Two Concepts in the Production of Liquid Fossil Fuels

Mary E. Chenoweth, Joseph F. Benzoni, William Micklish

May 1987
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Prepared for
The Argonne National Laboratory

RAND
PREFACE

This Note, which documents the results of an analysis of two novel approaches to the production of liquid fuels from coal and oil shale, was performed for the Argonne National Laboratory. The project's objective was to conduct in-depth analyses on two novel techniques that might offer major benefits over current fossil fuel technologies. The two concepts selected were lignin/coal/oil coprocessing and aboveground retorting of oil shale using high-power microwaves. The authors analyzed the potential product cost impacts of both concepts and investigated their implied economic chokepoints.

This research follows from earlier RAND work performed for the Department of Energy in which the authors were asked to identify novel concepts for the conversion and production of fossil fuels. This Note should be of interest to those concerned with the enhancement of domestic production of liquid fossil fuels through advanced technologies.
SUMMARY

The United States is largely dependent on liquid fossil transportation fuels that are derived from petroleum sources. However, the United States has vast domestic resources of solid fossil fuels, such as coal and oil shale, that can be converted into liquid fuels using technologies derived from processes that have existed for many years. At present, these technologies produce fuels that are considerably more expensive than petroleum products produced from conventional refining technologies. Without a significant cost reduction in these technologies, their use will not be economically practical.

In recent years, the stability in petroleum costs has reoriented the approach to research on coal and oil shale conversion within this country. There has been a deemphasis, particularly in the Department of Energy, on the commercial demonstration of conversion technologies in favor of basic research on innovative or very novel approaches to conversion technologies. Part of this latter effort has been led by the Argonne National Laboratory (ANL). In this study, RAND has assisted ANL in their effort to identify promising candidate technologies and to critically analyze in greater depth the most promising ones. This study analyzes two--lignin/coal/oil coprocessing and aboveground microwave retorting of oil shale--to assess their promise for significantly lowering the costs of producing liquid hydrocarbon fuels from domestic solid fossil resources.

Research in the processing of coal and oil has been vigorously pursued in the United States, Canada, and other countries for some years. Although promising, coal/oil coprocessing is not without its problems, which include coal/oil type incompatibilities and very severe operating conditions. In this Note, the concept of coal/oil coprocessing has been expanded into a more diverse approach termed "generalized coprocessing," defined as the processing of coal with other feedstocks, not necessarily petroleum only. In particular we analyze the processing of lignin (a by-product of papermaking), coal, and oil.
It has been shown that coal can be processed separately with lignin and oil; but the processing of all three feedstocks, to our knowledge, has never been attempted.

Technically, there are many reasons to suggest that this novel concept has promise: Lignin exhibits a catalytic quality as it undergoes liquefaction at temperatures as low as 300 degrees C. It appears to increase destabilization of the coal molecule at lower temperatures and moderates pressures compared with conventional liquefaction processes, making it more susceptible to hydrogenation and ultimately liquefaction. The addition of lignin to the coal/oil coprocessing technology is believed to improve chemical compatibility problems between paraffinic oil and aromatic coal molecules. The supply of lignin could be a limiting factor in the commercial realization of this process, given current conditions. However, the technical success of this concept could spur the search for sources of lignin other than the paper manufacturing industry.

Prior attempts at using electromagnetic energy to retort oil shale have focused upon slow heating using low power radio frequency or microwave radiation, generally applied to in-situ shale beds. Although these processes have been shown to be technically feasible, they consume large amounts of electricity, because of heat losses to the surrounding environment. The novelty of aboveground microwave retorting lies in the use of microwave devices sufficiently powerful to produce a high rate of retorting of the kerogen. The proposed process takes advantage of the low thermal conductivity of shale to minimize the amount of heat loss to the mineral portion of the oil shale and subsequent salt formation.

The energy benefits of this process lie mainly in the minimization of heat absorbance by the shale rock. The absolute maximum benefit, i.e., the difference between complete heating of the rock and no heating of the rock during the process of heating the kerogen to oil, is a significant reduction in heat requirement. In reality, the rock will be heated somewhat. The process proposed should cause less heat absorbance by the shale than other microwave processes because of rapid and selective heating of kerogen by microwave energy and its removal before a substantial amount of heat is conducted to the mineral matter.
In principle, this process could significantly reduce electricity consumption in comparison with slow electromagnetic heating processes such as in-situ radio frequency heating. At the same time, rapid heating is expected to produce 100 percent of the Fischer Assay amount of oil contained in the shale. In addition, the environmental impact will be decreased, relative to conventional retorting, because of reduced production of leachable salts and combustion products. It is recommended, therefore, that an engineering scale-up of this concept be undertaken to test the range of working parameters and thereby to establish the actual economic impacts to shale oil recovery by this method.
ACKNOWLEDGMENTS

The authors of this Note would like to express special thanks to those individuals whose contributions were invaluable in the course of our work. We appreciate the guidance provided by our project monitor, Mr. Thomas Wolsko. We would like to acknowledge those contributions made by our RAND colleagues Edward Merrow and Kenneth Phillips. As the first project leader of this research, Merrow provided the original framework and formulation of the research problem; he also played a significant role in forging several concepts explored in this report. We appreciate the invaluable suggestions and contributions made by our technical reviewer, Kenneth Phillips.
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I. INTRODUCTION

BACKGROUND

The United States has large markets for liquid hydrocarbon fuels, a substantial portion of which cannot be reasonably served by other means. For many years the domestic production of petroleum has fallen short of consumption and this situation is expected to worsen. Given our growing reliance on foreign sources of petroleum oil, the search for economical alternatives to conventional hydrocarbon supplies becomes increasingly important for this nation's energy security. In this study, we assess two alternatives to conventional liquid fuel supplies that employ novel technologies.

An appropriate response to the trend of importing petroleum oil is to use more domestic resources. Enhanced oil recovery (EOR) techniques including the potential use of heavy oil/tar sands will be applied to U.S. oil fields to increase total recovery and prolong the availability of domestic supplies. Forecasts have concluded, however, that such EOR activities and new frontier areas will not be able to reverse the slow but steady decline in domestic oil production. Therefore, other resources must be tapped if domestic liquid fuel supplies are to be available in sufficient quantity to bring U.S. dependence on imported liquid fuels to tolerable levels. Coal and oil shale, among other domestic resources, are two alternative fossil fuel resources that can be used to produce quality liquid fuel supplies on a scale consistent with the future national need.

Conversion processes for coal and oil shale have been in existence for some time. Coal-derived gas was used extensively in the 19th and 20th centuries. In addition, conversion processes for the production of synthetic fuels were available before the time that naturally available liquid and gaseous fossil fuels were discovered.\(^1\) The later discovery of vast quantities of relatively inexpensive oil preempted continued development of synfuels and led to a petroleum-based energy economy in the 20th century.

\(^1\)Crow and Hager (1985).
Research in fossil energy, in particular solid fossil fuels, has decreased. Ten years ago eleven cents out of every federal R&D dollar went to energy research; today that figure has dropped to three cents with the deepest cuts faced by fossil energy.² Concurrently, fossil energy research in the private sector has decreased sharply. Faced with these trends, it is imperative that the U.S. government ensure that promising innovative research in these areas continues.

Although conversion technologies for solid fossil fuels have existed for some time, they all suffer from deficiencies or "chokepoints" that render them economically, technologically, or environmentally unattractive. The relative stability of current petroleum prices offers the United States the opportunity to concentrate on more novel and innovative technologies that directly address these difficulties.³

PROJECT OBJECTIVES

Our objective for this project was to conduct in-depth analyses on two novel technologies that might offer major benefits over current fossil fuel technologies. In this research, a novel concept has been defined as a technical approach that has the potential of significantly reducing the product cost of fossil fuels and that falls into one of the following categories:

- An entirely new technical approach to fossil energy production or conversion;
- An approach that was previously explored and discarded for reasons now inappropriate;

³In this Note current technology refers to systems that have been brought close to commercial production. These include Exxon Donor Solvent, H-coal, and Solvent Refined Coal for coal liquefaction and Tosco II and Union B retorts for oil shale.
• Concepts presently being explored at a low level of support but whose promise might be underestimated; and
• New applications of existing technology.

We developed a list of novel and revolutionary technical approaches to fossil energy production and conversion in several of the areas that fall under the purview of the Department of Energy's (DOE) Office of Oil, Gas, Shale and Coal Liquids (OGSCL). We then screened these candidate technologies to isolate those that appeared to hold the most promise for providing major benefits. Following this process and discussions with Argonne National Laboratory (ANL), two technologies—lignin/coal/oil coprocessing and aboveground retorting of oil shale using microwave energy—were selected for in-depth analyses. (For further information on this screening process for concept evaluation refer to Appendix A.)

The concepts described in this Note largely lacked the necessary data to conduct the rigorous cost analyses one desires in any technical assessment of a concept. The technologies we discuss in depth are novel and have not received extensive experimental investigation. Therefore, the lack of key data is not surprising. In the absence of such data, we often had to turn to technical indicators that pointed to the direction of cost change and made calculations whenever possible to bound important economic problems.

TWO NOVEL CONCEPTS

It has been shown that coal and oil, and coal and lignin, can be processed to produce liquid fuels; the processing of all three feedstocks to our knowledge has never been attempted. Intuitively, the latter approach offers significant advantages over the first process such as addressing problems of solvent and coal incompatibility and severe operating conditions required to produce these liquid fuels, two areas that if addressed could bring major benefits to the processing of

---

*Depending on the process involved, some indicators included temperatures, pressures, rate of heating, efficiencies, power required, etc.*
coal and oil. On the other hand, lignin volume requirements for this concept could pose an economic problem if the process were adopted on a commercial scale—a single industry largely determines the supply of lignin, which is relatively small given the potential demand this novel process would require.

Prior attempts at using electromagnetic energy to retort oil shale have focused upon slow heating using low power radio frequency or microwave radiation, generally applied to in-situ shale beds. Although these processes have been shown to be technically feasible, they are high consumers of electricity because of heat losses to the surrounding environment. In contrast, the high power microwave radiation retort described in this Note might greatly reduce heat loss; calculations indicate that the theoretical maximum reduction in the retorting energy requirements possible by this method (based on no heating of the shale rock) is six times less than other methods (based on completely heating the rock). In addition, the quality of oil produced is expected to be good, and the environmental impact is expected to be lessened relative to conventional retorting processes.

ORGANIZATION OF THE NOTE

In Sec. II we analyze lignin/coal/oil coprocessing and in Sec. III we offer the results of the study of aboveground microwave retorting of oil shale. Section IV provides our conclusions and recommendations on these two novel technologies. Appendix A describes the methods used to choose novel technologies for further consideration; Appendix B presents a typology of novel technologies and a survey of candidate technologies; Appendix C explains the analysis performed to approximate lignin supplies required for conversion of coal using the lignin/coal/oil process; and Appendix D contains calculations that accompany discussions in Sec. III.
II. LIGNIN/COAL/OIL COPROCESSING: AN EXAMPLE OF GENERALIZED COPROCESSING

As an abundant source of fossil fuels in the United States, coal offers an important potential supply of liquid transportation fuels for this nation if cost-effective technologies for its conversion can be found. German advances made by 1913 in direct and indirect liquefaction are the basis of modern coal conversion technologies. The first type of liquefaction directly converts coal to liquids in the presence of hydrogen and is called direct liquefaction.\(^1\) Indirect processes transform two gasified coal constituents—carbon monoxide and hydrogen—into liquids through catalytic reactions. (Direct liquefaction will only be discussed in this section.) This section describes a novel approach to producing coal liquids that uses the additional feedstocks or starting materials, oil and lignin. The oils used in the processing of coal and oil are the heavier, less economically valuable oils, such as refining residua, heavier oils, or bitumen—a pitch-like material. Lignin is a naturally occurring polymer found in woody plants, thought to be molecularly similar to coal.

Coal/oil coprocessing, or the simultaneous processing of oil and coal, has been considered a bridging process between current refining technologies and direct liquefaction processes for the production of more valuable liquids from solid feedstocks. We explore a novel approach to coprocessing, one in which it is generalized to include the processing of different feedstocks with coal, not just oil. As an example, we study the use of lignin with coal and oil.

In the remainder of this section we briefly describe liquefaction and conventional coprocessing technologies and end with a detailed analysis of lignin/coal/oil coprocessing. Before we discuss the concept of generalized coprocessing, we will note the current coal reserves in the United States and explain the basic economic limitations of

\(^1\)Direct liquefaction technologies include pyrolysis, solvent extraction, and catalytic liquefaction processes. For more discussion, see Nowacki (1979).
conventional direct liquefaction processes. The discussion of
generalized coprocessing and disadvantages of current coal/oil
coprocessing technologies leads to the topic of generalized
coprocessing, and, in particular, lignin/coal liquefaction. These
subsections prepare for the feature topic—the proposed novel technology
of lignin/coal/oil coprocessing. We explain this hybrid technology and
then analyze its potential to save costs; we also suggest areas of
future research for this novel concept. The section concludes with a
suggested research agenda necessary to further develop this concept.

COAL RESERVE BASE

Estimates of the U.S. coal reserve base indicate that if economic
processes are developed for the conversion of coal to liquid fuels, the
supply potential of coal liquids would far surpass the current known
worldwide reserve base of petroleum oil. Vast coal reserves are located
in many areas of this country, as shown in Fig. 1. Of the four ranks
of coal—anthracite, bituminous, subbituminous, and lignite—bituminous
coals produce the highest liquid yields. Anthracite tends to primarily
produce gas and is generally not considered to be a good liquefaction
coal.

The U.S. Geological Survey estimates the total identified coal
resources at 1600 billion tons with perhaps the same amount in
unidentified resources. By comparison, in 1978 we consumed and exported
a total of 0.6 billion tons of coal. Estimates of coal, heavy oil,
bitumen, and crude petroleum reserves are shown in Table 1, along with
calculated barrel equivalents of coal liquids. As a rough
approximation of potential liquid hydrocarbons, we calculated the liquid
c coal equivalents of these reserves. These figures represent theoretical
upper limits of coal liquids that could be produced from these reserves.
(Because conversion yields differ for each process, for these purposes
we assumed that coal was completely converted to liquid.) Note that if
c coal could be completely converted into liquid fuels with no loss of
energy, the resulting potential liquid hydrocarbon supply would be over

\(^2\)Nowacki (1979), p. 4.  
\(^3\)For original data, see Nowacki (1979), p. 5.  
\(^4\)For the conversion of coal into coal liquids, conversion factors
listed in Table 1 are based on heating value and represent upper
Fig. 1 -- Coal reserves in the United States

250 times greater than our annual consumption of oil in 1979 (6.8 billion barrels). Such ratios offer important implications for the future U.S. supplies of liquid fuels. Coal, if economically converted to liquids, could greatly augment our current fuel supplies.

limits. In reality, yields will be lower. Conversion factors were found in Energy Factbook (1980), p. 777.
6World crude oil data are from Hanchey and Holcomb (1985), pp. 1-22.
Table 1
DEMONSTRATED COAL, HEAVY OIL, AND BITUMEN RESERVE BASE
OF THE UNITED STATES

<table>
<thead>
<tr>
<th>Resource</th>
<th>Billion Tons</th>
<th>Billion Barrels</th>
<th>Conversion Factor Used for Tons to Barrel Equivalents</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bituminous</td>
<td>233</td>
<td>1048</td>
<td>4.498</td>
</tr>
<tr>
<td>Subbituminous</td>
<td>165</td>
<td>742</td>
<td>4.498</td>
</tr>
<tr>
<td>Lignite</td>
<td>28</td>
<td>126</td>
<td>4.498</td>
</tr>
<tr>
<td>Anthracite</td>
<td>7</td>
<td>31</td>
<td>4.361</td>
</tr>
<tr>
<td>Total</td>
<td>433</td>
<td>1947</td>
<td></td>
</tr>
</tbody>
</table>

Oil reserves
- Heavy oil, bitumen, U.S. 27
- Crude oil, U.S. 30
- Crude oil, world 670

LIQUEFACTION AND CURRENT ECONOMIC CONSTRAINTS

The conventional approach to direct coal liquefaction has been the conversion of coal to liquids using coal as the only feedstock. In these processes, coal is dissolved in coal-derived solvents produced downstream and recycled forward, heated to 425-450 degrees C in the presence of hydrogen under pressures of 1500-2000 psi. The residence time or average time material remains within the reactor ranges between 30 and 90 minutes. The severity of these conditions reflects the difficult problem of converting coal, a relatively hydrogen-poor solid material, into a liquid, more hydrogen-rich fuel. The introduction of other feedstocks in liquefaction technologies, such as coal/oil coprocessing, addresses some but not all of these problems.

