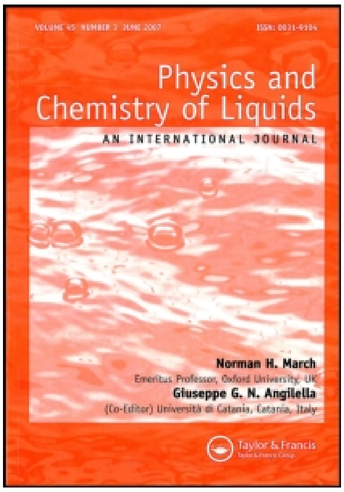

Physics and Chemistry of Liquids
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646857

Excess Volume of Ternary Water Diacetone Alcohol 2-Propanol as a Function of Pressure, Temperature and Composition

C. Boned[a]; M. Moha-ouchane[a]; J. Jose[b]
[a] Laboratoire Haute Pression, Faculté des Sciences, Pau, France [b] Laboratoire de Chimie Analytique 1 (LICAS), Université Claude Bernard, Villeurbanne Cedex, France

To cite this Article Boned, C. , Moha-ouchane, M. and Jose, J.(2000) 'Excess Volume of Ternary Water Diacetone Alcohol 2-Propanol as a Function of Pressure, Temperature and Composition', Physics and Chemistry of Liquids, 38: 1, 113 — 136
To link to this Article: DOI: 10.1080/00319100008045302
URL: http://dx.doi.org/10.1080/00319100008045302

Phys. Chem. Liq., 2000, Vol. 38, pp. 113-136
Reprints available directly from the publisher
Photocopying permitted by license only

Published by license under
the Gordon and Breach Science
Publishers imprint.
Printed in Malaysia.

EXCESS VOLUME OF TERNARY WATER + DIACETONE ALCOHOL + 2-PROPANOL AS A FUNCTION OF PRESSURE, TEMPERATURE AND COMPOSITION

C. BONED[a,]*, M. MOHA-OUCHANE[a] and J. JOSE[b]

[a] *Laboratoire Haute Pression, Faculté des Sciences, Avenue de l'Université, 64000, Pau, France;*
[b] *Laboratoire de Chimie Analytique 1 (LICAS), Université Claude Bernard, Lyon 1, 69622 Villeurbanne Cedex, France*

(Received 8 October 1998)

The excess volume V^E of the ternary water + diacetone alcohol (or DAA) + 2-propanol and of the three binaries water + DAA, water + 2-propanol and DAA + 2-propanol was evaluated from experimental density data (2772 values) as a function of the pressure P (between 0.1 MPa and 65 MPa), the temperature T (303.15 K, 323.15 K and 343.15 K) and the composition. Various representative models are discussed. It is possible to account for the values of the density with an average absolute deviation of about 0.06% in the experimental P-T domain.

Keywords: Density; excess volume; water; alcohol; ternary mixtures

INTRODUCTION

Whereas there is a very significant volume of data describing variations of excess volume V^E of binary systems at atmospheric pressure, according to the temperature, composition and chemical nature of the components, studies on the influence of the pressure are less common. Some data relating to binary systems can be found in the literature

---

*Corresponding author.

but with regard to the ternary systems there are practically no experimental values. We chose the ternary system water + 2-propanol + diacetone alcohol (or DAA, or 4-hydroxy-4-methyl-2-pen- tanone) as the interactions between water and each alcohol are particularly important.

EXPERIMENTAL TECHNIQUES

Values of ρ between 0.1 MPa and 65 MPa are measured using an Anton Paar DMA60 + DMA 601 resonance densitometer with an additional 512P cell. Details of the calibration have been described by Lagourette *et al.* [1] and water is used as calibration substance (we used the values given by Kell and Whalley [2]). Error on temperature T is ± 0.05 K. The error on pressure P is estimated to be ± 0.05 MPa (except at $P = 0.1$ MPa). The error on ρ is less than 0.1 kg/m^3 (except at $P = 0.1$ MPa where it is estimated to be below 0.03 kg/m^3) which corresponds to the estimate made by Papaioannou *et al.* [3] with identical apparatus (with the 512 cell limited to 40 MPa, instead of the more recent 512P cell limited to 70 MPa). The water (H_2O, molecular weight $M = 18.015$ g/mole) is distilled water. The DAA used is commercially available ($C_6H_{12}O_2$: Interchim, purity > 99%, molecular weight $M = 116.16$ g/mole) and the 2-propanol used is also commercially available (C_3H_8O: Sigma-Aldrich, purity > 99.5%, molecular weight $M = 60.1$ g/mole). The mixtures were prepared by weighing at atmospheric pressure and ambient temperature so as to obtain the molar fractions $x_i = 0.1$, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8 and 0.9 (with $\sum_{i=1}^{3} x_i = 1$).

EXPERIMENTAL RESULTS

Measurements of ρ were taken at 303.15 K, 323.15 K and 343.15 K, and within the pressure range from atmospheric pressure 0.1 MPa to 65 MPa by step of 5 MPa. The values are indicated in Table I [4]. There are 2772 experimental determinations. 42 values of ρ for each pure substance (126 values at all for the 3 pure substances including the 42 values for water which is the substance used for the calibration), 378 experimental values of ρ have been obtained for each of the 3 binary

TABLE I Variations of density ρ *versus* temperature, pressure and composition ($x_p = 1 - x_w - x_d$) x_w: water molar fraction, x_d: DAA molar fraction, x_p: 2-propanol molar fraction

			303.15K	323.15K	343.15K			303.15K	323.15K	343.15K
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
0.1	0	1	929.11	910.76	892.38	0	0.5	872.74	853.86	834.69
5	0	1	932.75	914.72	896.80	0	0.5	876.53	858.19	839.52
10	0	1	936.13	918.49	901.07	0	0.5	880.23	862.36	844.27
15	0	1	939.45	922.18	905.20	0	0.5	883.75	866.23	848.71
20	0	1	942.62	925.69	909.10	0	0.5	887.14	870.05	852.92
25	0	1	945.73	929.17	912.86	0	0.5	890.45	873.69	856.95
30	0	1	948.70	932.47	916.44	0	0.5	893.57	877.19	860.73
35	0	1	951.65	935.59	919.78	0	0.5	896.68	880.46	864.38
40	1	1	954.53	938.68	923.10	0	0.5	899.60	883.76	867.91
45	0	1	957.22	941.65	926.29	0	0.5	902.50	886.92	871.35
50	0	1	959.89	944.49	929.45	0	0.5	905.28	889.88	874.57
55	0	1	962.55	947.31	932.44	0	0.5	907.98	892.93	877.81
60	0	1	965.02	950.05	935.35	0	0.5	910.60	895.75	880.88
65	0	1	967.36	952.67	938.24	0	0.5	913.05	898.41	883.87
0.1	0	0.9	919.88	901.52	882.96	0	0.4	857.70	838.89	819.52
5	0	0.9	923.58	905.48	887.32	0	0.4	861.54	843.29	824.46
10	0	0.9	927.07	909.36	891.70	0	0.4	865.29	847.69	829.31
15	0	0.9	930.43	912.67	895.89	0	0.4	868.92	851.53	833.86
20	0	0.9	933.56	916.55	899.78	0	0.4	872.30	855.35	838.23
25	0	0.9	936.71	920.03	903.60	0	0.4	875.67	859.09	842.25
30	0	0.9	939.74	923.39	907.12	0	0.4	878.84	862.70	846.19
35	0	0.9	942.69	926.50	910.62	0	0.4	882.00	865.97	849.89
40	0	0.9	945.56	929.54	913.99	0	0.4	884.98	869.32	853.47
45	0	0.9	948.31	932.56	917.28	0	0.4	887.93	872.55	856.97
50	0	0.9	950.98	935.50	920.34	0	0.4	890.65	875.65	860.28
55	0	0.9	953.63	938.37	923.38	0	0.4	893.45	878.51	863.47
60	0	0.9	956.16	941.06	926.34	0	0.4	896.08	881.42	866.54
65	0	0.9	958.55	943.68	929.28	0	0.4	898.63	884.23	869.63
0.1	0	0.8	909.68	891.25	872.52	0	0.3	840.74	822.11	802.53
5	0	0.8	913.37	895.32	877.09	0	0.3	844.75	826.65	807.79
10	0	0.8	916.87	899.19	881.41	0	0.3	848.61	831.00	812.79
15	0	0.8	920.28	902.94	885.65	0	0.3	852.28	835.11	817.45
20	0	0.8	923.51	906.49	889.65	0	0.3	855.83	839.03	821.87
25	0	0.8	926.72	910.14	893.47	0	0.3	859.24	842.88	826.10
30	0	0.8	929.69	913.43	897.10	0	0.3	862.47	846.54	829.98
35	0	0.8	932.69	916.54	900.65	0	0.3	865.63	849.91	833.85
40	0	0.8	935.57	919.69	904.02	0	0.3	868.71	853.26	837.42
45	0	0.8	938.37	922.76	907.26	0	0.3	871.66	856.43	840.97
50	0	0.8	941.03	925.60	910.47	0	0.3	874.43	859.58	844.39
55	0	0.8	943.74	928.41	913.51	0	0.3	877.23	862.61	847.63
60	0	0.8	946.21	931.26	916.42	0	0.3	879.91	865.50	850.75
65	0	0.8	948.71	933.93	919.41	0	0.3	882.45	868.27	853.84
0.1	0	0.7	898.56	879.90	860.95	0	0.2	821.89	803.39	783.72
5	0	0.7	902.30	884.02	865.68	0	0.2	826.05	808.03	789.13
10	0	0.7	905.79	887.95	870.21	0	0.2	829.96	812.54	794.24
15	0	0.7	909.21	891.74	874.45	0	0.2	833.85	816.75	799.11
20	0	0.7	912.54	895.46	878.50	0	0.2	837.39	820.72	803.63
25	0	0.7	915.75	899.05	882.42	0	0.2	840.85	824.73	807.91

TABLE I (Continued)

