

SIMULATION OF A COAL/PETROLEUM RESID COPROCESSING PILOT PLANT SCHEME

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ABSTRACT

Coproprocessing involves the conversion of coal and heavy oil in the presence of hydrogen to products that can be further upgraded into commercial fuels; the upgrading can be carried out in an existing refining operation. This relatively new concept eliminates or significantly reduces the need for expensive solvent recycle, and thus has the potential for improved economic performance over current direct coal liquefaction processes.

A simulation of a single-stage coprocessing pilot plant involving the simultaneous conversion of resid and coal has been carried out using the ASPEN PLUS simulator. Limited experimental data were available for only one run, and results should be considered preliminary. The Assay Data Analysis and Pseudocomponent Correlation System of ASPEN PLUS has been used to develop a set of pseudocomponents for coal/resid liquids and to estimate corresponding physical and thermodynamic properties. Correlations based on coal liquids and petroleum liquids have been utilized. For some process equipment, petroleum liquids correlations are better than coal liquids correlations. Testing for the presence of a free water phase and the treatment of heavy resid as a single high-boiling pseudocomponent has improved simulator performance.

INTRODUCTION

Serious research efforts are under way to develop alternative energy sources in order to prevent petroleum supply disruptions from having adverse impact upon the economy of those countries dependent on external supplies of petroleum. During the last twenty years, considerable work has been done in attempts to understand the scientific and technological applications of coal conversion schemes for production of liquid fuels to supplement dwindling petroleum reserves. A relatively new concept addressing this issue involves coprocessing heavy oil with relatively low-cost coals to produce liquid distillates. A review of coal-oil coprocessing technology has been given by Cugini [1]. This review addresses the state of the technology, and several important research efforts required to advance the technology beyond the current level of knowledge. One important area of research required to improve understanding of coprocessing technology is the characterization of the heavy nondistillable feedstock and product oils. Estimations of physical and thermodynamic properties of feedstock and product oils are required to design the commercial reactor and the product separation train and to estimate material flows and compositions for internal and external streams of the coprocessing plant.

There are several sources of information related to coprocessing of coal and heavy oil in bench-scale and pilot-plant operations [2-4]. However, insufficient information is given in these reports to properly characterize the thermodynamic and physical properties of the liquid products as a function of operating conditions. Coal and heavy oil coprocessing in a continuous bench-scale plant has recently been initiated at UOP, Inc. and the Signal Research Center, Inc. [5]. The work related to this study has been described in sufficient detail to allow characterization of the products. The present effort describes the results of a simulation of the single-stage coprocessing bench-scale unit at Signal Research Center, Inc., using the ASPEN PLUS simulator to determine physical and thermodynamic properties

of the liquid products. This bench-scale work forms a preliminary basis to conduct process modeling studies for a range of operating conditions used with this concept.

EXPERIMENTAL

The simulation studies were based on one catalytic coprocessing run at the UOP pilot plant (Figure 1). Two parts of Lloydminster resid to one part of Illinois No. 6 coal constituted the feed. The Lloydminster resid was fractionated so that 95% of the resid consisted of 950°F+ material. Total feed rate to the reactor was 3.75 lb in 12 hours. Coal conversion defined as toluene insolubles was 86%. Operating conditions for the separators include the following: high-pressure separator--temperature = 302°F, pressure = 3114.7 psia; three-phase separator--temperature = 86°F, pressure = 3014.7 psia; low-pressure separator--temperature = 284°F, pressure = 19.7 psia; debutanizer--temperature = 43°F, pressure = 17.7 psia; vacuum fractionator--temperature = 608°F, pressure = 0.94 psia. The effluent from the low-pressure separator and gases from the vacuum pump are sent to the debutanizer, where C₆+ material is condensed and off-gases are sampled. The vacuum fractionator is a packed column that operates with an overhead reflux. Dimensions of the debutanizer and vacuum fractionator towers and of their packing were not available. For lack of better information on the number of equivalent theoretical plates in the debutanizer and vacuum fractionator, simulations for both units were performed as simple flash calculations.

