research to attain the goal of making liquid fuels from coal competitive with those from oil.

The major objectives of the coal liquefaction program are as follows:

- Develop a data base that industry can use to commercialize coal liquefaction technology when needed.
- Direct Liquefaction: Develop improved lower-cost process options that provide higher liquid yields and improved product quality at lower capital cost.
- Indirect Liquefaction: Develop improved technology to product liquid hydrocarbon or alcohol fuels from coal-derived synthesis gas through more efficient, cost-effective processes.

DOE's current coal liquefaction program with research areas is outlined in Figure 3-1. As shown in the figure, the three main program elements are direct liquefaction, indirect liquefaction, and support studies/engineering evaluations. The research areas of interest within each main program element are also shown in the figure.

The objectives of the current program for each research area within each main program element are described further in Figures 3-2, 3-3, and 3-4 for direct liquefaction, indirect liquefaction, and support studies, respectively.

3.8.2 Advanced Research and Technology Development Program

The Advanced Research and Technology Development (AR&TD) Program is a key coal activity within the Office of Fossil Energy (OFE) of DOE. Focused on the fundamental chemistry, physics, and engineering of coal-based processes and on the materials and devices that must be developed to bring such processes to fruition, the AR&TD Program fulfills the needs of basic and applied research within the scope and definition of overall OFE goals. The AR&TD Program is complementary to the line program in coal liquefaction in the OFE. Whereas the line program is oriented toward device and process development, the AR&TD Program is science

Figure 3-1. U.S. Department of Energy Coal Liquefaction Program

The Current Program with Research Areas

Direct Liquefaction	Indirect Liquefaction	Support Studies/Engineering Evaluations
o Advanced Multi-Staged Processing	o Synthesis Gas Conversion to Methanol	o Instruments/Components Research
o Coprocessing	o Synthesis Gas Conversion to Liquid Hydrocarbon Fuels	o Fundamental Research Support
o Novel Concept Research	o Synthesis Gas Conversion to Alcohol Fuels	o Liquefaction Technology Data Base
o Generic Research	o Light Hydrocarbon Gas Conversion	
	o Product Upgrading	
	o Reactor System Hydrodynamics and PCT Properties	
	o Biological Processing	

Figure 3-2. Objectives of the Current DOE Program in Direct Liquefaction

- o Advanced Multi-Stage Processing
  - Investigate and prove the technical feasibility of advanced multi-stage coal liquefaction concepts having significant potential for state-of-the-art advancements.

- o Coprocessing
  - Evaluate and develop transitional technology having potential for near-term production of coal liquids using, to a large extent, existing petroleum refining facilities and technology.

- o Novel Concept Research
  - Identify and evaluate truly innovative concepts for enhanced direct coal Liquefaction, such as concepts which
    - Operate at substantially lower pressures
    - Operate at low temperatures
    - Reduce or eliminate solvent recycle.

- o Generic Research
  - Perform generic research with potential for widespread applicability to the coal liquefaction technology data base.

Figure 3-3. Objectives of the Current DOE Program in Indirect Liquefaction

- Synthesis Gas Conversion to Methanol
  - Prove the technical feasibility of liquid-phase technology for methanol production at the PDU scale.
- Synthesis Gas Conversion to Liquid Hydrocarbon Fuels
  - Identify new or modified catalysts or other innovative approaches which can simultaneously improve conversion activity and suppress adverse yields of wax and light hydrocarbon gases.
- Synthesis Gas Conversion to Alcohol Fuels
  - Conduct research on innovative homogeneous and/or heterogeneous catalysts for production of ethanol or higher-molecular-weight fuel alcohols at moderate pressures and temperatures.
- Light Hydrocarbon Gas Conversion
  - Investigate novel approaches for the direct conversion of light paraffinic hydrocarbon gases to alcohol or hydrocarbon liquid fuels.
- Product Upgrading
  - Evaluate alternative techniques for efficient upgrading of products from conventional and advanced synthesis gas conversion processes.
- Reactor System Hydrodynamics and PCT Properties
  - Develop a fundamental technical data base on the hydrodynamics of synthesis gas conversion in two- and three-phase systems and on the system characteristics which affect hydrodynamics and transfer properties.
- Biological Processing
  - Investigate the potential for use of bioorganisms for the selective and efficient conversion of synthesis gas to liquid fuels at mild conditions.