			303.15K	323.15K	343.15K			303.15K	323.15K	343.15K
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
30	0	0.7	918.83	902.40	886.10	0	0.2	844.14	828.39	811.96
35	0	0.7	921.83	905.61	889.60	0	0.2	847.40	831.81	815.82
40	0	0.7	924.70	908.81	893.07	0	0.2	850.53	835.27	819.55
45	0	0.7	927.55	911.94	896.42	0	0.2	853.48	838.60	823.20
50	0	0.7	930.27	914.88	899.58	0	0.2	856.35	841.74	826.57
55	0	0.7	932.87	917.75	902.67	0	0.2	859.21	844.76	829.86
60	0	0.7	935.50	920.54	905.68	0	0.2	861.88	847.65	833.08
65	0	0.7	937.94	923.21	908.79	0	0.2	864.53	850.47	836.22
0.1	0	0.6	886.02	867.53	848.35	0	0.1	800.66	782.19	762.56
5	0	0.6	889.98	871.76	853.19	0	0.1	805.03	787.05	768.13
10	0	0.6	893.63	875.79	857.83	0	0.1	809.15	791.77	773.45
15	0	0.6	896.99	879.69	862.12	0	0.1	813.04	796.14	778.42
20	0	0.6	900.38	883.40	866.33	0	0.1	816.69	800.21	783.15
25	0	0.6	903.59	887.04	870.30	0	0.1	820.26	804.27	787.54
30	0	0.6	906.71	890.44	874.04	0	0.1	823.70	807.98	791.74
35	0	0.6	909.77	893.71	877.64	0	0.1	827.01	811.56	795.65
40	0	0.6	912.64	896.85	881.05	0	0.1	830.19	815.01	799.37
45	0	0.6	915.54	900.03	884.45	0	0.1	833.19	818.34	803.08
50	0	0.6	918.26	903.03	887.72	0	0.1	836.11	821.53	806.55
55	0	0.6	920.97	905.95	890.75	0	0.1	838.97	824.60	809.89
60	0	0.6	923.59	908.74	893.82	0	0.1	841.69	827.60	813.15
65	0	0.6	926.04	911.46	896.92	0	0.1	844.29	830.47	816.30
0.1	0	0	776.41	758.32	738.63	0.1	0.5	885.92	867.31	848.03
5	0	0	780.89	763.39	744.40	0.1	0.5	889.66	871.49	852.70
10	0	0	785.12	768.10	750.04	0.1	0.5	893.25	875.41	857.18
15	0	0	789.16	772.68	755.22	0.1	0.5	896.61	879.25	861.52
20	0	0	792.96	776.91	760.05	0.1	0.5	899.89	882.86	865.57
25	0	0	796.58	781.02	764.59	0.1	0.5	903.04	886.44	869.44
30	0	0	800.07	784.94	768.90	0.1	0.5	906.12	889.79	873.17
35	0	0	803.49	788.46	772.91	0.1	0.5	909.11	893.01	876.66
40	0	0	806.67	792.02	776.74	0.1	0.5	911.98	896.15	880.08
45	0	0	809.77	795.45	780.49	0.1	0.5	914.83	899.22	883.48
50	0	0	812.75	798.69	784.06	0.1	0.5	917.44	902.05	886.64
55	0	0	815.65	801.81	787.51	0.1	0.5	920.09	904.97	889.72
60	0	0	818.37	804.86	790.72	0.1	0.5	922.55	907.71	892.68
65	0	0	821.07	807.73	793.96	0.1	0.5	925.05	910.37	895.67
0.1	0.1	0.9	933.75	915.38	896.71	0.1	0.4	870.45	851.75	832.31
5	0.1	0.9	937.23	919.13	901.02	0.1	0.4	874.29	856.03	837.20
10	0.1	0.9	940.61	922.84	905.24	0.1	0.4	877.89	860.11	841.78
15	0.1	0.9	943.76	926.43	909.21	0.1	0.4	881.35	864.00	846.12
20	0.1	0.9	946.89	929.88	913.00	0.1	0.4	884.68	867.61	850.33
25	0.1	0.9	949.88	933.20	916.66	0.1	0.4	887.94	871.30	854.24
30	0.1	0.9	952.85	936.39	920.13	0.1	0.4	890.96	874.75	858.03
35	0.1	0.9	955.75	939.45	923.52	0.1	0.4	894.06	878.02	861.62
40	0.1	0.9	958.46	942.44	926.84	0.1	0.4	896.88	881.16	865.09
45	0.1	0.9	961.16	945.46	929.97	0.1	0.4	899.78	884.33	868.49
50	0.1	0.9	963.78	948.30	933.03	0.1	0.4	902.50	887.32	871.70
55	0.1	0.9	966.37	951.01	936.02	0.1	0.4	905.20	890.24	874.89
60	0.1	0.9	968.79	953.76	938.88	0.1	0.4	907.71	893.01	877.81
65	0.1	0.9	971.13	956.38	941.71	0.1	0.4	910.20	895.73	880.72

TABLE I (Continued)

			303.15K	323.15K	343.15K			303.15K	323.15K	343.15K
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
0.1	0.1	0.8	923.54	904.89	886.43	0.1	0.3	852.98	834.24	814.61
5	0.1	0.8	927.07	908.80	890.79	0.1	0.3	856.88	838.68	819.60
10	0.1	0.8	930.40	912.62	894.95	0.1	0.3	860.63	842.87	824.39
15	0.1	0.8	933.71	916.20	899.03	0.1	0.3	864.20	846.76	828.89
20	0.1	0.8	936.72	919.71	902.87	0.1	0.3	867.53	850.58	833.26
25	0.1	0.8	939.88	923.08	906.52	0.1	0.3	870.84	854.26	837.22
30	0.1	0.8	942.80	926.32	910.10	0.1	0.3	874.01	857.82	841.06
35	0.1	0.8	945.64	929.33	913.49	0.1	0.3	877.12	861.08	844.71
40	0.1	0.8	948.46	932.37	916.75	0.1	0.3	880.09	864.33	848.28
45	0.1	0.8	951.15	935.39	919.99	0.1	0.3	882.93	867.50	851.78
50	0.1	0.8	953.77	938.23	923.05	0.1	0.3	885.70	870.49	855.04
55	0.1	0.8	956.37	940.99	926.04	0.1	0.3	888.35	873.46	858.17
60	0.1	0.8	958.84	943.68	928.89	0.1	0.3	892.11	876.19	861.29
65	0.1	0.8	961.12	946.24	931.78	0.1	0.3	893.46	878.96	864.44
0.1	0.1	0.7	912.19	893.59	874.85	0.1	0.2	833.51	814.64	794.88
5	0.1	0.7	915.83	897.66	879.31	0.1	0.2	837.51	819.07	800.13
10	0.1	0.7	919.27	901.42	883.74	0.1	0.2	841.32	823.42	805.08
15	0.1	0.7	922.52	905.05	887.82	0.1	0.2	844.94	827.58	809.74
20	0.1	0.7	925.70	908.56	891.77	0.1	0.2	848.43	831.39	814.10
25	0.1	0.7	928.79	912.09	895.53	0.1	0.2	851.79	835.18	818.22
30	0.1	0.7	931.76	915.34	898.99	0.1	0.2	854.96	838.84	822.16
35	0.1	0.7	934.71	918.50	902.44	0.1	0.2	858.11	842.11	825.96
40	0.1	0.7	937.53	921.59	905.80	0.1	0.2	861.14	845.46	829.59
45	0.1	0.7	940.28	924.61	909.04	0.1	0.2	864.04	848.68	833.14
50	0.1	0.7	942.89	927.45	912.04	0.1	0.2	866.75	851.77	836.34
55	0.1	0.7	945.49	930.21	915.08	0.1	0.2	869.66	854.79	839.74
60	0.1	0.7	948.01	932.95	918.10	0.1	0.2	872.20	857.64	842.61
65	0.1	0.7	950.45	935.59	920.74	0.1	0.2	874.77	860.43	845.57
0.1	0.1	0.6	899.81	881.04	861.97	0.1	0.1	811.08	792.46	772.52
5	0.1	0.6	903.50	885.22	866.54	0.1	0.1	815.29	797.22	777.98
10	0.1	0.6	906.88	889.14	871.02	0.1	0.1	819.31	801.66	783.20
15	0.1	0.6	910.30	892.83	875.15	0.1	0.1	823.03	805.87	787.96
20	0.1	0.6	913.47	896.38	879.15	0.1	0.1	826.57	809.79	792.53
25	0.1	0.6	916.62	899.92	882.96	0.1	0.1	830.09	813.74	796.75
30	0.1	0.6	919.65	903.27	886.59	0.1	0.1	833.37	817.40	800.79
35	0.1	0.6	922.59	906.43	890.09	0.1	0.1	836.52	820.87	804.65
40	0.1	0.6	925.41	909.52	893.45	0.1	0.1	839.60	824.27	808.38
45	0.1	0.6	928.15	912.59	896.80	0.1	0.1	842.60	827.49	811.98
50	0.1	0.6	930.87	915.48	899.91	0.1	0.1	845.47	830.63	815.34
55	0.1	0.6	933.47	918.24	902.99	0.1	0.1	848.27	833.60	818.57
60	0.1	0.6	935.99	921.03	905.95	0.1	0.1	850.88	836.54	821.74
65	0.1	0.6	938.38	923.70	908.95	0.1	0.1	853.48	839.36	826.12
0.1	0.1	0	785.58	766.91	746.75	0.2	0.4	884.61	865.90	846.41
5	0.1	0	789.89	771.87	752.53	0.2	0.4	888.24	870.02	851.03
10	0.1	0	794.07	776.53	757.89	0.2	0.4	891.73	873.83	855.40
15	0.1	0	797.95	780.95	762.97	0.2	0.4	895.08	877.57	859.85
20	0.1	0	801.65	785.07	767.64	0.2	0.4	898.25	881.12	863.63
25	0.1	0	805.27	789.02	772.13	0.2	0.4	901.30	884.60	867.38
30	0.1	0	808.49	792.78	776.27	0.2	0.4	904.32	887.89	870.95
35	0.1	0	811.85	796.41	780.18	0.2	0.4	907.26	891.00	874.50

TABLE I (Continued)