Experimental values were available for the three-phase separator vapor stream, the debutanizer vapor stream, the vacuum fractionator bottoms, and a combined composition for the vacuum fractionator overhead and debutanizer bottoms. Gas phase components were analyzed by gas chromatography. Boiling point distributions were obtained by gas chromatographic simulated distillation.

FLOWSHEET SIMULATION METHODOLOGY

The ASPEN PLUS simulator [6] has been used for the flowsheet analysis of the separator system downstream from the reactor in the UOP coprocessing pilot plant (Figure 1). ASPEN PLUS was originally designed for the analysis of fossil fuel conversion processes, although it has proven useful for many process industries. It has been used to develop a reference data base system of thermophysical properties of coal liquids needed for vapor-liquid equilibrium and for heat and material balance calculations [2], and to simulate the preliminary separator system downstream from the reactor in the SRC-II coal liquefaction process [7].

A preliminary step in the simulation process is the development of an ASPEN flowsheet. Figure 2 represents the ASPEN flowsheet for the separator system following the reactor in the coprocessing pilot-plant flow diagram in Figure 1. The outlet stream from the reactor is the process feed stream for the simulation. The five separators include a high-pressure separator, a three-phase separator, a low-pressure separator, a debutanizer, and a vacuum fractionator. All are modeled as flash units where vapor-liquid equilibrium calculations are performed to produce vapor and liquid outlet streams. Mixer units are used to combine material streams into one stream. Names associated with the streams, and unit operation models for ASPEN identification purposes are designated in Figure 2.

Thermophysical properties used for the simulations were based on three ASPEN PLUS option sets. The option set used for lighter components up to C₆ is based primarily on the Redlich-Kwong-Soave thermophysical model. Heavier components were analyzed using the ASPEN PLUS Assay Data Analysis and Pseudocomponent Correlation System for petroleum liquids and coal liquids. Standard API procedures were used for the petroleum liquids thermophysical models, and correlations developed primarily at Aspen Technology, Inc., were used for the coal liquids thermophysical models. From assay analyses of the vacuum fractionator process streams, a set of

20 pseudocomponents was developed to represent the heavier components; each pseudocomponent represents about a 50°F cut of liquid distillate. Since the process feed stream included both coal and petroleum liquids, separate simulation runs were made where thermophysical properties for each pseudocomponent were analyzed using the correlation option set for coal liquids and the set for petroleum liquids. Comparisons have been made for the performance of each option set.

RESULTS AND DISCUSSION

The composition for the process feed stream to the high-pressure separator is estimated from experimental product slates for the vacuum fractionator, the debutanizer, and the three-phase separator (see Table 1). Gases and identified compounds range from H₂ to pentane. Liquid distillates are represented as pseudocomponents and range from an average boiling point of 111°F to 1091°F (pseudocomponents are prefaced by PC and followed by the average boiling point of the approximately 50°F cut). The initial boiling point (IBP) of the liquid distillate is 69.8°F. For conciseness in the presentation of results, the pseudocomponents have been expressed as four distillate fractions (see Table 2).

TABLE 1. Process Feed Stream Composition for High Pressure Separator

Components*	Flows (lb/hr)	Components*	Flows (lb/hr)
H ₂	0.0593	PC428	0.0075
CO	0.0051	PC477	0.0168
H ₂ S	0.0552	PC525	0.0163
CH ₄	0.0637	PC575	0.0163
C ₂ H ₆	0.0364	PC625	0.0167
C ₃ H ₈	0.0255	PC675	0.0174
C ₄ H ₁₀	0.0105	PC725	0.0158
I-C ₄ H ₁₀	0.0028	PC774	0.0139
C ₅ H ₁₂	0.0034	PC825	0.0128
I-C ₅ H ₁₂	0.0034	PC876	0.0113
H ₂ O	0.0108	PC924	0.0113
PC111	0.0052	PC973	0.0048
PC176	0.0017	PC1022	0.0034
PC226	0.0020	PC1091	0.0503
PC276	0.0025	Ash	0.0108
PC327	0.0041	Unconverted Coal	0.0135
PC376	0.0053	Nondistillate Solids	0.0456

*Pseudocomponents are 50°F cuts and are represented by PC followed by the average boiling point.

