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Enthalpy of Ternary Mixtures of Quinoline/1-Methylnaphthalene/*trans*-Decalin and Quaternary Mixtures of *m*-Cresol/Quinoline/1-Methylnaphthalene/*trans*-Decalin between 291 and 650 K at Pressures of 207 and 10342 kPa

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Experimental enthalpy measurements were made on three ternary mixtures of quinoline, 1-methylnaphthalene, and *trans*-decalin and three quaternary mixtures of *m*-cresol, quinoline, 1-methylnaphthalene, and *trans*-decalin at temperatures between 291.5 and 650 K and pressures of 206.8 and 10342.1 kPa using a reference fluid boil-off calorimeter. Enthalpy values predicted by the Soave–Redlich–Kwong equation of state agreed reasonably well with the experimental data for the ternary mixtures. However, due to the strong interaction between the acidic oxygen of *m*-cresol and the basic nitrogen of quinoline, predicted enthalpy values did not agree very well with the experimental quaternary data.

Introduction

As part of a continuing effort (*1*–*6*) to obtain enthalpies for coal-derived liquid model compounds, enthalpy measurements were obtained on three ternary mixtures of quinoline/1-methylnaphthalene/*trans*-decalin and on three quaternary mixtures of *m*-cresol/quinoline/1-methylnaphthalene/*trans*-decalin. Enthalpy measurements are important not only to provide basic data for use in engineering design and analysis, but also to check thermodynamic property correlations.

The ternary mixtures consist of a basic nitrogen compound present with an aromatic and a naphthenic compound. The quaternary mixtures also contain an acidic oxygen present with a basic nitrogen compound, an aromatic compound, and a naphthenic compound. The quaternary mixtures include interactions between the acidic oxygen in *m*-cresol and the basic nitrogen in quinoline in the presence of model hydrocarbons representative of coal-derived liquids. In a previous paper (*6*) data were reported for the *m*-cresol/1-methylnaphthalene/*trans*-decalin system, a mixture of an acidic oxygen compound with an aromatic and a naphthenic compound. Enthalpy measurements have been completed on each of the four pure fluids (*1*, *3*, *4*, *7*, *8*); however, no enthalpy measurements have to date been reported for the quinoline/1-methylnaphthalene/*trans*-decalin ternary system or for any quaternary mixtures of coal-derived liquid model compound mixtures. These multicomponent system data are useful in studying the common assumption of characterizing actual undefined mixtures, such as coal-derived liquids, in terms of lumped compound types.

Experimental Section

The experimental data were obtained with a Freon-11 ($CFCl_3$) reference fluid boil-off calorimeter system, which was discussed in detail in previous papers (*1*, *3*). In summary, energy is transferred from the flowing test fluid to boiling Freon-11. The quantity of energy transferred is determined from the mass of Freon-11 that is boiled off. Based on tests using water and *n*-heptane as standard fluids, the accuracy of the calorimeter was determined to be ±1% of the measured enthalpy difference or 3 kJ/g, whichever is greater (*1*, *3*).

The *m*-cresol, purchased from Sigma Chemical Co., and the 1-methylnaphthalene, purchased from Aldrich Chemical Co., were certified as 99+ mol %. Quinoline was initially purchased from Aldrich with a purity of 96%, with isoquinoline being a major impurity. Using a Perkin-Elmer spinning band distillation column we were able to increase the purity to at least 98.5%, with the remaining material being essentially isoquinoline. We were later able to obtain 99+% synthetic quinoline from J. T. Baker Co. with no isoquinoline. It was not possible to obtain significant quantities of high-purity *trans*-decalin, and thus the *trans*-decalin was produced chemically from a mixture of *cis*- + *trans*-decalin purchased from Fischer Chemical Co. An anhydrous aluminum chloride catalyst was used to convert the *cis*-decalin until a mixture composition of from 0.93 to 0.96 mole fraction *trans*-decalin was obtained. Details of this process are reported elsewhere (*9*). The *trans*-decalin was further distilled in a Perkin-Elmer spinning band distillation column to at least 99+%. The final chemicals, after distillation, were always greater than 98.6% pure, as determined by gas chromatography.

The compositions of each of the six mixtures as obtained from direct weighing of the samples are reported in Table I. These compositions neglect impurities in each of the pure fluids. Also reported in the table are the purities of each of the compounds that were added to form each mixture, as determined by gas chromatography. During the course of the experimental runs for each mixture the sample composition was periodically checked by gas chromatography. Samples were taken during liquid-, vapor-, and two-phase runs to ensure constant composition. Compositions determined by gas chromatography varied less than 0.1 mol % of each component; therefore the mixture composition was assumed to be constant.