			303.15K	323.15K	343.15K			303.15K	323.15K	343.15K
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
40	0.1	0	814.98	799.75	783.96	0.2	0.4	910.07	894.08	877.86
45	0.1	0	818.03	803.13	787.60	0.2	0.4	912.76	897.15	881.15
50	0.1	0	820.95	806.27	791.07	0.2	0.4	915.43	900.04	884.25
55	0.1	0	823.80	809.39	794.41	0.2	0.4	918.02	902.85	887.29
60	0.1	0	826.52	812.33	797.62	0.2	0.4	920.54	905.53	890.24
65	0.1	0	829.12	815.20	800.87	0.2	0.4	923.03	908.25	893.29
0.1	0.2	0.8	938.06	919.78	901.15	0.2	0.3	866.69	848.11	828.43
5	0.2	0.8	941.49	923.59	905.41	0.2	0.3	870.37	852.28	833.26
10	0.2	0.8	944.71	927.14	909.46	0.2	0.3	873.91	856.31	837.73
15	0.2	0.8	947.86	930.62	913.39	0.2	0.3	877.32	860.10	842.01
20	0.2	0.8	950.87	933.91	917.07	0.2	0.3	880.60	863.65	845.95
25	0.2	0.8	953.82	937.23	920.61	0.2	0.3	883.69	867.28	849.97
30	0.2	0.8	956.63	940.37	924.03	0.2	0.3	886.71	870.62	853.65
35	0.2	0.8	959.41	943.26	927.26	0.2	0.3	889.70	873.73	857.19
40	0.2	0.8	962.13	946.25	930.42	0.2	0.3	892.51	876.87	860.60
45	0.2	0.8	964.99	949.11	933.55	0.2	0.3	895.31	879.93	863.94
50	0.2	0.8	967.22	951.84	936.56	0.2	0.3	898.02	882.87	867.10
55	0.2	0.8	969.82	954.55	939.49	0.2	0.3	900.72	885.73	870.18
60	0.2	0.8	972.18	957.14	942.35	0.2	0.3	903.13	888.47	873.24
65	0.2	0.8	974.53	959.81	945.24	0.2	0.3	905.63	891.13	876.18
0.1	0.2	0.7	926.87	908.37	889.67	0.2	0.2	846.29	827.53	807.55
5	0.2	0.7	930.34	912.11	893.93	0.2	0.2	850.18	831.96	812.59
10	0.2	0.7	933.68	915.77	898.09	0.2	0.2	853.83	835.98	817.22
15	0.2	0.7	936.77	919.30	902.01	0.2	0.2	857.29	839.93	821.66
20	0.2	0.7	939.78	922.64	905.69	0.2	0.2	860.56	843.75	825.92
25	0.2	0.7	942.78	925.96	909.34	0.2	0.2	863.81	847.32	829.99
30	0.2	0.7	945.64	929.15	912.75	0.2	0.2	866.88	850.88	833.66
35	0.2	0.7	948.54	932.10	916.04	0.2	0.2	869.93	853.98	837.36
40	0.2	0.7	951.19	935.09	919.30	0.2	0.2	872.85	857.22	840.88
45	0.2	0.7	953.83	937.95	922.43	0.2	0.2	875.69	860.29	844.32
50	0.2	0.7	956.39	940.73	925.44	0.2	0.2	878.40	863.33	847.53
55	0.2	0.7	958.94	943.44	928.37	0.2	0.2	881.15	866.24	850.82
60	0.2	0.7	961.35	946.13	931.28	0.2	0.2	883.72	868.98	853.65
65	0.2	0.7	963.73	948.61	933.95	0.2	0.2	886.19	871.67	856.56
0.1	0.2	0.6	914.16	895.87	876.90	0.2	0.1	822.81	804.31	784.05
5	0.2	0.6	917.79	899.78	881.20	0.2	0.1	826.75	808.74	789.13
10	0.2	0.6	921.07	903.32	885.36	0.2	0.1	830.56	812.97	794.03
15	0.2	0.6	924.27	906.96	889.44	0.2	0.1	834.12	817.02	798.57
20	0.2	0.6	927.28	910.35	893.17	0.2	0.1	837.50	820.83	802.93
25	0.2	0.6	930.38	913.72	896.88	0.2	0.1	840.85	824.51	807.05
30	0.2	0.6	933.24	917.02	900.35	0.2	0.1	843.97	828.06	810.93
35	0.2	0.6	936.08	919.92	903.63	0.2	0.1	847.07	831.43	814.58
40	0.2	0.6	938.73	923.01	906.89	0.2	0.1	850.04	834.67	818.14
45	0.2	0.6	941.48	925.86	910.13	0.2	0.1	852.93	837.78	821.69
50	0.2	0.6	944.04	928.70	913.07	0.2	0.1	855.70	840.88	824.95
55	0.2	0.6	946.53	931.46	916.11	0.2	0.1	858.34	843.73	828.08
60	0.2	0.6	948.99	934.09	919.02	0.2	0.1	860.95	846.57	831.13
65	0.2	0.6	951.35	936.68	921.74	0.2	0.1	863.44	849.39	834.28
0.1	0.2	0.5	900.36	881.80	862.79	0.2	0	794.91	775.98	755.73
5	0.2	0.5	903.99	885.76	867.19	0.2	0	799.17	780.78	761.30

TABLE I (Continued)

			303.15K	323.15K	343.15K			303.15K	323.15K	343.15K
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
10	0.2	0.5	907.37	889.47	871.56	0.2	0	803.13	785.28	766.45
15	0.2	0.5	910.57	893.10	875.64	0.2	0	806.90	789.54	771.37
20	0.2	0.5	913.74	896.60	879.59	0.2	0	810.39	793.51	775.93
25	0.2	0.5	916.79	900.03	883.29	0.2	0	813.90	797.40	780.15
30	0.2	0.5	919.75	903.27	886.81	0.2	0	817.18	801.00	784.19
35	0.2	0.5	922.65	906.32	890.20	0.2	0	820.38	804.47	788.05
40	0.2	0.5	925.41	909.36	893.51	0.2	0	823.51	807.81	791.77
45	0.2	0.5	928.10	912.26	896.74	0.2	0	826.40	811.14	795.31
50	0.2	0.5	930.71	915.15	899.85	0.2	0	829.27	814.22	798.67
55	0.2	0.5	933.31	917.97	902.83	0.2	0	832.01	817.29	801.96
60	0.2	0.5	935.72	920.65	905.74	0.2	0	834.68	820.13	805.12
65	0.2	0.5	938.11	923.27	908.62	0.2	0	837.22	823.00	808.26
0.1	0.3	0.7	943.36	925.22	906.46	0.3	0.2	860.49	841.72	821.95
5	0.3	0.7	946.73	928.87	910.66	0.3	0.2	864.17	845.78	826.62
10	0.3	0.7	949.90	932.31	914.61	0.3	0.2	867.71	849.70	831.30
15	0.3	0.7	952.83	935.62	918.37	0.3	0.2	871.06	853.48	835.43
20	0.3	0.7	955.79	938.86	921.89	0.3	0.2	874.23	857.03	839.42
25	0.3	0.7	958.63	942.13	925.39	0.3	0.2	877.37	860.61	843.22
30	0.3	0.7	961.44	945.10	928.70	0.3	0.2	880.33	863.84	846.89
35	0.3	0.7	964.17	947.95	931.82	0.3	0.2	883.27	867.00	850.38
40	0.3	0.7	966.67	950.88	934.92	0.3	0.2	886.03	870.14	853.79
45	0.3	0.7	969.20	953.69	938.00	0.3	0.2	888.77	873.09	857.08
50	0.3	0.7	971.71	956.36	940.85	0.3	0.2	891.37	876.03	860.18
55	0.3	0.7	974.14	958.97	943.73	0.3	0.2	894.02	878.78	863.26
60	0.3	0.7	976.51	961.50	946.48	0.3	0.2	896.48	881.52	866.16
65	0.3	0.7	978.85	964.01	949.26	0.3	0.2	898.92	884.18	869.15
0.1	0.3	0.6	930.47	912.01	893.14	0.3	0.1	836.12	817.24	797.30
5	0.3	0.6	933.84	915.65	897.17	0.3	0.1	839.96	821.57	802.29
10	0.3	0.6	936.95	919.14	901.17	0.3	0.1	843.55	825.69	806.97
15	0.3	0.6	939.99	922.51	904.93	0.3	0.1	847.06	829.53	811.35
20	0.3	0.6	942.95	925.80	908.56	0.3	0.1	850.33	833.23	815.50
25	0.3	0.6	945.78	929.06	912.10	0.3	0.1	853.53	836.81	819.46
30	0.3	0.6	948.59	932.15	915.47	0.3	0.1	856.59	840.25	823.18
35	0.3	0.6	951.32	934.99	918.64	0.3	0.1	859.64	843.46	826.72
40	0.3	0.6	953.98	937.92	921.80	0.3	0.1	862.50	846.65	830.29
45	0.3	0.6	956.56	940.72	924.88	0.3	0.1	865.29	849.71	833.62
50	0.3	0.6	959.07	943.45	927.72	0.3	0.1	868.00	852.69	836.77
55	0.3	0.6	961.51	946.11	930.65	0.3	0.1	870.69	855.55	839.96
60	0.3	0.6	963.81	948.69	933.39	0.3	0.1	873.15	858.23	842.96
65	0.3	0.6	966.21	951.20	936.18	0.3	0.1	875.64	861.00	845.95
0.1	0.3	0.5	916.72	898.20	879.12	0.3	0	805.92	787.00	766.76
5	0.3	0.5	920.14	901.95	883.21	0.3	0	809.97	791.59	772.11
10	0.3	0.5	923.36	905.55	887.37	0.3	0	813.72	795.93	777.06
15	0.3	0.5	926.50	909.02	891.23	0.3	0	817.44	799.98	781.76
20	0.3	0.5	929.46	912.26	894.96	0.3	0	820.87	803.84	786.06
25	0.3	0.5	932.40	915.52	898.50	0.3	0	824.17	807.62	790.23
30	0.3	0.5	935.26	918.71	901.92	0.3	0	827.34	811.12	794.11
35	0.3	0.5	937.99	921.71	905.15	0.3	0	830.43	814.43	797.86
40	0.3	0.5	940.65	924.59	908.35	0.3	0	833.45	817.72	801.47
45	0.3	0.5	943.28	927.44	911.43	0.3	0	836.34	820.94	804.91

TABLE I (Continued)

