DE-79819-78

DOE/PC/79819--T2

DOE/PC/79819--T2

DE93 008632

**BENCH-SCALE DEVELOPMENT OF COAL/OIL
CO-PROCESSING TECHNOLOGY
FINAL SUMMARY REPORT**

J.E. Duddy
S.V. Panvelker
R.J. Parker (ARC)
M.S. Pramanik
G.A. Popper

WORK PERFORMED UNDER CONTRACT

DE-AC22-87PC79819

**HYDROCARBON RESEARCH, INC.
100 Overlook Center
Princeton, New Jersey 08540**

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

December 1991

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

**BENCH-SCALE DEVELOPMENT OF
COAL/OIL CO-PROCESSING TECHNOLOGY
FINAL SUMMARY REPORT**

**J.E. Duddy
S.V. Panvelker
R.J. Parker (ARC)
M.S. Pramanik
G.A. Popper**

**WORK PERFORMED UNDER CONTRACT
DE-AC22-87PC79819**

**HYDROCARBON RESEARCH, INC.
100 Overlook Center
Princeton, New Jersey 08540**

December 1991

TABLE OF CONTENTS

Section

I	Introduction	1
II	Objectives and Scope of Work	3
III	Continuous Bench-Scale Operations	7
IV	Laboratory Support	12
V	Conceptual Commercial Plant Design and Economics	14
VI	Recommendations for Future Work	16

Section I - Introduction

Hydrocarbon Research Inc. (HRI) is developing advanced, catalytic, two-stage technology for co-processing coal and petroleum derived oil. HRI's Coal/Oil Co-Processing technology uses the ebullated-bed reactor, invented by HRI, and in commercial operation since 1968 in the H-Oil® Process for upgrading petroleum derived residual oils. The ebullated-bed reactor has also been successfully demonstrated for processing coal slurries at the 200 TPD H-Coal® pilot plant operations in Catlettsburg, Kentucky from 1980 through 1982. Co-Processing of coal with petroleum derived residual oil was first demonstrated at the bench-scale in 1974, and HRI has been working on bench-scale development continuously since 1985. Scale-up of the co-processing technology from the 50 lb/day bench-scale to the 3 TPD PDU-scale was successfully demonstrated in 1989.

In coal/oil co-processing, coal is slurried with petroleum derived oil. Petroleum derived oils which can be used include atmospheric and vacuum residua, FCC slurry oils, heavy crudes, tar sands bitumen or shale oil. HRI has evaluated both single and two-stage (in series) reactor configurations. A two-stage configuration is preferred to obtain high conversions, high distillate yields and good product quality. The effluent from the first-stage reactor flows directly to the second-stage reactor, without interstage separation. Both reactors use commercially available NiMo or CoMo extrudate hydroprocessing catalysts. The unconverted residual oil, unconverted coal and ash is rejected via simple vacuum distillation. The resulting vacuum bottoms slurry is limited to a maximum solids content of about 50 W%, to maintain a pumpable slurry. Typically, co-processing operations are performed on a once-through basis, with a maximum dry coal concentration in the feed slurry of about 40 W%. Higher coal concentrations (up to 67 W% dry coal) have been demonstrated with the addition of a small amount of process-derived atmospheric bottoms recycle to the feed slurry.

Interest in the development of coal/oil co-processing technology is based on potential economic and technical advantages relative to coal liquefaction or hydroconversion of petroleum residua alone. Co-processing can provide a means to accelerate the acceptance of coal-derived liquids in conventional oil refineries. This introduction of coal liquids to the market would replace imported oil and utilize domestic resources and labor. Capital requirements for the production of these liquid fuels would certainly be minimized through efficient utilization of reactor volume and existing refining infrastructure. Perhaps even more important than these factors, however, is the apparent synergy which results in the combined processing of coal and petroleum derived oil. This synergy occurs through greater conversion to distillate liquids through combined processing, and removal of the petroleum derived contaminant metals (primarily nickel and vanadium) on the coal solids. This is especially important in the ebullated-bed reactor system in protecting the extrudate catalyst from metals poisoning, and extending catalyst life.

