

DIRECT LIQUEFACTION OF LOW-RANK COAL

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DIRECT LIQUEFACTION OF LOW-RANK COAL

EXECUTIVE SUMMARY

A multistep direct liquefaction process specifically aimed at low-rank coals (LRCs) has been developed at the Energy & Environmental Research Center (EERC). The process consists of a preconversion treatment to prepare the coal for solubilization, solubilization of the coal in the solvent, and polishing using a phenolic solvent or solvent blend to complete solubilization of the remaining material. The product of these three steps can then be upgraded during a traditional hydrogenation step.

This project addresses two research questions necessary for the further development and scaleup of this process: 1) determination of the recyclability of the solvent used during solubilization and 2) determination of the minimum severity required for effective hydrotreatment of the liquid product. The project is being performed as two tasks, the first consisting of ten recycle tests and the second consisting of twelve hydrotreatment tests performed at various conditions.

Several activities were performed during this quarter.

- A paper entitled "Solvent Recyclability in a Multistep Direct Liquefaction Process" was presented at the 1995 Coal Liquefaction and Gas Conversion Contractors' Review Conference that was held in Pittsburgh, PA, August 29-31, 1995.
- The Task 1 solvent recyclability tests were completed.
- The Task 1 quality assurance/quality control checks were performed.
- The first seven Task 2 hydrotreatability tests were completed.

Analysis of the Task 1 data indicates that 1) the multistep process produces adequate quantities of excess solvent for recycle and 2) the product slates of all of the tests were fairly consistent.

DIRECT LIQUEFACTION OF LOW-RANK COAL

1.0 INTRODUCTION

Direct liquefaction research at the Energy & Environmental Research Center (EERC) has, for a number of years, concentrated on developing a direct liquefaction process specifically for low-rank coals (LRCs) through the use of hydrogen-donating solvents and solvents similar to coal-derived liquids, the water-gas shift reaction, and lower-severity reaction conditions. The underlying assumption of all of the research was that advantage could be taken of the reactivity and specific qualities of LRCs to produce a tetrahydrofuran (THF)-soluble material that might be easier to upgrade than the soluble residuum produced during direct liquefaction of high-rank coals. A multistep approach was taken to produce the THF-soluble material, consisting of the following: 1) preconversion treatment to prepare the coal for solubilization; 2) solubilization of the coal in the solvent; and, 3) polishing to complete solubilization of the remaining material. The product of these three steps can then be upgraded during a traditional hydrogenation step.

To provide a preliminary comparison between the EERC process and existing direct liquefaction processes, product slurry produced during solubilization (Step 2) and polishing (Step 3) (i.e., without the Step 1 pretreatment) was catalytically hydrotreated to equilibrium, based upon hydrogen uptake. The hydrotreatment was performed in this manner to define the practical upper limit of the product's hydrotreatability. The results were positive, and further tests were performed incorporating the pretreatment step (Step 1). Steps 1 through 3 (pretreatment, solubilization, and polishing) were performed in an integrated fashion. The products were catalytically hydrotreated to demonstrate the maximum hydrotreatability of the solubilized slurry and to provide products that could be compared to the products of existing processes.

The results of the EERC's research indicated that additional studies to more fully develop this process were justified. Two areas were targeted for further research: 1) determination of the recyclability of the solvent used during solubilization, and 2) determination of the minimum severity required for hydrotreatment of the liquid product. This project addresses these two areas.

2.0 GOALS AND OBJECTIVES

The project goals are to determine the recyclability of the solvent used during solubilization and to determine the minimum hydrotreatment severity required to upgrade the liquid product of the multistep EERC process.

The project is being performed as two tasks. The first task consisted of ten recycle tests. Lignite was solubilized via the pretreatment, solubilization, and polishing steps. The product of these three steps was combined with a vehicle solvent and the resulting stream distilled to remove water, solubilization solvent, and oxygenated light, coal-derived liquids. The overheads were further distilled to separate the water and light oil streams. The light oil was then recycled for use as the solubilization solvent in the next test sequence. The analyses of the products of these tests are used to characterize the recycle solvent stream and to calculate mass and material balances.

The second task consists of a series of twelve hydrotreatment tests at various conditions. The tests are being performed according to a statistically designed experimental matrix to enable the identification and evaluation of the most effective low-severity hydrotreatment conditions. Analyses of the products of these tests are used to characterize the hydrotreated product and to calculate mass and material balances.

