Compressibility of Liquids. Rule of Noncrossing V−P Curvatures

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Weight analysis of the liquid introduced into the stainless steel bomb under pressures up to 1 kbar in the temperature range of 20−50 °C at the interval of 10 °C was performed for 1,4-dioxane, acetonitrile, toluene, ethyl acetate, chlorobenzene, and *n*-hexane. The coefficients of the Tait equation were determined for all of the solvents at each temperature. There was a clear linear relation between the tangent bulk modulus ($1/\beta_T$) at atmospheric pressure (1 bar) and the secant bulk modulus at 1 kbar; $1/\beta_T = 0.9865 \times (1000\ V_0/\Delta V_{1kbar}) - 4559$ was found at a wide range of temperatures for different liquids, including glycerol and even mercury. This represents the rule of noncrossing P−V curvatures for the liquids. Using the correlation, it is possible to predict the coefficients (*C*, *B*) of the Tait equation from the experimental data of β_T at 1 bar or, on the other hand, to calculate the value of β_T from experimental compression at high pressures.

Introduction

The compressibility of liquid data reflecting the balance of the intermolecular attraction and repulsion energies are now available from direct experimental measurement only. This property of liquid substantially determines the changes in the partial molar volume of the compound. The solvent electrostriction in electrolyte solution also changes such parameters as the dielectric permittivity, refraction index, and viscosity under elevated pressures. The compressibility allows one to make a more accurate prediction of the effect of acceleration of the reaction under conditions of pressure and changes of the activation and reaction volumes.[1–4] It has been established[3] that there is an absence of correlation between the values of β_T and a set of other physical characteristics of liquids, such as the thermal coefficient of the expansion (α_p), molar volume (*V*), enthalpy ($\Delta_{vap}H$), and free energy ($\Delta_{vap}G$) of vaporization. Reliable correlations have not been noted with additional consideration for the voids at the packing of molecules in the liquid ($V - V_W$)/*V* and taking into account the van der Waals volume (V_W) as well as some other parameters.[5]

There is a lot of data on the compressibility of liquids at ambient pressure.[1,3,6–14] Reliable compressibility data of organic solvents at elevated temperatures and pressures is scanty. Comparative analysis for many liquids was performed by Cibulka et al.[7–14] There are various well-known methods for measuring the volume of the liquids under elevated pressure, which is conditionally divided into three groups, (1) methods using piezometers of constant capacity. (2) methods using piezometers of variable capacity, and (3) methods for hydrostatic weighing under pressure.[1,15,16] The dependency of sound velocity from the change of density of the pressurized liquid underlies the basis for reliable calculation of the adiabatic compressibility of liquids. However, using these data for calculating the isothermal compressibility at selected values of pressure (*P*) and temperature (*T*) requires
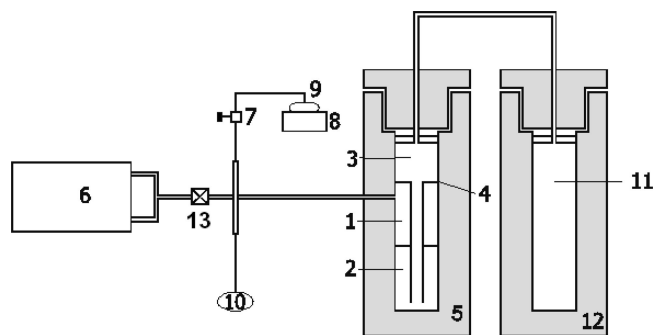

Figure 1. Device for determination of the liquid compressibility.

knowledge of the ratio of heat capacities (C_p/C_S) at these conditions. All of these methods require the account of volumetric deformation of the vessel under pressure, which is faced with difficulties. The method of hydrostatic weighing in liquids at a high pressure does not depend on problems of the volumetric deformation of the pressurized vessel but requires account of the sinker deformation.[16]

Experimental Section

Our proposed method to determination the liquid compressibility is based on the weighting of compressed liquid. The device (Figure 1) contains a thick-walled cylinder 5 made from stainless steel with welded siphon 4. In the bottom reservoir of cylinder 5 is located mercury seal 2, separating the hydraulic liquid (HL) in volume 1 and the liquid under investigation in volume 3.

The volume 1 of cylinder 5 is connected by a system of capillary tubes made from stainless steel, with a manometer 10 and with a compressor 6, through a high-pressure stopcock 13. The hydraulic liquid from reservoir 1 with controlled velocity by means of the valve of fine regulation 7 can be directed in receiver 9, located on the analytical balance 8. The long-term precision of the measurements is achieved when volume of the liquid under consideration is not less than 1/2 of the total

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TABLE 1: Experimental Data for the Determination of Toluene Compressibility under Excess Pressure up to 1 kbar at 20°C