In general, to convert coal into more valuable liquid products, its long carbon chains mainly composed of aromatic rings must be broken into smaller fragments. Coal liquefaction processes typically rupture the carbon-carbon bonds in coal to form very reactive fragments. To produce high liquid yields these reactive fragments must be stabilized as small
entities to inhibit these chains from reforming or polymerizing. (Polymerization can result in a solid and less reactive mass.) Stabilization occurs by adding hydrogen atoms usually supplied by a hydrogen-donating solvent or by gaseous hydrogen. Hydrogen requirements are central to chemical and economic requirements for direct liquefaction processes.

It has been shown that coal liquids can produce high quality, high octane gasoline. An important chemical difference between the starting material coal and one of the desired products such as gasoline is its hydrogen content, measured in terms of its hydrogen-to-carbon ratio. The addition of hydrogen to coal to form liquids and ultimately end-product fuel is very costly in part because of hydrogen production costs. For this reason, relative costs can be inferred from the hydrogen content of respective materials. All things being equal, the greater the difference in hydrogen-to-carbon ratios between starting material and end product, the greater the associated costs in producing the end product.

Table 2 describes the molecular composition of the different types of coal, oil, lignin, and some desired end-use products. In particular,

<table>
<thead>
<tr>
<th>Fuel</th>
<th>C</th>
<th>H</th>
<th>O</th>
<th>N</th>
<th>S</th>
<th>H/C (atomic ratio)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bituminous</td>
<td>85</td>
<td>5</td>
<td>8</td>
<td>1</td>
<td>1</td>
<td>0.7</td>
</tr>
<tr>
<td>Subbituminous</td>
<td>75</td>
<td>5</td>
<td>17</td>
<td>2</td>
<td>1</td>
<td>0.8</td>
</tr>
<tr>
<td>Lignite</td>
<td>70</td>
<td>6</td>
<td>21</td>
<td>1</td>
<td>2</td>
<td>1.0</td>
</tr>
<tr>
<td>Bitumen</td>
<td>84</td>
<td>10</td>
<td>1</td>
<td>--</td>
<td>5</td>
<td>1.5</td>
</tr>
<tr>
<td>Lignin</td>
<td>56</td>
<td>6</td>
<td>35</td>
<td>&lt;1</td>
<td>&lt;1</td>
<td>1.3</td>
</tr>
<tr>
<td>Crude oil</td>
<td>86</td>
<td>13</td>
<td>--</td>
<td>--</td>
<td>1</td>
<td>1.8</td>
</tr>
<tr>
<td>Gasoline</td>
<td>86</td>
<td>14</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>2.0</td>
</tr>
</tbody>
</table>

\(^7\)Coal data represent averages of samples 33544 and 33546. See Philp et al. (1982), p. 147. Values for bituminous and subbituminous coals (Alberts) represent averages.

\(^8\)Data for bitumen represent averages.

it is important to note the last column, hydrogen-to-carbon ratio. Bitumen, a coprocessing feedstock, has a higher ratio than coal and gasoline has an even higher ratio. The proposed novel technology described here uses the compound lignin, which has a higher hydrogen-to-carbon ratio than bitumen. Coal has a much lower hydrogen/carbon ratio than gasoline.

One of the major economic drawbacks of coal liquefaction processes is their high hydrogen requirement. This requirement translates to cost in the following way: The largest single component in product cost for coal liquids produced from current liquefaction processes is capital requirements. As Table 3 shows, over 50 percent of the product cost of coal liquefaction is due to capital costs; thus, changes in capital costs will have the single most important economic impact on the production of coal liquids.

(The category for "other capital

<table>
<thead>
<tr>
<th>Cost Category</th>
<th>Percentage of Total Plant Costs (Before Hydrotreating)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plant investment</td>
<td>59</td>
</tr>
<tr>
<td>Other capital required</td>
<td>17</td>
</tr>
<tr>
<td>Capital recovery</td>
<td>10</td>
</tr>
<tr>
<td>Coal</td>
<td>7</td>
</tr>
<tr>
<td>Other operating costs</td>
<td>7</td>
</tr>
</tbody>
</table>

Cost data for coprocessing technologies are not as complete or available in the open literature as the more near-commercial processes. Thus, although cost information for conventional technologies is referenced, should data for coprocessing technologies become available in the future, they should be used instead.

Table 3 costs represent average costs for EDS, H-Coal, HRI, and Lummus ITSL coal liquefaction processes. See El Sawy et al. (1986).
required" includes start-up costs, working capital, initial charge of catalyst and chemical, etc.)

Information in Table 4\(^\text{12}\) represents plant construction cost or capital for on-site facilities.\(^\text{13}\) Looking just at these capital costs, nearly one-third can be attributed to hydrogen-related infrastructure, which is concerned with the generation, recovery, purification, and use of hydrogen.

THE CONCEPT OF GENERALIZED COPROCESSING

Earlier coal conversion research was primarily concerned with using coal as a single feedstock and converting it to liquids. This represents the traditional approach to liquefaction. Coal/oil coprocessing has been considered as a bridging technology between refining processes and direct liquefaction technologies. It is the conversion of coal and oil feedstocks into liquid hydrocarbon fuels. Coprocessing research has gained momentum in recent years. The use of

<table>
<thead>
<tr>
<th>Cost Item</th>
<th>EDS</th>
<th>H-Coal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Liquefaction</td>
<td>31</td>
<td>40</td>
</tr>
<tr>
<td>Hydrogen generation, compression, and recovery</td>
<td>33</td>
<td>29</td>
</tr>
<tr>
<td>Other</td>
<td>36</td>
<td>31</td>
</tr>
</tbody>
</table>

\(^\text{12}\) Hemming, Holmes and Teper (1983), pp. 57, 58, 60.
\(^\text{13}\) EDS figures represent an average of once-through and bottoms recycle.
other feedstocks, such as oil, in the conversion of coal to liquids has brought very important cost implications to liquefaction processes. The novel concept we propose below takes the coprocessing approach still further.

In recent years, the processing of coal with feedstocks other than oil has been explored with varying degrees of success; one example includes lignin. Coal/oil coprocessing is a concept with important economic implications that we will discuss; we propose a more general category for this kind of liquefaction process that will include these and other important variations, some of which may address problems in the current approaches to coal/oil coprocessing. We have given the name "generalized coprocessing" to this more inclusive type of technology. Thus, in this Note generalized coprocessing is considered to be any technology that processes two or more feedstocks for the production of liquid hydrocarbon fuels from coal.

**Coal/Oil Coprocessing**

Coal/oil coprocessing can be considered as a reorientation of an already commercialized process in which small amounts of coal are added to heavy oils to produce lighter oils. Coal added in amounts of 5 percent by weight to heavy oil has been shown in the CANMET Hydrocracking Process to give benefits such as demetallization and the reduction of coking. In this case, the oil primarily benefits from the addition of coal in the processing of oil. The concept of coprocessing is thought to reverse this with the oil conferring benefits to the coal liquefaction process. In coal/oil coprocessing, coal and oil are blended up front to form one process stream from which coal liquids and oil are produced. Coal-to-oil ratios can be as high as 2:3, although a ratio of 1:2 is more common. Above these limits, viscosity problems, among other factors, become too severe.\(^\text{14}\) Table 5 lists ratios of coal to oil in various processes.

---

\(^{14}\) Coprocessing research in the United States has been conducted at the following institutions: Auburn University, Chevron Research Company, Exxon Research and Engineering Company, Hydrocarbon Research, Inc., Lummus Crest, Inc., Mobil Oil Corporation, Universal Oil Products, Inc., and others. In Canada, research has been conducted by CANMET.
Table 5
COAL/OIL AMOUNTS IN VARIOUS TECHNOLOGIES
FOR THE PRODUCTION OF LIQUID FUELS

<table>
<thead>
<tr>
<th>Type of Process</th>
<th>Stage of Research</th>
<th>Coal (%)</th>
<th>Oil (%)</th>
<th>Lignin (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Addition of coal to heavy</td>
<td>Commercialized</td>
<td>5</td>
<td>95</td>
<td>--</td>
</tr>
<tr>
<td>oil for demetallization, reduced coking</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Coal/oil coprocessing</td>
<td>Ongoing</td>
<td>40-50</td>
<td>50-60</td>
<td>--</td>
</tr>
<tr>
<td>Coal/oil/lignin processing</td>
<td>Proposed novel</td>
<td>TBD&lt;sup&gt;a&lt;/sup&gt;</td>
<td>TBD&lt;sup&gt;a&lt;/sup&gt;</td>
<td>TBD&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>concept</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<sup>a</sup>To be determined

**Advantages to Coal/Oil coprocessing.** The petroleum feedstock fraction in coal/oil coprocessing acts as a solvent for the coal and contributes to the final product volume. Coprocessing has several economically interesting benefits over conventional liquefaction technologies; for example it claims to:

- Use existing refinery capacity and infrastructure;
- Eliminate solvent recycle;
- Have a less-complex technology relative to conventional approaches;
- Require lower capital investment;
- Be able to be commercialized in a minimum amount of time; and
- Require less hydrogen for production of liquid product.

Coal/oil coprocessing offers significant advantages over most other direct coal liquefaction processes in that although operating conditions are similar to conventional technologies, much less capital investment is required up front. The approach affords a link from the present petroleum-based technology to a synfuels/coal-based technology by using
as much existing infrastructure and technology as possible, in particular those refineries equipped to handle heavy oil and bitumen. Use of existing infrastructure alone greatly affects product cost because it addresses the major cost category—capital cost or plant investment.

Another major advantage claimed by coal/oil coprocessing is the ability to process its feedstocks in a single pass, thus eliminating the recycling of solvent required by most conventional direct liquefaction processes. Solvent must be recycled in those cases because the process is entirely self-supporting, i.e., solvent required for dissolving coal comes from the liquid product, which is recycled back to the front of the process. Solvent recycle is expensive because it lowers throughput or process yield, adds to infrastructural requirements, and lowers efficiencies by requiring that the solvent be rehydrogenated, reheated, and repumped. Heavy oil and bitumen provide lower-cost solvents and because they are liquid at the liquefaction temperatures, they can dissolve the coal, which removes the need to recycle liquid product. Because they are also more hydrogen-rich to begin with, hydrogen requirements for the process are theoretically lower.

Unresolved Problems in Coal/Oil Coprocessing. Despite its many advantages, several drawbacks to coal/oil coprocessing have either not been entirely resolved or have been newly introduced to the liquefaction process. Some of these include severe operating conditions and variability in product yield because of feedstock mismatch, or oil incompatibility with the coal.

The operating parameters established by Auburn University research indicates that optimal yields with coal/oil coprocessing can be obtained when liquefaction occurs at 425 degrees C under hydrogen pressures of 1250 psig with a long residence time (60+ minutes) and a hydrogenation catalyst of small particle size.\(^\text{15}\) These conditions are similar to processes such as the Exxon Donor Solvent (EDS) process and H-Coal process, and have similar attendant problems, such as costly equipment requirements, erosion-corrosion, etc.

\(^{15}\)Curtis, Tsai, and Guin (n.d.).
The extent of liquefaction with coal/oil coprocessing and the yields of distillable oils vary, depending on the type of oil and process conditions. Past work by Moschopedis et al. showed that the optimal blend of operating parameters depends on the type of petroleum-based material used.\textsuperscript{16} Coal conversions from 51 weight percent to 87 weight percent have been observed depending on the severity of the coprocessing conditions.\textsuperscript{17}

There is a larger heavier-fraction yield (pitch) for coal/oil coprocessing than for conventional direct liquefaction technologies, which could imply greater costs downstream to upgrade to lighter fractions.\textsuperscript{18} The introduction of a recycle solvent stream in coprocessing technologies has been considered. Recycling solvent helps to boost yields by supplying a molecularly more compatible solvent for the coal than oil can provide and simultaneously upgrades coal liquids by exposing them longer to hydrogenation. Paraflinic petroleum fractions are less compatible with an aromatic coal than are aromatic coal-derived solvents. Some coprocessing technologies require two liquefaction steps rather than one. If recycling or two-stage liquefaction is being investigated for coprocessing technologies, then other variations to coprocessing that should be less expensive deserve attention.

A solvent that is compatible with coal and yet has the advantages of residual oil, such as acting as a once-through solvent, has an excellent chance of enhancing coal liquefaction economics. Lignin, a naturally occurring chemical compound, provides an interesting possibility for such a solvent, and at the very least may open the door to a new family of coal-dissolving solvents.

\textbf{Lignin/Coal Liquefaction}

Lignin is a naturally occurring polymer found in wood and woody plants in amounts up to 25-30 percent. The pulp and paper industry is

\begin{flushleft}\textsuperscript{16} Moschopedis, Hawkins, and Speight (1982), pp. 213-228. \\
\textsuperscript{17} Kelly et al. (1984). \\
\textsuperscript{18} Kelly et al. (1984).\end{flushleft}
the principal producer of lignin. In the manufacturing process of isolating cellulose fibers from lignin for the production of paper products (lignin is thought to discolor paper), it is separated in the form of black liquor. Among many of the diverse commercial applications of lignin-derived compounds (such as the source of vanillin, an artificial food flavoring), its most common application is as an additive for oil drilling mud. Lignin supplies are collocated with pulp and papermaking mills, e.g., in the Northwest, and are typically not adjacent to coal fields.

There is probably no single structure that fully represents lignin; however, it is known to be aromatic. Unlike some coal solvents that are not derived from coal, lignin is chemically compatible with coal and, in fact, has been postulated as the geological precursor to coal.19 (Our proposed novel concept of lignin/coal/oil coprocessing takes advantage of this characteristic to address some of the problems encountered in coal/oil coprocessing.)

The coprocessing or coliquefaction of lignin and coal to liquid products has been observed20 and explored.21 Researchers at the University of Connecticut were able to conduct, albeit on a very small scale, the liquefaction of lignin and bituminous coal at more moderate operating conditions than are typical of liquefaction processes, achieving liquid product at temperatures of 300 degrees C and pressures as low as 1500 psi. Coal conversion yields up to 66 percent were achieved.22 Hydrogen was supplied as gaseous hydrogen in addition to a hydroaromatic solvent or hydrogen donor solvent. Although most of the experiments used an acid catalyst, several runs offered evidence that a catalyst may not be necessary. Resid processing with lignin has also been investigated and patented.23

19 Hayatsu et al. (1979).
21 The Mobil Oil Corporation holds U.S. patent number 4,409,089 describing a coal/lignin liquefaction process.
22 Coughlin and Davoudzadeh (1983).
23 Mobil Oil Corporation patent 4,409,089.
Effects of Lignin

Lignin is thought to induce liquefaction at lower temperatures because of the very reactive compounds formed by its own liquefaction. Some additional coal liquefaction might also be attributed to dissolution in the better solvent provided by lignin liquefaction products formed in the reaction. Lignin can be liquefied to produce phenolic-like substances up to as much as 38-42 percent of total liquid yield. Researchers hypothesize that thermal degradation of lignin produces substituted phenoxy radicals at temperatures too low (300 degrees C) for reactive coal molecules to form substantially from temperature effects alone. The lignin-derived radicals are thought to cause depolymerization of coal molecules by attacking weaker carbon bonds holding coal fragments together. Other studies also indicate that when coal is liquefied in the presence of phenol, extensive depolymerization occurs. On the other hand, when coal liquefaction is conducted in nonphenolic solvents, such as toluene, xylene, or tetralin, repolymerization of the coal radical fragments can readily occur and can lead to the formation of a solid, relatively unreactive mass.

No coal liquefaction was observed when previously hydrocracked lignin was used. This finding suggests that lignin be considered, like oil, as a once-through solvent or that research be conducted on ways to reactiv ate liquid solvent formed from the liquefaction of lignin. The latter route would be considerably more desirable from the point of view of feedstock requirement although this would require research. From laboratory tests, yields monotonically increased with the ratio of lignin to coal to a certain threshold. In fact, the best results reported occurred with lignin to coal ratios of 1:1, which is quite high and probably economically infeasible, considering lignin's availability. If not separated from the product stream lignin would probably contribute to the final product but is not a true catalyst in that it is consumed by the reaction and cannot be reactivated. It does, however, have transitory catalytic qualities in the liquefaction setting.
LIGNIN/COAL/OIL COPROCESSING NOVEL CONCEPT

The proposed novel concept, lignin/coal/oil coprocessing, addresses the problems of coal/solvent incompatibility and the harsh operating conditions seen in coal/oil coprocessing.\(^2\)\(^4\) This concept makes use of two direct coal liquefaction technologies, coal/oil coprocessing and coal/lignin coliquefaction or depolymerization, to form a hybrid technology that we call lignin/coal/oil coprocessing. In the proposed case, lignin, molecularly better suited as a solvent for coal than oil, would assume a role as a solvent adjunct. Although more aromatic than oil, one of its chemical disadvantages is its high oxygen content, which, if not removed upstream, will cause a high consumption of expensive hydrogen downstream. Thus it should be used in moderate amounts.

