J. M. Sørensen
W. Arlt

LIQUID-LIQUID EQUILIBRIUM DATA COLLECTION

Ternary and Quaternary Systems

Chemistry Data Series
Vol. V, Part 3

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Chemistry Data Series
Vol. V, Part 3

Published by DECHEMA
Deutsche Gesellschaft für Chemisches Apparatewesen

Editors: Dieter Behrens, Reiner Eckermann

ISBN 3-921 567-19-X

Printed by Schön & Wetzel GmbH, Frankfurt/Main, F. R. Germany

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 for 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 of 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 open literature. For that reason the most urgent requirement regarding the publication of data is to offer critically evaluated and reliable data. This will be the goal of the series.

DECHEMA gives an 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.

After that a successful group contribution method for the prediction of vapor-liquid equilibria (UNIFAC) had been presented to the scientific community several years ago, the needs for a similar treatment of liquid-liquid equilibria 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.

During this cooperation, 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 as Volume V of this series, in 3 parts giving not only measured data but also evaluated correlation constants and recommended values. We hope that this gives an instrument that will allow the users to solve their problems considerably more easily aid more quickly than before.

Frankfurt/Main, October 1980

Dieter Behrens
Reiner Eckermann

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Vol. V, Part 2

(1) C4H4O FURAN

(2) C5H4O2 FURFURAL

(3) H2O WATER

KARMILCHIK A.YA., EFIMOVA L.S.
ZH.FIZ.KHIM. 42(1968)2305

TEMPERATURE = 20.0 DEG C TYPE OF SYSTEM = 2

EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE (1)	(2)	(3)	RIGHT PHASE (1)	(2)	(3)
82.319	15.697	1.984	0.217	0.481	99.301
76.026	21.546	2.428	0.191	0.599	99.210
61.143	34.656	4.201	0.138	0.818	99.044
40.646	50.700	8.654	0.111	1.001	98.889
26.444	62.452	11.104	0.056	1.373	98.571

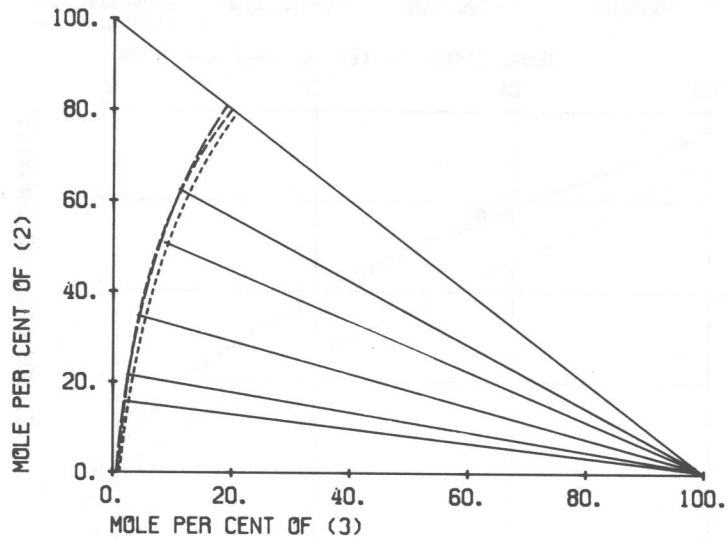
SPECIFIC MODEL PARAMETERS IN KELVIN

I J	UNIQUAC AIJ	AJI	NRTL(ALPHA=.2) AIJ	AJI
1 2	89.315	56.826	147.60	143.59
1 3	658.76	401.26	863.39	1161.8
2 3	225.83	77.206	69.031	1160.8

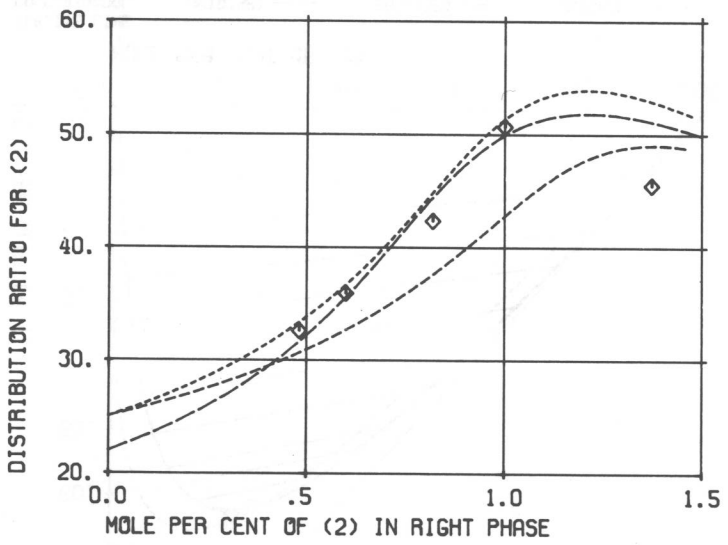
R1 = 2.3361 R2 = 3.1680 R3 = 0.9200
Q1 = 1.816 Q2 = 2.484 Q3 = 1.400

