J. M. Sørensen
W. Arlt

LIQUID-LIQUID EQUILIBRIUM DATA COLLECTION

Binary Systems

Chemistry Data Series

Vol. V, Part 1

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PREFACE OF AUTHORS

This work consists of three Parts. Part 1 deals with binary liquid-liquid equilibria and Parts 2 and 3 with ternary liquid-liquid equilibria.

The correlation and prediction of liquid-liquid equilibria (LLE) using models for liquid phase non-idealities is a subject within chemical engineering which is not yet mastered quantitatively. Model inadequacies may be one explanation to this. However, this work will show that the results may be quite good with existing models if they are used properly, i. e. if the parameter estimation procedure as well as the data base are chosen carefully.

In addition to an extensive collection of experimental binary, ternary, and quaternary data, this work contains NRTL and UNIQUAC parameters reduced from these data. For each ternary system (Parts 2 and 3) we give a specific set of NRTL and UNIQUAC parameters which emphasizes the distribution ratio of the solute at small concentrations. This makes semi-quantitative extraction calculations possible for many systems. We also include a table of UNIQUAC parameters for the A-B interaction reduced from a large number of systems with components A, B, and any third component. These parameters will usually be better for predicting multicomponent LLE than parameters based on VLE-data.

The work leading to these three books began in 1977 at Instituttet for Kemiteknik, Lyngby, Denmark, where Aa. Fredenslund started a project called „A Group Contribution Method for Predicting Liquid-Liquid Equilibria". An initial step in this project was to investigate how existing molecular models (e.g. NRTL and UNIQUAC) behave in LLE calculations. For this purpose a parameter estimation procedure and a large data base were established.

The above project was close to research plans at Dortmund University, and a collaboration was initiated in 1978 with the purpose of jointly preparing this publication. The data collection was extended, and the authors have profited very much by the experience of J. Gmehling and U. Onken who started Volume I of this series („Vapor-Liquid Equilibrium Data Collection").

Simultaneously the parameter estimation procedure was further refined under daily guidance by Aa. Fredenslund and P. Rasmussen to whom the authors are very grateful.

The parameter estimation from ternary LLE-data involves many numerical problems. The authors wish to thank M. L. Michelsen (Instituttet for Kemiteknik, Lyngby) for providing the final version of the estimation procedure. Finally, we dedicate special thanks to T. Magnussen (DECHEMA, Frankfurt/Main) for many fruitful discussions and help and to colleagues and students in Lyngby and Dortmund for various kinds of assistance.

The authors hope that the data collection and parameters of this work will be of use to the industry. We also hope that the availability of the large amount of data will facilitate the testing of new models at universities.

The authors

PREFACE OF EDITORS

Subjects of the DECHEMA Chemistry Data Series are the physical and thermodynamic property data of chemical compounds and mixtures essentially for the fluid state covering PVT data, heat capacity, and entropy data, phase equilibrium data, transport and interfacial tension data.

The main purpose is to provide chemists and engineers with data for process design and development. For computer based calculations in process design appropriate correlation methods and accurate data must be used. These are only in some cases available in the open literature. For that reason the most urgent requirement regarding the publication of data is to offer classified and critically evaluated data, thus giving an impression which of them are reliable or not. This will be the goal of the series.

DECHEMA gives the opportunity to authors especially from universities to publish not only their theoretical results, but also their measured or compiled data, most often a large amount, that would otherwise never have been published.

After that a successful group contribition method for the prediction of vapor-liquid equilibria (UNIFAC) had been presented to the scientific sommunity several years ago, it was the need for a similar treatment of liquid-liquid equilibria that led to a cooperation between the Dechema Data Compiler Development Group and Professor Fredenslund at the Instituttet for Kemiteknik in Lyngby, who has much experience in this field.

Before this background, J. M. Sørensen and W. Arlt, who is already co-author of Volume I of the Series, have collected the mutual solubility data of more than 2000 binary, ternary and quaternary mixtures of organic liquids.

This compilation is now being published in 3 parts of Volume V of this series, giving not only measured data but also evaluated correlation constants and recommended values. We hope that this gives particularly the users an instrument that will allow them to solve their problems considerably more easily and more quickly than before.