The lignin coal/oil hybrid integrates its two constituent technologies so that their respective advantages complement each other or address the deficiencies of each separately. The potential benefits of this technology are that it:

- May provide a superior coal/solvent system;
- Moderates operating temperatures;
- May decrease sensitivity of coal and oil combinations;
- May increase product yield; and
- Reduces catalyst requirement.

Determining the optimal placement of lignin (or lignin-like solvent) in the liquefaction process requires laboratory research; however, this study suggests at least three possibilities for its addition. Lignin could be (a) mixed initially together with coal and oil, (b) added to the spent reaction mixture to increase product yield, or (c) processed first with coal, with oil added to the unreacted coal

\(^{2\text{A}}\) A concept not described here addresses the hydrogen cost issue by the elimination of purified hydrogen. (See App. B.)
after the liquid product is removed. These options are illustrated in Fig. 2. Note that the first option indicates lowest capital costs (less complexity, probably less capital requirements) but highest volume requirements of lignin.

The first and third options suggest a greater requirement of lignin and have the disadvantage of introducing a significant amount of oxygen to the mixture. A higher introduction of oxygen to the product stream, and an associated higher hydrogen cost of its removal, or requirement of an oxygen removal step is a consequence of using greater amounts of lignin. Thus, there will be a tradeoff that must be further explored.

**Fig. 2 -- Proposed lignin/coal/oil liquefaction processes**
with analyses augmented by laboratory findings to better understand lignin's benefit compared with its costs. Lignin/coal liquefaction, like coal/oil coprocessing, works best with the lower rank coals, such as subbituminous and lignite. However, one of the concerns with using lignin in large amounts is that its high oxygen-to-carbon ratio (0.3-0.5),\textsuperscript{25} slightly more than lignite, would cause high consumption of hydrogen during hydrotreating steps further downstream. For every oxygen atom removed from solution by hydrotreatment, two hydrogen atoms are consumed in the formation of water. Additionally, higher oxygen levels in the final product tend to lower the energy content of the resulting fuel. More research in this area is suggested.

Potential Cost Savings Associated with Novel Concept

We believe that lignin/coal/oil coprocessing could reduce capital costs even more than coal/oil coprocessing. For instance, more moderate operating conditions can lead to less costly equipment, all other things being equal. The novel concept promises to reduce costs of coal/oil coprocessing even further because it requires less severe operating conditions. In addition, if the new process is integrated into an existing plant, substantial savings can accrue through lower capital costs—a primary contributor to product cost for liquefaction processes using coal alone as a feedstock. If the addition of lignin reduces the sensitivity of coal/oil combinations, this may introduce a flexibility factor in the major feedstock materials, which may have cost implications.

The addition of lignin to the processing of coal and oil, however, is not without some cost penalty. Any additional feedstock to a process stream adds complexity and requires more handling of materials, such as storage, piping, and blending. The distance between the lignin supply source and the processing plant is also a consideration that may become important. Chemically, lignin holds promise as a solvent adjunct. However, feedstock collocation and the decoupling of lignin from papermaking processes are important issues for the economic feasibility

\textsuperscript{25}Sternhell (1964), pp. 1236-1244.
of this novel concept. Much depends on how much lignin is required for the process.

Availability of Lignin

The availability of lignin must be explored and is an important consideration in the viability of this process. The types of coal to which this novel concept would apply are probably bituminous and subbituminous, which constitute the major portion of our coal resources.

This process would require fewer infrastructural changes than would conventional liquefaction facilities, but somewhat more than coprocessing facilities. Specific requirements would include:

- Linking the process with a lignin source;
- Setting up distribution links for the transportation, storage, and handling of lignin and coal at the refinery; and
- Making modifications to refining plants.

Lignin, coal, and oil are not collocated, therefore transportation costs should be an important consideration. Collocation of coal and oil is a current concern; the collocation of the three feedstocks only compounds the problem and therefore introduces with greater certainty the costs of hauling at least one of the feedstocks. If relatively small ratios of lignin to coal and oil are required, these hauling costs will be less important. If the requirement of lignin to a unit of coal is large, lignin's attractiveness as an adjunct solvent will be severely curtailed.

Environmentally, lignin should not contribute to certain problems that coal poses; for instance, it contains essentially no sulfur or nitrogen. (Associated cost savings due to their absence is associated with the fact that they are often removed by reacting with hydrogen, forming hydrogen sulfide and ammonia.) Waste concerns should be minimal with the introduction of lignin. Even if lignin were diverted from the product stream, it need not create a disposal problem because it could be burned in the liquefaction plant for additional fuel.
Earlier in this section, we explored the key economic chokepoints of liquefaction in general and coprocessing in particular. A severe constraint to the proposed novel process was an inadequate supply base for lignin, which, if put into the coprocessing context, may limit the amount of lignin available per unit of coal. We investigated the implications of theoretical amounts of lignin required to produce liquid fuels from the coprocessing of lignin, coal, and oil (see App. C). Many other critical issues hinge on this single parameter that ultimately can be resolved only in the laboratory. Issues influenced by the required amount of lignin include lignin supply requirements, sensitivity of process economics to lignin supply costs, and hydrogen and catalyst requirements, among others.

We determined that it was important to test the reasonableness of our assumption that lignin could be used in lignin/coal/oil coprocessing. Because the important data were missing (necessary ratios of lignin to coal and oil), we made several calculations to investigate whether the current supply of lignin is an important constraint to the further development of this process. It is essential to emphasize that these calculations were based on current supplies of lignin. We concluded that even if coprocessing could be integrated with current pulp and paper operations with the complete diversion of lignin consumption to the liquefaction industry, with the existing levels of lignin production, it would be difficult to depend on this novel process to supply large amounts of our transportation fuels. This suggests that if laboratory results are encouraging, the supply problem would need to be further investigated.

To investigate lignin supply requirements, in the absence of data, we set up a hypothetical scenario. As a best case, we proposed that the entire production of lignin be diverted to the production of liquid fuels from coal. Furthermore, we proposed that an arbitrary amount of the transportation fuels required in this country be produced from a lignin/coal/oil coprocessing technology. We assumed that this process would be an important contributor to liquid fuel production. To reflect that importance, we chose to require that 25 percent of this nation's transportation fuels be produced with the lignin/coal/oil coprocessing technology.
In addition to the above assumptions, the contribution that lignin made to the final product volume was ignored. Calculations were then made that describe total lignin supply requirements, based on various lignin-to-coal weight percentages. The results of our calculations, described in Appendix C, show that to produce this amount of fuel from the existing supplies of lignin of a major supplier, the maximum lignin-to-coal ratio currently possible was 1:19 (5 percent by weight, moisture-ash-free coal), a rather small amount.

One assumption in our analysis was to divert all lignin produced to lignin/coal/oil coprocessing. For that to occur, changes to the pulp and papermaking processes would be necessary. The primary producer of lignin, the pulp and paper industry, extracts lignin as a by-product of the kraft and soda processes for the production of pulp and paper. Lignin is separated from the cellulose in the form of black liquor, which contains all the inorganic chemicals used for pulping. To recover the inorganic materials used in pulping, this black liquor is burned in a recovery furnace that generates steam to run the mill (lignin's heating value is about 11,000-12,000 Btu/lb).\(^{26}\) The economics of the pulping process depend on being able to recover those inorganic chemicals. The kraft and soda pulp mills consume about three million tons of lignin annually in the recovery of chemicals and heat.\(^{27}\)

Relatively small amounts of lignin are purified for commercial applications and used outside the papermaking industry.

For lignin/coal/oil coprocessing to contribute significantly to this nation's transportation fuel needs, as one option, the supply of lignin would not only have to be decoupled from the pulp and papermaking industry but possibly also increased. To use lignin from the pulping process to produce coal liquids, either papermaking processes would need to be altered or liquefaction plants collocated with pulping mills to address the problem of recovering inorganics from the process and energy from lignin. If a liquefaction plant were collocated with a pulping plant, the inorganics extracted from lignin residues could be recycled.

\(^{26}\)Vincent (1980), pp. 299-300.
\(^{27}\)Pearl (1967), p. 308.
back to the pulping plant. Likewise, unreacted coal from the liquefaction plant might be able to substitute as the lignin fuel currently used in the pulping and papermaking operations. With a different allocation of resources, those components more easily converted to liquids would be diverted to liquid hydrocarbon production and those unreactive components, such as spent coal, could be used as energy. Feedstock transportation costs, among others, make this situation unlikely.

The supply of lignin can arguably be expanded. Lignin is derived from a renewable resource and can be isolated from materials other than wood, such as cotton hulls, grasses, etc. Few processes discard separated lignin from these sources, however, and may not be a reliable source of lignin.

Despite these problems, lignin offers new opportunities for direct coal liquefaction. Further investigation of this process could shed light on the nature of other desirable solvent adjuncts for coal/oil coprocessing approaches. Lignin may represent a new class of solvent additives for coprocessing technologies and therefore should still be considered worthy of future chemical study.

RESEARCH REQUIREMENTS

Technical uncertainties surround this technology. To our knowledge, it has not gone beyond the conceptualization stage. Because very little research has been conducted on the lignin/coal/oil coprocessing approach, much needs to be done. Basic research would be required as a first step. However, some suggested research activities worthy of particular attention include:

- Establishing chemical/technical feasibility. Lab-scale tests would be appropriate.
- Establishing an optimal amount of lignin. One would like to use as little as possible for as much catalytic reactivity as possible. These findings will indicate the total resource requirement for lignin.
• Determining optimal temperatures and pressures. There are indications that both will be lowered from conventional processes.
• Exploring the various possible process schemes described above with particular emphasis on using lignin with less reactive fractions.
• Better understanding the role of lignin. Investigate (a) whether reactivity can be restored so that lignin could act as a type of catalyst or (b) if other types of materials could mimic lignin's action.
• Searching for more available or less expensive lignin-like solvents.
• Assessing the increased complexity resulting from the introduction of lignin.

From the above, it is reasonable to assume that this process would not be available for commercial applications within the next ten years because of the uncertainties that still exist. Assuming that the process works, technically it has a good chance of reducing costs and thus of being "successful." We suggest that the economic success of the lignin/coal/oil coprocessing concept be further explored in conjunction with the determination of key laboratory data, as described in the above list.
III. THE ABOVEGROUND RETORTING OF OIL SHALE USING MICROWAVE ENERGY

SHALE RESOURCE ESTIMATES AND PHYSICAL CHARACTERISTICS

Oil shale retorting is the process of heating crushed oil shale to produce a liquid hydrocarbon fuel called shale oil. In this section, a novel technology for aboveground retorting of oil shale that makes use of microwave electromagnetic energy is described and assessed.

This section begins with a brief survey of oil shale resources and a description of the physical characteristics of oil shale. After reviewing some of the technical considerations involved in processing oil shale, the process of retorting oil shale with high power microwave energy is described, and inherent tradeoffs are considered. The section includes a look at the economic impact of this process and at its impact on the environment and existing infrastructures. Finally, we make recommendations concerning future research necessary to resolve key issues for this process.

Resource Estimates

Oil shale deposits have been found on all of the inhabited continents. The extent of the worldwide resources cannot be accurately determined, but it appears to be very large indeed (Table 6).¹ In 1965, the U.S. Geological Survey (USGS) estimated that the world's oil shale deposits constituted over 4 quadrillion tons, which has a total shale oil yield of over 2 quadrillion barrels.

The oil shale deposits in the United States are located mainly in the central and eastern states, Colorado, Utah, and Wyoming (the Green River formation), and Alaska, as shown in Fig. 3. The deposits in the Green River formation in Colorado, Utah, and Wyoming (Fig. 4) are particularly noteworthy because they contain the largest concentration of potential shale oil in the world. Because deposits in the central and eastern United States underlie a larger area, they appear more

Table 6

ESTIMATES OF IN-PLACE OIL SHALE RESOURCES

<table>
<thead>
<tr>
<th>Location</th>
<th>Area Underlain by Organic Shales (Square Miles)</th>
<th>Oil-Equivalent Deposits in Trillions of Barrels</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>25-100 GPTa</td>
</tr>
<tr>
<td>Africa</td>
<td>5.0 million</td>
<td>4</td>
</tr>
<tr>
<td>Asia</td>
<td>7.0 million</td>
<td>5.5</td>
</tr>
<tr>
<td>Australia</td>
<td>1.2 million</td>
<td>1</td>
</tr>
<tr>
<td>Europe</td>
<td>1.6 million</td>
<td>1.5</td>
</tr>
<tr>
<td>South America</td>
<td>2.4 million</td>
<td>2</td>
</tr>
<tr>
<td>North America</td>
<td>3.0 million</td>
<td>3</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>17</td>
</tr>
</tbody>
</table>

|                | 1.6 million                                     | 2             | 28         | 168       |
| United States  |                                                 |               |            |           |
| Green River    | 16 thousand                                     | 1.2           | 4          | 8         |
| formation      |                                                 |               |            |           |

\(^a\) Gallons per ton.

Fig. 3 -- U.S. oil shale deposits
impressive on maps than do those of the Green River. However, they contain less than half the oil shale in the Green River formation and do not yield as much oil on a unit basis because of a lower proportion of hydrogen to carbon in their organic component.\(^2\)

The Green River formation is a geologic entity underlying about 34,000 square miles of terrain in northwestern Colorado, southwestern Wyoming, and northeastern Utah. Oil shale resources have been found under about 17,000 square miles of the basins of the Green River formation. The principal deposits are found in the Piceance, Uinta, Green River, and Washakie basins as shown in Fig. 4. The total resource within this area is estimated at about 8 trillion barrels.

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\(^2\)Without the introduction of exogenous hydrogen, shales with lower hydrogen-to-carbon ratios will produce smaller proportions of oil relative to the kerogen present than shales with higher hydrogen-to-carbon ratios.
Physical Characteristics

When considering the production of oil from oil shale, it is helpful to understand the resource's characteristics. Oil shale is a sedimentary rock containing organic matter that was formed by the simultaneous deposition of mineral silt and organic debris on lakebeds and sea bottoms. As the raw materials accumulated, heat and pressure transformed them into a stable mixture of inorganic minerals and solidified organic sludge. The formation processes that yielded petroleum, tar sands, and coal were conceptually similar but differed with respect to certain physical and chemical conditions. In oil shale, these conditions resulted in the formation of chemical bonds between individual organic molecules. The large size of the molecules formed by this bonding prevents them from dissolving in normal solvents. However, when heated in processes known as pyrolysis or destructive distillation, the bonds rupture, forming smaller liquid or gaseous molecules. The smaller molecules can then be separated from the inorganic matrix, which remains behind as spent shale waste product.

About 10 percent of the organic component in oil shale is a solid substance called bitumen, which can be dissolved in certain solvents. However, most of the organic component is a solid material called kerogen, which is insoluble in most standard petroleum solvents. Kerogen is composed of carbon and hydrogen molecules crosslinked together by sulfur and oxygen atoms to form relatively large three-dimensional macromolecules. These macromolecules are embedded within the finer grained inorganic or mineral matrix of the oil shale. This organic continuous phase gives kerogen-rich oil shale most of its physical strength and stability. When the organic matrix is removed from very rich oil shale, the mineral residue has little cohesive strength and is easily crushed to a fine powder. The mineral and organic constituents of oil shale are listed in Table 7.³

When kerogen is heated above 400 degrees F, chemical bonds between and within the individual organic molecules begin to rupture, forming smaller molecules. Three combustible products are formed from the

Table 7
COMPOSITION AND PYROLYSIS PRODUCTS OF TYPICAL COLORADO OIL SHALE

<table>
<thead>
<tr>
<th>Mineral Constituents</th>
<th>Weight Percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dolomite</td>
<td>32</td>
</tr>
<tr>
<td>Calcite</td>
<td>16</td>
</tr>
<tr>
<td>Quartz</td>
<td>15</td>
</tr>
<tr>
<td>Illite</td>
<td>19</td>
</tr>
<tr>
<td>Low-albite</td>
<td>10</td>
</tr>
<tr>
<td>Adularia</td>
<td>6</td>
</tr>
<tr>
<td>Pyrite</td>
<td>1</td>
</tr>
<tr>
<td>Acalcime</td>
<td>1</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>100</strong></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Element</th>
<th>Weight Percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon</td>
<td>76.5</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>10.3</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>2.5</td>
</tr>
<tr>
<td>Sulphur</td>
<td>1.2</td>
</tr>
<tr>
<td>Oxygen</td>
<td>9.5</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>100.0</strong></td>
</tr>
</tbody>
</table>

Yields from Fischer Assay Pyrolysis\(^a\)

<table>
<thead>
<tr>
<th>Decomposition Product</th>
<th>Weight Percent of Organic Constituent in Raw Shale</th>
<th>Weight Percent of Total Raw Shale</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oil</td>
<td>63</td>
<td>10.4</td>
</tr>
<tr>
<td>Noncondensible gas...</td>
<td>15</td>
<td>2.5</td>
</tr>
<tr>
<td>Fixed-carbon residue.</td>
<td>13</td>
<td>2.2</td>
</tr>
<tr>
<td>Water vapor</td>
<td>9</td>
<td>1.4</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>100</strong></td>
<td><strong>16.5</strong></td>
</tr>
</tbody>
</table>

\(^a\)26.7 gal/ton shale pyrolyzed by Fischer Assay procedure at 500 degree C.
pyrolysis of kerogen: vaporized oil, which can be condensed by cooling; a gaseous mixture containing hydrogen, oxides of carbon, hydrogen sulfide, and hydrocarbon gases such as methane; and a coke-like solid residue, which remains behind. The relative proportions of evolved oil, gases, and coke largely depend on factors such as the pyrolysis temperature, atmospheric conditions in the retort, and, to a lesser extent, the organic content of the raw shale.