P, bar	m_1, g	m_2, g	m_3, g	Δm_{1-2}, g	$V_{p,\text{Water}}$, cm^3	$d_{p,\text{HL}}$, g cm^{-3}	Δm_{1-3}, g	$d_{p,\text{toluene}}$, g cm^{-3}	$\Delta V/V_0$ toluene
0	0	0	0	0	82.523028	0.8831	0	0.8668	0
50	0.283	0.232	0.381	0.051	82.335287	0.8858	0.097	0.8705	0.00431
100	0.561	0.463	0.749	0.097	82.150380	0.8883	0.188	0.8742	0.00842
150	0.831	0.691	1.105	0.139	81.968220	0.8908	0.275	0.8776	0.01235
200	1.094	0.917	1.449	0.176	81.788729	0.8932	0.356	0.8810	0.01612
250	1.351	1.141	1.783	0.209	81.611829	0.8956	0.432	0.8842	0.01973
300	1.602	1.363	2.106	0.239	81.437447	0.8979	0.504	0.8874	0.02320
350	1.848	1.583	2.419	0.265	81.265511	0.9001	0.572	0.8904	0.02652
400	2.088	1.799	2.724	0.288	81.095954	0.9022	0.636	0.8934	0.02972
450	2.323	2.014	3.019	0.309	80.928711	0.9044	0.697	0.8962	0.03281
500	2.553	2.226	3.307	0.326	80.763719	0.9064	0.754	0.8990	0.03579
550	2.778	2.436	3.587	0.342	80.600920	0.9085	0.810	0.9017	0.03867
600	2.998	2.642	3.861	0.355	80.440255	0.9104	0.862	0.9043	0.04147
650	3.213	2.846	4.127	0.367	80.281669	0.9124	0.913	0.9069	0.04419
700	3.425	3.048	4.387	0.377	80.125109	0.9143	0.962	0.9094	0.04684
750	3.633	3.246	4.643	0.387	79.970524	0.9162	1.010	0.9119	0.04943
800	3.836	3.441	4.893	0.395	79.817865	0.918	1.057	0.9143	0.05198
850	4.037	3.633	5.139	0.403	79.667084	0.9199	1.103	0.9167	0.05448
900	4.233	3.822	5.382	0.411	79.518136	0.9217	1.149	0.9191	0.05695
950	4.427	4.008	5.621	0.419	79.370977	0.9235	1.194	0.9215	0.05940
1000	4.618	4.191	5.858	0.427	79.225564	0.9253	1.240	0.9239	0.06183

volume. To provide, this effect the volume of the liquid under consideration can be increased (3 + 11, Figure 1). All values of the volumes of the liquids are defined at the filling.

In the first cycle of measurements, capacities 1, 3, and 11 are filled up by HL, for instance, vacuum oil. The same HL is kept in the reservoir of the compressor 6. The cylinders 5 and 12 are placed in a thermostat (±0.02 °C). The pressure up to 1000 bar is created by compressor 6. Then high-pressure stopcock 13 is closed, and the system is held for temperature equalizing for approximately 1 h until reaching constant readings of the manometer 10. Then pressure in the system is gradually decreased (about 2 bar/min) by the fine regulation valve 7, directing hydraulic liquid through the capillary tubing to receiver 9, located on the balance 8. It allows definition of the dependence of mass as a function of pressure ($m-P$) in the entire interval of the falling pressure. The appropriate velocity of the HL release ($\partial m/\partial t$) for temperature retention is checked by the constancy of the readings of manometer 10, when valve 7 is quickly closed. The sensitivity of the manometer 10 was ±0.1 bar or about ±0.01 °C on the temperature scale. Since

the volume of compression of all liquids in the system (Figure 1) equals the volume of expansion at the pressure release, it follows that

$$m_{1,p}/d_p = \sum \Delta v_{\text{HL/HL}} = \Delta v_{1,p} + \Delta v_{2,p} + \Delta v_{3,p} + \Delta v_{11,p} +$$
$$\Delta v_{\text{cap.},p} + \Delta v_{\text{def.},p} \quad (1)$$

where $m_{1,p}$ is the mass of HL escaped from the reservoir 1, determined by weighing, d_p is the density of HL at pressure (P) and temperature (T), $\Delta v_{1,p}$ and $\Delta v_{\text{cap.},p}$ are the volume changes at compression of HL in volume 1 and in the external capillary tubes (room temperature control 22 ± 1 °C), respectively, $\Delta v_{2,p}$ is the volume change at compression of mercury in volume 2, $\Delta v_{3,p}$ and $\Delta v_{11,p}$ are the volume changes at compression of HL in reservoirs 3 and 11, and $\Delta v_{\text{def.},p}$ is the volume of the system deformation under high pressure.