Frankfurt/Main, October 1979

Dieter Behrens
Reiner Eckermann

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LIST OF SYMBOLS

Symbols only used in the model expressions are not explained here.

a	activity or model parameters
$\triangle G$	molar Gibbs energy of mixing
$\triangle G_{id}$	ideal molar Gibbs energy of mixing
G^E	excess molar Gibbs energy
n	number of moles
P	pressure
q	constant in the UNIQUAC model
r	constant in the UNIQUAC model
R	the gas constant
T	temperature
x	liquid mole fraction
y	vapor mole fraction

Greek symbols

α	NRTL non-randomness parameter
γ	activity coefficient

Superscripts

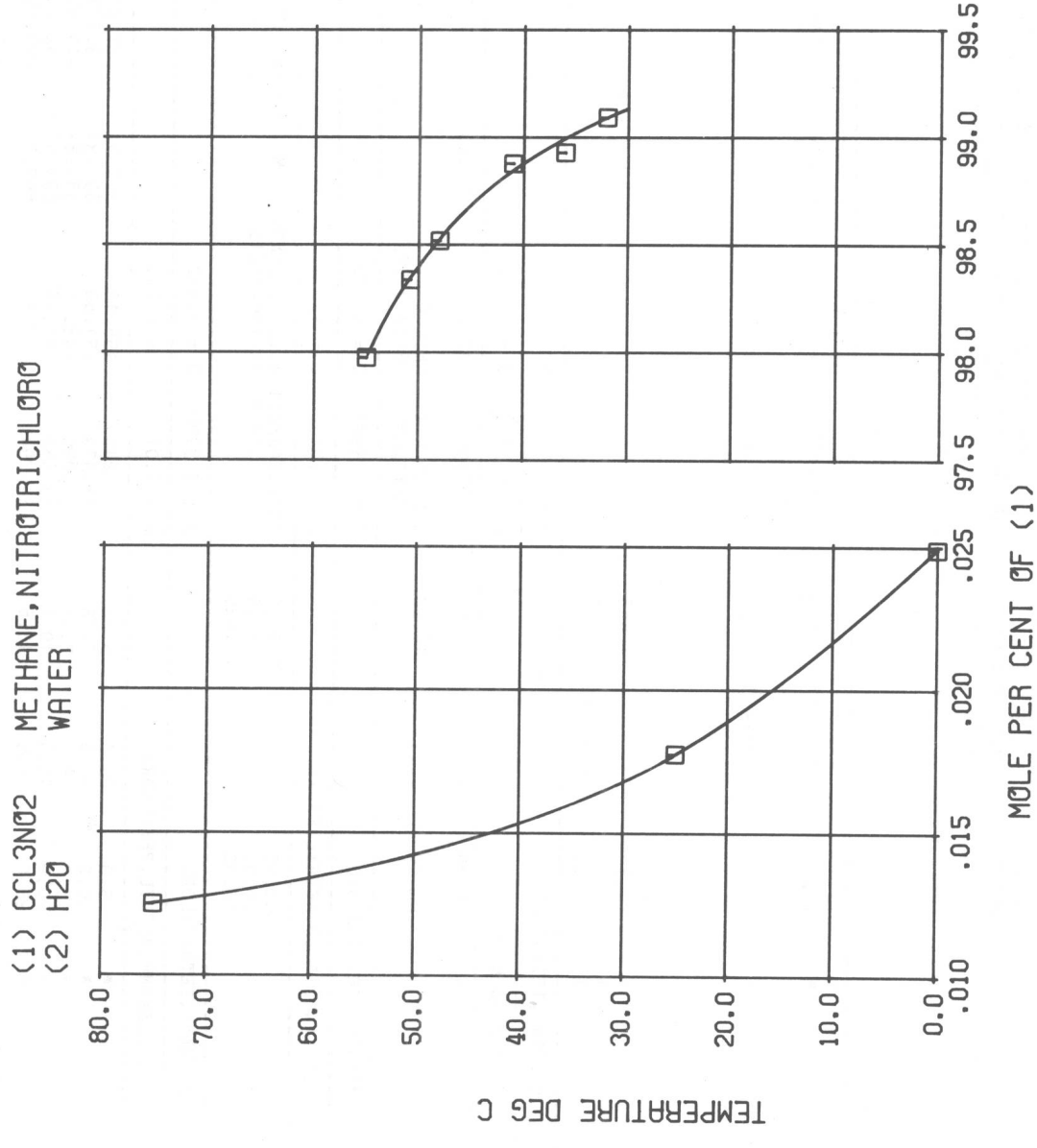
I, II	phases

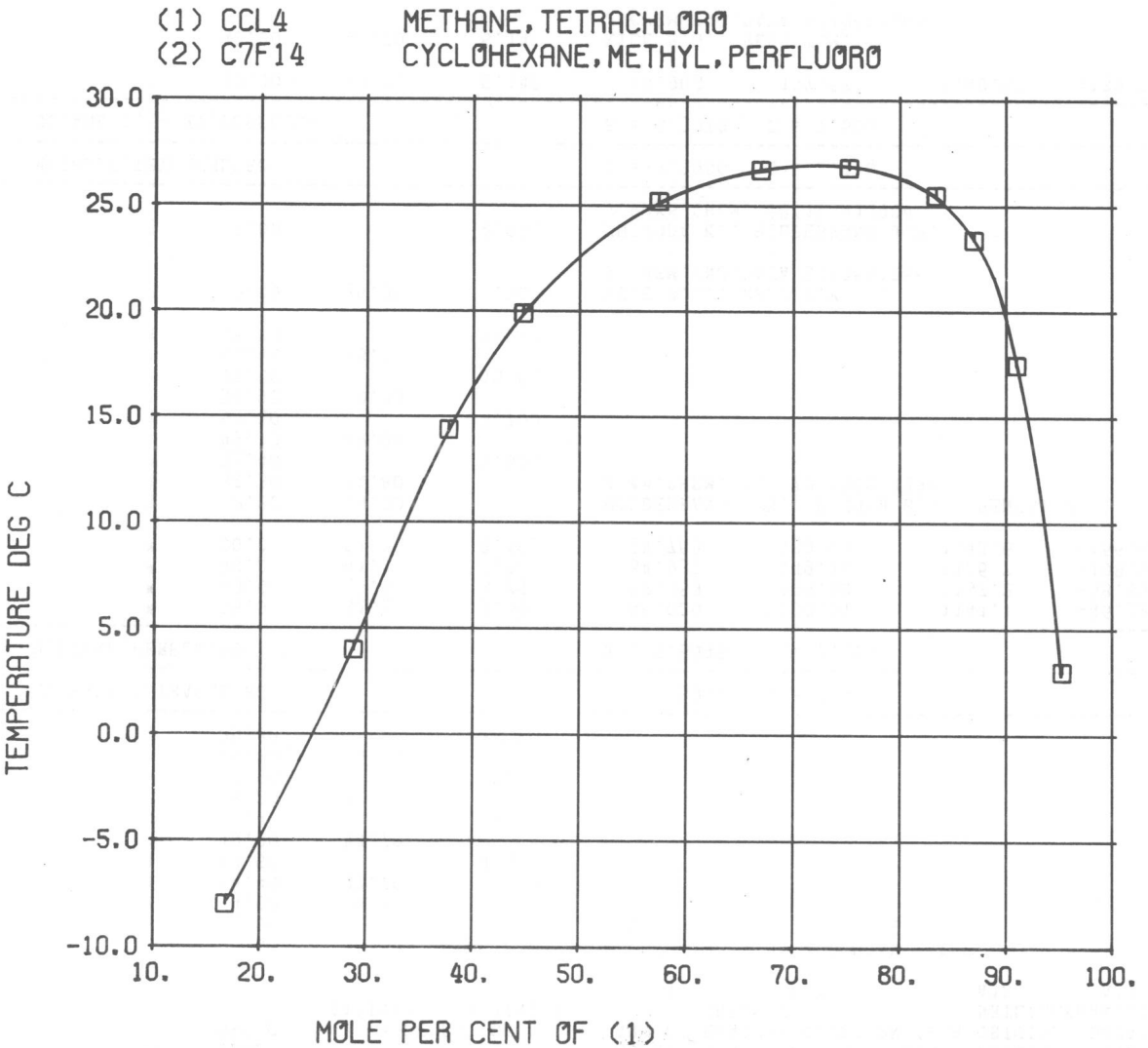
Subscripts

1, 2, 3	components
i, j	components

		REF	TEMP DEG C	SOLUBILITIES MOLE PCT OF (1)IN(2)	(2)IN(1)	MODEL PARAMETERS IN KELVIN (PROCESSED DATA) AND LITERATURE CITATION (FOR ORIGINAL DATA) UNIQUAC A12	UNIQUAC A21	NRTL(ALPHA=.2) A12	NRTL(ALPHA=.2) A21
(1) CCL3NO2	METHANE,NITROTRICHLORO					R = 3.7475	Q = 3.204		
(2) H2O	WATER					R = 0.9200	Q = 1.400		
		*	30.0	0.168E-01	0.858	767.69	393.18	952.03	2127.4
		*	40.0	0.154E-01	1.11	733.47	419.59	912.04	2239.3
