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Biomass Thermochemical Conversion Program 1987 Annual Report



January 1988

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Biomass Program Office

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute**



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BIOMASS THERMOCHEMICAL
CONVERSION PROGRAM
1987 ANNUAL REPORT

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Pacific Northwest Laboratory
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EXECUTIVE SUMMARY

Biomass feedstocks such as wood represent an important renewable energy resource in the United States. The Congressional Office of Technology Assessment has estimated that, with proper resource management and the development of efficient conversion processes, the potential contribution of biomass to U.S. energy demand could range as high as 17 quadrillion Btu per year--about 20% of current U.S. energy consumption. Similarly, the Energy Research Advisory Board (ERAB) has estimated a potential of approximately 10 quadrillion Btu per year.

Wood and crop residues constitute a vast majority of the biomass feedstocks available for conversion, and thermochemical processes are well suited for conversion of these materials. Thermochemical conversion processes can generate a variety of products such as gasoline hydrocarbon fuels, natural gas substitutes, or heat energy for electric power generation. The U.S. Department of Energy (DOE) is sponsoring research on biomass conversion technologies through its Biomass Thermochemical Conversion Program.

The Program is part of DOE's Biofuels and Municipal Waste Technology Division. Pacific Northwest Laboratory (PNL) has been designated the Technical Field Management Office for the Biomass Thermochemical Conversion Program with overall responsibility for the Program. This report briefly describes the Thermochemical Conversion Program structure and summarizes the activities and major accomplishments during fiscal year 1987.

PROGRAM OBJECTIVE AND RESEARCH AREAS

The objective of the Biomass Thermochemical Conversion Program is to generate a base of scientific data and conversion process information that will lead to establishment of cost-effective processes for conversion of biomass resources into clean fuels. To accomplish this objective, in fiscal year 1987 the Thermochemical Conversion Program sponsored research activities in the following four areas:

1. Liquid Hydrocarbon Fuels Technology
2. Gasification Technology
3. Direct Combustion Technology
4. Program Support Activities

In this report, an overview of the Thermochemical Conversion Program is presented. Specific research projects are then described. Major accomplishments for 1987 are summarized below and discussed in more detail in the text.

MAJOR ACCOMPLISHMENTS IN 1987

The Thermochemical Conversion Program made substantial progress in converting biomass into gasoline hydrocarbons. Progress included improvements in producing intermediate biocrude oils and improvements in upgrading the crudes to gasoline hydrocarbons. Specific accomplishments include:

- production of the first pure biocrude oil at University of Arizona using an extruder reactor
- successful hydrotreating of less stable biocrude oils in a single step process using a non-isothermal reactor
- improvement of yields and mass balances for a zeolite upgrading process.

The Thermochemical Conversion Program also made progress in the area of biomass gasification and program support. Specific highlights include:

- completion of design and initiation of assembly of a bench scale, semi-continuous reactor for gasifying high moisture biomass feedstocks
- identification of a method for catalytically eliminating tars from the gases produced by biomass gasifiers
- completion of a technoeconomic analysis of conceptual biomass to gasoline processes
- served as Operating Agent for the IEA Bioenergy Agreement in the area of conversion.

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BIOMASS THERMOCHEMICAL CONVERSION PROGRAM
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PROGRAM DESCRIPTION

INTRODUCTION

Biomass is a renewable energy resource broadly defined as sunlight which has been stored as chemical energy in plants and trees through the process of photosynthesis. As such, biomass can collect enormous amounts of solar energy over large land areas and store this energy until harvested for use. Principal biomass resources include forest wood, short rotation tree crops, herbaceous plant crops, grain, oilseed and sugar crops, crop residues, and many types of wastes.

Until the discovery of large quantities of low cost petroleum and natural gas in the early 1900's, wood supplied a significant proportion of this country's energy requirements for heat and power. Combustion of wood in various sized boilers produced steam to heat buildings, powered industrial machinery, and even powered transportation vehicles such as ships, trains and farm machinery.

Today it is estimated^(a) that biomass provides about 2.7 quadrillion Btu's of energy per year in the United States. This corresponds with about 3.6% of the annual U.S. energy demand. This energy comes primarily from the combustion of wood and other forms of biomass to provide steam and process heat. This contribution is very significant in relation to other energy resources. Nuclear power generation, for example, provides^(b) about 3.8% of the nation's energy supply.

With proper resource management and the development of efficient conversion processes, it has been estimated that biomass resources can provide an even

(a) Office of Policy, Planning, and Analysis. 1983. Energy Projections to the Year 2010. DOE/PE-0029, U.S. Department of Energy, Washington, D.C.

(b) Haggin, J., and J. H. Krieger. Chem. and Eng. News, p. 28, March 14, 1984.

greater fraction of this nation's energy supply. The Office of Technology Assessment has estimated^(c) the potential contribution of biomass to the U.S. energy supply could range as high as 17 quadrillion Btu per year, about 20% of current U.S. energy consumption. Similarly, the Energy Research Advisory Board (ERAB) has estimated^(d) that biomass could potentially supply the nation with about 10 quadrillion Btu by the year 2000. As an abundant, renewable, domestic energy resource, biomass can help the United States reduce its strategic dependence on imported oil and natural gas. In addition, biomass is the only renewable energy technology capable of addressing the need for liquid transportation fuels.^(e)

PROGRAM ORGANIZATION

For biomass to attain its full potential as a solar energy option, a technology base must be developed for converting biomass into fuels that can be easily substituted for natural gas and liquid hydrocarbons derived from petroleum. The U.S. Department of Energy (DOE) is encouraging the development of this technology base through research sponsored by its Biomass Thermochemical Conversion Program. This Program is part of DOE's Biofuels and Municipal Waste Technology Division, Office of Renewable Technologies. Pacific Northwest Laboratory (PNL)^(f) has been designated as the Technical Field Management Office for the Biomass Thermochemical Conversion Program. As such, PNL has responsibility for overall management of the Program.

The Biomass Thermochemical Conversion Program began in 1977 as one aspect of DOE's overall effort to develop renewable energy resources in the U.S. The recognized potential of biomass as a significant contributor to meet the nation's energy demand suggested the need for research into more efficient processes for converting the resource into usable energy. The relationship

(c) Office of Technology Assessment. 1980. Energy from Biological Processes. Vol. I, Biomass Resource Base, U.S. Governmental Printing Office, Washington, D.C.

(d) Solar Energy Research and Development: Federal and Private Sector Roles, Draft Report to the Solar R&D Panel of the Energy Research Advisory Board, September 2, 1982.

(e) Collins, P. Energy From Biomass: Building on a Generic Technology Base. November 27, 1984, p. 5. ANL/CNSV-TM-157.

(f) Operated for the U.S. Department of Energy by Battelle Memorial Institute.

of the Biomass Thermochemical Conversion Program to other programs within DOE's Biofuels and Municipal Waste Technology Division is shown in Figure 1.

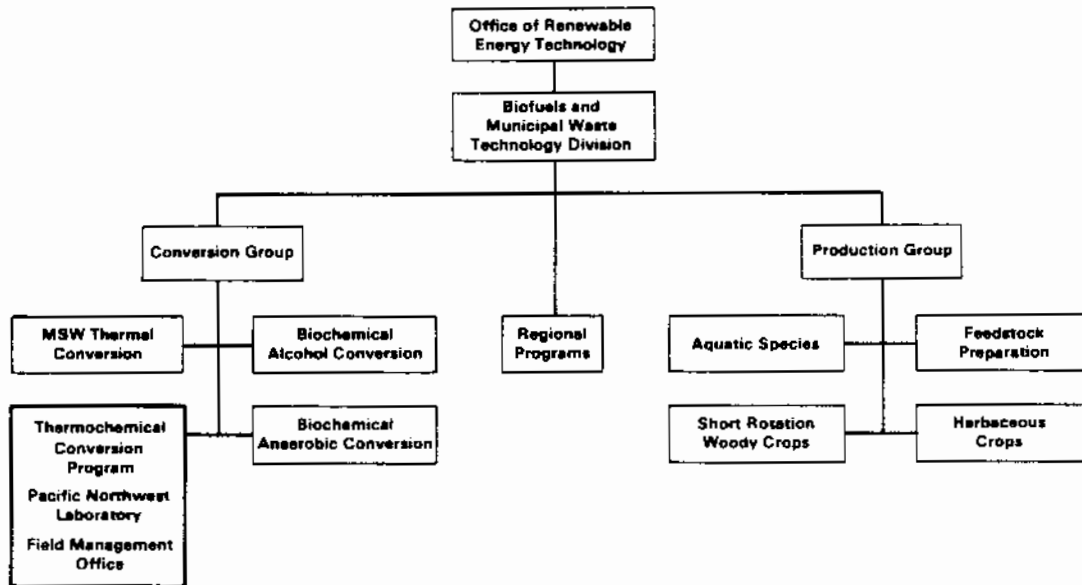


FIGURE 1. Relationship of Biomass Thermochemical Conversion Program to Biofuels and Municipal Waste Technology Division Organization

OBJECTIVE AND GOAL

The objective of the Biomass Thermochemical Conversion Program is to generate a base of scientific data and biomass conversion process information that will lead to establishment of cost-effective processes for conversion of biomass resources into clean fuels. Areas of research included in the program are gasification, liquid fuels production, and combustion of biomass. The goal of the program is to improve the data base for biomass conversion by investigating those parameters that are critical to thermal conversion processes.

RATIONALE

Thermochemical conversion processes employ elevated temperatures to convert biomass materials to more useful energy forms. Process examples include:

- liquefaction to produce biocrude oils for upgrading into gasoline and diesel fuels
- gasification to produce low-, intermediate-, or high-energy fuel gas
- gasification to produce synthesis gas for the production of high-value liquid and gaseous fuels
- combustion to produce heat, steam, electricity, direct mechanical power, and combinations of these.

Wood and crop residues comprise 96% of biomass feedstocks available for conversion to liquid and gaseous fuels. Thermochemical processes are well suited for conversion of these materials. These processes can convert 85 to 95% of the organic material in such feedstocks with high efficiency and relatively little sensitivity to variations in the feed material. As illustrated in Figure 2, thermochemical conversion processes can provide a wide spectrum of versatile products consistent with U.S. energy use patterns.