In the second cycle of measurements, the reservoirs 3 and 11 are filled with double-distilled water, and measurements are conducted similar to those for the first cycle. In this case, the
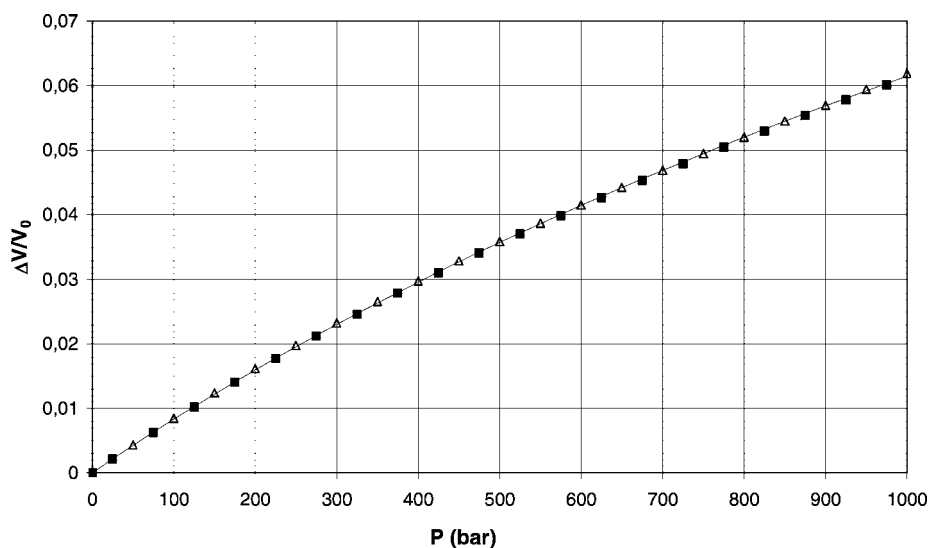

Figure 2. Compressibility of toluene at 20 °C. Solid line corresponds to precise data[11] of the compressibility of toluene, and the points (■) and (△) correspond to data of repeated measurements of this work.

TABLE 2: Tangent Bulk Modulus Values at Atmospheric Pressure ($1/\beta_0$, bar) and Secant Bulk Modulus Values at 1000 bar ($V_0/\Delta V$) of the Liquids and Coefficients (C,B) of the Tait Equation at 25°C until Other Temperature Indicated

N^0	liquid	experimental data					calcd.	
		C	B, bar	$1/\beta_0$, bar	$V_0/\Delta V$	ref.	$V_0/\Delta V$	error %[a]
1	diethyl ether	0.0951	489	5145	9.4483	9	9.7668	3.26
2	di-*n*-propyl ether	0.0860	488	5679	10.4349	9	10.3140	−1.17
3	diisopropyl ether	0.0888	485	5458	10.0574	9	10.0876	0.30
4	di-*n*-butyl ether	0.0906	750	8271	13.0170	9	12.9701	−0.36
5	methyl *tert*-butyl ether	0.0916	527	5750	10.2561	9	10.3868	1.26
6	ethyl *tert*-butyl ether	0.0865	485	5605	10.3289	9	10.2382	−0.89
7	methyl *tert*-pentyl ether	0.0872	641	7353	12.1996	9	12.0294	−1.41
8	ethylene glycol dimethyl ether	0.0926	837	9034	13.7330	9	13.7520	0.14
9	diethylene glycol dimethyl ether	0.0897	1098	12244	17.2230	9	17.0414	−1.07
10	tetraethylene glycol dimethyl ether	0.0862	1372	15918	21.1927	9	20.8062	−1.86
11	anisole	0.0959	1460	15228	19.9907	9	20.0991	0.54
12	tetrahydrofuran	0.0930	921	9906	14.6314	9	14.6456	0.10
13	1,4-dioxane (25)	0.0893	1192	13349	18.3827	b	18.1737	−1.15
14	1,4-dioxane (30)	0.0893	1153	12916	17.9367	b	17.7300	−1.17
15	1,4-dioxane (40)	0.0893	1080	12093	17.0846	b	16.8866	−1.17
16	1,4-dioxane (50)	0.0893	996	11152	16.1075	b	15.9224	−1.16
17	furan	0.0945	950	10053	14.7178	9	14.7962	0.53
18	2-propanone	0.0992	767	7732	12.0782	9	12.4178	2.73
19	2-butanone	0.0950	818	8614	13.1839	9	13.3216	1.03
20	2-pentanone	0.0874	858	9812	14.8025	9	14.5492	−1.74
21	3-pentanone	0.0946	908	9596	14.2328	9	14.3279	0.66
22	2-hexanone	0.0852	1017	11932	17.1327	9	16.7216	−2.46
23	4-methyl-2-pentanone	0.0991	1000	10089	14.5568	9	14.8331	1.86
24	4-heptanone	0.1024	1222	11933	16.3329	9	16.7227	2.33
25	cyclopentanone	0.0985	957	9721	14.1985	9	14.4560	1.78
26	cyclohexanone	0.0916	1388	15154	20.1239	9	20.0233	−0.50
27	butanal	0.0918	1041	11343	16.1843	9	16.1181	−0.41