TABLE 2. Process Feed Stream Distillate Fractions Expressed as Pseudocomponents.

Distillate Fraction	Pseudocomponent Range	Flows (lb/hr)
IBP-350°F	PC111-PC327	0.0155
350°-450°F	PC376-PC428	0.0128
450°-950°F	PC477-PC924	0.149
950°F+	PC973-PC1091	0.0585

Three-Phase Separator

For the three-phase separator overheads, the effect of the correlation option is given in Table 3. There is some improvement using the petroleum-liquids option set. Use of the coal-liquids option set results in an overall error for total mass flow of overhead of 1.86%, and the petroleum-liquids option set results in an error of 0.73%.

TABLE 3. Comparison of Calculated and Experimental Overhead Flows (lb/hr) for the Three-Phase Separator as a Function of Physical Properties Calculations Method

Components	Correlation Option Set		
	Coal Liquids	Petroleum Liquids	Experimental
H ₂	0.0591	0.0589	0.0587
CO	0.0051	0.0051	0.00513
H ₂ S	0.0516	0.0511	0.0498
CH ₄	0.0629	0.0626	0.0621
C ₂ H ₆	0.0349	0.0346	0.0340
C ₃ H ₈	0.0233	0.0229	0.0225
C ₄ H ₁₀	0.0086	0.0084	0.00849
I-C ₄ H ₁₀	0.0024	0.0023	0.00212
C ₅ H ₁₂	0.0023	0.0022	0.00263
I-C ₅ H ₁₂	0.0025	0.0023	0.00263

Debutanizer

For the debutanizer overheads, the effect of the correlation option is given in Table 4. There is improvement using the petroleum-liquids option set. Use of the coal-liquids option set results in an overall error for total mass flow of overhead of 16.4%, and the petroleum-liquids option set results in an error of only 1.37%.

TABLE 4. Comparison of Calculated and Experimental Overhead Flows (lb/hr) for the Debutanizer as a Function of Physical Properties Calculations Method

Components	Correlations Options Set		
	Coal Liquids	Petroleum Liquids	Experimental
H ₂	2.31 x 10 ⁻⁴	3.91 x 10 ⁻⁴	6.09 x 10 ⁻⁴
CO	2.79 x 10 ⁻⁵	4.59 x 10 ⁻⁵	--
H ₂ S	3.5 x 10 ⁻³	3.9 x 10 ⁻³	--
CH ₄	7.84 x 10 ⁻⁴	1.1 x 10 ⁻³	1.60 x 10 ⁻³
C ₂ H ₆	1.4 x 10 ⁻³	1.8 x 10 ⁻³	2.35 x 10 ⁻³
C ₃ H ₈	2.1 x 10 ⁻³	2.5 x 10 ⁻³	2.96 x 10 ⁻³
C ₄ H ₁₀	1.5 x 10 ⁻³	1.8 x 10 ⁻³	1.95 x 10 ⁻³
I-C ₄ H ₁₀	3.34 x 10 ⁻⁴	4.02 x 10 ⁻⁴	6.51 x 10 ⁻⁴
C ₅ H ₁₂	6.15 x 10 ⁻⁴	7.41 x 10 ⁻⁴	8.08 x 10 ⁻⁴
I-C ₅ H ₁₂	6.10 x 10 ⁻⁴	7.31 x 10 ⁻⁴	8.08 x 10 ⁻⁴
C ₆ ⁺	1.05 x 10 ⁻³	1.35 x 10 ⁻³	2.88 x 10 ⁻³