MEAN DEV. BETWEEN CALC. AND EXP. CONC. IN MOLE PCT

UNIQUAC (SPECIFIC PARAMETERS)	0.17
NRTL (SPECIFIC PARAMETERS)	0.22
UNIQUAC (COMMON PARAMETERS)	0.55



EXP.TIE LINE ——
CALC.BINODAL UNIQ(SP) —— NRTL(SP) --- UNIQ(CO) -----



EXP. DISTR.RATIO ◇
CALC.DISTR.RATIO UNIQ(SP) —— NRTL(SP) --- UNIQ(CO) -----

C_4H_4O-$C_5H_4O_2$

(1) C4H8O2S SULFONE,TETRAMETHYLENE

(2) C4H4S THIOPHENE

(3) C6H14 HEXANE

DE FRE R.M.
THESIS GENT 1976

TEMPERATURE = 25.0 DEG C TYPE OF SYSTEM = 1

EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE			RIGHT PHASE		
(1)	(2)	(3)	(1)	(2)	(3)
98.611	0.0	1.389	0.144	0.0	99.856
79.155	18.752	2.092	1.077	12.307	86.615
70.497	26.832	2.671	0.429	18.489	81.082
63.546	33.223	3.231	0.572	23.987	75.441
55.940	39.699	4.360	1.001	30.423	68.576
49.943	44.718	5.339	1.430	36.456	62.114
42.615	50.680	6.705	2.220	42.856	54.924
34.734	55.939	9.327	3.960	49.256	46.784
30.111	58.397	11.491	5.575	52.733	41.692
23.594	60.765	15.641	9.015	57.052	33.932

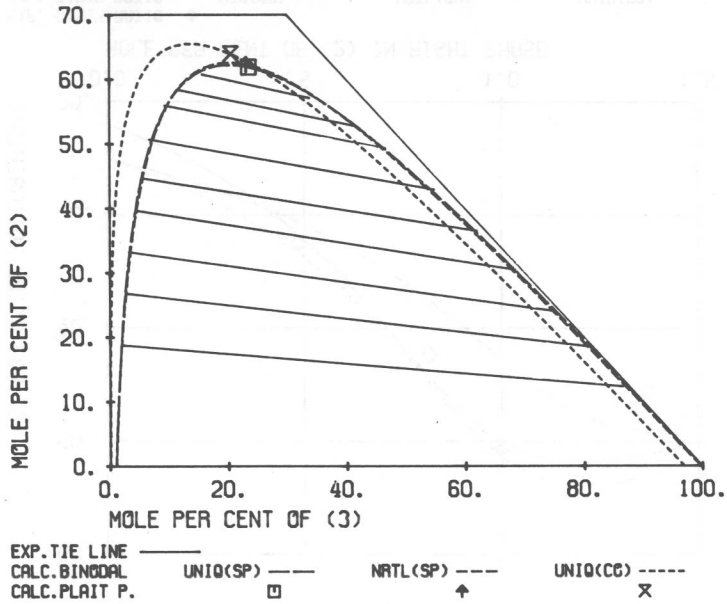
SPECIFIC MODEL PARAMETERS IN KELVIN

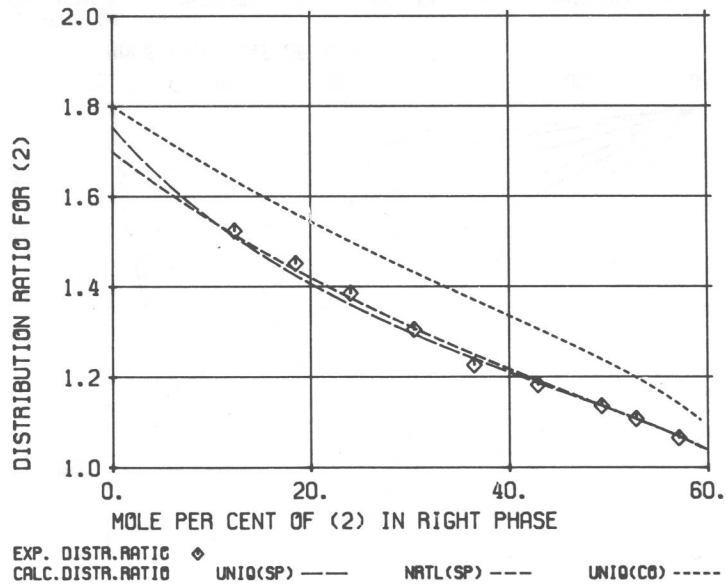
		UNIQUAC		NRTL(ALPHA=.2)	
I	J	AIJ	AJI	AIJ	AJI
1	2	-30.246	-58.263	-196.89	535.19
1	3	98.540	575.52	869.12	1221.0
2	3	-80.273	65.528	236.58	127.82

