Biomass Thermochemical Conversion Program 1986 Annual Report

January 1987

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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
1986 ANNUAL REPORT

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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 1986.

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, the Thermochemical Conversion Program is sponsoring research activities in the following four areas:

1. Liquid Hydrocarbon Fuels Technology	3. Direct Combustion Technology
2. Gasification 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 1986 are summarized below and discussed in more detail in the text.

MAJOR ACCOMPLISHMENTS IN 1986

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

- measured octane numbers for gasoline from biomass
- improved hydrotreating of unstable pyrolysis oils
- successfully produced biocrude oils in extruder/static mixer reactor
- produced first biomass-derived gasoline using zeolite catalysts.

The Thermochemical Conversion Program completed experimental research in four medium-Btu gasifiers. Basic research was also conducted on an efficient method for converting moist biomass to fuel gases. Specific highlights include:

- completed extended term test of down-draft gasifier
- achieved high conversion of wet biomass to methane.

The Thermochemical Conversion Program also conducted research in the area of biomass combustion and Program support. Highlights of these activities include:

- successfully generated first electricity for TVA grid using biomass fired gas turbine generator
- served as Operating Agent for the IEA Bioenergy Agreement in the area of conversion.

CONTENTS

EXECUTIVE SUMMARY	iii
PROGRAM DESCRIPTION	1
INTRODUCTION	1
ORGANIZATION	2
OBJECTIVE AND GOAL	3
RATIONALE	4
APPROACH	6
PROGRAM RESEARCH PROJECTS AND PROGRESS	8
LIQUID HYDROCARBON FUELS TECHNOLOGY	8
Biocrude Production	11
Product Upgrading Research	17
GASIFICATION TECHNOLOGY	21
1986 Gasification Program	23
Medium-Btu Gasifiers	24
Medium-Btu Gasification Research	32
DIRECT COMBUSTION TECHNOLOGY	35
PROGRAM SUPPORT ACTIVITIES	38
APPENDIX - BIOMASS THERMOCHEMICAL CONVERSION PROGRAM RECENT PUBLICATIONS	A.1

FIGURES

1	Relationship of Biomass Thermochemical Conversion Program to Biofuels and Municipal Waste Technology Division Organization	3
2	Thermochemical Conversion Can Provide a Wide Spectrum of Products	4
3	Biomass is Far More Volatile Than Coal	5
4	Biomass Chars Gasify Very Rapidly	5
5	Program Approach	7
6	Industry Participation in the DOE Program	7
7	Biomass is Converted First to a Biocrude Oil and Then Upgraded to Gasoline Hydrocarbons	9
8	Liquid Fuels Projects Sponsored by the Thermochemical Conversion Program	11
9	Schematic Diagram of Entrained-Flow Pyrolysis Reactor at Georgia Tech Research Corporation	12
10	View of Georgia Tech Research Corporation Pyrolysis Reactor	13
11	Schematic Diagram of High Pressure Liquefaction Unit at University of Arizona	14
12	View of Extruder/Static Mixer Reactor at University of Arizona, Tucson, Arizona	15
13	Pyrolysis of Wood in a Methane Atmosphere	16
14	View of Reactor at Solar Energy Research Institute Used to Produce and Upgrade Biocrude Vapors	20
15	Thermochemical Gasification of Biomass	21
16	Gasification Research Projects	24
17	View of Battelle-Columbus Gasifier	25
18	Schematic Diagram of Battelle-Columbus Gasifier	26
19	Schematic Diagram of Indirectly Heated Gasifier at University of Missouri-Rolla	27
20	View of Gasifier at University of Missouri-Rolla	28

FIGURES (cont'd)

21	Schematic Diagram of Gasifier at Institute of Gas Technology	29
22	Carbon Conversion Versus Gasification Temperature (Maple Feed)	30
23	Photograph of SGI Gasifier	31
24	Conceptual Diagram of SGI Down-Draft Gasifier	32
25	Comparison of Experimental vs. Equilibrium Gas Composition . . .	33
26	Bench-Scale Fluidized Bed Gasifier at Pacific Northwest Laboratory	34
27	Direct Combustion Research	35
28	Schematic Diagram of Combustor/Gas Turbine at Aerospace Research Corporation	36
29	View of Gas Turbine and Generator System	37
30	Program Support Research Projects	39
31	Organization of the International Energy Agency Bioenergy Agreement	41