Each of the three main products of kerogen decomposition is a potential source of energy. Crude shale oil can be burned directly as a fuel or it can be refined to produce fuels similar to those obtained from conventional petroleum crude oils. The undiluted off-gas from kerogen pyrolysis has a heating value similar to natural gas and could be burned as plant fuel in the oil shale facility or pipelined to other areas for commercial or industrial applications. The coke residue is also a potential source of energy but a poor fuel in comparison with coal or the raw shale itself. The heating value of the coke is about 2 percent that of coal.

The effect of heating the mineral material in shale is to form salts. Before returning the spent shale to the environment and for handling purposes, it must be cooled, usually with water. The salts then dissolve in the cooling water and must be removed before disposal of the water. It is this salt formation and subsequent leaching into the groundwater that is the primary environmental problem in shale pyrolysis. Spent shale will be produced in enormous quantities by an oil shale industry of any significant size. Some potential uses exist for the spent shale generated as a by-product of aboveground pyrolysis, such as conversion to cement or building materials, but the output from a single commercial scale facility will far exceed the market demand for such material. Nearly all of the spent shale will have to be disposed of as waste.

Embedded within some shale deposits are materials of commercial value other than kerogen. These include nahcolite (sodium bicarbonate or commercial baking soda), which may be processed to yield soda ash, a common raw material for glass production, and a potential means of removing sulfur compounds from power plant exhaust gases; dawsonite
(dihydroxy sodium aluminum carbonate), a potential source of alumina, which can be converted to aluminum; and trona (hydrated mixture of sodium carbonate and sodium bicarbonate), a source of soda ash for glass production. In the Piceance basin alone it is estimated that there are reserves of 300 billion barrels of shale oil, 30 billion tons of nahcolite, and 20 billion tons of dawsonite, which could produce 7 billion tons of soda ash and 7 billion tons of alumina.⁴

TECHNICAL CONSIDERATIONS FOR PROCESSING OIL SHALE

Converting raw oil shale in the ground to finished fuels and other products for consumer markets involves a series of processing steps centered on an overall processing approach. Two generic approaches exist to process the resource: in-situ processes and aboveground processes. Discussions of in-situ processes often distinguish between "true" in-situ processes, which involve only the drilling of wells, and "modified" in-situ processes, which require some mining to develop underground retort chambers. Distinctions of this type are unimportant here.

Within the two generic processing approaches, three methods of heat transport can be used to break down the highly crosslinked, high molecular weight organic material present in the shale: conduction, convection, and radiation. The first two methods typically involve a combustion source and are generally termed conventional heating; the third typically involves the absorption of electromagnetic radiation. Mineral matter in shale conducts heat poorly; as expected for poor conductors, it has a low thermal conductivity and low thermal diffusivity. As kerogen, which is a better heat conductor than the surrounding mineral matter, is vaporized the thermal conductivity of the remaining shale decreases.⁵ Thus when shale is at pyrolysis temperatures and as kerogen leaves the shale particle as oil vapor, to raise the temperature of the remaining shale an incremental amount will require higher levels of energy intensities.

⁵DuBow (1977).
General Retorting Methods

The steps involved in true in-situ (TIS) retorting include (1) fracturing the deposit to improve fluid flow, (2) providing heat for pyrolysis, and (3) recovering the oil and gases through wells. The main advantage in this process is the elimination of aboveground disposal requirements of waste material. One important drawback to this method is that efficient in-situ processing requires a highly permeable deposit for flow of oil. For this reason the Green River formation shales are virtually eliminated as candidates for TIS processing. A second drawback to this method is the associated heat losses, making this a highly energy intensive process.

In modified in-situ (MIS) retorting the permeability of the shale deposit is increased by mining some shale from the deposit and then blasting the remainder into the void thus created. Heat is then supplied to the shale bed and oil and gas are recovered. Although the mining to create voids in the shale bed enhances the recovery of shale oil in this process, it also increases the materials handling problems relative to TIS processes. Heat loss also remains a problem in this process. Yields from in-situ processes are typically lower than aboveground retorting processes.

Literally hundreds of aboveground retorts have been invented in the 600-year history of oil shale development. All surface retorts are similar in that the retorting process actually occurs in a metal vessel in an aboveground facility. Such a configuration has the advantage that process flows and operating parameters can be easily controlled and manipulated to obtain a maximum yield of oil. However, the surface processes also suffer the disadvantage that large quantities of raw oil shale must be mined, transported to the retort area, processed, and disposed of in an environmentally acceptable manner.

Methods of Heat Transport in Oil Shale

Heat can be provided to the shale bed by any of the three heat transport methods noted above. Both conventional and electromagnetic heat sources have been supplied to shale formations, resulting in pyrolysis products.
Aboveground retorts that use conduction (and some convection) heat transport mechanisms are generally called directly heated processes. In these processes, the shale is heated by the direct combustion of a portion of the organic material.\(^6\) Retorts that use convection (and some conduction) mechanisms are called indirectly heated processes. In these processes, the shale is heated by the transfer of heat from solid or gaseous heat carrying media.\(^7\) A further distinction can be made between processes that rely upon gaseous and solid heating media, but no such distinction is needed here.

Various strategies have been developed in the past for using electromagnetic energy to heat shales and oil sands. Among the most notable are filament heating, ohmic heating, microwave heating, and radio frequency heating. The earliest known use of electromagnetic energy to heat shale involved the use of filaments.\(^8\) Heating by filaments relies upon thermal diffusion (conduction) from a filament placed in the ground. Although successful in producing some oil, the method is quite inefficient, since thermal diffusion is a poor means of energy transport in these materials—some regions of the shale are overheated and others are underheated. This causes a large amount of coking and cracking of products before the oil reaches the recovery well, which can decrease oil yield, in addition to thermal energy losses in regions that are underheated.

Ohmic heating, which relies upon radiative heat transfer, uses an anode and cathode inserted in the shale bed with a current passed between the two. The method works quite well at low temperatures (<200 degrees C), but at higher temperatures steam is produced, which promotes

\(^6\)Three commercial retorts, Gas Combustion (U.S. Bureau of Mines), Paraho Kiln (Development Engineering, Inc.), and Union Retort "A" (Union Oil Company of California), make use of this heating mechanism.

\(^7\)Six commercial retorts make use of this mechanism: Union Retort "B" (Union Oil Company of California), Paraho Kiln-alternate (Development Engineering, Inc.), PetroSix (Brazil), and Union Steam Gas Recirculation System (Union Oil) use heating by gaseous media; Lurgi-Ruhrgas (Lurgi and Ruhrgas, A.G.) and TOSCO II (Oil Shale Corporation) use heating by solid media.

arcing over a narrow path. When only a narrow path is heated the effect is similar to filament heating. Another problem with this technique is that a large area must be mined to implant the anode and cathode.

Processes that are radiation intensive generally involve the absorption of electromagnetic radiation in the radio frequency or microwave region of the spectrum. Radiative heating typically relies upon the conversion of electromagnetic energy into thermal energy by the material. Electromagnetic radiation is applied to a material and a portion of that energy is absorbed by the material through dielectric or magnetic mechanisms. The absorbed energy is then transformed directly into heat energy. A familiar version of this process occurs in the domestic microwave oven. Bench scale testing of the use of electromagnetic radiative heating of shale has been conducted by Lawrence Livermore Laboratory\(^9\) and the Illinois Institute of Technology Research Institute (IITRI).\(^10\)

The use of oscillating electric fields in the microwave or radio frequency band to heat shale has an advantage over ohmic heating in that it does not rely upon the thermal diffusivity of the shale to transport energy and is not limited by the arcing problem of ohmic heating. Experiments conducted using microwave energy at 2.45 GHz indicate that the highest yields are obtainable when heating rates are high and when sweep gases are present.\(^11\)

The amount of heat energy that can be transferred to a medium by electromagnetic radiation at a given frequency depends upon physical properties of the material such as the dielectric constant. The dielectric constant is one measure of the behavior of a material in an electric field. For mathematical simplicity, the dielectric constant is taken to be composed of two parts—a real dielectric part and a conductivity or imaginary part. Metals have a conductivity that is large compared with the real dielectric constant; the situation is reversed in dielectric materials. The usual physical picture of

---

\(^9\)Mallon (1980).
\(^10\)Bridges, Taflove, and Snow (1978).
absorption of radiation by dielectric materials has the bound atomic charges in the material behaving as mass-spring systems that are excited by the impinging radiation and that have certain natural frequencies for which absorption is a maximum. At radiation frequencies other than these natural frequencies, the radiation passes through or is reflected by the material. A graph of the amount of radiation that is absorbed by a material at a given frequency is called an absorption spectrum.

Water, which is contained in the raw oil shale, has a very rich absorption spectrum. General bands of absorption resonances exist in water in the radio frequency and microwave regions. However, light in the visible spectrum mainly passes through water. The microwave frequency range of 0.3 to 300 GHz encompasses the vibrational and rotational frequency range of typical organic bonds, and it is the excitation of these resonances and the resonances in water that give rise to the dielectric heating of oil shale. For water in shale, absorption resonances have been found at 915 MHz, 2375 MHz, and 2430 MHz, which are all in the microwave region of the electromagnetic spectrum. A typical absorption spectrum for oil shale in the frequency range 2-4 GHz is shown in Fig. 5.

The Federal Communications Commission (FCC) has assigned four microwave bands for industrial heating in which there is no limit to the permitted radiation. These bands are centered around 915 MHz, 2450 MHz, 5800 MHz, and 22,000 MHz. Industrial power sources are available at only the first two frequencies.

**Economic and Environmental Chokepoints**

The chokepoints in the general development of a commercial scale oil shale industry are economic and environmental. Materials handling costs and heat energy costs for these processes make them economically unattractive. In conventional aboveground retorting processes, mining, hauling, and crushing of raw oil shale and disposal of spent shale account for about two-thirds of the total cost of producing refinery feedstock from an oil shale deposit; retorting accounts for the rest.

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12Bauman (1962).
Fig. 5 -- Typical absorption spectra of Green River shale over the frequency range 2-4 GHz

In conventional retorting processes, the slow mechanisms of heat transport (conduction and convection) transfer heat unevenly and generate much waste heat. An enormous amount of energy is also lost to the mineral substrate. In addition, waste heat is largely responsible for the main environmental problem in shale pyrolysis--leachable salts. The heating of the mineral portion of the shale causes the formation of salts, which then dissolve in water that is used to cool the spent shale. To prevent these salts from leaching into the local groundwater, the water must be cleaned before it is released to the environment.

We have defined an energy efficient process as one that produces a maximum amount of shale oil for a given amount of energy input; an efficient materials handling process is one that produces a maximum amount of shale oil for a given amount of material that must be mined, hauled, crushed, and disposed of. From these definitions, as noted in the previous subsection, aboveground processes are relatively energy
efficient but are quite inefficient in terms of materials handling. In contrast, in-situ processes have higher materials handling efficiencies but are relatively energy inefficient. Higher costs are associated with inefficiencies. One approach to producing shale oil more economically would be to investigate processes that optimally balance energy and materials handling efficiencies. This situation is illustrated in Fig. 6. As shown, processes such as in-situ processes have relatively low materials handling problems but unfortunately are less energy efficient than aboveground processes, which conversely must cope with relatively greater materials handling problems.

Fig. 6 -- Relative efficiencies of various shale oil processes
DESCRIPTION OF THE PROCESS

The proposed oil shale conversion process has potential to more evenly balance energy and materials handling efficiencies relative to conventional retorting processes. The proposed novel concept integrates strip mining with an on-site high power microwave retort. After mining the ore with a mining machine such as a dragline, the ore would be crushed by a hammermill, then moved by conveyor to the retort, which would rapidly pyrolyze the kerogen to vapor and gas. Spent shale would be continuously conveyed from the retort to the back of the mine-cut and the mine backfilled.\textsuperscript{13} A schematic comparison of this process with a conventional aboveground retorting process is shown in Fig. 7.

Two features of this process are distinctive: the combination of the retort with the mining machine, and the high power microwave retort. Strip mining is an established mining method. This process proposes to integrate the mining machine with a high power microwave retort. By conducting the retorting on-site, shale hauling costs can be reduced. By using a high power retort that heats the shale quickly, the kerogen in the shale can be pyrolyzed quickly, thereby reducing coking. The effect of the microwave heating is to selectively heat the kerogen rather than the inorganic material, thus reducing heat losses to the mineral matter and thereby decreasing overall retorting energy requirements relative to conventional aboveground retorting processes. This process has the additional benefit of reducing the amount of salts produced and therefore reducing the environmental problems associated with this process. Because the microwave retort is the least developed element of the process (large strip-mining machines are commercially available and the technology is well-established), we analyzed this part of the process in depth. Although the integration of the mining and retorting facility was beyond the scope of this work it is recommended that the problem be further investigated.

\textsuperscript{13}For further discussion of waste disposal and land use issues, see Rubenson and Pei (1983b).
Fig. 7 -- Comparison of proposed microwave process with conventional aboveground retorting
TRADEOFFS TO BE CONSIDERED

In this subsection, some of the technical aspects of the use of high power microwave radiation to retort oil shale will be discussed. We will identify a set of tradeoffs in the physical parameters, such as frequency of radiation to be used, shale heating rate, or shale oil production rate.

The dielectric heating process converts electromagnetic energy supplied by an oscillator or generator into thermal energy to heat oil shale. Energy is absorbed from the electric field as the field does mechanical work by rotating the polar molecules in the shale. Some of the rotational energy of the molecules is dissipated in the shale as heat, and it is this dissipation of energy that provides the heat necessary for pyrolysis. There are many advantages to using dielectric heating as a means for recovering oil from shale. Dielectric heating techniques make it possible to heat shale in a uniform manner, under controlled conditions. This in turn can reduce the amount of coking that occurs, increase the overall yield of carbonaceous material obtained from a given grade of shale, and decrease the pour point of the raw shale oil. Moreover, it is expected that environmental problems will be less severe with dielectric heating methods.

The ability of an electromagnetic field to heat shale, measured in power, depends upon both the magnitude and the frequency of the applied field. If the shale is treated as a dielectric material, then the power per unit volume absorbed by the shale from the field is given by

\[ P = \frac{1}{2} \varepsilon'' \omega E^2 \]

where \( \varepsilon'' \) is the imaginary part of the dielectric constant, \( \omega \) is the angular frequency of the applied fields, and \( E \) is the electric field. Thus for a given material the absorbed power can be increased by increasing either the frequency of radiation (\( \omega \)) or the electric field (\( E \)).

It has been found that the magnitude of the electric field that can be used to heat the shale is limited by the electrical breakdown characteristics of the shale. This breakdown occurs when field
strengths (E) reach a point where arcing across a narrow path in the
shale effectively shorts out the system. The electrical breakdown of
shale at frequencies of interest is unknown, but if this process is
conducted in air, then the electrical breakdown of air determines the
highest electric field strength that could be used. The electrical
breakdown of air occurs at field strengths of approximately 30 peak
kV/cm.\(^{14}\) This breakdown voltage is approximately proportional to
pressure and inversely proportional to absolute temperature. Thus,
because the electric field will be constrained by the conditions
described above, to maximize the power absorbed by the shale, radiation
of the highest possible frequency at which absorption takes place should
be used.

