cycle (IGCC) is capable of producing both electrical power and elemental sulfur or sulfur compounds such as sulfuric acid. However, the main product from the IGCC is electrical power with the sulfur products being more properly classified as byproducts which, while perhaps being a marketable commodity, is not a principal reason to build and operate an IGCC. While the IGCC itself is not considered to be a coal refinery, there are several coal refinery concepts discussed in this report which are based on the coal gasification process and which produce electrical power along with one or more additional major products.

Coal preparation or beneficiation plants are typically designed to yield a product that is combusted for the production of steam. Thus, coal preparation plants generally address a single market and are therefore not considered to be coal refineries. They have been used however in conjunction with other processes to create concepts that are considered to be coal refineries.

A third general type of facility that has been excluded from consideration in this report is that of cogeneration. Although this type of facility generally has two products (typically electricity and steam) that supply different markets, only one basic coal-based processes (usually simple combustion) is involved in the production of these products. The steam product is supplied to its market by simply by-passing the turbine or extraction from the turbine-steam cycle used to generate electricity. Therefore, cogeneration is really using a single product (steam) in two ways and thus not considered to be a coal refinery concept.

The coal refinery concepts presented in this report are listed and briefly summarized below. More detailed descriptions are included in the appendices.

### 3.2.1 Devolatilization Concepts

CALDERON

The Calderon Energy Co. has a proprietary multi-stage devolatilization process that pyrolyzes coal to produce a rich fuel gas and a residual char. The rich gas is then converted into methanol, which is both a chemical feedstock and potential utility/transportation fuel. The char is gasified with air to yield a lean fuel gas. This lean gas is conditioned for use in a gas turbine to generate electricity. A 24 ton per day process development unit (PDU) has been undergoing testing at Alliance, Ohio.

CHARFUEL

This concept uses the short residence time decomposition and volatilization of coal. Two types of products are generated, one being a char-liquid slurry for utility/industrial combustion purposes, and the other being a slate of liquids (naphtha, BTX, methanol) that can be used as power plant fuels or upgraded to produce chemical feedstocks.

COAL LIQUID AND COKE MILD GASIFICATION

Coal is continuously processed under mild conditions to extract high-quality liquids from coal and to generate char. The coal-derived liquids can be blended with petroleum stocks in special formulations to produce gasolines and diesel fuels. The char is converted into form coke usable in a blast furnace for reduction of iron ore. The process is being demonstrated in a 1,000 pound per hour pilot plant near Bristol, Virginia.

ENCOAL

This conversion concept, using a rotary grate pyrolysis unit, yields a liquid product and a dry and fast-burning process-derived solid fuel. The coal derived liquid is similar to a low-sulfur No. 6 fuel oil, while the process derived solid has enough volatiles for use as a boiler fuel. This process, originally developed by SGI International and the Shell Mining Company in the mid-1980s, has proceeded through a 2.5 tons per day process development unit. To date, most of the work has been with Powder River Basin subbituminous coals. Construction has begun on a 1,000 ton per day demonstration unit near Gillette, Wyoming as part of DOE's Clean Coal Technology Demonstration Program.

HYDROCARB

The two steps of this concept involve the hydropyrolysis of coal to produce methane, with the subsequent thermal decomposition of the methane to carbon black and hydrogen, with the hydrogen recycled. A potential third step converts the carbon monoxide formed in the hydropyrolysis step to methanol which could be directly used as a fuel or, if economics warranted, could be converted to other products such as gasoline. The main products are a clean carbon black fuel and coproducts consisting of a hydrogen-rich gas and methanol if the third step is incorporated into the concept. The ash-free, sulfur-free, and nitrogen-free particulate carbon black alone or in slurry form can be used as a heavy duty fuel for transportation or for utility power plants. Components of this concept have been tested at bench scale.

IGT MILD GASIFICATION

The IGT Mild Gasification (IMG) process utilizes a fluidized-bed coal pyrolysis reactor to produce char and coal-derived liquids. The char and liquids can be upgraded to produce form coke, electrode binders, roofing material, and chemicals. The process has been evaluated at the 100 pound per hour process research unit (PRU) scale, and has progressed to the point that a 24 tons per day cost-shared PDU has been selected for award under a competitive DOE Program Research and Development Announcement (PRDA).

MARSHALL OWEN

Marshall Owen Enterprises plans to use a continuous carbonization process licensed by the Coalite Group of Great Britain. This concept uses a bituminous coal to produce a smokeless char that can be used to replace wood due to its lower emissions of pollutants. The process also yields a coal-derived liquid that can be upgraded to become a chemical feedstock, industrial fuel, and/or transportation fuel. The Coalite process has been in commercial operation for over 50 years in England although the application there has used a batch operation scheme.

SFUEL

A smokeless fuel (SFUEL) concept has been proposed by the Institute of Chemical Processing of Coal (Poland) for the production of smokeless char fuel and a fuel gas for electric power production. The smokeless char would be used extensively in Poland where a large fraction of the energy market (including residential heating) is currently supplied by coal. Circulating fluidized bed (CFB) technology is used in the devolatilization process. A 7.2 ton per day CFB test reactor has been constructed in Poland to demonstrate the technical viability of the concept. A 200 ton per day pilot plant is being commissioned with startup expected in 1993.