HRI, under contract to the U.S. Department of Energy (DOE Contract No. DE-AC22-87PC79819) has performed a cost-shared research and development project for, "Bench-Scale Development of Coal/Oil Co-Processing Technology." The period of performance for this project covered in excess of four years, from September 1987, through December 1991. The co-sponsors of this program have included:

U.S. Department of Energy
Alberta Research Council
Mitsui SRC Development Co., Ltd.
Ruhrkohle Oel und Gas, GMBH
Electric Power Research Institute
Ontario-Ohio Synthetic Fuels Corporation, Ltd.
HRI, Inc.

The work featured a coordinated program of continuous bench-scale operations and engineering assessments of results at HRI, with laboratory-scale support provided by Alberta Research Council. This final report presents a summary of the work completed as a part of this project. Details on specific elements of this work are contained in the following topical reports:

- 1) Duddy, J.E., Pramanik, M.S. and G.A. Popper, "Bench-Scale Development of Coal/Oil Co-Processing Technology - Conceptual Commercial Plant Design and Economics," DE-79819-TOP-01, September 1990.
- 2) Duddy, J.E. and S.V. Panvelker, "Bench-Scale Development of Coal/Oil Co-Processing Technology - Continuous Bench-Scale Operations," DE-79819-TOP-02, December 1991.
- 3) Parker, R.J., "Bench-Scale Development of Coal/Oil Co-Processing Technology - Laboratory Support," DE-79819-TOP-03, December 1991.

Section II - Objectives and Scope of Work

The goal of this project was to expand the data base for HRI's Coal/Oil Co-Processing Technology and to allow for process optimization. The specific objectives were to:

- define process performance at commercially viable catalyst cost and activity levels
- determine the dependence of process performance on changes in feedstock characteristics
- improve the effectiveness of the catalyst system
- expand the data base to include other coal and petroleum feedstocks
- update economic assessments

To meet these stated objectives, work was performed in three main area:

- Continuous Bench-Scale Operations
- Laboratory Support
- Conceptual Commercial Plant Design and Economics

CONTINUOUS BENCH-SCALE OPERATIONS

Continuous bench-scale operations represent the smallest scale of operation available to obtain coal/oil co-processing process performance data, adequate in quality to develop meaningful process models and to project commercial performance. As a part of this project, HRI modified an existing single-stage, bench-scale unit (Unit No. 238) to handle two-stage operations. An existing two-stage bench-scale unit (Unit No. 227) was also available for operations in this project. The bench-scale operations were separated into two major areas:

1. Expansion of Feedstock Data Base

A critical aspect to the understanding of coal/oil co-processing and the feedstock interactions involved is to have an adequate data base. Bench-scale operations in this area were designed to evaluate different feedstocks combinations. Each run was scheduled to operate continuously for a total of 15-25 days. Based on HRI's past experience in continuous bench-scale operations, a number of operating conditions can be evaluated over the course of a single run. Each condition covers 3-7 operating days depending upon

specific requirements for the condition. In addition to evaluating feedstock combinations, the runs can evaluate the effect of other process variables including: temperature, relative space velocity, coal concentration and recycle.

2. Process Improvement

This area of continuous bench-scale operations was performed to evaluate the potential for improvement in process performance and/or costs through use of process improvements. Process improvements evaluated include alternative operating modes, feedstock pretreatment and alternative catalyst systems.

In addition to these continuous bench-scale operations, the scope of work included activities to modify the existing bench-scale unit and to perform non-routine analyses of co-processing products.

LABORATORY SUPPORT

Smaller-scale laboratory support work was performed by Alberta Research Council (ARC) as an integral part of this bench-scale development program. In general the laboratory support work represents activities undertaken to screen alternatives for subsequent bench-scale testing. Additionally, laboratory support work is performed to provide improved fundamental understanding of a particular aspect of the coal/oil co-processing technology. For this program there were two major elements to the laboratory support work at ARC.