R1 = 4.0358 R2 = 2.8569 R3 = 4.4998
Q1 = 3.200 Q2 = 2.140 Q3 = 3.856

MEAN DEV. BETWEEN CALC. AND EXP. CONC. IN MOLE PCT

UNIQUAC (SPECIFIC PARAMETERS)	0.28
NRTL (SPECIFIC PARAMETERS)	0.37
UNIQUAC (COMMON PARAMETERS)	2.52





2
C_4H_4S-$C_4H_8O_2S$

(1) C4H8O2S SULFONE,TETRAMETHYLENE

(2) C4H4S THIOPHENE

(3) C7H16 HEPTANE

HANSON C., PATEL A.N., CHANG-KAKOTI D.K.
J.APPL.CHEM. 19(1969)320

TEMPERATURE = 20.5 DEG C TYPE OF SYSTEM = 1

EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE			RIGHT PHASE		
(1)	(2)	(3)	(1)	(2)	(3)
90.386	7.512	2.102	0.829	4.736	94.435
83.417	14.297	2.286	1.400	9.526	89.074
72.865	24.370	2.765	1.463	17.418	81.119
67.893	29.168	2.939	1.533	22.014	76.452
56.456	40.191	3.354	1.589	31.540	66.872
47.264	47.749	4.986	2.355	39.466	58.179
37.024	55.692	7.285	3.411	48.057	48.532
30.609	59.248	10.143	5.010	53.394	41.596
27.981	60.515	11.504	6.154	56.028	37.819

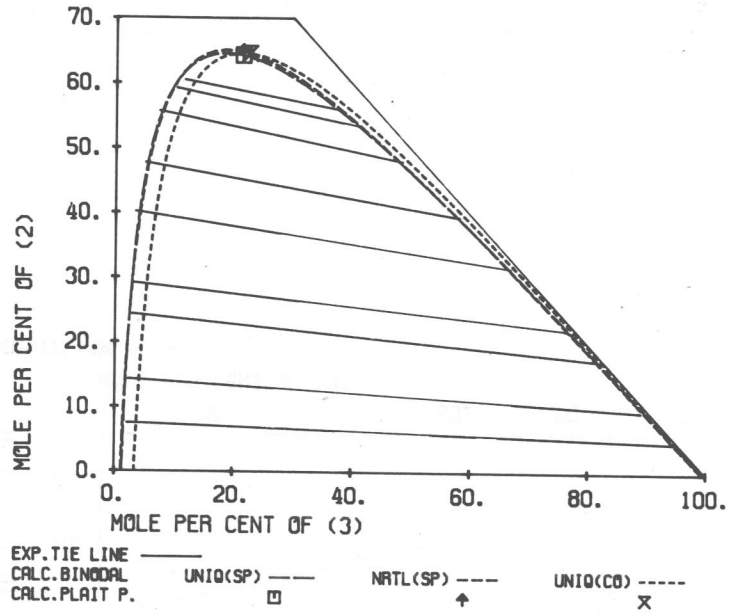
SPECIFIC MODEL PARAMETERS IN KELVIN

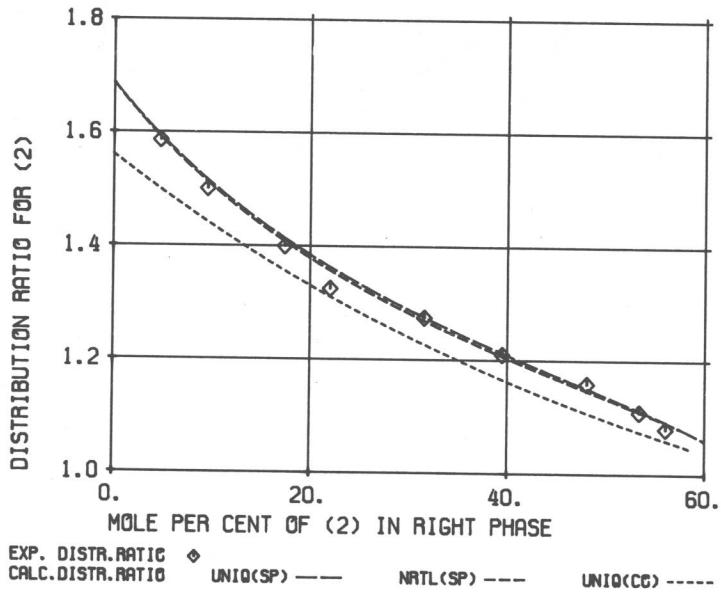
		UNIQUAC		NRTL(ALPHA=.2)	
I	J	AIJ	AJI	AIJ	AJI
1	2	-350.04	37.841	-821.07	783.45
1	3	70.172	434.12	849.98	1012.4
2	3	-67.753	-164.60	280.30	-442.52