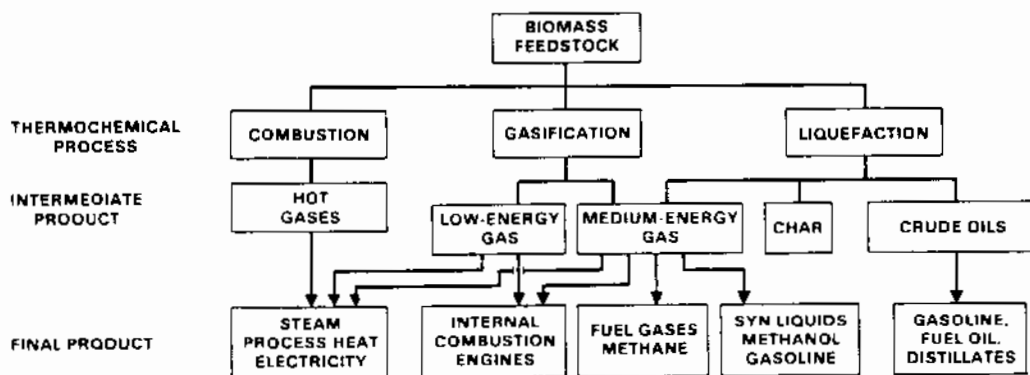


FIGURE 2. Thermochemical Conversion Can Provide a Wide Spectrum of Products

Compared to other solid fuels such as coal, biomass feedstocks have unique properties that offer potential advantages for thermochemical conversion processes. These properties include:

- high volatility - Biomass feedstocks contain a high proportion of volatile material, 70 to 90% for wood compared to 30 to 45% for typical coals. This means that a large fraction of most biomass feedstocks can be devolatilized rapidly at relatively low

temperatures. Figure 3 presents a comparison of wood and one typical coal in terms of weight loss due to devolatilization at various temperatures.

- high char reactivity - Biomass chars gasify rapidly in the presence of steam at relatively low temperatures. This property is illustrated in Figure 4, which compares the char reactivity of some biomass feedstocks with peat and coal chars at 800°C and 300 psi in the presence of steam.

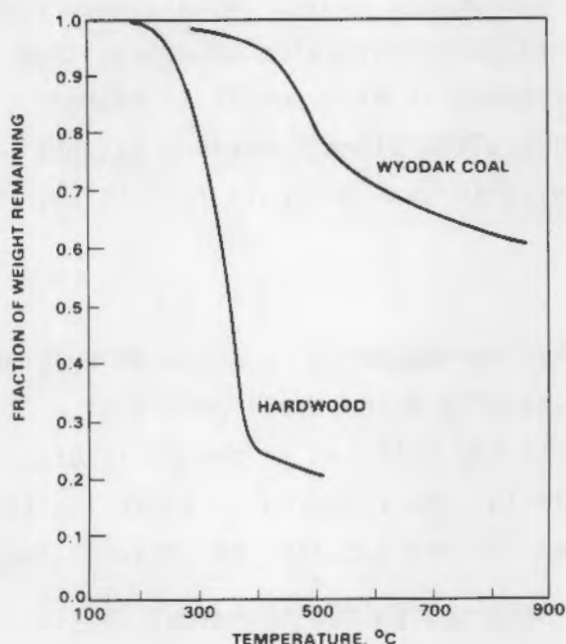


FIGURE 3. Biomass is More Volatile Than Coal (g)

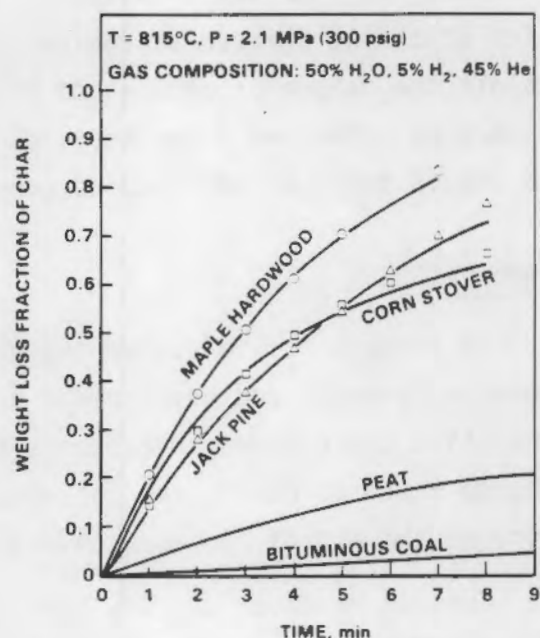


FIGURE 4. Biomass Chars Gasify Very Rapidly (h)

- low sulfur content - Typical wood feedstocks contain less than 0.2% sulfur, which greatly reduces gas cleanup costs and allows biomass to be reacted in the presence of catalysts without sulfur poisoning problems.

-
- (g) Antal, M. J. 1978. Biomass Energy Enhancement. A report to the President's Council on Environmental Quality. Princeton University, Princeton, New Jersey.
- (h) Institute of Gas Technology. 1981. Development of Hydroconversion of Biomass to Synthetic Fuels. DOE/CS/83004-4. U.S. Department of Energy, Washington, D.C.

- low ash content - Wood and most other biomass feedstocks contain less than 3.0% ash. Ash removal systems are simplified and ash disposal costs are reduced.

The combination of these four properties makes it possible to convert biomass feedstocks into gaseous and liquid fuels at faster rates and lower temperatures, and with fewer waste disposal problems, than most coals.

Thus, biomass feedstocks have some potential advantages when compared to fossil feedstocks. However, biomass is a widely distributed resource with a relatively high moisture content. This disadvantage limits the distance over which it can be shipped economically. Developing conversion processes that exploit the unique properties of biomass feedstocks will result in higher conversion rates and lower costs. This will allow biomass-derived gaseous and liquid fuels to be produced competitively in smaller-scale facilities.

APPROACH

To achieve its objective and goal, the Thermochemical Conversion Program sponsors research and development activities with high payoff potential. Innovative basic research concepts are initially selected on the basis of program research needs, the concepts' potential contribution to advancing the state-of-the-art of biomass conversion, and the availability of research funds.

Initial research usually consists of determining the technical feasibility of individual concepts in bench-scale experiments. Concepts passing the preliminary technical feasibility test are studied in continuous process research units. These small-scale research units permit further investigation of the concept under realistic conditions in a continuous, dynamic, interactive mode. This research stage allows the evaluation of variations in operating parameters in a continuous process environment and the determination of material and energy balances critical to establishing the potential of the process. Individual concepts are then combined into an integrated process. The integrated process is evaluated to verify process feasibility at a reasonable scale. This research approach is shown conceptually in Figure 5.

The interaction between the federal government and private industry in funding research on evolving technologies is shown in Figure 6. To assure

the maximum opportunity for technology transfer to the private sector, industrial interest and involvement is sought at all stages of research. The government role is primarily to explore basic research concepts and establish the technical feasibility of promising technologies. These early stages of research involve risks that are too great and lead times that are too long to be supported by industry. As the data base for evolving technologies becomes better defined, cost-sharing by the government and private industry becomes more practical.

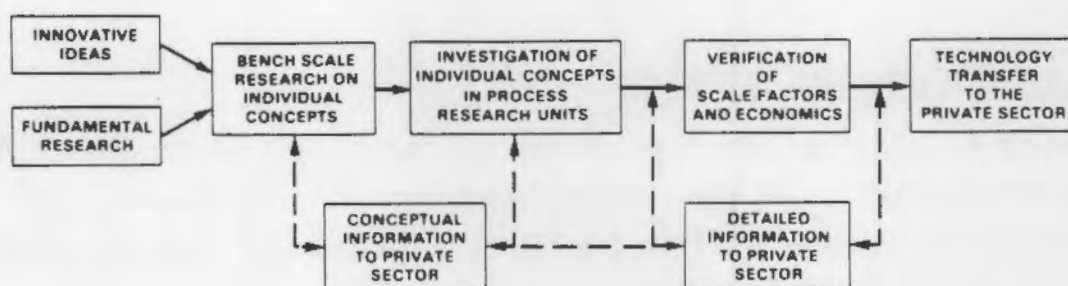


FIGURE 5. Program Approach

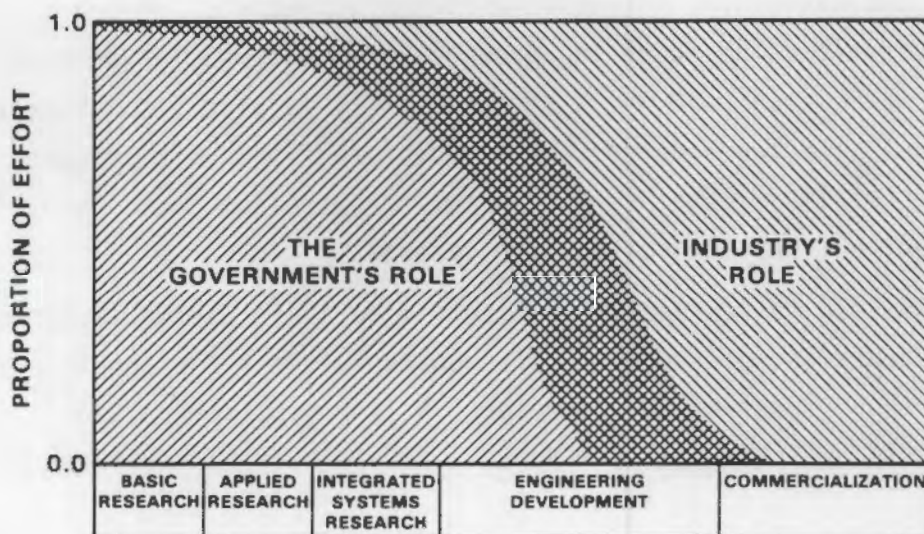


FIGURE 6. Industry Participation in the DOE Program

PROGRAM RESEARCH PROJECTS AND PROGRESS

The Biomass Thermochemical Conversion Program sponsored research in four major areas during FY87:

1. liquid hydrocarbon fuels technology
2. gasification technology
3. combustion technology
4. program support research

The following subsections present describe the specific projects conducted in each of these areas. Publications from these projects are listed in the Appendix.

LIQUID HYDROCARBON FUELS TECHNOLOGY

During 1987, the primary emphasis of the Thermochemical Conversion Program has been the production of gasoline hydrocarbon fuels from biomass. The gasoline product is made by first generating a biocrude oil and then upgrading that intermediate to form gasoline, as shown in Figure 7.

Liquid hydrocarbon fuels offer several advantages over the biomass resource itself. First, the hydrocarbon product is truly compatible with hydrocarbons from petroleum sources. The upgrading step removes oxygen from the biomass structure and chemically adjusts the elemental ratio of carbon to hydrogen of the biocrude oil. The resulting product is a mixture of hydrocarbons rich in aromatics such as toluene which are chemically identical to hydrocarbons from conventional petroleum sources. The hydrocarbon products produced in this manner are totally compatible with existing petroleum-based motor fuels and do not require separate distribution systems.