Figure 3-4. Objectives of the Current DOE Program in Support Studies/Engineering Evaluations

- o Instruments/Components Research
  - Provide adequate instrument and component technology support to the overall liquefaction program through
    - = Identifying critical needs/assessing specific deficiencies
    - = Developing project requirements
    - = Performing research and development
    - = Facilitating information availability
- o Fundamental Research Support
  - Perform fundamental research to enhance understanding of process chemistry, to improve catalyst effectiveness, and to develop guidance for future research.
- o Liquefaction Technology Data Base
  - Develop a computerized information system whose contents include
    - = Liquefaction process data
    - = Economic evaluation models
    - = Process simulation models
    - = Supporting studies
      - -- Upgrading data
      - -- Pyrolysis assay data
    - = Process cost data
    - = Feedstock characteristics
    - = Bibliographic search capability

oriented, sponsoring fundamental investigations. Project activity in coal liquefaction is focused on fundamental studies where significant efforts are directed to the determination of physical, chemical, and thermodynamic properties; to research in biological coal beneficiation and solubilization; and to the enhancement of catalyst activity/selectivity.

The objectives of the AR&TD coal liquefaction subprogram are to:

- Undertake fundamental studies to improve understanding of coal liquefaction, reactions of synthesis gas, effects of mineral matter in coal, and properties of coal liquids
- Explore new and evolving concepts in coal liquefaction, such as biological conversion and novel catalytic systems that offer the potential for substantially improved performance compared to present systems.

The liquefaction subprogram is based on the generic research needs described above: fundamental studies; novel concepts; physical, chemical, and thermodynamic (PCT) properties; and advanced catalysts. Figure 3-5 identifies specific technical subjects under investigation within each of the primary research areas.

3.8.3 Pittsburgh Energy Technology Center Program

DOE's Pittsburgh Energy Technology Center (PETC) sponsors a significant research program in coal liquefaction and participates in all aspects of DOE's program. The first site visit conducted by the COLIRN expert panel was to PETC to learn the details of PETC's program. The main elements of PETC's program are outlined in Figure 3-6.

Figure 3-5. Research Areas of DOE's Advanced Research and Technology Development Program in Coal Liquefaction

Fundamental Studies	o Reaction Chemistry o Kinetics & Mechanisms o Coal Structure o Sample Bank
Novel Concepts	o Indirect Liquefaction o Biological Approaches o Mild Liquefaction o H_2O-H_2S-CO Chemistry
PCT Properties	o Defined Systems o Undefined Systems o Characterization, Methodology Data Correlation
Advanced Catalysts	o Improved Activity o Improved Selectivity o Anchored Homogeneous o Novel Approaches

Figure 3-6. Pittsburgh Energy Technology Center In-House R&D Program in Coal Liquefaction

DIRECT LIQUEFACTION	INDIRECT LIQUEFACTION	ADVANCED RESEARCH
o Coprocessing	o C_1 chemistry for liquid fuels (oxyhydrochlorination)	o Mechanism of direct coal liquefaction
o Low solvent-to-coal process	o Slurry-phase Fischer-Tropsch catalysis	o Properties of coal, treated coals, and coal slurries
o Catalytic upgrading	o Fischer-Tropsch reactor modeling	o Fundamental studies of coprocessing
		o Bioconversion of coal
		o Structural definition of synthetic fuels
		o C_1 chemistry for liquid fuels (low temperature methanol synthesis)

3.9 OTHER CONCLUSIONS AND RECOMMENDATIONS

3.9.1 Opinions and Comments

In general, and for each liquefaction technology, most panel members expressed the opinion that the processes that will eventually be commercialized are not those currently under development. In the words of one panel member, "we haven't yet invented the chemistry that will ultimately be commercialized." This thinking is reflected in the highest-priority recommendations, many of which are for fundamental or applied research that will lead to new processes.

On the other hand, the panel recognized that DOE must continue to develop the current processes as a major short-term objective. Process development units (PDUs), although expensive to operate, are necessary to perform this function as proof-of-concept units (POCs). They also serve to test equipment and instrumentation, and provide information for economic evaluations. Most important, these large units are necessary to maintain preparedness for commercialization. Thus, the highest-priority recommendations are relatively balanced between fundamental research and process development/applied research.

Much of the recommended fundamental research is applicable to more than one technology. A prime example is research on coal structure, which applies equally to direct liquefaction, pyrolysis, coprocessing, and bioliquefaction. The list of recommendations contains several other research areas that cut across technology boundaries. Nevertheless, funding for these investigations is included in the budget for a particular technology, and understanding this, the panel has placed each fundamental research recommendation into one of the technologies to which it applies. The reader should understand that many of these fundamental programs should not be limited to the technology category in which they have been placed. Also, fundamental programs may be divided into two areas: supportive (or evolutionary) research, which provides basic

information for processes under development, and explorative (or revolutionary) research, which lays the foundation for new and better processes.