3.0 ACCOMPLISHMENTS

3.1 Work Planned for This Quarter

The project has been extended; originally, no work was planned for this quarter.

3.1.1 Task 1 Operations

During the July–September quarter, four runs were made and two quality assurance/quality control (QA/QC) check samples were prepared as part of the Task 1 experimental matrix. These runs are summarized in Tables 1 and 2 and are discussed in more detail in the following paragraphs.

3.1.1.1 Test 9

Test 9 was performed as Runs N630, N631, and N632. (Runs N630 and N631, performed during the April–June quarter, are discussed here as part of Test 9.) During Run N630, approximately 0.214 kg of moisture- and ash-free (maf) Freedom lignite was slurried with 0.424 kg of the recycle solvent obtained from the Run N629 distillation. The autoclave was charged with slurry, CO, and H₂S, which served as the reaction promoter. The pretreatment was performed at 424 K (151°C) and 1.02×10^7 N/m² (1477 psig) for 30 min. The conditions were then increased to 647 K (374°C) and 2.25×10^7 N/m² (3267 psig) for 60 min. for the solubilization step. The unit was quenched and the product recovered and sampled. The overall material balance for the run was 99.8%.

The N630 product slurry was polished during Run N631. The product slurry was charged to a preheated autoclave containing 0.105 kg of recycle solvent and H₂. The polishing step was performed at average conditions of 713 K (440°C) and 2.59×10^7 N/m² (3761 psig) for 20 min. The reaction was quenched and the product recovered and sampled. The polishing step overall material balance was 98.2%.

The polished product slurry from N631 was combined with 0.47 kg HAO61 heavy fraction and distilled during Run N632. Water was removed from the organics and the distillation performed at the end point conditions of 7.6×10^3 N/m² (1.1 psig) and 495 K (222°C) pot temperature to separate the light oil fraction from the heavier fraction. The light oil fraction was recycled for use as the feed solvent for Test 10. An overall mass balance of 96.3% was achieved for the Test 9 distillation step.

TABLE 1

Summary of Tests Performed July Through September 1995

Run No.	Description	Mass Balance, %
N632	Task 1, Test 9 – Distillation of polished product	96.3
N633	Task 1, Test 10 – Pretreatment and solubilization steps	100.4
N634	Task 1, Test 10 – Polishing step	98.8
N635	Task 1 – QA/QC liquid product check sample	NA ^a
N636	Task 1 – QA/QC feed slurry check sample	NA
N637	Task 1, Test 10 – Distillation of polished product	97.1
N638	Task 2, Test 1 – Hydrotreatment feed preparation	NA
N639	Task 2 – Catalyst preparation	99.0
N640	Task 2 – Catalyst preparation	96.4
N641	Task 2, Test 2	96.9
N642	Task 2, Test 3	95.2
N643	Task 2 – Catalyst preparation (possible feed gas contamination)	NA
N644	Task 2 – Catalyst preparation	96.1
N645	Task 2, Test 4	97.9
N646	Task 2, Test 5	98.3
N647	Task 2 – Catalyst preparation	96.4
N648	Task 2, Test 6 (thermocouple failed)	NA
N649	Task 2, Test 6	98.6
N650	Task 2, Test 7	99.9
N651	Task 2, Test 8 (stirrer leaked)	NA

^a Not applicable.

3.1.1.2 Test 10

Runs N633, N634, and N637 made up Test 10. The pretreatment and solubilization steps were performed as Run N633. Approximately 0.191 kg of maf Freedom lignite was slurried with 0.325 kg of recycle solvent obtained from the N632 distillation. The slurry was cold-charged to the autoclave with CO and H₂S, the reaction promoter. The pretreatment was performed at an average temperature of 424 K (151°C) and an average pressure of 1.01×10^7 N/m² (1471 psig) for 30 min. Solubilization was performed at 647 K (374°C) and 2.22×10^7 N/m² (3226 psig) for 60 min. After the unit was quenched, the product was recovered and sampled. The overall material balance for these two integrated steps was 100.4%.

