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THERMODYNAMIC PROPERTIES OF THE MIXTURES OF A DIOXANONANE ISOMER + *n*-OCTANE AT 298.15 K. Part II. Excess molar volumes and excess isentropic and isothermal compressibilities (measurements of density and speed of sound)

HISAKI NAKAI **, KATSUTOSHI TAMURA *, HIDEO OGAWA *** and SACHIO MURAKAMI

Department of Chemistry, Faculty of Science, Osaka City University, 3-3-138, Sugimoto, Sumiyoshi-ku, Osaka, 558 (Japan)

YOSHIO TOSHIYASU

Science Education Institute of Osaka Prefecture, 4-13-23, Karita-cho, Sumiyoshi-ku, Osaka, 558 (Japan)

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ABSTRACT

Densities, ρ, and speeds of sound, u, of six mixtures of an isomer of the diethers 2,*m*- or 3,*n*-dioxanonane ($m = 5$, 6, 7, 8; $n = 6$, 7) + *n*-octane were measured at 298.15 K. The densities provided excess molar volumes V^E, and excess isothermal compressibilities, κ_T^E, of the mixtures were estimated using heat capacities and isentropic compressibilities, κ_S^E, obtained from measurements of density and speed of sound. The excess volumes were positive and parabolic, ranging from 0.5 to 0.7 cm^3 mol^{-1} and depending on both the density and dipole moment, μ, of the pure diethers. The excess isentropic compressibilities are positive and parabolic, except for the 2,8-dioxanonane + *n*-octane system, where the curve is flattened at large diether mole fractions and the maximum shifts to the small mole fractions. The excess isothermal compressibilities are almost parallel to the excess isentropic compressibilities but are about 20% higher. The speeds of sound, isentropic and isothermal compressibilities of the pure diethers are all very similar to each other and strongly correlated with their densities.

INTRODUCTION

Only a few thermodynamic studies on mixtures containing a polyether have been reported [1,2] because such compounds are not readily available.

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* To whom correspondence should be addressed.
** Present address: Kiyo Software Service Co., Ltd., Wakayama, Wakayama, Japan.
*** Present address: Faculty of Science and Engineering, Tokyo Denki University, Hiki-gun, Saitama 350-03, Japan.

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We are interested in the way in which the positions of functional groups (in this case two ether oxygen atoms) in isomers affect the thermodynamic properties of mixtures. In a previous paper [3] we reported excess enthalpies and excess heat capacities of binary mixtures of six dioxanonane isomers with *n*-octane. Thermodynamic properties of these mixtures are explained in terms of the dipole moments of the pure substances, and the behavior of excess heat capacities of the mixtures discussed in terms of fluctuations of local concentration. In this paper, we report excess volumes and isentropic and isothermal compressibilities obtained from the measurements of density and speed of sound.

EXPERIMENTAL

The diethers, prepared by the Williamson synthesis, and *n*-octane were fractionally distilled as described previously [3].

Density was measured by a vibrational densimeter (Anton Paar DMA 602) as described elsewhere [4]. The speed of sound was measured by the "sing-around" method (Cho-Onpa Kogyo Co., UVM-2) which has been described previously [5]. The error in the density measurements is less than 10^{-5} g cm^{-3}, and the accuracy of the speed of sound is ± 0.6 m s^{-1}. The relative precisions of excess parameters for these mixtures are within 0.3 m s^{-1}.

RESULTS AND DISCUSSION

The physical properties of the pure materials are presented in Table 1. The properties of the isomers are all very similar to each other. Speeds of sound, u, isentropic and isothermal compressibilities, κ_S and κ_T, are corre-

TABLE 1

Thermodynamic properties of dioxanonane isomers and octane at 298.15 K

Material	ρ (g cm^{-3})	C_p (J K^{-1} mol^{-1})	α (kK^{-1})	μ (D)	u (m s^{-1})	κ_S (T Pa^{-1})	κ_T (T Pa^{-1})
Octane	0.69855	254.2	1.15	0.0	1172.9	1040.5	1294.2
Dioxanonane							
2,5-	0.84199	278.8	1.17	1.56	1201.2	823.1	1053.0
2,6-	0.83744	279.0	1.12	1.48	1179.8	857.9	1069.5
2,7-	0.84147	280.4	1.12	1.63	1198.7	827.0	1036.6
2,8-	0.85298	278.2	1.04	1.73	1242.0	760.0	939.7
3,6-	0.83307	278.2	1.13	1.50	1171.1	875.3	1093.2
3,7-	0.83127	285.2	1.12	1.49	1156.8	898.9	1107.4