		*	50.0	0.142E-01	1.59	672.19	450.00	838.57	2362.1
		1	0.0	0.2489E-01		THOMPSON T.G., BLACK J.H.			
		1	25.00	0.1776E-01		IND.ENG.CHEM. 12(1920)1066			
		1	32.00		0.9069				
		1	36.00		1.070				
		1	41.00		1.121				
		1	48.00		1.481				
		1	50.80		1.663				
		1	55.00		2.025				
		1	75.00	0.1250E-01					
(1) CCL4	METHANE,TETRACHLORO					R = 3.3900	Q = 2.910		
(2) CH2O2	FORMIC ACID					R = 1.5280	Q = 1.532		
		*	25.00	2.037	10.28	405.96	226.66	285.50	981.65
		1	25.00	2.037	10.28	GORDON N.E., REID E.E. J.PHYS.CHEM. 26(1922)773			
(1) CCL4	METHANE,TETRACHLORO					R = 3.3900	Q = 2.910		
(2) C2H3NO	ISOCYANIC ACID,METHYL ESTER					R = 2.0152	Q = 1.601		
		1	50.00		39.36	MOGILYANSKII A.I., LOGINOVA M.M., KOGAN L.M.			
		1	60.00		14.90	ZH.FIZ.KHIM. 43(1969)1915			
		1	70.00		3.429				
(1) CCL4	METHANE,TETRACHLORO					R = 3.3900	Q = 2.910		
(2) C7F14	CYCLOHEXANE,METHYL,PERFLUORO					R = 7.0735	Q = 6.440		
		*	5.0	29.5	5.22	37.210	155.16	892.41	27.379
		*	10.0	33.6	6.38	35.479	151.60	891.31	3.7499
		*	15.0	38.4	7.97	33.050	148.70	887.83	-19.445
		*	20.0	44.9	10.1	33.652	141.16	894.93	-52.069
		*	25.0	56.9	15.6	32.294	132.39	892.72	-93.662

