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Heats of transport from diffusion thermoeffect measurements on binary liquid mixtures of carbon tetrachloride with benzene, toluene, 2-propanone, *n*-hexane, and *n*-octane

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Measurements of heats of transport are reported for binary liquid mixtures containing carbon tetrachloride and benzene, toluene, 2-propanone, *n*-hexane, or *n*-octane at 298 K and ambient pressure. Additionally, measurements were made at 308 K for carbon tetrachloride with benzene, toluene, and 2-propanone. The corresponding heat-mass Onsager coefficients are also reported, from which thermal diffusion ratios have been calculated based on Onsager reciprocity. These data will be used in a companion paper to report a modified Enskog method for correlation and prediction of heats of transport.

I. INTRODUCTION

Not only will a temperature gradient give rise to a heat flux in fluid mixtures, but so also will composition gradients (more appropriately, isothermal chemical potential gradients). This latter effect, known as the diffusion thermoeffect or the Dufour effect, is characterized by a transport property called the heat of transport. The heat of transport of component *i*, Q_i^* , relates the magnitude of the produced heat flux to the diffusional flux of component *i* under isothermal conditions. For many years the diffusion thermoeffect was thought to be too small for measurement in liquid mixtures because thermal conduction opposes build up of the mass-flux-induced temperature gradient and because heats of mixing were thought to obscure the effect. Ingle and Horne¹ showed mathematically that the diffusion thermoeffect should be measurable in carefully designed experiments and Rowley and Horne² were able to measure heats of transport in carbon tetrachloride + cyclohexane mixtures accurately enough to verify Onsager reciprocity. Subsequently, Rowley and co-workers have used the diffusion thermoeffect to measure heats of transport in ideal liquid mixtures,^{3,4} ternary liquid mixtures,⁵ and recently in nonideal liquid mixtures.⁶ These studies have shown that while it is true that measured temperature gradients induced in initially isothermal liquid mixtures are small due to opposition by thermal conduction, the heat of transport itself may be large. As heats of transport contain significant information concerning the actual energy-diffusional relationship of molecular interactions, their measurement is of interest; values obtained may enhance our understanding and development of generalized molecular transport models and theories.

Previous studies of the diffusion thermoeffect have focused on development and verification of the method for accurate determination of liquid mixture heats of transport.^{1,2,7} Most of the mixtures studied were nearly ideal in the thermodynamic sense. Recently, Rowley *et al.*⁶ published results of six nonideal mixtures containing carbon tetrachloride and a paraffin. In this work we report binary measurements made on nonideal systems containing carbon tetrachloride and either an aromatic or ketone constituent

with emphasis on the composition dependence of the heat of transport. These two studies provide sufficient data that preliminary testing of models may begin. In the companion paper to this one, we present a simple revision to the square-well Enskog model and test its predictive and correlational capabilities.

II. EXPERIMENTAL

All chemicals used in this study were specified by the manufacturer to be 99 + % pure and were used without further purification. Mixtures were prepared gravimetrically in glass-stoppered flasks.

The diffusion thermoeffect technique for measurement of heats of transport was utilized in this study. As before, the cell was a modification of the boundary-sharpening or Tiselius diffusion cell. The cell and experimental technique were the same as those used previously and are adequately described elsewhere.^{5,6} For clarity of discussion, only the basic principles of the experiment are repeated here. To measure the heat of transport of a binary mixture of mean mass fraction w_1 , two mixtures of different compositions (such that the average composition is w_1) are used to create an initial interface in the bore of a large stopcock. Subsequently, the interface is lowered into the cell and sharpened by withdrawal of fluid at cell half-height. Because the withdrawal rate is faster than diffusion, the sharpened interface is stable until the pumps are turned off at the start of the experiment. Temperature differences between two tiny thermocouples, located equidistant above and below the interface, are monitored as a function of time. Temperature differences rather than absolute temperatures were used in accordance with the studies of Ingle and Horne,¹ which show that heat of mixing effects provide less background response in this mode, and to eliminate the need for a thermocouple reference since one thermocouple can be referenced to the other. Initial nonuniformities in fluid temperature have been shown⁴⁻⁶ not to interfere with the analysis procedure for data obtained subsequent to the first 800 s of the experiment. The ΔT vs time data are logged at 100 s intervals over the normal 7200 s duration of the experiment and subsequently analyzed to obtain the heats of transport.