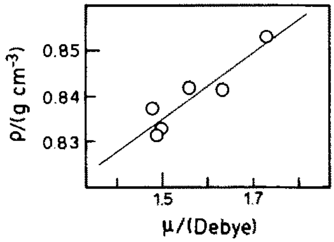

Fig. 1. Linear relation of density, ρ, to dipole moment, μ, for dioxanonane isomers

lated with the densities, ρ, which in turn are highly correlated with the dipole moment, μ, of the diether. The relation of ρ to μ is almost linear (Fig. 1) and the standard deviation is within 0.003 g cm^{-3}. ρ at $\mu = 0.0$ is 0.725 and coincidentally agrees with the value ($\rho = 0.713$) for n-nonane.

Excess volumes in Table 2 are estimated from ρ by the following equation

$$V^{E} = [xM_1 + (1-x)M_2]/\rho_{mix} - xM_1/\rho_1 - (1-x)M_2/\rho_2 \qquad (1)$$

where M_1 and M_2 are the molar masses of the diether and n-octane, respectively. In this table, the suffixes obs and calc identify the values observed and calculated by the smoothed function, respectively. The smoothed function for an excess property is estimated by the least-squares method

$$X^{E}(\text{unit}) = x(1-x)\sum A_i(1-2x)^{i-1} \qquad (2)$$

where x is the mole fraction of the diether, and X^E is the excess property. The coefficients A_i are given in Table 5.

In Fig. 2, the V^E curves for the mixtures are seen to be positive and parabolic, with values of the same order as the molar volumes of the pure diethers, as expected.

Table 3 lists ρ, u, κ_S, and excess isentropic compressibility, κ_S^E, for these mixtures estimated from the following relations

$$\kappa_S = 1/(u\rho^2) \qquad (3)$$

$$\kappa_S^E = \kappa_S - \kappa_S^{id} \qquad (4)$$

$$\kappa_S^{id} = \kappa_T^{id} - (\alpha^{id})^2 V^{id} T / C_p^{id} \qquad (5)$$

where

$$\kappa_T^{id} = \phi\kappa_{T,1} + (1-\phi)\kappa_{T,2} \qquad (6)$$

$$\alpha^{id} = \phi\alpha_1 + (1-\phi)\alpha_2,$$

and V^{id} and C_p^{id} are represented by

$$X^{id} = xX_1 + (1-x)X_2.$$

TABLE 2

Molar excess volume of a dioxanonane isomer + n-octane mixtures at 298.15 K

Mole fraction diether x	V^E (obs.) (cm^3 mol^{-1})	V^E (calc.) (cm^3 mol^{-1})	Diff.
x 2,5-dioxanonane + (1 − x) n-octane			
0.09997	0.2422	0.2432	−0.0010
0.19428	0.4080	0.4070	0.0010
0.29799	0.5236	0.5241	−0.0005
0.39333	0.5811	0.5806	0.0005
0.49421	0.5923	0.5922	0.0001
0.59577	0.5560	0.5574	−0.0014
0.69285	0.4833	0.4828	0.0005
0.79532	0.3631	0.3622	0.0009
0.89707	0.2025	0.2016	0.0009
0.93972	0.1204	0.1227	−0.0023
x 2,6-dioxanonane + (1 − x) n-octane			
0.09939	0.2258	0.2276	−0.0018
0.19567	0.3849	0.3838	0.0011
0.30109	0.4956	0.4947	0.0009
0.39755	0.5485	0.5485	−0.0000
0.49723	0.5607	0.5611	−0.0004
0.59479	0.5302	0.5333	−0.0031
0.69758	0.4655	0.4619	0.0036
0.79727	0.3514	0.3512	0.0002
0.90132	0.1903	0.1920	−0.0017
x 2,7-dioxanonane + (1 − x) n-octane			
0.10036	0.2347	0.2367	−0.0020
0.19696	0.3980	0.3960	0.0020
0.29991	0.5082	0.5074	0.0008
0.39589	0.5640	0.5651	−0.0011
0.49697	0.5796	0.5816	−0.0020
0.59628	0.5559	0.5540	0.0019
0.69560	0.4833	0.4823	0.0010
0.79650	0.3624	0.3640	−0.0016
0.89822	0.2012	0.2008	0.0004
0.94522	0.1123	0.1121	0.0002
x 2,8-dioxanonane + (1 − x) n-octane			
0.10115	0.2168	0.2190	−0.0022
0.19590	0.3627	0.3595	0.0032
0.29954	0.4565	0.4567	−0.0002
0.39445	0.4987	0.5023	−0.0036
0.49353	0.5130	0.5107	0.0023
0.59272	0.4824	0.4812	0.0012
0.69369	0.4126	0.4136	−0.0010
0.79309	0.3104	0.3112	−0.0008
0.89444	0.1740	0.1732	0.0008