TABLES

1	Liquid Hydrocarbon Fuels from Biomass can be Produced with High Efficiencies	10
2	Comparison of the Energy Densities of Hydrocarbon Fuels from Biomass with Fuel Alcohols	10
3	Comparison of Biocrude Oil Products	17
4	Comparison of Typical Constituents of Biomass Catalytic Liquefaction Biocrude Oil and Hydrotreated Product	18
5	Comparison of Properties of Biocrude Oils and Hydrotreated Product	19

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, power industrial machinery, and even power 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 energy generation, for example, provides^(b) about 3.8% of the nation's energy supply.

(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.

With proper resource management and the development of efficient conversion processes, it has been estimated that biomass resources can provide an even 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 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 Field Management Office for the Biomass Thermochemical Conversion Program. As such, PNL has responsibility for overall management of the Program.

(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.

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 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 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:

- combustion to produce heat, steam, electricity, direct mechanical power, and combinations of these
- 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
- liquefaction to produce biocrude oils for upgrading into gasoline compatible hydrocarbon fuels.

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 great 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 pyrolyzed (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.

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

FIGURE 4. Biomass Chars Gasify Very Rapidly

- 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.
- 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.

- 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.

FIGURE 5. Program Approach

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. In 1986, for example, DOE issued a Request for Proposals to cost-share the scale-up of medium-Btu gasification technology.

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 FY86:

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 1986, the primary emphasis of the Thermochemical Conversion Program has been on the production of liquid hydrocarbon fuels from biomass. Liquid fuels from biomass offer several advantages over the biomass resources itself. Perhaps most important, liquid fuels have a higher energy density. 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. The higher energy density of liquid fuels from biomass allows the products to be transported more economically and to be more easily stored. Liquid fuels also match existing end-use patterns, particularly in the transportation sector. Biomass is the only renewable energy technology capable of addressing this need for transportation fuels.^(e)

The objective of the research sponsored by the Thermochemical Conversion Program is to produce gasoline-compatible liquid hydrocarbon fuels. As shown in Figure 7, the biomass resource is first converted to a biocrude oil and then upgraded using state-of-the-art catalytic methods. The catalytic upgrading step removes chemically bound oxygen from the biocrude oil and

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

yields liquid hydrocarbon products which are virtually identical to hydrocarbon products derived from petroleum. As shown in Table 1, the hydrocarbon fuels can be produced from biomass with high yields and with high overall thermal efficiencies. Preliminary economic analysis also indicates low production costs.^(h)

TABLE 1. Liquid Hydrocarbon Fuels from Biomass can be Produced with High Efficiencies

	<u>Theoretical</u>	<u>Goals</u>
Yield, gal/ton	90-130	75-100
Energy Efficiency, %	70-85	60-70
Gasoline Cost, \$/gal Wood @\$40/dry ton	---	1.00

Unlike ethanol, mixed alcohols, or other oxygenates which can be made from biomass, the hydrocarbon fuels produced in this manner are totally compatible with existing petroleum-based motor fuels. The hydrocarbon fuels also have significantly higher energy densities than alcohols due to the elimination of bound oxygen, as shown in Table 2. The liquid hydrocarbon products produced using this approach would serve as a gasoline blending stock or could potentially be used directly if factors such as octane number and volatility were appropriately adjusted.

