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Isothermal compressibility and sound velocity of binary liquid systems: Application of hard sphere models

J D PANDEY, P JAIN and V VYAS
Department of Chemistry, University of Allahabad, Allahabad 211 002, India

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Abstract. Sound velocity and density were measured in six binary liquid mixtures namely, *n*-heptane + toluene (I); *n*-heptane + *n*-hexane (II); toluene + *n*-hexane (III); cyclohexane + *n*-heptane (IV); cyclohexane + *n*-hexane (V), and *n*-decane + *n*-hexane (VI) at 298·15 K. The experimental isothermal compressibility has been evaluated from measured values of density and sound velocity. The isothermal compressibility of these mixtures has been calculated theoretically using different models for the hard sphere equation of state and also using Flory's statistical theory. The computed values of isothermal compressibility were also compared with the experimentally evaluated values. A satisfactory agreement has been observed.

Keywords. Isothermal compressibility; sound velocity; density; binary liquid systems; hard sphere equation of state.

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1. Introduction

The prime object of sound velocity and density measurements in liquid systems is to estimate the value of isentropic compressibility (β_s) which cannot be done by any other method. Isentropic compressibility has been widely used to study the molecular interactions through its excess value. On the other hand, it can also be used to deduce other useful thermodynamic properties e.g. isothermal compressibility, (β_T), heat capacity ratio (γ), internal pressure (P_i), cohesive energy density, (CED) etc. Latter parameters are only accessible from β_s provided the values of thermal expansion coefficient, (α) and heat capacity at constant pressure (C_p) are known. Isothermal compressibility and heat capacities ratio are the two key parameters in molecular thermodynamics of fluid phase equilibria. Various hard sphere equations of state have been applied to evaluate the values of β_T, γ, and sound velocity (c) of pure liquids under varying physical conditions [1–5]. Such models were also applied to a few binary liquid mixtures [6–7] for estimating β_T. The theoretical values of β_T compared with the experimental values which were obtained from β_s, α, and C_p data. Experimental measurement of β_T for some binary mixtures have been reported earlier [8–9]. In the present work, the results of experimental measurements of sound velocity (c) in and density (ρ) of six binary liquid mixtures, namely—*n*-heptane + toluene (I); *n*-heptane + *n*-hexane (II); toluene + *n*-hexane (III); cyclohexane + *n*-heptane (IV); cyclohexane + *n*-hexane (V) and *n*-decane + *n*-hexane (VI) are reported at 298·15 K. Further, density and sound velocity data were employed to evaluate β_T with the help of empirical equation derived. β_T values were also estimated with the help of various important hard sphere models. Another approach i.e. Flory's statistical theory [10–11] is also used to compute β_T values of binary liquid systems under the present investigation. This method has also been used earlier by a few workers [12–15]. The set of β_T values obtained are compared with the experimental β_T value deduced from the derived equation. With the help of sound velocity and density data, the values of γ and β_s for all the systems are also computed. This is entirely a new approach for estimating isothermal compressibility of liquid mixtures.

2. Experimental

All the organic liquids used were of analytic reagent grade and were obtained from BDH Chemicals Ltd., England. These chemicals were further purified by double distillation. Densities and speeds of sound were measured at 298·15 K. Densities were determined with a precalibrated bicapillary pyknometer with an accuracy of ± 0·3 kg/m^3 (approx. 0·05%). Speed of sound measurements were made with a single crystal variable path ultrasonic interferometer at 2 MHz frequency and the data were accurate to ± 0·01 per cent.

The purity of the samples were checked by comparing the measured density of the compounds with those reported in literature [16]. The measured densities and speeds of sound along with the literature values are presented in table 1.