R1 = 4.0358 R2 = 2.8569 R3 = 5.1742
Q1 = 3.200 Q2 = 2.140 Q3 = 4.396

MEAN DEV. BETWEEN CALC. AND EXP. CONC. IN MOLE PCT

UNIQUAC (SPECIFIC PARAMETERS)	0.35
NRTL (SPECIFIC PARAMETERS)	0.34
UNIQUAC (COMMON PARAMETERS)	1.33





C_4H_4S-$C_4H_8O_2S$

(1) C7H16 HEPTANE

(2) C7H8 TOLUENE

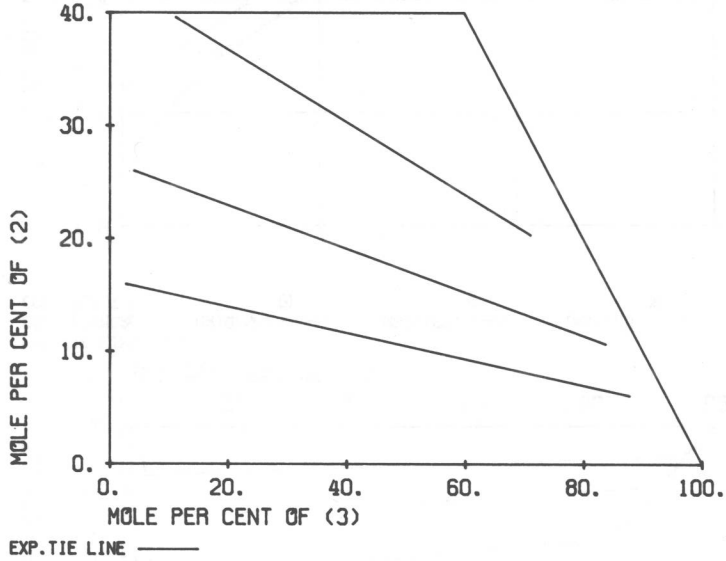
(3) C4H6N2S THIAZOLE,2-AMINO,4-METHYL

RAWAT B.S., GULATI I.B.
J.APPL.CHEM.BIOTECHNOL. 26(1976)425

TEMPERATURE = 50.0 DEG C TYPE OF SYSTEM = 1

EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE			RIGHT PHASE		
(1)	(2)	(3)	(1)	(2)	(3)
81.266	15.950	2.784	6.146	6.076	87.779
69.857	25.994	4.149	5.563	10.665	83.773
49.263	39.590	11.147	8.489	20.324	71.187



(1) C7H8 TOLUENE

(2) C4H6O 3-BUTEN-2-ONE

(3) H2O WATER

FROLOV A.F., LOGINOVA M.A., KIRSANOVA G.N.
ZH.PRIKL.KHIM.(LENINGRAD) 42(1969)2095

TEMPERATURE = 25.0 DEG C TYPE OF SYSTEM = 2

EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE			RIGHT PHASE		
(1)	(2)	(3)	(1)	(2)	(3)
79.349	20.602	0.049	0.002	0.843	99.155
56.848	42.695	0.457	0.004	1.821	98.175
36.564	62.060	1.376	0.004	2.861	97.135
21.398	72.632	5.970	0.002	4.013	95.985
17.446	74.794	7.760	0.002	4.618	95.380