TABLE 2. Comparison of the Energy Densities of Hydrocarbon Fuels from Biomass with Fuel Alcohols

<u>Fuel</u>	<u>Higher Heating Value</u>	
	<u>Btu/lb</u>	<u>Btu/gal</u>
Hydrocarbon Fuel from Biomass*	18,900	125,6200
Ethanol	12,600	84,300
Methanol	9,640	63,500

* Hydrotreated liquid produced at PNL. See p. 17

(h) Diebold, J. P., R. J. Evans, B. E. Levie, T. A. Milne, and J. W. Scahill. Low Pressure Upgrading of Primary Pyrolysis Oils from Biomass, Quarterly Report January-March 1986. Solar Energy Research Institute, Golden, Colorado

In 1986, the Thermochemical Conversion Program conducted research on both the production of biocrude oils and on the upgrading of those products into liquid hydrocarbon fuels. Projects funded by the Program are shown in Figure 8 and are discussed below.

FIGURE 8. Liquid Fuels 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 three projects in 1986 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. In addition, Brookhaven National Laboratory completed research on a process which directly produces liquid hydrocarbons. This work is described below.

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 9 and 10, consists of an entrained, upflow reactor and an oil recovery system that allows partial on-stream fractionation of the product.

FIGURE 9. Schematic Diagram of Entrained-Flow Pyrolysis Reactor at Georgia Tech Research Corporation

In previous research, GTRC performed experiments which established the interdependence of the reaction temperature and residence time on pyrolysis

FIGURE 10. View of Georgia Tech Research Corporation Pyrolysis Reactor

oil yield. Tests were performed over a temperature range of 400 to 550°C under varying residence times in the reactor. In 1986, GTRC completed additional test runs to optimize biocrude oils. Yields of biocrude oils up to approximately 66% on a thermal basis were achieved at temperatures of about 520°C and short residence times. Theoretical modeling studies initiated in 1986 suggest that liquid yields may increase further if residence times can be adjusted appropriately.

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 11 and 12. 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 11. Schematic Diagram of High Pressure Liquefaction Unit at University of Arizona

The University of Arizona has completed construction of the research unit and has begun experimental operation. Results to date show that the

FIGURE 12. View of Extruder/Static Mixer Reactor at University of Arizona, Tucson, Arizona

unit can be operated consistently and reliably over a variety of experimental conditions. Experiments at temperatures over the range 300–350°C, pressures of 1500–3000 psi, and a variety of feedstock concentrations have successfully been performed. The biocrude product produced is low in oxygen content and has a high heating value. The experimental results in 1986 suggest that the operating pressure may be reduced and that the CO/H₂ reducing gas may be reduced or eliminated without major impact on product quality. Research will continue on this process in 1987.

BROOKHAVEN NATIONAL LABORATORY, Upton, New York, completed an investigation of the effects which various reactive gases have on the products from biomass pyrolysis reactions. By using relatively high temperatures (1000°C), a range of light hydrocarbons and noncondensable gases can be generated. These products are significantly different than the biocrude oils formed at GTRC or University of Arizona and could potentially be used directly as octane enhancers or upgraded into gasoline. As shown in Figure 13, the yields of hydrocarbon products can be significantly altered when the reaction is conducted in the presence of methane. Using a bench-scale, entrained downflow reactor, biomass is pyrolyzed at temperatures of 800 to 1100°C in methane at pressures of 50 to 500 psi. With methane present, the yields of high-value products such as ethylene, benzene,

FIGURE 13. Pyrolysis of Wood in a Methane Atmosphere

toluene, and xylene can be up to about 2.5 times greater than those where an inert gas such as helium is used. Up to 50% of the carbon in the feedstock can be converted to liquid hydrocarbons. Gas phase products of CO and H₂ are also formed. During 1986, Brookhaven National Lab completed mechanistic studies on the source of this effect. The enhancement of liquid hydrocarbon yields appears to arise from the formation of free radicals from the biomass decomposition which subsequently react with methane.

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 3, 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 3. 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	1.5%	1.4%
<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.

PACIFIC NORTHWEST LABORATORY, 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 4 and 5. 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 by low pressure processes if an initial stabilization step is added. During 1986, PNL found that a mild hydrotreating step with a nickel catalyst or cobalt/molybdenum at relatively low temperatures

TABLE 4. 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 5. Comparison of Properties of Biocrude Oils and Hydrotreated Product

	Biocrude Oil	Refined Biomass Oil
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

(260-280°C) stabilized the low pressure biocrude oils. Following stabilization, the biocrude oils produced in the low pressure processes can be upgraded in the same way as those from the high pressure ones. The ability to successfully deal with a wide range of biocrude oils gives this upgrading approach great flexibility. Research is continuing in 1987.