3. Theoretical

The isothermal compressibilities of six binary liquid mixtures were evaluated using the following rigid sphere equations of state:

$$\frac{PV}{NkT}=\frac{1+y+y^2}{(1-y)^3} \quad [17] \tag{1}$$

$$\frac{PV}{NkT}=\frac{1+2y+3y^2}{(1-y)^2} \quad [18\text{–}19] \tag{2}$$

$$\frac{PV}{NkT}=\frac{1+y+y^2-y^3}{(1-y)^3} \quad [20] \tag{3}$$

Table 1. Density (ρ) and sound velocity (c) of pure component liquids at 298·15 K.

Component	$\rho \times 10^{-3}$(kg m^{-3}) Present study	Lit.	c(ms^{-1}) Present study	Lit.
Toluene	0·8626	0·8625	1304·3	1304·0
Cyclohexane	0·7736	0·7738	1252·0	1253·3
n-Hexane	0·6548	0·6550	1075·8	1076·0
n-Heptane	0·6790	0·6795	1131·0	1131·0
n-Decane	0·7260	0·7263	1125·0	1225·2

$$\frac{PV}{NkT}=\frac{1}{(1-y)^4} \qquad [21] \tag{4}$$

$$\frac{PV}{NkT}=\frac{1}{(1-y)^2} \qquad [22] \tag{5}$$

$$\frac{PV}{NkT}=\frac{1+y^2/8}{(1-y)^2} \qquad [23] \tag{6}$$

$$\frac{PV}{NkT}=1+4y+10y^2+18{\cdot}36y^3+28{\cdot}2y^4+39{\cdot}5y^4+\ldots \quad [24] \tag{7}$$

where, P V, T, N and k are respectively the pressure, volume, absolute temperature, Avogadro's number and Boltzmann constant. y is the packing fraction equal to $\pi a^3 N/6V$ in all cases, a being the hard sphere diameter of the molecule. The expressions for the isothermal compressibility, β_T, corresponding to eqs (1) to (7) are obtained as:

$$\beta_T=\frac{V}{RT}\frac{(1-y)^4}{(1+2y)^2} \tag{8}$$

$$\beta_T=\frac{V}{RT}\frac{(1-y)^3}{1+5y+9y^2-3y^3} \tag{9}$$

$$\beta_T=\frac{V}{RT}\frac{(1-y)^4}{1+4y+4y^2-4y^3+y^4} \tag{10}$$

$$\beta_T=\frac{V}{RT}\frac{(1-y)^5}{1+3y} \tag{11}$$

$$\beta_T=\frac{V}{RT}\frac{(1-y)^3}{1+y} \tag{12}$$

$$\beta_T=\frac{V}{RT}\frac{8(1-y)^3}{8+8y+3y^2-y^3} \tag{13}$$

$$\beta_T=\frac{V}{RT}(1+8y+30y^2+73{\cdot}44y^3+141{\cdot}0y^4+273{\cdot}0y^5)^{-1} \tag{14}$$

Flory's theory yields the following expression [10–11] for the isothermal compressibility:

$$\beta_T=\frac{\alpha_T\tilde{V}^2}{P^*} \tag{15}$$

where, $\tilde{V}$ and P^* are respectively the reduced volume and characteristic pressure of the liquid mixture. Values of $\tilde{V}$ and P^* were evaluated by the procedure detailed out by Flory and used by other workers [12–15].

In 1966, Mc-Gowan [25] suggested the following relationship between isothermal compressibility and surface tension (σ):

$$\beta_T\sigma^{3/2}=1{\cdot}33\times 10^{-8} \quad \text{(cgs units)} \tag{16}$$

The Auerbach relationship [26] between speed of sound (c) and surface tension (σ),

$$c = \left(\frac{\sigma}{6{\cdot}4 \times 10^{-4}\rho}\right)^{2/3} \tag{17}$$

has been employed by a number of workers during recent years [12–15, 27] in the case of pure liquids, solutions and non-electrolytes, molten salts and liquid metals. From (14) and (15), one gets,

$$\beta_T = \frac{1{\cdot}33 \times 10^{-8}}{(6{\cdot}3 \times 10^{-4} c^{3/2} \cdot \rho)^{3/2}} \tag{18}$$

This expression gives a direct relationship between isothermal compressibility, speed of sound and density. This relation has been tested for a variety of liquids under varying physical conditions.