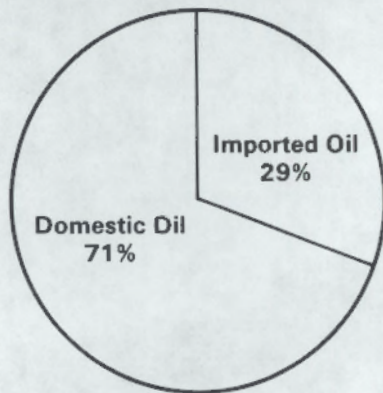
The ability to replace these types of fuels is particularly important to ensure a secure energy supply for the United States. As shown in Figure 8, the percentage of petroleum which the U.S. imports is expected⁽ⁱ⁾ to increase significantly over the next few years. Liquid hydrocarbon fuels from biomass

(i) Office of Policy, Planning, and Analysis. December 1985. National Energy Policy Plan Projections to 2010. DOE/PE-0029/3, U.S. Department of Energy, Washington, D.C.

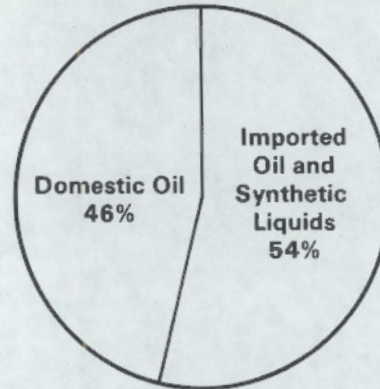


FIGURE 7. Biomass is Converted First to a Biocrude Oil
and Then Upgraded to Gasoline Hydrocarbons

1985 (30.1 Quadrillion Btu/yr)



2010 (33.2 Quadrillion Btu/yr)



Source: National Energy Policy Plan, V

FIGURE 8. Liquid Hydrocarbon Fuels from Biomass are Needed to Avoid Increased Reliance on Imported Oil

could help reduce the vulnerability of our transportation systems on fluctuations in the world market situation. (j)

In addition, the hydrocarbon fuels derived from biomass have much higher energy densities than the resource itself. The energy content of wood, for instance, is about 8500 Btu/lb, roughly half that of liquid hydrocarbon fuels. The effective bulk energy density of biomass solids on a volumetric basis is even lower if the void space between the solids is considered. A cubic foot of dry wood chips for instance, has an energy content of about 90,000 Btu. This is less than one-tenth the energy density of gasoline which contains about 928,000 Btu/ft³. A comparison of the energy densities of various fuels is shown in Figure 9. The higher energy density of liquid fuels from biomass allows the products to be transported more economically and to be more easily stored. Finally, hydrocarbon fuels can be produced from biomass with high yields and thermal efficiencies. Theoretical yields of hydrocarbon fuels range from 90 to 130 gallons per ton of dry biomass depending on what type of conceptual process is selected. While yields in actual systems will likely be lower, it is estimated that goals of 75-100 gallons/ton can realistically be achieved. Coupled with the high energy density of the liquid product, the

(j) United States Department of Energy. March 1987. Energy Security. A Report to the President of the United States. DOE/S-0057, U.S. Department of Energy, Washington, D.C.

integrated conversion process provides an effective means for potentially generating liquid transportation fuels.

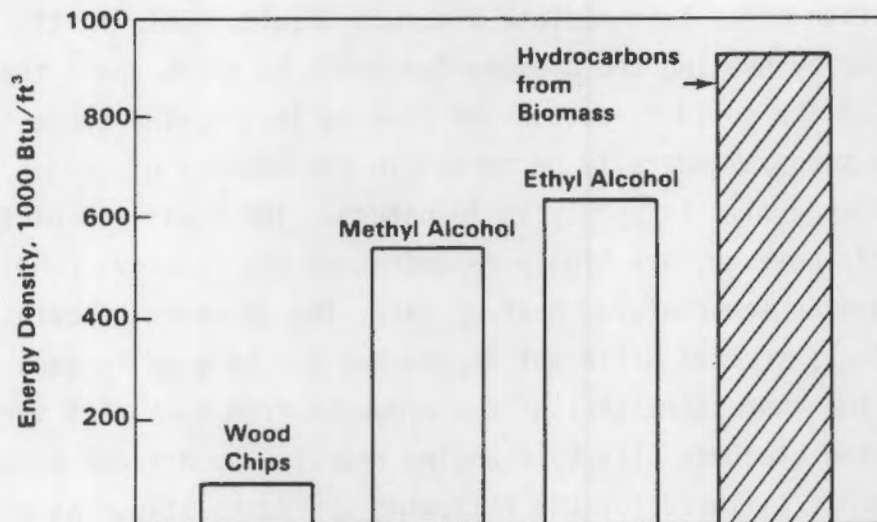


FIGURE 9. Comparison of the Energy Densities of Wood Chips and Liquid Fuels from Biomass

The Thermochemical Conversion Program is sponsoring research on methods both to convert biomass into biocrude oils and to upgrade the intermediate products into hydrocarbon fuels. The projects being funded by the Program are shown in Figure 10.

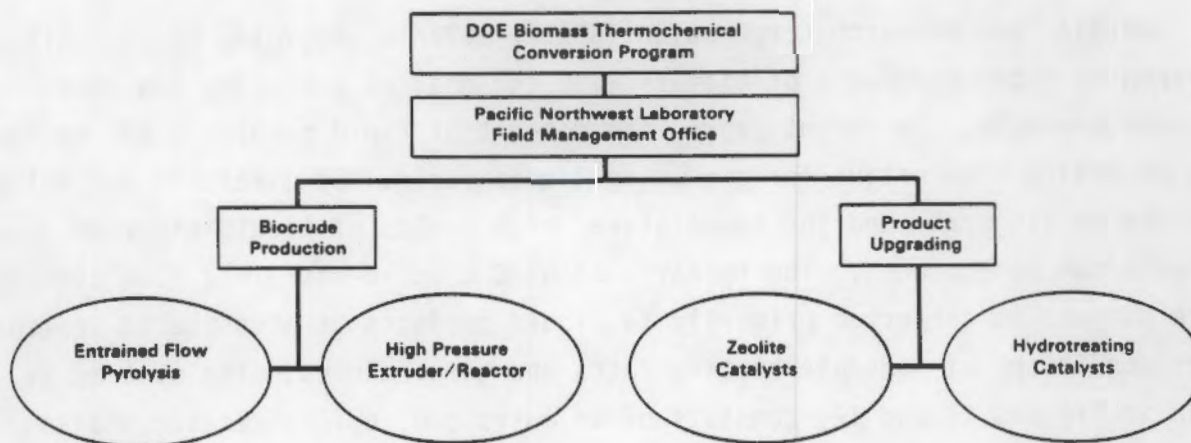


FIGURE 10. Liquid Fuels Research Projects Sponsored by the Thermochemical Conversion Program

Biocrude Production

The initial step in the conversion of biomass to hydrocarbon fuels is the production of an intermediate biocrude liquid. This initial step is accomplished by heating the biomass feedstock to break apart the internal structure of the solid feedstock and then collecting the crude liquid product. Since this step is generally performed in the absence of oxygen, the production of the biocrude oils is pyrolytic in nature. The qualities of the biocrude oils formed, however, are highly dependent on the reactive conditions including factors such as temperature, heat-up rate, the presence of catalysts, and others. Thus, several different approaches can be used to generate biocrude oils, and the characteristics of the products from each will vary. This ability to change the biocrude oils by changing reactive conditions allows the products to be somewhat tailored for the following upgrading step. As will be described in greater detail later, the biocrude oils are quite different chemically than petroleum crude oils, and different refining and upgrading procedures are necessary.

The Thermochemical Conversion Program sponsored two projects in 1987 on the production of biocrude oils. These projects included a low pressure process at Georgia Tech Research Corporation and a high pressure process at University of Arizona.

Low Pressure, Entrained Flow Reactor

Georgia Tech Research Corporation (GTRC), Atlanta, Georgia, is conducting research on rapid pyrolysis of biomass with the goal of producing low cost biocrude products. In recent years, the concept of rapid pyrolysis has emerged as a promising alternative for producing liquid fuels. By carefully controlling both the heating rate and the temperature, high yields of liquid biocrude products can be produced. The research at GTRC uses an entrained flow pyrolyzer where biomass is converted primarily to liquid products at atmospheric pressure under conditions of moderate heating rates and temperatures. The system, as shown in Figures 11 and 12, consists of an entrained, upflow reactor and an oil recovery system that allows partial on-stream fractionation of the product.

In 1987, GTRC performed experiments to further explore the effect of residence time on pyrolysis oil yield. Wood was fed into the entrained reactor at various points to alter the residence time. Maximum oil yields of about 65% on a thermal basis were produced by introducing the wood at an intermediate point. For longer residence times, the oil yields decreased due to secondary decomposition, and at shorter times the oil yields decreased due to incomplete reaction.

During 1987, GTRC also initiated research to upgrade the product oils. Screening studies of approximately twenty catalysts were performed to determine which candidates appeared to be most active in generating a gasoline product. The tests were performed in a batch autoclave apparatus. The candidates included various modified zeolite catalysts. During 1988, the most promising catalysts will be tested in a fixed catalyst bed immediately downstream from the entrained reactor. Modifications to the entrained reactor have been initiated.

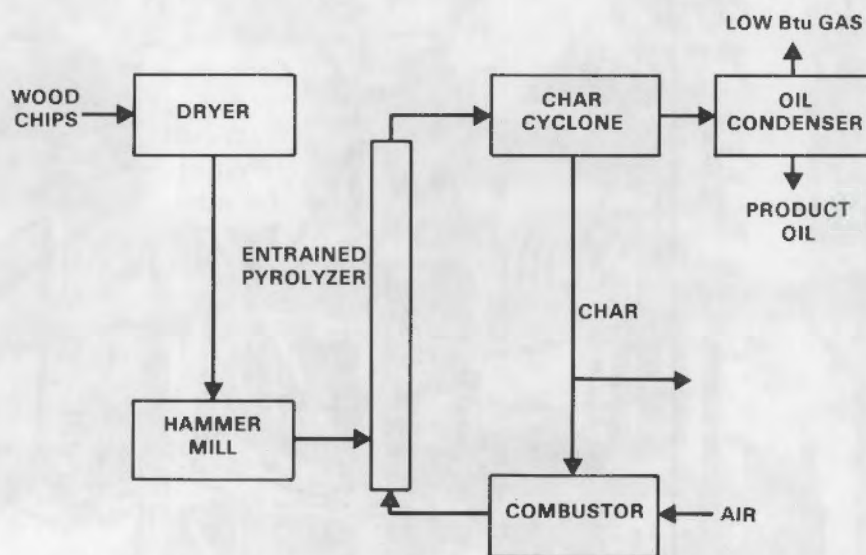


FIGURE 11. Schematic Diagram of Entrained-Flow Pyrolysis Reactor at Georgia Tech Research Corporation



FIGURE 12. View of Georgia Tech Research Corporation Pyrolysis Reactor

High Pressure Extruder Feeder Reactor

The University of Arizona, Tucson, Arizona, is conducting research on an advanced high pressure process to produce biocrude oils which have relatively low oxygen content. In this process, wood is mixed with recycled biocrude oil and injected into a pressurized reactor (3000 psi) in the presence of a carbonate catalyst and a carbon monoxide or hydrogen reducing gas. The research uses a laboratory scale extruder/static mixer reactor as shown in Figures 13 and 14. The modified extruder-feeder system is capable of handling slurries of at least 60% wood solids by weight in biocrude oil. Conventional systems, by comparison, typically cannot handle slurries containing over about 20% wood. The static mixer located in the vertical reactor provides the capability of mixing the concentrated slurries.

