Kinetics of Short Contact Time Coal Liquefaction

Part II: Effect of Nature of Coal and Solvent

A bituminous coal (Powhatan No. 5) and a subbituminous coal (Belle Ayr) were liquefied in the presence of hydrogen donor solvents. Statistical analyses of the data showed that for contact times up to 10 min, the coal conversion, measured in terms of tetrahydrofuran solubles, was not significantly different for the two coals. However, the subbituminous coal gave more asphaltenes than the bituminous coal.

The hydrogen-donating capacity of the solvent was varied by the addition of hydrogenated phenanthrene to SRC-II recycle solvent. The results indicated that there are two routes for the formation of oils from coal. One is by adduct formation giving asphaltenes and preasphaltenes, which in turn gives oils, and the other is direct oil formation from coal. The preferred route depends on the hydrogen-donating capacity of the solvent. The kinetic model, proposed in Part I of this paper, is extended to account for the change in the donor capacity of the solvent.

J. S. ABICHANDANI,
J. H. WIELAND, and
Y. T. SHAH
Department of Chemical and Petroleum Engineering
University of Pittsburgh
Pittsburgh, PA 15261
and
D. C. CRONAUER
Gulf Research and Development Co.
Pittsburgh, PA 15230

SCOPE

Coal liquefaction can be characterized as occurring in three stages, namely dissolution, hydrogen transfer, and hydrogenation. In each of these stages the nature of the solvent can affect the rate of reaction and the selectivity of the products. Solvent components which affect the behavior of coal liquefaction processes can be classified as hydrogen donors, hydrogen shuttlers, and components that aid in solubilization. Recent studies have shown that poor hydrogen-donating solvents form adducts with coal to give insoluble products. This appears to occur to a lesser extent with good donor solvents. It is likely that for the case of good hydrogen donor solvents the formation of soluble products compensate for the adduct formation. Kinetic measurements at short contact times (up to 10 min) with solvents of varying hydrogen donor capacity have been made to provide a better understanding of the processes of liquefaction and adduct formation.

CONCLUSIONS AND SIGNIFICANCE

In a stirred tank batch reactor, there appears to be no significant difference in the conversion of Powhatan No. 5 bituminous and Belle Ayr subbituminous coal to pentane and tetrahydrofuran (THF) solubles for contact times up to 10 min. More asphaltenes are recovered from the liquefaction of Belle Ayr coal than from that of Powhatan coal. Both the coals, when contacted with SRC-II recycle solvent, show an initial effective loss of solvent.

The initial depletion of oils progressively decreases with an increase in the hydrogen-donating capacity of the solvent, indicating that the initial loss of solvent is independent of the nature of coal, but it is dependent on the hydrogen-donating capacity of the solvent. Oils can be formed either directly from coal or by an indirect route via asphaltenes and preasphaltenes through adduct formation. The latter results in an initial loss of solvent. The former route gains preference over the latter as the hydrogen-donating capacity of the solvent is increased.

A single kinetic model entailing both direct and indirect routes is shown to adequately represent the data obtained with two coals and three solvents. Emphasis in this model has been directed toward the chemical and thermal changes occurring during the heat up period in a coal liquefaction preheater. The model accounts for changes for the contact times up to 10 min.

INTRODUCTION

The extent of coal liquefaction is greatly influenced by coal characteristics such as rank, petrographic composition, mineral matter content, and geological history. Variation in the liquid yields has been observed for different petrographic compositions (Fisher et al., 1942). Storch and his coworkers (1943) worked with a number of coals and concluded that two compositional parameters should be considered in coal liquefaction—the petrography and MAF (moisture and ash free) carbon content of coal.

J. S. Abichandani is presently with Schneider Consulting Engineers, Pittsburgh, PA 15222.

TABLE 1. ANALYSIS OF BELLE AYR COAL

Proximate Analysis	As Rec'd	Dry
% Moisture	27.50	—
% Ash	5.58	7.70
% Volatile	32.61	44.98
% Fixed Carbon	34.31	47.32
Calorific Value	**As Rec'd**	**Dry**
Btu	11,217	11,962
Ultimate Analysis		**Dry**
% Moisture		—
% Carbon		65.56
% Hydrogen		4.74
% Nitrogen		1.16
% Chlorine		0.01
% Sulfur		.41
% Oxygen (diff)		20.42
% Ash		7.70

SI Conversion: kJ = Btu × 1.055

TABLE 2. ANALYSIS OF SOLVENTS

	HPh	SRC-II
Elemental Analysis		
Carbon	91.08%	87.44
Hydrogen	8.83%	8.65
Nitrogen	0.01%	1.39
Oxygen	0.00%	2.28
Sulfur	0.08%	0.25
Physical Properties		
Specific Gravity	1.038	1.023
Viscosity @ 298 K	10.80 cs	—
Distillation Data (K)		
(Modified ASTM D-1160; Watson K = 10.0)		
% Vol. Distilled		
10	543	518
50	596	596
90	624	694
100	663	747
Donatable Hydrogen (wt %)	1.58	0.55

TABLE 3. EFFECT OF NATURE OF COAL ON THE RECOVERY OF PENTANE SOLUBLES

Coal*/Solvent**	Temp. (*K*)	Time (min) 2	4	6	10
P/I	723	−0.02	0.04	0.05	−0.02
B/I	723	−0.44	−0.17	−0.09	0.20
P/I	673	−0.25	−0.14	−0.51	−0.13
B/I	673	−0.15	−0.09	−0.07	0.08
P/I	573	−0.09	−0.16	−0.21	0.06
B/I	573	0.00	−0.04	−0.08	−0.04
P/II	673	0.03	0.08	0.08	0.34
B/II	673	0.21	0.27	0.25	0.33
P/II	623	−0.03	−0.07	−0.11	0.16
B/II	623	0.06	0.06	0.06	0.24
P/II	573	−0.05	−0.07	−0.09	0.05
B/II	573	0.14	−0.04	−0.3	0.17

* P = Powhatan coal; B = Belle Ayr coal.
** I = SRC recycle solvent; II = SRC recycle solvent (0.6) + HPh (0.4).