4. Results and discussion

The experimental values of sound velocity (c) and density (ρ) of all the pure component liquids of various binary systems at 298·15 K have been determined and recorded in table 1 along with the literature values. The agreement between the two set of values is good. Density and sound velocity of six binary liquid mixtures (I–VI) at 298·15 K have been measured as a function of composition, and the values are recorded in columns two and three of tables 2–7. Sound velocity and density data, obtained for all the six binaries, have been utilized to estimate the experimental values of isothermal compressibilities (β_T) from (18). These values are reported in column four of each table (tables 2–7) for the respective systems mentioned. Two sets of data are generated for computing the theoretical values of β_T for all the binary mixtures. In one set various hard sphere models, vide eqs (1)–(7), have been used. The corresponding values of compressibility have been computed from eqs (8)–(14). As (12) and (13) corresponding to models (5) due to Frisch [22] and (6) due to Henderson [23] did not yield satisfactory values, we have not reported β_T values obtained from these equations. Theoretical values of β_T computed from (8)–(11) and (14) for various binary liquid mixtures under the present investigation are presented in columns five, six, seven, eight and nine respectively of each table (tables 2–7). A second set of theoretical β_T values are computed from Flory's statistical theory using (15). These values are recorded in the last column of each table (tables 2–7). The experimental values of isothermal compressibility of all the six binary liquid mixtures were estimated from sound velocity and density data in conjunction with (18). The values of β_T, thus obtained are plotted against mole fraction of one component in the mixture. These plots are shown in figure 1. In the case of system (I), the experimental values of β_T increased continuously (table 2) on increasing the mole fraction of the first component n-heptane. A similar trend is also obtained theoretically, as is evident from the results of columns five to nine of table 2. The values of β_T computed from Flory theory for this system also show a similar trend (last column of the table 2). It has been observed that in the case of system I i.e. n-heptane and toluene the value of β_T increases continuously with the increasing molar fraction of component 1 while in the other systems it decreases. The probable reason for this is that in this system the two components have equal number of carbon atoms i.e. one is the cyclic analogue of

Table 2. Density, sound velocity and isothermal compressibility of binary system – *n*-heptane (x_1) + toluene (x_2) (I) at 298·15 K.

x_1	$\rho \times 10^{-3}$ (kg m^{-3})	c (ms^{-1})	$\beta_T \times 10^{12}$ (m^{2}N^{-1}) (Exptl.)	$\beta_T \times 10^{12}$ (Theoretical) (m^{2}N^{-1})					
				Eq. (8)	Eq. (9)	Eq. (10)	Eq. (11)	Eq. (14)	Eq. (15)
0·2979	0·8693	1247·3	112·30	102·17	142·05	112·77	85·42	147·23	167·04
0·3162	0·8667	1243·0	113·62	103·06	143·19	113·68	86·21	148·33	170·04
0·3325	0·8637	1240·5	114·73	103·80	144·17	114·49	86·85	149·29	172·04
0·3519	0·8601	1237·2	116·15	104·53	145·15	115·28	87·47	150·26	173·74
0·3707	0·8564	1233·7	117·65	105·78	146·80	116·63	88·58	151·81	175·50
0·3902	0·8532	1230·0	119·19	106·76	147·99	117·69	89·43	153·03	177·40
0·4029	0·8500	1228·0	120·23	107·53	149·01	118·53	90·10	154·03	179·45
0·4282	0·8468	1224·0	121·80	108·51	148·98	119·60	90·96	155·29	181·26
0·4484	0·8433	1220·0	123·47	109·62	151·72	120·79	91·93	156·67	183·92
0·4684	0·8394	1216·0	125·25	110·82	153·25	122·09	92·98	158·16	185·32
0·4874	0·8363	1213·8	126·46	111·58	154·26	122·92	93·62	159·17	187·47
0·5023	0·8225	1211·0	130·33	114·60	158·19	126·19	96·25	163·04	182·06
0·5227	0·8188	1206·5	130·32	115·93	162·74	127·63	97·43	164·67	183·73
0·5442	0·8153	1203·7	133·87	116·90	161·17	128·68	98·27	165·94	189·81
0·5646	0·8122	1199·0	135·83	118·17	162·78	130·05	99·37	167·48	187·61