SOLAR ENERGY RESEARCH INSTITUTE, 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. During 1986, SERI completed modification of 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 14.

In 1987, research is continuing to determine catalyst lifetimes, the extent of carbon deposition, and related factors in order to optimize reaction conditions.

FIGURE 14. View of Reactor at Solar Energy Research Institute
Used to Produce and Upgrade Biocrude Vapors

GASIFICATION TECHNOLOGY

Gasification of biomass can be achieved by reacting biomass, at moderately high temperatures, to produce a combustible gas containing large quantities of hydrogen and carbon monoxide. Heat is provided to the reactor either by combusting a portion of the biomass with air or oxygen, or by indirectly heating the reactor.

If air is used to provide heat for the gasifier, the nitrogen in the air significantly dilutes the product gas, and a low-Btu gas is produced as shown in Figure 15. Low-Btu gas typically has a heating value ranging from 90 to 200 Btu/scf. Today, low-Btu gasification of biomass 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%.

FIGURE 15. Thermochemical Gasification of Biomass

If nitrogen is eliminated from the product gas of a biomass gasifier by heating indirectly or with oxygen, a medium-Btu gas can be produced as shown in Figure 15. Medium-Btu gas has a heating value typically ranging from 300 to 600 Btu/scf and is much more versatile than low-Btu gas.

Because of its higher heating value, it possesses the following advantages over low-Btu gas:

- 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.

Medium-Btu fuel gas can also be produced from coal. The major disadvantage of coal gasification is that coal is not very reactive. Large quantities of oxygen are required to achieve sufficiently high reactor temperatures and reasonable reaction rates. Oxygen plants, however, are expensive to build and operate. This dictates that medium-Btu coal gasification plants be constructed with very large capacities to take advantage of economies of scale.

Because biomass is a distributed resource, transportation costs limit the amount of biomass that can be delivered to a central facility. Consequently, biomass gasifiers are limited to a maximum capacity of about 2,000 dry tons of wood per day. It is therefore necessary to reduce or eliminate the requirement for oxygen to achieve a cost-effective process at a smaller scale. Fortunately, biomass is much more reactive than other solid fuels such as lignite or coal and can be gasified at lower temperatures. Less heat is required for reaction due to the higher volatile content, and the remaining chars are also more reactive, as shown in Figures 3 and 4. This reduces

the oxygen requirement for medium-Btu gasifiers that are directly heated with oxygen.

Furthermore, the high reactivity and lower heating requirements make it possible to indirectly heat the biomass in the reactor without using expensive, exotic heat exchanger materials, thereby eliminating the requirement for oxygen. This approach offers the opportunity for significantly reducing gasification costs. In addition, the low sulfur content of most biomass feedstocks makes it possible to gasify biomass in the presence of catalysts without catalyst poisoning problems. As a result, smaller conversion facilities can be economically feasible if the conversion process capitalizes on the unique characteristics of the biomass feedstock. The lower sulfur content also eliminates the need for a costly gas cleanup system to remove sulfur from the product gas.

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.

1986 Gasification Program

The gasification projects sponsored by the Biomass Thermochemical Conversion Program in FY86 focused on two primary areas as shown in Figure 16. Work on four medium-Btu biomass gasification process research units (PRU's) concentrated on completing data collection and preparing final reports. Efforts at the Institute of Gas Technology and Battelle Columbus Laboratory consisted of data reduction and report writing. University of Missouri-Rolla completed final experimental tests and started preparation of a final report. SynGas, Inc., under contract to Solar Energy Research Institute, completed an extended experimental run in the oxygen-blown mode and a final report is being written.

In addition to the research with the PRU's, gasification studies were also conducted to resolve more basic research issues. This research included examination of the low temperature gasification of high moisture biomass feedstocks and studies of methods to improve the quality of product gases. The projects are shown in Figure 16 and are discussed in more detail below.