TABLE 4. EFFECT OF NATURE OF COAL ON THE RECOVERY OF TOLUENE SOLUBLES

Coal*/Solvent**	Temp. (K)	Time (min) 2	4	6	10
P/I	723	0.57	0.21	0.41	0.48
B/I	723	0.41	0.50	0.50	0.61
P/I	673	0.19	0.27	0.20	0.29
B/I	673	0.26	0.33	0.34	0.48
P/I	573	0.12	0.10	0.02	0.22
B/I	573	0.21	0.16	0.13	0.21
P/II	673	0.18	0.26	0.31	0.57
B/II	673	0.47	0.53	0.52	0.52
P/II	623	0.08	0.04	−0.02	0.23
B/II	623	0.19	0.21	0.19	0.39
P/II	573	0.03	0.04	0.00	0.15
B/II	573	0.29	0.11	0.10	0.31

* P = Powhatan coal; B = Belle Ayr coal.
** I = SRC recycle solvent; II = SRC recycle solvent (0.6) + HPh (0.4).

Whitehurst et al. (1977) compared the behavior of different rank coals at short times (3 min) and long times (90 min) and concluded that at short reaction times, coals in the 75–85 MAF carbon range give about the same conversion; while at long times, the conversion is high for all coals having less than 85% MAF carbon; at higher levels, conversion falls sharply.

The solvent components which affect the behavior of the coal liquefaction process can be classified as hydrogen donors, hydrogen shuttlers, and components that aid in solubilizing. A hydrogen donor is a species which can give up one or more hydrogens to another reactant such as free radicals formed during coal pyrolysis. The principal source of hydrogen is partially hydrogenated aromatic hydrocarbons. Seshadri et al. (1978) have used Carbon-13 nuclear magnetic resonance spectroscopy to quantitatively define the hydrogen-donating capacity (H_s) of a solvent. Crounauer and Ruberto (1979) experimented with Belle Ayr coal and three solvents with H_s values of 0.15, 1.01, and 2.25. They reported that the solvent with the least hydrogen-donating capacity gave an initial observable loss of solvent (oil). No such depletion of oil was observed for the other two solvents.

Petrakis and Grandy (1978), Epperly (1978), and Whitehurst et al. (1980) studied the effects of the hydrogen-donating capacity of the solvent by adding tetralin in different proportions to the solvent. They all reported that the conversion (defined as THF solubles) is responsive to the concentration of hydroaromatic H-donors up to a particular concentration level, beyond which further H-donor addition has little beneficial effect. These results are consistent with the EDS development studies reported by Fant (1978) of Exxon. While an extensive study of liquefaction with various donor solvents has been on-going at a number of laboratories, reference is made herein to the work of Curtis et al. (1981), Derbyshire et al. (1981), Furlong et al. (1982), Miller et al. (1982), King and Stock (1981), and Wright et al. (1975), for recent publications in this area.

Preheaters used in coal liquefaction are usually in the form of helical coils. Schweighardt et al. (1976) have reported that the presence of even small amounts of asphaltenes or preasphaltenes in the coal-oil slurry increases the viscosity of the slurry fifty-fold. Pumping of such high viscosity material is one of the major engineering problems associated with the operation of such preheaters. It was shown in the previous paper that at least part of the adducts are asphaltenes and preasphaltenes; therefore, the level of adduction may be of critical importance to the design of preheaters. Since the hydrogen-donating capacity of the solvent is involved with the amount of adduct formation, the nature of the solvent influences the efficiency of the preheater.

EXPERIMENTAL

The experimental procedure and the analytical technique employed are the same as those previously described (Part I). In addition to the bituminous coal (Powhatan No. 5), a subbituminous coal (Belle Ayr) was used. The analysis of the Belle Ayr coal is given in Table 1.

The nature of the solvent was altered by the addition of partially hydrogenated phenanthrene (HPh) and SRC-II recycle solvent. Table 2 shows the analysis of the solvents. Two such mixed solvents were prepared with

TABLE 5. EFFECT OF NATURE OF COAL ON THE RECOVERY OF TETRAHYDROFURAN SOLUBLES

Coal*/Solvent**	Temp. (K)	Time (min) 2	4	6	10
P/I	723	0.76	0.42	0.64	0.65
B/I	723	0.63	0.70	0.63	0.72
P/I	673	0.42	0.50	0.53	0.59
B/I	673	0.52	0.49	0.52	0.63
P/I	573	0.25	0.23	0.16	0.40
B/I	573	0.27	0.23	0.22	0.34
P/II	673	0.44	0.49	0.52	0.79
B/II	673	0.57	0.59	0.56	0.65
P/II	623	0.23	0.15	0.18	0.39
B/II	623	0.23	0.23	0.23	0.41
P/II	573	0.16	0.18	0.15	0.23
B/II	573	0.29	0.11	0.09	0.31

* P = Powhatan coal; B = Belle Ayr coal.