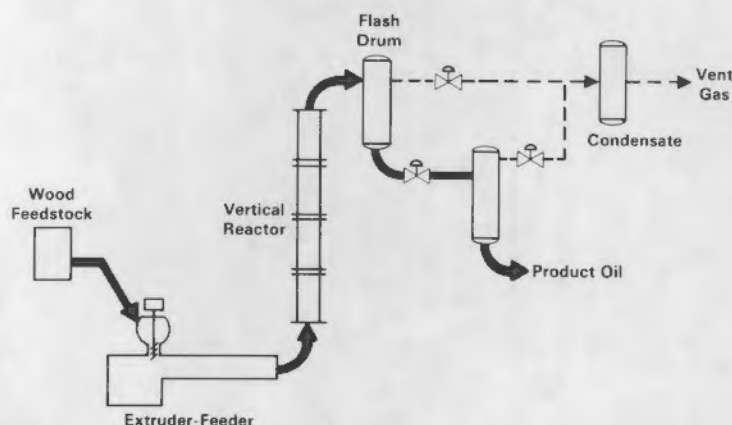


FIGURE 13. Schematic Diagram of High Pressure Liquefaction Unit at University of Arizona

During 1987, the research unit was operated to collect experimental data. The experimental apparatus operated reliably over a variety of conditions including a temperature range of 300-400°C, pressures of 1000-3000 psi, and feedstock slurry concentrations of up to 65% by weight. Mass balances exceeding 95% are now attainable on a regular basis. During 1987, the university also produced the first "all-Arizona" biocrude oil. The Arizona oil is produced from a starting slurry of about 60% wood and 40% biocrude oil produced previously at Albany, Oregon. By mixing the product oil with fresh wood and recycling, the previous product is diluted to a very low concentration. The

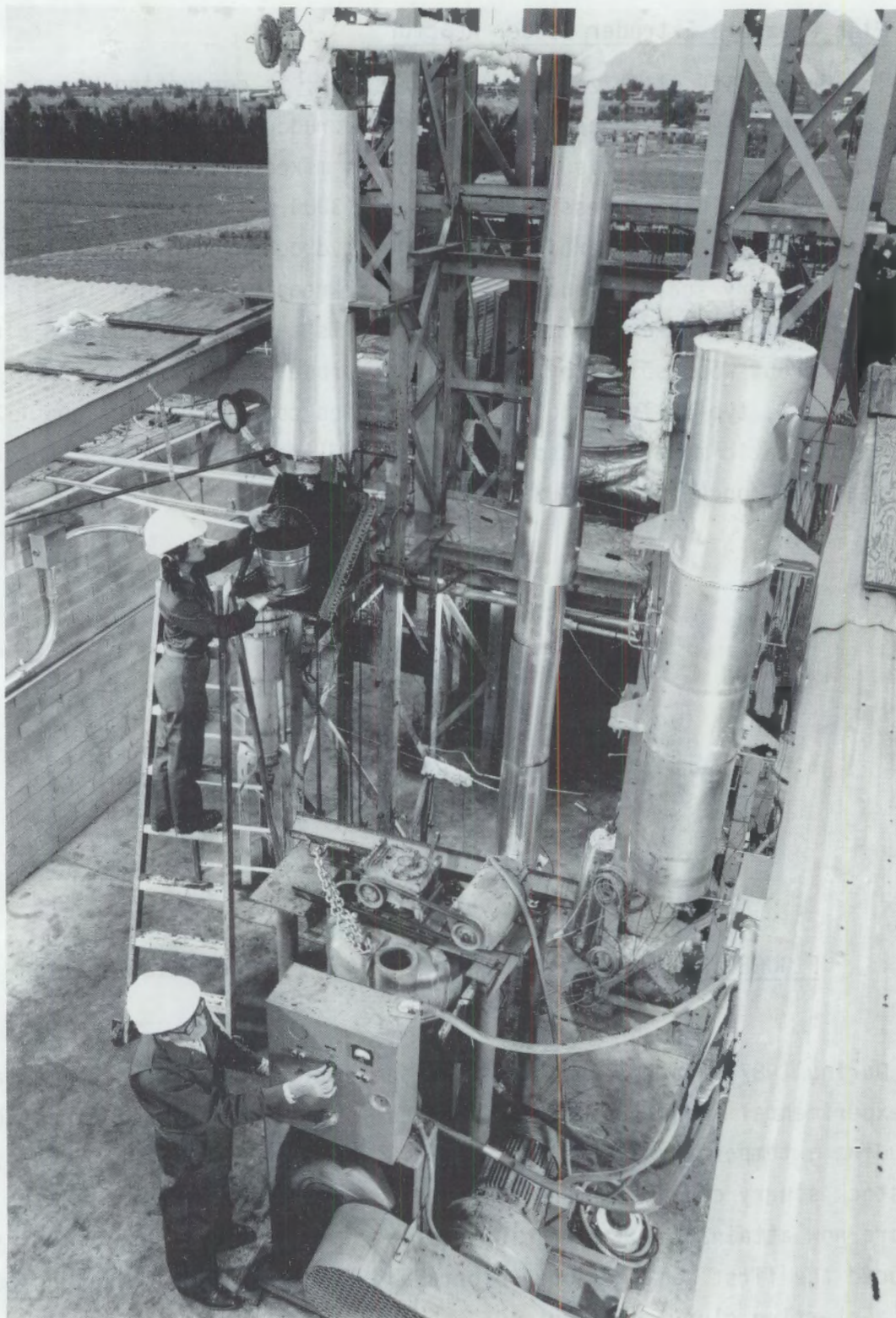


FIGURE 14. View of Extruder/Static Mixer Reactor at University of Arizona, Tucson, Arizona

university completed four successive recycles, leaving a product containing approximately 95% Arizona oil. The product is currently being analyzed. Further tests in the experimental reactor will be conducted in 1988.

Product Upgrading Research

After the biocrude oils are produced, they can then be upgraded to gasoline compatible hydrocarbons. The biocrude oils are substantially different chemically from crude petroleum, and different methods of upgrading are required.

As expected, the characteristics of the biocrude products are highly dependent on the reactive conditions in the conversion process. As shown in Table 1, the characteristics of the biocrudes produced in the low pressure and high pressure processes described above are very different. For this reason several different approaches to product upgrading may be suitable.

TABLE 1. Comparison of Biocrude Oil Products

<u>Elemental Analysis (MAF)</u>	<u>High Pressure Process (PERC-Albany TR12)</u>	<u>Low Pressure Process (Georgia Tech #11)</u>
Carbon	81.0%	59.2%
Hydrogen	10.2%	7.0%
Oxygen	8.8%	33.8%
Nitrogen	0.1%	0.1%
Sulfur	45 ppm	73 ppm
<u>Moisture and Heating Value</u>		
% H ₂ O	7.3	19.7
raw HHV	14,200 Btu/lb	7,950 Btu/lb
MAF HHV	15,300 Btu/lb	9,800 Btu/lb
<u>Other Properties</u>		
viscosity cps @ 40°C	400,000	62
density g/ml @ 23°C	1.14	1.24
pourpoint	27°C	-15°C

The Thermochemical Conversion Program is conducting research on two promising approaches to product upgrading. Pacific Northwest Laboratory is examining methods of selectively hydrotreating the crude oils to form liquid hydrocarbon products. Solar Energy Research Institute is conducting research on zeolite catalysts to form a similar product slate. These projects are described below.

Hydrotreating and Hydrodeoxygenation Research

Pacific Northwest Laboratory (PNL), Richland, Washington, is conducting research on the catalytic hydrotreating of biocrude oils. The hydrotreating step is performed in a selective manner to remove bound oxygen from the biocrude oils but retain the aromatic nature of the products. The aromatic products tend to increase the octane number of the resulting hydrocarbon fuel.

The research uses nickel/molybdenum and cobalt/molybdenum catalysts in a continuously fed, bench-scale reactor to hydrotreat the liquefaction crude oils. Initial research focused on the high pressure liquefaction oils due to their lower initial oxygen content and higher stability. Hydrotreating under conditions of 350 to 400°C at pressures of 2000 psi selectively eliminates oxygen and yields primarily hydrocarbon materials. Comparisons of key constituents and relative properties of both the crude and upgraded products are shown in Tables 2 and 3. Research indicates that the research and motor octane numbers for the hydrocarbon product are 78.1 and 72.8 respectively, about the same as straight run gasoline from petroleum sources.

The hydrotreating process is also suitable for upgrading the less stable biocrude oils produced from low pressure processes such as that at Georgia Tech Research Corporation. Previously, PNL had shown that the oils could be upgraded in a two step process by first stabilizing them at relatively low temperatures and then hydrotreating them using the same methods as those for the high pressure biocrude oils. In 1987, PNL showed the less stable oils could be upgraded in a single step using a non-isothermal reactor. The oils were fed into a reactor with a temperature gradient across the catalyst bed ranging from 270°C at the bottom to 400°C at the top. With a cobalt/molybdenum catalyst and pressures of about 2000 psig, a gasoline product was formed directly from the biocrude oil. The single step process should conserve hydrogen, minimize waste streams, and lower capital costs by eliminating the need for a second hydrotreater in a conversion facility. In 1987, PNL also redesigned their hydrotreater to allow product recycle. The recycle of partially reacted product back to the reactor should allow better control of hydrogen consumption. Modifications to the reactor will be completed in 1988.