TABLE I. Thermophysical properties of the pure components.

A. Equations property	Equation							Units
Molar volume (Ref. 8)	$V = A + BT$							cm ³ /mol
Heat capacity (Ref. 8)	$C_p = C + DT + ET^2$							J/mol K
Thermal conductivity (Ref. 9)	$k = F + GT$							mW/m K
B. Constants component	A	B	C	D	10 ³ ·E	F	G	
Carbon tetrachloride	62.39	0.116	47.07	0.530	-0.8	163.60	-0.218	
Benzene	57.80	0.106	83.96	0.112	0.2	238.70	-0.323	
Toluene	72.55	0.115	41.93	0.540	-0.5	216.26	-0.292	
2-Propanone	43.61	0.102	243.25	-0.975	2.0	308.66	-0.502	
n-Hexane	79.10	0.176	266.48	-0.820	2.0	217.79	-0.328	
n-Octane	108.46	0.185	194.80	-0.004	0.7	208.56	-0.277	

III. ANALYSIS PROCEDURE

As before²⁻⁴ a numerical procedure was used to solve the partial differential equations describing the diffusion thermoeffect with isothermal initial and vanishing flux boundary conditions. The corresponding composition and temperature equations, simplified¹ for the cell geometry, types of fluids being studied, and experimental arrangement, are

$$\rho \left(\frac{\partial w_1}{\partial t} \right) = \left\{ \frac{\partial [\rho D (\partial w_1 / \partial z)]}{\partial z} \right\} \quad (1)$$

$$\rho C_p \left(\frac{\partial T}{\partial t} \right) = \left\{ \frac{\partial [k (\partial T / \partial z)]}{\partial z} \right\} + \left\{ \frac{\partial [\rho D Q^* (\partial w_1 / \partial z)]}{\partial z} \right\} + \left[\frac{\rho D M^3}{(M_1 M_2)^2} \right] \left(\frac{\partial^2 H^E}{\partial x_1^2} \right) \left(\frac{\partial w_1}{\partial z} \right)^2, \quad (2)$$

where ρ is density, C_p is specific heat, k is thermal conductivity, D is mutual diffusivity, M is mean molecular weight of the mixture, M_i is molecular weight of component i , x_1 is mole fraction of component 1, and H^E is the molar excess enthalpy. A Crank-Nicholson technique was used and the numerical solution was integrated into a nonlinear, weight-

ed, least-squares fitting program which allowed determination of Q^* from the measured temperature differences (at fixed and known thermocouple locations) as a function of time. Temperatures calculated at the grid points corresponding to the locations of the centers of the thermocouple beads and at times corresponding to measured times were subtracted from each other. Experimental temperature differences were used to adjust Q^* until a minimum in the sum of the squares of the residuals was obtained.

Also required in the analysis are the values of the thermophysical properties appearing in Eqs. (1) and (2). Table I contains relations used to compute the pure component properties which were then used in conjunction with the mixture relations given in Table II to compute the thermophysical properties at each grid point composition in the numerical solution. Only the mean temperature of the run was used in the evaluation of the properties since the induced temperature profiles are small (less than ± 0.3 K deviation from the initial value) and lead to insignificant changes in property values. This also serves to decouple Eq. (1) from Eq. (2), allowing solution of the composition equation at each time row, independent of the temperature equation so-

TABLE II. Thermophysical properties for mixtures.