** I = SRC recycle solvent; II = SRC recycle solvent (0.6) + HPh (0.4).

TABLE 6. F_{exp} FOR EFFECT OF NATURE OF COAL

	Time (min) 2	4	6	10
Pentane Solubles	−0.1	−0.18	−0.46	−0.51
Toluene Solubles	−33.51	−65.11	−56.89	−31.5
THF Solubles	−1.86	−1.71	−0.038	−0.01
Error Estimates	Mean Square	Degrees of Freedom	F_{tab}	
Pentane Solubles	0.087	32	4.15	
Toluene Solubles	0.013	32	4.15	
THF Solubles	0.007	32	4.15	

0.16 and 0.4 weight fractions of HPh. The donatable hydrogen, H_s, for each solvent was obtained by nuclear resonance spectroscopy by the procedure outlined by Seshadri et al. (1978). H_s values of 0.55, 0.7, and 1.0 were obtained for SRC-II recycle solvent, 16 wt % HPh, and 40 wt % HPh solvents.

RESULTS AND DISCUSSION

Effect of the Nature of Coal

The liquefaction rates for the Powhatan bituminous coal and the Belle Ayr subbituminous coal were determined at short times (up to 10 min). The solvents were a SRC-II recycle solvent and a mixture of 60 wt % SRC-II recycle solvent with 40 wt % HPh. The product distributions in terms of pentane, toluene, and THF solubles are shown in Tables 3 to 5.

The statistical F test was employed to verify the difference in the product distribution for the two coals. The computed F values (F_{exp}) for the effect of the nature of coal are reported in Table 6. In this analysis, Powhatan coal was taken as the higher level, i.e., a negative value for F_{exp} implies that Belle Ayr coal gives more soluble product than Powhatan coal.

As seen from Table 6 there is no significant change in the production of pentane solubles for the two coals; however, a much higher level of toluene solubles results when liquefying Belle Ayr coal than when liquefying Powhatan coal. This is indicated by the highly significant F_{exp} values (numerically much greater than F_{tab}) obtained for toluene solubles. Note also that the yields of THF solubles do not show a significant difference for the two coals. For reference, toluene solubles include pentane solubles, and THF solubles include toluene solubles. Hence, the overall conversion (based on THF solubles) is almost the same for the two coals. for contact times up to 10 min; however, Belle Ayr coal gives much greater yields of asphaltenes than Powhatan coal. Consequently Powhatan coal produces more preasphaltenes.

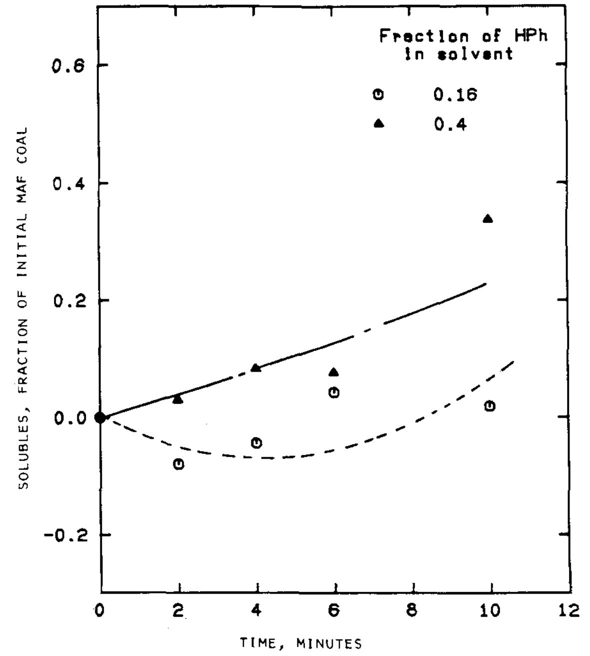

Figure 1. Effect of solvent on the conversion of Powhatan to pentane solubles at 673 K and S/C = 2.6.

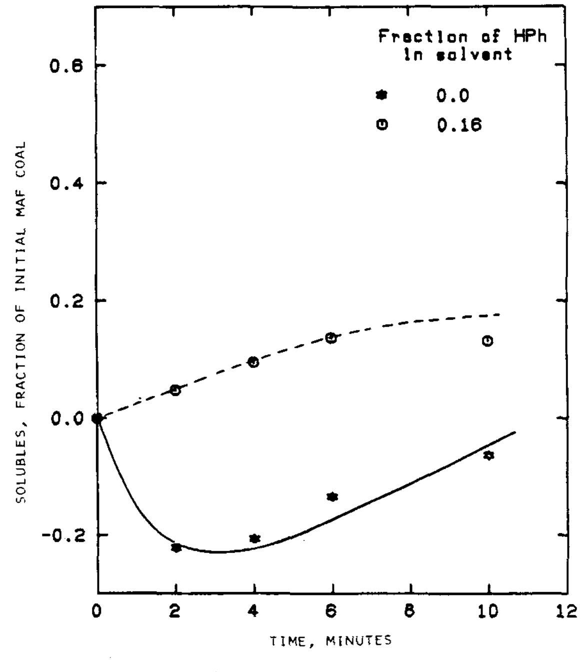

Figure 2. Effect of solvent on the conversion of Powhatan coal to pentane solubles at 723 K and S/C = 2.3.