TABLE 2. Comparison of Typical Constituents of Biomass Catalytic Liquefaction Biocrude Oil and Hydrotreated Product

<u>Biomass Crude Product</u>	<u>Refined Biomass Oil</u>
Cyclic Keytones	Cyclohexane
Unsaturated Cyclic Keytones	Alkyl-Cyclohexanes (to C-10)
Alkyl-Phenols	Benzene
Methoxy-Phenols	Toluene
Di-Phenols	Xylene
Napthols	Other Alkyl-Benzenes

TABLE 3. Comparison of Properties of Biocrude Oils and Hydrotreated Product

	<u>Biocrude Oil</u>	<u>Refined Biomass Oil</u>
Hydrogen to Carbon Ratio	1.26	1.61
Oxygen Content (% MAF Basis)	10	0.3
Density (g/cm ³)	1.14	0.83
Viscosity (cPs @35°C)	100,000	1.1
Heating Value (HHV, MAF Basis)	15,300	18,900

Zeolite Upgrading Research

Solar Energy Research Institute (SERI), Golden, Colorado, is conducting research on the use of shape-selective zeolite catalysts to upgrade biocrude oils from low pressure processes. The zeolite catalysts are efficient at removing bound oxygen from small molecules and have been used to produce gasoline-like hydrocarbons from a variety of oxygenated feedstocks such as methanol. The biocrude oil vapors are reacted on the catalyst directly downstream from the pyrolysis reactor, eliminating the need for intermediate condensation of the biocrude products. This zeolite catalyst process could potentially eliminate the need for hydrogen in the upgrading process.

Preliminary studies at SERI using mass spectroscopic techniques showed that zeolite catalysts are reactive with respect to the biomass vapors and will produce hydrocarbon products. SERI modified their ablative pyrolysis unit to include a slipstream reactor so the biocrude vapors could be directly upgraded. Using a zeolite catalyst donated by Mobil Corporation, SERI has now shown that the catalyst can effectively deoxygenate biomass vapors to produce hydrocarbons. The experimental reactor at SERI is shown in Figure 15.

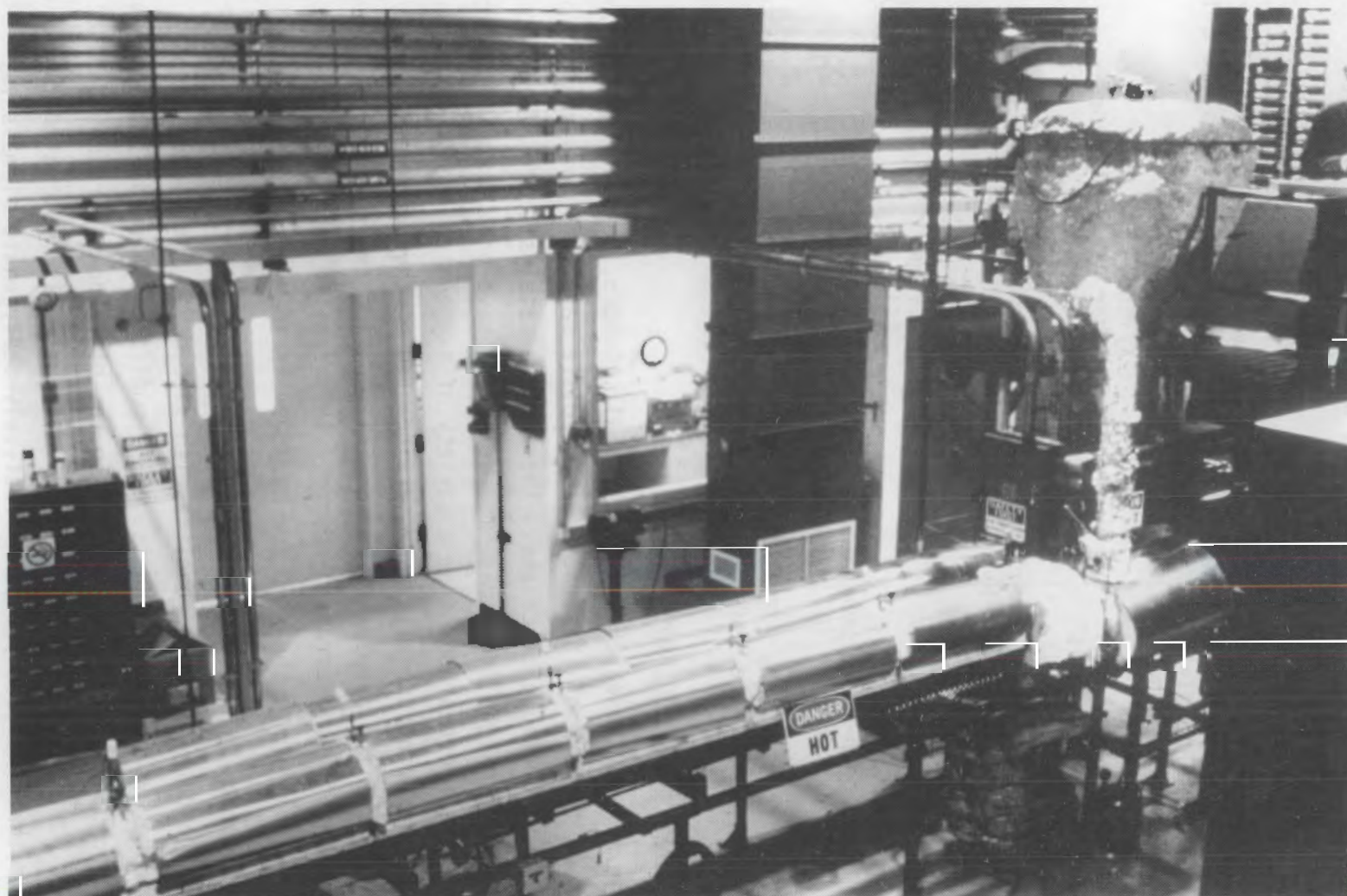


FIGURE 15. View of Reactor at Solar Energy Research Institute
Used to Produce and Upgrade Biocrude Vapors

In 1987, SERI successfully operated the experimental reactor to collect data over a variety of conditions. Mass balances for the integrated wood-to-gasoline process were determined within 95% closure. High yields of product consisting of a mixture of gasoline and partially reacted olefins were also obtained. Yields of this mixed product equivalent to approximately 50 gallons per ton of dry biomass were collected, and the production of carbon in the process was low. Research is continuing in an effort to reach a goal of about 70 gallons/ton of pure gasoline components.

SERI also completed a technoeconomic analysis of the conceptual biomass-to-gasoline process. Based on 1000 ton per day facility and a wood feedstock cost of \$34/dry ton (\$2.00/MM Btu), the production costs for a high octane gasoline were estimated to be approximately \$0.95 per gallon. The estimate assumes that the research goals for product yields can be obtained.

GASIFICATION TECHNOLOGY

Biomass resources can also be gasified to form fuel gases which can substitute for natural gas or oil. Gasification is achieved by reacting the biomass at moderately high temperatures to form a combustible gas containing large quantities of hydrogen, carbon monoxide, methane and other short-chain hydrocarbons.

Over the past few years, the primary emphasis of gasification research sponsored by the Biomass Thermochemical Conversion Program has been to exploit the natural advantages of biomass to produce medium-Btu gas. The medium-Btu gas, depending on process concept, could be used either as a fuel gas or as a synthesis gas for production of methane or liquid fuels. Medium-Btu gas is produced by volatilizing biomass using heat either from a source external to the gasifier or from the partial combustion of biomass with pure oxygen inside the reactor. The product gas typically has a heating value ranging from 300 to 600 Btu/SCF. Because of its high heating value, medium-Btu gas is very versatile.

- Medium-Btu gas can be substituted for natural gas or oil in most retrofit applications without significant derating problems.

- Medium-Btu gas produces a higher flame temperature than low-Btu gas, making it suitable for retrofitting processes where this is a critical factor.
- Medium-Btu gas has two to five times the energy density of low-Btu gas, allowing it to be transported moderate distances by pipeline at a reasonable cost.
- Medium-Btu gas can be used for the synthesis of high-value products such as pipeline gas or liquid transportation fuels.

Low-Btu fuel gases, by comparison, have heating values between 90 and 200 Btu/SCF. Low-Btu gases are produced by partially combusting the biomass resource with air. The inert nitrogen in the air dilutes the fuel gases and reduces the heating value.

Today, low Btu gasification is considered to be a commercial or near-commercial technology. Low-Btu gas can be substituted for natural gas and oil to fire boilers, subject to the following limitations:

- Low-Btu gasifiers must be close-coupled to boilers to take advantage of the high temperature of the gas leaving the gasifier.
- Burning low-Btu gas in a boiler designed for oil or natural gas will frequently result in boiler derating unless expensive modifications are made to the boiler.
- Low-Btu gas can also be used instead of gasoline and diesel oil to fuel internal combustion engines, provided that the gas is sufficiently cleaned and cooled. However, the efficiency of the engine will be reduced by about 20%.

1987 Gasification Program

The Thermochemical Conversion Program sponsored research in 1987 on two topics related to producing medium-Btu fuel gases. This research includes work on a novel low temperature process to gasify high-moisture feedstocks and work on catalysts to reduce tars from biomass gasifiers. These projects are shown in Figure 16.

In addition to the research, the Thermochemical Conversion Program completed peer review and preparation of final reports on four medium-Btu

gasification process research units (PRUs) which were funded through 1988. The final revisions to the reports are nearing completion, and they will be published in 1988. The four PRUs are described in greater detail elsewhere^(k) and include:

- an oxygen-blown, fluidized bed gasifier at Institute of Gas Technology, Chicago, IL
- a fixed-bed, oxygen-blown gasifier at SynGas, Incorporated, Golden, CO
- an indirectly heated, fluidized bed gasifier at University of Missouri, Rolla, MO
- an indirectly heated, dual fluidized bed gasifier at Battelle Columbus Laboratory, Columbus, OH.

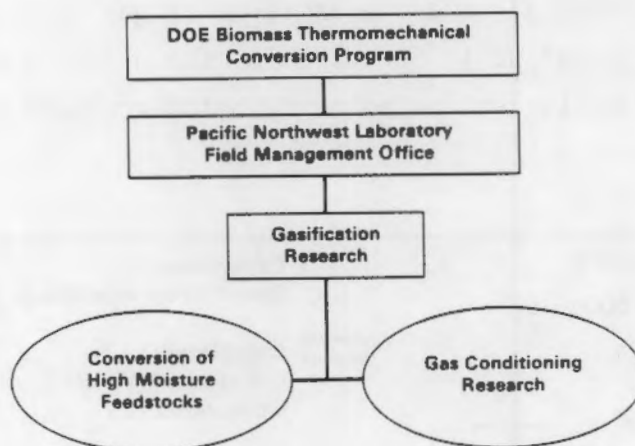


FIGURE 16. Gasification Research Projects

Gasification of High Moisture Biomass Feedstocks

Pacific Northwest Laboratory, Richland, Washington, is developing a process for the gasification of high moisture biomass feedstocks. The concept under investigation uses a commercial nickel catalyst and alkali carbonates at moderate temperatures (350-425°C) and pressures in the range of 4000 psig.