A. Equations property (units)	Equation													Excess term				
Density (g/cm ³)	$\rho = M / (x_1 V_1^0 + x_2 V_2^0 + V^E)$													$V^E = x_1 x_2 \sum_{n=0}^2 A_n (1 - 2x_1)^n$				
Heat capacity (J/g K)	$C_p = (x_1 C_{p,1}^0 + x_2 C_{p,2}^0 + C_p^E) / M$													$C_p^E = x_1 x_2 \sum_{n=0}^2 B_n (1 - 2x_1)^n$				
Excess enthalpy (J/mol)														$H^E = x_1 x_2 \sum_{n=0}^2 C_n (1 - 2x_1)^n$				
Thermal conductivity (mW/m K)	$k = w_1 k_1^0 + w_2 k_2^0 + k^E$													$k^E = w_1 w_2 [G_{11}(k_{11} - k_1^0) / (w_1 + w_2 G_{11}) + G_{22}(k_{22} - k_2^0) / (w_2 + w_1 G_{22})]$				
Diffusion coefficient (m ² /s)	$D \times 10^9 = \sum_{n=0}^2 D_n x_1^n$																	
B. Constants component 2	T	A ₀	A ₁	A ₂	B ₀	B ₁	B ₂	B ₃	C ₀	C ₁	C ₂	C ₃	k ₁₂	G ₁₂	G ₂₁	D ₀	D ₁	D ₂
Benzene ¹⁰⁻¹³	298.15	0.0215	0.0247	0.0264	-16.040	-47.73	11.30	-2.90	457.9	-11.3	28.0	-8.1	132.9	1.003	0.958	1.412	0.402	0.104
	308.15	0.0215	0.0247	0.0264	-16.040	-47.73	11.30	-2.90	377.7	48.2	84.5	-22.6	125.4	1.002	0.959	1.650	0.551	0.038
Toluene ^{11,12,14,15}	298.15	-0.1712	-0.0379	0.0059	4.469	1.17	1.25	-0.89	-67.6	-30.3	5.4	-4.1	120.3	1.036	0.952	1.450	0.549	0.173
	308.15	-0.1324	0.0019	0.0408	4.469	1.17	1.25	-0.89	22.0	-6.8	30.4	22.0	115.7	1.035	0.954	1.695	0.753	0.062
2-Propanone ^{11,12,16-18}	298.15	-0.4419	0.4445	0.2931	17.482	-0.94	0.46	-0.65	637.3	962.6	707.5	323.4	105.5	0.708	1.014	1.684	-1.557	3.471
	308.15	-0.2989	0.4542	0.1394	17.482	-0.94	0.46	-0.65	1021.9	941.9	717.5	309.1	97.0	0.716	1.013	2.033	-1.835	3.754
n-Hexane ^{11,12,19,20}	298.15	0.1777	0.1127	0.0309	-4.680	-2.85	0.00	26.96	1267.1	270.4	35.4	-226.8	102.9	0.737	1.168	1.482	1.632	0.739
n-Octane ^{11,12,21,22}	298.15	1.2538	0.3037	0.1342	-7.100	-4.31	11.67	-8.06	1493.5	424.1	101.2	221.3	106.2	0.628	1.336	1.233	1.283	0.063

lution. Because the relaxation time of diffusion is slow compared to that of thermal conduction, the composition profile of the previous time step could be used in the temperature solution without loss of accuracy. Initially, very short time steps (2 s) were used when the composition gradient was steep. These were gradually expanded to 100 s steps as the composition gradient was diminished.

IV. RESULTS

The heats of transport of carbon tetrachloride obtained in this study are shown in Table III. A Monte Carlo error analysis program was developed to determine the expected precision of the heats of transport. One thousand replicated runs, generated by including random normal deviate errors based on the uncertainty of each of the experimental variables, were analyzed in the same manner as the actual experimental data. To minimize computer CPU time, the analytical approximate solutions to Eqs. (1) and (2) by Ingle and Horne¹ were used in these simulations rather than the complete numerical solution used in the actual data analysis.