It is noted that the above similarities in the depth of conversion of Belle Ayr subbituminous and Powhatan bituminous coals in this batch stirred-tank reactor are not consistent with those of liquefaction runs made using these coals in a flow unit (CSTR). Namely, Cronauer and Ruberto (1979) and Abichandani et al. (1982) reported a greater extent of conversion of the higher rank coal to pyridine solubles using hydrogenated anthracene oil as the solvent in a CSTR unit. This difference of conversion levels may be due to a combination of reactor configuration, liquefaction solvent, and/or extraction solvent.

Effect of the Nature of Solvent

Relatively few studies have reported initial depletion of oils (Wright et al., 1975; Longanbach et al., 1978; Traeger et al., 1979;

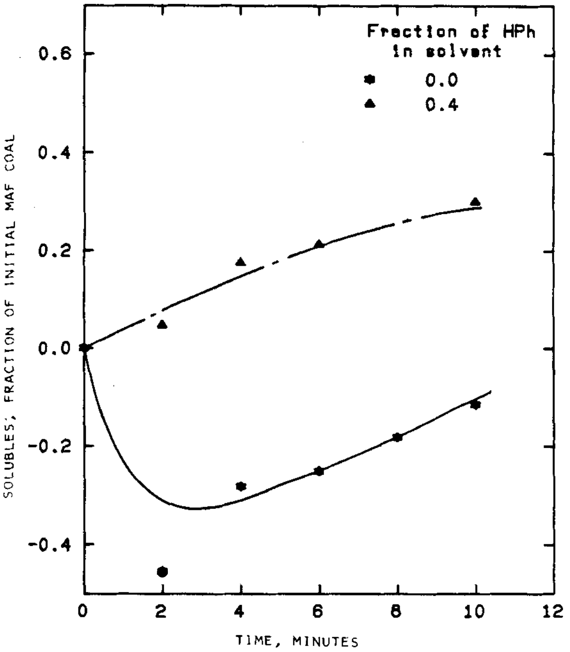

Figure 3. Effect of solvent on the conversion of Powhatan coal to pentane solubles at 723 K and *S/C* = 4.2.

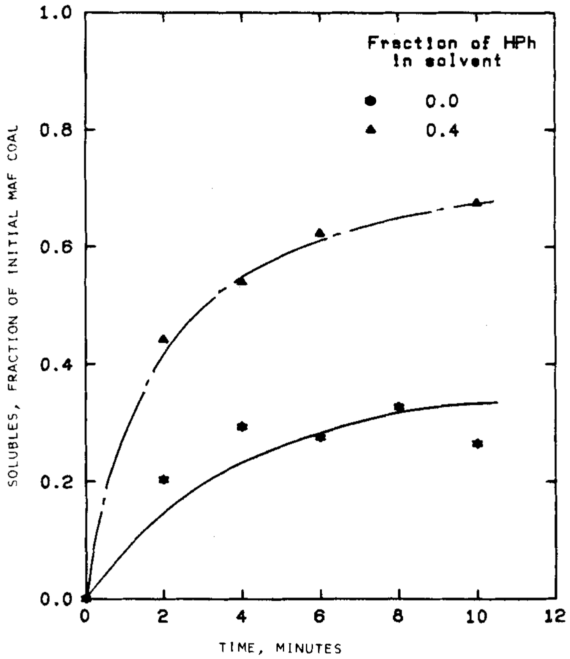

Figure 4. Effect of solvent on the conversion of Powhatan coal to toluene solubles at 723 K and *S/C* = 4.2.

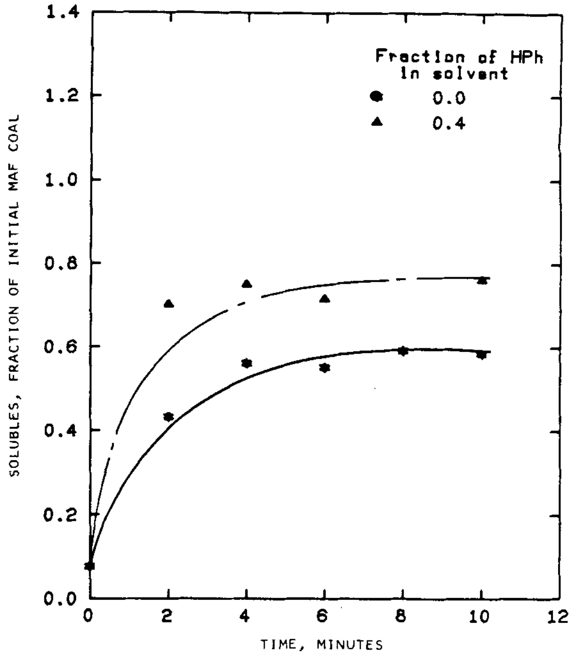

Figure 5. Effect of Solvent on the conversion of Powhatan coal to THF solubles at 723 K and *S/C* = 4.2.