The first part involved feedstock reactivity screening of feedstocks in a batch autoclave. Standard operating conditions have been developed to best simulate conditions in the ebullated-bed reactor, including:

Temperature, °F	806
Feed: Diluent Ratio	1:2
Feed: Catalyst Ratio	1:1
Time, minutes	100

Batch autoclave reactivity screening focused on the optimal integration of coal/oil co-processing with ARC's oil agglomeration technology, as a coal pretreatment step. The scope of work also included production of adequate quantities of oil agglomerated coal for bench-scale testing.

The other major element of the laboratory support work involved screening of catalysts, using the batch autoclave, as well as fixed-bed catalytic reactors. Catalysts screened included dispersed iron-based catalysts and spent catalysts from prior coal/oil co-processing operations. Spent catalyst activity was screened to evaluate the potential for catalyst re-use through cascading.

In addition to these major activities in laboratory support, other work was performed to provide additional feedstock and product characterization.

CONCEPTUAL COMMERCIAL PLANT DESIGN AND ECONOMICS

The conceptual commercial plant design was based on a preliminary engineering design of a co-processing plant. This preliminary design was for a full-scale co-processing production facility, containing two maximum sized reactor trains. The design work included a heat and material balance simulation, major equipment specifications, utility and operating cost estimates. Although the engineering work was preliminary and abbreviated in nature, it was performed in sufficient detail to support a meaningful budget-type cost estimate. The design detail, however, was not sufficient to proceed to detailed engineering or to purchase equipment. A summary of the work included follows:

- *Design Basis* of the sections within the battery limits was outlined by general criteria and included the quantity and properties of the charge stocks and products.
- *Battery Limits* for the preliminary design of the HRI Coal/Oil Co-Processing Technology included all areas from slurry mixing to rough splitting of the product fractions. The specific process steps included were:
 - Coal slurry mixing
 - Preheat and reaction
 - Primary separation
 - Recycle hydrogen purification and compression
 - Make-up hydrogen compression
 - Product fractionation
- *Process Description* explaining the process flow through the process steps.
- *Process Flow Sheets* showing the major equipment.
- *Overall Material Balance* showing the composition and rates of feed and product streams.
- *Utility and Catalyst Consumptions* were estimated.
- *Battery Limits Capital Cost Estimates* was budgetary or feasibility type with vendor input on major equipment or recent quotations for similar equipment. The remainder of commodity materials and labor were factored based on statistical techniques developed for the H-Oil® Process.

- *Duty Specifications and Curve-Type Cost Estimates* for all ancillary process such as hydrogen manufacture, sour water treating, gas clean-up, sulfur plant, etc.
- *Overall Plant Balances and Product Costs* were developed for complete co-processing facilities based on HRI's Process Planning Model.

The economic screening studies were performed in two-parts. The first part defined a complete, grass-roots, stand-alone co-processing facility. This facility depicted was devoted entirely to co-processing. The primary feedstocks to this plant were coal and petroleum derived residuum. The plant included all facilities necessary to produce finished quality products (gasoline and utility turbine fuel) in an environmentally acceptable manner. Product costs were calculated and related to an equivalent cost for light crude oil (West Texas Intermediate).

The other part of the economic screening studies evaluated co-processing in a refinery situation. Three grass-roots refineries were depicted representing the transition from processing all light crude, to a mix of light and heavy crudes, to the addition of coal and co-processing. Economics were evaluated in terms of the light crude price required to make co-processing the lowest cost option.

The fundamental basis for the bench-scale research and development of coal/oil co-processing technology is derived from HRI's prior Coal/Oil Co-Processing Program supported by private industry. Performed from 1985-1987, this work included feedstock characterization, microautoclave and autoclave reactivity screening, continuous bench-scale operations, product characterization, economic screening and commercial planning. Reports on this work are available from EPRI (EPRI AP-5101, March 1987, and EPRI GS-6357, April 1989).

In parallel with this bench-scale research and development program, HRI has also been working on the commercialization of coal/oil co-processing as a part of the Clean Coal Technology Program. The Ohio Clean Fuels Prototype Commercial Coal/Oil Co-Processing Plant Project was selected by DOE in the first Clean Coal Technology solicitation. This project is for a first-of-a-kind commercial co-processing plant to produce about 12,000 BPD of clean liquid fuels using HRI's technology. As a part of this project, HRI successfully completed scale-up of co-processing to the 3 TPD Process Development Unit scale in 1989 (DOE Report No. DE/PC/79797-T4 (Vol.2).