FIGURE 16. Gasification Research Projects

Medium-Btu Gasifiers

BATTELLE-COLUMBUS LABORATORIES (BCL), Columbus, Ohio, is conducting research to determine the feasibility of using a dual bed, entrained gasifier to produce a medium-Btu fuel gas. The gasifier is shown in Figure 17. The concept provides for indirect heating of the gasifier by circulating a low density, hot incandescent sand from a combustor to the gasifier, thus eliminating the need for using purified oxygen. As shown in Figure 18, the entrained sand and any char leaving the gasifier is separated from the product gas in a cyclone separator. Both the sand and char are transferred to an entrained bed combustor where the char is burned with air. The sand is heated to a high temperature and recirculated back to the gasifier to provide heat for the gasification reaction.

In previous research, BCL successfully produced a medium-Btu fuel gas with a heating value of about 475 Btu/scf using either wood chips or coarsely shredded bark. This heating value did not vary significantly for feedstocks containing from 8% to 40% moisture on a wet basis. Gasifier throughputs of up to 1800 lb/hr/ft² were achieved, and the PRU operated continuously for extended periods up to 72 continuous hours. Wood feedstocks ranging from chips to sawdust to hogged fuel were successfully gasified. Research has also shown that gasifier tars can successfully be destroyed by injecting them

FIGURE 17. View of Battelle-Columbus Gasifier

directly into the combustor vessel. In 1986, BCL completed analysis of their data and prepared a draft final report. The draft is in review and will be published in 1987.

FIGURE 18. Schematic Diagram of Battelle-Columbus Gasifier

THE UNIVERSITY OF MISSOURI, Rolla, Missouri, is investigating the technical feasibility of using an internal heat exchanger to provide heat indirectly to a fluidized bed gasifier. In the conceptual design, a portion of the feedstock or product gas would be burned to provide heat for the fire-tube heat exchanger. This eliminates the need for pure oxygen in heating the gasifier. The university has constructed a 20-in. fluidized bed gasifier with an internal heat exchanger as shown in Figures 19 and 20. The configuration selected for actual testing uses thirty 1-in. diameter U-tubes spaced on a 2-in. pitch. The system has been operated at feed rates up to 400 lb/hr (10% moisture wood) and temperatures up to 1450°F. The university also completed an extended term test. Continuous, uninterrupted operation of the gasifier was achieved for a period of five days and yielded valuable information on material and energy balances which helped confirm the operational feasibility of this concept.

In 1986, the University of Missouri completed the collection of data to determine the effect of recycling raw product gas. The recycle of product

Indirectly Fired Wood Gasifier

Gasifier Internal Heat Exchanger

FIGURE 19. Schematic Diagram of Indirectly Heated Gasifier at University of Missouri-Rolla

gas would reduce steam requirements by displacing part of the steam needed for fluidizing the bed of the gasifier. The recycling could also improve conversion of tars. The use of recycle gas was found to have a significant impact on the hydrogen/carbon monoxide ratio of the final product gas. At a bed temperature of 760°C, for example, the H_2/CO ratio of the product gas changes from about 0.5 to about 1.4 when the fluidizing steam is partially replaced by recycle gas. The change may arise primarily from the water shift gas reaction. It is important to note that the recycle gas must go through a plenum at about 1100°C before it reaches the fluidized bed, so that the reactions may be taking place before reaching the bed itself. The University of Missouri also began preparation of a final report which should be completed in early 1987.

THE INSTITUTE OF GAS TECHNOLOGY (IGT), Chicago, Illinois, is conducting research to determine the minimum oxygen requirements for gasifying biomass

FIGURE 20. View of Gasifier at University of Missouri-Rolla

to produce a medium-Btu gas. A 12-in. diameter fluidized bed research gasifier capable of operation at pressures up to 500 psia has been constructed and is shown conceptually in Figure 21. Wood is fed from a pressurized lockhopper to a pressurized, live-bottom feed hopper and is metered into the gasifier unit. Wood feedstock throughputs of about 1000 lb/hr/ft² have been achieved. At these feed rates, a pressure of 325 psia, and a temperature of about 1500°F, the total carbon conversion to gas exceeded 88%. Under these conditions, steam requirements were in the range of 0.5 to 0.8 lb/lb wood. Perhaps most important, the oxygen requirements over a variety of conditions were found to be relatively low. Under the conditions above, the oxygen requirement was 0.21 to 0.34 lb/lb wood.