TABLE 7. MODEL PARAMETERS FOR BELLE AYR COAL WITH SRC RECYCLE SOLVENT

Parameters (min^{-1})	Temperature (K) 723	673	623	573	Activation Energy (kcal/gmol)
k_1 ($C_1 + 0 \rightarrow P$)	0.32	0.16	0.03	0.02	16.5
k_2 ($C_2 + 0 \rightarrow A$)	17.89	7.78	6.17	3.84	7.9
k_3 ($C_1 \rightarrow 0$)	0.04	0.04	0.01	0.0	8.3
k_4 ($C_2 \rightarrow 0$)	9.83	3.67	1.66	1.56	10.1
k_5 ($C_1 + C_2 \rightarrow G$)	0.028	0.010	0.006	0.006	7.9
k_6 ($P \rightarrow A$)	0.17	0.16	0.16	0.15	0.8
k_7 ($A \rightarrow 0$)	0.08	0.08	0.04	0.03	6.1
k_8 ($P \rightarrow$ char)	0.16	0.16	0.01	0.0	30.1
C_1 as fraction of initial coal	0.51	0.55	0.81	0.82	

Cronauer and Ruberto, 1979; Cronauer et al., 1979). Cronauer and Ruberto (1979) used Belle Ayr coal and reported that oil depletion was strongly influenced by the nature of the solvent used. At a temperature level of 723 K, anthracene oil (AO) showed a marked loss of oils, while no oils depletion was seen when hydrogenated anthracene oil (HAO) or hydrogenated phenanthrene (HPh) were used as solvent. H_s values for AO, HAO, and HPh were reported as 0.15, 1.01, and 2.25. Hence, oil depletion was observed only for the poor donating solvent.

Experiments were conducted at temperatures of 573, 623, 673 and 723 K, for both Powhatan and Belle Ayr coals. The principal results are shown in Figures 1 through 5.

Very little difference in the product distribution is observed at the two lower levels of temperature (573 and 623 K). This is not surprising since the conversion of coal to soluble products is very small and any solvent effect would be small and perhaps masked by experimental error. However, it is noted that, at such low temperature levels, even the mixed solvents do show adduct formation as indicated by a loss of solvent.

The coal liquefaction behavior is striking for the three solvents at temperature levels of 673 K and above. Figure 1 compares the pentane-soluble fraction of the products at 673 K for the two mixed solvents. The solvent with 16 wt % HPh does show some initial loss of oils, but this loss is no longer observed for the 40 wt % HPh solvent. When the 16 wt % solvent is used at 723 K, the oils depletion is not observed as seen from Figure 2. The oils depletion vanishes at a lower temperature (673 K) for the solvent with higher hydrogen-donating capacity (40 wt % HPh) and at a higher temperature (723 K) for the solvent with lower hydrogen-donating capacity (16 wt % HPh). Therefore, the initial oils depletion progressively decreases with an increase in the hydrogen-donating capacity of the solvent. This result is also evident from an examination of Figure 3.

It appears that in coal liquefaction there are two major routes for the formation of oils from coal; one is by the reaction of the solvent with coal fragments resulting in adduct formation giving asphaltenes and preasphaltenes which in turn are converted to oils, and the other is direct formation of oils from coal. Coal may also

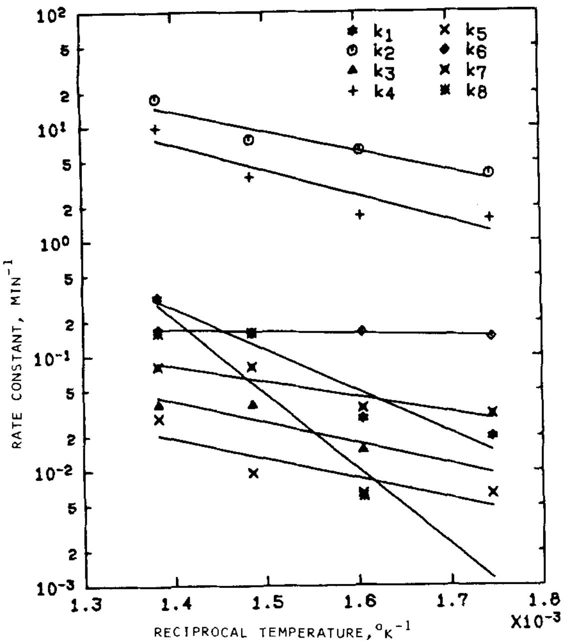

Figure 6. Arrhenius fits of the rate constants of Belle Ayr coal using SRC solvent.

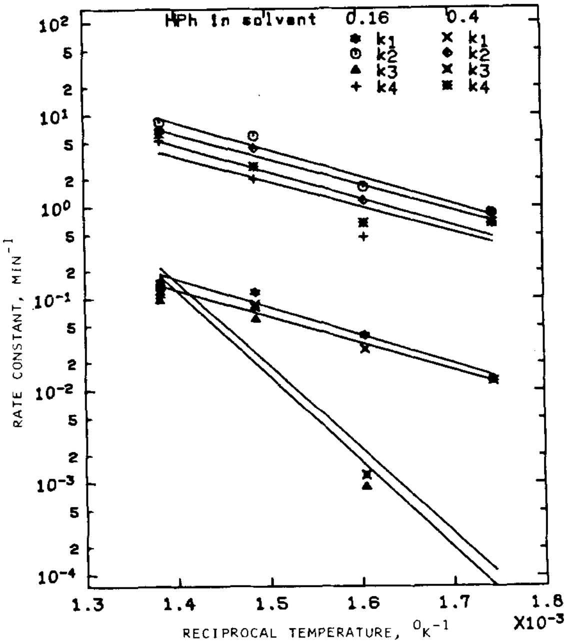

Figure 7. Arrhenius fits of the rate constants of Powhatan coal using mixed solvents.