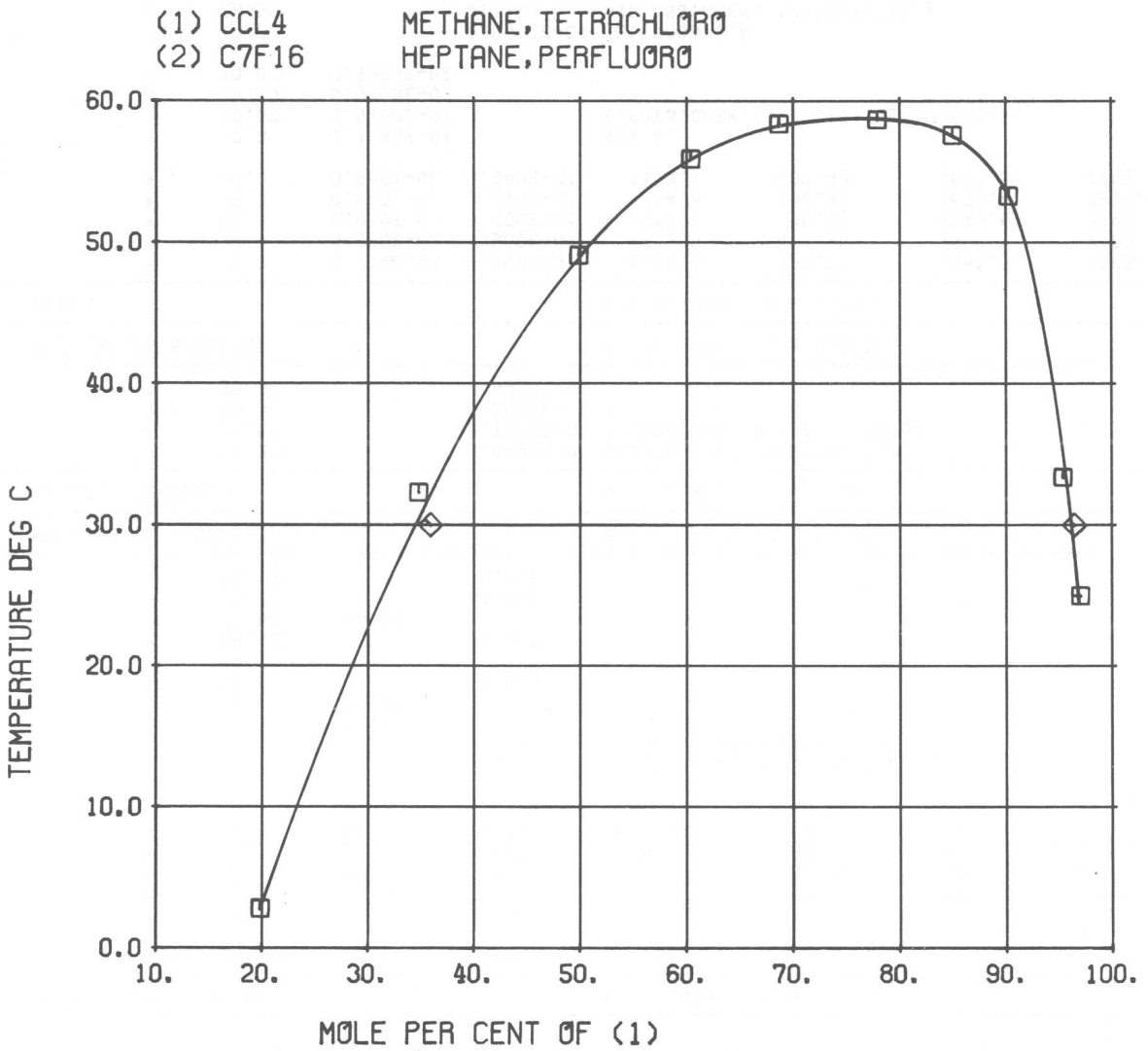
CCl_3NO_2


(1) CCL3NØ2 METHANE,NITRØTRICHLØRØ
(2) H2Ø WATER
REF
1
TEMPERATURE DEG C
80.0
70.0
60.0
50.0
40.0
30.0
20.0
10.0
0.0
.010
.015
.020
.025
MØLE PER CENT ØF (1)
97.5
98.0
98.5
99.0
99.5


(1) CCL4 METHANE, TETRACHLORO
(2) C7F14 CYCLOHEXANE, METHYL, PERFLUORO
REF
1
30.0
25.0
20.0
15.0
10.0
5.0
0.0
-5.0
-10.0
TEMPERATURE DEG C
10.
20.
30.
40.
50.
60.
70.
80.
90.
100.
MOLE PER CENT OF (1)