FIGURE 21. Schematic Diagram of Gasifier at Institute of Gas Technology

IGT has completed an extensive matrix of tests to determine the effect of various experimental parameters such as pressure, temperature, and feed rates on product yields. The tests were conducted over the range 1390 to 1800°F, 100 to 300 psig, and feed rates up to 1030 lb/hr. The effect of

the temperature of the fluidized bed on the carbon conversion is shown in Figure 22. IGT has also completed continuous, extended-term tests of up to three days in length. The tests confirm the mass and energy balances of previous shorter term runs and also confirm the ability of the facility to operate at pressure for extended periods. During 1986, IGT prepared a draft final report describing the progress to date. The draft report is being peer reviewed and will be published in 1987.

FIGURE 22. Carbon Conversion Versus Gasification Temperature (Maple Feed)

SOLAR ENERGY RESEARCH INSTITUTE (SERI), Golden, Colorado, is conducting research on the use of a pressurized, down-draft, oxygen-blown gasifier to produce medium-Btu fuel gas. During 1986, the research effort centered on the operation in an oxygen-blown mode of a 20 ton/day down-draft gasifier built by SynGas Incorporated (SGI). The SGI gasifier was operated under subcontract to SERI. A view of the SGI unit is shown in Figure 23, and the gasifier is shown schematically in Figure 24.

The conversion of the gasifier from air-blown to oxygen-blown capability was completed at the beginning of FY86, and testing to determine mass and energy balances, oxygen consumption and other experimental parameters was conducted. The gasifier was successfully operated at atmospheric pressure over a variety of conditions, and wood feed rates up to 2000 lb/hr. were achieved. Oxygen consumption was typically 0.25 lb. O_2 /lb. wood, and the

FIGURE 23. Photograph of SGI Gasifier

gas produced had a higher heating value of about 300 Btu/scf. SGI also completed an extended duration test totaling 63 continuous hours of operation. A final report describing this work will be issued in 1987.

FIGURE 24. Conceptual Diagram of SGI Down-Draft Gasifier

Medium-Btu Gasification Research

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. 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, aquatic plants, and

food processing wastes 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 ($\approx 40\%$), hydrogen ($\approx 8\%$) and carbon dioxide ($\approx 52\%$) and could therefore be upgraded to a high-Btu gas by simply removing CO_2 .

During 1986, reaction parameter tests and kinetic studies were carried out using a one-liter batch reactor system. These tests indicate that the ash content of some of the biomass feedstocks tested may provide adequate catalytic activity to eliminate the need for added alkali catalyst. Other tests which were carried out indicate that under proper catalyst loadings, the reaction of biomass is nearly complete at temperatures significantly below 400°C with residence times of 10-15 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 25, this gas composition is close to the estimated equilibrium composition for these conditions.

FIGURE 25. Comparison of Experimental vs. Equilibrium Gas Composition

PACIFIC NORTHWEST LABORATORY (PNL), Richland, Washington, 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 should be removed for many end uses.

During 1986, PNL conducted basic research to test the effects of different fluidizing contact gases on the quality of the gasifier product gas. Using a bench-scale fluidized bed reactor shown schematically in Figure 26, experiments were performed for fluidizing gases including, carbon dioxide, methane, and hydrogen. The experiments were conducted over a temperature range of 650-800°C and at residence times of both one and two seconds. The experiments showed that the fluidizing gases could alter the spectrum of product gases, which is consistent with results from the gasifier at University of Missouri. Tar formation, while low in all cases, remained relatively unchanged as a function of contacting gas. These results indicate that pyrolytic decomposition is the initial reaction occurring in the gasifier. The subsequent steam reforming reaction with chars and tars is slower. As a result, there may be a good opportunity to reduce steam requirements in gasifier systems by replacing the fluidizing steam with other, potentially less expensive gases.