28	3-methylbutanal	0.0941	832	8845	13.4668	9	13.5583	0.68
29	heptanal	0.1022	1130	11052	15.4293	9	15.8199	2.47
30	octanal	0.1027	1216	11838	16.2208	9	16.6253	2.43
31	benzaldehyde	0.0940	1541	16382	21.2577	9	21.2817	0.11
32	formic acid	0.0963	1494	15510	20.2593	9	20.3881	0.63
33	acetic acid	0.0924	1005	10879	15.6719	9	15.6426	−0.19
34	propionic acid	0.0913	987	10812	15.6545	9	15.5740	−0.52
35	isobutyric acid	0.0874	870	9961	14.9612	9	14.7019	−1.76
36	capric acid (80)	0.0939	959	10216	14.9120	9	14.9636	0.34
37	methyl acetate	0.0879	757	8615	13.5170	9	13.3226	−1.46
38	ethyl acetate	0.0894	743	8305	13.1120	9	13.0050	−0.82
39	ethyl acetate (20)	0.0891	792	8881	13.7357	9	13.5952	−1.03
40	ethyl acetate (30)	0.0891	726	8149	12.9590	b	12.8451	−0.89
41	ethyl acetate (40)	0.0891	666	7472	12.2369	b	12.1514	−0.70
42	ethyl acetate (50)	0.0891	601	6744	11.4527	b	11.4054	−0.41
43	triacetin	0.0988	1983	20064	24.7791	9	25.0547	1.10
44	isopentyl propionate	0.0854	900	10544	15.6778	9	15.2993	−2.47
45	methyl caproate	0.0914	694	7595	12.2628	9	12.2774	0.12
46	*n*-nonyl caproate	0.0939	813	8661	13.2819	9	13.3698	0.66
47	tricaproin	0.0956	1470	15379	20.1597	9	20.2539	0.46
48	2-ethylhexyl benzoate	0.0978	1382	14125	18.7735	9	18.9689	1.03
49	methyl methacrylate	0.0943	998	10582	15.2765	9	15.3383	0.40
50	*n*-nonyl methacrylate	0.0935	1200	12842	17.6533	9	17.6541	0.00
51	methyl oleate	0.0968	1492	15411	20.1370	9	20.2867	0.74
52	diethyl dibenzylmalonate	0.1038	2028	19537	24.0332	9	24.5147	1.96
53	di-*n*-butyl phthalate	0.0917	1585	17281	22.2908	9	22.2029	−0.40
54	propylene carbonate	0.0930	1816	19532	24.5183	9	24.5095	−0.04
55	methanol	0.0979	806	8231	12.6598	7	12.9291	2.08
56	ethanol	0.0950	849	8930	13.5159	7	13.6454	0.95
57	1-propanol	0.0921	943	10239	15.0209	7	14.9868	−0.23
58	1-butanol	0.0910	1005	11045	15.9127	7	15.8127	−0.63
59	1-pentanol	0.0952	1099	11545	16.2345	7	16.3251	0.55
60	1-heptanol	0.0989	2043	20659	25.3808	7	25.6644	1.11
61	1-octanol	0.0940	1264	13452	18.2591	7	18.2792	0.11
62	1-undecanol (40)	0.1002	1153	11517	15.9938	10	16.2969	1.86
63	1-dodecanol	0.0908	1294	14250	19.2325	10	19.0970	−0.71
64	1-tetradecanol (60)	0.0955	1155	12096	16.7900	10	16.8894	0.59
65	1-hexadecanol (75)	0.0908	1027	11318	16.2065	10	16.0925	−0.71
66	2-propanol	0.0890	792	8904	13.7656	10	13.6188	−1.08
67	2-butanol	0.0890	930	10448	15.3866	10	15.2010	−1.22
68	2-methyl-1-propanol	0.0866	853	9858	14.8954	10	14.5964	−2.05