Section III - Continuous Bench-Scale Operations

Continuous bench-scale operations represent the smallest scale of operation available to obtain coal/oil co-processing process performance data, adequate in quality to develop meaningful process models and to project commercial performance. The bench-scale operations were separated into two major areas:

1. Expansion of Feedstock Data Base

A critical aspect to the understanding of coal/oil co-processing and the feedstock interactions involved is to have an adequate data base. Bench-scale operations in this area were designed to evaluate different feedstocks combinations. Each run was scheduled to operate continuously for a total of 15-25 days. Based on HRI's past experience in continuous bench-scale operations, a number of operating conditions can be evaluated over the course of a single run. Each condition covers 3-7 operating days depending upon specific requirements for the condition. In addition to evaluating feedstock combinations, the runs can evaluate the effect of other process variables including: temperature, relative space velocity, coal concentration and recycle.

2. Process Improvement

This area of continuous bench-scale operations was performed to evaluate the potential for improvement in process performance and/or costs through use of process improvements.

Seven coal feedstocks were tested in this program. The coals tested were from the United States, Germany, Japan and Canada. The coal qualities covered a wide range of rank, ranging from a very high rank bituminous coal to low rank sub-bituminous coals and lignite. The ash content of the coals processed ranged from low, about 5 W% of dry coal (Westerholt, Ohio and Black Thunder), to intermediate, about 10-13 W% of dry coal (Texas Lignite, Taiheiyo and New Mexico), to high, about 18 W% on dry coal (Forestburg). The coals in general had very low sulfur, ranging from 0.3-2.3 W% on dry coal. This is largely due to the fact that most of these coals come from active mining operations and there is a very limited market for high sulfur coal.

Four oil feedstocks were tested in this program. The oils came from the United States, Canada, Venezuela and Mexico, and were all derived from poor quality heavy crudes. All of the feedstocks had very high sulfur (4.4-5.7 W%), nitrogen (0.5-0.8 W%) and metals (385-982 wppm) contents. The Maya and Cold Lake vacuum residua had the same hydrogen contents (9.9 W%) and similar Conradson Carbon Residues (CCR) of about 25 W%. In comparing the Maya and Hondo atmospheric residua, the Hondo is of higher quality, with higher API gravity, higher

hydrogen content and lower CCR. The Orinoco residuum is of slightly poorer quality than Maya residuum.

The continuous bench-scale operations completed as a part of this project included ten separate runs, covering a total of 223 operating days. In six of the runs different feedstock combinations were evaluated over a total of 133 operating days. The base feedstock combination for this bench-scale program was Texas Lignite and Maya residuum. This feedstock combination represents a logical combination for processing at a U.S. Gulf Coast location, where over 45% of all U.S. refining capacity is located. In addition to this base feedstock combination, the following feedstock combinations were evaluated:

Texas Lignite/Cold Lake
Texas Lignite/Hondo
Westerholt/Cold Lake
Taiheiyo/Maya
Forestburg/Cold Lake
Black Thunder/Cold Lake
New Mexico/Hondo
Ohio/Maya
Ohio/Orinoco

The foreign coals, Westerholt (Germany), Taiheiyo (Japan) and Forestburg (Canada), were tested in recognition of the contributions of Ruhrkohle, Mitsui SRC Development Co., Ltd., and the Alberta Research Council to the project.