TABLE 2 (continued)

Mole fraction diether x	V^E (obs.) ($cm^3\ mol^{-1}$)	V^E (calc.) ($cm^3\ mol^{-1}$)	Diff.
x 3,6-dioxanonane + (1 − x) *n*-octane			
0.09882	0.2595	0.2611	−0.0016
0.19654	0.4514	0.4488	0.0026
0.30052	0.5799	0.5811	−0.0012
0.39483	0.6457	0.6468	−0.0011
0.49437	0.6657	0.6643	0.0014
0.59060	0.6327	0.6330	−0.0003
0.69024	0.5521	0.5523	−0.0002
0.78690	0.4281	0.4281	−0.0000
0.88373	0.2597	0.2593	0.0004
0.94746	0.1242	0.1247	−0.0005
x 3,7-dioxanonane + (1 − x) *n*-octane			
0.09949	0.2530	0.2547	−0.0017
0.19616	0.4301	0.4293	0.0008
0.29611	0.5506	0.5488	0.0018
0.40037	0.6160	0.6169	−0.0009
0.49752	0.6318	0.6329	−0.0011
0.59374	0.6043	0.6051	−0.0008
0.69932	0.5263	0.5243	0.0020
0.79396	0.4064	0.4061	0.0003
0.89444	0.2322	0.2330	−0.0008
0.94383	0.1297	0.1303	−0.0006

where ϕ is the volume fraction of the diether, X_1 and X_2 are propeties of the pure liquids, and X^{id} the corresponding property of the ideal mixing state.

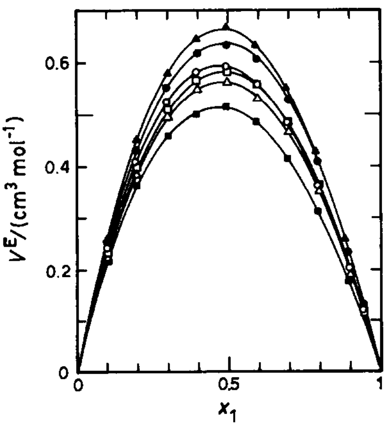

Fig. 2. Excess volumes of x-dioxanonane isomer + (1 − x) *n*-octane: ○, 2,5-dioxanonane + *n*-octane; ●, 2,6-dioxanonane + *n*-octane; □, 2,7-dioxanonane + *n*-octane; ■, 2,8-dioxanonane + *n*-octane; ▲, 3,6-dioxanonane + *n*-octane; △, 3,7-dioxanonane + *n*-octane.

TABLE 3
Density, speed of sound, isentropic compressibility, and excess isentropic compressibility of a dioxanonane isomer + n-octane at 298.15 K