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TABLE 2: Continued

N^0	liquid	experimental data					calcd.	
		C	B, bar	$1/\beta_0$, bar	$V_0/\Delta V$	ref.	$V_0/\Delta V$	error %[a]
69	2-methyl-2-propanol (60)	0.0834	498	5977	10.8962	10	10.6195	−2.61
70	2-pentanol	0.0995	1015	10199	14.6531	10	14.9458	1.96
71	3-pentanol	0.0952	1013	10644	15.3003	10	15.4018	0.66
72	4-methyl-2-pentanol	0.0827	858	10379	15.6559	10	15.1303	−3.47
73	2-octanol	0.0905	1084	11982	16.9110	10	16.7729	−0.82
74	3-octanol	0.0937	1155	12329	17.1141	10	17.1285	0.08
75	3-methyl-1-heptanol	0.0945	1244	13167	17.9409	10	17.9872	0.26
76	2-methyl-3-heptanol	0.0960	1154	12026	16.6966	10	16.8180	0.72
77	6-methyl-3-heptanol	0.0924	1083	11717	16.5415	10	16.5013	−0.24
78	3-methyl-4-heptanol	0.0907	1100	12127	17.0477	10	16.9215	−0.75
79	2,7-dimethyl-2-octanol	0.0832	1233	14823	20.2410	10	19.6841	−2.83
80	cyclopentanol	0.0862	1330	15434	20.6964	10	20.3102	−1.90
81	ethylene glycol	0.0950	2585	27206	32.1823	10	32.3733	0.59
82	1,2-propanediol	0.0984	2337	23766	28.5489	10	28.8482	1.04
83	1,3-propanediol	0.0964	2427	25184	30.0753	10	30.3013	0.75
84	glycerol	0.1143	4946	43286	47.5281	10	48.8508	2.71
85	2-methoxyethanol	0.0952	1338	14063	18.8313	10	18.9053	0.39
86	2-butoxyethanol	0.0913	1185	12981	17.9031	10	17.7966	−0.60
87	diethylene glicol	0.1008	2791	27703	32.4124	10	32.8826	1.43
88	eugenol	0.0856	1856	21689	27.1147	10	26.7199	−1.48
89	benzene (40)	0.0937	861	10368	15.0822	11	15.1190	0.24
90	hexadeuteriobenzene (40)	0.0930	878	9191	13.8504	11	13.9128	0.45
91	toluene	0.0937	1042	9443	14.1436	11	14.1713	0.20
92	toluene (20)	0.0901	1013	11247	16.1674	b	16.0197	−0.92
93	toluene (30)	0.0901	944	10477	15.3649	b	15.2307	−0.88
94	toluene (40)	0.0901	891	9896	14.7569	b	14.6353	−0.83
95	toluene (50)	0.0901	819	9096	13.9169	b	13.8155	−0.73
96	ethenyl-benzene	0.0928	1226	13208	18.0616	11	18.0292	−0.18
97	ethylbenzene	0.0884	1011	11436	16.4506	11	16.2134	−1.46
98	*p*-xylene	0.0845	946	11185	16.3938	11	15.9562	−2.74
99	cumene	0.0936	1112	11882	16.6558	11	16.6704	0.09
100	pseudocumene	0.0829	1011	12205	17.5535	11	17.0014	−3.25
101	mesitylene	0.0884	1125	12722	17.7804	11	17.5312	−1.42
102	naphthalene (120)	0.0911	594	6519	11.1177	11	11.1748	0.51
103	tetralin	0.0900	1518	16854	21.9403	11	21.7653	−0.80
104	*sec*-butylbenzene (50)	0.0922	931	10100	14.8717	11	14.8447	−0.18
105	1-methylnaphthalene	0.0920	1778	19329	24.3620	11	24.3015	−0.25
106	hexylbenzene	0.0835	1065	12766	18.1009	11	17.5763	−2.98
107	1,1-diphenylethane (50)	0.0891	1452	16294	21.4167	11	21.1912	−1.06
108	nonylbenzene	0.0844	1175	13916	19.2354	11	18.7547	−2.56
109	DHCH[c] (50)	0.0852	1886	22139	27.5935	11	27.1814	−1.52
110	1,1-diphenylheptane (50)	0.0868	1299	14967	20.1821	11	19.8317	−1.77
111	1-phenyl-3-(2-phenylethyl) undecane (50)	0.0936	1524	16287	21.1824	11	21.1840	0.01
112	1-pentadecylnaphthalene (70)	0.0888	1463	16466	21.6080	11	21.3674	−1.13
113	(3-octylundecyl)benzene (50)	0.0908	1268	13966	18.9428	11	18.8062	−0.73
114	1,1-diphenyltetradecane (50)	0.0863	1220	14142	19.3607	11	18.9859	−1.97
115	*n*-hexane	0.0943	587	6222	10.6586	8	10.8705	1.95
116	*n*-hexane (0)	0.0943	723	7670	12.2143	24	12.3543	1.13
117	*n*-hexane (40)	0.0943	515	5464	9.8310	24	10.0937	2.60
118	*n*-hexane (60)	0.0943	430	4561	8.8256	24	9.1684	3.74
119	*n*-hexane (20)	0.0882	549	6222	10.9305	b	10.8705	−0.55
120	*n*-hexane (30)	0.0882	500	5668	10.3210	b	10.3028	−0.18
121	*n*-hexane (40)	0.0882	458	5191	9.7914	b	9.8140	0.23
122	*n*-hexane (50)	0.0882	410	4648	9.1808	b	9.2576	0.83
123	heptane	0.0943	662	7017	11.5162	8	11.6851	1.45
124	*n*-heptane (0)	0.0943	799	8473	13.0651	24	13.1771	0.85
125	*n*-heptane (40)	0.0943	591	6263	10.7030	24	10.9125	1.92
126	*n*-heptane (60)	0.0943	505	5359	9.7150	24	9.9861	2.72
127	octane	0.0943	787	8343	12.9275	8	13.0439	0.89
128	*n*-octane (0)	0.0943	944	10007	14.6757	24	14.7491	0.50
129	*n*-octane (40)	0.0943	706	7486	12.0185	24	12.1657	1.21
130	*n*-octane (60)	0.0943	607	6437	10.8915	24	11.0908	1.80
131	pentane	0.0888	419	4711	9.2217	8	9.3221	1.08
132	hexane	0.0924	561	6078	10.5833	8	10.7229	1.30
133	heptane	0.0916	641	6997	11.6123	8	11.6646	0.45
134	octane	0.0928	739	7966	12.5940	8	12.6576	0.50
135	nonane	0.0943	848	8988	13.6073	8	13.7049	0.71
136	decane	0.0880	813	9244	14.1754	8	13.9672	−1.49
137	undecane	0.0903	837	9270	14.0882	8	13.9938	−0.67