Vacuum Fractionator

As a measure of simulation adequacy, total vapor and liquid flows computed by simulation were compared with experimental data. Total mass flow of overhead and

bottoms is less sensitive to the number of theoretical stages than are the individual distillate fraction flows. The effect of the correlation option set (coal-liquids vs. petroleum-liquids) on the vacuum fractionator simulation performance was determined. Since simulated distillate flows were in considerable disagreement with experimental values for the reported operating pressure of 0.94 psia, additional simulation runs were made to observe the effect of assumed column pressures. For the petroleum-liquids option set, a value of 4.5 psia gave the best match of calculated total overhead and bottom flows to experimental flows. Correspondingly, a value of 6.0 psia was found for the coal-liquids option set. Given that the reported column pressure, 0.94 psia, is closer to 4.5 psia than to 6.0 psia, this result gives an indirect indication that the petroleum-based option set better describes the experimental system. The actual operating pressure for the vacuum fractionator was known to increase above 0.94 psia during the experimental run, but no information is available as to the extent of increase. The results for the flows of the distillate fractions are presented in Tables 5-7. Table 5 represents results for vacuum bottoms flows, and Table 6, for vacuum overhead flows (calculated vacuum overhead flows also include values for debutanizer bottoms flows in order to agree with experimental measurements). Table 7 gives a comparison of the pressure and correlation option set in terms of an overall percentage error for both bottoms and overhead at the operating pressure of 0.94 psia. Use of the petroleum-liquids option set gives better agreement, although the percentage of error relative to experimental error is still considerable.

TABLE 5. Comparison of Calculated and Experimental Vacuum Bottoms Flows (lb/hr) as a Function of Physical Properties Calculation Method and System Pressure

Correlation Option Set	Boiling Point Range, °F	Calculated	Experimental
<u>Coal Liquids</u>			
P = 0.94 psia	IBP-350	1.52×10^{-6}	--
	350-450	8.46×10^{-5}	--
	450-950	5.60×10^{-3}	5.16×10^{-2}
	<u>950+</u>	<u>3.24×10^{-2}</u>	<u>5.92×10^{-2}</u>
	Total	3.80×10^{-2}	1.11×10^{-1}
P = 6.0 psia	IBP-350	5.38×10^{-5}	--
	350-450	2.94×10^{-4}	--
	450-950	5.47×10^{-2}	5.16×10^{-2}
	<u>950+</u>	<u>5.67×10^{-2}</u>	<u>5.92×10^{-2}</u>
	Total	1.12×10^{-1}	1.11×10^{-1}
<u>Petroleum Liquids</u>			
P = 0.94 psia	IBP-350	4.92×10^{-6}	--
	350-450	2.47×10^{-5}	--
	450-950	1.40×10^{-2}	5.16×10^{-2}
	<u>950+</u>	<u>5.38×10^{-2}</u>	<u>5.92×10^{-2}</u>
	Total	6.78×10^{-2}	1.11×10^{-1}
P = 4.5 psia	IBP-350	5.59×10^{-5}	--
	350-450	2.75×10^{-4}	--
	450-950	5.21×10^{-2}	5.16×10^{-2}
	<u>950+</u>	<u>5.79×10^{-2}</u>	<u>5.92×10^{-2}</u>
	Total	1.10×10^{-1}	1.11×10^{-1}

TABLE 6. Comparison of Calculated and Experimental Vacuum Overhead Flows (lb/hr) as a Function of Physical Properties Calculation Method and System Pressure