In addition to evaluating the co-processing reactivity of different feedstock combinations, four of the bench-scale runs examined potential process improvements. Process improvements were evaluated over 90 operating days including:

Low/High Temperature Staging
Feedstock Pretreatment (Oil Agglomeration)
Catalyst Cascading
Dispersed Iron Additive
Optimal Coal Concentration

In the course of these bench-scale operations, a wide range of operating conditions were evaluated. These operating conditions covered the following ranges:

• relative space velocity	- 1.0-2.4
• first-stage temperature	- 750-825°F
• second-stage temperature	- 810-825°F
• coal concentration	- 10-50 W% Dry Feed
• recycle:feed ratio	- 0-0.5
• dispersed iron additive	- 0-3 W% Dry Feed

Product yields and process performance results also covered a wide range. While it is generally desirable to operate at the highest levels of process performance practical, operation at lower severities provides valuable data for development of accurate process models. Following is the range of product yield and process performance results obtained in the bench-scale operations:

- 975°F⁺ Conversion - 66.0-93.2 W% MAF
- Coal Conversion - 81.4-96.5 W% MAF
- HDS - 69.0-97.7 W%
- HDN - 46.2-87.7 W%
- Demetallization - 85.0-99.9 W%
- Total C₄-975°F Distillate Yield - 58.7-80.1 W% MAF
- Net C₄-975°F Distillate Yield - 55.0-78.3 W% MAF
- C₁-C₃ Gas Yield - 3.0-5.3 W% Dry Feed
- Hydrogen Consumption - 2.8-5.4 W% Dry Feed

Following are the major conclusions from this program based on HRI's analysis of the data obtained:

- the base feedstock combination for this program, Texas Lignite and Maya residuum was found to be very reactive in co-processing. 975°F⁺ conversion as high as 93 W% MAF was obtained. Coal conversions in excess of 90 W% MAF were consistently obtained. Net yields of C₄-975°F liquids approached 80 W% MAF, and over five barrels per ton of MAF feed.
- distillate product qualities were very good including production of a mid-distillate product which meets the future diesel specification of 0.05 W% sulfur, with no further upgrading.
- 975°F⁺ conversion was found to increase with increasing coal concentration in once-through operations with Texas Lignite and Maya residuum. At 10 W% dry coal feed the 975°F⁺ conversion was 80.5 W% MAF. At 40 W% dry coal feed the 975°F⁺ conversion was 90.0 W% MAF.
- test results indicated that Hondo residuum was much more reactive in co-processing than either Maya or Cold Lake residua. The reactivity of the residua in co-processing can be related to the Conradson Carbon Residue, which provides an indirect measurement of the aromaticity of the oil feedstock. The observed difference in reactivity was significant, with a more than 30% increase in reaction rate, which can be translated to a 30% increase in reactor throughput at constant 975°F⁺ conversion.
- the Taiheiyo sub-bituminous coal feedstock was also very reactive in co-processing with Maya residuum. Extremely high coal conversion of

96.5 W% was demonstrated. This is the highest coal conversion ever achieved in HRI's Coal/Oil Co-Processing Technology.

- the Westerholt bituminous coal was the least reactive feedstock tested. Demonstration of 975°F⁺ conversion in excess of 90 W% MAF, required reducing the relative space velocity from 1.0 to 0.8, and a 10°F increase in reactor operating temperature, relative to operations with Texas Lignite. Since Westerholt is a relatively high rank bituminous coal (O/C atomic ratio of 0.08), the yield of distillate liquid products will be greater than that for a lower ranked coal.
- co-processing performance with New Mexico sub-bituminous coal and Hondo residuum was the best ever demonstrated in HRI's Coal/Oil Co-Processing Technology. 975°F⁺ conversion was in excess of 93 W% MAF. HDS was extremely high, 97 W% after 21 days of bench-scale operation. Even at moderate severity conditions, designed to produce 975°F⁺ conversion of about 75 W% MAF, conversion was in excess of 85 W% MAF.
- the observed rate of catalyst deactivation when processing Taiheiyu and New Mexico sub-bituminous coals was extremely low, less than half the rate observed for Texas Lignite and Maya residuum. Both coals were notable for very high hydrogen contents (H/C atomic ratios of 1.00 for Taiheiyu coal and 0.90 for New Mexico coal). The low deactivation rate is related to this high hydrogen content and also evidenced by lower carbon loadings on recovered catalysts from these operations.
- recovered catalysts showed extremely low levels of petroleum-derived metals contaminants (Ni+V), especially when the high metals contents of the feed oils (up to 1,000 wppm) is considered. Co-Processing these oils with coal will lead to reduced catalyst consumption compared to processing oil only.
- an accurate kinetic model for 975°F⁺ conversion was developed. Preliminary analysis of the model parameters indicates that it may be possible to relate them to feedstock properties including coal rank, coal hydrogen content, aromaticity of the oil feed and metals content of the oil feed.
- low/high reactor temperature staging was identified as a potentially attractive process improvement. Temperature staging improved the selectivity to liquid products and product quality. Carbon loading and H/C atomic ratio on the recovered catalyst both indicated the improved hydrogenation atmosphere and reduced catalyst deactivation rate.
- the bench-scale simulation of catalyst cascading was very successful. About the same level of 975°F⁺ conversion, coal conversion and HDS was obtained compared to operation with all fresh catalyst. The only noticeable decrease in performance was HDN. The cascading operation offers the potential to reduce