TABLE 2: Continued

N^0	liquid	experimental data					calcd.	
		C	B, bar	$1/\beta_0$, bar	$V_0/\Delta V$	ref.	$V_0/\Delta V$	error %[a]
138	dodecane	0.0905	907	10016	14.8604	8	14.7583	−0.69
139	tridecane (40)	0.0880	840	9552	14.5002	8	14.2832	−1.52
140	tetradecane	0.0901	981	10888	15.7910	8	15.6518	−0.89
141	pentadecane (40)	0.0885	968	10936	15.9230	8	15.7012	−1.41
142	hexadecane	0.0905	1044	11540	16.4499	8	16.3200	−0.80
143	isopentane	0.0880	376	4270	8.7552	12	8.8702	1.30
144	neopentane	0.0865	274	3165	7.5191	12	7.7379	2.83
145	2-methylpentane	0.0895	499	5575	10.1554	12	10.2075	0.51
146	3-methylpentane	0.0879	513	5830	10.5113	12	10.4688	−0.41
147	2,2-dimethylbutane	0.0890	452	5080	9.6318	12	9.7002	0.71
148	2,3-dimethyl-butane	0.0874	489	5590	10.2686	12	10.2228	−0.45
149	3-ethylpentane	0.0919	680	7400	12.0336	12	12.0776	0.36
150	2,2,3-trimethylbutane	0.0902	608	6747	11.4075	12	11.4085	0.01
151	isooctane	0.0878	572	6512	11.2641	12	11.1676	−0.86
152	heptadecane	0.0877	1035	11794	16.8563	12	16.5802	−1.67
153	octadecane (80)	0.0878	800	9110	14.0433	12	13.8299	−1.54
154	7-hexyltridecane (50)	0.0888	929	10454	15.4035	12	15.2075	−1.29
155	eicosane (120)	0.0867	599	6910	11.7463	12	11.5757	−1.47
156	9-octyl-heptadecane (50)	0.0922	1117	12112	16.9606	12	16.9059	−0.32
157	triacontane (120)	0.0862	718	8335	13.3042	12	13.0361	−2.06
158	squalane	0.0902	1222	13549	18.5426	12	18.3786	−0.89
159	11-decylheneicosane (40)	0.0886	1143	12903	17.9618	12	17.7171	−1.38
160	13-dodecylhexacosane (40)	0.0895	1207	13490	18.5192	12	18.3185	−1.10
161	tetracontane (160)	0.0868	625	7208	12.0677	12	11.8808	−1.57
162	isoprene	0.0991	809	8162	12.5378	12	12.8584	2.49
163	amilene	0.0961	519	5406	9.6971	12	10.0343	3.36
164	1,5-hexadiene	0.0841	509	6054	10.9438	12	10.6983	−2.29
165	1-hexene	0.0854	512	5997	10.8149	12	10.6399	−1.64
166	1-octene	0.0895	677	7560	12.3114	12	12.2416	−0.57
167	squalene	0.0912	1395	15284	20.2722	12	20.1565	−0.57
168	cyclopentane	0.0886	677	7642	12.4459	12	12.3256	−0.98
169	methylcyclopentane	0.0880	671	7623	12.4519	12	12.3061	−1.18
170	cyclohexane	0.0852	750	8808	13.8597	12	13.5204	−2.51
171	methylcyclohexane	0.0870	773	8883	13.8443	12	13.5973	−1.82
172	cycloheptane	0.0909	1006	11067	15.9376	12	15.8353	−0.65
173	cyclooctane (50)	0.0866	926	10692	15.7667	12	15.4508	−2.04
174	*trans*-decalin	0.0921	1253	13595	18.4941	12	18.4258	−0.37
175	butylcyclohexane	0.1008	1133	11246	15.6872	12	16.0187	2.07
176	perhydrochrysene (40)	0.0847	1657	19567	25.0091	12	24.5456	−1.89
177	DPPH[d] (40)	0.0905	1466	16202	21.2521	12	21.0975	−0.73
178	1-(1-decahydronaphthyl) pentadecane (80)	0.0871	1114	13073	18.2145	12	17.8910	−1.81
179	CPPD[e] (40)	0.0927	1402	15124	20.0378	12	19.9929	−0.22
180	CHHU[f] (40)	0.0922	1447	15686	20.6356	12	20.5688	−0.32
181	9-(2-cyclohexylethyl) heptadecane (40)	0.0898	1274	14185	19.2183	12	19.0299	−0.99
182	9-(3-cyclopentylpropyl) heptadecane (40)	0.0921	1246	13526	18.4249	12	18.3552	−0.38
183	DHNU[g] (40)	0.0820	1451	17691	23.2588	12	22.6231	−2.81
184	1,4-cyclohexadiene	0.0884	974	11018	16.0122	12	15.7850	−1.44
185	dichloromethane	0.0976	946	9693	14.2038	13	14.4273	1.55
186	chloroform	0.0957	923	9640	14.2303	13	14.3730	0.99
187	tetrachloromethane	0.0933	827	8861	13.5174	13	13.5747	0.42
188	bromomethane	0.0976	662	6789	11.1386	13	11.4515	2.73
189	bromoform (50)	0.1035	1165	11254	15.5892	13	16.0267	2.73
190	iodomethane	0.0955	908	9511	14.1055	13	14.2408	0.95
191	1,1-dichloroethane	0.0980	856	8740	13.1910	13	13.4507	1.93
192	1,2-dichloroethane	0.0960	1199	12486	17.1691	13	17.2893	0.70
193	1,1,1-trichloroethane	0.0987	916	9276	13.7230	13	14.0000	1.98
194	1,1,2-trichloroethane	0.0992	1374	13857	18.4412	13	18.6942	1.35
195	1,1,2,2-tetrachloroethane	0.1000	1643	16423	21.0270	13	21.3237	1.39
196	trichloroethene	0.1010	1160	11487	15.9285	13	16.2656	2.07
197	tetrachloroethene	0.1017	1366	13429	17.8965	13	18.2557	1.97
198	bromoethane	0.0943	702	7442	11.9689	13	12.1206	1.25
199	1-bromopropane	0.0889	782	8796	13.6580	13	13.5081	−1.11
200	2-bromopropane	0.0902	678	7520	12.2395	13	12.2006	−0.32
201	1-iodopropane	0.1010	1117	11058	15.4826	13	15.8260	2.17
202	1-chlorobutane	0.0964	811	8411	12.9122	13	13.1136	1.54
203	1-bromobutane	0.0977	934	9559	14.0628	13	14.2900	1.59
204	1-iodobutane	0.1006	1163	11565	16.0258	13	16.3456	1.96
205	1-chloropentane	0.0966	874	9049	13.5742	13	13.7674	1.40
206	1-bromopentane	0.0899	953	10610	15.5146	13	15.3670	−0.96

