COAL LIQUEFACTION/RESID HYDROCRACKING VIA TWO-STAGE INTEGRATED CO-PROCESSING

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BACKGROUND

Lummus Crest Inc. (LCI), a subsidiary of Combustion Engineering Inc., has been developing technology for the simultaneous processing of coal and heavy petroleum liquids under a joint development contract with the U. S. Department of Energy. The LCI co-processing route is an outgrowth of its Integrated Two-Stage Liquefaction (ITSL) technology developed over the past decade by LCI for coal liquefaction. A 33-month R&D contract was initiated in October 1984 with the objective of determining the technical and economic feasibility of coal liquefaction via the LCI co-processing route.

The project was formulated into five major program tasks as follows:

- Task 1: Project Management Plan
- Task 2: Feedstock Analysis
- Task 3: Co-Processing Reactivity Screening
- Task 4: Continuous Bench-Scale Operations
- Task 5: Cost Estimate of Conceptual Commercial Facility

The first three tasks have been completed and the continuous Bench-Scale Operations task has recently been initiated. The balance of this paper will describe experimental methods, the LCI co-processing approach and the results of recent bench-scale unit operations.

SOME JUSTIFICATIONS FOR CO-PROCESSING

Since co-processing inherently requires two separate feedstocks, namely coal and petroleum resid, it is possible to assess any potential process advantages from two viewpoints. From the refiner's viewpoint, the aromatic-rich, coal-derived extracts, being well known hydrogen donor solvents, can improve the hydroprocessing conversion of heavy, low grade petroleum feedstocks. On a constant energy cost basis, the syncrude cost contribution from a coal feedstock may be less than that from a petroleum feedstock.

For example, if one assumes a typical net syncrude yield of 0.0005 M^3/Kg (3.0 bbl/ton) for a run-of-mine bituminous coal priced at \$0.033/Kg (\$30/ton), then the coal feedstock cost of the coal syncrude is about \$62/M^3 (\$10.00/bbl). This compares to petroleum crude prices, even under the current suppressed spot market, in excess of \$74-100/M^3 (\$12-16/bbl). The situation is even more pronounced in the case of a typical subbituminous coal. Although the net yield of liquids from subbituminous coal is lower than that from bituminous coal (0.0003 M^3/Kg vs. 0.0005 M^3/Kg for bituminous), the corresponding subbituminous R.O.M. coal cost is not proportionately lower but rather about 73 percent lower than that of the bituminous coal (\$0.0088/Kg vs. \$0.033/Kg). This translates to a subbituminous coal feedstock cost of the coal liquids of about \$32.6/M^3 (\$5.20/bbl). In both cases, it is envisioned that a relatively low level of coal-derived liquids would be blended with petroleum resid feedstock so as not to greatly alter the downstream refinery processability of the petroleum-coal liquid mixtures.

From a coal liquefaction plant owner's viewpoint, the introduction of petroleum resid allows for a reduction in the large and costly solvent recycle systems. Additional advantages include:

- o Reduces net hydrogen consumption and correspondingly reduced hydrogen production costs;
- o Avoids the need for a costly deashing step;
- o Provides for a more rapid introduction of coal into the domestic energy networks; and
- o Allows the consideration of smaller-scale, less capital intensive plant sizes in an over-the-fence concept rather than in a grass-roots, mega-project concept.

LCI CO-PROCESSING APPROACH

Of the three key co-processing routes - thermal, thermocatalytic, biochemical - the LCI approach represents a hybrid of the first two in that it consists of a two-stage method. The first-stage is a thermal reaction system paralleling the Short Contact Time (SCT) reaction system developed for LCI's ITSL Process. The second-stage consists of a catalytic reaction system based on LCI's proprietary expanded-bed technology known as LC-Fining[sm]. The SCT reactor is close-coupled to the LC-Fining[sm] reactor to allow for rapid stabilization of coal extracts by the LC-Fining[sm] catalyst thereby minimizing undesirable free radical condensation reactions.

Figures 1 and 2 show two alternative flowschemes depending upon the source and type of the petroleum resid. The scheme shown in Figure 1 is predicated on the use of a heavy refinery stream such as the unconverted resid from a catalytic hydrocracker. In this scheme, the petroleum feedstock is blended with the coal and a recycle gas oil stream prior to the first-stage, SCT thermal reactor.