TABLE 2

Run Conditions for Task 1, Tests 1 Through 10

Processing Step	1	2	3	4	5	6	7	8	9	10
Test Number										
Pretreatment	N602	N607	N611	N614	N617	N621	N624	N627	N630	N633
Temperature, K (°C)	425 (152)	426 (153)	424 (151)	425 (152)	422 (149)	423 (150)	424 (151)	421 (148)	424 (151)	424 (151)
Pressure, N/m ² × 10 ⁶ (psig)	9.67 (1402)	9.63 (1397)	9.74 (1412)	9.82 (1424)	10.05 (1458)	10.16 (1474)	10.07 (1461)	9.82 (1424)	10.18 (1477)	10.14 (1471)
Time, min	35	30	30	30	30	31	30	30	30	30
Gas	CO	CO	CO	CO	CO	CO	CO	CO	CO	CO
Additive	H ₂ S	H ₂ S	H ₂ S	H ₂ S	H ₂ S	H ₂ S	H ₂ S	H ₂ S	H ₂ S	H ₂ S
Solubilization	N602	N607	N611	N614	N617	N621	N624	N627	N630	N633
Temperature, K (°C)	650 (377)	650 (377)	646 (373)	645 (372)	645 (372)	647 (374)	647 (374)	647 (374)	647 (374)	647 (374)
Pressure, N/m ² × 10 ⁶ (psig)	23.98 (3478)	24.27 (3520)	24.55 (3560)	23.72 (3440)	23.38 (3391)	24.08 (3493)	23.78 (3449)	23.03 (3340)	22.53 (3267)	22.24 (3226)
Time, min	60	60	60	60	60	60	60	60	60	60
Polishing	N603	N609	N612	N615	N618	N622	N625	N628	N631	N634
Temperature, K (°C)	714 (441)	713 (440)	717 (434)	720 (437)	721 (438)	725 (442)	726 (443)	726 (443)	723 (440)	717 (434)
Pressure, N/m ² × 10 ⁶ (psig)	31.75 (4605)	31.30 (4540)	31.54 (4575)	29.61 (4294)	27.59 (4001)	28.99 (4204)	28.59 (4147)	27.49 (3987)	25.93 (3761)	24.30 (3525)
Time, min	20	20	20	20	20	20	20	20	20	20
Gas	H ₂	H ₂	H ₂	H ₂	H ₂	H ₂	H ₂	H ₂	H ₂	H ₂
Additive	Feed Solvent	RS-606	RS-610	RS-613	RS-616	RS-620	RS-623	RS-626	RS-629	RS-632
Distillation	N606	N610	N613	N616	N620	N623	N626	N629	N632	N637
Temp., pot, K (°C)	494 (221)	492 (219)	492 (219)	494 (221)	494 (221)	496 (223)	496 (223)	495 (222)	495 (222)	495 (222)
Pressure, N/m ² × 10 ⁶ (psig)	0.008 (1.19)	0.007 (1.10)	0.007 (1.10)	0.007 (1.09)	0.008 (1.10)	0.007 (1.09)	0.007 (1.09)	0.007 (1.09)	0.007 (1.10)	0.007 (1.08)

The N633 product slurry was charged to a preheated autoclave containing 0.097 kg recycle solvent and H₂ for Run N634. The average polishing conditions were 707 K (434°C) and 2.43×10^7 N/m² (3525 psig) for 20 min. The reaction was quenched and product recovered. An overall material balance of 98.8% was achieved for the polishing step.

The polished product slurry from N634 was combined with 0.43 kg HAO61 heavy fraction and distilled during Run N637. Water was removed from the organics and the distillation performed at the end point conditions of 495 K (222°C) pot temperature and 7.0×10^3 N/m² (1.08 psig) to separate the light oil fraction from the heavier fraction. An overall mass balance of 97.1% was achieved for the Test 10 distillation.

3.1.1.3 Task 1 Quality Assurance/Quality Control

Process measurement instrumentation was calibrated according to the schedule listed in the project QA/QC plan.

A technical systems audit (TSA) was performed. The TSA consisted of a thorough qualitative evaluation of the ability of the technical, operations, and analytical personnel and systems, including the chain-of-custody protocol and data transfer schemes, to produce high-quality data meeting the quality assurance objectives of mass and material balances of $\geq 95\%$. It was decided that the technical systems did produce high-quality data and were capable of flagging nonvalid results.