Mole fraction diether x	ρ (g cm^{-3})	u (m s^{-1})	κ_S (T Pa^{-1})	κ_S^E(obs.) (T Pa^{-1})	κ_S^E(calc.) (T Pa^{-1})	Diff. (T Pa^{-1})
x 2,5-dioxanonane + $(1-x)$ n-octane						
0.00000	0.698540	1172.93	1040.55			
0.09997	0.711307	1170.33	1026.43	6.53	6.50	0.03
0.19428	0.723687	1169.14	1010.93	10.73	10.73	−0.00
0.29799	0.737686	1169.08	991.83	13.55	13.57	−0.02
0.39333	0.750867	1170.13	972.68	14.77	14.73	0.04
0.49421	0.765149	1172.50	950.66	14.55	14.66	−0.11
0.59577	0.779876	1175.82	927.45	13.52	13.45	0.07
0.69285	0.794263	1180.22	903.88	11.40	11.36	0.04
0.79532	0.809810	1186.01	877.90	8.30	8.32	−0.02
0.89707	0.825616	1192.91	851.16	4.54	4.53	0.01
0.93972	0.832369	1196.22	839.58	2.67	2.74	−0.07
1.00000	0.841989	1201.20	823.12			
x 2,6-dioxanonane + $(1-x)$ n-octane						
0.00000	0.698540	1172.94	1040.54			
0.09939	0.710928	1168.56	1030.08	6.61	6.68	−0.07
0.19567	0.723242	1165.43	1018.00	11.29	11.26	0.03
0.30109	0.737080	1163.32	1002.50	14.41	14.40	0.01
0.39755	0.750041	1162.49	986.58	15.76	15.75	0.01
0.49723	0.763717	1162.70	968.58	15.84	15.81	0.03
0.59479	0.777398	1163.96	949.47	14.65	14.69	−0.04
0.69758	0.792102	1166.32	928.07	12.39	12.40	−0.01
0.79727	0.806696	1169.69	906.04	9.16	9.19	−0.03
0.90132	0.822301	1174.23	881.98	4.97	4.90	0.07
1.00000	0.837438	1179.77	857.92			
x 2,7-dioxanonane + $(1-x)$ n-octane						
0.00000	0.698552	1173.05	1040.32			
0.10036	0.711362	1169.91	1027.09	6.84	6.88	−0.04
0.19696	0.724032	1168.19	1012.08	11.43	11.44	−0.01
0.29991	0.737902	1167.71	993.88	14.43	14.39	0.04
0.39589	0.751138	1168.50	975.04	15.62	15.60	0.02
0.49697	0.765381	1170.53	953.57	15.56	15.51	0.05
0.59628	0.779674	1173.88	930.76	14.06	14.24	−0.18
0.69560	0.794331	1178.03	907.16	12.08	11.97	0.11
0.79650	0.809595	1183.67	881.60	8.77	8.77	−0.00
0.89822	0.825339	1190.50	854.89	4.80	4.75	0.05
0.94522	0.832745	1194.20	842.04	2.57	2.65	−0.08
1.00000	0.841469	1198.73	827.03			

κ_T and excess isothermal compressibilities for these mixtures, κ_T^E, are presented in Table 4. κ_T and κ_T^E are estimated by the following relations

$$\kappa_T = \kappa_S + \alpha^2 VT/C_p \qquad (7)$$

$$\kappa_T^E = \kappa_T - \kappa_T^{id} \qquad (8)$$

TABLE 3 (continued)

Mole fraction diether x	ρ ($g\ cm^{-3}$)	u ($m\ s^{-1}$)	κ_S ($T\ Pa^{-1}$)	κ_S^E(obs.) ($T\ Pa^{-1}$)	κ_S^E(calc.) ($T\ Pa^{-1}$)	Diff. ($T\ Pa^{-1}$)
x 2,8-dioxanonane + $(1-x)$ *n*-octane						
0.00000	0.698545	1173.03	1040.37			
0.10115	0.712479	1173.99	1018.36	3.83	3.97	−0.14
0.19590	0.725889	1176.01	996.12	6.31	6.16	0.15
0.29954	0.740984	1179.85	969.47	7.27	7.20	0.07
0.39445	0.755143	1184.71	943.52	7.11	7.22	−0.11
0.49353	0.770214	1190.92	915.43	6.49	6.56	−0.07
0.59272	0.785681	1198.30	886.38	5.49	5.44	0.05
0.69369	0.801798	1207.14	855.90	4.11	4.05	0.06
0.79309	0.818024	1217.12	825.22	2.64	2.61	0.03
0.89444	0.834945	1228.68	793.35	1.13	1.22	−0.09
1.00000	0.852979	1242.03	759.97			
x 3,6-dioxanonane + $(1-x)$ *n*-octane						
0.00000	0.698536	1172.96	1040.50			
0.09882	0.710344	1167.44	1032.91	7.91	7.95	−0.04
0.19654	0.722341	1163.21	1023.16	13.67	13.63	0.04
0.30052	0.735491	1160.06	1010.32	17.53	17.52	0.01
0.39483	0.747706	1158.36	996.75	19.28	19.31	−0.03
0.49437	0.760895	1157.62	980.72	19.59	19.58	0.01
0.59060	0.773967	1157.99	963.53	18.38	18.40	−0.02
0.69024	0.787815	1159.41	944.28	15.85	15.82	0.03
0.78690	0.801572	1161.91	924.09	12.06	12.09	−0.03
0.88373	0.815681	1165.38	902.71	7.27	7.23	0.04
0.94746	0.825157	1168.33	887.83	3.41	3.45	−0.04
1.00000	0.833074	1171.07	875.29			
x 3,7-dioxanonane + $(1-x)$ *n*-octane						
0.00000	0.698562	1173.05	1040.31			
0.09949	0.710335	1165.93	1035.61	8.04	8.02	0.02
0.19616	0.722106	1160.35	1028.54	14.28	14.30	−0.02
0.29611	0.734598	1155.82	1019.00	18.34	18.35	−0.01
0.40037	0.747960	1152.37	1006.79	20.55	20.52	0.03
0.49752	0.760684	1150.34	993.44	20.84	20.84	0.00
0.59374	0.773555	1149.40	978.51	19.62	19.65	−0.03
0.69932	0.788000	1149.50	960.40	16.78	16.76	0.02
0.79396	0.801281	1150.68	942.55	12.80	12.81	−0.01
0.89444	0.815719	1153.04	922.09	7.28	7.28	−0.00
0.94383	0.822945	1154.63	911.47	4.08	4.06	0.02
1.00000	0.831268	1156.84	898.90			