Table 8. Model Parameters for Powhatan Coal with Mixed Solvents*

Temp. (K)	Parameter (min^{-1}) k_1 I**	k_1 II**	k_2 I	k_2 II	k_3 I	k_3 II	k_4 I	k_4 II
723	0.15	0.13	8.45	7.01	0.10	0.12	5.33	6.42
673	0.12	0.09	5.97	4.39	0.06	0.08	2.03	2.77
623	0.04	0.03	1.65	1.17	0.00	0.00	0.47	0.67
573	0.01	0.01	0.86	0.83	0.0	0.0	0.64	0.67
Activation Energy (kcal/gmol)	1.4	13.2	13.4	12.65	42.5	41.5	12.5	13.3

---

* Rate constants $k_5 - k_8$ are the same for all solvents and are as shown in Table II (Part I).
** I = SRC Recycle (0.84) + HPh (0.16).
II = SRC Recycle (0.6) + HPh (0.4).

Table 9. Model Parameters for Belle Ayr Coal with Mixed Solvent*

Temp. (K)	Parameter (min^{-1}) k_1	k_2	k_3	k_4
723	0.18	9.67	0.07	18.19
673	0.09	4.32	0.07	6.61
623	0.01	3.04	0.02	3.37
573	0.01	2.11	0.0	2.82
Activation Energy (kcal/gmol)	16.6	8.0	10.8	10.1

---

* Mixed Solvent = SRC Recycle (0.6) + HPh (0.4). Rate constants k_5-k_8 are the same as shown in Table 8.

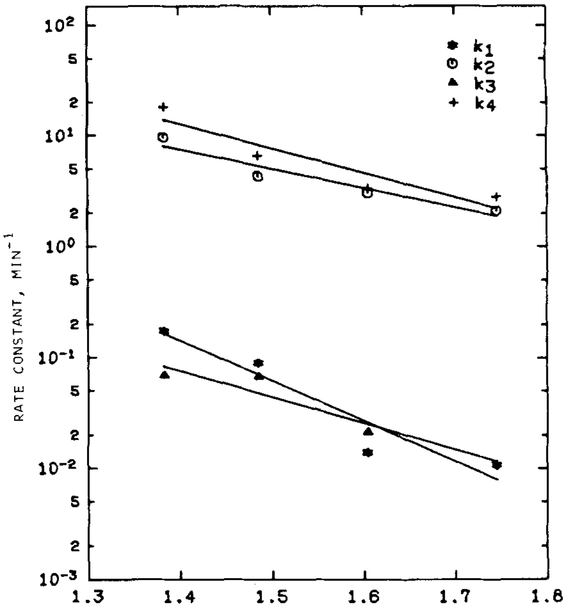

Figure 8. Arrhenius fits of the rate constants of Belle Ayr coal using mixed solvents.

be directly converted to preasphaltenes and asphaltenes, but this cannot be separated from the adduction-type reaction due to the inability to analyze the molecular structures of these fractions. The preferred route depends on the hydrogen-donating capacity of the solvent. The preference for the direct route appears to increase with an increase in the H_s value of the solvent. This explanation conforms with the conceptual picture of coal liquefaction given by Whitehurst et al. (1977). They state that if hydrogen is available (as in the case of good donor solvents) the coal free radicals will preferably combine with the available hydrogen to form stable low molecular weight species.

Figures 4 and 5 show that the toluene and THF solubles progressively increase with an increase in the hydrogen-donating capacity of the solvent. This is expected in that a better hydrogen donor solvent would have a stabilizing effect on the free radicals due to a greater availability of hydrogen.

Kinetics Model

The previously proposed kinetic model was tested against the data for the two coals with mixed solvents. The parameter values

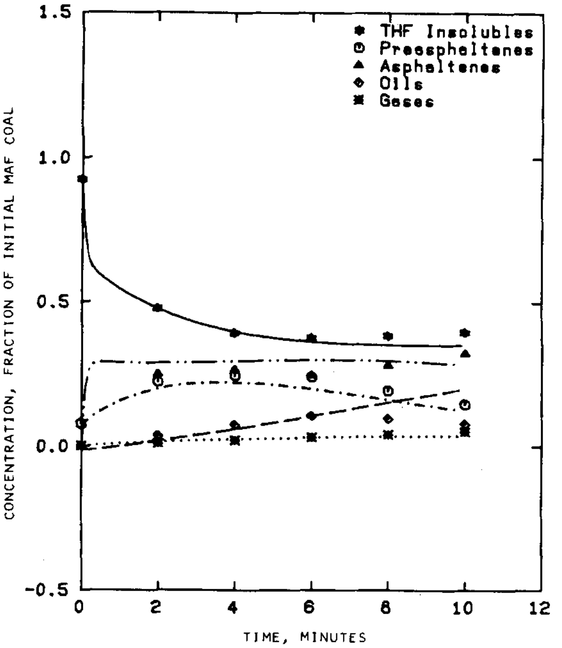

Figure 9. Model fit for Powhatan coal liquefaction; solvent of SRC recycle (0.84) Plus HPH (0.16) at 723 K.

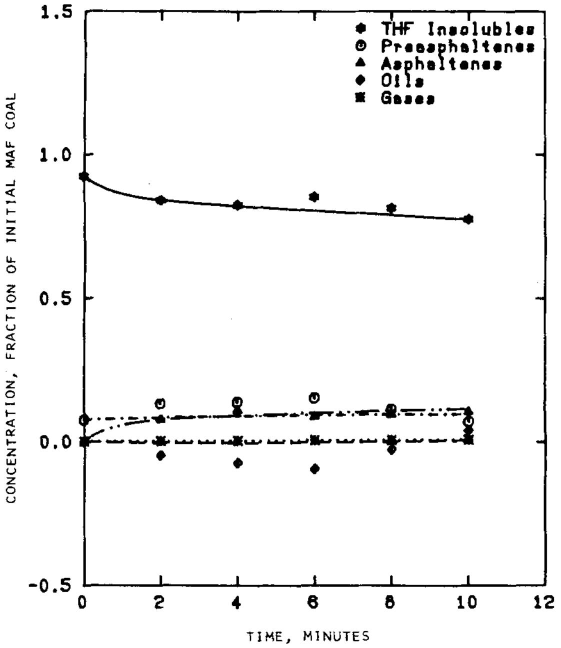

Figure 10. Model fit for Powhatan coal liquefaction; solvent of SRC recycle (0.6) Plus HPH (0.4) at 573 K.

for Belle Ayr coal and SRC-II recycle solvent are shown in Table 7. Figure 6 shows the Arrhenius plot for these constants.