An audit of data quality (ADQ) was performed for each of the ten Task 1 tests. Each data point was examined to determine whether it was a feasible number within the range of analytical and/or system error. All of the data were judged to be feasible.

Each Task 1 test was then evaluated with respect to meeting the quality assurance objectives of mass and material balances of $\geq 95\%$. Because all of the Task 1 tests met these criteria, none of the tests was repeated.

Quality control check samples were prepared and analyzed to check the analytical data accuracy and precision. A feed slurry check sample was prepared from known quantities of certified-analysis cresylic acid solvent and as-received lignite. The as-prepared analysis, the accuracy requirements, and the as-analyzed results for this analysis are presented in Table 3. The feed slurry check sample met the accuracy criteria for all analyses except carbon. Because the other analyses were within the tolerance limits, the error was thought to be with the lignite carbon analysis used to calculate the as-prepared feed slurry composition. A second carbon-hydrogen-nitrogen analysis was performed for the coal, but the results were not available for the preparation of this report.

Two certified gas mixtures were submitted for gas chromatography (GC) analysis as gaseous product samples, N635 PG-1 and N635 PG-2. The as-analyzed results of these check samples are compared with the certified gas composition in Table 4. As the table shows, all analyses are within the $\pm 2\%$ accuracy limit. Finally, a sample of cresylic acid sample was submitted as the

TABLE 3

Analysis of Task 1 Feed Slurry Check Sample

Analysis	As Prepared, wt%	Accuracy	
		Requirements, wt%	As Analyzed, wt%
C	61.57	±0.5	62.93
H	7.08	±0.5	7.14
N	0.49	±0.5	0.62
S	0.20	±0.5	0.29
Ash	2.62	±1.0	2.39
H ₂ O	15.95	±1.39	15.63
THFI ^a	29.90	— ^b	33.31

^a THF insolubility analysis.

^b Cannot be calculated because of lack of accepted standard reference material.

liquid product check sample, N634. The as-analyzed results are compared to the cresylic acid analysis in Table 5. As Table 5 shows, the liquid product check sample also met the project accuracy criteria.

3.1.1.4 Discussion

The material balances for the Task 1 solvent recyclability tests are presented in Table 6. The recoveries for each of the steps are similar. The liquid balance for the pretreatment/solubilization step ranged from 90.4% to 91.7% and, for the polishing step, ranged from 95.9% to 97.7%. The overall mass balances for the pretreatment/solubilization step ranged from 96.0% to 100.4%, for the polishing step from 98.2% to 99.2%, and for the distillation step from 97.1% to 99.6%. The consistency of the mass balances is indicative of the operational stability of the system. Overall mass balances of at least 96.0% indicate that there were no leaks or spills that might skew the data. Therefore, the data reliably describe the process.

Table 7 shows the solvent yields for each of the Task 1 tests. As discussed in the April–June 1995 quarterly report, during the original distillation some of the distillable material from Test 3 was not collected. The distillation bottoms were redistilled and the additional material collected was added to the recycle solvent stream used in the Test 6 feed slurry. The table shows that as a result of the lower fraction of light distillate present in Tests 4 and 5, the solvent recovery dropped from approximately 15% excess solvent to about 5% excess solvent. Excess solvent was produced in each of the tests, with an average excess solvent production for all tests of 16.81 wt%. Excluding the low solvent balances for Tests 4 and 5, the average amount of excess solvent produced is 19.48 wt%.

The preliminary product yield structures of the ten Task 1 tests are shown in Table 8. The values are expressed in weight percentages equivalent to the quantity of maf lignite fed to the pretreatment/solubilization step of each test. Approximately 20 wt% of the maf lignite fed to the system went toward the production of gaseous products and water, while about 80 wt% of the maf lignite went toward the production of liquid products. The table shows that, in general, the product slates for each test are fairly consistent.

TABLE 4

Analysis of Task 1 Gas Product Check Samples^a

Analysis	Gas 1		Gas 2	
	N635 PG-1	Standard 1	N635 PG-2	Standard 2
Hydrogen			38.97	38.63
Carbon Dioxide	7.53	7.22	35.27	35.71
Propane	0.27	0.11		
Acetylene	0.15	0.20		
<i>iso</i> -Butane	0.10	0.10		
<i>n</i> -Butane	0.09	0.10		
Ethylene	0.18	0.18		
Ethane	0.30	0.31	0.96	0.97
Oxygen/Argon	0.40	0.35		
Nitrogen	84.24	84.83	15.60	15.59
Methane	6.65	6.50	0.94	0.90
Carbon Monoxide			8.26	8.20

^a All values are in mol%.