An effect that depends upon radiation frequency and competes
directly with the absorbed power is the decay length of the electric
field in the material, also known as the attenuation length. The
attenuation length of an electric field in shale depends on the
frequency of radiation and to a lesser extent on the composition of the
shale. The exact functional dependence of the attenuation length on
frequency differs for good conductors versus poor conductors, and
whether or not the radiation is near an absorption resonance. In all
cases, however, the attenuation length is inversely proportional to some
power of the frequency of radiation. For a dielectric material, near a
frequency of high absorption, the attenuation length of the electric
field (\(\delta\)) is proportional to

\[
\delta \sim \frac{\varepsilon''}{(\varepsilon' \times \omega)}
\]

where \(\varepsilon'\) is the real part of the dielectric constant, \(\varepsilon''\) is the
imaginary part of the dielectric constant, and \(\omega\) is the frequency of the
radiation.\(^{15}\) From this discussion it can be seen that an optimal
frequency of radiation and electric field strength must be determined
that will produce a maximum yield of shale oil with minimal absorption
of electromagnetic radiation by the shale rock.

\(^{14}\)Reference Data for Radio Engineers (1968).
\(^{15}\)Jackson (1982), pp. 284-298.
The greater the power absorbed per volume of shale, the higher the heating rate. It has been found in experiments conducted by Lawrence Livermore Laboratory\textsuperscript{16} that oil yield from pyrolysis of oil shale increases as the heating rate increases and decreases as the pressure increases. This is shown in Fig. 8. Heating rates of 100-700 degrees C/hr and a pressure of 1 atmosphere produced oil yields of 97-100 percent of Fischer Assay, respectively. However, as shown in Fig. 9,\textsuperscript{17} the temperature at which the maximum oil yield is attained rises as the heating rate rises.

\textsuperscript{16}Burnham and Singleton (1983).
\textsuperscript{17}Burnham and Singleton (1983).
Fig. 9 -- Fraction of oil evolved as a function of temperature for several heating rates
The increase in yield with increased heating rate can be explained by considering the pyrolysis mechanism. The exact mechanism involved in pyrolysis of oil shale is complex but is believed to involve the following sequence of first-order kinetic reactions:\textsuperscript{18}

\[
\begin{array}{c}
\text{Kerogen} \rightarrow \text{Gas} \\
\downarrow \\
\text{Rubberoid} \rightarrow \text{Oil} \\
\downarrow \\
\text{Bitumen} \rightarrow \text{Oil} \\
\downarrow \\
\text{Semicoke} \rightarrow \text{High Molecular Weight Oil} \rightarrow \text{Gas + Oil} \\
\downarrow \\
\text{Coke} \rightarrow \text{Oil}
\end{array}
\]

The formation of semicoke and coke is a reaction sequence in competition with the formation of oil and gas. By rapid heating, the formation of gas and oil is favored over the formation of semicoke and coke. A reaction of secondary importance in reducing the oil yield is the thermal decomposition (cracking) of kerogen. Although the total amount of carbonaceous material produced from the shale remains fairly constant, the cracking of kerogen increases the ratio of gas to oil and has been found to increase with increasing particle size.\textsuperscript{19} This increase in the gas-to-oil ratio occurs because oil vapors take longer to exit larger particles and therefore spend more time within the pyrolysis region.\textsuperscript{20}

The oil cracking effect and the attenuation length effect both dictate smaller particle size for maximum oil yield. But smaller particles mean higher crushing costs. Hence, shale particle size is also an important parameter to optimize.

\textsuperscript{18}Dumbaugh et al. (1977); Johnson et al. (1975).
\textsuperscript{19}Galloway and Sandholtz (1979).
\textsuperscript{20}Burnham (1981).
Note that in the final stages of oil evolution, i.e., as the oil yield increases towards 100 percent of Fischer Assay, the relative amount of high molecular weight organic material produced from semicoke also increases. The effect of the high molecular weight material is to increase the pour point of the resultant shale oil, which in turn affects the means of transporting the product to a refinery. Hence a tradeoff to be considered is the total oil yield obtained for economic value versus the pour point and handling considerations of the resultant oil.

Two other parameters that are influenced by the heating rate and are of importance to the amount of coking that occurs are the permeability and porosity of the mineral substrate. It has been found experimentally that, as shale is heated, the chemical reactions produce enough permeability to allow the evolved gases and oil to flow through the shale.\(^{21}\) Increasing the heating rate allows the liquid organic material to quickly vaporize and therefore rapidly exit from the material. In fact, the permeability of the mineral matter is such that, if the kerogen could be pyrolyzed without the formation of any coke, the remaining minerals would not form a self-supporting structure and therefore would tend to collapse. It is the crosslinked organic material that gives shale its rigidity.

The heat requirements for retorting oil shale and for the thermal decomposition of the minerals present in oil shale have been estimated.\(^ {22}\) The total heat required to heat and retort kerogen from 77 degrees F to 891 degrees F (retorting conditions) is approximately 530 Btu/lb. The mineral heat requirement over the same temperature range is approximately 230 Btu/lb. Some uncertainty exists in these figures, since several endothermic and exothermic reactions take place as the temperature of shale is raised. The endothermic reactions are the result of evolution of hydrate water and decomposition of the minerals. The exothermic reactions are attributed to the combustion of the carbonaceous materials. The degree to which these reactions influence

\(^ {22}\) Cook (1970), p. 133.
the heat requirements depends upon retort conditions, such as the amount of oxygen present during retorting, and upon grade and composition of the shale and the amount of water bound as hydrates in the minerals.

If the microwave energy is selectively absorbed by the kerogen, as is suggested by Judzis, Williams, and Hiatt\textsuperscript{23} and shown in Fig. 10, then only a fraction of the mineral matter will be directly heated by the microwave energy. The heating of the mineral matter will thus rely mainly upon conduction and convection mechanisms, which are relatively slow on the time scales involved in dielectric heating. In addition, the thermal conductivity and thermal diffusivity, which are measures of the ability of a material to conduct heat, both decrease with increasing shale grade and with increasing temperature as shown in Figs. 11 and 12.\textsuperscript{24} The thermal conductivity ranges from a high of about 1.0

\textsuperscript{23}Judzis, Williams, and Hiatt (1977).
\textsuperscript{24}DuBow (1977).
Fig. 11 -- Thermal conductivity vs temperature for five different grades of oil shale

Fig. 12 -- Thermal diffusivity vs temperature for oil shale
Btu/ft-hr-deg F for 6.8 gal/ton shale to about 0.40 Btu/ft-hr-deg F for 48 gal/ton shale and decreases monotonically with increasing temperature.

If only a fraction of the mineral matter is actually heated in this process—or equivalently the mineral matter experiences only a fractional temperature rise—then the heat requirements will be reduced. The amount of reduction depends on the richness of the shale, the exact individual heat requirements of the mineral matter and the oil, the thermal conductivity, and the heating rate. The process of retorting oil shale using high power microwave radiation is proposed on the merits of such a decrease in the relative heating of the mineral matter. From the above discussions, theoretically, if shale is subjected to microwave energy in which kerogen is quickly heated to pyrolytic temperatures, oil vapors have exited the shale particle before significant thermal energy has been dissipated to the mineral matter.

The daily peak power requirements can be obtained by combining the energy absorbed per ton of shale with the desired daily production rate, the conversion efficiency of electricity to microwaves, and the conversion efficiency of microwaves to heat energy. The change in each of these variables necessary to decrease the peak power requirement is described below.

<table>
<thead>
<tr>
<th>Change in variables needed to reduce peak power requirements</th>
</tr>
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<tbody>
<tr>
<td>Variable</td>
</tr>
<tr>
<td>-------------------------------------------------------------</td>
</tr>
<tr>
<td>1. Fractional temp rise of minerals, f</td>
</tr>
<tr>
<td>2. Conversion efficiency electricity--microwaves, $\eta_{mc}$</td>
</tr>
<tr>
<td>3. Conversion efficiency microwaves--heat, $\eta_{ht}$</td>
</tr>
<tr>
<td>4. Production rate, Z</td>
</tr>
<tr>
<td>5. Number of hours per production day, T</td>
</tr>
</tbody>
</table>
As an example, consider 20 gal/ton shale, \( \eta_{mc} = 0.8 \), \( \eta_{ht} = 0.8 \), \( Z = 10,000 \) bbl/prod-day, and \( T = 24 \) hour/prod-day. These are rather modest values in terms of production rate and shale richness. Also, the conversion efficiencies are assumed to be conservative estimates of the true efficiencies. Then as the fraction of heat \( (f) \) absorbed by the mineral matter ranges from no heat absorbed \( (f = 0) \) to complete mineral heating \( (f = 1) \) the absorbed energy/barrel required for production ranges from (App. D):

(a) Energy/barrel \( (f = 0) = 0.172 \) million Btu/bbl
(b) Energy/barrel \( (f = 1) = 1.06 \) million Btu/bbl

These values represent extremes: (a) in the best case, the rock absorbs no heat, and (b) in the worst case, the rock is completely heated to retort conditions. In reality, although it is hypothesized that energy absorbed by the mineral matter will be low for the reasons already stated, the actual amount absorbed given the conditions proposed must still be experimentally determined. The difference in the theoretical maximum and minimum amount of energy absorbed by the kerogen and mineral matter per barrel of oil produced is six-fold; this calculated range represents a bounded region in which the actual peak power required by the retort can be expected to lie for the conditions proposed. This peak power requirement will determine the power capacity required by the device, i.e., what must be supplied by the power grid. Taking into account the daily production rate and accounting for the conversion efficiencies, the above energy values imply the following peak power requirements (App. D):

\[
\begin{align*}
\text{Power}(f = 0) &= 33 \text{ MW} \\
\text{Power}(f = 0.5) &= 118 \text{ MW} \\
\text{Power}(f = 1) &= 203 \text{ MW}
\end{align*}
\]

In the case of our example described previously, if the rock absorbs no energy in the conversion of kerogen to oil vapor, a device will require
from the power grid a peak power of 33 MW. On the other hand, if the rock were to be completely heated during the retorting process, a device will require from the grid a peak power of 203 MW. Thus, the degree to which rock is heated (or not heated) during retorting will have important cost implications. Ten MW microwave devices are commercially available; experimental microwave devices generating 1000 MW or 1 GW have been proposed but not as yet built.

Assuming a continuous mode of operation, the device residence time can be determined from the maximum temperature rise divided by the heat rate. The greater the heating rate, the smaller the process time. The product of the volume flow rate and the residence time gives the minimum volume of the device. Therefore, a shorter residence time will have two positive effects: it will decrease the amount of heat lost in the spent shale and will decrease the size of the retort required.

To optimize this process, tradeoffs will be made among many physical parameters. A frequency of microwave radiation must be determined that falls within the range allowable by the FCC and that coincides with an absorption maximum for kerogen. This frequency must be high enough to provide rapid heating of the kerogen but not so high that the attenuation length is small enough to require excessive crushing of the oil shale. Experimental studies will have to be performed to make these tradeoffs and to determine the engineering and economic feasibility of this process.

ECONOMIC IMPACT

Five basic steps are involved in the production of transportation fuels from oil shale: (1) shale mining and crushing, (2) ore transport, (3) retorting, (4) upgrading, and (5) spent shale disposal. The technology as proposed here combines the mining and crushing, retorting, and spent shale disposal into one operation, hence it

- Virtually eliminates costs associated with hauling; and
- Reduces spent shale disposal costs.
Disposal costs are assumed to be lower, since hauling costs are also reduced in that step, and waste heat recovery is expected to be minimal. It is believed that each of the three steps—(1) shale mining, crushing and disposal; (2) retorting; and (3) upgrading of raw shale oil—contributes about one-third of the cost of producing raw shale oil. Additional costs are incurred in operation of utilities and general (offsite) facilities. Water requirements are expected to be reduced in this concept since

- Waste heat disposal will be reduced; and
- Dust generation will be reduced.

The lower water requirements and the smaller leachable salt formation should reduce the environmental impact. Both of these attributes are important because a likely location for commercial sized oil shale conversion plants will be in arid areas.

The largest benefit to be derived from the use of high power microwave retorting is the reduction in energy consumed in pyrolysis, hence lower electricity costs. A study conducted by IITRI in 1984 concluded that the operating costs for an in-situ radio frequency retort ranged from $9.40 to $22.00 per barrel of oil depending upon whether power was obtained from a dedicated coal-fired power plant or was purchased from a local utility. In this analysis 85 percent of Fischer Assay oil recovery from 25 gal/ton shale was assumed. The radio frequency radiation was produced from solid state generators of 1 MW size with an efficiency of 90 percent. No capital was generated from by-products. The power costs alone ranged from $4.00/barrel for power obtained from a dedicated plant (no demand charge) to $16.50/barrel for purchased power. The capital investment ranged from $40,000 per barrel/day capacity to $20,000 per barrel/day capacity for the respective cases.²⁶

Assuming a fourfold decrease in the amount of energy required to pyrolyze the kerogen using high power microwaves, based on previously

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described calculations, the reduction in electricity costs would more than offset the increased mining costs associated with the aboveground process. Indeed, at current electricity costs of 10-15 mils/kWh in the Bonneville Power area,\textsuperscript{27} the electricity cost per barrel (neglecting demand charge) ranges from about $3.75/barrel for full mineral heating, to $0.63/barrel for no mineral heating. The price with full mineral heating, $3.75/barrel, should be compared with the price obtained by IITRI, $4.00/barrel, in the above example with a dedicated power plant. The discrepancy is due to rounding.

An in-depth analysis of all costs of production should be performed after bench scale or pilot plant tests are conducted from this process. In particular, it is important to determine how much energy is absorbed by the mineral matter when retorting with high power microwaves.

ENVIRONMENTAL/INFRASTRUCTURE IMPACT

There is a clear lack of existing infrastructure for the described process as is the case with most other retorting processes. Although there are power generation facilities in regions with the richest shale resources, the ability to provide the required power is limited. Therefore, construction of a high power microwave retorting facility would require the construction of a supporting power generating facility, dedicated or not. In the Bonneville Power area (which services the Green River formation), two nuclear power generating facilities are presently on hold, pending an increase in demand sufficient to warrant such large generating capacity.\textsuperscript{28}

In addition to a lack of power generation facilities, there is a lack of general facilities capable of supporting such an operation. In an environmental impact statement conducted in 1977 by the U.S. Energy Research and Development Administration, the costs for community expansion resulting from development of a 45,000 barrel/day oil shale facility were estimated to be $5 million annually.\textsuperscript{29}

\textsuperscript{27}Personal communication from the Pacific Gas & Electric Company.
\textsuperscript{28}Personal communication from the Pacific Gas & Electric Company.
The water resources needed to operate such a facility must also be treated carefully. Existing water pollution control technology may be applicable to most of the waste streams encountered in oil shale processing. In comparison with conventional aboveground retorting processes, this process is expected to produce waste water that is

- Less hazardous; and
- Smaller in volume.

It may be necessary to conduct research on waste water generated from this process to determine the amount of priority pollutants and toxics present, and to ensure safe reuse or discharge of the water. Although the environmental problems posed are expected to be less severe than in conventional retorting processes, purification of process waste water, spent shale compaction waste water, and mine dewatering waste water poses problems that will have to be resolved before commercialization of this process.

CONCLUSIONS AND RECOMMENDATIONS

Aboveground retorting of oil shale using high power microwave radiation is technically feasible, and it is believed that by integrating the retorting and mining portions of oil shale processing, energy efficiencies will increase, and materials handling efficiencies will be improved relative to other aboveground retorting processes. The retorting portion of this process makes use of the dielectric heating mechanisms involved in microwave heating to rapidly pyrolyze the kerogen in shale, and also takes advantage of the low thermal conductivity of shale to minimize the amount of heat lost to the spent shale. Low heat loss has the added benefit of reducing the amount of leachable salts formed in the spent shale and, therefore, reducing environmental hazards. The integration of mining and retorting has the effect of virtually eliminating hauling costs.

It appears that this process could substantially decrease the cost of recovery of shale oil. Electricity costs for retorting could theoretically be relatively low, although engineering tests must be conducted to assess the true cost.
We conclude that a key to the success of aboveground retorting of microwave energy is the attainment of high heating rates. If it can be demonstrated that heating rates on the order of 1000 degree C per minute can be achieved, we believe this process could provide significant improvements in efficiencies and economics in aboveground retorting. It is recommended, therefore, that an engineering scale-up of this concept be undertaken to test the range of working parameters and thereby estimate the actual economic impact to shale oil recovery by this method.
IV. CONCLUSIONS AND RECOMMENDATIONS

We have conducted in-depth analyses of lignin/coal/oil coprocessing and aboveground microwave retorting of oil shale. Our purpose has been to assess their promise for significantly lowering the cost of producing liquid hydrocarbon fuels from domestic solid fossil fuels.

We determined that an important economic consideration for the success of lignin/coal/oil coprocessing was the amount of lignin required. Technically, there are many reasons to suggest that this novel concept has promise: Lignin exhibits a catalytic quality as it and coal are liquefied at temperatures as low as 300 degrees C. It appears to increase destabilization of the coal molecule at lower temperatures and moderates pressures better than conventional liquefaction processes, making it more susceptible to hydrogenation and ultimately liquefaction.