FIGURE 26. Bench-Scale Fluidized Bed Gasifier at Pacific Northwest Laboratory

DIRECT COMBUSTION TECHNOLOGY

Direct combustion of biomass feedstocks, particularly wood, is already 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 widespread 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 examine unique, innovative combustion systems as shown in Figure 27.

FIGURE 27. Direct Combustion Research

The Biomass Thermochemical Conversion Program is currently sponsoring direct combustion research to determine the technical feasibility of converting the heat released from biomass combustion directly into mechanical power. By directly producing mechanical power without the use of an intermediate working fluid such as steam in a boiler/steam turbine system, high conversion efficiencies can be realized. In addition, costs associated with the working fluid, such as boilers and condensers, are eliminated, allowing small engines to operate economically.

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 28. 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.

FIGURE 28. Schematic Diagram of Combustor/Gas Turbine at Aerospace Research Corporation

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 29. 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. A majority of the funds for the unit are being provided by the private sector. Electric power generated by the unit will also be sold to TVA.

FIGURE 29. View of Gas Turbine and Generator System

In 1986, Aerospace Research Corporation completed initial shakedown of the 3-MW system and began generating electrical power. Power generation to the TVA grid can occur only when the generator is operating in a constant speed synchronous mode, and special start-up procedures were formulated to achieve stable operating speeds. Continuous operation of the turbine for long periods of time was hampered by operational problems including problems with the feeding systems and pressure losses in the ducting system which lowered operating efficiencies. Modifications to these systems will be completed in 1987, and operation of the 3-MW unit will continue.

Aerospace Research Corporation also conducted research on a second project aimed specifically at testing the suitability of using animal manures as feedstocks for gas turbine systems. The first stage of the project included the characterization of a bovine manures from New Mexico. The manures were found to contain 25-49% ash on a dry basis and were high in chlorine, but the ash softening point was slightly higher than that for wood. Based on the analysis, the manures were deemed to be suitable for further testing. At the end of FY 1986, one test of the manure feedstock in the smaller 375 kW turbine system was performed. The test, totaling about 12 hours in duration, showed that the turbine will operate on these feedstocks. The long term effects of the chloride and sulfur in the combustion gases on turbine life, however, remain uncertain. Additional testing will be conducted in 1987.

PROGRAM SUPPORT ACTIVITIES

During FY86, 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 30.

FIGURE 30. Program Support Research Projects

SCIENCE APPLICATIONS INTERNATIONAL CORPORATION (SAIC), Arlington, Virginia, conducted engineering and economic studies to assess advanced biomass conversion concepts. During 1986, SAIC completed an analysis of the production of ethanol using thermochemical conversion processes. In this conceptual process, biomass would be gasified to form a synthesis gas. The synthesis gas would be reformed sequentially to methanol and then ethanol. For a 1000 ton/day facility, ethanol production costs range from \$0.82-\$1.47 depending on process modifications and assuming utility financing. Feedstock costs for this analysis were assumed to be \$25/ton. The analysis suggests that thermal conversion of wood to ethanol may be competitive with other conversion methods.

SAIC also completed draft analyses in two other areas. Analyses of the four medium-Btu gasifiers described previously were updated using final data reported by these investigators. SAIC also completed a draft analysis of producing liquid hydrocarbons from wood. The analysis assumed that the biocrude oils would be produced in a low pressure pyrolysis process and upgraded using hydrotreating techniques. The draft of both reports are being peer reviewed and should be published in 1987.

PACIFIC NORTHWEST LABORATORY (PNL), Richland, Washington, served as the Operating Agent for cooperative biomass conversion activities sponsored by the International Energy Agency's Bioenergy Agreement. Under this Agreement, eleven 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 31.