Correlation Option Set	Boiling Point Range, °F	Calculated	Experimental
<u>Coal Liquids</u>			
P = 0.94 psia	IBP-350	1.07×10^{-2}	1.34×10^{-2}
	350-450	1.28×10^{-2}	1.90×10^{-2}
	450-950	1.43×10^{-1}	8.97×10^{-2}
	950+	2.60×10^{-2}	8.17×10^{-5}
	Total	1.93×10^{-1}	1.22×10^{-1}
P = 6.0 psia	IBP-350	1.07×10^{-2}	1.34×10^{-2}
	350-450	1.25×10^{-2}	1.90×10^{-2}
	450-950	9.38×10^{-2}	8.97×10^{-2}
	950+	1.70×10^{-3}	8.17×10^{-5}
	Total	1.19×10^{-1}	1.22×10^{-1}
<u>Petroleum Liquids</u>			
P = 0.94 psia	IBP-350	1.05×10^{-2}	1.34×10^{-2}
	350-450	1.28×10^{-2}	1.90×10^{-2}
	450-950	1.34×10^{-1}	8.97×10^{-2}
	950+	4.60×10^{-3}	8.17×10^{-5}
	Total	1.62×10^{-1}	1.22×10^{-1}
P = 4.5 psia	IBP-350	1.05×10^{-2}	1.34×10^{-2}
	350-450	1.25×10^{-2}	1.90×10^{-2}
	450-950	9.64×10^{-2}	8.97×10^{-2}
	950+	5.08×10^{-3}	8.17×10^{-5}
	Total	1.24×10^{-1}	1.22×10^{-1}

TABLE 7. Comparison of Physical Properties Calculation Method on Vacuum Fractionator Effluent Stream Mass Flow Rates

Correlation Option Set	% Error*	
	Vacuum Bottoms	Vacuum Overhead
<u>Coal Liquids</u>		
P = 0.94 psia	65.8	58.2
<u>Petroleum Liquids</u>		
P = 0.94	38.9	32.8

*Absolute value of (Calculated - Experimental)/Experimental.

Effects of pressure and correlation option set on pseudocomponent composition for the vacuum fractionator are reflected in Figures 3-6. Figures 3 and 4 represent the effect on the vacuum bottoms stream, and Figures 5 and 6, the effect on the overhead stream. These figures indicate that the petroleum-liquids option set gives better values than the coal-liquids option set and that the effect of pressure is considerable.

Low Pressure Separator

A temperature of 284°F was used for the operating condition of the low-pressure separator in the simulator. Since, experimentally, a temperature range of 248°F-284°F was given, it was decided to make a simulation run at the lower temperature to determine any effects of the assumed temperature on the effluent flow rates. Results for the lower temperature indicate only a small increase in bottoms flows of 1.3% and an decrease in overhead flows of 11.8%.

Effect of Free Water Phase on the Debutanizer

Simulations were carried out on the debutanizer to determine the impact of the presence of a free water phase (Table 8). Without the invocation of the option to test for the presence of free water, no distillate and only a small amount of lighter gases are predicted by the simulator to be present in the debutanizer bottoms. With the test for the presence of water invoked, the presence of a free water phase is confirmed, and results indicate a substantial increase in bottoms flow and a decrease in overhead. Both of these predictions agree with experiment, as shown in Table 8. All simulations have tested for the presence of a free water phase.

TABLE 8. Effect of Treatment of Water on the Debutanizer Effluent Product Flows (lb/hr)

Products	Assumed Absence of Free Water Phase		Assumed Presence of Free Water Phase		Experimental Overhead
	Overhead	Bottoms	Overhead	Bottoms	
Gases-C ₃	6.93 x 10 ⁻³	3.90 x 10 ⁻⁶	1.11 x 10 ⁻²	1.16 x 10 ⁻³	1.18 x 10 ⁻²
IBP-350°F	3.79 x 10 ⁻³	0	1.05 x 10 ⁻³	4.86 x 10 ⁻³	
350°-450°F	1.53 x 10 ⁻³	0	1.08 x 10 ⁻⁷	1.16 x 10 ⁻³	2.88 x 10 ⁻³
450°-950°F	1.31 x 10 ⁻³	0	4.69 x 10 ⁻⁸	8.46 x 10 ⁻⁴	
950°F+	1.02 x 10 ⁻⁷	0	0	6.52 x 10 ⁻⁸	

