

TECHNOECONOMIC ASSESSMENT OF ADVANCED CONCEPTS FOR DIRECT LIQUEFACTION OF SUBBITUMINOUS COALS

Michael Peluso
LDP Associates
32 Albert E. Bonacci Drive
Hamilton Square, N. J. 08690

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INTRODUCTION

For the past three years a group headed by the Center for Applied Energy Research of the University of Kentucky has been investigating, under DOE sponsorship, the use of Advanced Concepts for improving the Integrated Two Stage Liquefaction process practiced at the Wilsonville, Alabama pilot plant (1). Among the concepts tested at the batch scale level were: (1) extraction, dewaxing and hydrotreating of the distillate recycle solvent to improve coal conversion, (2) oil agglomeration of the feed coal to liquefaction to reduce its ash content and improve supported catalyst life, and (3) preparation and use of alternate dispersed and particulate catalysts to improve process performance and/or reduce dispersed catalyst cost.

BASE CASE ASSESSMENT - WILSONVILLE RUN# 263J

Wilsonville pilot plant operation with Black Thunder subbituminous coal was chosen as the basis for defining state-of-the art TSL technology. During Wilsonville Runs #262 & 263, the pilot plant unit was operated in the so-called hybrid mode with dispersed iron and molybdenum catalysts used in the first reaction stage and a supported nickel-moly catalyst used in the second stage ebullated bed reactor. Material balance period #263J was chosen as the basis for developing a baseline conceptual commercial plant case against which the results of this program could be compared (2). An all-distillate product Base Case was formulated in which resid extinction was achieved in the system via a reduction in reactor space velocities as predicted by first order reaction kinetics. All liquefaction distillates are assumed to be upgraded to a common basis (all-gasoline finished product) so that consistent comparisons are assured (3). The Base Case conceptual commercial plant is an all-coal facility located at a mine-mouth Wyoming location. The hydrogen needed for liquefaction is generated by water slurry gasification of ash concentrate from the ROSE unit and coal. Light hydrocarbon gases produced in liquefaction and upgrading are used to close the fuel gas balance. Any excess gas is used to generate hydrogen via the steam reforming process. It is assumed that the electricity needed to help operate the plant is purchased from a nearby utility power plant. A simplified block flow diagram of the conceptual commercial plant is shown in Figure 1. The plant converts 17,929 T/D of Black Thunder coal (MF basis) fed to liquefaction into 68,100 barrels per day (BPSD) of gasoline product. An additional 5,204 T/D of Black Thunder must be gasified in order to meet the plants hydrogen requirements. Overall MAF coal conversion for the Base Case is 92%. A high process solvent to coal ratio of 2.33 is employed since significant quantities of both IOM and ash are recirculated via the ashy recycle technique. Recycled ash is approximately 3.3 times the quantity of ash rejected from the process via the ash concentrate. As a result, the effective concentration of moly on coal to liquefaction is approximately 430 ppm at the Base Case fresh addition level of 100 ppm. Four liquefaction reactor trains in parallel are required to process the 17,929 T/D of coal to liquefaction. Reactor gas rates were determined based on the estimated average reactor partial pressures which existed during Wilsonville Run #263J and the recycle hydrogen gas purity. Actual reactor residence times and space velocities were also based on estimated WR#263J operation with appropriate corrections for the required resid plus IOM conversion level. Organic rejection (i.e. resid, IOM & DAS) from the liquefaction process amounts to 14.5% on an MF coal basis.

ADVANCED CONCEPTS CASE

Three main process variations from the Base Case are incorporated in the ACC. They are:

1. Coal impregnation with moly salt via incipient wetness.
2. Oil agglomeration of the feed coal at low pH, and
3. Distillate solvent quality improvement via solvent extraction, solvent dewaxing and hydrotreating. Used together these three techniques seek to significantly increase product yield per unit of coal feed while reducing solvent recycle rate and liquefaction system additive costs.