Coefficients of the smoothed equations of excess properties A_i are also listed in Table 5.

Figure 3 illustrates the excess isothermal compressibility, κ_T^E, for these mixtures. The curves are positive and parabolic, except for the 2,8-isomer mixture, the values of which are flattened in the diether-rich region and consequently the maximum shifts to the region of smaller diether mole fraction. In the large mole fraction region, a less compressible structure formed by dipole–dipole alignment is expected. Such behaviour is also found in the 2,5,8-trioxanonane + *n*-octane mixture [6].

The excess isentropic compressibilities of these mixtures are not greatly different from the excess isothermal compressibilities but are a little lower.

TABLE 4

Excess isothermal compressibilities for dioxanonane isomer + *n*-octane mixtures at 298.15 K

Mole fraction diether x	κ_T (T Pa^{-1})	κ_T^E(obs.) (T Pa^{-1})	κ_T^E(calc) (T Pa^{-1})	Diff. (T Pa^{-1})
x 2,5-dioxanonane + $(1-x)$ *n*-octane				
0.00000	1294.2			
0.09997	1278.2	7.3	7.3	−0.0
0.19428	1261.0	12.1	12.1	−0.0
0.29799	1239.8	15.4	15.4	0.0
0.39333	1218.5	16.9	16.8	0.1
0.49421	1194.2	16.7	16.9	−0.2
0.59577	1168.5	15.7	15.6	0.1
0.69285	1142.5	13.3	13.3	−0.0
0.79532	1113.8	9.8	9.9	−0.1
0.89707	1084.3	5.5	5.5	0.0
0.93972	1071.4	3.3	3.3	−0.0
1.00000	1053.0			
x 2,6-dioxanonane + $(1-x)$ *n*-octane				
0.00000	1294.2			
0.09939	1279.8	7.2	7.4	−0.2
0.19567	1264.0	12.5	12.4	0.1
0.30109	1244.2	16.0	16.0	0.0
0.39755	1224.4	17.6	17.5	0.1
0.49723	1202.2	17.7	17.7	0.0
0.59479	1179.0	16.5	16.5	−0.0
0.69758	1153.2	14.0	14.1	−0.1
0.79727	1126.9	10.5	10.5	−0.0
0.90132	1098.2	5.8	5.7	0.1
1.00000	1069.5			
x 2,7-dioxanonane + $(1-x)$ *n*-octane				
0.00000	1294.0			
0.10036	1276.4	7.3	7.4	−0.1
0.19696	1257.3	12.4	12.4	−0.0
0.29991	1234.7	15.8	15.7	0.1
0.39589	1211.8	17.2	17.2	0.0
0.49697	1185.9	17.3	17.2	0.1
0.59628	1158.7	15.7	15.9	−0.2
0.69560	1130.7	13.6	13.5	0.1
0.79650	1100.6	10.0	10.0	−0.0
0.89822	1069.3	5.6	5.5	0.1
0.94522	1054.2	3.0	3.1	−0.1
1.00000	1036.6			

TABLE 4 (continued)