Opinions expressed by individual panel members at the final meeting in McLean, Virginia, on July 13-14, 1988, that disagreed with the consensus of the panel are as follows:

- The area of alternative liquefaction chemistries is not adequately represented in this document because these chemistries are not in the current processing schemes and research on them is not being funded by DOE.
- The DOE liquefaction program lacks the means to test new basic findings at the next scale of development. Without such a capability, research in fundamental areas is dead-ended and will be unable to modify significantly the current processes under development.
- Commercialization of pyrolysis depends on utilization of the char. It is a mistake to fund pyrolysis under liquefaction because the liquefaction community will not address the char utilization problem.
- Direct conversion of methane should be included among the high-priority recommendations because of its potential impact on fuels production.

3.9.2 DOE Procedures and Policies

The panel offered and received recommendations that relate to DOE procedures and policies. These recommendations, if implemented, could have a profound effect on the liquefaction program.

A major concern is that the DOE solicitation procedure channels research and thereby stifles new ideas. This results from the RFP procedure that request research in areas specified by DOE. New ideas in liquefaction, or which can be applied to liquefaction but which are not anticipated by the RFP, would be considered as not responsive. Instead, many of the proposals are for "me too" research in the belief that such work will be funded.

Therefore, the panel recommended that DOE increase funding for unsolicited proposals to encourage and fund research on new liquefaction chemistries.

Other recommendations concerning DOE procedures are:

- DOE should establish standardized procedures to evaluate new chemistries and process concepts in order to weed out programs of limited potential and make the best use of the available funds in programs of high potential.
- Establish a standardized procedure for communication or linking of basic and applied/process development research so that each understands the significant findings or needs of the other. This will speed the application of basic research to process development.
- One panel member recommended that universities should participate in each large development program, such as at LaPorte or Wilsonville. The universities could provide support in such areas as microautoclave tests, catalyst screening, and analyses. In this way, the university becomes familiar with the development program, and graduate students get first-hand experience in industrial programs.

The Wilsonville PDU is the largest-scale unit in operation in the direct liquefaction program. It is the unit that tests the best process(es) under development, and it does so in continuous-flow, integrated, steady-state runs of several weeks duration, and at conditions most closely approximating commercial operation. A concern was voiced about the lack of accessibility of many contractors to fundamental information and materials from this program. There was also an underlying feeling that more information is obtainable than is now being generated at Wilsonville. Consequently, the following recommendations were made to more closely coordinate the laboratory programs with the Wilsonville operation:

- Establish a Wilsonville data and sample bank from which DOE contractors can obtain materials produced at PDU scale and at well-documented conditions. The quantities produced at Wilsonville are sufficiently large to supply several contractors with the identical materials, if necessary,

eliminating many of the problems associated with interpreting results from laboratories that use different solvents, many of which are not representative of streams produced during liquefaction.

- This data/sample bank should be administered by an organization other than the Wilsonville operations, so that its sole responsibility will be the collecting of samples, documenting the run conditions that produced the samples, disseminating information regarding the samples available, and responding to requests for samples.

- More extensive analyses should be performed on the Wilsonville streams (product and internal) to obtain more fundamental understanding of what is happening in the direct liquefaction process. These analyses should be performed by a group with strong organic chemistry expertise.

3.10 PEER REVIEWERS' COMMENTS

A draft of this report was sent to ten outside reviewers who were chosen for their extensive experience in coal-liquefaction research and development. The following people served as peer reviewers of this report:

1. Mr. Seymour Alpert, Electric Power Research institute
2. Dr. Raymond Anderson, National Institute for Petroleum and Energy Research
3. Dr. David Gray, MITRE Corporation
4. Dr. Gerald Huffman, University of Kentucky
5. Dr. Alex Mills, University of Delaware
6. Mr. Eric Reichl, Consultant
7. Dr. George Roberts, Air Products and Chemicals
8. Dr. David Schmalzer, Argonne National Laboratory
9. Dr. Howard Stephens, Sandia National Laboratory
10. Dr. Duayne Whitehurst, Mobil Oil

In most instances peer reviewers' comments were incorporated within this report, particularly those which dealt with corrections or specific changes. Opposing and supporting viewpoints, and comments of a general nature are included in Appendix F. Numerous comments were received from the reviewers that this report provides a comprehensive and authoritative review of the status of coal liquefaction science and technology. The report was also considered to be generally well written by knowledgeable individuals. The choice of panel members was considered excellent and ensured that the total status of coal liquefaction technologies would be exhaustively reviewed. The reviewers also thought that the major advances of recent years are described in a clear manner, including the reasoning underlying individual developments. By and large, the reviewers expressed their agreement with the panel's recommendations and priorities.

The most important <u>opposing</u> comments are abstracted and summarized in the next sections, with replies by the principal investigator. Some of these opposing views concern statements made in the review chapters 4-9.