- catalyst replacement cost by up to 50%.
- co-processing was demonstrated using coal cleaned via oil agglomeration. This represents a logical combination of process steps and would be very attractive for co-processing of poor quality coals.
- use of dispersed iron additive in HRI's Coal/Oil Co-Processing Technology is not attractive. The dispersed iron catalyst is not very active at the relatively low temperatures employed in the ebullated-bed.
- recycle of atmospheric bottoms with its residuum content enhanced by high severity atmospheric distillation was not effective in increasing 975°F+ conversion. To be effective a more traditional vacuum bottoms recycle would be required, with a higher concentration of unconverted 975°F+ liquid.

The operations with Texas Lignite and Maya residuum, and New Mexico sub-bituminous coal and Hondo residuum gave further implicit evidence of the synergy which exists on 975°F+ conversion in co-processing. The operation in Bench Run No. 9, which varied coal concentration in once-through operation, yielded much higher conversions than would be expected if the feedstock reactivities were merely additive. For example, if the conversions were additive, 975°F+ conversion, with 40 W% coal feed would be expected to be about 84 W% MAF. Actual data showed 975°F+ conversion at 40 W% dry coal feed to be 90 W% MAF.

In Bench Run No. 6, with New Mexico sub-bituminous coal and Hondo residuum, very high 975°F+ conversion was obtained in the first operating condition (93 W% MAF). The remaining 975°F+ liquid product contained 0.07 W% sulfur. The feed oil, Hondo residuum contained well over 5 W% sulfur. In oil only operation, at this high conversion operation, the sulfur content of the unconverted residuum would be much higher, at least as high as the feed sulfur content. The only possible explanation for this performance observed in co-processing is essentially complete conversion of the oil feed to distillable liquid products.

The results of this program have been very successful in extending HRI's data base for a number of feedstock combinations in co-processing and providing the groundwork for determining the dependence of the measured levels of process performance on changes in fundamental feedstock characteristics. A number of opportunities have also been identified for improvement of the existing catalyst system through catalyst cascading, temperature staging and selective utilization of coal feedstocks.

Section IV - Laboratory Support

Alberta Research Council has devised an autoclave procedure which permits the screening of feedstocks and certain operating conditions under simulated single stage ebullated bed conditions.

For this program, an autoclave test program was designed to study the effects of:

- coal/oil agglomerates (deashed coal) as co-processing feedstocks,
- iron-based additives, and
- catalyst cascading - screening tests,

on process performance, as measured by coal conversion and 975°F+ conversion.

It is observed that raw coal and deashed coal agglomerates have similar performance. Mild thermal and steam stripping treatments of agglomerates have no detrimental effect on process performance. Also, Cold Lake resid and Athabasca bitumen behavior is similar. Based on twelve screening tests using coal agglomerates, a large batch of bulk agglomerates (80 kg) was prepared, and shipped to HRI at Lawrenceville, NJ, for bench unit evaluation. Tests were also performed to ensure the processibility of bulk agglomerates and to confirm their suitability as a feedstock for continuous flow of the bench unit test. There were not significant differences between the bulk agglomerates and untreated small batch agglomerates.