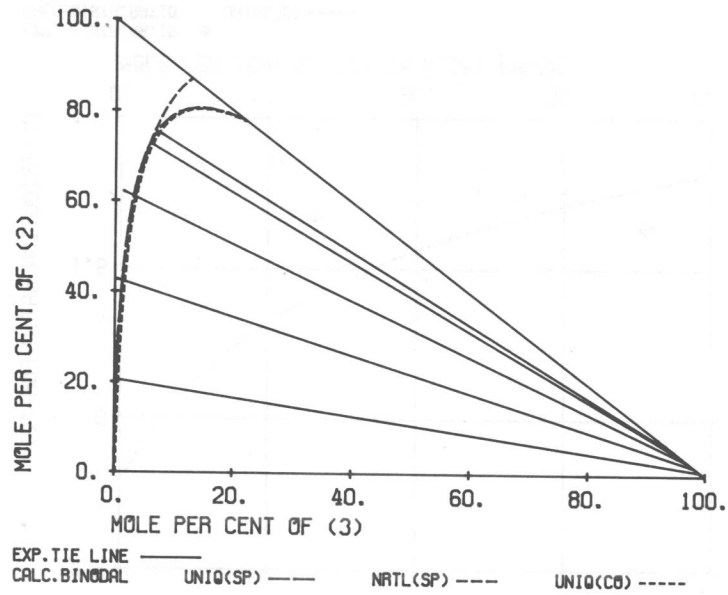
SPECIFIC MODEL PARAMETERS IN KELVIN

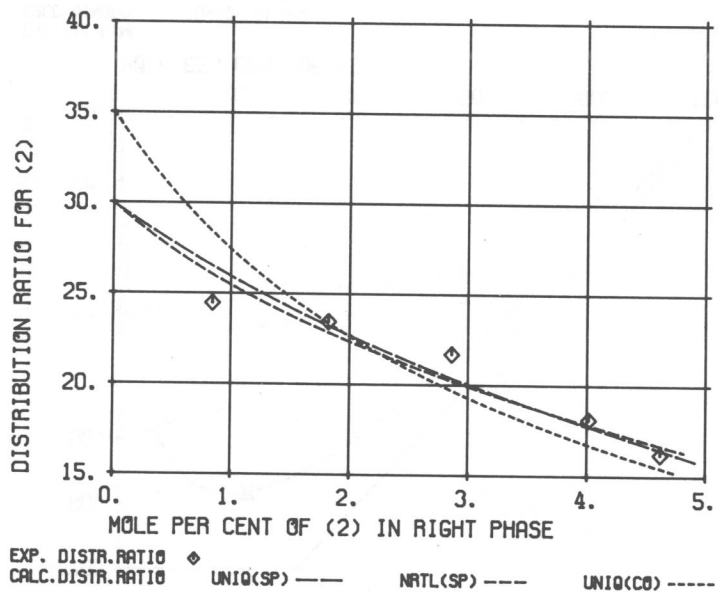
		UNIQUAC		NRTL(ALPHA=.2)	
I	J	AIJ	AJI	AIJ	AJI
1	2	-100.55	87.426	-145.04	73.134
1	3	1277.0	714.85	1861.0	1355.7
2	3	410.03	0.30367	328.52	683.01

R1 = 3.9228 R2 = 3.0178 R3 = 0.9200
Q1 = 2.968 Q2 = 2.664 Q3 = 1.400

MEAN DEV. BETWEEN CALC. AND EXP. CONC. IN MOLE PCT

UNIQUAC (SPECIFIC PARAMETERS)	0.47
NRTL (SPECIFIC PARAMETERS)	0.56
UNIQUAC (COMMON PARAMETERS)	0.66





C_4H_6O-C_7H_8

(1) C7H16 HEPTANE

(2) C6H6 BENZENE

(3) C4H6O2S SULFONE,DIETHENYL

RAWAT B.S., GULATI I.B.
J.APPL.CHEM.BIOTECHNOL. 26(1976)425

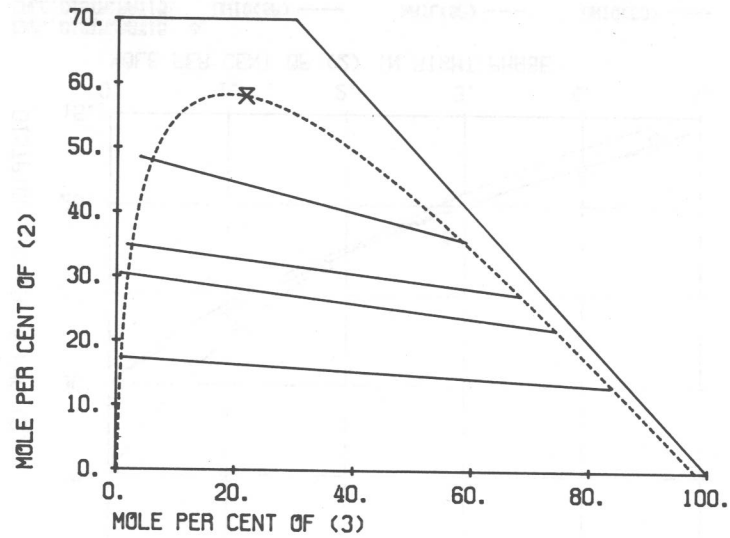
TEMPERATURE = 25.0 DEG C TYPE OF SYSTEM = 1