Using the following uncertainties in measured and input variables, ± 2.6 mK in T , $\pm 1\%$ in ρ , $\pm 4\%$ in D , $\pm 1\%$ in C_p , and $\pm 4\%$ in k , we obtained an uncertainty in fitted heats of transport of about $\pm 4\%$. Nearly $\pm 1.6\%$ of this results from uncertainties in the measured temperature. A worst possible case analysis indicated a maximum possible error of about $\pm 9\%$. Since the Monte Carlo simulations do not account for possible experimental and procedural errors, we estimate $\pm 5\%$ to be the experimental precision. Rowley and Horne,² using a different type of cell, were able to obtain heats of transport which agreed within $\pm 2\%$ of values obtained from thermal diffusion data, but we estimate the accuracy of the results here at about $\pm 6\%$. Replicate experiments were performed on the carbon tetrachloride + benzene and *n*-hexane systems to experimentally test the reproducibility of the measurements. The replicates were all within $\pm 2.9\%$ of the mean values.

Thermal diffusion factors α are indications of the degree of separation that can be achieved by thermal diffusion. Heats of transport and thermal diffusion factors are not independent, but are directly related by Onsager reciprocity,

TABLE III. Heats of transport of carbon tetrachloride (1) in mixtures at ambient pressure.

Comp. #2	$T(K)$	w_1	x_1	$Q^*(kJ/kg)$	$1 + \Gamma_{11}$	$10^7 \cdot \Omega_{10}(kg/m s)$	α
Benzene	298.15	0.1	0.0534	22.6	0.985	2.101	2.409
	298.15	0.3	0.1787	28.5	0.959	6.042	2.249
	298.15	0.5	0.3368	30.5	0.940	7.370	1.698
	298.15	0.7	0.5423	31.5	0.935	5.930	1.159
	298.15	0.9	0.8205	33.9	0.965	2.384	0.742
	308.15	0.1	0.0534	22.8	0.987	2.367	2.348
	308.15	0.3	0.1787	29.1	0.959	6.922	2.221
	308.15	0.5	0.3368	31.7	0.941	8.626	1.707
	308.15	0.7	0.5423	32.0	0.937	6.788	1.137
	308.15	0.9	0.8205	34.9	0.966	2.759	0.739
Toluene	298.15	0.1	0.0624	20.5	1.000	2.104	1.879
	298.15	0.3	0.2043	25.2	0.993	5.803	1.789
	298.15	0.5	0.3746	26.9	0.986	7.037	1.444
	298.15	0.7	0.5829	29.0	0.980	6.008	1.140
	298.15	0.9	0.8453	32.3	0.983	2.645	0.889
	308.15	0.1	0.0624	21.0	0.999	2.395	1.864
	308.15	0.3	0.2043	25.9	0.993	6.672	1.780
	308.15	0.5	0.3746	27.4	0.986	8.072	1.423
	308.15	0.7	0.5829	29.9	0.980	7.002	1.138
	308.15	0.9	0.8435	33.1	0.983	3.052	0.881
2-Propanone	298.15	0.1	0.0403	25.1	0.955	3.852	3.561
	298.15	0.3	0.1393	27.3	0.849	9.193	2.842
	298.15	0.5	0.2741	27.9	0.726	10.284	2.064
	298.15	0.7	0.4684	30.0	0.594	8.292	1.491
	298.15	0.9	0.7726	33.2	0.582	3.044	0.798
	308.15	0.1	0.0403	26.1	0.955	4.211	3.583
	308.15	0.3	0.1393	28.4	0.852	10.103	2.853
	308.15	0.5	0.2741	28.8	0.729	11.363	2.054
	308.15	0.7	0.4684	30.5	0.601	9.187	1.451
	308.15	0.9	0.7726	34.3	0.595	3.528	0.780
<i>n</i> -Hexane	298.15	0.1	0.0586	25.9	0.985	3.588	2.545
	298.15	0.3	0.1936	33.0	0.948	10.259	2.522
	298.15	0.5	0.3591	36.8	0.900	12.980	2.150
	298.15	0.7	0.5666	44.5	0.849	12.233	1.925
	298.15	0.9	0.8345	55.8	0.855	5.415	1.594
<i>n</i> -Octane	298.15	0.1	0.0762	19.6	1.003	1.942	1.510
	298.15	0.3	0.2414	28.0	1.000	6.324	1.839
	298.15	0.5	0.4262	34.1	0.980	9.048	1.923
	298.15	0.7	0.6341	41.0	0.925	9.278	2.042
	298.15	0.9	0.8699	52.8	0.858	5.196	2.333