The effects of iron oxide (Fe_2O_3) and iron sulfide were evaluated at two loadings (2 and 5 W% daf feed). The coal conversion for the Forestburg coal/Cold Lake resid and Texas lignite/Maya crude was unchanged in the presence of additives as compared to the tests with just the supported catalyst. The Forestburg coal conversion appeared slightly better than that for Texas Lignite/Maya but, the differences were statistically insignificant. At high Fe_2O_3 (5%) concentration, there was a negative impact on 975°F+ conversion as observed for both feedstock pairs. Depression in liquid yield and hydrogen sulfide evolution were also noticed.

Activity of the fresh and spent catalyst were evaluated in a 1.6 cm i.d. bench-scale continuous flow trickle bed reactor system at the Alberta Research Council. Selection of the operating ranges were based on the Brox-Behnken experimental design matrix. Catalyst activity was severely curtailed for both the first and second stage catalysts (see below). Nitrogen removal was reduced to an almost constant level, whereas sulfur removal and asphaltene conversion were on an average greater for the second stage catalysts. The H/C ratio of the product liquid was improved slightly but was essentially constant for the first and second stage catalysts. The second stage catalysts therefore retained considerable activity for cracking reactions but somewhat reduced activity for those reactions which proceed via hydrogenation e.g. HDN. The

substantial residual activities of the second stage catalysts suggest that there would be a potential benefit if the spent second stage catalysts were transferred to the first stage.

Spent Catalyst Activity at High Severity Conditions

<u>Catalyst</u>	<u>Fresh</u>	<u>1A</u>	<u>1E</u>	<u>2C</u>	<u>2D</u>	<u>2E</u>	<u>2F</u>
HDS, %	98	47	63	77	70	73	73
HDN, %	86	28	24	20	23	28	20
Asphaltene, %	95	35	36	60	56	57	58
H/C Ratio	1.59	1.38	1.41	1.42	1.40	1.40	1.36

Spent first stage catalysts had lost about 2/3 of their surface area and pore volume. Second stage catalysts had even lower surface area, only 12-23% of the fresh catalyst. Deactivation of catalysts in coal liquefaction has been attributed to simultaneous pore more plugging by coke, and shell progressive pore mouth poisoning by metals. In spite of higher carbon content and much lower surface area, second stage catalysts showed better activities than the first stage catalysts. The first stage catalysts are exposed to more coal derived metals such as iron and titanium. This suggests that the loss in the activity of first stage catalysts is primarily due to the shell progressive pore mouth poisoning mechanism by these metals.

Section V - Conceptual Commercial Plant Design and Economics

As a part of this project, HRI has completed a conceptual commercial co-processing plant design and economics studies. The conceptual commercial plant design is based on process performance demonstrated in bench-scale operations at HRI on co-processing of Texas Lignite and Maya residuum. For this conceptual design, the weight ratio of oil to dry coal feedstock is 2:1. At this oil to coal ratio, no recycle slurry is required, and operations are performed on a once-through basis. The co-processing performance levels were set to obtain high 975°F+ conversion levels (88 W% MAF), high liquid yields and good quality products.

The co-processing unit capacity is based on providing two parallel reactor trains, each containing two commercial sized reactors. Based on this criteria, the total co-processing unit through-put is 4800 TPD, including 1584 TPD of dry Texas Lignite and 3216 TPD (17,490 BPD) of Maya residuum. The total erected cost estimated for this co-processing unit is \$270-million (1990-dollars, including a 25% contingency).

Economics of co-processing were assessed in two different ways. First, co-processing was evaluated in a complete, grass-roots, stand-alone plant, devoted entirely to co-processing. Coal and residuum are purchased, and all facilities necessary to produce finished quality products in an environmentally acceptable manner are specified. For this evaluation the total liquid product is 22,869 BPD or 4.8 barrels per ton of dry feed. The total erected cost of the facility is \$578-million (1990-dollars, including a 25% contingency). Net operating costs for the plant are \$17/barrel. Including the cost of capital and relating the finished co-processing products to the cost of crude, shows the equivalent crude cost for co-processing to be \$22-25/barrel.