• **Coal Impregnation: Moly Salt via Incipient Wetness**

The cost of dispersed iron and moly catalysts in the Base Case accounts for almost \$3/Bbl. of gasoline product selling price. A significant portion of this cost results from the use of an expensive moly source, the oil soluble Molyvan L. A significant reduction in moly cost is achieved when a much cheaper moly salt such as Ammonium Octomolybdate is used. Even when processing costs for preparing the salt solution, impregnating a small portion of the feed coal to liquefaction and driving off the extra water added to the coal are added, the cost of the impregnated moly is still only about 25% of the cost of using Molyvan L. Experimental results indicate that performance with moly impregnated coals is approximately equivalent to the performance with Molyvan L. The cost of using moly for the ACC drops below the cost of using particulate iron oxide at the 1 wt.% on MF coal dosage level of the Base Case. Therefore, the use of iron is questionable and has not been included in the ACC.

• **Oil Agglomeration at Low pH**

Results indicate that the use of oil agglomeration at low pH can remove approximately 50% of the ash in Black thunder coal. Ash reduction at the front end of the liquefaction process reduces organic rejection at the back end of the process, thereby increasing product yield. It also reduces the ash recirculation rate within the process while still maintaining the same catalyst recycle enhancement factor as in the Base Case. At low pH, potential supported catalyst poisons, such as calcium, sodium, magnesium and potassium are also removed. For the ACC it has been assumed that the second stage reactor supported catalyst replacement rate can be reduced by 30%. Tests have indicated that impregnated iron and moly are retained on the coal during agglomeration. The oil agglomeration process is well suited for liquefaction. Distillate recycle solvent can be used as the agglomerating agent. Sour water can be used as makeup water to the system and the slurry reject of solids and dissolved salts can be utilized in the gasification slurry mixing systems. In fact, the dissolved salts may even act as a catalyst in the gasification process. A significant amount of sulfuric acid is consumed in the oil agglomeration unit.

• **Distillate Solvent Quality Improvement**

For the ACC, three process steps are used to treat the waxy distillate recycle solvent used in the Base Case. These processes are Solvent Extraction, Solvent Dewaxing and Hydrotreating. In combination these processes effectively remove and recover the waxy material from the distillate solvent and enhance its donor solvent capabilities (see Figure 3). Both solvent extraction and solvent dewaxing are commercial processes used in the petroleum refining industry.

The benefit of applying these three processes are:

1. Reduction of distillate solvent recycle while improving quality
2. Recovery of a valuable byproduct wax
3. Increased product yield via coal conversion improvement.

It is estimated that the wax yield on MAF coal is 4 wt.%. However, this wax builds up in the distillate recycle solvent until its cracking rate equals production rate. Based on Wilsonville Run#263J data the wax concentration in the distillate recycle solvent is estimated to be approximately 24 wt.%. Removal of a substantial portion of the wax, significantly reduces the distillate solvent recycle rate. The wax that is removed and recovered is a valuable material with an estimated selling price (34¢/lb.) more than double that of gasoline. The solvent extraction process is used upstream of the solvent dewaxing process as a means of significantly reducing the feedrate and the cost of the much more expensive solvent dewaxing process. In the solvent extraction process, a solvent such as N-Methyl-2-Pyrrolidone is used to absorb aromatics from the feed stream. The paraffinic wax is not absorbed and passes thru the unit. For the ACC approximately 70% of the distillate solvent feed to the extraction unit is absorbed, thereby reducing the solvent dewaxing unit feedrate by a factor of 3. In the solvent dewaxing process,

the paraffinic wax is separated from the feed stream by chilling, precipitation and filtration in the presence of a suitable solvent such as methyl ethyl ketone (MEK). When wax production is desired, as in the ACC, a three stage filtration system is used along with a wax finishing step. Conventional fixed bed hydrotreating is used to make the final improvement in distillate solvent quality. For the ACC, a single train system operating at conditions favorable to aromatics hydrogenation (650 to 7500F & 1,800 psig) is used.

The average MAF coal conversion improved 2.4 percentage points when the distillate recycle solvent was fully dewaxed and hydrotreated as compared to using the as-is distillate solvent. Hydrogen uptake in the small scale hydrotreating runs was less than half of what was expected based on Exxon EDS operation. For the ACC it is assumed that the full dewaxing and hydrotreating of the distillate recycle solvent will improve MAF coal conversion by 3 percentage points (95% vs 92% in Base Case). This improvement further increases product yield and reduces the IOM recycle rate.