In modeling the data for the mixed solvents, it was assumed that the nature of the soluble products is the same as that obtained with SRC-II recycle solvent. Consequently, the rate at which preasphaltenes and asphaltenes react is independent of the nature of the solvent. Hence, the rate constants k_5 to k_8 for each coal remain the same as previously reported. The optimized parameters with their activation energies for the mixed solvents are shown in Tables 8 and 9. Figures 7 and 8 show the Arrhenius fits to these constants.

The model gave an overall average error of 3.4% in conversions. Some of the model fits are shown in Figures 9 through 12. In these figures the lines are the model-predicted results.

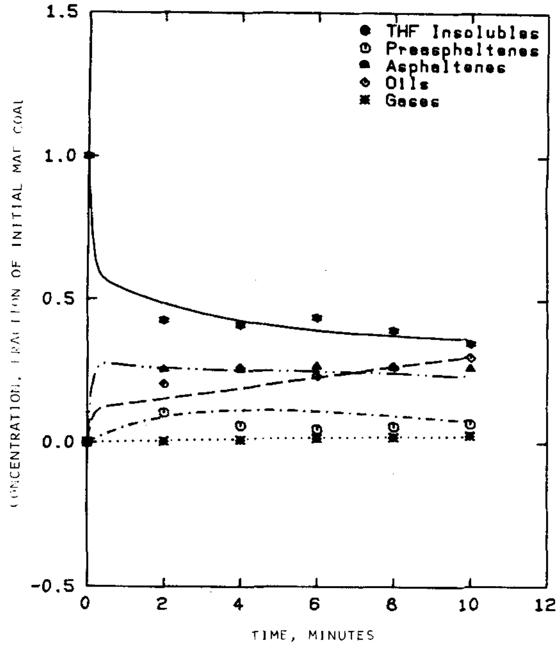

Figure 11. Model fit of Belle Ayr coal liquefaction; solvent of SRC recycle (0.6) Plus HPH (0.4) at 623 K.

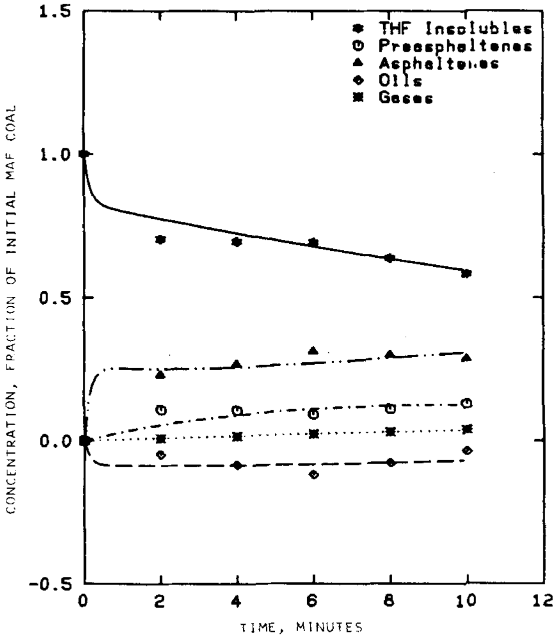

Figure 12. Model fit of Belle Ayr coal liquefaction; solvent of SRC recycle at 623 K.

With an increase in the hydrogen-donating capacity of the solvent, it is expected that the formation of oils by the direct route would be preferred over the indirect route. Consequently, with an increase in the H_s value of the solvent, rate constants k_1 and k_2 should decrease while k_3 and k_4 should increase. Examination of the rate constants reported in Tables 7 to 9 and Table 11 of Part I shows that this is indeed the case.

ACKNOWLEDGMENTS

Funding of this work was provided by the Department of Energy under Project No. DE-AC22-80PC30080, and we wish to acknowledge the coordination of S. Akhtar of the Pittsburgh Energy Technical Center. We wish to also acknowledge the comments of R. I. McNeil and A. Bruce King of Gulf Research & Development Co.

LITERATURE CITED

Abichandani, J. S., Y. T. Shah, D. C. Cronauer and R. G. Ruberto, "Kinetics of Thermal Liquefaction of Coal," *Fuel*, **61**, p. 276 (1982).

Abichandani, J. S., J. H. Wieland, Y. T. Shah, and D. C. Cronauer, "Kinetics of Short Contact Time Coal Liquefaction, I: Effect of Operating Variables," **30** (Mar., 1984).

Cronauer, D. C., and R. G. Ruberto, "Investigation of Mechanisms of Reactions Involving Oxygen-Containing Compounds in Coal," EPRI-AF-913 (1979).

Cronauer, D. C., D. M. Jewell, Y. T. Shah, R. J. Modi, and K. S. Seshadri, "Isomerization and Adduction of Hydrogen Donor Solvents under Conditions of Coal Liquefaction," *ACS, Div. Fuel Chem. Prep.*, **24**, p. 166 (1979).