(k) Schiefelbein, G. F., D. J. Stevens, and M. A. Gerber. Biomass Thermochemical Conversion Program 1986 Annual Report. Pacific Northwest Laboratory, Richland, WA. Report No. PNL-6282.

Biomass is gasified in slurry form without air or oxygen to produce a methane rich product which has a higher heating value of up to 560 Btu/SCF (930 Btu/SCF on a CO₂ free basis).

A number of high moisture feedstocks, including grasses, agricultural residues, and aquatic plants, have been tested in the system. Feedstocks containing up to 95 wt% moisture can be gasified without drying or dewatering. Methane production rates have been achieved that are at least two orders of magnitude higher than those obtained via biological processes. It is interesting to note that the product gas is composed almost entirely of methane (≈40%), hydrogen (≈8%) and carbon dioxide (≈52%) and could therefore be upgraded to a high-Btu gas by simply removing CO₂. With proper catalyst loadings, the reaction of biomass is nearly complete at temperatures significantly below 400°C with residence times of less than 60 minutes. The low temperature operation yields substantial benefits in terms of gas quality with methane contents in the raw gas of up to 52 vol%. As shown in Figure 17, this gas composition is close to the estimated equilibrium composition for these conditions.

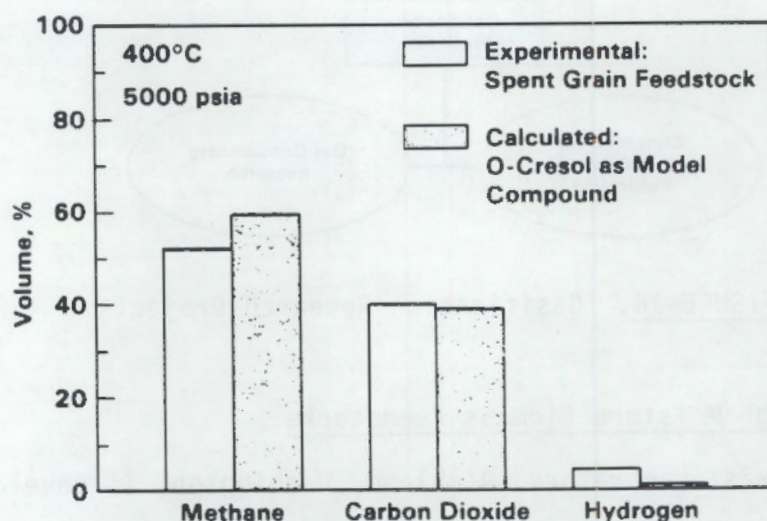


FIGURE 17. Comparison of Experimental vs. Equilibrium Gas Composition

During 1987, PNL continued research on the gasification process. Additional screening studies were performed in a batch autoclave reactor to determine the effects of catalyst loading, temperature, and reaction time on

product yields. PNL also designed and began procurement of a semi-continuous reactor system for this concept. The new reactor will allow data to be collected under steady state conditions which are not possible in the batch autoclave. A schematic diagram of the reactor is shown in Figure 18.

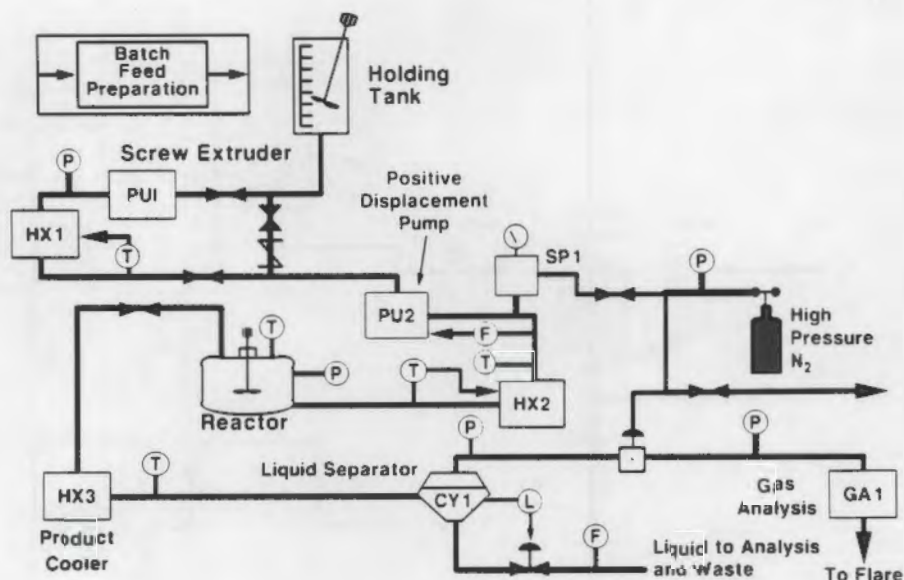


FIGURE 18. Schematic Diagram of the Semi-Continuous Reactor for Gasifying High Moisture Feedstocks

Gas Conditioning Research

Pacific Northwest Laboratory (PNL), is also conducting research on methods to condition and upgrade the quality of product gases from biomass gasifiers. The raw product gases from these units frequently contain condensible tars and oils which must be removed prior to use. The formation of tars also lowers the overall conversion efficiency to fuel gases.

During 1987, PNL conducted research on a method to catalytically eliminate the unwanted tars. Using the apparatus shown in Figure 19, researchers first generated tar-laden gases in a small fluidized bed reactor. The tar laden gas was passed through a catalyst bed downstream under steam reforming conditions. The treatment leads to the reduction of the tars with 20 or more carbon atoms to less than 100 ppm. These higher molecular weight tars are particularly difficult to convert in conventional systems. The technique

also eliminates over 90% of the tars with eight to twenty carbon atoms. The reaction of the tars not only increases process efficiency but also eliminates a potential source of problems. PNL is currently applying for a patent on this process. After the patent filing is complete, details relating to the experimental conditions will be published. Research on the process will continue in 1988.

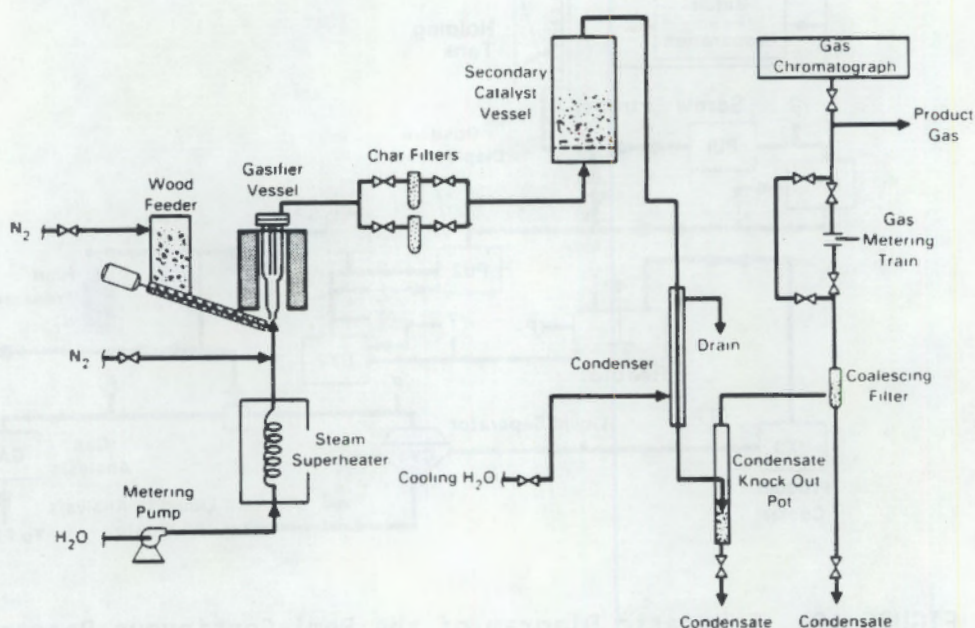


FIGURE 19. Bench-Scale Fluidized Bed Gasifier at Pacific Northwest Laboratory

DIRECT COMBUSTION TECHNOLOGY

Direct combustion of biomass feedstocks, particularly wood, is widely practiced by the private sector, especially in the forest products industry. Prior to the introduction of extensive natural gas distribution systems and cheap imported crude oil following World War II, many forest products companies utilized wood wastes from their operations to supply a large portion of their energy needs. In the current era of uncertain energy costs, there is interest in returning to self-sufficiency in fuel supplies. Many types of direct combustion equipment, such as wood-fired boilers and various types of burners, are commercially available for this purpose. Therefore, direct combustion research funded by the Biomass Thermochemical Conversion Program is focused on a unique, innovative combustion system as shown in Figure 20.

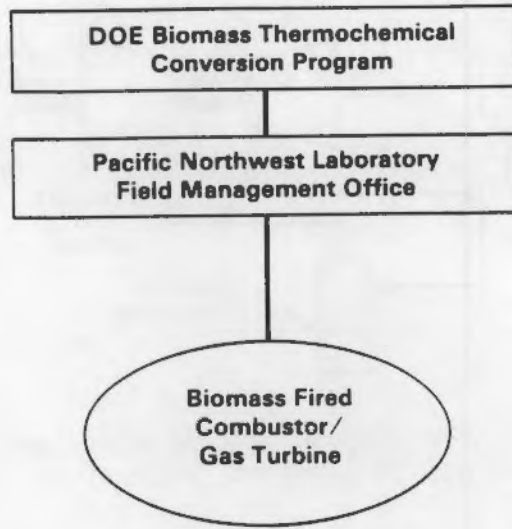


FIGURE 20. Direct Combustion Research

Combustor/Gas Turbine

Aerospace Research Corporation, Roanoke, Virginia, is conducting research on a directly-fired gas turbine generating system using wood feedstock. The objective of this project is to determine whether a gas turbine can be practically fueled with solid biomass fuels such as wood. In the Aerospace system, the integral jet engine combustor is replaced with an external combustor utilizing wood as fuel. Hot combustion gases from a pressurized wood-fired suspension burner pass through a series of cyclones to remove particulate matter and are injected directly into the gas turbine. A schematic diagram of this concept is shown in Figure 21. In previous trials using a 375-kW combustor/gas turbine, over 500 hours of operation were completed with no significant signs of erosion or corrosion to the turbine blades. Tests showed that particulate loading in the 375-kW system was very low.

Aerospace Research Corporation completed construction of a 3-MW combustor/gas turbine and electrical generator unit similar in design to the earlier system. The 3-MW unit is shown in Figure 22. The large unit was initially constructed at Roanoke, Virginia, where it underwent preliminary shakedown. The unit was then moved to Red Boiling Springs, Tennessee, for long-term testing.