As Operating Agent for the Biomass Conversion Annex, PNL coordinated the international efforts and administered funding for individual projects. During 1986, ten cooperative projects were active. These ten 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 31. Organization of the International Energy Agency Bioenergy Agreement

TABLE 21. Classification of the Incision of the River into the River

Incision	River	River	River	River

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.5
Gasification	p. A.6 - A.10
Combustion	p. A.11
Program Support	p. A.12

LIQUID HYDROCARBON FUELS

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

Knight, J. A., C. W. Gorton, R. J. Kovac, and C. J. Newman. "Entrained Flow Pyrolysis of Biomass." In Proceedings of the 1985 Biomass Thermochemical Conversion Contractors' Meeting, Minneapolis, Minnesota, 15-16 October 1985. CONF-8510167 (NTIS).

Gorton, C. W., and J. A. Knight. "Oil from Biomass by Entrained Flow Pyrolysis." In Proceedings of the Sixth Symposium on Biotechnology for Fuels and Chemicals. In press.

Knight, J. A., and C. W. Gorton. "Oil Production by Entrained Flow Pyrolysis of Biomass." In Proceedings of the Third Biomass Liquefaction Experts Meeting. University of Sherbrooke, Sherbrooke, Quebec, Canada, 1983. 138-46.

Knight, J. A., C. W. Gorton, and R. J. Kovac. "Entrained Flow Pyrolysis of Biomass." In Proceedings of the 16th Biomass Thermochemical Conversion Contractors' Meeting, (pp 287-97) Portland, Oregon, 8-9 May 1984. CONF-8405157 (NTIS).

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

White, D. H., D. Wolf, N. W. Andrews, D. Joshi, J. Wong, and G. Davenport. "Real-Time Microprocessor Control and Data Acquisition of Biomass Direct Liquefaction Systems." Presented at the Second National Conference on Microcomputer Applications for Conservation and Renewable Energy, Tucson, Arizona, 21-23 October 1986. In Press.

White, D. H., and D. Wolf. "A Continuous Extruder-Feeder for Reactor Systems for Biomass Fuels Processing." Presented at Energy from Biomass and Waste X, sponsored by the Institute of Gas Technology, Washington, D.C., 7-10 April 1986. In Press.

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LIQUID HYDROCARBON FUELS (cont'd)

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- Ghoddoussi, M. G. "Solvolysis of Biomass at Elevated Temperatures." Master of Science Thesis, University of Arizona, December 1983.
- Homaidan, A. "Modeling of Extruder-Feeder for Biomass Concentrated Slurries." Master of Science Thesis, University of Arizona, May 1984.
- Khan, M. B. "Gas Dispersion in Viscous Non-Newtonian Fluids Using an Extruder-Feeder." Master of Science Thesis, University of Arizona, August 1984.
- Lezzar, A. "Effects of Void Volume on the Friction and Rheology of Concentrated Slurries," Master of Science Thesis, University of Arizona, December 1983.
- Shlapak, N. A. "Microprocessor-Based Real-Time Process Control of Biomass Liquefaction." Master of Science Thesis, University of Arizona, August 1984.
- White, D. H. "Development of Extruder-Feeder Biomass Liquefaction System." In Proceedings of the 16th Biomass Thermochemical Conversion Contractors' Meeting, Portland, Oregon, 8-9 May 1984. CONF-8405157 (NTIS).
- Project Title:** Flash Pyrolysis and Hydropyrolysis of Biomass - Brookhaven National Laboratory
- Steinberg, M., P. T. Fallon, and M. S. Sundaram. "Flash Pyrolysis of Biomass with Reactive and Non-Reactive Gas." Biomass (9) (1986): 293-315.
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LIQUID HYDROCARBON FUELS (cont'd)

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- Project Title:** Upgrading of Liquid Fuels from Biomass - Pacific Northwest Laboratory
- Elliott, D. C., and E. G. Baker. "Hydrotreating Biomass Liquids to Produce Hydrocarbon Fuels." Presented at Energy from Biomass and Waste X, sponsored by the Institute of Gas Technology, Washington, D.C., 7-10 April 1986. In Press.
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GASIFICATION

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

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