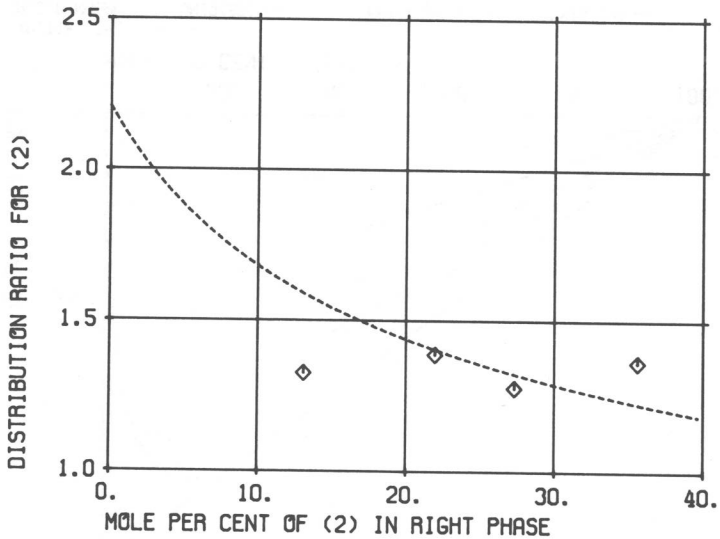
EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE			RIGHT PHASE		
(1)	(2)	(3)	(1)	(2)	(3)
81.920	17.345	0.735	3.330	13.087	83.583
68.969	30.421	0.610	3.754	21.934	74.312
63.453	34.858	1.688	4.356	27.298	68.345
47.698	48.487	3.815	5.343	35.588	59.068

MEAN DEV. BETWEEN CALC. AND EXP. CONC. IN MOLE PCT

UNIQUAC (COMMON PARAMETERS) 1.43





(1) C7H16 HEPTANE

(2) C7H8 TOLUENE

(3) C4H6O2S SULFONE,DIETHENYL

RAWAT B.S., GULATI I.B.
J.APPL.CHEM.BIOTECHNOL. 26(1976)425

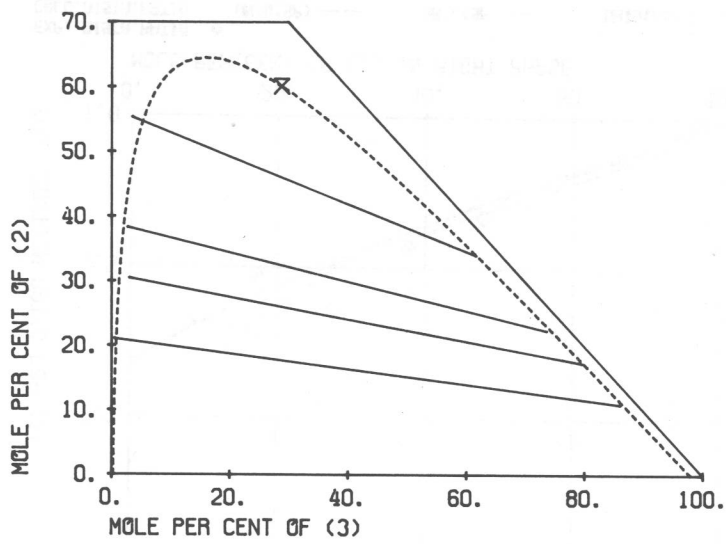
TEMPERATURE = 25.0 DEG C TYPE OF SYSTEM = 1

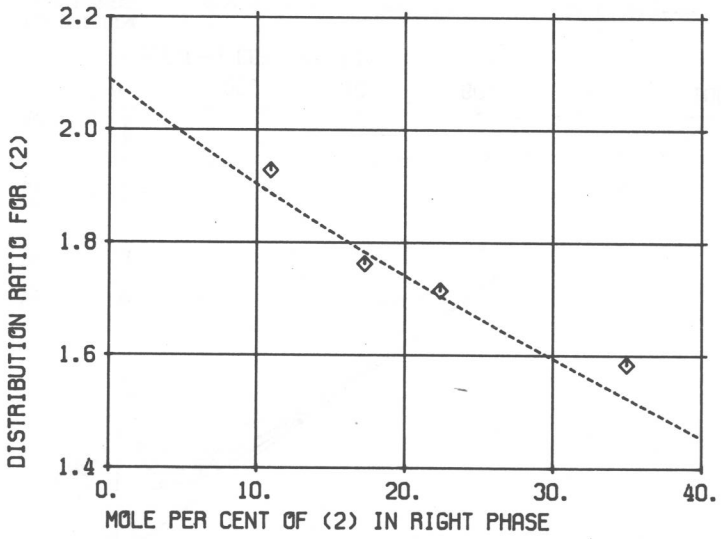
EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE			RIGHT PHASE		
(1)	(2)	(3)	(1)	(2)	(3)
78.513	21.045	0.442	2.922	10.915	86.163
66.579	30.452	2.969	3.408	17.278	79.314
58.976	38.332	2.692	3.900	22.358	73.742
41.255	55.321	3.424	5.929	34.934	59.137