Results and Discussion

Results for the enthalpy of the 60.00% quinoline/30.00% 1-methylnaphthalene/10.00% *trans*-decalin, the 20.00% quinoline/40.00% 1-methylnaphthalene/40.00% *trans*-decalin, and the 40.00% quinoline/10.00% 1-methylnaphthalene/50.00% *trans*-decalin ternary mixtures are respectively reported in Tables II, III, and IV. Enthalpies for the 30.00% *m*-cresol/15.00% quinoline/40.00% 1-methylnaphthalene/15.00% *trans*-decalin, the 15.00% *m*-cresol/30.00% quinoline/40.00% 1-methylnaphthalene/15.00% *trans*-decalin, and the 20.00% *m*-cresol/20.00% quinoline/40.00% 1-methylnaphthalene/20.00% *trans*-decalin quaternary mixtures are respectively presented in Tables V, VI, and VII. Measurements for all mixtures were obtained along isobars of 206.8 and 10 342.1 kPa over a temperature range of 291.5–650 K.

The reported enthalpy values are the experimental values corrected to a reference $H = 0$ at 291.5 K and 0.1013 MPa. The pressure of the boiling reference fluid is not controlled but varies with the barometric pressure and thus the outlet tem-

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Table I. Compositions and Analyses of Ternary and Quaternary Mixtures[a]

Ternary 1

60.00 mol % quinoline, 98.7 mol % quinoline; the impurity is isoquinoline 1.3 mol %, Aldrich

30.00 mol % 1-methylnaphthalene, 99.4 mol % 1-methylnaphthalene; one major impurity (unknown); Aldrich

10.00 mol % trans-decalin, 99.0 mol % *trans*-decalin; the one major impurity is assumed to be *cis*-decalin; Fisher

Ternary 2

20.00 mol % quinoline, 98.6 mol % quinoline; the impurity is isoquinoline 1.4 mol %; Aldrich

40.00 mol % 1-methylnaphthalene, 99.4 mol % 1-methylnaphthalene, one major impurity (unknown); Aldrich

40.00 mol % trans-decalin, 99.1 mol % *trans*-decalin; the one major impurity is assumed to be *cis*-decalin; Fisher

Ternary 3

40.00 mol % quinoline, 99.1 mol % quinoline; the impurity is unknown; synthetically made; Baker

10.00 mol % 1-methylnaphthalene, 99.4 mol % 1-methylnaphthalene; one major impurity (unknown); Aldrich

50.00 mol % trans-decalin, 99.2 mol % *trans*-decalin; the one major impurity is assumed to be *cis*-decalin; Fisher

Quaternary 1

30 mol % m-cresol; 99.9 mol % *m*-cresol; the major impurity assumed to be one of the cresol isomers; Sigma

15 mol % quinoline, 98.4 mol % quinoline; the impurity is isoquinoline 1.6 mol %; Aldrich

40 mol % 1-methylnaphthalene, 99.4 mol % 1-methylnaphthalene; one major impurity (unknown); Aldrich

15 mol % trans-decalin, 99.1 mol % *trans*-decalin; the one major impurity is assumed to be *cis*-decalin; Fisher

Quaternary 2

15 mol % m-cresol, 99.9 mol % *m*-cresol; the major impurity assumed to be one of the cresol isomers; Sigma

30 mol % quinoline, 99.1 mol % quinoline; the impurity is unknown; synthetically made; Baker

40 mol % 1-methylnaphthalene, 99.4 mol % 1-methylnaphthalene; one major impurity (unknown); Aldrich

15 mol % trans-decalin, 99.1 mol % *trans*-decalin; the one major impurity is assumed to be *cis*-decalin; Fisher

Quaternary 3

20 mol % m-cresol, 99.9 mol % *m*-cresol; the major impurity assumed to be one of the cresol isomers; Sigma

20 mol % quinoline, 99.1 mol % quinoline; the impurity is unknown; synthetically made; Baker

40 mol % 1-methylnaphthalene, 99.4 mol % 1-methylnaphthalene; one major impurity (unknown); Aldrich

20 mol % trans-decalin, 99.1 mol % *trans*-decalin; the one major impurity is assumed to be *cis*-decalin; Fisher