The addition of lignin to the coal/oil coprocessing technology is also believed to reduce the problem of incompatibility between paraffinic oil and aromatic coal molecules. The chemical compatibility problem between solvent and coal is thought to be partly responsible for the sensitivities of yield and optimal operating conditions to coal/oil combinations in conventional coprocessing.

The economic feasibility of this process depends on the required lignin resource base. If this novel concept were to satisfy 25 percent of the U.S. demand for transportation fuels, given the total amount of lignin currently consumed by the pulp and paper industry, the maximum lignin to coal weight ratio possible is 1:19. However, the technical success of this concept could spur the search for other sources of lignin. A research agenda that would address key questions would include determining chemical feasibility, process yields, product spectrum, and the optimum lignin/coal/oil ratios. Part of the latter determination will depend on where lignin is introduced to the process. A search for solvents that have similar effects to lignin on coal/oil coprocessing should also be conducted.
The process of retorting oil shale using high power electromagnetic radiation in the microwave portion of the spectrum has been shown to be scientifically sound and technically feasible. Component steps in the process of producing shale oil from this process are well developed or involve technologies that have been used in other applications. Strip mining techniques are well developed. Electromagnetic absorption spectra of oil shale have been obtained and resonances observed in the microwave region. Microwave devices of sufficient power are commercially available, and upgrading techniques and facilities are also well established. The novelty of this concept lies in the use of microwave devices sufficiently powerful to produce a high rate of retorting of the kerogen, thereby taking advantage of the low thermal conductivity of shale to minimize the amount of heat loss to the mineral portion of the oil shale and subsequent salt formation.

Prior attempts at using electromagnetic energy to retort oil shale have focused upon low power or slow heating using radio frequency or microwave radiation, generally in in-situ applications. Although these processes have been shown to be technically feasible they consume large amounts of electricity because of heat losses to the surrounding environment. In contrast, the high power microwave energy envisioned for use in this application would reduce the amount of exoteric heat loss.

Studies have been performed on microwave absorption, thermal conductivity, and reaction sequences involved in the production of shale oil. In addition, basic research has been conducted on the effect of heating rate on oil yield; such research has shown that yield increases with heating rate. Engineering research must still be performed to produce a microwave retort design that optimizes the yield of shale oil for a given radiation frequency and power level. Investigations of impedance matching will also be required to maximize the conversion of microwave energy to heat energy. Further, the environmental impact of such a process must be accurately determined. This impact is expected to be less than for other commercially tested means of producing shale oil from oil shale. This type of research could conceivably be completed within a five year period.
In principle, this process could significantly reduce electricity consumption in comparison with slow electromagnetic heating processes such as in-situ radio frequency heating. At the same time, rapid heating is expected to produce 100 percent of the Fisher Assay amount of oil contained in the shale. The oil produced may be of higher quality and have a lower pour point than shale oil produced by conventional retorting schemes. In addition, the environmental impact will be decreased, relative to conventional aboveground retorting, because of reduced production of leachable salts and combustion products. It is recommended, therefore, that an engineering scale-up of this concept be undertaken to test the range of working parameters and thereby to establish the actual economic impacts to shale oil recovery by this method.
Appendix A

IDENTIFICATION AND SELECTION OF NOVEL CONCEPTS

SEARCH FOR NOVEL CONCEPTS

This research began by establishing general criteria for what would be considered "novel." It was preferable that a novel concept address one or more of the key cost driving elements in current commercial systems, such as hydrogen use in the production of coal liquids or energy consumption in the retorting of oil shale. Novel concepts taken singly must affect economically significant portions of the total system to have important effects on product costs. Concepts that contributed marginally to cost reduction were not considered for further analysis, since significant cost reduction was a primary requirement.

To identify, explore, and develop ideas for novel concepts we made patent and literature searches, and held discussions with research scientists and engineers in-house at RAND, in the academic world, and in a variety of industrial and government laboratories. In addition, a workshop sponsored by ANL provided a forum for the exchange of ideas with other experts on advanced, innovative concepts. Some time was also devoted to brainstorming sessions in-house, resulting in several promising candidates, including the two described in the body of this Note.

SCREENING CRITERIA FOR SELECTION OF THE MOST PROMISING CONCEPTS

General Background

We defined a novel concept as a technical approach that could potentially significantly reduce the cost of fossil fuel production and that falls into one of the following categories:

- An entirely new technical approach to fossil energy production or conversion;
• An approach that was previously explored and discarded for reasons now inappropriate;
• Concepts presently being explored at a low level of support but whose promise is seriously underestimated; and
• New applications of existing technology.

To select a few of the most promising candidates for further study, screening criteria provided by ANL were expanded by RAND. Our guiding principle was the requirement that the screening criteria be commensurate with the quality of the data available on the novel concepts. This requirement cannot be overstated. It is nearly always preferable in any technical assessment to perform quantitative analyses whenever data permit; however, in the absence of necessary data one may be forced to resort to less rigorous methods of analysis.

To determine how rigorous our method of evaluation of an innovative concept could reasonably be, we characterized concepts in terms of a parameter common to all—i.e., the extent of research and development conducted to date. This is illustrated in Table A.1. Ideas fit into one of three basic categories of research and development: previous research and development performed, ongoing R&D, and research conducted at a relatively low level. These categories suggest that the resolution of the evaluation criteria will depend on the amount and type of

Table A.1

<table>
<thead>
<tr>
<th>Previous R&amp;D</th>
<th>Ongoing R&amp;D</th>
<th>Little or No R&amp;D</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a) Rejected concepts or those put on indefinite hold</td>
<td>(c) Underemphasized by DOE, others</td>
<td>(d) Basic science known</td>
</tr>
<tr>
<td>(b) New applications of proven technologies</td>
<td></td>
<td>(e) Basic science new</td>
</tr>
</tbody>
</table>
information available. Although it may be possible to quantitatively analyze items in technology category (a), items in category (d) may not be as amenable to the same methods. Examples of concepts that fall into these categories include:

(a) Coal pyrolysis unit collocated with an electrical utility;
(b) Membrane technology as oxygen enrichment for end-use devices or in-situ gasification;
(c) Aboveground retorting with high power microwave power;
(d) Coal/lignin/oil coprocessing;
(e) Liquid clathrates.

On the basis of available data, it is thus unlikely that liquid clathrates or novel forms of coprocessing could be analytically evaluated with the same rigor as a coal pyrolysis unit collocated with an electrical utility.

One of our first tasks was to identify and characterize the potential positive and negative aspects of different technical approaches, as well as to assess the breadth and depth of existing information pertaining to the concepts. A representation of these efforts appears in App. B. Technical descriptions for selected technologies were given, along with their role in the recovery/conversion process. These brief descriptions and the screening criteria were of assistance in selecting concepts for Phase II.

**Screening Criteria**

Screening criteria developed by RAND and ANL were used to identify concepts from Phase I that offered promise for the enhanced production of U.S. fossil fuels. These criteria included:

- The current level of technical understanding or uncertainty for a concept;
- Its potential economic and resource impact;
• The research status and requirements associated with the
technical approach; and
• Its use of existing infrastructure in addition to environmental
impact.

Level of Technical Understanding. The current level of technical
understanding that exists for a concept can be measured in terms of the
scientific, technical, and engineering uncertainties associated with it.
An examination of these categories yields more information than just a
concept's technical plausibility or likelihood of succeeding. They also
suggest the research requirements and associated risks in bringing the
technical approach from the conceptual stage to development. For any
novel concept basic types of uncertainties include:

• Scientific, in which basic knowledge is unknown;
• Technical, in which basic knowledge or science is known, but
  the technology is unproven; and
• Engineering, in which the basic knowledge is known, the
technology is proven, but the particular application in mind or
  device desired is unproven.

We began by examining the scientific principles governing the
technology. As a first step, it is useful to determine whether the
basic science underlying a novel concept is known and to identify the
magnitude of technical or engineering uncertainties. Different sets of
concerns that can have important implications for research are suggested
by each type of uncertainty. For example, the issues involved in
analyzing a proposed large-scale mining and retorting unit will be quite
different from those pertinent to a proposed new chemical process.
These distinctions were helpful in identifying gaps of knowledge in
current technology. Later, when promising concepts were identified, the
economically significant uncertainties were explored in greater depth.

Economic and Resource Impact. Novel concepts were screened
according to their likely economic and resource impact. Consideration
was given to the estimated economic impact on product cost, whether the
approach opened up other energy resources, or if the concept had other applications. For instance, horizontal well techniques for enhanced oil recovery have the potential of opening up greater oil reserves in known fields. Additional applications of a novel concept can dramatically increase the cost-effectiveness of research funding and can benefit other energy-related technology areas.

Our goal was to measure criteria quantitatively whenever data permitted, such as the expected reduction in product costs in terms of percentage decrease. The resolution of our information dictated how refined our evaluation criteria could reasonably be. In measuring potential cost impact, we used units that in some way mirrored or took into consideration the inherent uncertainty in the concept itself. Thus, our evaluations very often involved bounded values rather than discrete quantities.

Another important consideration is that when comparing novel concepts with currently existing technologies, one must recognize that the true effect of the integration of a novel concept is very difficult to predict. Without actually testing a concept, unseen problems and sometimes benefits very often arise that will distort an estimation. Nonetheless, it is useful to understand the direction of cost change. When it proved too difficult to predict the magnitude of effect on a given system by the introduction of innovation, we turned to technical indicators that at least pointed in the direction of change. Depending on the process involved, some indicators include temperatures, pressures, rate of heating, efficiencies, power required, etc.

**Research Status and Requirements.** Research requirements flow from the previously discussed uncertainties concerning the technical success of a concept. From the types of unknowns surrounding a technology, one can roughly gauge how much effort is required to bring the idea from the conceptual stage to a completely integrated process. For example, a concept with scientific uncertainties requires a qualitatively different agenda than those ideas involving engineering uncertainties, and it will probably take longer to commercialize than a concept based on proven technology.
It is very difficult to estimate the time required to resolve key issues for an innovative concept, since this is a function of other variables, such as the level of funding, complexity of the unknown, etc. Similar arguments can be made for the cost of research and expected chance of success. The cost of the research program will be governed by the kind of issues that must be resolved. Of great interest to policymakers is the chance of a concept's success, i.e., will it deliver what it promises. To evaluate this criterion, one must have adequate information on which to base this judgment. Although this information may be available for some cases (e.g., novel concepts requiring scientific breakthroughs have an associated small probability of success), for others this information is lacking.

It is desirable for novel concepts to continue to show promise as research progresses. To monitor research results and be guided by the conclusions reached, there are certain minimal goals that should be reached before going ahead. Research goals should address the most significant economic and technical problems. Once basic and fundamental questions are favorably satisfied, a new research agenda is advisable following yet another level of basic concerns and so on. It is reasonable to expect that at some point the concept has been explored sufficiently such that if market conditions proved favorable, the private sector could commercialize the concept.

**Infrastructure Availability and Environmental Concerns.** On a broad systems level, a novel concept was evaluated in terms of infrastructure availability and environmental concerns. One of the more important criteria a concept can be measured by is the amount of existing infrastructure it is capable of using in the production of liquid fuels. Billions of dollars have been invested in the petroleum, mining, and fuel distribution industries. Many more billions of dollars are implied by near-commercialization approaches to the enhanced production of domestic liquid fossil fuels. Because much of the product cost is capital-related, use of existing infrastructure can be one of the most important avenues available to cutting product cost. This notion is a very important economic component in coal/oil coprocessing, for instance.
New fossil fuels also involve important socioeconomic issues such as the expected impact in emissions, land use, waste disposal, worker safety, general problems, or benefits. In particular, if environmental problems were addressed or incurred by the novel concept, this was noted. For example, a benefit of rapidly retorting shale is the reduced production of leachable salts, thus lessening environmental impact, relative to conventional retorting technologies.

Although institutional barriers are of concern to novel concepts, their impact was not very clear, since these obstructions are more subject to change than the previously mentioned criteria. Very often technological advances can even force institutional change.

APPLICATION OF SCREENING CRITERIA

The above criteria were applied to the list of novel concepts identified in Phase I. From this exercise and after consultation with Argonne, two candidate technologies were chosen for in-depth analysis in Phase II. At this point we explored in greater depth areas that conceivably were economic or technical bottlenecks, i.e., those areas that have great influence over the success of a given technology.
Appendix B

EXAMPLES OF NOVEL CONCEPTS CONSIDERED

BACKGROUND

This appendix presents selected concepts identified during Phase I and a brief discussion of key economic issues, suggested agenda, etc. The discussions indicate the type of analysis performed in our initial assessment of each novel concept's economic promise.

The following technical discussions describe a number of novel approaches to producing, transforming, or using fossil fuels. The goal of our efforts has been to bring to the attention of OGSCL technologies that we believe can significantly lower the costs of or can significantly increase the availability of fossil energy resources. Some of these technologies are highly speculative, although we do not believe that any of them are technically unreasonable. Others are well-known technologies being used in a new way. Some systems would require basic research before any commercial application;¹ others would require only development work or systems integration.

To clarify what we mean by a "novel concept," we have developed a typology and have applied it to each of the concepts described in this Note.

TYPOLOGY OF NOVEL CONCEPTS

Class I: Revival of Previously Rejected Concept

The least innovative concepts that we include are those that have been worked on by government or industry in the past but have been abandoned for reasons that may have been good at the time but are now inappropriate. For example, the flash pyrolysis of coal has been previously researched. However, we would encourage a reexamination of such a system but in the context of a collocated electric power plant/refinery.²

¹Assuming, of course, that the results of the research and subsequent development were favorable for the technology's ultimate commercial introduction.
²EPRI has done some work in this area.
Class II: In Development, but Underemphasized

In the same vein are technologies currently under development by industry or by DOE, but that in our estimation are significantly underemphasized relative to their promise. An example is the use of horizontal wells for heavy oil and tar sands production. Industry has done some preliminary work on the technology, but largely because of the collapse of R&D funding in the past few years, interest in it has decreased.

Class III: First-Time Integration or New Application of Proven Technologies

This type of novel concept involves the first-time integration of off-the-shelf technologies or a new application of existing technologies. For example, the use of membranes to enrich air for internal combustion engines falls into this category. Although some modest development of appropriate membranes will be necessary, most of the required work is integration of available technologies.

Class IV: Major Development Necessary for Commercialization

Concepts in this category require a good deal of R&D before their feasibility can be established but, unlike the next category, do not require the use of revolutionary technology. An example in this category is our coal/oil/lignin coprocessing scheme. Although the idea looks promising in a number of important respects, it will require a fair amount of experimentation and analysis before a commercial design could be completed.

Class V: Radically New System--Basic Issues to Be Resolved

Technologies in this category entail the use of fundamentally new ideas, usually based on new developments in science. These technologies are the most speculative of those examined but sometimes promise the greatest improvements in cost. An example in our work would be the use of liquid clathrates for coal liquefaction.
NOVEL CONCEPTS IDENTIFIED

Some of the novel concepts from Phase I of this research considered as promising candidate technologies are listed in Table B.1. They are grouped according to the typology previously described.

1. ADVANCED STIRLING ENGINE

Concept Type

Class I: Revival of previously rejected concept.

Original Developer

N. V. Philips Gloeilampenfabrieken of The Netherlands. Both GM and Ford have worked with Philips under licensing agreements.

Status

A free piston Stirling engine designed to investigate the dynamics of this concept has been under test at the NASA Lewis Research Center for several years. Since 1972, a team composed of Mechanical Technology Incorporated, United Stirling of Sweden, and AM General has been conducting development work aimed at producing an automotive power plant.