CCl_4

	REF	TEMP DEG C	SOLUBILITIES MOLE PCT OF (1)IN(2)	(2)IN(1)	MODEL PARAMETERS IN KELVIN (PROCESSED DATA) AND LITERATURE CITATION (FOR ORIGINAL DATA) UNIQUAC A12	UNIQUAC A21	NRTL(ALPHA=.2) A12	NRTL(ALPHA=.2) A21
(CONTINUED)								
	1	-8.00	16.80		HILDEBRAND J.H., COCHRAN D.R.F.			
	1	3.00		4.700	J.AM.CHEM.SOC. 71(1949)22			
	1	4.00	28.80					
	1	14.40	37.70					
	1	17.50		9.000				
	1	19.90	44.70					
	1	23.40		13.00				
	1	25.20	57.40					
	1	25.50		16.60				
	1	26.70	66.90					
	1	26.80		24.80				
(1) CCL4	METHANE,TETRACHLORO				R = 3.3900 Q = 2.910			
(2) C7F16	HEPTANE,PERFLUORO				R = 7.8645 Q = 7.360			
	*	25.0	31.3	3.10	45.029	150.91	1141.1	-46.328
	*	30.0	34.5	3.75	42.089	149.60	1137.2	-68.594
	*	40.0	41.4	5.52	34.937	149.46	1126.7	-108.54
	*	50.0	51.1	8.15	34.768	139.64	1142.6	-166.50
	1	2.80	19.90		HILDEBRAND J.H., FISHER B.B., BENESI H.A.			
	1	32.30	34.80		J.AM.CHEM.SOC. 72(1950)4348			
	1	33.40		4.600				
	1	49.10	49.90					
	1	53.30		9.700				
	1	55.90	60.40					
	1	57.60		15.00				
	1	58.40	68.70					
	1	58.70		22.10				
	2	30.00	35.95	3.507	KYLE B.G., REED T.M. J.CHEM.ENG.DATA 5(1960)266			
	3	25.00		2.990	SHINODA K., HILDEBRAND J.H. J.PHYS.CHEM. 68(1964)3904			
(1) CCL4	METHANE,TETRACHLORO				R = 3.3900 Q = 2.910			
(2) C8F16O	OCTANE,1,8-OXY,PERFLUORO				R = 8.3279 Q = 7.600			
	*	30.00	43.70	5.148	40.903	137.67	1140.6	-139.72
	1	30.00	43.70	5.147	KYLE B.G., REED T.M. J.CHEM.ENG.DATA 5(1960)266			