TABLE 9. Effect of Treatment of Heavy Resid on Vacuum Bottoms

Product	Heavy Residue Treated as	
	Solid Material (Flows, lb/hr)	Pseudocomponent (Flows, lb/hr)
Gases-C ₃	0	1.44 x 10 ⁻⁸
IBP-350°F	0	1.52 x 10 ⁻⁶
350°-450°F	0	8.47 x 10 ⁻⁶
450°-950°F	0	5.59 x 10 ⁻³
950°F+	0	3.25 x 10 ⁻²
Nondistillate Solid	0.0456	0

Effect of Treatment of Heavy Resid on Vacuum Bottoms

The method of treatment of heavy resid has an impact on predicted composition and flow rate of vacuum bottoms as reflected in Table 9. The data are for the simulation of the vacuum fractionator at the pressure of 0.94 psia and using coal-liquids correlations. When the heavy resid is treated as a nondistillate solid, i.e., material with negligible vapor pressure, no distillates are predicted to appear in the vacuum bottoms. When the heavy resid is treated as a 1091°F pseudo-

component, distillate products are predicted in the bottoms. This treatment of the resid fraction corresponds to that of McKeegan and Klunder [8], who also assigned a single normal boiling point to the nondistillate material in their simulation of the separator system in the SRC-II coal liquefaction process (although they used a much higher temperature). The plot in Figure 4 for $P = 0.94$ psia, as well as for $P = 6.0$ psia, reflects the presence of heavy resid treated as the 1091°F pseudocomponent. These plots are in line with the experimental observation of the presence of significant amounts of liquid distillate product in the vacuum bottoms stream. All simulation results for the vacuum fractionator presented in Tables 5-7 and Figures 3-6 have treated the heavy resid as a 1091°F pseudocomponent.

CONCLUSIONS

The very preliminary results reported here indicate that the use of petroleum-liquids correlations may result in an improvement over coal-liquids correlations in the simulation of the coprocessing of Lloydminster with Illinois No. 6 coal. Agreement between simulation and experiment is improved by using a higher assumed pressure than the experimental pressure for the vacuum fractionator, by treating the presence of water as a free water phase, and by treating the heavy resid as 1091°F distillate rather than an inert solid material. It is necessary to obtain better definition of the separation equipment used and the operating conditions employed, and to acquire a larger data set in order to evaluate the present capability for simulating the separation steps in coprocessing.

ACKNOWLEDGMENT

The authors would like to thank UOP, Inc., for supplying the experimental data that served as the basis for the simulation studies, and Charles Luebke of UOP, Inc., and Carl Lea of Signal Research Center, Inc., for many discussions relative to process operation and data analysis.

DISCLAIMER

Reference in this report to any specific product, process, or service is to facilitate understanding and does not necessarily imply its endorsement or favoring by the United States Department of Energy.

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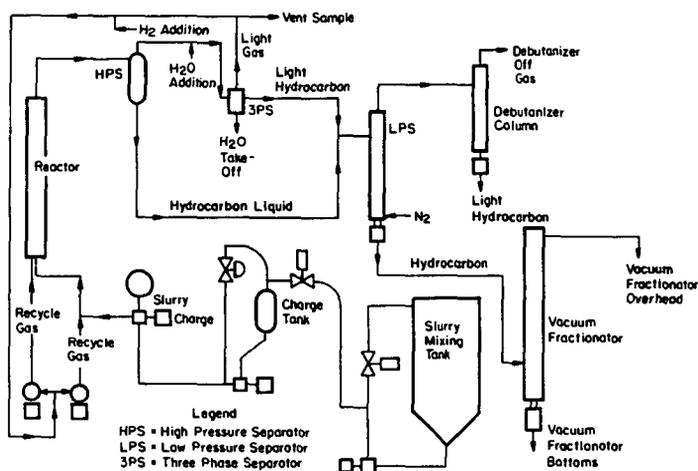


Figure 1 - UOP Coprocessing Pilot-plant Flow Diagram.