$\Omega_{10} = \Omega_{01}$. The Onsager heat-mass coefficient Ω_{01} can be calculated from⁵

$$\Omega_{01} = \rho D Q_1^* M_1 M_2 w_1 w_2 / [MRT(1 + \Gamma_{11})], \quad (3)$$

where Γ_{11} is the so-called thermodynamic factor, related to the activity coefficient of component 1, γ_1 , by

$$\Gamma_{11} = \left(\frac{\partial \ln \gamma_1}{\partial \ln x_1} \right)_{T,P}. \quad (4)$$

Values of Γ_{11} were obtained from activity coefficient data¹² in accordance with the above two equations and are compiled in Table III. The coefficient Ω_{10} can be related to the thermal diffusion factor by

$$\Omega_{10} = \rho D \alpha x_1 x_2. \quad (5)$$

Equating Eqs. (3) and (5) on the basis of Onsager reciprocity yields

$$\alpha = Q_1^* (M_1 M_2)^2 / [M^3 RT(1 + \Gamma_{11})]. \quad (6)$$

Values of α obtained from the measured heats of transport in conjunction with Eq. (6) are also given in Table III.

Story and Turner²³ also reported thermal diffusion factors on this system measured in a flow thermal diffusion cell at the same temperatures reported here. Their results are compared to ours in Fig. 1. Agreement is acceptable at compositions below 50 mol % carbon tetrachloride, but deviate significantly at higher compositions. They only made one measurement at 308.15 K, but the very slight decrease in thermal diffusion factor with an increase in temperature is in agreement with our results. Guzzi and Tyrrell²⁴ have also reported thermal diffusion factors at 298.15 K, their results are also shown in Fig. 1. Again agreement in the lower carbon tetrachloride composition range is not terrific, but acceptable. Drickamer *et al.*²⁵ reported values at 313 K. Although this does not correspond to the temperature for the data reported here, our results indicate very little temperature dependence for α and so a direct comparison is not unreasonable. As shown in Fig. 1 there is considerable dis-

agreement and scatter between all four of the data sets except in the region between $x_1 = 0.2$ and $x_1 = 0.4$. Our data show a stronger composition dependence over the entire composition range than the others. Story and Turner, on the other hand, show a rapid decrease in α near the pure component limits.

V. DISCUSSION OF RESULTS

It is of interest to plot the heats of transport as a function of composition to ascertain the possibility of obtaining a pure component value for the heat of transport of carbon tetrachloride independent of which system the value was determined from. Rather than plot Q_1^* , we choose to plot MQ_1^* in Figs. 2 and 3 because we find it to be more linear in mole fraction. Values of MQ_1^* were fitted to a second-order polynomial equation in mole fraction using a least-squares regression program; the equation and resultant coefficients are shown in Table IV. The smoothing curves represented by these equations are the lines given in Fig. 2 at 298.15 K and in Fig. 3 at 308.15 K.

From Fig. 2 it is obvious that Q_1^* in the pure carbon tetrachloride limit is not the same for the five systems. Within the experimental uncertainty, benzene, toluene, and 2-propanone yield the same value while the two paraffins yield a different value. The infinite dilution values for Q_1^* are remarkably similar for all the systems studied. Since for a binary system $Q_2^* = -Q_1^*$, this infinite dilution value represents the pure component limit for the second component. Further studies are mandated to test this observation on other groups of mixtures (each group containing a different common component). If similar results are found for those groups, it may be possible to develop a predictive technique based on tabulated infinite dilution values.