The scheme shown in Figure 2 is predicated on the use of a virgin vacuum residua which is fed directly into the LC-Finer[sm] along with the SCT coal extract. The first-stage reactor of the LC-Finer[sm] can be operated to simultaneously optimize the production of a) a donor solvent-rich gas oil recycle stream; b) an unconverted but hydrotreated recycle resid stream having improved solvency for coal; and c) hydrocracked C_5-524°C (C_5-975°F) distillates.

EXPERIMENTAL APPROACH

The experimental approach to obtaining key process data required for the preliminary design and estimate of a conceptual commercial facility has been accomplished in a variety of test units. Initial work for screening candidate coal and petroleum feedstocks was carried out in microautoclave reaction systems shown schematically in Figure 3. This was followed by testing in a continuous, close-coupled test unit under once-through conditions utilizing solvents characteristic in composition to what is expected at steady-state, but synthetically generated. The test unit, shown schematically in Figure 4, consists of an SCT reaction system comprised of a 6.2 mm i.d. by 343 cm long horizontal coil heater followed by a vertical SCT reactor having a volume of 118 cc. The SCT

(sm) LC-Fining is a service mark of Lummus Crest Inc. for engineering, marketing and technical services related to hydrocracking and hydrodesulfurization processes for reduced crude and residual oils.

system, which could be operated with either SCT reactors described above, was close-coupled to a stirred autoclave catalytic hydrocracker. The continuous bench-scale unit being operated in Task 4 consists of the above SCT reactor systems close-coupled to a small diameter, expanded-bed LC-Fining^{sm} reactor system. This unit can operate in a recycle mode which consists of batch collection of hydrotreated liquids from the LC-Finer^{sm} followed by batch slurrying of the coal and the thus collected recycle liquids. Both SCT and LC-Finer^{sm} reactors are operated continuously.

RESULTS OF BENCH-SCALE TESTING

Process variables studies, carried out in the continuous close-coupled test units, have recently been completed. The bulk of the test scans concentrated on scouting of both SCT and LC-Fining^{sm} reaction conditions close to that of the anticipated region of preferred commercial operations. Tests were made with two coal-petroleum combinations that were selected from the analysis of the microautoclave test data. These combinations consisted of a bituminous coal - Pittsburgh seam with Athabasca vacuum resid - and a subbituminous coal - Wyodak with Arab Heavy vacuum resid. During the scans, two sets of data were obtained indicating the effect of LC-Fining^{sm} reactor temperature and solvent/coal ratio on co-processing performance.

EFFECT OF TEMPERATURE

The impact of LC-Fining^{sm} temperature at constant SCT reaction conditions is shown in Table 1. From these data, we have calculated pseudo-kinetic rate constants for each of the performance criteria based on simplified kinetic models. These values are indicated below:

Co-Processing (SCT & LC-F) Performance Parameter	Pseudo-Rate Constant, hr^{-1}		
	404C	416C	432C
Desulfurization	0.75	1.09	1.67
Demetallization	0.55	0.90	1.12
524C+ Conversion	0.54	0.72	1.11
Denitrogenation	0.29	0.43	0.90

The feedstock in all tests consisted of 25 percent coal/37.5 percent hydrotreated petroleum resid/37.5 percent coal-derived gas oil. The order of reactivity as measured by the specific co-processing performance parameters in descending order is:

Desulfurization > Demetallization > Conversion > Denitrogenation

It is also interesting to note that in all three tests, the preasphaltene content in the product of the single-stage LC-Finer^{sm} has been reduced to less than 2 percent.

EFFECT OF SOLVENT QUALITY AND SOLVENT/COAL RATIO

The impact of solvent/coal ratio and solvent quality at constant SCT and LC-Fining^{sm} reaction conditions is shown in Table 2. The following interesting observations have been made based on the data shown:

- o At constant coal slurry concentration, reducing the ratio of the solvent (524^{o}C- boiling range) to coal from 1.5/1 to 1.0/1 had a minimal effect on observed coal conversions.

- However, the reduction in the said solvent/coal ratio did adversely affect syncrude characteristics and net distillate yields.