Problems Addressed

End-use devices must be considered as an integral part of any new fuel development. Present day spark ignition and diesel ignition internal combustion engines have very stringent fuel requirements especially with regard to octane number, cetane number, pour point, and amount of impurities. Burning a fuel in these engines that is outside of their standards typically requires one or several add-on devices. On the other hand, liquids from coal or shale are, at present, economically unattractive to upgrade to the rigid standards of petroleum fuel look-alikes. These new fuels could be made more economically viable if the expensive processing and upgrading requirements such as hydrotreating could be reduced. This in turn can be accomplished by designing and building engines that are capable of burning a broader range of fuels with less rigid requirements for octane number, cetane number, etc. The
### Table B.1

**NOVEL CONCEPTS IN THE PRODUCTION AND CONVERSION OF FOSSIL FUELS**

<table>
<thead>
<tr>
<th>Class I: Previous R&amp;D; concept previously rejected</th>
<th>Use-Category</th>
</tr>
</thead>
<tbody>
<tr>
<td>Advanced Stirling engines</td>
<td>End-use device</td>
</tr>
<tr>
<td>Coal pyrolysis integrated with electrical power generation processes</td>
<td>Conversion</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Class II: Ongoing R&amp;D; concept underemphasized</th>
<th>Use-Category</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beneficiation of coal through supercritical explosive coal shattering</td>
<td>Beneficiation</td>
</tr>
<tr>
<td>Coal liquid production using a supercritical gas extraction approach</td>
<td>Conversion</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Class III: Previous R&amp;D; new application or integration of concept</th>
<th>Use-Category</th>
</tr>
</thead>
<tbody>
<tr>
<td>Enhanced heavy oil/tar sands production using horizontal wells</td>
<td>Resource extraction</td>
</tr>
<tr>
<td>Integrated surface mining/microwave retorting approach to shale oil production</td>
<td>Conversion</td>
</tr>
<tr>
<td>Modification of the internal combustion engine: enriched oxygen supply to the combustion chamber with simple membrane device</td>
<td>End-use device</td>
</tr>
<tr>
<td>Production of biopolymers aboveground for enhanced oil recovery.</td>
<td>Resource extraction</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Class IV: Little or no R&amp;D to date; basic principles known</th>
<th>Use-Category</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal liquefaction with elimination of purified hydrogen</td>
<td>Conversion</td>
</tr>
<tr>
<td>Generalized coprocessing: Lignin/coal/oil coprocessing</td>
<td>Conversion</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Class V: Little or no R&amp;D to date; basic principles not well understood</th>
<th>Use-Category</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low severity coal liquefaction using liquid clathrates</td>
<td>Conversion</td>
</tr>
</tbody>
</table>
Stirling engine is one of two end-use devices that have been identified as having this multifuel capability and that have high thermal efficiencies.

Description of the Proposed System

The Stirling engine is an incarnation of the Carnot heat cycle in which heat from the combustion of an air-fuel mixture is converted to mechanical work. The Stirling engine is an external combustion engine that, in contrast to a standard internal combustion engine, has a working fluid and a heat source maintained separately from one another.

Key Features of the Proposed System

Since the combustion of fuel occurs external to the working fluid, the combustion can be controlled at the source. This provides a distinct advantage over conventional internal combustion engines in that:

- A wide spectrum of fuels can be burned; and
- The combustion process can be strictly controlled to keep emissions low.

In addition, maintaining the temperature of the coolant reservoir at a constant low level permits a high thermal efficiency.

Potential Problems

Potential limitations of the Stirling engine include its large size and hence large cooling requirements, making it unsuitable for small vehicles. The large size of the engine is a result of the number of ancillary devices required to run it. The cooling requirements are directly related to the size of the engine and to thermal efficiency.
Key Items for Further Investigation

- Components from nonstrategic materials such as ceramics must be designed and fabricated;
- A system of seals for containment of the working fluid must be developed; and
- Optimum specific power outputs must be obtained.

II. BENEFICIATION OF COAL THROUGH SUPERCRITICAL EXPLOSIVE COAL SHATTERING

Concept Type

Class II: In development but underemphasized.

Original Developer

Consolidated Natural Gas Research Co. (CNG) of Pittsburgh, Pennsylvania.

Status

A continuously fed 10 ton per day unit, specifically designed to test this technique, is presently being operated by CNG.

Problems Addressed

The coprocessing of coal and heavy or residual oils is one of the most promising routes to the introduction of direct coal liquefaction. The best candidate coals for coprocessing are low-volatile sub-bituminous coals and lignite. These low rank coals, however, generally contain substantial mineral impurities that would complicate coprocessing and render it less cost-effective. It is especially desirable to reduce the sulfur content of the coal; most sulfur in low rank coals tends to be pyritic rather than organic. The proposed candidate technology would effectively reduce the mineral content of coal, pulverize it to a very fine size, and thereby prepare it for coprocessing or other liquefaction processes.
Description of the Proposed System

Explosive coal shattering is a means of separating impurities from coal by selectively breaking the coal into microscopic fragments without significantly breaking up the impurities. This is achieved by subjecting the coal to supercritical water at high pressure. The water "vapor" saturates the pores of the coal but not the mineral impurities. The pressure of the system is then suddenly dropped and the water expands, violently pulverizing the coal in the process. The end results of this process are twofold: coarse mineral products and coal fines. These two can be readily separated by mechanical means. This process is expected to allow the removal of over 50 percent of the sulphur and other ash-forming minerals from the coal.

Key Features of the Proposed System

- This process is expected to work best with coal containing mainly pyritic sulphur, such as bituminous coal or lignite.
- A significant portion of the sulphur and ash-forming minerals can thus be removed from these coals without expensive hydrotreating.
- Once the coal has been pulverized in this fashion the coal and minerals can be separated mechanically, since the coal particles produced are much smaller in size than the impurity particles.
- The small size of the resultant coal fines could also help reduce some of the solids handling problems associated with coal. That is, by mixing the coal fines with oil or water, a slurry can be made that is readily transported. The coal-oil slurry is suitable for direct refinery processing. The coal-water slurry can be burned directly in power plants or large fixed diesels. In these systems, the combustion process is slow, the temperature is high, and fairly complete combustion can take place.
Potential Problems

Potential limitations of this process include:

- The coal fines produced may be wet and therefore an extra water removal step may be needed in a refining/upgrading process.
- Vessels that can process a large volume of solid coal at high temperatures and pressures must be designed and built.

Key Items for Further Investigation

Further pilot research should be conducted to determine the optimum use for coal beneficiated in this manner.

III. COAL LIQUEFACTION WITH ELIMINATION OF PURIFIED HYDROGEN

Concept Type

Class IV: Major development necessary before commercialization.

Original Developer

Sandia National Laboratory.

Status

Basic research.

Problems Addressed

Current direct coal liquefaction processes require purified hydrogen under high pressure to hydrogenate either the solvent or a coal/solvent slurry. This proposed technique eliminates the direct use of hydrogen gas in the hydroaromatic solvent production step and in the primary coal liquefaction stages. Reduced temperatures in the first step and pressures in both stages have been observed. The economic impact of the described technology resides principally in capital cost reductions, a component that accounts for nearly 50 percent of the product cost.
Description of the Technology

This technology builds from past research in the use of the water-gas shift reaction for the generation of in-situ hydrogen for solvent production, but claims novelty in the separation of the solvent production step from the liquefaction phase. Each step is operated according to its own optimal thermodynamic requirements. This discovery, made possible by a better understanding of the chemistry involved, uses milder reaction conditions than conventional approaches.

In the first stage, the hydrogen donor solvent is produced via the water-gas shift reaction at 240 degrees C and 500 psi pressure. Although liquefaction temperatures remain unchanged (greater than 400 degrees C) to promote bond rupture in the coal and transfer of hydrogen from the HDS to coal fragments, pressures of 500 psi were used, a substantial reduction from the several thousand psi required in conventional processes.

Key Features of the Proposed System

Principal features of this process include the following:

- Two-staged process separates the solvent production from the liquefaction reaction and each is operated at optimum conditions. Pressures are reduced for both stages and temperatures in the first step as well.
- Purified hydrogen is eliminated altogether. In-situ hydrogen is generated from steam and synthesis gas.
- Solvent recycle is required.

Potential Problems

Relatively few technical problems are apparent. Despite the novelty of this approach, it builds from a large knowledge base in two-stage liquefaction and therefore should involve less risk.
Key Items for Further Investigation

Although much appears promising with this technology, still more must be explored before a sound economic evaluation may be feasible. Several suggested areas for further investigation include:

- Characterization of product;
- Better understanding of pressure requirements for liquefaction stage;
- Materials and heat balances; and
- Scale-up tests.

IV. COAL LIQUID PRODUCTION USING A SUPERCRITICAL GAS EXTRACTION APPROACH

Concept Type

Class II: In development, but underemphasized.

Original Developer

National Coal Board (NCB), Great Britain.

Status

A continuously fed 120 kilogram per day unit has been operated by the NCB in England since 1977, and a 25 ton per day plant has been scheduled for operation.

Problems Addressed

Coal pyrolysis liquids evolved at or below 400 degree C do not volatilize to any extent, whereas at higher temperatures they tend to decompose. In the presence of a compressed gas substances volatilize more readily, the more so the greater the gas density. The compressed gas thus acts as a strong solvent. At high pressures the density of a gas is a maximum at its critical temperature where its density approaches that of a liquid. By carrying out the pyrolysis in the presence of an extractive supercritical gas with a critical temperature close to but below the pyrolysis temperature, the gas acts as a strong solvent and causes the liquids to volatilize. Large increases in volatility can be realized by this process.
Description of the Proposed System

Supercritical gas extraction is a special low temperature pyrolysis procedure that makes use of the ability of a gas near its critical temperature to extract relatively involatile substances. The extraction gas should have a critical temperature at or slightly below the desired pyrolysis temperature, the range 300-400 degrees C being most desirable. Toluene, whose critical temperature is 319 degrees C, has proved suitable. The low molar mass liquids formed by thermal depolymerization and that are soluble in the gas phase are removed directly as they are formed. The product is a low melting glassy solid that represents the hydrogen-rich fraction of the coal. The residue is a porous char with a high reactivity and calorific value similar to that of the coal from which it was derived.

Key Features of the Proposed System

- The extract is essentially free from mineral matter and solvent, and contains less nitrogen and sulphur than the coal from which it was derived;
- The hydrogen-rich extract is easily hydrocracked to petroleum-like products at a relatively high efficiency;
- The solvent is virtually 100 percent recoverable.

Potential Problems

- The pressures required to run this process are relatively high (100-400 atm);
- Extract yield is strongly dependent on coal residence time, hence there is a slow turnaround time.

Key Items for Further Investigation

Further research should be conducted to test the viability of other extractants that have a critical temperature in the desired range but with a critical pressure in a lower range.
The feasibility of combining this process with explosive coal shattering is recommended.

V. COAL PYROLYSIS INTEGRATED WITH ELECTRICAL POWER GENERATION PROCESSES

Concept Type

Class I: Revival of previously rejected concept.

Original Developer

Lurgi-Ruhrgas.

Status

Pilot tested.

Problems Addressed

Recent coal liquefaction technologies have emphasized maximum production of hydrocarbon liquids. This has led to large, high pressure reactors using an expensive catalyst and large amounts of hydrogen. The proposed technology would reduce the cost of reactors and the amount of catalyst and hydrogen required to produce liquid fuels.

Description of the Proposed System

In the proposed system fast pyrolysis at low pressure is used to separate coal into a carbon-rich component (char) and a hydrogen-rich (hydrocarbon) component. The char is fed directly to a steamboiler and the hydrocarbon stream is upgraded at a conventional refinery.

Key Features of the Proposed System

- Fast pyrolysis is a fairly well developed technology (e.g. Lurgi-Ruhrgas, Occidental Flash Pyrolysis) using low pressure (0.1-0.3 MPa), high throughput (1-3 sec residence time) reactors that should be much cheaper than those of other technologies such as H-Coal (19 MPa, 0.5-1.2 hr).
Collocation with a coal fired power plant will allow use of existing coal transport/storage infrastructure.

Hydrogen production facilities are not required at the power plant site, reducing investment cost.

Total hydrogen consumption to produce liquid fuels would be cut by as much as half compared with other liquefaction technologies.

Liquids produced could be upgraded to high density jet fuels for military applications or high octane gasoline at modern petroleum refineries.

Key Items for Further Investigation

Determination of optimum furnace type for coal char combustion.

VI. ENHANCED HEAVY OIL/TAR SANDS PRODUCTION USING HORIZONTAL WELLS

Concept Type

Class III: New integration or new application of proven technology.

Original Developer

Oil industry.

Status

Very modest and declining development effort within the private sector exists. Drilling techniques have been proven, but no commercial applications to heavy oil or tar sands have yet been made.

Problems Addressed

Recovery of heavy oils and tar sands by thermal methods is generally very inefficient in terms of the use of energy. This in turn is a major driver of production costs. The proposed approach to heavy oil and tar sands production promises better steam/oil ratios, improved overall recovery of oil, a more rapid production profile, and significantly improved costs.
Description of Proposed System

Over the past several years improvements in drilling technology have made the drilling of horizontal wells more practical; that is, wells that start from a vertical or near-vertical position are able to turn and run into an oil reservoir horizontally rather than vertically. This type of well is promising in all phases of oil production: primary (for use in controlling coning of gas or water, or increasing of production rates), secondary, or tertiary (primarily with thermal EOR).

Under favorable conditions, horizontal wells can contact a much larger reservoir area than a vertical well, thus offering many of the advantages of mine-assisted recovery techniques without the expense of the associated tunneling.

Key Features of the Proposed System

Analysis discloses that horizontal wells are most effective in the following situations:

- In reservoirs where vertical permeability is better than horizontal permeability, such as fields where vertical fractures are suspected. Such fractures are common, for example, in Devonian shales, or geothermal fields. In cases where vertical permeability is poor, such as where shale streaks are present, horizontal well production is likely to be disappointing. Problems with vertical permeability appear to have contributed to at least one field test failure.
- In reservoirs where horizontal well length can be large compared with reservoir thickness. In such fields, a horizontal well will be exposed to a much greater area of the reservoir than would a vertical well. The result will be improved productivity.
- Where horizontal wells can be accurately placed, and successfully completed. Simulations have disclosed that deviation in the drilling of horizontal wells from their intended location can result in a decrease in sweep efficiency and production. Such sensitivity is often greater than for a
vertical well. For example, if a horizontal well could not be located correctly relative to the oil/water contact, the result would be disastrous. In addition, horizontal wells experience more severe completion problems in unconsolidated sands because the holes cave in.

Potential Problems

Although simulations of thermal recovery with horizontal wells almost universally show potential benefits, drilling and completing such wells is not without difficulty. First, drilling costs per foot are substantially greater than for vertical wells, perhaps two to three times as much. In addition, it has often been difficult to understand why some horizontal wells produce according to expectations, and others do not, sometimes even in the same field.

Key Items for Further Investigation

Until field test results can be fully understood, simulator studies will remain interesting and hopeful rather than definitive predictions. In fact, there is probably a great deal of understanding to be gained just by extending the conventional techniques of well testing and reservoir engineering to horizontal wells.

On the basis of the criteria we have developed, horizontal wells seem one of the most promising of novel concepts in EOR and tar sands. We believe that substantial progress could be gained by addressing drilling and completion problems, as well as by funding work on extension of reservoir engineering to the horizontal well case.

VII. GENERALIZED COPROCESSING: LIGNIN/COAL/OIL COPROCESSING

Concept Type

Class IV: Major development necessary to commercialization.

ORIGINAl CONCEPT DEVELOPMENT

The RAND Corporation.
Status

Early stages of research.

Problems Addressed

Coal/oil coprocessing offers significant advantages over most other direct coal liquefaction processes in that although operating conditions are similar to conventional technologies, much less up front capital investment is required. Recent research by the United States and Canada points to potential problem areas that may affect coprocessing economics, for instance, coal/solvent incompatibilities with resulting lower product yields and sensitivities to coal and oil combinations. The proposed technology addresses the problem of coal/solvent incompatibility and severe operating conditions.

Description of Proposed System

The proposed technology makes use of two direct coal liquefaction technologies, coal/oil coprocessing and coal/lignin depolymerization,\(^3\) to form a hybrid technology called lignin/coal/oil coprocessing. As the name implies the basis for this technology is coprocessing, a well-known process, with lignin assuming the role as a solvent adjunct. Lignin is molecularly better suited as a solvent for coal than oil, but is oxygen-rich and should be used only in moderate amounts. To determine the optimal placement of lignin (or lignin-like solvent) in the system will require further research; however, there are at least three additional schemes:

- Mixed initially together with coal and oil;
- Added to the unreacted coal to increase product yield;
- Processed first with coal, then after product removal, oil added to the unreacted coal.

---

\(^3\)Process patent taken out by Mobil Oil Corporation, U.S. No. 4,409,089.
The first and second scheme probably hold most promise, though again, this would need to be confirmed empirically.

Key Features of the Proposed System

The coal/oil/lignin hybrid integrates its two constituent technologies so that their respective advantages complement each other or address the deficiencies each have separately. The potential benefits of this technology are that it:

- Provides a superior coal/solvent system;
- Moderates operating temperatures,
- Decreases sensitivity of coal and oil combinations,
- Increases product yield; and
- Reduces catalyst requirement.

Potential Problems

Anticipated concerns with this technology that will require further analysis and research to assess their importance and better understand costs are:

- Increased complexity because of introduction of lignin;
- Resource base for lignin; and
- Fate of the oxygen on lignin molecule.

Key Items for Further Investigation

Technical uncertainties surrounding this technology are great since, to our knowledge, it has not gone beyond the conceptualization stage. Suggested research agenda items include:

- Conduct laboratory-scale trials according to process schemes described;
- Determine lignin’s resource base; and
- Explore lignin-like solvents that contain less oxygen.
VIII. INTEGRATED SURFACE MINING/HIGH POWER MICROWAVE RETORTING OF OIL SHALE

Concept Type

Class III: First-time integration or new application of proven technologies.