			303.15 K	323.15 K	343.15 K			303.15 K	323.15 K	343.15 K
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
50	0.3	0.5	945.73	930.17	914.43	0.3	0	839.05	823.75	808.16
55	0.3	0.5	948.22	932.88	917.36	0.3	0	841.74	826.77	811.40
60	0.3	0.5	950.63	935.46	920.21	0.3	0	844.30	829.55	814.45
65	0.3	0.5	952.97	938.02	922.94	0.3	0	846.95	832.26	817.48
0.1	0.3	0.4	900.41	881.69	862.30	0.4	0.6	949.43	930.94	912.15
5	0.3	0.4	903.88	885.54	867.01	0.4	0.6	952.63	934.47	916.08
10	0.3	0.4	907.16	889.14	870.86	0.4	0.6	955.64	937.70	919.82
15	0.3	0.4	910.35	892.77	874.83	0.4	0.6	958.57	940.96	923.41
20	0.3	0.4	913.36	896.06	878.56	0.4	0.6	961.32	944.03	926.88
25	0.3	0.4	916.35	899.43	882.21	0.4	0.6	964.04	947.14	930.21
30	0.3	0.4	919.21	902.61	885.72	0.4	0.6	966.69	950.06	933.36
35	0.3	0.4	921.99	905.56	889.00	0.4	0.6	969.31	952.85	936.43
40	0.3	0.4	924.81	908.54	892.21	0.4	0.6	971.92	955.57	939.43
45	0.3	0.4	927.39	911.45	895.34	0.4	0.6	974.40	958.37	942.35
50	0.3	0.4	929.95	914.23	898.39	0.4	0.6	976.80	960.94	945.19
55	0.3	0.4	932.43	916.88	901.37	0.4	0.6	979.18	963.49	947.96
60	0.3	0.4	934.79	919.51	904.17	0.4	0.6	981.43	965.97	950.66
65	0.3	0.4	937.18	922.12	907.16	0.4	0.6	983.61	968.37	953.39
0.1	0.3	0.3	881.99	863.24	843.49	0.4	0.5	935.22	916.63	897.69
5	0.3	0.3	885.51	867.04	848.00	0.4	0.5	938.43	920.22	901.62
10	0.3	0.3	888.89	870.85	852.37	0.4	0.5	941.49	923.60	905.51
15	0.3	0.3	892.14	874.48	856.50	0.4	0.5	944.42	926.86	909.16
20	0.3	0.3	895.20	877.92	860.33	0.4	0.5	947.27	930.04	912.62
25	0.3	0.3	898.30	881.34	864.08	0.4	0.5	950.04	933.20	916.06
30	0.3	0.3	901.21	884.63	867.60	0.4	0.5	952.80	936.17	919.31
35	0.3	0.3	904.04	887.68	870.93	0.4	0.5	955.42	938.96	922.44
40	0.3	0.3	906.80	890.77	874.29	0.4	0.5	957.97	941.73	925.43
45	0.3	0.3	909.54	893.67	877.52	0.4	0.5	960.50	944.48	928.40
50	0.3	0.3	912.10	896.50	880.57	0.4	0.5	962.85	947.10	931.24
55	0.3	0.3	915.02	899.21	883.55	0.4	0.5	965.28	949.65	934.06
60	0.3	0.3	917.42	901.84	886.51	0.4	0.5	967.53	952.12	936.76
65	0.3	0.3	919.81	904.39	889.39	0.4	0.5	969.76	954.58	939.49
0.1	0.4	0.4	918.47	900.00	881.18	0.5	0.5	956.20	937.79	918.98
5	0.4	0.4	921.78	903.53	885.16	0.5	0.5	959.19	941.00	922.70
10	0.4	0.4	924.94	906.97	889.21	0.5	0.5	962.04	944.12	926.27
15	0.4	0.4	927.98	910.38	892.91	0.5	0.5	964.75	947.27	929.71
20	0.4	0.4	930.88	913.61	896.48	0.5	0.5	967.44	950.24	933.06
25	0.4	0.4	933.82	916.88	900.02	0.5	0.5	970.11	953.18	936.18
30	0.4	0.4	936.57	919.96	903.33	0.5	0.5	972.65	956.05	939.22
35	0.4	0.4	939.36	922.85	906.55	0.5	0.5	975.22	958.68	942.19
40	0.4	0.4	942.07	925.73	909.65	0.5	0.5	977.66	961.34	945.07
45	0.4	0.4	944.65	928.58	912.62	0.5	0.5	980.03	963.93	947.88
50	0.4	0.4	947.15	931.26	915.68	0.5	0.5	982.32	966.45	950.57
55	0.4	0.4	949.64	933.91	918.50	0.5	0.5	984.65	968.94	953.28
60	0.4	0.4	952.06	936.49	921.24	0.5	0.5	986.80	971.36	955.93
65	0.4	0.4	954.40	939.00	924.02	0.5	0.5	988.92	973.60	958.49
0.1	0.4	0.3	899.92	881.20	861.76	0.5	0.4	939.59	921.14	902.18
5	0.4	0.3	903.28	884.95	865.95	0.5	0.4	942.63	924.51	905.95
10	0.4	0.3	906.45	888.49	870.00	0.5	0.4	945.58	927.74	909.63
15	0.4	0.3	909.53	891.96	873.91	0.5	0.4	948.35	930.83	913.06

TABLE I (Continued)

			303.15K	323.15K	343.15K			303.15K	323.15K	343.15K
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
20	0.4	0.3	912.49	895.19	877.64	0.5	0.4	951.09	933.96	916.42
25	0.4	0.3	915.42	898.50	881.07	0.5	0.4	953.82	936.90	919.69
30	0.4	0.3	918.17	901.58	884.53	0.5	0.4	956.41	939.77	922.84
35	0.4	0.3	921.01	904.42	887.76	0.5	0.4	958.98	942.45	925.80
40	0.4	0.3	923.66	907.40	890.91	0.5	0.4	961.42	945.22	928.74
45	0.4	0.3	926.19	910.20	894.04	0.5	0.4	963.89	947.86	931.66
50	0.4	0.3	928.64	912.87	896.93	0.5	0.4	966.29	950.42	934.34
55	0.4	0.3	931.07	915.52	899.80	0.5	0.4	968.51	952.92	937.16
60	0.4	0.3	933.48	918.15	902.65	0.5	0.4	970.76	955.39	939.69
65	0.4	0.3	935.76	920.60	905.43	0.5	0.4	972.88	957.85	942.36
0.1	0.4	0.2	876.87	858.09	838.47	0.5	0.3	920.21	901.68	882.58
5	0.4	0.2	880.34	861.94	842.82	0.5	0.3	923.30	905.16	886.45
10	0.4	0.2	883.66	865.69	847.07	0.5	0.3	926.25	908.49	890.18
15	0.4	0.2	886.80	869.21	851.09	0.5	0.3	929.18	911.69	893.77
20	0.4	0.2	889.81	872.55	854.87	0.5	0.3	931.97	914.81	897.29
25	0.4	0.2	892.79	875.86	858.51	0.5	0.3	934.75	917.91	900.62
30	0.4	0.2	895.65	879.04	861.97	0.5	0.3	937.34	920.83	903.81
35	0.4	0.2	898.48	882.03	865.25	0.5	0.3	939.96	923.62	906.82
40	0.4	0.2	901.13	885.01	868.45	0.5	0.3	942.50	926.33	909.81
45	0.4	0.2	903.76	887.86	871.68	0.5	0.3	944.98	929.02	912.78
50	0.4	0.2	906.26	890.69	874.67	0.5	0.3	947.37	931.58	915.62
55	0.4	0.2	908.85	893.34	877.60	0.5	0.3	949.70	934.13	918.33
60	0.4	0.2	911.20	895.97	880.44	0.5	0.3	951.95	936.60	920.97
65	0.4	0.2	913.54	898.47	883.22	0.5	0.3	954.12	939.00	923.64
0.1	0.4	0.1	850.53	831.91	812.18	0.5	0.2	896.71	878.06	858.68
5	0.4	0.1	854.16	835.86	816.69	0.5	0.2	899.90	881.69	862.76
10	0.4	0.1	857.64	839.72	821.10	0.5	0.2	903.01	885.12	866.70
15	0.4	0.1	860.88	843.45	825.33	0.5	0.2	905.93	888.37	870.34
20	0.4	0.1	864.04	846.94	829.26	0.5	0.2	908.83	891.60	873.90
25	0.4	0.1	867.08	850.41	833.01	0.5	0.2	911.60	894.76	877.34
30	0.4	0.1	870.09	853.70	836.57	0.5	0.2	914.30	897.78	880.64
35	0.4	0.1	872.92	856.69	840.00	0.5	0.2	917.03	900.61	883.75
40	0.4	0.1	875.73	859.77	843.31	0.5	0.2	919.57	903.43	886.90
45	0.4	0.1	878.41	862.73	846.59	0.5	0.2	922.09	906.12	889.87
50	0.4	0.1	881.02	865.61	849.58	0.5	0.2	924.54	908.73	892.65
55	0.4	0.1	883.55	868.36	852.61	0.5	0.2	926.92	911.28	895.57
60	0.4	0.1	886.01	870.98	855.56	0.5	0.2	929.22	913.85	898.31
65	0.4	0.1	888.39	873.53	858.38	0.5	0.2	931.44	916.25	901.09
0.1	0.4	0	818.57	799.87	779.90	0.5	0.1	868.87	850.34	830.75
5	0.4	0	822.41	804.09	784.77	0.5	0.1	872.22	854.13	834.99
10	0.4	0	826.10	808.21	789.50	0.5	0.1	875.49	857.72	839.13
15	0.4	0	829.61	812.20	793.88	0.5	0.1	878.63	861.13	843.04
20	0.4	0	832.88	815.85	798.08	0.5	0.1	881.58	864.46	846.76
25	0.4	0	836.01	819.42	801.93	0.5	0.1	884.45	867.72	850.24
30	0.4	0	839.08	822.86	805.70	0.5	0.1	887.31	870.84	853.70
35	0.4	0	842.12	826.07	809.34	0.5	0.1	890.03	873.78	856.92
40	0.4	0	845.03	829.20	812.69	0.5	0.1	892.62	876.65	860.06
45	0.4	0	847.82	832.31	816.08	0.5	0.1	895.25	879.50	863.19
50	0.4	0	850.47	835.18	819.23	0.5	0.1	897.70	882.22	866.13
55	0.4	0	853.06	838.04	822.41	0.5	0.1	900.23	884.81	868.94

TABLE I (Continued)