Table 3. Density, sound velocity and isothermal compressibility of binary system – *n*-heptane (x_1) + *n*-hexane (x_2) (II) at 298·15 K.

x_1	$\rho \times 10^{-3}$ (kg m^{-3})	c (ms^{-2})	$\beta_T \times 10^{12}$ (m^{2}N^{-1}) (Exptl.)	$\beta_T \times 10^{12}$ (Theor.) (m^{2}N^{-1})					
				Eq. (8)	Eq. (9)	Eq. (10)	Eq. (11)	Eq. (14)	Eq. (15)
0·3388	0·6614	1091·1	228·53	168·31	223·88	183·50	144·64	224·74	198·04
0·3598	0·6628	1092·2	227·29	167·83	223·42	183·01	144·18	212·40	198·23
0·3782	0·6606	1093·9	227·63	168·47	224·31	183·73	144·72	225·37	196·65
0·4001	0·6639	1094·4	223·54	167·56	223·28	182·76	143·08	224·37	196·30
0·4193	0·6636	1095·1	225·53	167·81	223·69	183·05	144·07	224·83	196·51
0·4394	0·6646	1096·5	224·37	157·44	223·33	182·63	143·70	224·55	196·24
0·4604	0·6656	1097·0	223·64	167·31	223·25	182·56	143·55	225·54	193·15
0·4782	0·6663	1098·5	222·60	166·67	222·92	182·22	143·21	224·30	190·26
0·4986	0·6672	1099·6	221·65	166·72	222·70	181·96	142·94	224·15	195·35
0·5191	0·6681	1101·8	220·21	165·53	221·40	180·74	141·79	223·10	195·29
0·5395	0·6693	1102·8	219·17	164·43	220·25	180·13	140·74	222·10	195·11
0·5594	0·6729	1104·0	216·89	164·60	220·36	179·76	140·93	222·12	195·85
0·5795	0·6716	1104·9	217·12	165·15	221·15	180·37	141·38	222·96	195·59
0·6025	0·6730	1105·6	216·13	164·87	220·89	180·10	141·10	222·77	194·42
0·6185	0·6736	1106·0	215·67	164·74	220·77	180·00	140·97	222·69	194·51

Table 4. Density, sound velocity and isothermal compressibility of binary system – toluene (x_1) + *n*-hexane (x_2) (III) at 298·15 K.

x_1	$\rho \times 10^{-3}$ ($\mathrm{kg\,m^{-3}}$)	c ($\mathrm{ms^{-1}}$)	$\beta_T \times 10^{12}$ ($\mathrm{m^2\,N^{-1}}$) (Exptl.)	$\beta_T \times 10^{12}$ (Theoretical) ($\mathrm{m^2\,N^{-1}}$)					
				Eq. (8)	Eq. (9)	Eq. (10)	Eq. (11)	Eq. (14)	Eq. (15)
0·4074	0·7936	1149·7	154·57	124·30	167·96	136·09	105·82	170·39	198·55
0·4299	0·7984	1153·3	152·10	123·09	166·48	134·81	104·73	168·99	196·33
0·4486	0·8018	1159·5	149·32	121·61	164·72	133·23	103·37	167·38	192·03
0·4704	0·8058	1163·3	147·13	120·56	163·43	132·11	102·42	166·18	191·78
0·4919	0·8111	1167·0	144·65	119·26	161·83	130·72	101·26	164·66	188·65
0·5114	0·8157	1172·0	142·05	117·83	160·09	129·20	99·96	163·04	188·32
0·5290	0·8188	1176·0	140·17	116·87	158·93	128·18	99·07	161·97	186·21
0·5491	0·8286	1181·0	137·63	115·47	157·21	126·66	97·81	160·36	180·27
0·5584	0·8278	1183·0	136·06	114·47	155·98	125·61	96·94	157·19	183·52
0·5880	0·8323	1188·0	133·68	113·37	154·64	124·44	95·94	157·94	176·05
0·6088	0·8368	1194·0	131·11	111·90	152·88	122·88	94·61	156·31	173·94
0·6275	0·8400	1199·1	129·12	110·81	151·57	121·72	93·62	155·41	175·31
0·6453	0·8512	1203·6	125·51	108·36	148·47	119·08	91·45	152·14	176·87
0·6637	0·8527	1208·2	124·12	107·70	147·70	118·39	90·84	151·46	174·23
0·6829	0·8537	1213·1	122·78	107·11	147·02	117·76	90·24	150·88	170·64