Curtis, C. W., J. A. Guin, J. Jeng, and A. R. Tarrer, "Coal Solvolysis with a Series of Coal-Derived Liquids," *Fuel*, **60**, p. 677 (1981).

Derbyshire, F. J., and D. D. Whitehurst, "Study of Coal Conversion in Polycondensed Aromatic Compounds," *Fuel*, **60**, p. 655 (1981).

Epperly, W. R., "EDS Coal Liquefaction Process Development, Phase IIIB/IV," Annual Technical Progress Report, FE-2893-IT (1978).

Fant, B. T., "EDS Coal Liquefaction Process Development," Final Technical Progress Report (Jan. 1, 1976–June 30, 1977), **1**, No. FE-2353-20.

Fisher, C. H., G. C. Sprunk, A. Einer, H. J. O'Donnell, L. Clark, and H. H. Storch, "Hydrogenation and Liquefaction of Coal, Part 2—Effect of Petrographic Composition and Rank of Coal," Technical Paper 52, U.S. Bureau of Mines (1942); P. H. Given, D. C. Cronauer, W. Spackman, H. L. Lovell, A. Davis, and B. Biswas, *Fuel*, **54**, p. 40 (1975).

Furlong, M. W., R. M. Baldwin, and R. L. Bain, "Reactivity of Coal Toward Hydrogenation—Ranking by Kinetic Measurements," *Fuel*, **61**, p. 116 (1982).

King, H., and L. M. Stock, "Hydrogen Transfer Reactions of 1,2 and 1,4-Dihydronaphthalene," *Fuel*, **60**, p. 748 (1981).

Longanbach, J. R., J. R. Droege, and S. P. Chauhan, "Short Residence Time Coal Liquefaction," EPRI-AF-780 (1978).

Miller, R. L., H. F. Silver, and R. J. Hurtubise, *Ind. Eng. Chem. Process Des. Dev.*, "Upgrading of Recycle Solvent Used in the Direct Liquefaction of Wyodak Coal," **21**, p. 170 (1982).

Petrakis, L., and D. W. Grandy, "An ESR Investigation of Free Radicals in Coals and Coal Conversion," *ACS*, *Prep.*, Fuel Div., 146th Nat. ACS Meeting, Miami, p. 147 (1978).

Schweighardt, F. K., H. L. Retcofsky, and R. Raymond, "Asphaltenes from Coal Liquefaction," ACS Div. of Fuel Chem., *Preprints*, **21**, (7), p. 27 (1976).

Seshadri, K. S., R. G. Ruberto, D. M. Jewell, and H. P. Malone, "Application of Carbon-13 Nuclear Magnetic Resonance Spectroscopy to Coal Chemistry: Calculation of Transferable Hydrogen," *Fuel*, **57**, p. 549 (1978).

Storch, H. H., C. H. Fisher, C. O. Hawk, and A. Eisner, "Hydrogenation and Liquefaction of Coal," Technical Paper 642, U.S. Bureau of Mines (1943).

Traeger, R. K., T. C. Bickel, and R. M. Curlee, "Preheater Studies in Coal Liquefaction," SAND-79-0150 (1979).

Whitehurst, D. D., M. Farcasiu, T. O. Mitchell, and J. J. Dickert, Jr., "The Nature and Origin of Asphaltenes in Process Coals," EPRI-AF-480 (1977).

Whitehurst, D. D., T. O. Mitchesll, and M. Farcasiu, *Coal Liquefaction*, Academic Press, New York (1980).

Wright, C. H., R. E. Perrussel, and G. R. Pastor, "Development of a Process for Producing an Ashless, Low-Sulfur Fuel from Coal Autoclave Experiments," R&D Report No. 53, Interim Report No. 6, Contract No. 14-01-0001-496 (1975).

Manuscript received January 12, 1983; revision received May 26, and accepted June 10, 1983.

Coupled Boundary Layer Transport Involving Particles

Particle deposition by diffusiophoresis and thermophoresis is described using the linearized Fickian multicomponent diffusion equations. For these cases, the diagonal component of the diffusivity matrix for the particle is expected to be much smaller than the other diagonal components. Intuitive arguments, dimensional analyses and the standard interfacial transport theories in multicomponent form suggest that the larger boundary layer due to the larger diffusivity component describes the transition between bulk and surface values for both components.

J. A. SHAEIWITZ

Department of Chemical Engineering
University of Illinois
Urbana, IL 61801

SCOPE

Coupled transport is characterized by a flux caused by a driving force not normally associated with the flux in question. The formalism for describing coupled transport processes involves a matrix of diffusivities, yielding straightforward but somewhat more complex computational procedures. Two examples of coupled transport are multicomponent diffusion and thermal diffusion. It has been shown that diffusiophoresis, the migration of a particle in a solute gradient, is a coupled transport process, describable as a multicomponent diffusion phenomenon. When particles are involved, thermal diffusion is often called thermophoresis; and this too has been modeled as a multicomponent diffusion phenomenon. These effects appear as a "cross term" in the coupled transport model. Both diffusiophoresis and thermophoresis have been suggested as methods for enhancing particle capture from gas and liquid streams. The description of coupled transport in the boundary layer provides a straightforward method for modeling particle collection by these mechanisms.

CONCLUSIONS AND SIGNIFICANCE

A general formalism for coupled boundary layer transport for low flux situations is presented. Even though in the particle migration situations of interest the uncoupled boundary layer thicknesses differ by about two orders of magnitude, it is shown