Mole fraction diether x	κ_T (T Pa^{-1})	κ_T^E(obs.) (T Pa^{-1})	κ_T^E(calc) (T Pa^{-1})	Diff. (T Pa^{-1})
x 2,8-dioxanonane + $(1-x)$ n-octane				
0.00000	1294.0			
0.10115	1264.1	4.3	4.5	−0.2
0.19590	1234.7	7.2	7.0	0.2
0.29954	1200.2	8.4	8.3	0.1
0.39445	1167.1	8.4	8.5	−0.1
0.49353	1131.7	7.8	7.9	−0.1
0.59272	1095.3	6.8	6.7	0.1
0.69369	1057.5	5.2	5.2	0.0
0.79309	1019.7	3.5	3.5	0.0
0.89444	980.6	1.7	1.7	−0.0
1.00000	939.7			
x 3,6-dioxanonane + $(1-x)$ n-octane				
0.00000	1294.2			
0.09882	1283.6	8.7	8.9	−0.2
0.19654	1270.9	15.3	15.2	0.1
0.30052	1254.8	19.7	19.7	0.0
0.39483	1238.0	21.8	21.8	0.0
0.49437	1218.5	22.2	22.2	0.0
0.59060	1197.9	20.9	21.0	−0.1
0.69024	1174.9	18.1	18.1	−0.0
0.78690	1151.0	13.9	14.0	−0.1
0.88373	1125.7	8.5	8.4	0.1
0.94746	1108.2	4.1	4.0	0.1
1.00000	1093.2			
x 3,7-dioxanonane + $(1-x)$ n-octane				
0.00000	1294.0			
0.09949	1284.8	9.0	9.1	−0.1
0.19616	1273.7	15.5	15.5	−0.0
0.29611	1259.9	20.0	20.0	0.0
0.40037	1243.1	22.5	22.5	0.0
0.49752	1225.4	23.0	22.9	0.1
0.59374	1206.2	21.7	21.8	−0.1
0.69932	1183.2	18.6	18.7	−0.1
0.79396	1161.0	14.3	14.4	−0.1
0.89444	1135.9	8.3	8.2	0.1
0.94383	1122.9	4.7	4.6	0.1
1.00000	1107.4			

In conclusion, the excess volumes of the mixtures show very simple parabolic behaviour with no anomalous features. The excess compressibility of the 2,8-dioxanonane mixture shows a complicated dependence on concentration. This may be due to a favoured dipole–dipole rearrangement occuring in the various configurations of the long central alkyl chain

TABLE 5

Coefficients of eqn. (2) for excess volume, and excess isentropic and isothermal compressibilities of x m, n-dioxanonane isomer + $(1-x)$ n-octane mixtures at 298.15 K, with standard deviation s

m,n-	X^E	A_1	A_2	A_3	A_4	s
2,5-	V^E	2.3656	0.2884	0.1198	0.0575	0.0014
	κ_S^E	58.49	14.48	3.30	–	0.06
	κ_T^E	67.3	13.9	4.4	–	0.08
2,6-	V^E	2.2435	0.1911	0.1668	0.0754	0.0025
	κ_S^E	63.17	12.16	2.61	–	0.07
	κ_T^E	70.7	11.6	3.5	–	0.10
2,7-	V^E	2.3255	0.1444	0.1307	0.1914	0.0020
	κ_S^E	61.94	15.14	3.33	–	0.09
	κ_T^E	68.8	13.9	3.5	–	0.12
2,8-	V^E	2.0397	0.2671	0.1258	0.1491	0.0027
	κ_S^E	26.00	19.35	3.59	–	0.12
	κ_T^E	31.3	19.4	4.0	–	0.12
3,6-	V^E	2.6549	0.2221	0.1093	0.0553	0.0015
	κ_S^E	78.19	11.97	2.32	–	0.04
	κ_T^E	88.6	11.1	3.0	–	0.08
3,7-	V^E	2.5309	0.1497	0.1931	0.1323	0.0015
	κ_S^E	83.30	10.30	3.14	–	0.02
	κ_T^E	91.7	8.8	3.7	–	0.07

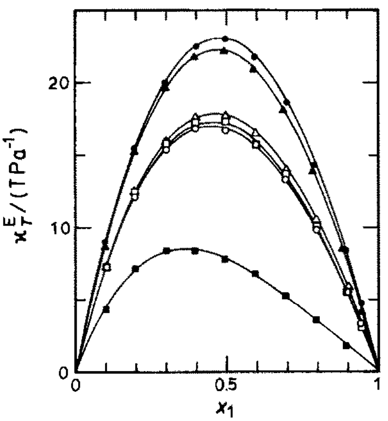

Fig. 3. Excess isothermal compressibilities of x dioxanonane isomer + $(1-x)$ n-octane: ○, 2,5-dioxanonane + n-octane; ●, 2,6-dioxanonane + n-octane; □, 2,7-dioxanonane + n-octane; ■, 2,8-dioxanonane + n-octane; ▲, 3,6-dioxanonane + n-octane; △, 3,7-dioxanonane + n-octane.

between the two oxygen atoms of the diether molecule induced by mixing with *n*-octane.

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