ANALYSIS OF WILSONVILLE LOW RANK COAL LIQUEFACTION DATA

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ABSTRACT

The use of both dispersed and supported catalysts for the liquefaction of low rank coals in the Wilsonville, AL facility has been evaluated using a statistical approach. This analysis was undertaken to identify the most important operating variables and their effects upon coal conversion and product distributions. The preferred two-stage configuration for the liquefaction of low rank coal consists of a slurry first-stage reactor followed by an ebullated-bed second stage reactor. The presence of a dispersed catalyst promotes the initial liquefaction steps. An ebullated-bed second stage reactor is effective for hydrocracking the coal-derived products and hydrogenating a stream to be recycled as liquefaction solvent. The effect of process configuration upon coal conversion and product distributions is significant.

INTRODUCTION AND BACKGROUND

The principal goal of this study of Wilsonville low rank coal conversion data was to identify the primary operating variables and their effects upon conversion and product distributions. The goals of Wilsonville operation with low rank coal were to demonstrate operation with increasing yields, to provide an overall design concept, and to evaluate different coals. A total of 41 run periods was made with several low rank coals over a wide range of conditions. Only those periods that were lined-out were included in this study. In addition, the runs were not carried out using a statistical approach, so there may be some confounding of results.

The Department of Energy Advanced Two-Stage Coal Liquefaction Facility was located in Wilsonville, Alabama. The Wilsonville pilot plant was run by Southern Company Services (SCS) with funding by the United States Department of Energy (DOE), the Electric Power Research Institute (EPRI), and Amoco Corporation. The unit capacity was about 6 tons of coal per day. Only the recent runs with low rank coals using the close-coupled, integrated two-stage liquefaction (CC-ITSL) mode with and without interstage separation with "ashy recycle" are discussed herein. "Ashy recycle" refers to the recycle of a portion of the mineral matter and unconverted coal with the recycle solvent to the feed tank and first reactor. Solids separation was achieved using a Kerr-McGee ROSE-SR℠ unit. A description of the process has been reported.[1]

Four low rank coals were considered in this data analysis (three Wyoming subbituminous coals and a Texas lignite).

Consideration was made for differences in catalyst bed configurations. The pilot plant had two reactors. The reactor volumes were varied (50, 75, or 100%), and an ebullated supported-catalyst bed was used in none, one, or both reactors. In this paper, a "thermal" stage (T) indicates that no supported catalyst was used; however, the feed to the stage included red mud (an iron ore with limited catalyst activity). A "catalytic" stage (C) refers to the use of an ebullated-bed reactor. A "soluble catalyst" stage (S) refers to the use of a oil-soluble catalyst precursor. Combinations were used; for example, a T/C mode indicates that the first reactor was thermal and the second was catalytic. In addition, when a soluble catalyst precursor was added to the feed, it would be carried into the second stage. When the second stage was catalytic, the designation was S/C. Each of these modes called for different reactor temperatures, and because of the lack of experimental design, reactor temperatures were confounded with reactor configuration.

The data analysis was based on t-tests of the primary dependent variables grouped by reactor mode (T/C, C/C, C/T, S/T, and S/C). The primary assumptions were that differences between the coals were minimal and any effects of temperature, hydrogen partial pressure, and reactor volumes were lumped into the mode distinction.

SUMMARY OF WILSONVILLE LOW RANK LIQUEFACTION RUNS

Overall coal conversion ranged between 87 and 95% (all yields and conversions were calculated on a moisture-ash-free basis: MAF). The average of the statistical group was 92.2%. This conversion was calculated as the MAF portion of the feed coal that was converted to creosote soluble materials. The conversion of the organic portion of the feed coal to total products other than solids recovered from the ROSE-SR unit was reported as 100%-energy rejection. This energy rejection included unconverted coal plus any heavy materials carried along with the solids in the reject stream from the ROSE-SR unit. Energy rejections ranged from about 9 to 24% with an average of 19%. This rejected energy isn't really lost in that the stream could be gasified along with additional coal to generate needed hydrogen and heat for the process.