Original Developer

The RAND Corporation.

Status

Early stages of research.

Problems Addressed

Surface oil shale production schemes have resisted major cost reduction because their costs are distributed in a number of major subsystems, none of which appeared subject to elimination or major cost improvement. Major subsystem cost areas include:

1. Shale mining and crushing;
2. Ore transport to retorting complex;
3. Retorting battery;
4. Upgrading facilities; and
5. Spent shale disposal;

The proposed technology addresses these problems by combining items 1, 3, and 5 and eliminating item 2 and probably 4.

Description of the Proposed System

Using a strip mining approach to oil shale mining, it is proposed to combine a large mining machine such as a dragline with a high powered microwave shale retort for rapid conversion of kerogen to oil. After shale ore was mined using the conventional mining machine, it would be

\(^4\)For an analysis of the feasibility of large-scale strip mining of oil shale in the Piceance Basin, see Pei and Rubenson (1982); and Rubenson and Pei (1983a, 1983b).
dumped on a conveyor and moved to an integrated hammermill for crushing. It would then be conveyed continuously to and through a microwave retort that used multiple high power transmitters to flash the kerogen contained in the marlstone to vapor and gas. The vapor fraction would be immediately distilled and moved by flexible piping out of the mine. The gas fraction would be transported by flexible piping to power a gas turbine for electricity production to contribute to the electricity needs of the overall system. Spent shale from the retort would be continuously conveyed to the back of the mine-cut to back-fill the mine.

**Key Features of the Proposed System**

The proposed approach to shale oil extraction promises some important cost advantages over current-technology surface oil shale systems:

- Shale handling as ore and spent shale is dramatically reduced. No shale leaves the mine after the initial mine face is established.
- Spent shale disposal costs are minimized along with the concomitant water consumption.
- Raw shale oil quality is expected to be much better than with any combustion retort. Oil quality should enable pipelining of crude out of the area without partial refining (upgrading) and without extensive use of pour point depressants. The oil should be acceptable as feedstock for most refineries.
- Capital costs should be dramatically reduced because of the elimination of costs associated with shale pile runoff impoundment, upgrading facilities, and lower expected costs of the retorting battery.

**Potential Problems**

Electricity cost and consumption.
IX. LOW SEVERITY COAL LIQUEFACTION USING LIQUID CLATHRATES

Concept Type

Class V: Radically new system--basic issues to be resolved.

Original Concept Development

J. L. Atwood, Ph.D., University of Alabama.

Status

Basic research in university laboratories.

Problems Addressed

Capital cost is a prime factor in boosting unit product costs for hydrocarbon liquid fuels derived from solid fossil fuels such as coal. Substantial decreases in investment costs promise to have a profound effect on reducing final product cost for coal liquids.

Many factors contribute to capital costs, including high operating pressures and temperatures. A majority of direct coal liquefaction technologies require severe operating conditions that not only contribute to high plant capital costs but strain technical capabilities. Such conditions lead to the need for thicker walled reactors, expensive materials for coal contact areas, higher maintenance costs and energy intensive steps for heating and pressurization.

This proposed technology offers a new coal chemistry that operates under very mild conditions and suggests improved plant performance.

Description of the Proposed System

The proposed technology uses a special solution called liquid clathrates to dissolve coal particles and facilitate the coal's conversion to coal liquids under mild conditions. Liquid clathrates derive their name from crystalline "clathrates." Like crystals, liquid clathrates trap certain sized molecules; in this case, solvent, catalyst and coal particles. Because clathrates are liquid, particles and solvent can freely move through the structures thus allowing for a continuous reaction.
Energy must be supplied to the system to produce coal liquids. The requirements are lower for this process than many conventional approaches. Because the spaces between clathrate-forming chemicals are fairly small, coal, catalyst, and hydrogen are brought into very close contact via chemical rather than kinetic means (heat and pressure). Liquefaction by this process has been observed with temperatures as low as 40 degrees C and hydrogen pressures of one atmosphere. Conventional operating conditions require temperatures of 425-450 degrees C and pressures of 100-140 atmospheres.

**Key Features of the Proposed System**

The economic edge of the low temperature liquid clathrate process can be summarized as follows:

- Lower energy consumption in heating and pressurization;
- Reduced capital cost of process equipment;
- Reduced corrosion and materials handling problems;
- Potential for using high ash coal as feedstock.

Reduced corrosion and materials handling problems result from lowering operating temperatures and pressures. This process may be able to accommodate high ash coal as a feedstock material, since noncoal constituents will not enter into the clathrate layer but will drop to the bottom of the reactor with unreacted coal.

**Potential Problems**

Not enough is known about this process to firmly draw the line between potential problems and areas for suggested further investigation. Nevertheless, key items important for the economic success of this technology include the robustness of the clathrates and the reaction's residence time. Apart from the air and water sensitivity of the clathrates, which can lead to their degradation, the relationship

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*By "energy," we refer to process steps that require energy inputs, such as compressing hydrogen gas.*
between coal and appropriate clathrate is not well understood. Compatible combinations of feedstock and clathrate chemicals allow the clathrates to function as long as their integrity is maintained; incompatibility risks the clathrates being consumed by the reaction and must be avoided.

**Key Items for Further Investigation**

Many chemical and technical uncertainties surround this technology. More research will be required to perform economic evaluations of this process. Suggested areas for further investigation include:

- Further pursue basic research studies with liquid clathrate chemistry;
- Determine product yields;
- Using chemical engineering methods, design efficient, continuous reactor system with attention given to moving materials in and out, e.g., fresh coal, unreacted coal, product liquids, solvent;
- Further identify and study air- and water-stable, inexpensive liquid clathrates that are unreactive with coal;
- Determine energy balances for clathrate system;
- Identify suitable catalysts that can coexist with the clathrate system or be incorporated into it; and
- Explore coal rank dependence.

X. MODIFICATION OF THE INTERNAL COMBUSTION ENGINE: ENRICHED OXYGEN SUPPLY TO THE COMBUSTION CHAMBER WITH SIMPLE MEMBRANE DEVICE

**Concept Type**

Class III: First-time integration or new application of proven technologies.
Original Developer
The RAND Corporation.

Status
Early stages of research.

Problems Addressed
Hydrocarbons produced from alternative resources require considerable and expensive upgrading to meet present transportation fuel specifications. Currently, there are no end-use devices that can make use of less refined and therefore less expensive synfuels. The proposed technology would reduce the need for high quality fuels for internal combustion engines and thus promote the use of less refined fuels.

Description of the Proposed System
In the proposed system membrane air separation technology is used to increase the oxygen concentration in combustion air by 10 to 50 percent. A membrane of either the fiber bundle or polymer sandwich type would be affixed to the intake system of the engine to operate off the intake manifold vacuum. Should pressure drop be inadequate an exhaust turbocharger could be employed to provide additional pressure. The engine fuel control system would be programmed to adjust fuel flow for oxygen concentration as well as air flow to optimize combustion.

Key Features of the Proposed System
Oxygen enrichment will increase both the rate and efficiency of the combustion process. Potential benefits include:

- In spark ignition engines the ignition advance required for complete fuel combustion will be reduced, thereby reducing fuel octane requirement.
- In diesels the ignition lag should be reduced by the more reactive oxygen atmosphere, reducing octane demand. Heavier fuels will be more easily combusted and particulate emissions will be reduced.
For both engine types hydrocarbon emissions should be reduced and fuel efficiencies should increase as a result of more complete combustion of the fuel.

The system should readily lend itself to retrofitting of the existing vehicle infrastructure.

Potential Problems

Enriched air may increase combustion temperatures enough to require new engine materials (ceramics).

Key Items for Further Investigation

- Dynomometer tests to determine engine response to varying concentrations of oxygen and octane/cetane reduction; and
- A membrane system with proper pressure drop, flow, and enrichment characteristics.
Appendix C
THEORETICAL DETERMINATION OF LIGNIN SUPPLY REQUIREMENTS

The following analysis was performed to approximate lignin supplies necessary for the conversion of coal to liquids in the lignin/coal/oil coprocessing setting. Calculations and assumptions are described below.

In 1984, 9.72 million barrels per day of petroleum were consumed in the United States as transportation fuels (62 percent of the total petroleum supply). Suppose 25 percent of petroleum requirements are to be supplied by crude produced from coprocessing liquid products, or about 888 million barrels crude per year derived from the conversion of coal/bitumen.

Coprocessing crude yields from coal and bitumen were calculated in the following manner:

- Assume on a MAF weight basis for one ton feedstock that coal constitutes 33 percent and asphaltic bitumen 67 percent.
- Assume that 2.93 bbl/ton crude can be produced per ton coal and that the conversion factor for the production of crude from bitumen is 5.53 bbl/ton.

Disregarding any synergistic effects, the coprocessing yield of crude per ton feedstock was calculated to be:

\[(2.93)(0.33) + (5.53)(0.67) = 4.7 \text{ bbl/ton feedstock} \]

---

2One barrel of coprocessing crude is considered to be equivalent to one barrel petroleum crude.
3MAF refers to moisture-ash free coal.
or, for every 0.33 tons coal, 4.7 bbl coprocessing crude can be produced.

Thus, approximately 60 tons of coal are required to produce 25 percent of all transportation fuels from the coprocessing technology given above, specifically,

\[(887.5 \times 10^6) \times (0.33/4.7) = 62.3 \text{ or } \sim 60 \text{ MM tons coal required}\]

In this country, the largest producer of lignin is also its largest consumer. The kraft and soda pulp mills consume about three million tons of lignin annually to recover chemicals and heat.\(^6\) Thus, even if all three million tons of wood-derived polymer were diverted to the production of liquid fuels by coprocessing techniques, only 5 percent weight basis of lignin to coal would be possible. Conversely, if other ratios of lignin to coal were desired the calculated required supplies of lignin would be those shown in Table C.1 and illustrated in Fig. C.1.

Table C.1

<table>
<thead>
<tr>
<th>Lignin to Coal (MAF weight percent)</th>
<th>Lignin Supplies Required (MM tons)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>3</td>
</tr>
<tr>
<td>10</td>
<td>6</td>
</tr>
<tr>
<td>20</td>
<td>12</td>
</tr>
<tr>
<td>50</td>
<td>30</td>
</tr>
</tbody>
</table>

\(^6\)Pearl (1967), p. 308.
Fig. C.1 -- Lignin supply requirements based on lignin input requirements for conversion of 60 MM tons coal to coal liquids using novel process
Appendix D
SECTION III CALCULATIONS

The retorting heat requirements for oil shale, $\Delta H_{\text{shale}}$, can be written as a sum of the individual heat requirements for the mineral fraction, $\Delta H_{\text{mineral}}$, and for the shale oil fraction, $\Delta H_{\text{oil}}$. For a mass of raw shale, $m_{\text{shale}}$,

$$\Delta H_{\text{shale}} = \left( \frac{\Delta H_{\text{mineral}} \times m_{\text{mineral}} + \Delta H_{\text{oil}} \times m_{\text{oil}}}{m_{\text{shale}}} \right) \text{ in Btu/lb raw shale}$$

The ratio of the mass of shale oil to the mass of raw shale, $m_{\text{oil}} / m_{\text{shale}}$, can be written in terms of the shale richness, $R$. That is, given the shale richness in gallons of oil per ton of raw shale, the mass fraction of oil, $X$, is determined from:

$$X = R \times \left( \frac{1}{2000} \right) \times \left( \frac{1}{7.4805} \right) \times \rho_{\text{oil}} \text{ in lb oil/lb raw shale}$$

where $\rho_{\text{oil}}$ is the density of the shale oil in lb/ft$^3$. Thus the heat requirement becomes

$$\Delta H_{\text{shale}} = \Delta H_{\text{mineral}} \times (1 - X) + \Delta H_{\text{oil}} \times X \text{ in Btu/lb raw shale}$$

Define the fraction of mineral matter that is actually heated in the retorting process to be $f$. Then the amount of energy absorbed by raw shale can be written as

$$\Delta H_{\text{shale}} = f \times \Delta H_{\text{mineral}} \times (1 - X) + \Delta H_{\text{oil}} \times X \text{ in Btu/lb raw shale}$$

Clearly as $f$ decreases, the shale retorting heat requirements are lessened.
This can be converted to units of energy per barrel; for example:

\[
\Delta H_{\text{tot}} \equiv \Delta H_{\text{shale}} \times 2000 \times (1/R) \times 42 \quad \text{in Btu/barrel}
\]

By combining the total absorbed energy, \( \Delta H_{\text{tot}} \), with the desired daily production rate, \( Z \), the total daily energy requirements can be obtained. Including the efficiency of conversion from electricity to microwaves, \( \eta_{\text{mc}} \), and the efficiency of conversion from microwaves to heat energy, \( \eta_{\text{ht}} \), the minimum amount of power needed to heat the rock in a given time is

\[
\text{Power} = \frac{\text{absorbed energy}}{(\text{time} \times \text{efficiency})}
\]

\[
= \frac{\Delta H_{\text{tot}} \times Z}{[(\eta_{\text{mc}} \times \eta_{\text{ht}})] \times T} \quad \text{in Btu/hr}
\]

where \( T \) is the number of hours in a production day.

The mass and volume flow rates of material that must be processed in a production day can be determined from

\[
\text{mass-flow-rate} = \frac{1}{R} \times 42 \times Z \quad \text{in ton/day}
\]

\[
\text{volume-flow-rate} = \frac{\text{mass-flow-rate}}{\text{rock density}}
\]

\[
= \frac{42 \times Z}{(R \times \rho_{\text{rock}})} \quad \text{in m}^3/\text{day}
\]

where the rock density, \( \rho_{\text{rock}} \), is given in tons/m\(^3\). Using the number of hours in a production day, \( T \), these become:

\[
\text{mass-flow-rate} = \frac{42 \times Z}{(R \times T)} \quad \text{in ton/hour}
\]

or

\[
\text{volume-flow-rate} = \frac{42 \times Z}{(R \times T \times \rho_{\text{rock}})} \quad \text{in m}^3/\text{hour}
\]
As an example, consider 20 gal/ton shale, with an oil specific gravity of 0.93. Then the mass fraction of oil, $X$, is

$$X = 20 \times 0.93 \times 62.4/2000 \times 7.4805$$

$$= 0.0776 \text{ lbs oil/lb shale}$$

Using the mineral and oil heat requirements as cited in the text, the shale heat requirement can be written as

$$\Delta H_{\text{shale}} = (f \times 230 \times (1 - 0.0776)) + (530 \times 0.0776)$$

$$= (f \times 212) + 41 \text{ Btu/lb raw shale}$$

Thus the heat requirements for shale range from 41 Btu/lb for $f = 0$ (no heat absorbed by the mineral matter) to 253 Btu/lb for $f = 1$ (maximum heat absorbed by the mineral matter), a sixfold difference. This means that the maximum energy benefit between completely heating the shale versus removing the kerogen without any heating of the shale is a factor of six. In reality, this factor will be less than six; the exact value is a future research issue in that it must be determined experimentally. The process as described in the text is proposed mainly on the merits of less absorbance of heat to the shale or a decrease in $f$.

In terms of energy per barrel, the heat requirements range from:

$$\Delta H_{\text{tot}} = 41 \times 2000 \times (1/20) \times 42$$

$$= 1.72 \times 10^5 \text{ Btu/barrel}$$

for $f = 0$ to

$$\Delta H_{\text{tot}} = 253 \times 2,000 \times (1/20) \times 42$$

$$= 1.06 \times 10^5 \text{ Btu/barrel}$$
for $f = 1$. Assuming $\eta_{mc} = \eta_{ht} = 0.8$, a daily production rate ($Z$) of 10,000 barrels/day, and a daily production schedule ($T$) of 24 hours, these imply the peak power requirements of

\[
\text{Power}(f = 0) = 1.72 \times 10^9 / (0.8 \times 0.8 \times 24)
\]

\[
= 1.12 \times 10^8 \text{ Btu/hr}
\]

\[
= 32.8 \text{ MW}
\]

\[
\text{Power}(f = 0.5) = 6.17 \times 10^9 / (0.8 \times 0.8 \times 24)
\]

\[
= 4.02 \times 10^8 \text{ Btu/hr}
\]

\[
= 118 \text{ MW}
\]

\[
\text{Power}(f = 1) = 1.06 \times 10^{10} / (0.8 \times 0.8 \times 24)
\]

\[
= 6.90 \times 10^8 \text{ Btu/hr}
\]

\[
= 203 \text{ MW}
\]

Recall that the above calculations pertain to a particular grade of shale, R. Other grades, however, can be analyzed similarly.
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