MEAN DEV. BETWEEN CALC. AND EXP. CONC. IN MOLE PCT

UNIQUAC (COMMON PARAMETERS) 0.75





$C_4H_6O_2S$-C_7H_8

(1) C15H32 PENTADECANE

(2) C6H6 BENZENE

(3) C4H6O3 1,3-DIOXOLAN-2-ONE,4-METHYL

GRISHCHENKO N.F., ROGOZKIN V.A., PETRUNENKOVA A.I.
KHIM.TEKHNOL.TOPL.MASEL (1973)5,16

TEMPERATURE = 20.0 DEG C TYPE OF SYSTEM = 1

EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE			RIGHT PHASE		
(1)	(2)	(3)	(1)	(2)	(3)
87.021	12.979	0.0	0.094	7.707	92.198
73.442	26.038	0.520	0.139	17.087	82.775
54.360	44.751	0.889	0.271	27.241	72.488
44.736	53.907	1.357	0.440	37.716	61.844
33.773	63.553	2.675	0.950	47.769	51.282
21.539	74.867	3.594	1.909	67.013	31.078

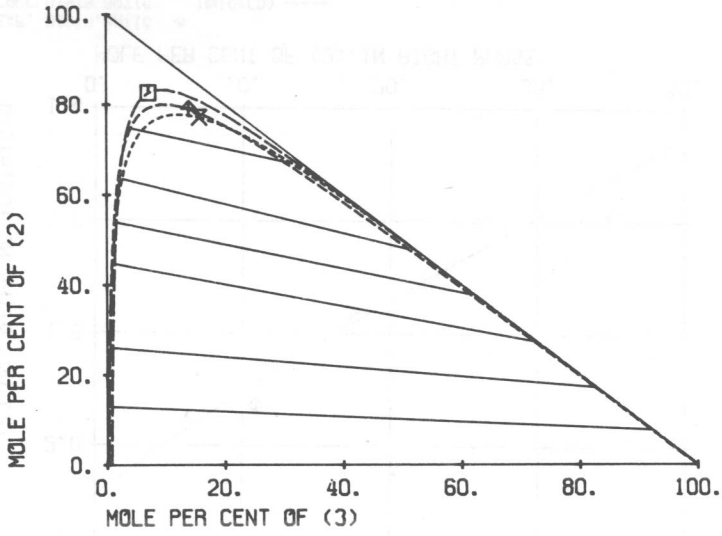
SPECIFIC MODEL PARAMETERS IN KELVIN

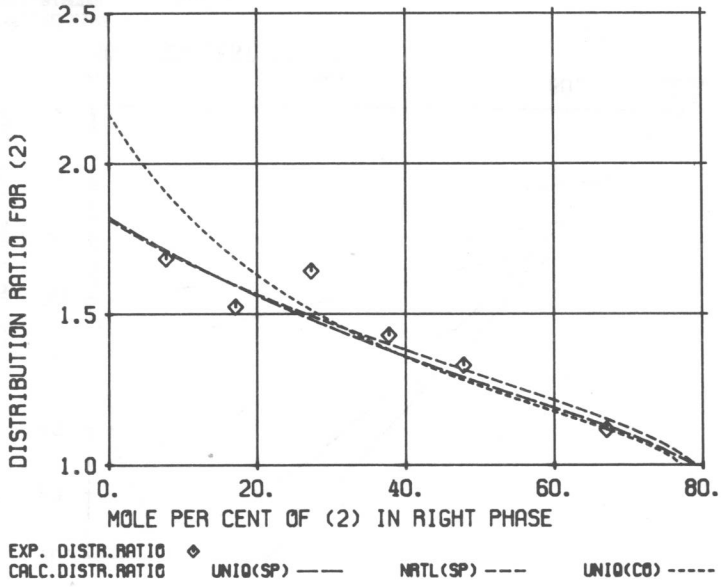
	UNIQUAC		NRTL(ALPHA=.2)	
I J	AIJ	AJI	AIJ	AJI
1 2	165.12	-24.752	-549.56	556.34
1 3	474.27	129.98	1223.4	1410.0
2 3	236.12	-13.422	945.97	-489.64