[a] The data are given in the following order: *amount of compound in mixture*, purity of the compound used; number and identification of impurities; where purchased.

perature of the calorimeter varies slightly from run to run. The enthalpy values reported have been corrected to a reference temperature of 291.5 K by using the heat capacity of the liquid obtained by extrapolating the liquid curve of figures such as Figures 1 and 2 to 291.5 K and taking the slope of the curve at 291.5 K. Temperature corrections were very small and never amounted to more than 1 kJ/kg. The values reported are also corrected to a pressure of 0.1013 MPa by using the correlation of Kesler and Lee (*10*, *11*) and the pseudocritical parameters for the mixture. These are computed by using the pure fluid critical properties in Table VIII obtained from ref *12* and *13* by applying the mixing rules proposed by Kesler and Lee (*10*, *11*). These values are listed in Table IX. Since these pressure corrections are quite small (less than 10 kJ/kg at the highest pressure), even a substantial error in the values obtained from the generalized correlation will have an insignificant effect on the values reported here.

Typical enthalpies for the 40.00% quinoline/10.00% 1-methylnaphthalene/50.00% *trans*-decalin ternary mixture and the 30.00% *m*-cresol/15.00% quinoline/40.00% 1-methylnaphthalene/15.00% *trans*-decaline quaternary mixture are illustrated graphically in Figures 1 and 2. The high-pressure results illustrate the effect of temperature on the liquid enthalpy, while the low-pressure data show the effect of vaporization on the sample. The data in the two-phase region, where the change in enthalpy with temperature is the greatest, are enthalpy values for a two-phase mixture with overall composition equal to that of the sample. These curves differ from results for pure fluids, where the two-phase line should be vertical. An estimated precision of ±0.5% is also illustrated in these figures, which is consistent with our estimated accuracy of ±1%. Considering these mixtures to be model coal liquid systems, the weight percent nitrogen and oxygen for each of these samples is also reported in Table IX.

Shown in the figures are the calculated results from the Soave (*14*) equation of state in conjunction with reported ideal gas enthalpies (*12*, *15*). In all calculations, interaction pa-

Table II. Enthalpy Data for the Ternary Mixture of 60.00% Quinoline, 30.00% 1-Methylnaphthalene, and 10.00% *trans*-Decalin

temp, K	press., kPa	enthalpy, kJ/kg	temp, K	press., kPa	enthalpy, kJ/kg
351.7	206.8	91.5	589.3	212.0	896.2
374.4	206.8	134.2	594.6	210.3	910.1
394.7	203.4	169.2	600.3	199.9	923.3
423.8	206.8	226.0	614.9	210.3	949.3
443.8	210.3	267.0	631.2	213.7	986.6
476.9	206.8	338.4	649.4	210.3	1018.3
503.6	210.3	405.1	373.8	10321.5	135.6
518.2	210.3	448.2	392.1	10314.6	166.0
521.9	210.3	471.8	412.6	10314.6	207.6
535.6	206.8	589.6	434.0	10397.3	244.8
540.0	208.6	730.3	450.4	10369.7	280.7
542.8	210.3	794.4	470.4	10355.9	321.1
543.4	206.8	780.4	489.6	10362.8	363.0
544.3	201.7	802.7	531.6	10342.1	459.7
548.7	208.6	810.2	558.3	10342.1	521.7
553.7	206.8	823.6	570.3	10342.1	543.3
562.5	206.8	845.6	613.5	10342.1	652.2
567.1	210.3	858.5	627.7	10342.1	686.0
574.6	206.8	865.7	648.1	10342.1	746.4

Table III. Enthalpy Data for the Ternary Mixture of 20.00% Quinoline, 40.00% 1-Methylnaphthalene, and 40.00% *trans*-Decalin