UNDEERC-AMAX MILD GASIFICATION
The Energy and Environmental Research Center of the University of North Dakota and the AMAX R&D Center have developed a mild gasification process that utilizes a fast fluidized-bed coal pyrolysis reactor to product char and coal-derived liquids. The solid and liquid products can be upgraded to produce metallurgical coke substitute, carbon black, and chemical feedstocks. The process is being evaluated at the 100 pound per hour process research unit scale. A conceptual design for a 1,000 TPD of coal demonstration plant is being prepared.

WRI-AMAX MILD GASIFICATION
The Western Research Institute (WRI) and the AMAX R&D Center have developed a mild gasification process to produce coal-derived liquids and a solid char. In the WRI-AMAX process, the solid char is upgraded into higher value products such as carbon powder for use in rubber tires or anode carbon for use in aluminum production. At the current time, liquids from the WRI-AMAX process will not be upgraded to highly refined products but will be used for coal dust suppression, manufacturing carbon anodes, and as a diesel fuel feedstock. Demonstration tests at the 50-100 pound per hour process development unit level have been carried out at WRI in Wyoming and AMAX R&D in Golden, Colorado.

### 3.2.1 Gasification Concepts

COPRODUCTION OF ACETIC ANHYDRIDE, ACETIC ACID, AND METHANOL
Tennessee Eastman Co. (TEC), a division of Eastman Kodak Co., developed and commercialized a process using synthesis gas from coal for the production of acetic anhydride, acetic acid and methanol. A 900 tons per day plant in Kingsport, TN started operations in 1983. This plant uses high-sulfur coal to produce about 500 million pounds of acetic anhydride per year, which is used for production of TEC's film and other products. While the acetic anhydride is the primary chemical produced in this plant, other chemicals, e.g., methanol and acetic acid, used in TEC's operation are also produced. Sulfur is recovered and sold as a byproduct from this process.

COPRODUCTION OF METHANOL AND ELECTRICITY
This gasification concept is an example of EPRI's definition of a coal refinery in which electrical generation becomes one element of a broader, resource-processing capability due to the coproduction of commercial chemicals (e.g., methanol). The methanol liquid-phase synthesis, using a single-pass (once-through) process, is integrated into a coal

gasification combined-cycle power plant. The liquid-phase methanol synthesis process developed by Air Products and Chemicals, Inc. has been demonstrated at 60 to 70 barrels per day in La Porte, TX, and will be integrated with the Cool Water IGCC facility.

COPRODUCTION OF SNG, ELECTRICITY, METHANOL, AND CHEMICAL INTERMEDIATES

This concept is based on operations at the Great Plains Coal Gasification Plant in Beulah, North Dakota. Incoming coal is first screened to separate coal fines, which are utilized in a conventional pulverized coal power plant to generate electricity. The remaining larger-sized coal is gasified to produce a synthesis gas that is converted to substitute natural gas (SNG). A small amount of the synthesis gas is diverted to a commercial methanol synthesis unit.

COPRODUCTION OF UREA AND ELECTRICITY

This concept, advanced by the Tennessee Valley Authority (TVA), would integrate the production of urea with power generation in a combined cycle system. Coal is gasified to produce a synthesis gas containing hydrogen and carbon monoxide. The hydrogen is then separated from the synthesis gas through the use of the membrane gas-separation technology. The hydrogen reacts with nitrogen to form ammonia and then with carbon dioxide to form urea, which can be used as a chemical feedstock for materials such as fertilizer. The remainder of the synthesis gas is sent to an advanced combined-cycle power plant and burned to produce electricity. Ammonia production from coal was demonstrated in the 1980s by TVA using a 150 tons per day Texaco gasifier.

DUAL PRODUCTION OF AMMONIA AND ELECTRICITY

This process integrates current technology for ammonia (chemical feedstock for fertilizer, etc.) production with a high-performance electric generation technology. Coal is gasified to produce a synthesis gas stream, with a large fraction of the syngas stream feeding an advanced combined-cycle power plant. The remaining fraction of the syngas is cleaned of pollutants and adjusted in composition for the production of ammonia. A representative large-scale (1,000 tons per day) coal-to-ammonia plant is operated by Ube Industries, Ltd. (Japan).

ONCE-THROUGH FISCHER-TROPSCH PROCESS WITH POWER GENERATION

A single-pass Fischer-Tropsch (F-T) technology is combined with combined-cycle power generation to coproduce chemical feedstocks/liquid fuels such as naphtha and middle distillates with electrical power production. Coal is gasified to produce synthesis gas, which is then catalytically converted to liquid hydrocarbons and/or oxygenated fuels. Any unreacted synthesis gas and light hydrocarbons would be used to run power turbines rather than be recycled for further conversion into liquids. The MITRE Corp has developed models for DOE for the integration of the once-through F-T process; an example of a commercial F-T plant is the Sasol facility in South Africa.