The yield of distillable liquids (C_4+distillate) averaged 54.7% with a range of about 47 to 57%. That of resid (material boiling above 1000°F) was about 5.5% with a range of 0 to 13%. It was regarded that the resid could have been further converted to distillate by being recycled to extinction in a commercial facility; therefore, the overall yield of liquids was projected to be about 60% with a maximum approaching 65%. The yields of light hydrocarbons (C_1-C_3) was sizeable with a range of 4.2 to 14.2% MAF. The average was 8.3% based on total MAF coal feed; this average calculated on a basis of coal converted to C_4+distillate was 15.2%.

Hydrogen consumption is a significant factor in determining the cost of coal conversion to liquids. In the ideal case, all hydrogen should be consumed in generating distillate liquids. However, light hydrocarbon gases are generated and hetero-atom reduction (deoxygenation, desulfurization, and denitrogenation) add to hydrogen consumption. The level of hydrogen consumption in these low rank coal runs averaged 5.4% on an MAF coal feed basis (9.9% on a basis of coal conversion to C_4+distillate). The range of hydrogen consumption was 4.3 to 7.7% MAF.

STATISTICAL ANALYSIS OF WILSONVILLE RESULTS

Designations and Definitions of Variables:

The following measures of yield/quality (with units in square brackets) were considered in this statistical evaluation:

1. Overall coal conversion [wt% MAF coal],
2. Energy rejection [ratio of combustibles in the ROSE-SR reject stream, "ash concentrate," to that of the feed coal, given as %],
3. Resid+unconverted coal conversion [wt% MAF feed],
4. Individual distillate fraction yields [wt% MAF coal],
5. Overall resid (1000°F+) yield [wt% MAF coal],
6. Hydrogen yield/consumption [wt% MAF coal], and
7. Hydrogen efficiency [C4+ distillate/hydrogen consumed].

Table 1 summarizes the averages of the nominally independent variables for each of the modes considered. Note that temperatures for the soluble catalyst runs were higher in both stages than for the other run periods.

Overall Coal Conversion (wt% MAF):

Figure 1 shows coal conversion as a function of operating mode. The highest level of coal conversion was achieved in the C/C mode. The C/T mode has very poor conversion compared to the other modes. Significant differences exist between C/C and C/T, C/C and S/C, and C/T and S/T modes. Conversion is improved by either switching to the soluble catalyst in the first stage or adding supported catalyst in the second stage.

Energy Rejection:

Reactor operating mode has little effect on energy rejection as shown in Figure 2. The best (lowest) level of energy rejection was achieved in the C/C mode; this is consistent with this mode having the highest level of coal conversion. Only the difference between soluble catalyst and supported catalyst when using supported catalyst in the second stage is significant at 90% confidence.

(Resid+Unconverted Coal) Conversion (Wt% MAF feed):

Figure 3 summarizes the resid + UC conversion data. Significant differences exist between C/C and C/T, C/C and S/C, S/C and S/T, and C/T and S/T modes. Highest conversions again occur in C/C mode. This would be anticipated considering that an active, supported catalyst was used in both stages.

Overall C1-C3 Yield (wt% MAF coal):

Figure 4 shows C1-C3 yield as a function of operating mode. The lowest gas yield was that of the C/T mode periods. The highest yield of light hydrocarbon gases occurred in the C/C mode. This mode generated more light gases than the S/C mode in spite of having much lower temperatures (826/681° vs. 841/810°F).

Overall C4+ Total Yield (wt% MAF coal):

Figure 5 shows C4+ total yield as a function of operating mode. None of the differences are significant at 90% confidence (note the scale is expanded). While the operating mode has no significant effect on C4+ total yield, it is interesting that the pattern of C4+ yields is similar to that of the C1-C3 yields as first stage and second stage modes are changed.

Overall Resid (1000°F+) Yield (wt% MAF coal):

Figure 6 shows resid (1000°F+) yield as a function of operating mode. None of the primary comparisons is statistically significant. The lowest yields occurred when operating in the S/C and C/C modes (averaging about 4%) while the averages of those of the other modes were in the range of 5% to 6%.