Table 5. Density, sound velocity and isothermal compressibility of binary system – cyclohexane (x_1) + *n*-heptane (x_2) (IV) at 298·15 K.

x_1	$\rho \times 10^{-3}$ (kg m^{-3})	c (ms^{-1})	$\beta_T \times 10^{12}$ (m^{2}N^{-1}) (Exptl.)	$\beta_T \times 10^{12}$ (Theor.) (m^2 N^{-1}) Eq. (8)	Eq. (9)	Eq. (10)	Eq. (11)	Eq. (14)	Eq. (15)
0·2227	0·7184	1156·0	177·27	144·22	196·11	158·17	122·30	199·83	193·14
0·2689	0·7214	1162·1	174·09	141·20	193·19	155·74	120·35	196·94	185·80
0·3158	0·7236	1167·8	171·40	140·10	190·70	153·69	118·72	194·47	182·34
0·3588	0·7191	1171·8	171·68	140·45	191·06	154·06	119·07	194·76	177·73
0·4016	0·7243	1177·2	168·09	139·44	189·46	152·90	118·30	192·95	176·65
0·4433	0·7307	1182·0	164·38	135·11	184·05	148·27	114·14	187·80	175·68
0·4861	0·7334	1187·9	161·71	133·20	181·55	146·18	112·79	185·31	172·90
0·5270	0·7361	1191·9	159·55	131·69	179·50	144·52	111·49	183·24	171·26
0·5642	0·7384	1196·0	157·58	130·22	177·55	142·92	110·24	181·28	169·50
0·6019	0·7412	1201·2	155·17	128·48	175·26	141·02	108·73	179·01	167·36
0·6414	9·7438	1205·8	153·03	126·93	173·20	139·68	107·40	176·94	164·55
0·6795	0·7463	1210·6	150·91	125·39	171·15	137·66	106·07	174·89	162·27
0·7164	0·7482	1215·9	148·86	123·92	169·22	136·06	104·80	172·97	160·89
0·7521	0·7518	1220·0	146·68	122·30	167·06	134·29	103·41	170·81	159·01
0·7877	0·7547	1224·0	144·77	120·89	165·17	132·75	102·20	168·90	158·48

Table 6. Density, sound velocity and isothermal compressibility of binary system – cyclohexane (x_1) + n-hexane (x_2) (V) at 298·15 K.