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TABLE 2: Continued

N^0	liquid	experimental data					calcd.	
		C	B, bar	$1/\beta_0$, bar	$V_0/\Delta V$	ref.	$V_0/\Delta V$	error %[a]
207	1-iodopentane	0.1005	1152	11459	15.9184	13	16.2370	1.96
208	fluorobenzene	0.0854	851	9966	15.0710	13	14.7070	−2.47
209	hexafluorobenzene	0.0850	714	8407	13.4427	13	13.1095	−2.54
210	chlorobenzene	0.0957	1275	13325	18.0476	13	18.1491	0.56
211	chlorobenzene (45)	0.0937	1098	11710	16.4721	25	16.4942	0.13
212	chlorobenzene (65)	0.0937	961	10250	14.9547	25	14.9981	0.29
213	chlorobenzene (85)	0.0937	835	8907	13.5477	25	13.6219	0.54
214	chlorobenzene (20)	0.0938	1278	13617	18.4359	b	18.4483	0.07
215	chlorobenzene (30)	0.0938	1202	12807	17.6013	b	17.6183	0.10
216	chlorobenzene (40)	0.0938	1129	12037	16.8069	b	16.8292	0.13
217	chlorobenzene (50)	0.0938	1060	11296	16.0390	b	16.0699	0.19
218	1,2-dichlorobenzene	0.0934	1525	16324	21.2298	13	21.2222	−0.04
219	chlorocyclohexane	0.0937	1110	11857	16.6286	13	16.6448	0.10
220	2,4-dichlorotoluene	0.0794	1550	19509	25.2833	13	24.4860	−3.26
221	bromobenzene	0.0974	1459	14981	19.6709	13	19.8460	0.88
222	bromobenzene (45)	0.0937	1247	13305	18.1181	25	18.1286	0.06
223	bromobenzene (65)	0.0937	1103	11769	16.5328	25	16.5546	0.13
224	bromobenzene (85)	0.0937	972	10368	15.0782	25	15.1190	0.27
225	2,4-dibromotoluene	0.0780	1391	17820	23.6509	13	22.7552	−3.94
226	*tert*-butylamine	0.0916	541	5906	10.4297	14	10.5467	1.11
227	aniline	0.0937	2007	21404	26.3794	14	26.4278	0.18
228	aniline (45)	0.0937	1798	19182	24.1242	25	24.1509	0.11
229	aniline (65)	0.0937	1606	17127	22.0317	25	22.0451	0.06
230	aniline (85)	0.0937	1429	15247	20.1116	25	20.1186	0.03
231	*o*-toluidine (50)	0.0849	1688	19872	25.3048	14	24.8579	−1.80
232	acetonitrile (20)	0.1025	942	9199	13.4957	b	13.9211	3.06
233	acetonitrile (30)	0.1025	871	8499	12.7635	b	13.2038	3.33
234	acetonitrile (40)	0.1025	812	7928	12.1622	b	12.6187	3.62
235	acetonitrile (50)	0.1025	743	7251	11.4465	b	11.9249	4.01
236	propanenitrile	0.0950	863	9083	13.6766	14	13.8022	0.91
237	butanenitrile	0.0922	934	10133	14.9051	14	14.8782	−0.18
238	2-methylpropanenitrile	0.0989	867	8772	13.1884	14	13.4835	2.19
239	benzonitrile	0.0989	1586	16040	20.6858	14	20.9312	1.17
240	pyridine (40)	0.0947	1282	13528	18.3014	14	18.3572	0.30
241	quinoline	0.0919	1972	21453	26.5225	14	26.4780	−0.17
242	nitromethane	0.0943	1300	13789	18.5900	14	18.6246	0.19
243	nitroethane	0.0965	1225	12689	17.3575	14	17.4974	0.80
244	1-nitropropane	0.0925	1149	12413	17.2561	14	17.2145	−0.24
245	2-nitropropane	0.0892	1005	11264	16.2270	14	16.0371	−1.18
246	2-methyl-2-nitropropane (40)	0.1032	991	9602	13.8876	14	14.3342	3.12
247	nitrobenzene	0.0932	1852	19876	24.8562	14	24.8621	0.02
248	nitrobenzene (45)	0.0937	1679	17908	22.8274	25	22.8454	0.08