It should be kept in mind that the high LC-Fining[sm] severity used in this particular test campaign was aimed at assessing per pass performance (e.g., conversion, desulfurization, etc.) limits during co-processing. In the conceptual commercial concept depicted in Figures 1 and 2, it is anticipated that the first-stage LC-Finer[sm] located immediately downstream of the SCT reactor will be operated under optimal conditions for generating recycle gas oil solvents having high hydrogen donor capacity. At these severities, it is anticipated that the 524^{o}C+ conversions would be lower than the values shown in Table 4. The additional required feed conversions, i.e., overall conversions in excess of 80 percent, could be achieved in a second-stage LC-Finer[sm] operated at higher, more conventional petroleum hydrocracking severities.

In test BSCL-20, a 524^{o}C- recycle solvent during co-processing was simulated by blending a neat coal-derived hydrogenated solvent with a neat petroleum-derived hydrogenated solvent in the same ratio as that of the coal and petroleum resid fed in that run. By comparing these results to those of test BSCL-9 made only with coal-derived 524^{o}C- solvent, it is possible to estimate the relative solvent quality index (SQI) of the petroleum-derived gas oil solvent in comparison to that of the coal-derived gas oil solvent. The SQI of the particular petroleum gas oil utilized in this test has been estimated to be about 60 percent of that of the coal-derived solvent based on relative solvent performance as measured by observed net distillate yields. Optimization of the petroleum resid hydrocracking step in which the simulated gas oil solvent was generated has the potential to increase the latter's SQI value closer to that of the coal-based solvent.

FUTURE WORK

The continuous bench-scale unit will be operated in the recycle mode to demonstrate the effect of solvent maintenance on co-processing performance as a function of feedstock types (Pittsburgh seam and Wyodak coals; Arab Heavy and Athabasca residua); coal/resid ratios; solvent/coal ratio and catalyst age. These data will serve as the basis for formulating a conceptual commercial plant design. Towards the end of the Task 4 experimental program, a catalyst life test at preferred co-processing conditions will be made to demonstrate the technical feasibility of the base case design.

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TABLE 1

EFFECT OF LC-FINING[SM] TEMPERATURE ON CLOSE-COUPLED CO-PROCESSING PERFORMANCE

SUMMARY OF TEST CONDITIONS AND RESULTS

Run No. (BSCL-)	9	10	11
I. Test Conditions			
SCT Temperature, ^{o}C (^{o}F)	449(840)	449(840)	449(840)
LC-Fining[sm] Temperature, ^{o}C (^{o}F)	432(810)	416(780)	404(760)
Coal Space Velocity to SCT, Kg/Hr/M^3	3400	3400	3400
Solvent/Coal Wt. Ratio	1.5	1.5	1.5
Resid/Coal Wt. Ratio	1.5	1.5	1.5
II. Test Results			
Coal Conversion, Wt.% MAF	96.0	93.8	93.7
524^{o}C+ (975F+) Conversion, Wt.%	75.8	67.3	58.4
Net Distillate Yield, Kg 524^{o}C-/100 Kg 524^{o}C+	58.1	55.9	48.3
Desulfurization, %	82.5	75.7	66.0
Denitrogenation, %	71.7	55.0	43.0
Resid Demetallization, %	76.0	72.0	59.0
Preasphaltenes Concentration, %	1.7	1.5	1.9

(1) Feedstocks: Pittsburgh seam coal; Prehydrotreated 524^{o}C+ Athabasca resid

(2) Solvent: 524^{o}C- gas oil characteristic of a coal-derived recycle solvent produced in LCI's ITSL PDU during Wyodak coal operations

(3) SCT Condition: 137 atm; 360 M^3 H_2/M^3 feed

(4) LC-Fining[sm] Conditions: Shell 324M catalyst; 137 atm; 0.4R hr^{-1}; 530 M^3 H_2/M^3 feed