x_1	$\rho \times 10^{-3}$ (kg m^{-3})	c (ms^{-1})	$\beta_T \times 10^{12}$ (m^{2}N^{-1}) (Exptl.)	$\beta_T \times 10^{12}$ (Theor.) (m^{2}N^{-1})					
				Eq. (8)	Eq. (9)	Eq. (10)	Eq. (11)	Eq. (14)	Eq. (15)
0·2189	0·6821	1110·0	209·98	154·04	204·90	168·11	132·42	205·65	195·63
0·2541	0·6860	1118·8	204·48	151·21	201·53	164·94	129·83	202·53	193·67
0·2981	0·6914	1134·9	195·70	146·41	195·87	159·87	125·41	197·35	191·33
0·3150	0·6932	1135·0	194·90	146·11	195·44	156·97	125·17	196·89	190·33
0·3647	0·6990	1142·3	189·72	143·46	192·18	156·71	123·86	193·79	186·84
0·3897	0·7022	1149·8	185·67	141·23	189·50	154·33	120·74	191·32	186·13
0·4258	0·7065	1155·4	181·98	139·31	187·15	152·29	119·03	189·08	184·02
0·4777	0·7129	1163·2	176·84	136·57	183·36	149·36	116·57	185·86	180·98
0·5029	0·7157	1170·2	173·51	134·72	181·54	147·39	114·88	183·80	178·93
0·5675	0·7232	1180·3	167·48	131·50	177·55	143·94	112·80	180·03	175·28
0·5799	0·7250	1182·7	166·09	130·71	176·58	143·10	111·28	179·11	173·33
0·6012	0·7275	1188·9	163·31	129·10	174·66	141·40	109·82	177·33	173·20
0·6695	0·7356	1200·0	157·29	125·88	170·65	137·94	106·94	173·52	168·71
0·7187	0·7415	1210·6	152·38	122·97	167·10	134·84	104·32	170·20	165·45
0·7888	0·7499	1221·8	146·75	119·80	163·16	131·44	101·49	166·20	160·74

Table 7. Density, sound velocity and isothermal compressibility of binary system – *n*-decane (x_1) + *n*-hexane (x_2) (VI) at 298·15 K.

x_1	$\rho \times 10^{-3}$ (kg m^{-3})	c (ms^{-1})	$\beta_T \times 10^{12}$ (m^{2}N^{-1}) (Exptl.)	$\beta_T \times 10^{12}$ (Theoretical) (m^{2}N^{-1})					
				Eq. (8)	Eq. (9)	Eq. (10)	Eq. (11)	Eq. (14)	Eq. (15)
0·0187	0·6663	1075·5	233·46	165·85	219·42	180·54	143·05	219·48	208·11
0·0588	0·6697	1080·1	229·47	166·02	220·33	180·88	142·91	220·83	205·15
0·1008	0·6730	1087·9	224·13	165·40	220·40	180·41	142·03	221·03	202·13
0·1436	0·6774	1094·6	218·90	164·72	220·33	179·86	141·12	221·97	199·63
0·1896	0·6777	1100·0	216·35	165·94	222·66	181·34	141·88	224·79	196·40
0·2348	0·6850	1108·4	209·28	163·84	220·86	179·36	139·68	223·67	194·15
0·2886	0·6883	1118·8	203·46	163·00	220·84	178·59	138·54	224·44	190·78
0·3350	0·6917	1127·5	198·47	162·12	220·58	177·83	137·42	224·87	188·26
0·3872	0·6943	1135·4	194·28	161·99	221·30	177·88	136·96	226·29	185·30
0·4449	0·6987	1142·8	189·65	161·63	221·76	177·69	136·29	227·48	182·48
0·5009	0·7012	1152·3	185·16	161·18	222·15	177·41	135·51	228·68	178·16
0·5627	0·7071	1161·9	179·46	159·83	221·17	176·16	133·94	228·82	177·39
0·6248	0·7099	1173·9	174·33	158·80	222·50	175·37	132·62	229·56	174·41
0·6944	0·7114	1184·1	170·42	158·29	222·75	175·69	132·25	231·85	170·71
0·7655	0·7144	1195·6	165·71	157·70	220·08	175·19	131·24	233·16	167·93