temp, K	press., kPa	enthalpy, kJ/kg	temp, K	press., kPa	enthalpy, kJ/kg
380.0	207.5	147.6	598.1	206.0	915.4
402.8	212.7	191.4	614.4	206.0	950.3
427.0	204.1	238.7	626.7	206.0	985.1
439.5	207.5	268.1	641.5	207.7	1014.5
464.5	205.8	320.9	646.4	206.0	1027.6
456.8	209.3	304.9	376.6	10300.0	146.2
480.4	205.8	354.7	384.6	10293.1	157.3
500.8	205.8	404.1	409.8	10341.4	211.8
515.5	206.0	472.7	438.9	10320.7	268.6
513.1	205.8	450.1	463.8	10204.2	328.1
516.0	206.0	481.0	482.3	10217.3	368.8
521.0	207.5	518.5	495.5	10355.2	391.8
522.2	207.5	534.1	505.7	10362.1	420.7
524.8	205.8	585.1	523.3	10362.1	451.9
527.4	207.5	617.8	536.2	10362.1	489.8
528.8	206.0	689.1	549.0	10313.9	520.7
531.4	206.0	737.7	566.9	10341.4	566.3
533.3	207.7	763.4	571.6	10348.3	573.2
537.5	207.7	778.3	583.9	10341.4	605.3
540.6	206.0	792.9	596.1	10341.4	642.8
547.1	209.5	804.2	608.2	10341.4	668.4
552.6	209.5	807.4	616.1	10341.4	694.2
558.5	207.7	820.1	627.3	10341.4	718.6
567.1	206.0	846.3	640.7	10341.4	752.5
574.6	206.0	861.3	646.4	10341.4	772.1
584.6	206.0	883.0			

Table IV. Enthalpy Data for the Ternary Mixture of 40.00% Quinoline, 10.00% 1-Methylnaphthalene, and 50.00% *trans*-Decalin

temp, K	press., kPa	enthalpy, kJ/kg	temp, K	press., kPa	enthalpy, kJ/kg
372.6	208.2	136.8	606.4	206.4	935.2
384.2	206.4	156.6	618.8	208.2	965.6
399.8	206.4	187.5	629.1	206.4	993.0
415.0	208.2	219.2	646.7	208.2	1033.0
430.9	209.9	248.1	377.0	10341.8	148.5
453.3	206.4	298.5	384.8	10355.6	166.5
466.5	206.4	325.8	403.2	10341.8	196.9
484.9	206.4	376.9	420.9	10341.8	241.1
495.1	206.4	400.9	448.7	10348.7	292.0
500.9	206.4	447.8	470.6	10383.2	345.2
508.1	206.4	507.1	485.9	10341.8	380.5
514.8	206.4	638.4	498.2	10341.8	404.9
516.6	206.4	656.4	505.7	10341.2	422.3
518.2	208.2	690.9	520.0	10341.2	454.5
519.4	208.2	707.9	533.7	10341.0	489.8
519.8	209.9	712.5	546.6	10344.7	519.5
525.9	206.4	758.5	565.2	10341.2	569.5
534.4	206.4	774.7	582.2	10341.2	609.9
540.4	206.4	785.9	603.6	10341.2	661.1
547.9	206.4	803.5	617.4	10348.1	705.1
557.9	206.4	827.4	626.8	10341.2	729.0
572.9	206.4	857.2	642.3	10341.2	769.4
591.1	206.4	900.5	648.0	10341.2	788.6

Table V. Enthalpy Data for the Quaternary Mixture of 30.00% *m*-Cresol 15.00% Quinoline, 40.00% 1-Methylnaphthalene, and 15.00% *trans*-Decalin

temp, K	press., kPa	enthalpy, kJ/kg	temp, K	press., kPa	enthalpy, kJ/kg
380.8	213.3	162.0	590.1	208.4	967.3
380.9	206.8	164.3	604.8	205.0	994.1
390.2	216.8	183.8	629.2	206.7	1051.6
409.6	208.1	223.1	644.1	208.4	1084.8
429.8	206.4	272.2	654.3	206.7	1104.0
454.8	208.1	326.3	356.3	10287.0	118.2
458.4	206.8	329.0	380.2	10328.3	168.8
464.2	209.8	345.0	403.5	10342.1	215.3
488.3	206.8	395.7	424.1	10342.1	267.5
491.7	215.0	412.6	445.0	10321.5	309.1
500.0	203.4	441.9	479.8	10300.8	382.7
506.9	208.6	460.7	503.3	10273.2	448.9
509.8	205.1	484.0	506.1	10307.7	448.8
515.9	206.8	525.9	521.1	10321.5	486.2
517.3	206.8	545.3	534.9	10321.5	525.0
520.9	212.0	575.4	545.9	10300.8	552.9
525.6	206.8	661.1	551.0	10321.5	562.5
525.8	210.2	680.2	557.1	10314.6	579.0
528.2	208.6	726.4	584.1	10321.5	645.6
528.6	206.8	768.1	599.1	10342.1	685.5
530.3	208.4	813.2	611.6	10342.1	714.0
533.0	213.6	839.0	611.7	10328.3	718.0
545.3	208.4	870.4	627.6	10342.1	761.8
562.1	206.7	904.7	639.1	10321.5	792.4
576.2	213.6	936.5	651.8	10342.1	824.8

rameters were set equal to zero. As can be seen from Figure 1, there is a good agreement between the calculated enthalpies and the experimental values in both the single- and two-phase regions for the ternary mixture (generally within 2%). In the two-phase region, enthalpy values were determined by using a flash calculation to determine the vapor–liquid equilibrium, and by calculating enthalpies of each phase from the resulting vapor/liquid mixture. Thus, in order for an equation of state to predict two-phase enthalpies, the equation must predict both vapor/liquid equilibrium and enthalpies of both phases.