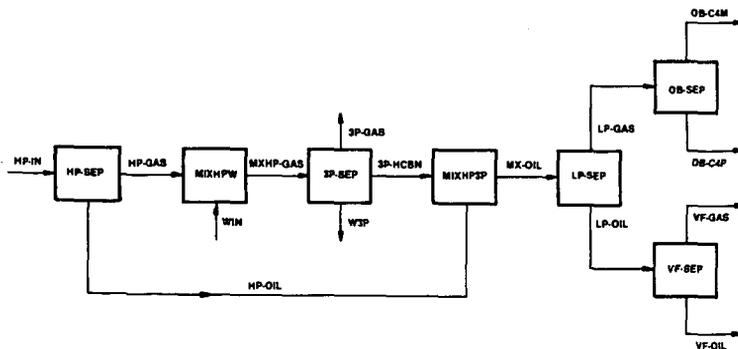


FIGURE 2. ASPEN FLOWSHEET OF SEPARATOR SYSTEM FOR UOP COPROCESSING PILOT-PLANT FLOW SCHEME.

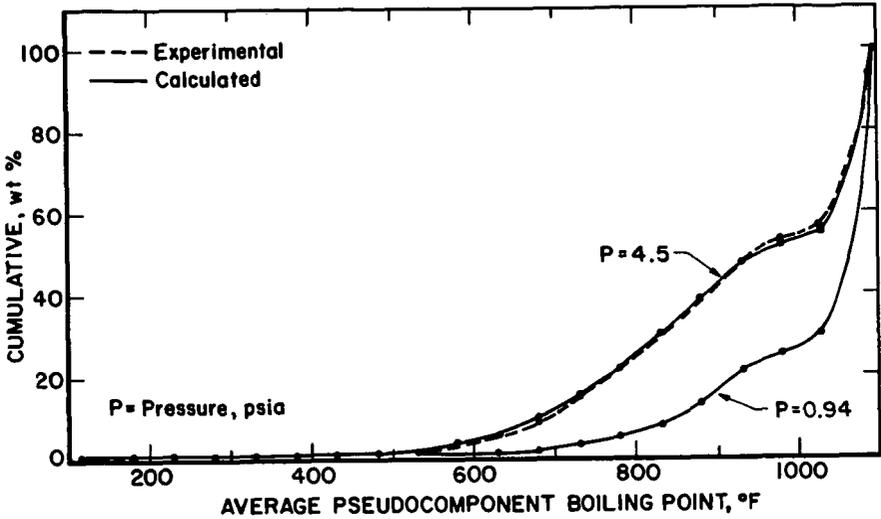


Figure 3 - Composition of Vacuum Bottoms Pseudocomponents Using Petroleum Liquids Correlations.

