

Thermal Diffusion in Some Binary Liquid Mixtures by the Flowing Cell Method

P. Poty, J. C. Legros, and G. Thomaes

Université Libre de Bruxelles, Belgium

(Z. Naturforsch. **29 a**, 1915–1916 [1974];
received September 27, 1974)

In this paper we present measurements of the Soret coefficient (D'/D) by means of a flowing cell technique for disulfide-3 methylpentane and for the three associated solutions: carbon tetrachloride-ethanol, carbon tetrachloride-isopropanol and water-isopropanol.

For our study^{1,2} of the hydrodynamic stability of a binary liquid mixture heated from below (Bénard problem), we have to know the thermal diffusion coefficient (D'/D), for liquids also called Soret coefficient, of the investigated mixtures.

The most interesting cases are the ones corresponding to $D'/D < 0$. (D'/D is positive when the denser component migrates towards the cold plate.) In this case, the Bénard system is stabilized by the concentration gradient, and under some conditions oscillatory behaviour has already been observed^{3,4}. That is the reason why we measured the Soret coefficient for some systems by means of the flowing cell technique we have already described^{5,6}.

These systems are: ethanol – carbon tetrachloride, isopropanol – carbon tetrachloride, isopropanol – water and carbon disulfide-3 methylpentane.

For the system CS_2 – 3 methylpentane, our results are in contradiction to the ones given by Brock⁷. We observed that this coefficient is small and positive at a mean temperature of 24 °C.

The following systems were chosen because, in 1950, Prigogine, de Brouckère and Amand⁸ reported that for associated solutions, the Soret coefficient D'/D was found to vary considerably with concentration, in contradistinction to what had been observed with perfect solutions. With inert solvents such as benzene and cyclohexane, the sign of D'/D for alcohol solutions was reversed when passing from low to high concentrations. The reversal ob-

served with inert solvents is probably related to an entropy increase due to destruction of complexes which takes place when a molecule of alcohol and a molecule of inert solvent exchange their positions. In this work, carbon tetrachloride was considered as an inert solvent. That is why we studied the two systems carbon tetrachloride with ethanol and with isopropanol. But we did not observe the expected reversal of sign: the observed Soret coefficient is always positive.

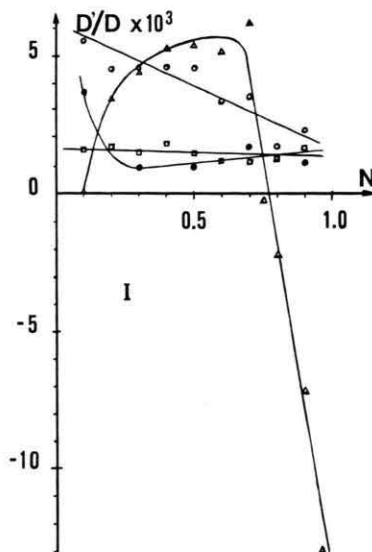


Fig. 1. Soret coefficients for the systems carbone disulfide-3 methylpentane (●), carbon tetrachloride-ethanol (○), carbon tetrachloride-isopropanol (□) and water-isopropanol (△) as a function of concentration; N is the mass fraction of the component named first. Mean temperature 24 °C, temperature difference = 10 °C, flow rates = 10 ml/h.

The last system studied was water – isopropanol. As expected, there is an inversion of D'/D for high concentrations in water. The results are reported in Table 1 and Figure 1. All products are “pro analysis” grade products from Merck and Baker Companies.

Reprint requests to Dr. J. C. Legros, Service de Chimie Physique E.P., Faculté des Sciences Appliquées, Université Libre de Bruxelles, 50, av. F. D. Roosevelt, B-1050 Bruxelles, Belgium.

¹ J. C. Legros, J. K. Platten, and P. G. Poty, Phys. Fluids **15**, 1383 [1972].

² J. C. Legros, P. G. Poty, and G. Thomaes, Physica **64**, 481 [1973].

³ a) J. K. Platten and G. Chavepeyer, Phys. Lett. **40 A**, 287 [1972]. — b) J. K. Platten, G. Chavepeyer, and J. Tellier, Phys. Lett. **44 A**, 479 [1973].

⁴ D. T. J. Hurle and E. Jakeman, J. Fluid Mech. **47**, 667 [1971].

⁵ G. Thomaes, J. Chem. Phys. **53**, 407 [1956].

⁶ J. C. Legros, D. Rasse, and G. Thomaes, Physica **57**, 585 [1972].

⁷ J. R. Brock, Symposium on the less common means of separation, Chem. Engns. Inst. (1963).