Overall Hydrogen Consumption (Wt% MAF coal):

Figure 7 shows hydrogen consumption as a function of operating mode. Significant differences exist between T/C and C/C, C/C and S/C, C/T and S/T, C/C and C/T, and S/C and S/T modes. Using supported catalyst in both stages consumes the most hydrogen, and the substitution of soluble catalyst for supported catalyst in the first stage resulted in reduced hydrogen consumption. It is interesting to note, however, that while the C/C mode consumes the most hydrogen, it is least efficient in putting that hydrogen into the distillate product.

CONCLUSIONS AND OBSERVATIONS

It is concluded that when liquefying low rank coals in a close-coupled, two-stage system similar to that at Wilsonville, the second stage should use supported catalyst in an ebullated bed configuration. The first stage should be catalytic, but the differences between soluble and supported catalysts are not particularly pronounced. (The use of a soluble catalyst was effective in limiting operating problems not discussed herein.) Operation with a supported catalyst in both stages generates more light oil fractions, but wastes hydrogen by making more light gases. Operation using a soluble precursor in the first stage makes more efficient use of hydrogen, but a slightly heavier product is generated.

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REFERENCES

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Table I. Variable Averages by Operating Mode

		Operating Mode				
Variable	All runs	C/T	C/C	T/C	S/T	S/C
CoalConc	29.2	30.1	32.7	30.4	24.7	29.8
Vol1	62.5	75	100	63.889	50	50
Temp1	832	784	826	837	836	841
SpRate1	33.8	28.6	23.9	31.4	37.6	46.9
Vol2	63.971	100	100	62.5	50	50
Temp2	774	792	681	760	809	810
SpRate2	33.3	21.4	23.9	31.7	37.6	46.9

1. Coal Concentration in Feed Slurry [wt% MF[2]], *CoalConc* - Coal concentration in the feed is defined as the concentration of coal on a MF basis in the slurry that is fed to the first reactor.

2. Relative Reactor Volume [%], *Vol 1 or Vol 2.* - Total liquid volume as a percent of full reactor volume.

3. Reactor Temperature [°F], *Temp1, Temp2* - Individual reactor temperatures are designated as the average temperature over the length of the reactor.

4. Space Rate [%], *SpRate1, SpRate2* - Space rate is calculated as the lb/hr feed rate of MF coal per ft^3 of single reactor volume and reported as a ratio to a standard value.