R1 =10.5694 R2 = 3.1878 R3 = 3.2815
Q1 = 8.716 Q2 = 2.400 Q3 = 2.736

MEAN DEV. BETWEEN CALC. AND EXP. CONC. IN MOLE PCT

UNIQUAC (SPECIFIC PARAMETERS)	0.90
NRTL (SPECIFIC PARAMETERS)	0.97
UNIQUAC (COMMON PARAMETERS)	1.07





(1) C15H32 PENTADECANE

(2) C7H8 TOLUENE

(3) C4H6O3 1,3-DIOXOLAN-2-ONE,4-METHYL

GRISHCHENKO N.F., ROGOZKIN V.A., PETRUNENKOVA A.I.
KHIM.TEKHNOL.TOPL.MASEL (1973)5,16

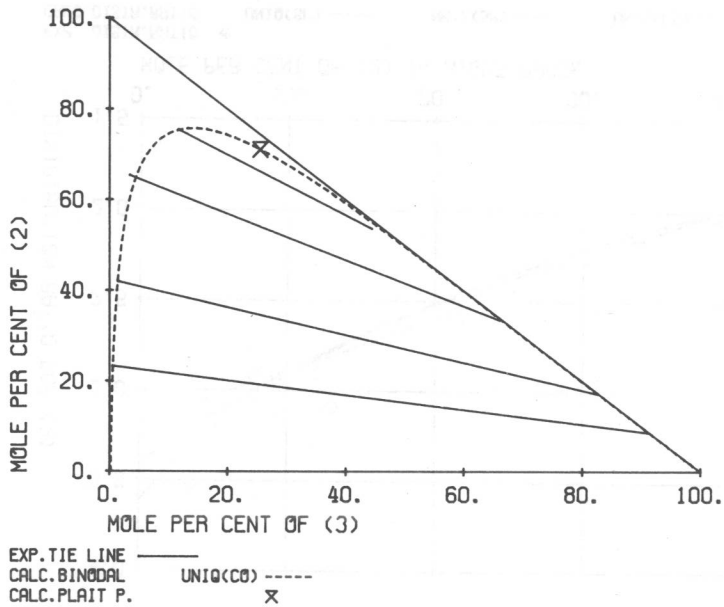
TEMPERATURE = 20.0 DEG C TYPE OF SYSTEM = 1

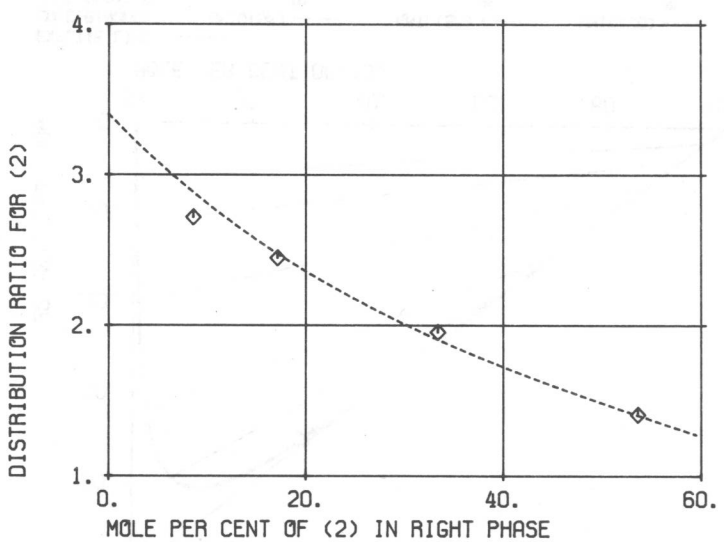
EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE			RIGHT PHASE		
(1)	(2)	(3)	(1)	(2)	(3)
76.128	23.332	0.540	0.095	8.578	91.326
56.635	41.951	1.414	0.142	17.130	82.728
31.222	65.339	3.439	0.467	33.400	66.132
12.793	75.495	11.712	1.861	53.616	44.524

MEAN DEV. BETWEEN CALC. AND EXP. CONC. IN MOLE PCT

UNIQUAC (COMMON PARAMETERS) 0.54





$C_4H_6O_3$-C_7H_8

(1) C15H32 PENTADECANE

(2) C8H10 BENZENE,1,2-DIMETHYL

(3) C4H6O3 1,3-DIOXOLAN-2-ONE,4-METHYL

GRISHCHENKO N.F., ROGOZKIN V.A., PETRUNENKOVA A.I.
KHIM.TEKHNOL.TOPL.MASEL (1973)5,16

TEMPERATURE = 20.0 DEG C TYPE OF SYSTEM = 1

EXPERIMENTAL TIE LINES IN MOLE PCT

LEFT PHASE			RIGHT PHASE		
(1)	(2)	(3)	(1)	(2)	(3)
87.073	12.537	0.389	0.096	3.567	96.337
77.119	22.144	0.737	0.097	7.627	92.276
59.081	40.092	0.827	0.194	12.979	86.828
33.297	64.486	2.217	0.219	29.055	70.725
14.637	78.701	6.662	1.236	38.882	59.882

SPECIFIC MODEL PARAMETERS IN KELVIN

	UNIQUAC		NRTL(ALPHA=.2)	
I J	AIJ	AJI	AIJ	AJI
1 2	87.041	-11.162	-409.26	762.23
1 3	493.32	283.37	1182.3	1419.1
2 3	196.02	16.442	667.28	-2.5237

R1 =10.5694 R2 = 4.6578 R3 = 3.2815
Q1 = 8.716 Q2 = 3.536 Q3 = 2.736

MEAN DEV. BETWEEN CALC. AND EXP. CONC. IN MOLE PCT

UNIQUAC (SPECIFIC PARAMETERS)	0.53
NRTL (SPECIFIC PARAMETERS)	0.54
UNIQUAC (COMMON PARAMETERS)	0.71