TABLE 5

Analysis of Task 1 Liquid Product Check Sample

Analysis	N634 Check Sample	Cresylic Acid	Accuracy
	Analysis, wt%	Analysis, wt%	Requirements, wt%
C	77.24	77.51	±0.5
H	7.66	7.52	±0.5
N	0.39	0.21	±0.5
S	0.0	0.0	±0.5
Ash	0.02	-- ^a	±1.0
H ₂ O	1.11	0.28	±1.39
THF ^b	0.03	--	-- ^c

^a Analysis not performed; sample is assumed to be ash-free and 100% THF-soluble.^b THF insolubility analysis.^c Cannot be calculated because of lack of accepted standard reference material.

TABLE 6

Mass and Material Balances for Task 1, Tests 1 Through 10

	Gas Balance, %	Liquid Balance, %	Overall Mass Balance, %
Test 1			
Pretreatment/Solubilization	122.6	91.4	96.0
Polishing	169.5	97.3	98.2
Distillation	NA ^a	NA	97.9
Test 2			
Pretreatment/Solubilization	135.8	91.7	98.6
Polishing	197.9	97.3	98.7
Distillation	NA	NA	98.7
Test 3			
Pretreatment/Solubilization	140.9	91.7	99.0
Polishing	205.6	97.7	99.2
Distillation	NA	NA	97.9
Test 4			
Pretreatment/Solubilization	132.1	91.6	98.7
Polishing	220.0	97.2	99.1
Distillation	NA	NA	98.1
Test 5			
Pretreatment/Solubilization	131.8	91.0	99.3
Polishing	196.1	97.2	99.0
Distillation	NA	NA	99.6
Test 6			
Pretreatment/Solubilization	138.9	90.8	99.8
Polishing	222.1	96.7	98.8
Distillation	NA	NA	97.2
Test 7			
Pretreatment/Solubilization	133.7	90.8	99.1
Polishing	219.4	96.7	98.8
Distillation	NA	NA	97.3
Test 8			
Pretreatment/Solubilization	133.8	90.5	99.7
Polishing	212.7	96.7	98.9
Distillation	NA	NA	97.4
Test 9			
Pretreatment/Solubilization	131.5	90.4	99.8
Polishing	210.6	95.9	98.2
Distillation	NA	NA	96.3
Test 10			
Pretreatment/Solubilization	129.8	90.7	100.4
Polishing	183.4	96.9	98.8
Distillation	NA	NA	97.1

^a Not applicable.

3.2 Work Performed During This Quarter

Three activities were performed during this quarter: 1) completion of the Task 1 tests; 2) completion of Tests 1 through 8 in Task 2; and 3) presentation of a paper at the Coal Liquefaction and Gas Conversion Contractors' Review conference held in Pittsburgh, PA, on August 29–31, 1995.

3.2.1 Task 2 Operations

Fourteen runs were made during the July–September quarter as part of Task 2. The Task 2 testing was performed according to a statistically designed experimental matrix. A full factorial matrix for three factors was chosen. In addition to the factorial matrix points, two tests were performed at center point conditions to test for system reproducibility and to provide a means by which to determine the linearity of the data. The Task 2 experimental matrix is given in Table 9.

3.2.1.1 Run N638 – Test 1, Hydrotreatment Feed Preparation

The liquid product from Task 1, Tests 1, 2, 4, and 6, were combined into a single sample to be used as the feedstock for the Task 2 tests. The composite sample was well mixed and sampled. The analysis of the composite feed is presented in Table 10.

3.2.1.2 Runs N639 and N640 – Catalyst Preparation

Shell 424 catalyst was activated using H_2S and H_2 at an average temperature of 698 K (425°C) and an average pressure of about $1.10 \times 10^7 \text{ N/m}^2$ (1600 psig) for 180 min.