			303.15 K	323.15 K	343.15 K			303.15 K	323.15 K	343.15 K
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
60	0.4	0	855.56	840.71	825.35	0.5	0.1	902.47	887.38	871.78
65	0.4	0	858.00	843.42	828.33	0.5	0.1	904.86	889.88	874.56
0.1	0.5	0	833.95	815.45	795.79	0.6	0	852.60	834.57	815.42
5	0.5	0	837.51	819.40	800.18	0.6	0	856.01	838.30	819.55
10	0.5	0	840.99	823.20	804.59	0.6	0	859.21	841.84	823.58
15	0.5	0	844.18	826.87	808.76	0.6	0	862.29	845.24	827.49
20	0.5	0	847.39	830.36	812.80	0.6	0	865.19	848.46	831.10
25	0.5	0	850.32	833.83	816.44	0.6	0	868.00	851.66	834.58
30	0.5	0	853.33	836.95	819.94	0.6	0	870.86	854.78	837.82
35	0.5	0	856.16	840.10	823.32	0.6	0	873.58	857.61	841.03
40	0.5	0	858.91	843.13	826.67	0.6	0	876.22	860.48	844.12
45	0.5	0	861.53	846.08	829.90	0.6	0	878.68	863.21	847.18
50	0.5	0	864.13	848.90	832.83	0.6	0	881.13	865.88	850.01
55	0.5	0	866.72	851.59	835.85	0.6	0	883.61	868.47	852.82
60	0.5	0	869.07	854.11	838.75	0.6	0	885.90	870.98	855.56
65	0.5	0	871.50	856.77	841.57	0.6	0	888.18	873.43	858.27
0.1	0.6	0.4	963.36	945.25	926.46	0.7	0.3	972.50	954.73	936.39
5	0.6	0.4	966.30	948.30	929.86	0.7	0.3	975.21	957.56	939.57
10	0.6	0.4	968.98	951.26	933.32	0.7	0.3	977.79	960.35	942.76
15	0.6	0.4	971.64	954.19	936.49	0.7	0.3	980.29	963.13	945.77
20	0.6	0.4	974.22	957.05	939.63	0.7	0.3	982.60	965.82	948.69
25	0.6	0.4	976.73	959.94	942.64	0.7	0.3	985.05	968.44	951.49
30	0.6	0.4	979.05	962.54	945.52	0.7	0.3	987.32	971.04	954.21
35	0.6	0.4	981.57	965.11	948.43	0.7	0.3	989.67	973.34	956.90
40	0.6	0.4	983.90	967.67	951.15	0.7	0.3	991.90	975.79	959.46
45	0.6	0.4	986.22	970.20	953.97	0.7	0.3	994.05	978.16	962.06
50	0.6	0.4	988.46	972.61	956.43	0.7	0.3	996.12	980.46	964.53
55	0.6	0.4	990.62	974.94	959.04	0.7	0.3	998.18	982.69	966.92
60	0.6	0.4	992.60	977.25	961.58	0.7	0.3	1000.22	984.84	969.24
65	0.6	0.4	994.73	979.44	964.09	0.7	0.3	1002.23	987.03	971.70
0.1	0.6	0.3	944.73	926.42	907.44	0.7	0.2	948.28	933.38	911.99
5	0.6	0.3	947.60	929.52	910.99	0.7	0.2	950.99	933.44	915.22
10	0.6	0.3	950.39	932.58	914.50	0.7	0.2	953.62	936.34	918.46
15	0.6	0.3	953.10	935.62	917.83	0.7	0.2	956.17	939.11	921.63
20	0.6	0.3	955.74	938.48	921.02	0.7	0.2	958.64	941.80	924.60
25	0.6	0.3	958.24	941.37	924.09	0.7	0.2	961.03	944.52	927.56
30	0.6	0.3	960.78	944.07	927.12	0.7	0.2	963.41	947.12	930.27
35	0.6	0.3	963.24	946.64	929.92	0.7	0.2	965.76	949.58	933.01
40	0.6	0.3	965.63	949.36	932.75	0.7	0.2	968.03	952.08	935.68
45	0.6	0.3	967.88	951.84	935.56	0.7	0.2	970.18	954.50	938.38
50	0.6	0.3	970.18	954.29	938.13	0.7	0.2	972.36	956.85	940.90
55	0.6	0.3	972.39	956.68	940.74	0.7	0.2	974.53	959.18	943.40
60	0.6	0.3	974.54	958.99	943.22	0.7	0.2	976.51	961.39	945.77
65	0.6	0.3	976.60	961.34	945.73	0.7	0.2	978.58	963.63	948.17
0.1	0.6	0.2	920.38	902.12	883.12	0.8	0	909.52	892.72	875.01
5	0.6	0.2	923.36	905.43	886.78	0.8	0	912.12	895.59	878.23
10	0.6	0.2	926.20	908.54	890.40	0.8	0	914.74	898.32	881.41
15	0.6	0.2	929.02	911.63	893.83	0.8	0	917.23	901.09	884.41
20	0.6	0.2	931.65	914.59	897.13	0.8	0	919.64	903.77	887.38
25	0.6	0.2	934.31	917.59	900.35	0.8	0	921.97	906.44	890.11

TABLE I (Continued)

			303.15 K	323.15 K	343.15 K			303.15 K	323.15 K	343.15 K
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
30	0.6	0.2	936.84	920.39	903.38	0.8	0	924.34	908.92	892.82
35	0.6	0.2	939.36	923.07	906.34	0.8	0	926.63	911.27	895.45
40	0.6	0.2	941.79	925.73	909.22	0.8	0	928.79	913.71	897.89
45	0.6	0.2	944.10	928.26	912.02	0.8	0	930.99	916.02	900.48
50	0.6	0.2	946.39	930.77	914.70	0.8	0	933.06	918.20	902.89
55	0.6	0.2	948.66	933.20	917.41	0.8	0	935.16	920.47	905.32
60	0.6	0.2	950.85	935.56	919.94	0.8	0	937.19	922.61	907.53
65	0.6	0.2	952.97	937.91	922.50	0.8	0	939.15	924.74	909.87
0.1	0.6	0.1	890.71	872.52	853.27	0.7	0.1	918.08	900.27	881.50
5	0.6	0.1	893.91	876.04	857.19	0.7	0.1	920.85	903.36	884.99
10	0.6	0.1	896.90	879.21	861.02	0.7	0.1	923.63	906.37	888.45
15	0.6	0.1	899.83	882.46	864.55	0.7	0.1	926.23	909.24	891.61
20	0.6	0.1	902.56	885.47	867.95	0.7	0.1	928.75	912.04	894.80
25	0.6	0.1	905.22	888.56	871.22	0.7	0.1	931.31	914.87	897.75
30	0.6	0.1	907.86	891.42	874.41	0.7	0.1	933.68	917.46	900.67
35	0.6	0.1	910.53	894.26	877.47	0.7	0.1	936.13	920.08	903.47
40	0.6	0.1	913.02	897.02	880.41	0.7	0.1	938.46	922.63	906.18
45	0.6	0.1	915.43	899.65	883.37	0.7	0.1	940.71	925.05	908.83
50	0.6	0.1	917.83	902.21	886.15	0.7	0.1	942.95	927.45	911.39
55	0.6	0.1	920.15	904.70	888.86	0.7	0.1	945.11	929.77	913.89
60	0.6	0.1	922.39	907.16	891.54	0.7	0.1	947.19	932.08	916.42
65	0.6	0.1	924.51	909.50	894.16	0.7	0.1	949.26	934.26	918.98
0.1	0.8	0.2	982.67	965.68	947.95	0.7	0	877.36	859.50	840.95
5	0.8	0.2	985.17	968.35	950.80	0.7	0	880.34	862.86	844.60
10	0.8	0.2	987.53	970.87	953.62	0.7	0	883.28	866.02	848.21
15	0.8	0.2	989.81	973.43	956.52	0.7	0	886.15	869.10	851.69
20	0.8	0.2	991.96	975.86	959.17	0.7	0	888.77	872.06	855.03
25	0.8	0.2	994.19	978.31	961.75	0.7	0	891.43	875.10	858.19
30	0.8	0.2	996.35	980.58	964.25	0.7	0	894.01	877.84	861.22
35	0.8	0.2	998.49	982.78	966.73	0.7	0	896.52	880.51	864.11
40	0.8	0.2	1000.61	985.01	969.03	0.7	0	898.95	883.22	866.99
45	0.8	0.2	1002.59	987.28	971.46	0.7	0	901.36	885.74	869.84
50	0.8	0.2	1004.61	989.41	973.71	0.7	0	903.64	888.25	872.46
55	0.8	0.2	1006.56	991.42	975.94	0.7	0	905.96	890.68	875.21
60	0.8	0.2	1008.49	993.52	978.21	0.7	0	908.09	893.03	877.68
65	0.8	0.2	1010.35	995.54	980.45	0.7	0	910.26	895.48	880.24
0.1	0.8	0.1	951.88	934.91	917.08	1	0	995.65	988.04	977.77
5	0.8	0.1	954.33	937.68	920.09	1	0	997.82	990.16	979.92
10	0.8	0.1	956.79	940.26	923.01	1	0	1000.01	992.31	982.09
15	0.8	0.1	959.17	942.86	925.96	1	0	1002.19	994.43	984.23
20	0.8	0.1	961.43	945.34	928.67	1	0	1004.34	996.53	986.35
25	0.8	0.1	963.71	947.85	931.35	1	0	1006.46	998.61	988.44
30	0.8	0.1	965.92	950.22	933.90	1	0	1008.57	1000.67	990.51
35	0.8	0.1	968.05	952.52	936.43	1	0	1010.65	1002.70	992.56
40	0.8	0.1	970.22	954.80	938.94	1	0	1012.72	1004.72	994.58
45	0.8	0.1	972.26	957.12	941.42	1	0	1014.76	1006.71	996.59
50	0.8	0.1	974.28	959.31	943.72	1	0	1016.78	1008.69	998.57
55	0.8	0.1	976.33	961.42	946.06	1	0	1018.79	1010.64	1000.53
60	0.8	0.1	978.26	963.57	948.32	1	0	1020.77	1012.58	1002.47
65	0.8	0.1	980.11	965.64	950.62	1	0	1022.74	1014.49	1004.40

TABLE I (Continued)

			$303.15K$	$323.15K$	$343.15K$
$P(MPa)$	x_w	x_d	$\rho(kg/m^3)$	$\rho(kg/m^3)$	$\rho(kg/m^3)$
0.1	0.9	0.1	993.35	979.15	963.15
5	0.9	0.1	996.01	981.43	965.63
10	0.9	0.1	998.04	983.63	968.01
15	0.9	0.1	1000.16	985.81	970.59
20	0.9	0.1	1002.20	988.02	972.92
25	0.9	0.1	1004.11	990.21	975.23
30	0.9	0.1	1006.10	992.32	977.41
35	0.9	0.1	1008.08	994.29	979.61
40	0.9	0.1	1009.98	996.31	981.75
45	0.9	0.1	1011.85	998.36	983.86
50	0.9	0.1	1013.66	1000.33	986.06
55	0.9	0.1	1015.55	1002.17	988.07
60	0.9	0.1	1017.32	1004.05	990.12
65	0.9	0.1	1019.01	1005.97	992.15
0.1	0.9	0	950.74	936.05	920.55
5	0.9	0	952.90	938.39	923.08
10	0.9	0	955.04	940.69	925.56
15	0.9	0	957.15	943.03	928.08
20	0.9	0	959.13	945.23	930.51
25	0.9	0	961.20	947.47	932.82
30	0.9	0	963.19	949.51	935.10
35	0.9	0	965.16	951.54	937.35
40	0.9	0	967.05	953.55	939.54
45	0.9	0	968.98	955.65	941.75
50	0.9	0	970.78	957.67	943.83
55	0.9	0	972.67	959.57	945.90
60	0.9	0	974.43	961.44	948.00
65	0.9	0	976.11	963.35	950.07

mixtures (1134 values at all for the 3 binary mixtures) and 1512 values for the ternary mixture, described by 36 compositions which cover the ternary diagram. The excess volume V^E is then calculated using the relation:

$$\rho = \frac{\sum_{i=1}^{3} x_i M_i}{\sum_{i=1}^{3} x_i V_i + V^E} \qquad (1)$$

$M = \sum_{i=1}^{3} x_i M_i$ is the equivalent molar mass of the mixture and $V_i = M_i/\rho_i$ is the molar volume of component (i).