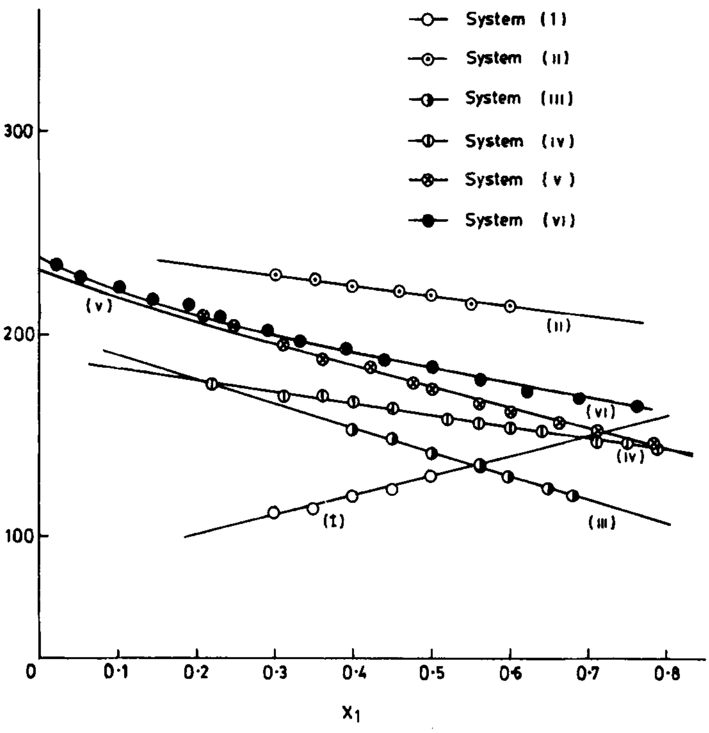

Figure 1. Isothermal compressibility β_T vs mole fraction (X_1) of component 1 for the systems (I) to (VI).

the other with aromatic characters, where π electrons play an important role in the molar interaction as is evident from their structures:

When the experimental and theoretically computed values of β_T for all other systems (II–IV) are examined (tables 3–7), it is observed that in each case β_T decreases on increasing the mole fraction of the first component. A similar trend in the variation of β_T has also been shown from Flory's theory (last column of each table).

Table 7 lists the average percentage deviations obtained in compressibility values for all the systems under the present study. From this table it is observed that (10) due to Carnahan and Starling [20] provides good agreement with the experimental values for all the systems, the observed deviations being not more than 18·12 per cent. Equations (9) due to Werthein and Thiel [18] and (14) due to Hoover and Ree [24] give better agreement with the experimental values specially for the system, n-heptane + n-hexane, the observed deviations being not more than 0·17 per cent for (9) and 0·48 per cent for Eq. (14). Carnahan and Starling equation (10) gives good agreement in the case of the system n-heptane + toluene, deviations being not more than 1·85 per cent. In the case of system IV (cyclohexane + n-heptane) a similar agreement from all the equations has been obtained. But in the case of system V (cyclohexane + n-hexane) eqs (9), (14) and (15) yield better agreement.

The present study demonstrates that Flory's statistical theory, vide (15), yields good agreement for almost all the systems except for I and III. In the hard sphere models attractive and repulsive forces are taken into account that give better agreement with

Table 8. Average percentage deviation of isothermal compressibility β_T, at 298·15 K with experimental values.

System	Eq. (8)	Eq. (9)	Eq. (10)	Eq. (11)	Eq. (14)	Eq. (15)
n-Heptane + toluene	10·9	−23·36	1·85	25·32	−27·33	−46·44
n-Heptane + *n*-hexane	24·99	−0·17	18·12	35·70	−0·48	−11·96
Toluene + *n*-hexane	16·29	−13·95	8·17	29·07	−16·15	−32·98
Cyclohexane + *n*-heptane	17·52	−12·43	9·45	30·16	−14·76	−7·17
Cyclohexane + *n*-hexane	22·97	−3·56	15·88	34·19	−4·75	−1·44
n-Decane + *n*-hexane	18·58	−10·85	10·63	30·97	−13·19	5·69

the observed β_T values. The superiority of Carnathan and Starling's hard sphere model has also been demonstrated earlier by other workers [28–29].

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