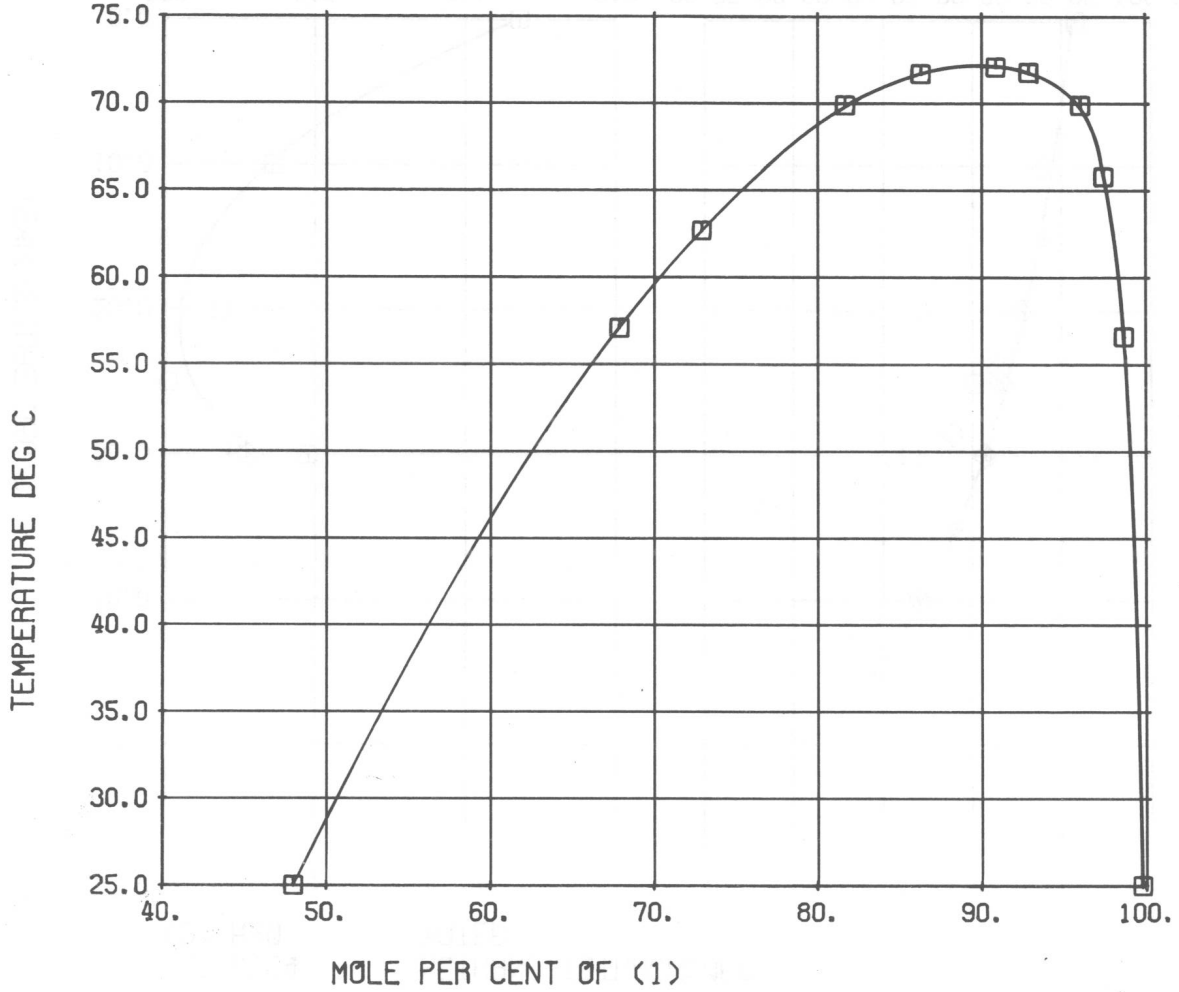

(1) CCL4 METHANE, TETRACHLORO
(2) C7F16 HEPTANE, PERFLUORO
REF
1
2
3
60.0
50.0
40.0
30.0
20.0
10.0
0.0
10.
20.
30.
40.
50.
60.
70.
80.
90.
100.
TEMPERATURE DEG C
MOLE PER CENT OF (1)

CCl_4

		REF	TEMP DEG C	SOLUBILITIES MOLE PCT OF (1)IN(2)	(2)IN(1)	MODEL PARAMETERS IN KELVIN (PROCESSED DATA) AND LITERATURE CITATION (FOR ORIGINAL DATA) UNIQUAC A12	A21	NRTL(ALPHA=.2) A12	A21
(1) CCL4	METHANE,TETRACHLORO					R = 3.3900	Q = 2.910		
(2) C21H8F2808	PENTAERYTRITOL,TETRAPERFLUORO BUTANOATE					R =20.6331	Q =18.560		
		*	25.0	48.0	0.230	3.2485	186.17	2110.4	-366.79
		*	30.0	50.0	0.329	-5.8161	196.00	2072.9	-395.47
		*	40.0	55.1	0.575	-16.083	204.32	2050.8	-465.17
		*	50.0	61.9	0.917	-18.881	199.86	2081.6	-559.79
		*	60.0	70.6	1.64	-21.484	192.19	2117.9	-676.81
		*	70.0	82.2	3.94	-26.564	185.60	2146.3	-811.21
		1	25.00	48.00	0.2300	SHINODA K., HILDEBRAND J.H.			
		1	56.60		1.292	J.PHYS.CHEM. 62(1958)481			
		1	57.10	67.87					
		1	62.70	72.94					
		1	65.80		2.524				
		1	69.90	81.71					