COMPARATIVE RESULTS

A simplified block flow diagram of the ACC is shown in Figure 2. At the same coal feedrate to liquefaction, gasoline production increases by 4.5% while a significant quantity of the valuable wax byproduct is also recovered. This increase in product yield is directly related to the reduction in Rose unit organic rejection by 6.0% on MF coal. At the same time, recycle solvent rate is reduced by 20% because of wax removal, lower feed ash and higher coal conversion. Moly catalyst recirculation enhancement remains constant. In order to achieve higher product yield, the required per pass resid plus IOM conversion increases in both reaction stages. This increased conversion is achieved by a space velocity reduction (predicted by first order kinetics) of approximately 15% versus the Base Case. Although reactor space velocities are lower, reactor weights are only slightly higher due to the offsetting effect of lower recycle solvent rates. Hydrogen consumption increases in proportion to the increased product rate. With the significant decrease in organic rejection, additional gasification of coal is required to close the hydrogen balance. The total electrical power requirement for the ACC increases by approximately 12% due to increased gasification quantities and the requirements of the added units.

The capital and operating cost estimates for the Base Case were developed using the relevant portions of previous liquefaction plant studies, as well as in-house information (4,5,6). Comparative process units capital costs for the ACC are shown in Table 1. Process units investment increases by \$452 million due to the added units and the increased gasification requirements. Interestingly, liquefaction system capital cost decreases despite the higher distillate production rate. As shown in Table 2, the total capital required increases by \$ 522 million over the Base Case. However, as shown in Table 3 net operating costs drop by approximately \$6.94 per barrel of gasoline product due to the lower liquefaction system additive costs and the significant impact of byproduct wax revenue. At a 15% capital charge factor, the required gasoline product selling price for the ACC is \$4.64/Bbl. lower than that for the Base Case.

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TABLE 1
BLACK THUNDER COAL STUDY
ADVANCED CONCEPTS CASE vs BASE CASE
DIFFERENCES IN PROCESS UNIT INVESTMENT

<u>UNIT</u>	<u>Millions of Mid '94 \$:Wyoming Location</u>
OIL AGGLOMERATION	+ 60
COAL SLURRY PREPARATION & DRYING	- 6
FIRST STAGE REACTION SYSTEM	- 23
SECOND STAGE REACTION SYSTEM	- 4
LETDOWN SYSTEM	- 3
VACUUM FRACTIONATION	- 14
ROSE UNIT	- 7
DISTILLATE SOLVENT EXTRACTION	+ 74
DISTILLATE SOLVENT DEWAXING	+ 207
DISTILLATE SOLVENT HYDROTREATING	+ 95
GAS PLANT HYDROGEN RECOVERY & RECYCLE	Same
ASH CONCENTRATE & COAL GASIFICATION	+ 49
OXYGEN PLANT	+ 17
STEAM REFORMER	- 8
MAKEUP HYDROGEN COMPRESSION	Same
UPGRADING UNITS	+ 15
Difference in Process Unit Investment	= + 452

TABLE 2
BLACK THUNDER COAL STUDY
ADVANCED CONCEPTS CASE vs BASE CASE
DIFFERENCES IN TOTAL CAPITAL REQUIREMENT

	<u>Millions of Mid '94 \$: Wyoming Location</u>
PROCESS UNITS	+ 452
OFFSITE UNITS	+ 54
WORKING CAPITAL	Same
START-UP COSTS	+ 8
INITIAL CATALYSTS & CHEMICALS	+ 8
Difference in Total Capital Required	= + 522