In addition to evaluation of co-processing as a stand-alone plant, the economics of co-processing in an integrated refinery operation were evaluated. For this part of the study, three refinery cases were depicted, representing the transition in refining all light crude oil (West Texas Intermediate) to the addition of heavy crude oil (Maya) to a refinery, and finally the addition of coal and co-processing. Following is a summary of the refinery cases:

Case	<u>1</u>	<u>2</u>	<u>3</u>
Description	Light Oil	Heavy Oil	Co-Processing
<u>Crude Oil, BPD</u>			
West Texas Intermediate	100,000	50,843	50,843
Maya	--	49,157	49,157
Coal, TPD	--	--	1,584
<u>Refined Products, BPD</u>	104,202	103,657	110,190
B/B Crude Oil	1.04	1.04	1.10
Total Erected Cost, \$/MM	265	720	920
<u>Net Operating Cost, \$/B</u>			
Crude Oil	19.19	16.92	15.92
Refining Cost	<u>1.97</u>	<u>3.10</u>	<u>3.95</u>
TOTAL	21.16	20.02	19.87

Crude oil prices shown above are based on average 1989 values for West Texas Intermediate (\$20/Barrel) and Maya (\$15/Barrel). With these prices, the co-processing option has the lowest net operating cost of \$19.87/barrel. When the cost of capital is considered, the light crude oil price required for the co-processing option to be the lowest cost option increases. If the cost of capital is inexpensive, as it can be with a highly leveraged utility financing basis, co-processing can be the lowest cost option at relatively low light oil prices, about \$25/barrel. Without leveraging, or other incentives, the light crude oil price required for co-processing to have the lowest product cost can be as high as \$30/barrel.

In evaluating the economics of co-processing, it is clear that the cost of capital is still a very significant component in the total product cost. Research efforts are in progress and should continue to reduce the capital cost per barrel of capacity for coal liquefaction. Co-Processing, however, has brought the economics of direct coal liquefaction close to economic levels. With low cost capital, the calculated equivalent crude costs for co-processing in the low \$20's/barrel, compares favorably with average 1989 prices of \$20/barrel for West Texas Intermediate. Private industry could be willing to accept the risks in commercializing co-processing if the mechanisms were in place to provide low cost capital (investment tax credits, accelerated depreciation, etc.).

Section VI - Recommendations for Future Work

Based on the successes of this program, following are recommendations for future work:

- work should be performed to further develop and optimize reactor temperature staging operations. This work offers many benefits in terms of improved selectivity to liquid products, improved product quality and reduced catalyst deactivation rate. This mode of operation also offers some very specific process design and thermal efficiency advantages in utilizing the exothermic heat of reaction to minimize preheat and quench requirements. It may also be possible to increase capacities per reaction train through improved hydrogen management practices and/or interstage separation.
- the work relating reactivity to feedstock properties is very encouraging. Data on more feedstocks is required to improve the statistical significance of the preliminary relationships developed. Specifically, more data is required on higher rank (bituminous), high sulfur coals.
- testing of different coal concentrations in once-through operation provided very dramatic results on Texas Lignite and Maya residuum. The conversion model developed suggests that this result can vary depending on the feedstocks selected. Further data is required to show this explicitly. In addition, as coal is a lower cost feedstock than oil, optimization of coal loading is important in maximizing utilization of low cost feedstocks.
- in parallel with this program Consolidation Coal Company's Research and Development Department has been developing stable carbon isotope analysis as a technique to distinguish between coal and oil derived products from co-processing. This data needs to be assessed from a modelling perspective to further improve the existing model and to enhance fundamental understanding of process chemistry.
- in this program only limited characterization of liquid products was performed to identify differences in quality as a function of the different feedstocks. More detailed characterization and refining testing is required to define the ultimate utilization and integration of co-processing in U.S. refineries. This is an especially important activity in light of recent environmental legislation regarding fuel products (Clean Air Act).
- the processing characteristics and options need to be clearly defined and evaluated for the vacuum bottoms from co-processing. As conversion processes are designed to transform undesirable fuel products to high value fuels, this tends to produce a more concentrated form of undesirable fuel. Effective, low-cost and environmentally acceptable utilization of the

END

DATE
FILMED

4 / 1 / 93