TABLE 2

EFFECT OF SOLVENT QUALITY AND SOLVENT/COAL RATIO ON CLOSE-COUPLED CO-PROCESSING PERFORMANCE

SUMMARY OF TEST CONDITIONS AND RESULTS

Run No. (BSCL-)	9	21	20
I. Test Conditions			
Solvent/Coal Wt. Ratio	1.5	1.0	1.5
Coal/Resid Wt. Ratio	0.67	0.50	0.67
Coal Slurry Concentration, Wt.%	25.0	25.0	25.0
524^{o}C- Solvent Composition, Wt.%			
ITSL*	100	100	40
Petroleum Gas Oil**	0	0	60
SCT Temperature, ^{o}C (^{o}F)	449(840)	449(840)	449(840)
LC-Fining^{sm} Temperature, ^{o}C (^{o}F)	432(810)	432(810)	432(810)
Coal Space Velocity to SCT, Kg/Hr/M^3	3400	3400	3400
II. Test Results			
Coal Conversion, Wt.% MAF	96.0	93.4	94.2
524^{o}C+ (975F+) Conversion, Wt.%	75.8	66.2	64.2
Net Distillate Yield, Kg 524^{o}C-/100 Kg 524^{o}C+	58.1	51.0	46.0
Desulfurization, %	82.5	75.0	77.2
Denitrogenation, %	71.7	65.0	67.1
Resid Demetallization, %	76.0	69.0	71.0
Preasphaltenes Concentration, %	1.7	2.2	2.0

* 524^{o}C- gas oil characteristic of a coal-derived recycle solvent produced in LCI's ITSL PDU during Wyodak coal operations.

** 524^{o}C- gas oil characteristic of a petroleum-derived recycle solvent produced during LC-Fining^{sm} of virgin Athabasca bitumen.

1. Feedstocks: Pittsburgh seam coal; prehydrotreated 524^{o}C+ Athabasca resid
2. SCT Conditions: 137 atm; 360 M^3 H_2/M^3 feed
3. LC-Fining^{sm} Conditions: Shell 324M catalyst; 137 atm; 0.4R Hr^{-1}; 530 M^3 H_2/M^3 feed