⁸ I. Prigogine, L. de Brouckère, and R. Amand, Physica **16**, 851 [1950].



Table 1.

Mean temperature: 24 °C

RUN	<i>N</i>	$\Delta N \times 10^4$	$D'/D \times 10^3$	RUN	<i>N</i>	$\Delta N \times 10^4$	$D'/D \times 10^3$
CS ₂ -3 methylpentane system							
1	0.9	3.71	+1.1	6	0.5	9.44	+1.01
2	0.9	3.68	+1.09	7	0.3	7.51	+0.95
3	0.7	13.43	+1.71	8	0.3	7.45	+0.95
4	0.7	13.39	+1.7	9	0.1	12.82	+3.8
5	0.5	9.35	+1	10	0.1	12.82	+3.66
CCl ₄ -C ₂ H ₅ OH system							
1	0.1	18.64	+5.52	10	0.5	42.97	+4.58
2	0.1	19.00	+5.63	11	0.6	29.65	+3.29
3	0.2	27.09	+4.52	12	0.6	29.75	+3.31
4	0.2	27.31	+4.55	13	0.7	27.67	+3.51
5	0.3	35.91	+4.56	14	0.7	27.61	+3.51
6	0.3	36.15	+4.59	15	0.8	10.35	+1.73
7	0.4	41.47	+4.61	16	0.8	10.39	+1.73
8	0.4	41.60	+4.62	17	0.9	7.98	+2.36
9	0.5	42.76	+4.56	18	0.9	8.00	+2.37
CCl ₄ -(CH ₃) ₂ CHOH system							
1	0.1	5.41	+1.60	17	0.5	14.18	+1.51
2	0.1	5.41	+1.60	18	0.5	14.14	+1.51
3	0.1	5.41	+1.60	19	0.5	14.10	+1.50
4	0.1	5.41	+1.60	20	0.5	14.21	+1.52
5	0.2	10.04	+1.67	21	0.6	11.15	+1.24
6	0.2	10.22	+1.70	22	0.6	11.17	+1.24
7	0.2	10.16	+1.69	23	0.6	11.33	+1.26
8	0.2	10.10	+1.68	24	0.6	11.19	+1.24
9	0.3	11.75	+1.49	25	0.7	9.41	+1.20
10	0.3	11.58	+1.47	26	0.7	9.48	+1.20
11	0.3	11.77	+1.49	27	0.7	9.50	+1.21
12	0.3	11.82	+1.50	28	0.7	9.49	+1.21
13	0.4	16.53	+1.84	29	0.8	7.68	+1.28
14	0.4	16.36	+1.82	30	0.8	7.70	+1.28
15	0.4	16.41	+1.82	31	0.9	5.84	+1.73
16	0.4	16.31	+1.81	32	0.9	5.89	+1.75
H ₂ O-(CH ₃) ₂ CHOH system							
1	0.9	22.96	-6.8	23	0.5	51.42	+5.5
2	0.9	24.09	-7.1	24	0.4	47.79	+5.3
3	0.9	25.10	-7.4	25	0.4	48.54	+5.4
4	0.9	22.92	-6.8	26	0.4	47.79	+5.3
5	0.9	23.88	-7.1	27	0.4	47.42	+5.3
6	0.9	24.49	-7.3	28	0.3	34.92	+4.4
7	0.8	15.44	-2.6	29	0.3	34.92	+4.4
8	0.8	15.10	-2.5	30	0.3	34.64	+4.4
9	0.8	15.70	-2.6	31	0.3	33.93	+4.3
10	0.8	14.00	-2.3	32	0.2	21.49	+3.6
11	0.7	48.00	+6.1	33	0.2	21.91	+3.7
12	0.7	48.96	+6.2	34	0.2	20.53	+3.4
13	0.7	50.57	+6.4	35	0.2	21.49	+3.6
14	0.7	49.75	+6.3	36	0.1	0	0
15	0.7	50.39	+6.4	37	0.1	0	0
16	0.6	46.46	+5.2	38	0.1	0.62	-0.2
17	0.6	46.58	+5.2	39	0.75	1.53	-0.2
18	0.6	45.57	+5.1	40	0.75	1.42	-0.2
19	0.6	44.90	+5.0	41	0.75	1.03	-0.2
20	0.5	50.03	+5.3	42	0.95	24.03	-13.5
21	0.5	52.25	+5.6	43	0.95	22.96	-12.9
22	0.5	51.02	+5.4	44	0.95	23.21	-13.0
				45	0.95	23.11	-13.0