Figure 1

Coal Conversion as a Function of Operating Mode

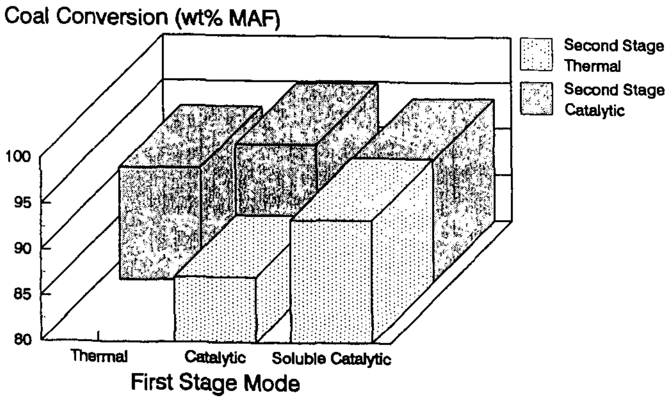

Figure 2
Energy Rejection as a Function of Operating Mode

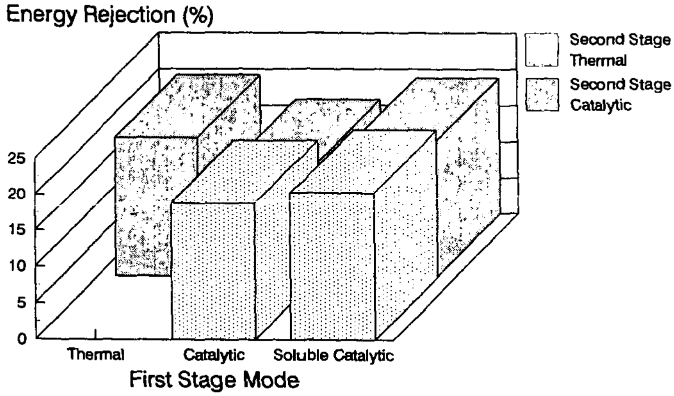

Figure 3
Resid+UC Conversion as a Function of Operating Mode

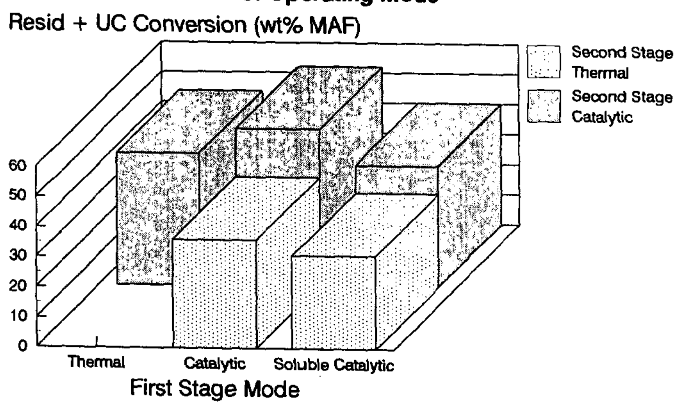

Figure 4
Overall C1-C3 Yield as a Function of Operating Mode

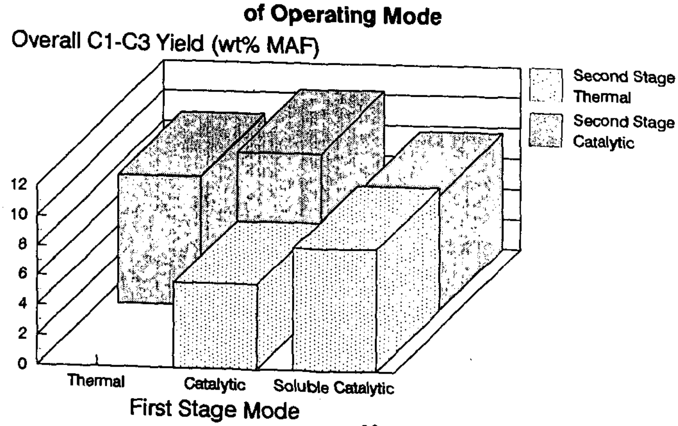

Figure 5

Overall C4+ Total Yield as a Function of Operating Mode

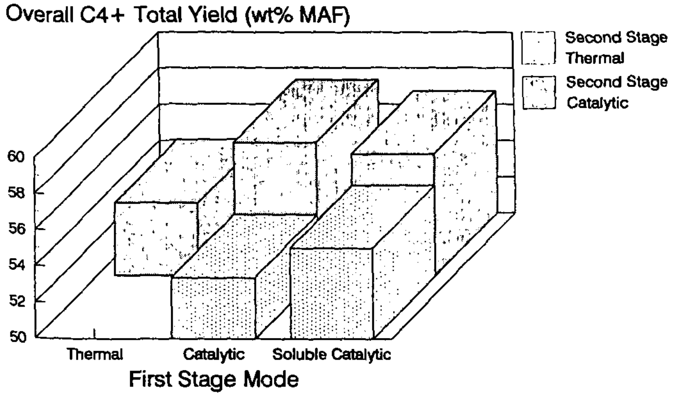

Figure 6

Overall Resid Yield as a Function of Operating Mode

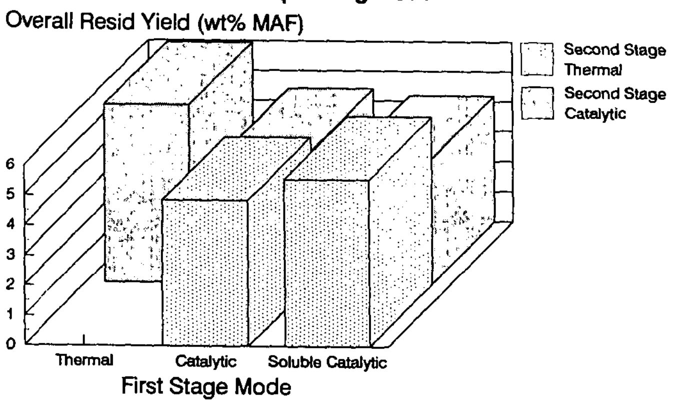

Figure 7

Overall Hydrogen Consumption as a Function of Operating Mode