As regards the density ρ the behavior is usual: ρ is observed to decrease with T, increase with P, and is monotonous with the composition x_i. Figure 1 represents the variations of ρ as a function of the molar fraction of water x_w at $T = 323.15\,\mathrm{K}$ for different values of pressure P, in the case of the binary water + DAA. The variations of ρ ac- cording to x_w are not linear. Figure 2 represents the variations of the excess volume V^E in the same conditions. There is a very marked effect. Figures 3 and 4 represent the variations of V^E for the other two systems. The effect is less marked for the binary water + 2-propanol and even less marked for the binary DAA + 2-propanol. It reflects decreasing attractive interactions. For the 3 binaries V^E is negative, whatever the pressure, the temperature and the composition. At a given composition and temperature set $|V^E|$ decreases when the pressure increases. At a given composition and pressure set $|V^E|$ decreases when the temperature increases. It is important to stress here that the significance of the minimum of the excess volume V^E needs to be interpreted with care. This quantity is relative to 1 mole of the mixture. From another point of view $\rho V^E/M$ is relative to the unit volume of the mixture. Figure 5 displays the variations of V^E and $\rho V^E/M$ for the binary water + DAA, at $T = 323.15\,\mathrm{K}$ and $P = 40\,\mathrm{MPa}$, as a function

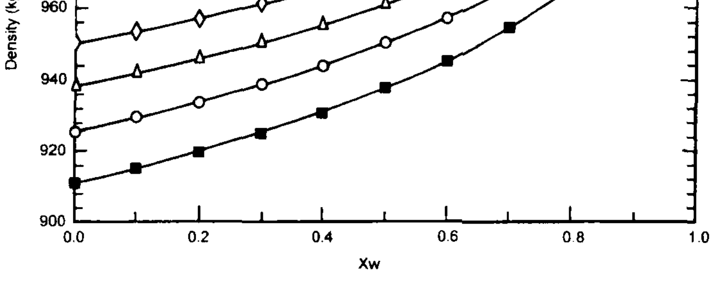

FIGURE 1 Binary Water + DAA. Variations of density ρ *versus* molar fraction of water x_w at $T = 323.15\,\mathrm{K}$ for different pressures (■ : 0.1 MPa, ○ : 20 MPa, ▲ : 40 MPa, ◇ : 60 MPa).

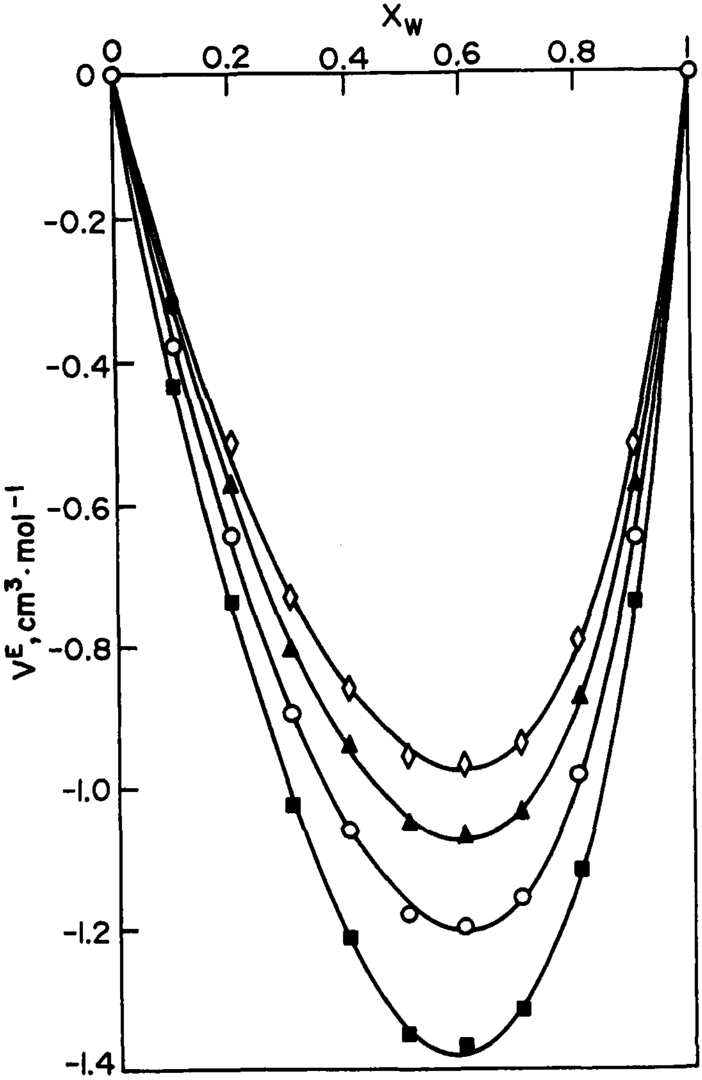

FIGURE 2 Binary Water + DAA. Variations of excess volume V^E *versus* molar fraction of water x_w at $T = 323.15$ K for different pressures (■ : 0.1 MPa, ○ : 20 MPa, ▲ : 40 MPa, ◇ : 60 MPa).

of the molar fraction of water x_w. There is a notable difference on the position of the two minima. Let us state here that for the 3 binaries the contraction $\rho V^E/M$ is maximum at $P = 1$ MPa and $T = 303.15$ K. One

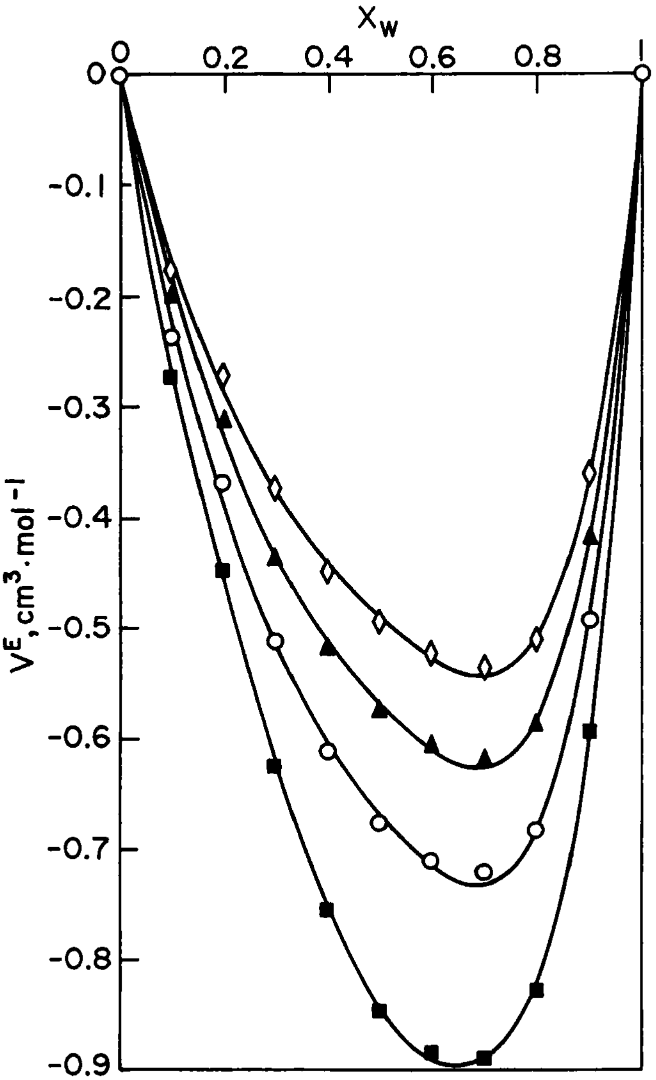

FIGURE 3 Binary Water+2-Propanol. Variations of excess volume V^E *versus* molar fraction of water x_w at $T = 323.15\,\mathrm{K}$ for different pressures (■ : 0.1 MPa, ○ : 20 MPa, ▲ : 40 MPa, ◇ : 60 MPa).

obtains 3.06% for the binary water + DAA at $x_w = 0.8$, 3.08% for the binary water + 2-propanol at $x_w = 0.8$ and 0.25% for the binary 2-propanol + DAA at $x_p = 0.6$. The maximum value of $\rho|V^E|/M$ is

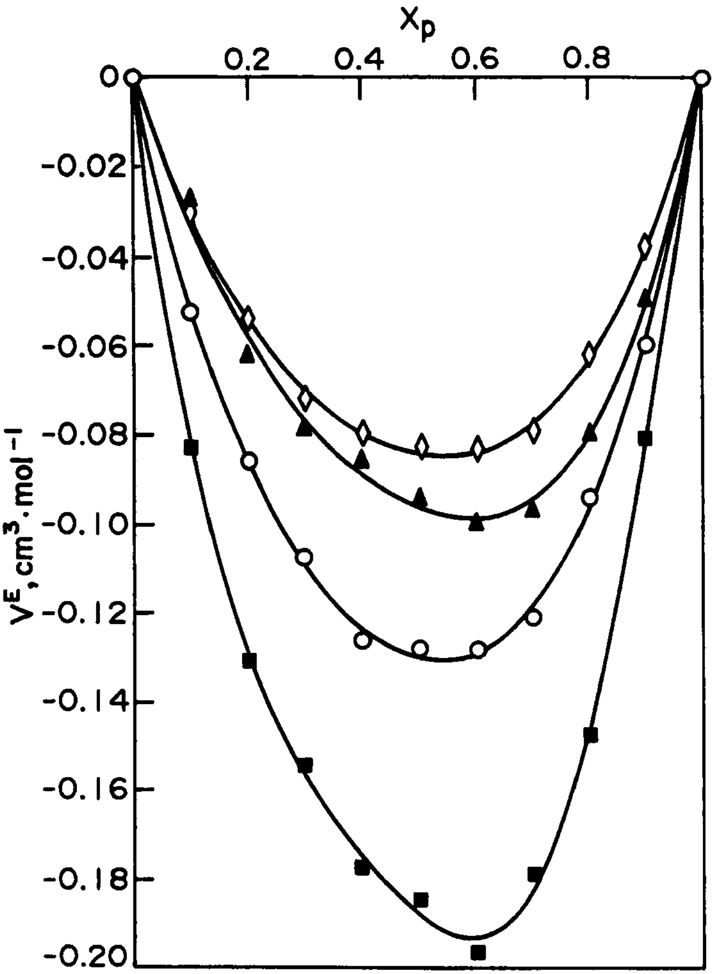

FIGURE 4 Binary 2-Propanol + DAA. Variations of excess volume V^E *versus* molar fraction of 2-propanol x_p at $T = 323.15$ K for different pressures (■ : 0.1 MPa, ○ : 20 MPa, ▲ : 40 MPa, ◇ : 60 MPa).

practically the same for the 2 binaries with water, whereas the maximum of $|V^E|$ is more marked for binary water + DAA than for binary water + 2-propanol (see Figs. 2 and 3). Which one of both quantities is really characteristic of the maximum effect of the