3.10.1 General (Opposing) Comments

a. There are fundamental problems in the DOE program that virtually assure the failure of basic research finding their way into process development. These problems include the absence of adequate resources at the process development and demonstration level. The panel, therefore, over-emphasized the need for and value of basic research given the inadequate resources provided for meaningful utilization of the products of basic research.

b. Process development and large pilot plant activities must receive greater resources than DOE has provided in recent years if there is to be any substantial likelihood of commercially deployable liquefaction technology.

c. DOE should have a few continuous-flow units in operation for process screening and process parameter studies. These units should range in scale from 0.5 to 2 tons/day to a fully integrated pilot plant of 100 to 200 tons/day that will be of a commercial process configuration.

   PI Reply: The need for screening units and scale-up facilities has been recognized by DOE. Design and construction of two PDUs will begin in 1989. A direct liquefaction PDU will have a capacity of 200 pounds of coal per day. The indirect liquefaction PDU will have a capacity of one barrel per day. This latter unit will have two independent reactor systems--one Fischer-Tropsch and the other to produce oxygenates.

d. CO/H_2 are produced more economically from natural gas, leading industry toward a concentrated R&D effort in CO/H_2 conversion. There is no need for DOE to interfere with these private-sector efforts.

e. Generally, R&D needs to be more "exploratory" and less "programmatic".

f. More importance should be given to innovative research.

g. Not enough consideration was given to measurement and control instrumentation.

3.10.2 Comments re: Direct Liquefaction

a. The research recommendations do not address the need for the data required for engineering design and process scale-up. In particular there is a lack of thermodynamic data. The panel placed too much emphasis on kinetic data.

b. Even if the (current) catalytic reactors were eliminated, the cost of product would not drop very much. To make further advances implies the discovery of some new approach which might allow operation in the 250-500 psig range.

c. An additional need for the operation of an integrated pilot plant is the development of meaningful environmental, safety, and health information.

d. The statement on Page 4-50, paragraph 3, asserts that the U.S. liquefaction processes use high surface-area supported catalysts operating at lower pressure than European (e.g., German) developers. This is inconsistent with the fact that processes developed in the 1970's included SRC-II, which used native coal minerals as catalyst and SRC-I, which was a thermal process.

PI reply: The statement referred to recent process developments. In the 1980's all liquefaction processes under development in the U.S. have used commercially-produced promoted hydrotreating catalysts.

e. A clarification of the liquid yields claimed in Tables 4-9 and 4-11 is needed. Does this include coal feed to the gasifier to generate hydrogen.

PI reply: These yields reflect the current processing philosophy of squeezing maximum liquid production out of the feed to the liquefaction process. These yields do not include the feed to the gasifier. Hydrogen can be produced by one of several methods, including gasification of coal.

f. An environmental issue not addressed is the restrictions on aromatic content in gasoline, such as the 0.8 percent benzene limit in California.

g. The analytical section of Chapter 4 should contain references to XAFS studies, variable angle spinning (VASS) and depolar dephasing. Low-temperature ashing, followed by x-ray diffraction or FTIR, is not a good way to study mineral matter.

h. More work is needed on the relationship of catalyst structure to catalyst performance.

i. Homogeneous catalysts used in the past, such as $ZnCl_2$, had great activity and deserve renewed attention.

j. Section 4.2.2, although informative, should not be used to present a comprehensive view of research in the chemistry and the mechanism of liquefaction reactions.

3.10.3 Comments re: Indirect Liquefaction

a. The review is narrowly limited to the conversion of CO/H_2, an area that is highly developed and well covered by R&D in private industry. The recommendations will do little to improve the economics of indirect coal liquefaction. The two major reasons are that coal gasification represents 4/5 of the total cost and that the CO/H_2 reaction is so efficient that further improvements will be irrelevant. The important subjects in indirect liquefaction are gasification and gas clean-up.

b. A more extensive evaluation of indirect liquefaction based on sulfur-resistant catalysts for the CO/H_2 conversion step should be recommended.

c. A somewhat more uncertain reduction in the cost of synthesis gas might be found in the use of air in lieu of oxygen.

d. Significant reduction in the cost of indirect liquefaction requires lower-cost synthesis gas, which probably means higher sulfur-content and, possibly, air-blown gas.

e. Given the excellent performance, long life, and low cost of methanol catalysts, there is little economic incentive for developing homogeneous liquid-phase catalysts.

f. The report should have placed greater emphasis on oxygenates, because of the surge in their use in transportation fuels.

g. Chapter 9 does not review overseas developments in indirect liquefaction.

3.10.4 Comments re: Pyrolysis

a. Major recommendations should be "What can be done with the tar and char" and "How is the reactor to be scaled up to get this same yield as obtained in small-scale equipment."

b. The swelling or "caking" tendency of coal is increased enormously by hydrogen, which simply fuses coal when in a dry state. Therefore, one can not be optimistic about the potential of hydropyrolysis.

c. Catalytic hydropyrolysis may simply be a new buzzword and should really be treated as part of the wider subject of innovative catalysts for hydrogenation at lower temperature.