The Soave equation of state has been developed for nonpolar fluids and, as was demonstrated in the previous study (*6*), was in error when used to estimate enthalpies of mixtures containing polar, associating fluids, such as *m*-cresol. However, mixtures of a basic nitrogen compound with hydrocarbons

Table VI. Enthalpy for the Quaternary Mixture of 15.00% *m*-Cresol, 30.00% Quinoline, 40.00% 1-Methylnaphthalene, and 15.00% *trans*-Decalin

temp, K	press., kPa	enthalpy, kJ/kg	temp, K	press., kPa	enthalpy, kJ/kg
382.3	209.4	156.3	633.1	204.2	1027.2
406.2	209.4	199.7	647.2	207.7	1061.2
433.4	212.8	262.5	375.7	10355.6	147.3
460.1	205.9	322.6	404.4	10362.5	203.3
473.1	212.8	347.5	423.4	10376.3	240.4
485.4	205.9	378.4	430.1	10341.5	248.1
495.4	213.9	403.1	442.4	10341.8	287.3
506.6	207.7	427.4	448.9	10341.5	291.3
510.9	209.4	439.0	462.9	10341.5	331.0
523.5	207.5	496.9	475.0	10341.5	358.9
526.8	207.5	532.3	479.9	10334.6	374.6
531.4	209.4	622.7	496.4	10348.4	401.4
533.4	137.0	709.4	509.3	10355.3	430.5
535.1	205.9	768.3	520.3	10341.5	458.1
539.0	205.9	826.2	531.4	10341.5	482.1
543.3	202.5	834.9	545.6	10334.6	524.5
554.4	204.2	857.2	568.6	10341.5	574.1
567.8	205.9	884.5	591.1	10341.5	637.0
585.5	205.9	925.4	605.1	10341.5	673.6
601.7	209.4	957.2	623.0	10341.5	722.0
619.4	207.7	991.5	640.1	10341.5	764.9
632.6	204.2	1027.9	651.3	10341.5	796.1

Table VII. Enthalpy for the Quaternary Mixture of 20.00% *m*-Cresol 20.00% Quinoline, 40.00% 1-Methylnaphthalene, and 20.00% *trans*-Decalin

temp, K	press., kPa	enthalpy, kJ/kg	temp, K	press., kPa	enthalpy, kJ/kg
375.2	213.6	146.3	618.5	206.6	1006.8
381.7	211.9	158.3	634.2	206.6	1044.5
406.7	206.7	208.4	646.0	206.6	1067.6
418.5	206.7	234.8	374.4	10341.1	154.3
436.5	208.4	275.9	386.8	10354.9	175.9
453.1	210.0	314.2	411.3	10341.1	229.8
466.8	206.7	344.8	438.2	10341.1	282.9
484.2	206.7	383.4	457.4	10341.1	331.6
502.1	210.0	427.0	477.4	10341.1	379.1
515.9	206.6	492.6	494.7	10341.1	415.7
518.0	208.3	514.4	502.3	10341.5	437.5
523.3	206.6	596.5	519.0	10362.2	471.5
525.3	206.6	624.8	531.9	10313.9	506.5
530.1	206.7	746.1	545.2	10320.8	542.8
531.2	206.6	769.4	558.6	10327.7	567.7
531.4	206.7	795.9	572.7	10341.5	614.6
538.4	206.6	835.5	585.6	10341.5	637.4
553.0	208.2	859.0	599.2	10348.4	676.3
565.0	206.6	891.9	611.6	10348.4	709.5
567.3	206.6	896.6	626.6	10341.5	752.2
576.7	206.6	914.0	640.2	10341.5	787.8
591.1	206.6	942.6	652.7	10341.5	821.9
604.7	206.6	974.5			