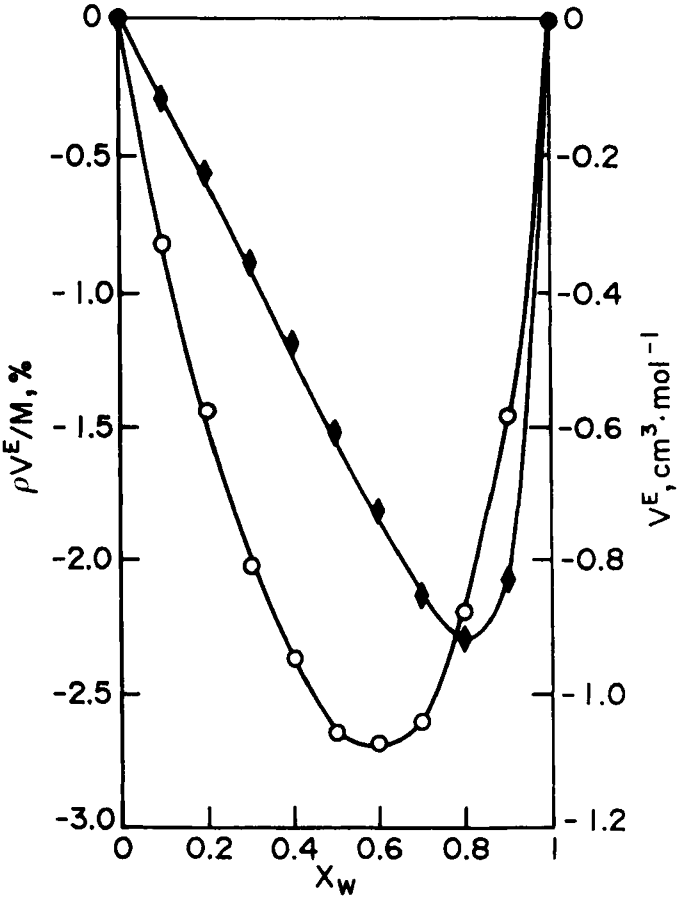

FIGURE 5 Binary Water + DAA. Variations of V^E and $\rho V^E/M$ *versus* molar fraction of water x_w at $T = 323.15$ K and $P = 40$ MPa (○: V^E is relative to 1 mole, ◆ : $\rho V^E/M$ is relative to 1 unit volume).

intermolecular interactions? From the excess volume point of view (a contraction effect in our case) $\rho V^E/M$ probably seems more significant. Figure 6 shows the variations of $\rho V^E/M$ according to the water content x_w at $P = 40$ MPa for the binary system water + DAA, at various temperatures.

With regard to the ternary, Figure 7 represents the surface V^E (x_w, x_d, x_p) in the ternary diagram, at $T = 323.15$ K and $P = 40$ MPa.

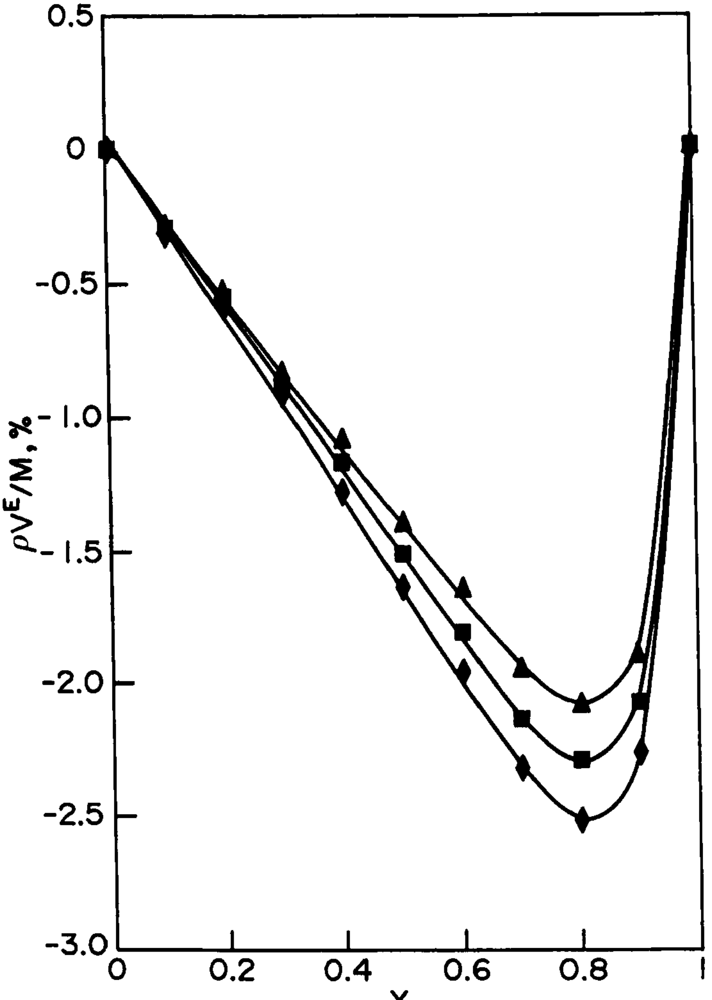

FIGURE 6 Binary water + DAA. Variations of $\rho V^E/M$ *versus* the molar ratio of water x_w at $P = 40$ MPa for different temperatures (▲ : 70°C, ■ : 50°C, ◆ : 30°C).

Figure 8 corresponds to the isoviscosity lines under the same conditions. Finally, Figure 9 represents surface $V^E(P, T)$ with $x_w = 0.3$, $x_d = 0.3$ and $x_p = 0.4$. As for the binaries, one notices that $|V^E|$ decreases if the pressure P increases or if the temperature T increases (see also Figs. 2 and 6 relative to the binary water + DAA).

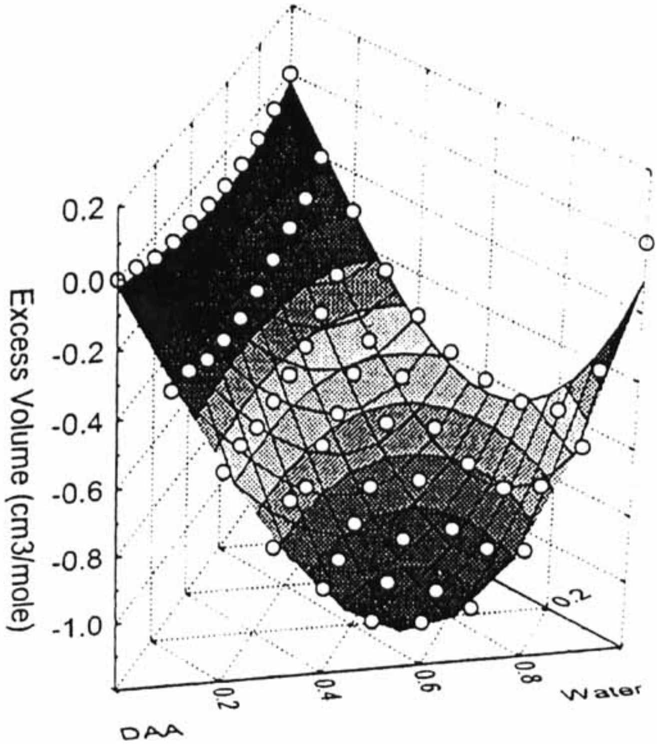

FIGURE 7 Surface $V^E(x_w, x_d, x_p)$ in the ternary diagram (P = 40 MPa, T = 323.15 K).

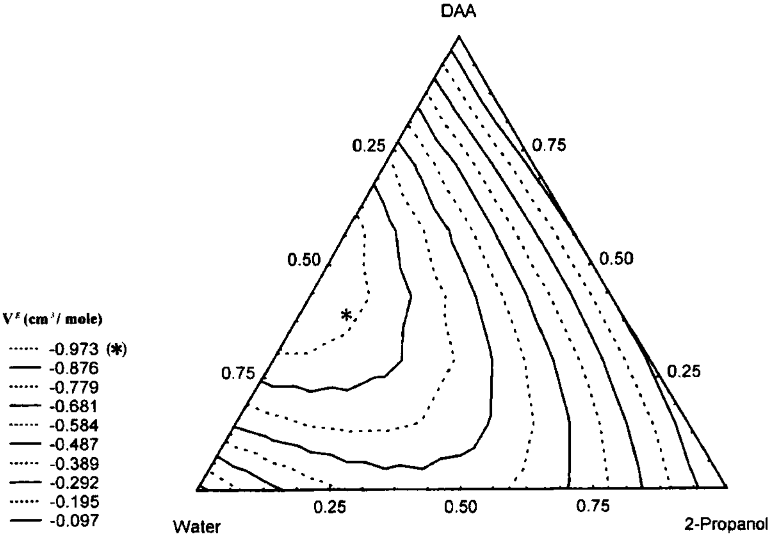

FIGURE 8 Iso-V^E lines in the ternary diagram (P = 40 MPa, T = 323.15 K).

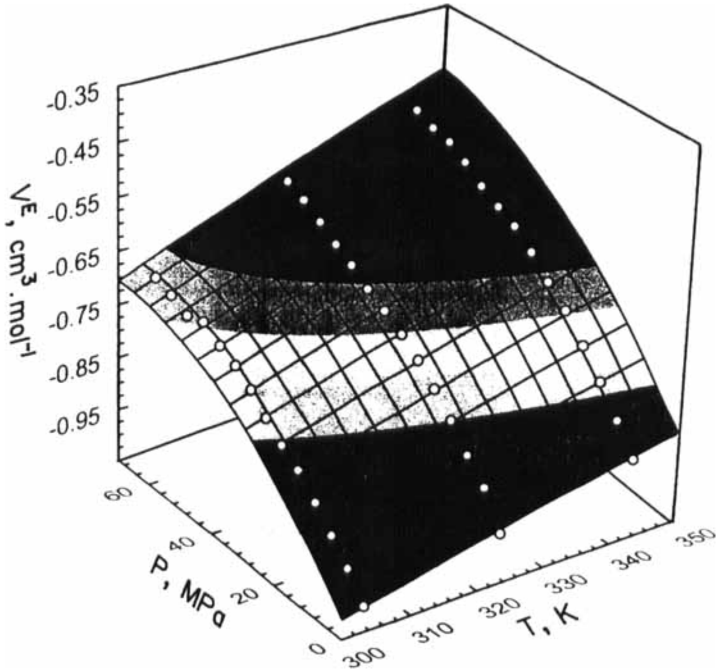

FIGURE 9 Surface $V^E(P, T)$ at $x_w = 0.3$, $x_d = 0.3$, $x_p = 0.4$.