TABLE 7

Distillate Yields and Solvent Balances for Task 1				
Hydrotreatable Solubles, wt% maf ^a			Solvent Yield, wt% maf	Solvent Balance, %
Test No.	Liquid Basis ^b	Gas Basis ^c		
1	79.52	87.02	42.01	116.42
2	71.23	86.49	68.17	127.75
3	86.00	86.08	46.27	118.57
4	86.02	85.00	13.67	105.47
5	82.61	83.08	17.12	106.80
6	83.35	79.63	49.53	120.00
7	79.57	83.83	39.52	115.84
8	72.86	79.86	44.88	118.07
9	75.90	79.44	45.03	117.96
10	81.60	81.69	47.20	121.22
Average	79.87	83.21	41.34	116.81

^a Weight percentage of maf coal fed to the system.

^b Yield calculated from liquid stream mass balance data.

^c Yield calculated by subtracting the gas yield from unity.

TABLE 8

Preliminary Yield Structures for Task 1 Solvent Recyclability Tests^a

	Test Number									
	1	2	3	4	5	6	7	8	9	10
<u>Gas Out</u>										
CO	-38.79	-39.06	-37.58	-38.91	-37.77	-36.87	-38.37	-37.99	-38.86	-35.13
H ₂	0.98	1.25	0.68	0.97	1.35	0.23	0.07	0.22	0.41	0.79
CO ₂	71.78	72.16	70.32	72.32	71.15	73.14	72.70	75.38	76.68	70.01
C1-C3	3.07	3.38	3.39	3.85	3.98	4.81	5.03	5.23	5.49	2.95
H ₂ S	-1.70	-1.71	-1.22	-0.76	0.06	0.38	-1.08	-0.73	-0.67	0.03
H ₂ O	-14.58	-9.68	-21.25	-22.83	-21.32	-23.53	-18.08	-14.24	-19.09	-19.03
Total Gas + Water	20.76	26.34	14.34	14.64	17.45	18.16	20.27	27.87	23.96	19.62
<u>Liquid Out</u>										
Liquid to HT Step ^b	79.52	71.23	86.00	86.02	82.61	83.35	79.57	72.86	75.90	81.60
Ash	-0.29	2.42	-0.36	-0.67	-0.06	-1.50	0.15	-0.72	0.14	-1.22
Total Liquids	79.23	73.65	85.64	85.35	82.55	81.85	79.72	72.14	76.04	80.38
Total, all products	99.99	99.99	99.98	99.99	100.00	100.01	99.99	100.01	100.00	100.00

^a Product slates given are based upon percentage of maf coal fed to the pretreatment/solubilization step. Positive values indicate production of a component; negative values indicate a consumption.

^b Liquid products that will be hydrotreated during the hydrotreatment (HT) step.

TABLE 9

Task 2 Experimental Matrix

Test No.	Temperature	Pressure	Residence Time, min
1	Multistep test to produce solubilized slurry for hydrotreatment		
2	698 K (425°C)	1.724×10^7 N/m ² (2500 psig)	73
3	718 K (445°C)	1.324×10^7 N/m ² (1920 psig)	112
4	698 K (425°C)	1.724×10^7 N/m ² (2500 psig)	73
5	718 K (445°C)	1.324×10^7 N/m ² (1920 psig)	34
6	678 K (405°C)	1.324×10^7 N/m ² (1920 psig)	112
7	678 K (405°C)	2.124×10^7 N/m ² (3080 psig)	34
8	718 K (445°C)	2.124×10^7 N/m ² (3080 psig)	112
9	678 K (405°C)	2.124×10^7 N/m ² (3080 psig)	112
10	678 K (405°C)	1.324×10^7 N/m ² (1920 psig)	34
11	718 K (445°C)	2.124×10^7 N/m ² (3080 psig)	34
12	Verification test at conditions indicated by statistical analysis		

TABLE 10

Analysis of Feedstock for Task 2 Hydrotreatment Severity Tests

Component	Analysis, wt%
C	87.85
H	6.42
N	1.08
S	0.40
Ash	2.83
THFI ^a	5.49

^a THF insolubles

3.2.1.3 Run N641 – Test 2

Approximately 0.225 kg of composite feed and 0.043 kg sulfided Shell 424 catalyst were charged to the 1-liter autoclave. Hydrotreatment took place at an average temperature of 698 K (425°C) and an average pressure of 1.67×10^7 N/m² (2419 psig) for 73 min. A liquid balance of 99.1%, a gas balance of about 44.3%, and an overall material balance of 96.9% were achieved for this test.