TABLE 3
BLACK THUNDER COAL STUDY
ADVANCED CONCEPTS CASE vs BASE CASE
DIFFERENCES IN ANNUAL OPERATING COST

	<u>\$/Bbl. of Gasoline</u>
TOTAL MF COAL: @ \$7.14/T	+ 0.152
PURCHASED ELECTRICAL POWER, @ 4¢/ Kw-hr.	+ 0.364
LIQUEFACTION SYSTEM ADDITIVES :	
- Iron Oxide @ 12¢/lb. & H2S @ 7.5¢/lb.	- 0.802
- Moly Impregnating/ Dispersed Catalyst	- 1.582
- Supported Ni-Moly @ \$3/lb.	- 0.782
MAKEUP DEASHING UNIT SOLVENT	- 0.222
OTHER LIQ'N CATALYSTS & CHEMICALS	+ 0.577
UPGRADING UNITS CATALYSTS & CHEM.	Same
RAW WATER @ \$ 2.50/1,000 Gallons	+ 0.035
ASH DISPOSAL @ \$5/Ton	- 0.007
OPERATING LABOR	+ 0.075
ADMINISTRATION & OPERATIONS SUPPORT	+ 0.013
MAINTENANCE @ 1.5% of TIC + ICC	+ 0.233
INSURANCE & LOCAL TAXES @ 1% of TPC + ICC	+ 0.156
BYPRODUCT CREDITS:	
• Ammonia, Sulfur & Phenols	- 0.019
• Fully Refined Paraffin Wax	- 5.133

Difference in Annual Operating Cost = - 6.942

Diff. in Annualized Capital Cost (15% Cap'l Charge Factor)= + 2.302

ADVANCED CONCEPTS CASE ADVANTAGE = - 4.640

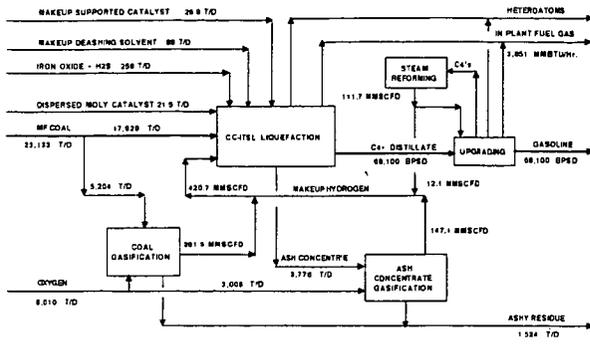


FIGURE 1
BLACK THUNDER LIQUEFACTION STUDY - HYBRID MODE
SIMPLIFIED BLOCK FLOW DIAGRAM - CONCEPTUAL COMMERCIAL PLANT
BASE CASE (SO₂-Free Ash, Wilsonville Run # 263J)

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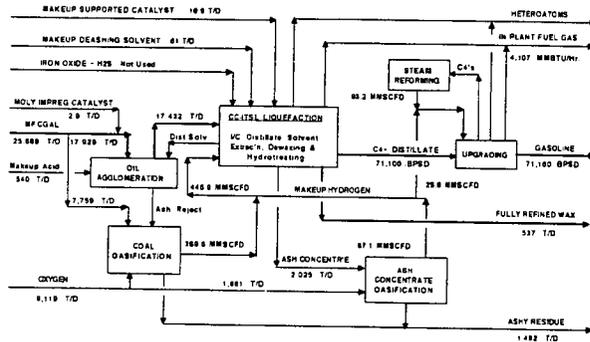


FIGURE 2
BLACK THUNDER LIQUEFACTION STUDY - HYBRID MODE
SIMPLIFIED BLOCK FLOW DIAGRAM - CONCEPTUAL COMMERCIAL PLANT
ADVANCED CONCEPTS CASE (SO₂-Free Ash, W'ville Run # 263J)

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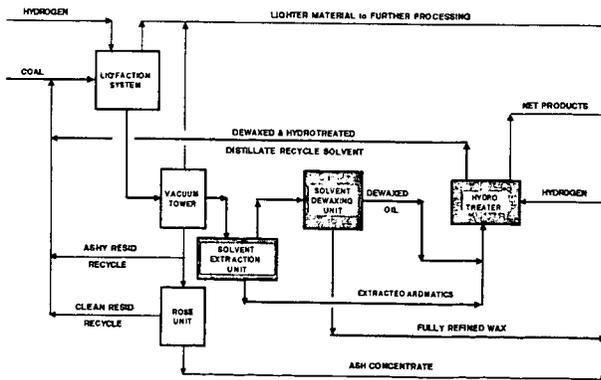


FIGURE 3
ADVANCED CONCEPTS CASE SIMPLIFIED BLOCK FLOW DIAGRAM
SOLVENT EXTRACTION, SOLVENT DEWAXING & HYDROTREATING
OF THE DISTILLATE RECYCLE SOLVENT

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