DISCUSSION

At atmospheric pressure there is a very large number of studies relating to the excess volume of binary mixtures according to composition and temperature. There are far fewer studies relating to ternary mixtures. One can refer for example to articles [5–9]. In all the cases the excess volume of the binary mixture is represented by a Redlich-Kister polynomial equation:

$$V_{ij}^E = x_i x_j \sum_{k=0}^{m} A_k (1 - 2x_i)^k \tag{2}$$

Numerical analysis of our results showed that it was enough for each P, T set to develop Eq. (2) with only 3 A_k parameters in order to have a good representation of V_{ij}^E. A higher degree order of the

Redlich-Kister polynomial equation does not improve the restitution of the excess volume of the binaries. It is then necessary to represent the excess volume V_{123}^E of the ternary mixture. In a recent work at atmospheric pressure, Oswald and Platel [7] discussed 5 different methods for the calculation of the excess volume V_{123}^E of a ternary from the excess volumes V_{ij}^E of the binaries and the molar fractions x_i of the pure substances (the interested reader may find the development of these 5 methods in references [4] and [7]). These methods are noted: RK [10], TS [11], KO [12], RAST [13] and LARK [14]. Knowing A_k for each binary $(i)+(j)$ and each P, T set then the 5 models were tested with the ternary and density ρ was calculated with Eq. (1). In order to characterize the performances the following quantities were defined:

$$\mathrm{dev}_\rho(i) = 100\left(1 - \frac{\rho_{\mathrm{calc}}(i)}{\rho_{\mathrm{exp}}(i)}\right) \quad \mathrm{AAD}_\rho = \text{absolute average deviation} = \frac{1}{Nb}\sum_{i=1}^{Nb} \mathrm{abs}(\mathrm{dev}_\rho(i))$$

$$\mathrm{Dmax}_\rho = \mathrm{MAX}(\mathrm{abs}(\mathrm{dev}_\rho(i)))$$

$$\mathrm{Bias}_\rho = \frac{1}{Nb}\sum_{i=1}^{Nb} \mathrm{dev}_\rho(i) \quad \text{where } Nb \text{ is the number of data.}$$

Table II relates to the 3 binaries and ternary mixtures whereas Table III relates only to the ternary. Let us mention here that if the mixture is assumed to be ideal (*i.e.*, $V^E = 0$), then $\mathrm{AAD}_\rho = 1.03\%$, $\mathrm{Dmax}_\rho = 3.05\%$ and $\mathrm{Bias}_\rho = 1.03\%$ are obtained considering all the mixtures (binaries and ternary). For the ternary system only $\mathrm{AAD}_\rho = 1.09\%$, $\mathrm{Dmax}_\rho = 3.05\%$ and $\mathrm{Bias}_\rho = 1.09\%$. Tables II and III show that RK and KO models give the best results. Model RK [10] generalizes a relation suggested by Redlich and Kister for the representation of some thermodynamic properties of mixtures. It is expressed as:

$$V_{123}^E = V_{12}^E + V_{13}^E + V_{23}^E \tag{3}$$

where V_{12}^E, V_{13}^E, V_{23}^E are excess volume calculated using Eq. (2) with the values x_i of ternary ($x_i + x_j = 1 - x_k$ where x_k is the molar fraction of the third component). Model KO [12] generalizes an equation relating

TABLE II Results obtained with 5 different models (3 binaries and ternary :2646 experimental data)

	RK	*TS*	*KO*	*RAST*	*LARK*
$\mathrm{AAD}_{VE}\%$	4.91	22.46	5.08	16.75	35.31
$\mathrm{AAD}_{\rho}\%$	0.05	0.25	0.05	0.19	0.36
$\mathrm{Dmax}_{\rho}\%$	0.42	1.49	0.42	1.25	1.66
$\mathrm{Bias}_{\rho}\%$	0.04	0.25	−0.03	0.18	0.35

TABLE III Results obtained with 5 different models (only the ternary :1512 experimental data)

	RK	*TS*	*KO*	*RAST*	*LARK*
$\mathrm{AAD}_{VE}\%$	5.31	36.03	5.62	26.03	58.53
$\mathrm{AAD}_{\rho}\%$	0.06	0.42	0.07	0.31	0.61
$\mathrm{Dmax}_{\rho}\%$	0.36	1.49	0.37	1.25	1.66
$\mathrm{Bias}_{\rho}\%$	0.05	0.42	−0.06	0.31	0.61

to the excess enthalpy and its expression is:

$$V^E_{123} = (x_1 + x_2)^2 V^E_{12} + (x_1 + x_3)^2 V^E_{13} + (x_2 + x_3)^2 V^E_{23} \quad (4)$$

where V^E_{ij} refers to the binary $(i) + (j)$ and is calculated starting from Eq. (2) using $x^0_i = 1 - x^0_j = x_i/x_i + x_j$. Our results confirm those of Oswald and Platel [7] at atmospheric pressure. The introduction of the pressure parameter does not seem to deteriorate the ability of these equations to evaluate V^E. Let us note here that for the RK model $\mathrm{AAD}_\rho = 0.06\%$ (Eq. (3) and Tab. III) which corresponds to an error of about $\pm 0.6\,\mathrm{kg/m^3}$ over the whole experimental P, T range domain, whereas experimental uncertainty is about $\pm 0.1\,\mathrm{kg/m^3}$. In Tables II and III the maximum value for AAD_{VE} is not indicated, since, in the vicinity of the pure substances, V^E tends to 0, especially for binary DAA + 2-propanol which is far from associative. Under these conditions the deviation from the calculated value can be important but it is not significant. On the other hand, AAD_ρ is significant for the density never tends to 0.

The previous results are satisfactory but it is necessary as a preliminary step to determine from each binary the values of the 3 coefficients A_0, A_1 and A_2 of Eq. (2) for each P, T set. The numerical analysis shows that these coefficients vary with P and T. Even if one limits oneself to the representation of the associated surfaces by planes, there are 3 coefficients per plan, therefore 9 per binary and 27 for the 3 binary ones. Moreover, as $A_i(P,T)$ surfaces are not planes there is a deterioration of the results compared to Tables II and III. Therefore we propose a total representation of the mixtures (the 3 binary ones and the ternary) which does not use the polynomial Redlich-Kister Eq. (2). We used the following expression:

$$V^E = \frac{a_1 x_w x_d + a_2 x_w x_p + a_3 x_d x_p + a_4 x_w x_p x_d}{1 + a_5 x_w + a_6 x_d + a_7 x_p}(1 + a_8\sqrt{T} + a_9 P) \qquad (5)$$

which involves only 9 parameters to describe the whole system in our P, T range. The results are indicated in Table IV. It will be noticed that for the binary DAA + 2-propanol, which is not very associative, AAD_{VE} presents a maximum but that AAD_ρ and Dmax_ρ are smaller. This is because V^E is small for this binary. By comparison with Table III relating to the ternary, one notes that values of AAD_ρ (approximately 0.05%) and AAD_{VE} (approximately 5%) are practically identical, but that Dmax_ρ is only 0.27% with Eq. (5) instead of 0.36% with the RK model. The interest of Eq. (5) is that it requires a smaller number of coefficients compared to the other methods. Comparable results can be obtained with $(1 + a_8 T + a_9 P)$ or $(1 + a_8/T + a_9 P)$ instead of $(1 + a_8\sqrt{T} + a_9 P)$ (with however coefficients a_1 to a_9 different).

Finally we use Eq. (5) with only the binary terms characteristic of the presence of water *i.e.*, the very strong interactions water + DAA and water + 2-propanol. The binary DAA + 2-propanol is assumed to be ideal, compared to the binary with water. Writing:

$$V^E = \frac{a_1 x_w x_d + a_2 x_w x_p}{1 + a_3 x_w} \qquad (6)$$

$\mathrm{AAD}_{VE} = 28.1\%$, $\mathrm{AAD}_\rho = 0.17\%$, $\mathrm{Dmax}_\rho = 1.40\%$ and $\mathrm{Bias}_\rho = 0.06\%$ are obtained, which is a considerable improvement compared to the

TABLE IV Results obtained using the Eq. (5) with 9 parameters (W: water, 2-Pr: 2-propanol)

	W + 2-*Pr*	*W* + *DAA*	2-*Pr*+*DAA*	3 *Binaries*	*Ternary*
AAD_{VE}%	10.39	5.81	23.46	13.21	4.8
AAD_ρ%	0.14	0.07	0.02	0.075	0.05
Dmax_ρ%	0.82	0.30	0.09	0.82	0.27
Bias_ρ%	0.08	0.00	0.01	0.03	− 0.00

case where the 3 binaries and the ternary are assumed to be ideal (1.03%, 3.05% and 1.03% for density) insofar as there are only 3 parameters to represent all the 2646 experimental values (the binary DAA + 2-propanol which is represented by 378 values being almost ideal). Finally while introducing into Eq. (6) the corrective term $(1 + a_4\sqrt{T} + a_5P)$ one passes to a relationship with 5 coefficients for which $\mathrm{AAD}_{VE} = 22.5\%$, $\mathrm{AAD}_\rho = 0.09\%$, $\mathrm{Dmax}_\rho = 0.93\%$ and $\mathrm{Bias}_\rho = 0.03\%$ which is a very satisfactory result.

References

[1] Lagourette, B., Boned, C., Saint-Guirons, H., Xans, P. and Zhou, H. (1992). *Measure. Sci. Technol.*, **3**, 699.
[2] Kell, G. and Whalley, E. (1975). *J. Chem. Phys.*, **62**, 3496.
[3] Papaioannou, D., Bridakis, M. and Panayotou, C. G. (1993). *J. Chem. Eng. Data*, **38**, 370.
[4] Moha-Ouchane, M. (1999). Thése de Doctorat, Université de Fés.
[5] Mamagakis, N. and Panayiotou, C. (1989). *Z. Phys. Chem.*, **162**, 57.
[6] Aminabhavi, T. M. and Joshi, S. S. (1992). *Indian J. Technology*, **30**, 197.
[7] Oswald, S. L. and Patel, S. G. (1994). *J. Chem. Soc. Faraday Trans.*, **90**, 1083.
[8] Olivé, F., Patil, K. R., Fernandez, J. and Coronas, A. (1995). *Thermochimica Acta*, **259**, 57.
[9] Olivé, F., Chaudhari, S. K., Patil, K. R. and Coronas, A. (1996). *Can. J. Chem. Eng.*, **74**, 163.
[10] Redlich, O. and Kister, A. T. (1948). *Ind. and Eng. Chem.*, **40**, 345.
[11] Tsao, C. C. and Smith, J. M. (1983). Chemical Engineering Progress, *Symposium Series*: Applied Thermodynamics, **49**, 107.
[12] Kohler, F. (1960). *Monasth. Chem.*, **91**, 738.
[13] Rastogi, R. P., Nath, J. and Das, S. S. (1977). *J. Chem. Eng. Data*, **22**, 249.
[14] Lark, B., Kaur, S. and Singh, S. (1981). *Indian. J. Chem. Sect. A*, **26**, 197.