3.2.1.4 Run N642 – Test 3

Approximately 0.225 kg composite feed and 0.043 kg of sulfided Shell 424 catalyst were charged to the 1-liter autoclave. Hydrotreatment took place at an average temperature of 716 K (443°C) and an average pressure of 1.33×10^7 N/m² (1930 psig) for 112 min. A liquid balance of 97.8%, a gas balance of 44.9%, and an overall material balance of 95.2% were achieved for this test.

3.2.1.5 Run N643 – Catalyst Preparation

This test was to be another Shell 424 catalyst activation run. The feed gas may have been contaminated with nitrogen, so the test was disregarded.

3.2.1.6 Run N644 – Catalyst Preparation

Shell 424 catalyst was activated using H₂S and H₂ at an average temperature of 699 K (426°C) and an average pressure of about 1.15×10^7 N/m² (1664 psig) for 180 min.

3.2.1.7 Run N645 – Test 4

Approximately 0.219 kg composite feed and 0.044 kg of sulfided Shell 424 catalyst were charged to the 1-liter autoclave. Hydrotreatment took place at an average temperature of 698 K (425°C) and an average pressure of 1.71×10^7 N/m² (2485 psig) for 73 min. A liquid balance of 100.3%, a gas balance of 41.8%, and an overall material balance of 97.9% were achieved for this test.

3.2.1.8 Run N646 – Test 5

Approximately 0.217 kg composite feed and 0.043 kg sulfided Shell 424 catalyst were charged to the 1-liter autoclave. Hydrotreatment took place at an average temperature of 715 K (442°C) and an average pressure of 1.34×10^7 N/m² (1937 psig) for 34 min. A liquid balance of 100.0%, a gas balance of 39.1%, and an overall material balance of 98.3% were achieved for this test.

3.2.1.9 Run N647 – Catalyst Preparation

Shell 424 catalyst was activated using H₂S and H₂ at an average temperature of 699 K (426°C) and an average pressure of about 1.14×10^7 N/m² (1656 psig) for 180 min.

3.2.1.10 Run N648 – Test 6

A thermocouple failed during this test and the test was repeated as Run N649.

3.2.1.11 Run N649 – Test 6

Approximately 0.217 kg of composite feed and 0.043 kg of sulfided Shell 424 catalyst were charged to the 1-liter autoclave. Hydrotreatment took place at an average temperature of 679 K

(406°C) and an average pressure of 1.33×10^7 N/m² (1934 psig) for 112 min. A liquid balance of 99.9%, a gas balance of about 53.5%, and an overall material balance of 98.6% were achieved for this test.

3.2.1.12 Run N650 – Test 7

Approximately 0.213 kg of composite feed and 0.043 kg of sulfided Shell 424 catalyst were charged to the 1-liter autoclave. Hydrotreatment took place at an average temperature of 677 K (404°C) and an average pressure of 2.12×10^7 N/m² (3081 psig) for 34 min. A liquid balance of 102.2%, a gas balance of 47.4%, and an overall material balance of 99.9% were achieved for this test.

3.2.1.13 Run N651 – Test 8

The stirrer leaked during this test, resulting in a 78% liquid recovery. Because this value did not meet the quality assurance criterion of a material balance of ≥ 95 wt%, the test was repeated as Run N658.

3.2.2 Presentation of Contractors' Review Conference Paper

A paper entitled "Solvent Recyclability in a Multistep Direct Liquefaction Process" was presented at the Coal Liquefaction and Gas Conversion Contractors' Review Conference that was held August 29–31, 1995.

3.3 Conclusions

- The multistep process produces approximately 19 wt% excess solvent.
- The product slates of each multistep test were fairly consistent over the course of the recycle testing.

4.0 FUTURE OBJECTIVES

- Complete speciation of the liquid products of Task 1, Tests 1, 4, 7, and 10 will be completed. This will indicate whether the recycle solvent was beginning to line out.
- Tests 8 through 11 from Task 2 will be completed.
- The results from the Task 2 tests will be statistically analyzed, and the conditions of the verification test will be selected based upon the results of the analysis.
- Task 2, Test 12 (the verification test) will be performed.
- All quality assurance checks will be completed.
- The final technical report will be prepared.