		1	69.95		3.941				
		1	71.68	86.30					
		1	71.82		7.094				
		1	72.09		9.116				
(1) CCL4	METHANE,TETRACHLORO					R = 3.3900	Q = 2.910		
(2) D20	WATER,DIDEUTERO					R = 0.9200	Q = 1.400		
		1	15.00		0.5497E-01	GLASOE P.K., SCHULTZ S.D.			
		1	25.00		0.7232E-01	J.CHEM.ENG.DATA 17(1972)66			
		1	30.00		0.8099E-01				
		1	35.00		0.9930E-01				
(1) CCL4	METHANE,TETRACHLORO					R = 3.3900	Q = 2.910		
(2) H20	WATER					R = 0.9200	Q = 1.400		
		*	0.0	0.114E-01	0.450E-01	1221.0	439.01	1646.3	1988.7
		*	10.0	0.970E-02	0.560E-01	1218.8	471.30	1650.1	2103.8
		*	20.0	0.910E-02	0.700E-01	1214.5	495.03	1645.6	2193.8
		*	25.0	0.920E-02	0.810E-01	1204.8	502.85	1630.4	2226.4
		*	30.0	0.960E-02	0.940E-01	1194.1	507.43	1611.8	2249.6
		1	0.0	0.1136E-01		REX A.			
		1	10.00	0.9720E-02		Z.PHYS.CHEM.(LEIPZIG) 55(1906)355			
		1	20.00	0.9368E-02					
		1	30.00	0.9954E-02					
		2	24.00		0.8532E-01	CLIFFORD C.W.			
		2	28.50		0.1109	IND.ENG.CHEM. 13(1921)628			
		3	25.00	0.9017E-02		GROSS P.M.			
						J.AM.CHEM.SOC. 51(1929)2362			