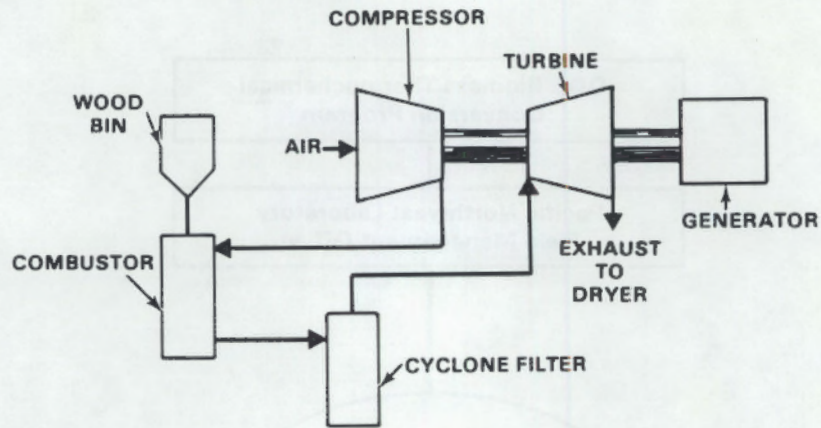


FIGURE 21. Schematic Diagram of Combustor/Gas Turbine at Aerospace Research Corporation

During 1987, the gas turbine system continued to operate in a shakedown mode. The system has accumulated over 500 hours of operation on wood and has been on-line with the TVA power grid intermittently for over 70 hours. The project has verified the performance of both the pressurized combustor and gas cleanup systems. Special procedures have been developed for bringing the system up to speed and synchronizing the generator with the TVA grid. The shakedown, however, has taken longer than expected due to problems with the unique, prototype system. The original seals in the pressurized feeder system had to be replaced with all-metal seals to allow continuous operation. Problems of turbine breakdown unrelated to the wood combustion system were also encountered.

A key remaining issue which will be examined in 1988 relates to gas particulate deposition. The particulate loadings achieved in the system (40 ppm with all particles less than 2.6 microns in diameter) are consistent with the specifications for natural gas and petroleum fuels to control erosion of the turbine components. However, constituents from the ash still produce a buildup of material on the first stage turbine blades and stators which eventually restricts the flow of gas through the turbine. At this time this problem is limiting system operation to about 1450°F. Alternative cleaning methods based on industry practices have been proposed for examination in 1988.



FIGURE 22. View of Gas Turbine and Generator System

PROGRAM SUPPORT ACTIVITIES

During FY87, the Biomass Thermochemical Conversion Program sponsored additional research activities with the goal of supporting major program elements. This research includes evaluating the technoeconomic feasibility of candidate conversion processes, and serving as the Operating Agent for cooperative conversion activities sponsored by the International Energy Agency's Bioenergy Agreement. Program support research projects are shown in Figure 23.

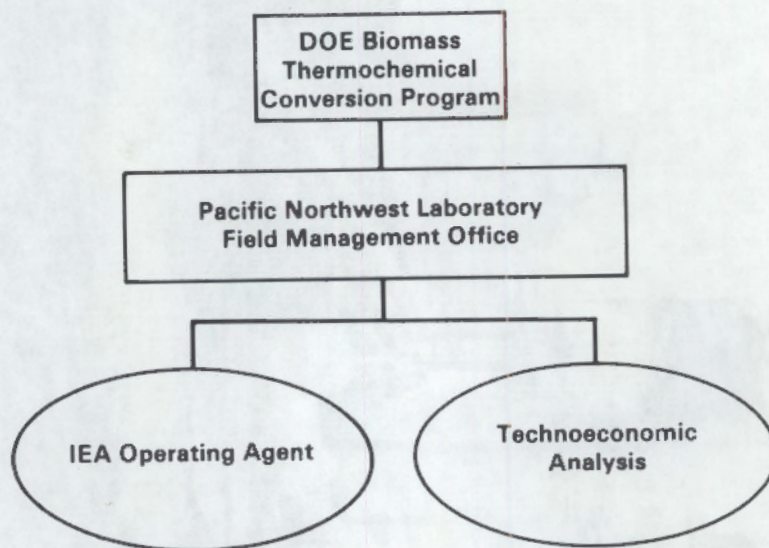


FIGURE 23. Program Support Research Projects

Technoeconomic Analysis

Science Applications International Corporation (SAIC), Arlington, Virginia, conducted engineering and economic studies to assess advanced biomass conversion concepts. During 1987, SAIC completed an analysis of the costs of producing gasoline from wood. The analysis examined two conceptual approaches for producing gasoline hydrocarbons. In the first, wood is rapidly pyrolyzed to form large quantities of biocrude vapors. The vapors are condensed, and the biocrude liquid is then hydrotreated to upgrade the intermediate to gasoline. In the second conceptual approach, the biomass is also pyrolyzed, and then the biocrude vapors are upgraded directly over zeolite catalysts without intermediate condensation. The zeolite catalysts also eliminate the need for hydrogen in the upgrading step.

The analysis of each conceptual process was performed based both on currently attainable experimental results and on research goals assumed to be attainable in the future. For a 1000 ton/day facility with wood feedstock costs of \$25 per dry ton, the gasoline production costs for the hydrotreating and zeolite processes, based on present results, were calculated to be \$1.81/gallon and \$2.21/gallon respectively. Assuming research goals can be attained, the costs are reduced to \$0.86/gallon and \$0.94/gallon for the future case. In either case, the costs are significantly less than the retail selling cost of \$1.56 per gallon projected⁽ⁱ⁾ for the year 2000.

IEA OPERATING AGENT

Pacific Northwest Laboratory (PNL), served as the Operating Agent for cooperative biomass conversion activities sponsored by the International Energy Agency's Bioenergy Agreement. Under this Agreement, twelve countries are exchanging technical information and coordinating research on topics of mutual interest. Areas of cooperation include short rotation forestry, conventional forestry, and biomass conversion. The organization of the IEA Bioenergy Agreement is shown in Figure 24.

As Operating Agent for the Biomass Conversion Annex, PNL coordinated the international efforts and administered funding for individual projects. During 1987, eleven cooperative projects were active. These eleven projects covered specific topics in the areas of thermal, biochemical and general conversion and addressed issues related to environmental concerns and the establishment of voluntary standards. The international cooperation improves the effectiveness of the research sponsored by improving the information exchange.

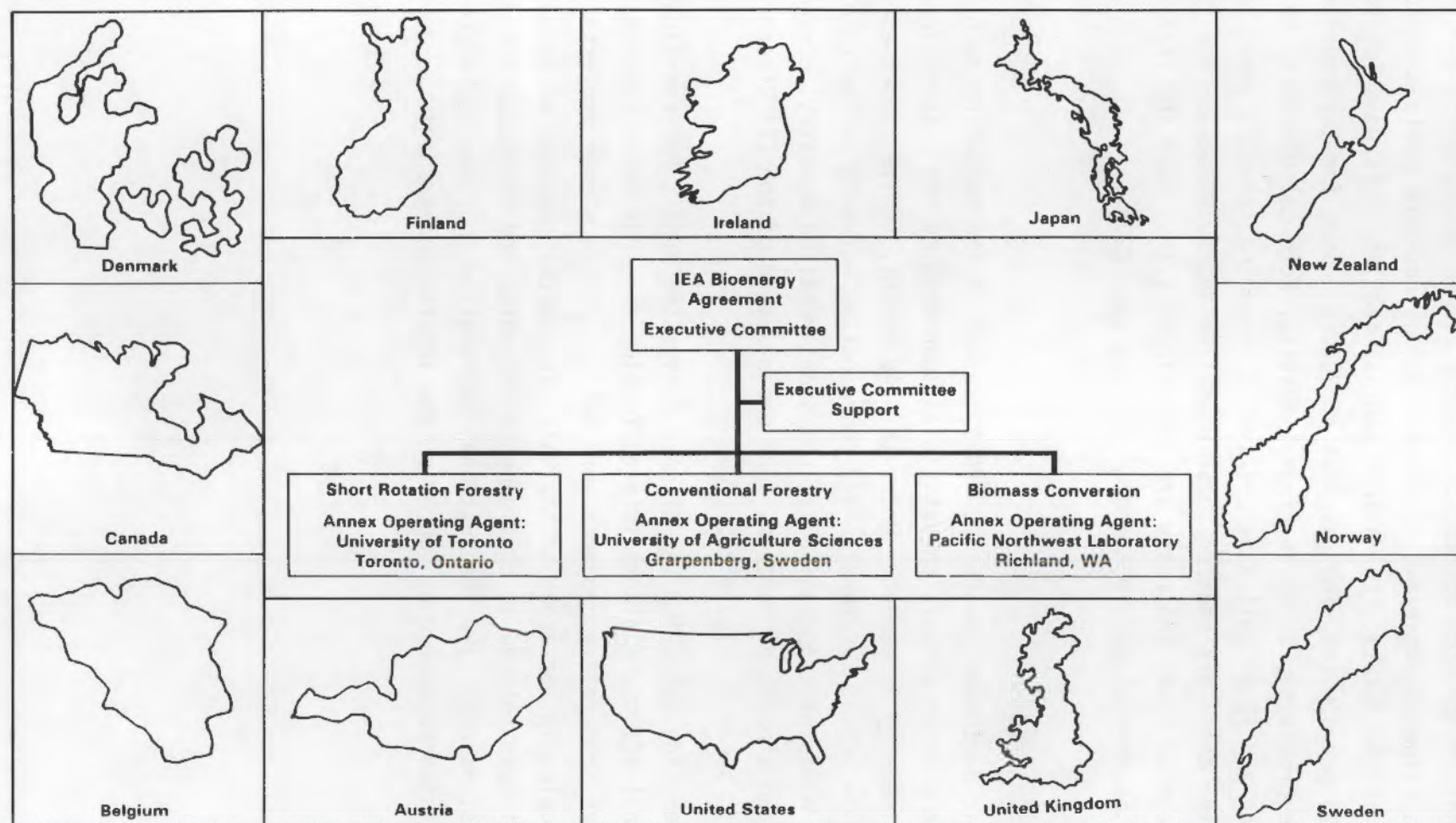


FIGURE 24. Organization of the International Energy Agency Bioenergy Agreement

APPENDIX

BIOMASS THERMOCHEMICAL CONVERSION PROGRAM

RECENT PUBLICATIONS

Recent publications from projects sponsored by the Biomass Thermochemical Program are listed below. The publications are listed in the following order:

Liquid Hydrocarbon Fuels	p. A.1 - A.4
Gasification	p. A.5 - A.9
Combustion	p. A.10
Program Support	p. A.11

LIQUID HYDROCARBON FUELS

Project Title: Production of Biocrude Oils Using an Entrained Bed Process Development Unit - Georgia Tech Research Corporation

Kovac, R. J., C. W. Gorton, D. J. O'Neil, and C. J. Newman. 1987. "Low Pressure Entrained Flow Pyrolysis of Biomass to Produce Liquid Fuels." In Proceedings of the 1987 Biomass Thermochemical Conversion Contractors' Review Meeting, pp. 23-39 CONF-8705212, National Technical Information Service, Springfield, Virginia.