Table VIII. Critical Constants

compd	formula	T_c, K	P_c, atm	acentric factor	ref
m-cresol	$C_6H_4CH_3OH$	705.80	45.05	0.464	*12, 13*
quinoline	C_9H_7N	800.10	57.00	0.318	*13*
1-methyl-naphthalene	$C_{10}H_7CH_3$	772.00	35.20	0.334	*12*
trans-decalin	$C_{10}H_{16}$	687.10	31.00	0.294	*12*

Table IX. Pseudoacentric and Critical Properties for the Mixtures

mixture	T_c, K	P_c, atm	acentric factor	av mol wt	wt % nitrogen	wt % oxygen
ternary 1	777.16	45.52	0.320	133.98	6.3	
ternary 2	740.37	36.38	0.315	138.01	2.0	
ternary 3	734.47	39.17	0.308	135.01	4.2	
quaternary 1	743.26	39.49	0.365	129.43	1.6	3.7
quaternary 2	755.78	40.76	0.343	132.59	3.2	1.8
quaternary 3	745.94	39.16	0.349	131.99	2.1	2.4

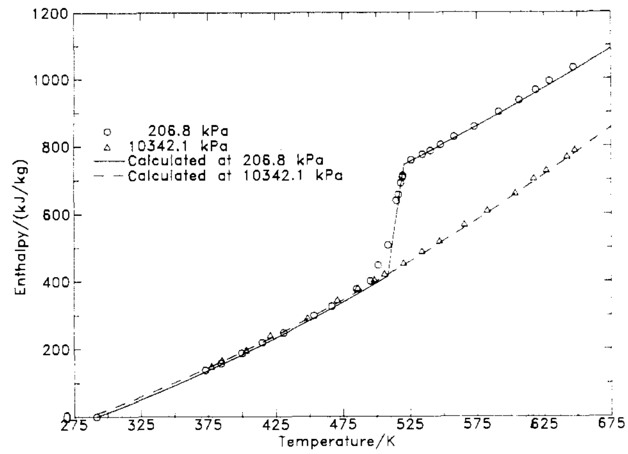

Figure 1. The enthalpy of the 40.00% (mol) quinoline/10.00% 1-methylnaphthalene/50.00% *trans*-decalin as a function of pressure and temperature relative to 291.5 K and 0.1013 kPa.

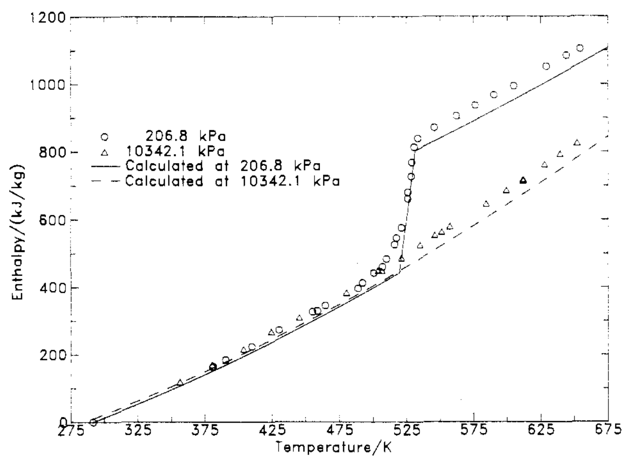

Figure 2. The enthalpy of the 30.00% (mol) *m*-cresol/15.00% quinoline/40.00% 1-methylnaphthalene/15.00% *trans*-decalin as a function of pressure and temperature relative to 291.5 K and 0.1013 kPa.

appear to be relatively ideal. For this reason, it appears that equations of state such as the Soave equation do a reasonable job in predicting phase behavior and enthalpies for such systems. However, for the quaternary mixture, illustrated in Figure 2, results contrast with previous results, where mixtures did not contain both *m*-cresol and quinoline. As reported previously (*6*) for *m*-cresol/methylnaphthalene/*trans*-decalin mixtures the agreement became worse as the *m*-cresol concentration increased. However, it was not until the *m*-cresol concentration reached 8.0% oxygen (60% *m*-cresol) that errors comparable to the errors shown on Figure 2 were observed (±8%). In the present study oxygen levels were always less than 4%. It thus appears that errors in the correlation result from both self-association of acidic oxygen, and association between acidic oxygen and basic nitrogen. Thus, correlations applicable to coal-derived liquids must take into account both of these effects.

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Registry No. Quinoline, 91-22-5; 1-methylnaphthalene, 90-12-0; *trans*-decalin, 493-02-7; *m*-cresol, 108-39-4.

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