While the observed temperature dependence of the heats of transport is small, it is definitely shown that Q_1^* decreases with increasing temperature. This trend is more

FIG. 1. Thermal diffusion factor α for carbon tetrachloride (1) + benzene mixtures determined in this study (+, 298.15 K; $\square$, 308.15 K) compared to values reported in Ref. 23 ($\diamond$, 298.15 K; $\blacklozenge$, 308.15 K), Ref. 24 (*, 298.15 K), and Ref. 25 (O, 313 K).

FIG. 2. Product of mean molecular weight and heat of transport at 298.15 K for carbon tetrachloride (1) and benzene (—; $\bullet$), toluene (---; $\blacksquare$), acetone benzene (···; $\blacktriangle$), *n*-hexane (— · —; +), and *n*-octane (---; *).

FIG. 3. Product of mean molecular weight and heat of transport at 308.15 K for carbon tetrachloride (1) and benzene (—; ●), toluene (---; ■), and acetone (---; ▲).

observable in terms of the Onsager coefficients illustrated in Fig. 4.

VI. CONCLUSIONS

Diffusion thermoeffect measurements have been made at 298.15 and 308.15 K for carbon tetrachloride plus benzene, toluene, and 2-propanone mixtures and at 298.15 K for carbon tetrachloride plus *n*-hexane and *n*-octane mixtures. From comparison of these ΔT vs t measurements to the numerical solution of the appropriate diffusion thermoeffect boundary value problem, heats of transport have been regressed. Calculation of Onsager coefficients and thermal diffusion factors was also carried out using activity coefficient information. Measured heats of transport are considered to be reproducible to about $\pm 5\%$ as supported by Monte Carlo error analysis procedures and limited replicate studies. Accuracy is assumed to be about $\pm 6\%$, although calculated thermal diffusion factors do not agree with previous literature values within the combined specified accuracy limits, except in the range of $x_1 = 0.2$ to $x_1 = 0.4$. However, none of the previous data sets are in agreement outside of this composition range.

TABLE IV. Coefficients for the smoothing equation $Q_1^*M = \sum_{n=0}^2 Q_n x_1^n$ (kJ/mol) for mixtures of the indicated component with carbon tetrachloride (1) at ambient pressure.

Comp. #2	<i>T</i> (K)	<i>Q</i> ₀	<i>Q</i> ₁	<i>Q</i> ₂	AAD (%)
Benzene	298.15	1.702	4.599	− 1.126	2.9
	308.15	1.719	4.821	− 1.233	3.5
Toluene	298.15	1.813	3.502	− 0.203	2.1
	308.15	1.860	3.609	− 0.222	2.2
2-Propanone	298.15	1.457	3.071	0.923	1.0
	308.15	1.539	2.900	1.242	1.3
<i>n</i> -Hexane	298.15	2.163	4.281	3.153	2.3
<i>n</i> -Octane	298.15	2.030	4.712	2.177	2.9

FIG. 4. Onsager coefficient Ω_{01} for carbon tetrachloride (1) and benzene (—; ○), toluene (---; □), and acetone (---; △). Open symbols are for 298.15 K; closed for 308.15 K.

The results show that heats of transport decrease with increasing temperature. Furthermore, it appears that all five systems yield approximately the same value for the infinite dilution value of the heat of transport of carbon tetrachloride. While the pure component limit of the heat of transport of carbon tetrachloride is similar for mixtures containing benzene, toluene, and 2-propanone, it differs that from that found for *n*-hexane and *n*-octane. It is hoped that these data will form the start of a data base from which models and theories for the heat of transport may be developed and tested. To this end, we present in a subsequent, but related, paper a revision to the square-well Enskog theory which is able to correlate the data well with one interaction coefficient or predict the data adequately with no mixture adjustable parameters.

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