Knight, J. A., C. W. Gorton, R. J. Kovac, and C. J. Newman. 1986. "Entrained Flow Pyrolysis of Biomass." In Proceedings of the 1985 Biomass Thermochemical Conversion Contractors' Meeting, pp. 99-113 CONF-8510167, National Technical Information Service, Springfield, Virginia.

Knight, J. A., C. W. Gorton, and R. J. Kovac. 1984. "Entrained Flow Pyrolysis of Biomass." In Proceedings of the 16th Biomass Thermochemical Conversion Contractors' Meeting, pp. 287-297 CONF-8405157, National Technical Information Service, Springfield, Virginia.

Gorton, C. W., and J. A. Knight. 1984. "Oil from Biomass by Entrained Flow Pyrolysis." In Proceedings of the Sixth Symposium on Biotechnology for Fuels and Chemicals, C. D. Scott, ed., Symposium No. 14, pp. 15-20. John Wiley & Sons, New York, New York.

Knight, J. A., and C. W. Gorton. 1983. "Oil Production by Entrained Flow Pyrolysis of Biomass." In Proceedings of the Third Biomass Liquefaction Experts Meeting, pp. 138-146, University of Sherbrooke, Sherbrooke, Quebec, Canada.

Project Title: Advanced Biomass Direct Liquefaction Extruder-Feeder Reactor Systems - University of Arizona

White, D. H., D. Wolf, G. Davenport, S. Mathews, M. Porter, and Y. Zhao. 1987. "An Advanced Extruder-Feeder Biomass Liquefaction Reactor System." In Proceedings of the 1987 Biomass Thermochemical Conversion Contractors' Review Meeting, pp. 5-21 CONF-8705212, National Technical Information Service, Springfield, Virginia.

White, D. H., and D. Wolf. 1987. "A Continuous Extruder-Feeder for Reactor Systems for Biomass Fuels Processing." In Proceedings of Energy from Biomass and Waste X, ed. D. L. Klass, pp. 1685-1688. Institute of Gas Technology, Chicago, Illinois.

White, D. H., D. Wolf, N. W. Andrews, D. Joshi, J. Wong, and G. Davenport. 1986. "Real-Time Microprocessor Control and Data Acquisition of Biomass Direct Liquefaction Systems." In Proceedings of the Second National Conference on Microcomputer Applications for Conservation and Renewable Energy, Donald E. Osborn, ed., University of Arizona, Tucson, Arizona,

LIQUID HYDROCARBON FUELS (cont'd)

- White, D. H., and D. Wolf. 1986. "An Advanced Extruder-Feeder Biomass Liquefaction Reactor System." In Proceedings of the 1985 Biomass Thermochemical Conversion Contractors' Meeting, pp. 115-140 CONF-8510167, National Technical Information Service, Springfield, Virginia.
- White, D. H., and D. Wolf. 1985. "An Extruder-Feeder for Reactor Systems for Biomass Fuels Processing." In Proceedings of the Fourth Technical Review Meeting, Energy from Biomass: Building on a Generic Technology Base, pp. 115-124 ANL/CNSV-TM-176, Argonne National Laboratory, Argonne, Illinois.
- White, D. H. 1984. "Development of Extruder-Feeder Biomass Liquefaction System." In Proceedings of the 16th Biomass Thermochemical Conversion Contractors' Meeting, pp. 375-397 CONF-8405157, National Technical Information Service, Springfield, Virginia.
- Khan, M. B. 1984. "Gas Dispersion in Viscous Non-Newtonian Fluids Using an Extruder-Feeder." Master of Science Thesis, University of Arizona, August 1984.
- Shlapak, N. A. 1984. "Microprocessor-Based Real-Time Process Control of Biomass Liquefaction." Master of Science Thesis, University of Arizona, August 1984.
- Homaidan, A. 1984. "Modeling of Extruder-Feeder for Biomass Concentrated Slurries." Master of Science Thesis, University of Arizona, May 1984.
- Ghoddoussi, M. G. 1983. "Solvolysis of Biomass at Elevated Temperatures." Master of Science Thesis, University of Arizona, December 1983.
- Lezzar, A. 1983. "Effects of Void Volume on the Friction and Rheology of Concentrated Slurries," Master of Science Thesis, University of Arizona, December 1983.

Project Title: Upgrading of Liquid Fuels from Biomass - Pacific Northwest Laboratory

- Elliott, D. C., and E. G. Baker. 1987. "Research on Hydrotreating Catalysts to Hydrodeoxygenate Biocrude Oils." In Proceedings of the 1987 Biomass Thermochemical Conversion Contractors' Review Meeting, pp. 81-97 CONF-8705212 National Technical Information Service, Springfield, Virginia.
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LIQUID HYDROCARBON FUELS (cont'd)

- Elliott, D. C., and E. G. Baker. 1986. "Upgrading Biomass Liquid Fuels." In Proceedings of the 1985 Biomass Thermochemical Conversion Contractors' Meeting, pp. 81-97 CONF-8510167, National Technical Information Service, Springfield, Virginia.
- Elliott, D. C., and E. G. Baker. 1985. "Hydrodeoxygenation of Wood-Derived Liquids to Produce Hydrocarbon Fuels." In Proceedings of the 20th Intersociety Energy Conversion Engineering Conference, pp. 1.586-1.592, #SAE P-164, Society of Automotive Engineers.
- Beckman, D., and D. C. Elliott. 1985. "Comparisons of the Yields and Properties of the Oil Products from Direct Thermochemical Biomass Liquefaction Processes." Canadian Journal of Chemical Engineering. 63:99-104.
- Elliott, D. C. 1984. "Bench-Scale Research in Biomass Direct Liquefaction." In Proceedings of the 16th Biomass Thermochemical Conversion Contractors' Meeting, pp. 399-422 CONF-8405157, National Technical Information Service, Springfield, Virginia.
- Elliott, D. C., and E. G. Baker. 1984. "Biomass Liquefaction Product Analysis and Upgrading." In Comptes Rendus de L'Atelier de Travail Sur la Liquefaction de la Biomasse, University of Sherbrooke, Sherbrooke, Quebec, Canada.
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- Schirmer, R. E., T. R. Pahl, and D. C. Elliott. 1984. "Analysis of a Thermochemically-Derived Wood Oil." Fuel 63:368-372.

Project Title: Upgrading of Biocrude Oils Using Zeolite Catalyst - Solar Energy Research Institute

- Diebold, J. P., and J. W. Scahill. 1987. "Upgrading Pyrolysis Vapors to Aromatic Gasoline with Zeolite Catalysis at Atmospheric Pressure." In Proceedings of the 1987 Biomass Thermochemical Conversion Contractors' Review Meeting, pp. 61-80, CONF-8705212, National Technical Information Service, Springfield, Virginia.
- Diebold, J. P., H. L. Chum, R. J. Evans, T. A. Milne, T. B. Reed, and J. W. Scahill. 1987. "Low Pressure Upgrading of Primary Pyrolysis Oils from Biomass and Organic Wastes." In Proceedings of Energy from Biomass and Wastes X, ed. D. L. Klass, pp. 801-830. Institute of Gas Technology, Chicago, Illinois.

LIQUID HYDROCARBON FUELS (cont'd)

- Evans, R. J., and T. A. Milne. 1986. Fundamental Pyrolysis Studies, Final Report 1 October 1980 - 30 December 1985. SERI/PR-234-3026, Solar Energy Research Institute, Golden, Colorado.
- Evans, R. J., T. A. Milne, and M. N. Soltys. 1986. "Direct Mass-Spectrometric Studies of the Pyrolysis of Carbonaceous Fuels. III. Primary Pyrolysis of Lignins." J. Anal. Appl. Pyrol. 9:207-236.
- Diebold, J. P., J. W. Scahill, and R. J. Evans. 1986. Entrained-Flow, Fast Ablative Pyrolysis of Biomass, Annual Report, 1 December 1984 - 31 December 1985. SERI/PR-234-3012, Solar Energy Research Institute, Golden, Colorado.
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- Diebold, J. P., J. W. Scahill, and R. J. Evans. 1986. "The Thermal and Catalytic Upgrading of Oxygenated, Primary Biomass Pyrolysis Oil Vapors." In Proceedings of the 1985 Biomass Thermochemical Conversion Contractors' Meeting, pp. 31-55, CONF-8510167, National Technical Information Service, Springfield, Virginia.
- Evans, R. J., and T. A. Milne. 1986. "Applied Mechanistic Studies of Biomass Pyrolysis." In Proceedings of the 1985 Biomass Thermochemical Conversion Contractors' Meeting, pp. 57-79 CONF-8510167, National Technical Information Service, Springfield, Virginia.
- Diebold, J. P. 1985. The Cracking Kinetics of Depolymerized Biomass Vapors in a Continuous Tubular Reactor, MS Thesis T-3007, Colorado School of Mines, Golden, Colorado.
- Diebold, J. P., and J. W. Scahill. 1984. "Ablative Entrained-Flow Fast Pyrolysis of Biomass." In Proceedings of the 16th Biomass Thermochemical Conversion Contractors' Meeting, pp. 319-347, CONF-8405157, National Technical Information Service, Springfield, Virginia.
- Evans, R. J., T. A. Milne, M. N. Soltys, and H. R. Schulten. 1984. "The Mass Spectrometric Behavior of Levoglucosan Under Different Ionization Conditions and Implications for Studies of Cellulose Pyrolysis." J. Anal. Appl. Pyr. 6:273-283.
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GASIFICATION

Project Title: Conversion of Forest Residues to a Methane-Rich Gas in a High Throughput Gasifier - Battelle Columbus Laboratories

Feldmann, H. F., M. A. Paisley, and H. R. Appelbaum. 1986. "Conversion of Forest Residues to a Methane-Rich Gas in a High Throughput Gasifier." In Proceedings of the 1985 Biomass Thermochemical Conversion Contractors' Meeting, pp. 339-360 CONF-8510167, National Technical Information Service, Springfield, Virginia.

Paisley, M. A., H. F. Feldmann, and W. D. Watt. 1985. Wood Gasification Combined Cycle Retrofit in a Conventional Pulp Mill Cogeneration System. American Institute of Chemical Engineers, 1985 Summer Meeting, Seattle, Washington.

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