(1) CCL4 METHANE, TETRACHLORO
(2) C21H8F2808 PENTAERYTRITOL, TETRAPERFLUORO BUTANOATE

REF
1



CCl_4

CCl_4

(1) CCL4
(2) H2Ø
METHANE, TETRACHLØRØ
WATER
TEMPERATURE DEG C
MØLE PER CENT ØF (1)
60.0
50.0
40.0
30.0
20.0
10.0
0.0
.009
.010
.011
.012
99.75
99.80
99.85
99.90
99.95
100.00
REF
1
2
3
4
5
6
7
8
9

	REF	TEMP DEG C	SOLUBILITIES MOLE PCT OF (1)IN(2)	(2)IN(1)	MODEL PARAMETERS IN KELVIN (PROCESSED DATA) AND LITERATURE CITATION (FOR ORIGINAL DATA) UNIQUAC A12	A21	NRTL(ALPHA=.2) A12	A21
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	4	20.00		0.7201E-01	J.AM.CHEM.SOC. 52(1930)3568			
	4	30.00		0.9298E-01				
	4	40.00		0.1296				
	4	50.00		0.2019				
	5	15.00	0.9017E-02		GROSS P.M., SAYLOR J.H.			
	5	30.00	0.9486E-02		J.AM.CHEM.SOC. 53(1931)1744			
	6	30.00	0.9547E-02		VAN ARKEL A.E., VLES S.E.			
					RECL.TRAV.CHIM.PAYS-BAS. 55(1936)407			
	7	0.0		0.4225E-01	STAVERMANN A.J.			
	7	25.00		0.9896E-01	RECL.TRAV.CHIM.PAYS-BAS. 60(1941)836			
	7	30.00		0.1347				
	8	25.00		0.8388E-01	JOHNSON J.R., CHRISTIAN S.D., AFFSPRUNG H.E.			
					J.CHEM.SOC. A(1966)77			
	9	15.00		0.5979E-01	GLASOE P.K., SCHULTZ S.D.			
	9	25.00		0.8292E-01	J.CHEM.ENG.DATA 17(1972)66			
	9	30.00		0.8966E-01				
	9	35.00		0.1099				
	9	45.00		0.1503				
(1) CD3NO2	METHANE,NITROTRIDEUTERO				R = 2.0086 Q = 1.868			
(2) C8H16	1-OCTENE				R = 5.6185 Q = 4.724			
	*	60.0	25.6	7.44	35.078	392.29	891.21	141.55
	*	65.0	29.3	8.97	30.453	387.54	883.71	119.31
	*	70.0	34.7	11.0	28.025	376.04	887.00	84.315
	*	75.0	42.2	14.0	26.879	360.55	896.86	41.041
	*	80.0	57.1	22.0	25.397	338.88	907.08	-16.999
	1	57.25	24.00		SPINOLO G., FERLONI P.			
	1	60.15		7.500	INT.DATA SER.,SEL.DATA MIXTURES A(2)(1974)69			
	1	67.35		9.800				
	1	68.65	33.00					
	1	78.75		18.40				
	1	79.15	52.20					
	1	80.65	62.00					
	1	80.75		27.50				
(1) CHBR3	METHANE,TRIBROMO				R = 3.3342 Q = 2.736			
(2) CH2O2	FORMIC ACID				R = 1.5280 Q = 1.532			
	*	25.00	4.405	11.86	410.24	172.27	312.86	774.35
	1	25.00	4.405	11.86	GORDON N.E., REID E.E.			
					J.PHYS.CHEM. 26(1922)773			

CD_3NO_2