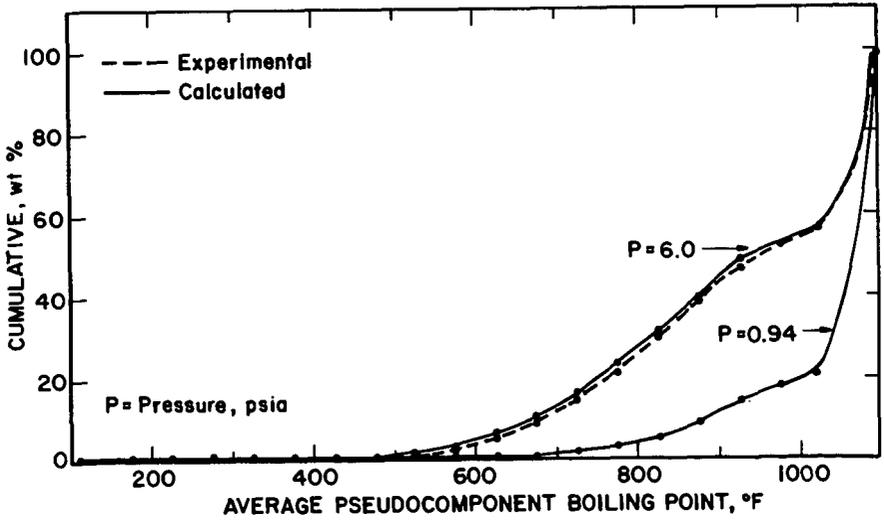


Figure 4 - Composition of Vacuum Bottoms Pseudocomponents Using Coal Liquids Correlations.

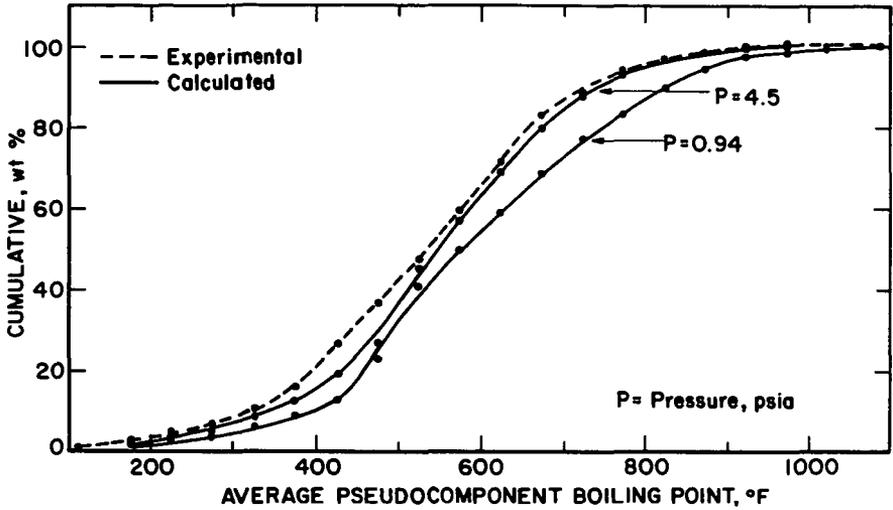


Figure 5 - Composition of Vacuum Overhead Using Petroleum Liquids Correlations.

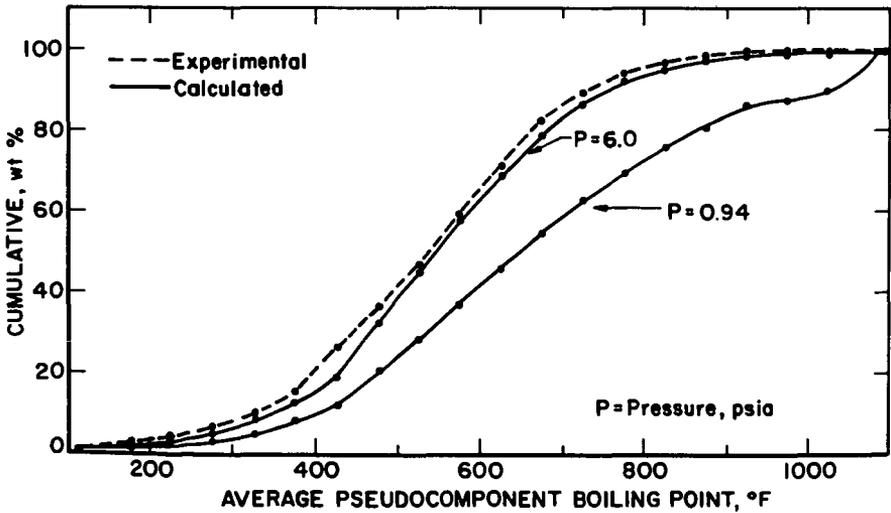


Figure 6 - Composition of Vacuum Overhead Using Coal Liquids Correlations.