Figure 1. SCHEMATIC OF LCI CO-PROCESSING CONCEPT WITH HYDROCRACKED PETROLEUM RESIDUA

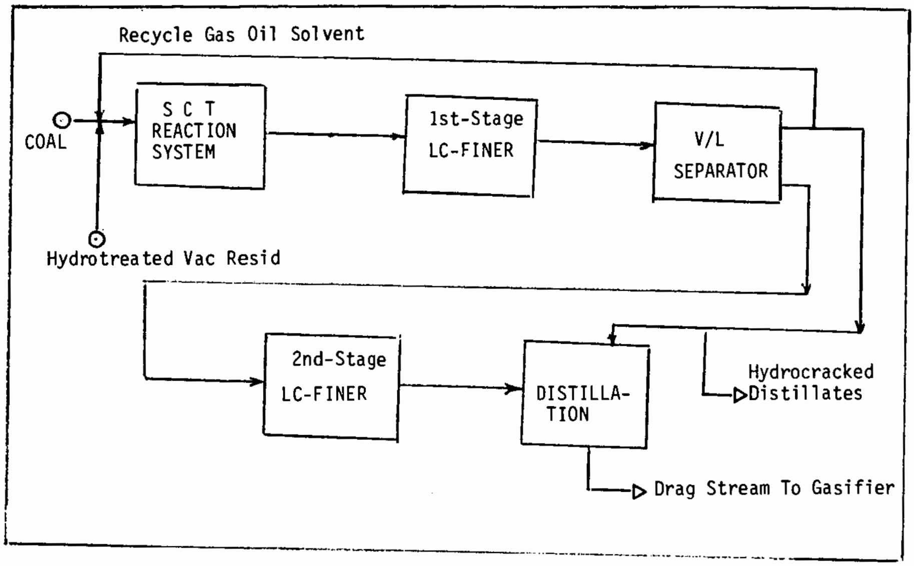


Figure 2. SCHEMATIC OF LCI CO-PROCESSING CONCEPT WITH VIRGIN PETROLEUM RESIDUA

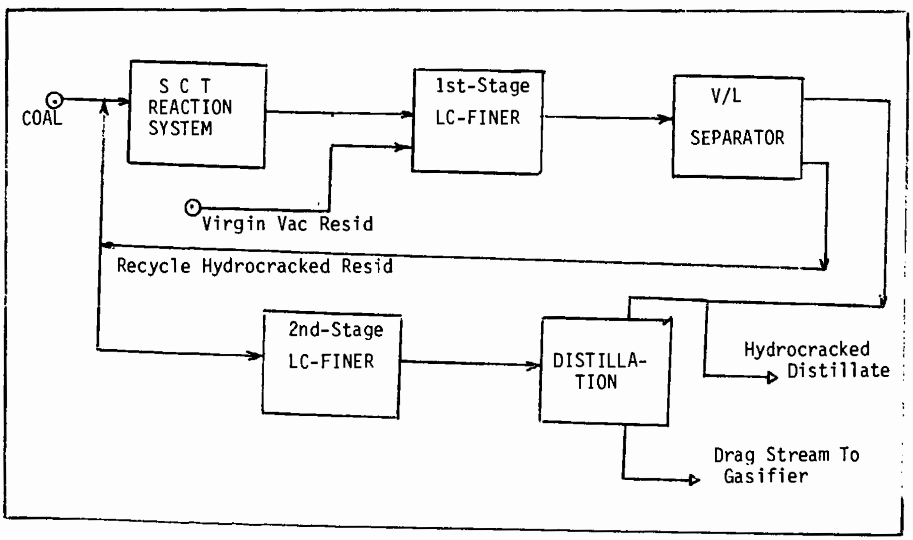
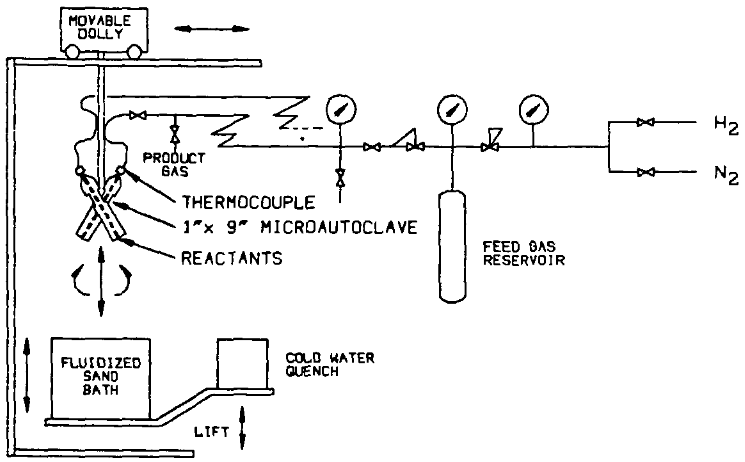


FIGURE 3
SCHEMATIC OF MICROAUTOCLAVE TEST UNIT



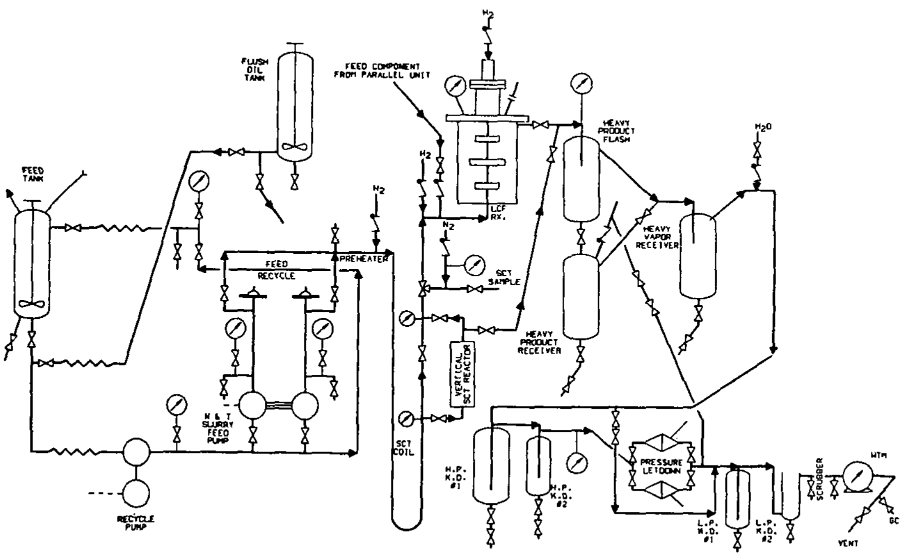


FIGURE 4 SCHEMATIC OF LCI'S CONTINUOUS CO-PROCESSING TEST UNIT