249	nitrobenzene (65)	0.0937	1505	16049	20.9309	25	20.9404	0.05
250	nitrobenzene (85)	0.0937	1342	14311	19.1518	25	19.1595	0.04
251	formamide	0.1053	2614	24816	29.3076	14	29.9242	2.06
252	*N*-methylformamide	0.0937	1581	16862	21.7625	14	21.7735	0.05
253	*N,N*-dimethylformamide	0.0982	1535	15622	20.2882	14	20.5029	1.05
254	*N,N*-dimethylacetamide	0.0921	1428	15505	20.4573	14	20.3830	−0.36
255	triethanolamine	0.1014	2736	26974	31.6484	14	32.1355	1.52
256	1-methylpyrrolidin-2-one	0.0911	1735	19048	24.1231	14	24.0136	−0.46
257	3-cyanopropanal	0.0906	1390	15339	20.3610	14	20.2129	−0.73
258	2-fluoroethanol	0.0974	1496	15358	20.0559	14	20.2323	0.87
259	2,2-difluoroethanol	0.0948	1294	13650	18.4248	14	18.4821	0.31
260	2,2,2-trifluoroethanol	0.0929	749	8064	12.6939	14	12.7580	0.50
261	2,2,3,3-tetrafluoropropanol	0.0837	1063	12702	18.0198	14	17.5107	−2.91
262	bis(disfluoromethyl) ether	0.0869	333	3832	8.2973	14	8.4214	1.47
263	HFE245mf[h]	0.0903	450	4982	9.4639	14	9.5998	1.42
264	HFE245mc[i]	0.0891	365	4098	8.5103	14	8.6940	2.11
265	pentafluorobenzonitrile	0.0847	1062	12547	17.8039	14	17.3519	−2.61
266	tetramethylstannane	0.0937	580	6186	10.6437	14	10.8336	1.75
267	tetramethylsilane	0.0904	305	3372	7.6056	14	7.9500	4.33
268	tetraethylsilane	0.0855	777	9093	14.1443	14	13.8125	−2.40
269	tetraethoxysilane	0.0880	728	8272	13.1439	14	12.9712	−1.33
270	octamethylcyclotetrasiloxane (50)	0.0859	472	5494	10.2343	14	10.1241	−1.09
271	dimethyl sulfoxide	0.1057	2010	19019	23.4327	14	23.9839	2.30
272	Hg	0.0640	16003	250000	257.732	26	260.676	1.13

[a] Calculated from $100[(V_0/\Delta V_{1kbar})_{calcd} - (V_0/\Delta V_{1kbar})_{exp}]/(V_0/\Delta V_{1kbar})_{exp}$. [b] This work. [c] 1,2,3,4,4a,7,8,9,10,11,12,12a-Dodecahydrochrysene. [d] 1,7-Dicyclopentyl-4-(3-cyclopentylpropyl)heptane. [e] 1-Cyclopentyl-4-(3-cyclopentylpropyl)dodecane. [f] 1-Cyclohexyl-3-(2-cyclohexylethyl)undecane. [g] 1,1-Bis-(decahydronaphtyl)undecane. [h] 2,2,2-Trifluoroethyldifluoromethyl ether. [i] Pentafluoroethylmethyl ether.

volume of expansion of all liquids in system at the pressure reduction is defined by (eq 2)

$$m_{2,p}/d_p = \sum \Delta v_{\text{HL/Water}} = \Delta v_{1,p} + \Delta v_{2,p} + \Delta v_{3,p} + \Delta v_{11,p} +$$
$$\Delta v_{\text{cap}.,p} + \Delta v_{\text{def}.,p} \quad (2)$$

where d_p is the density of HL at pressure (P) and temperature (T).

The compressibility of a liquid can be defined by the change of its volume or density

$$(V_0 - V_p)/V_0 = (d_p - d_0)/d_p \quad (3)$$

and the problem is brought to determine the value of ΔV or d_p in the chosen pressures range. The value $\Delta v_{\text{def}.,p}$ includes an elastic reversible change of the volume of all parts of the stainless steel device at increasing pressure and is difficult to
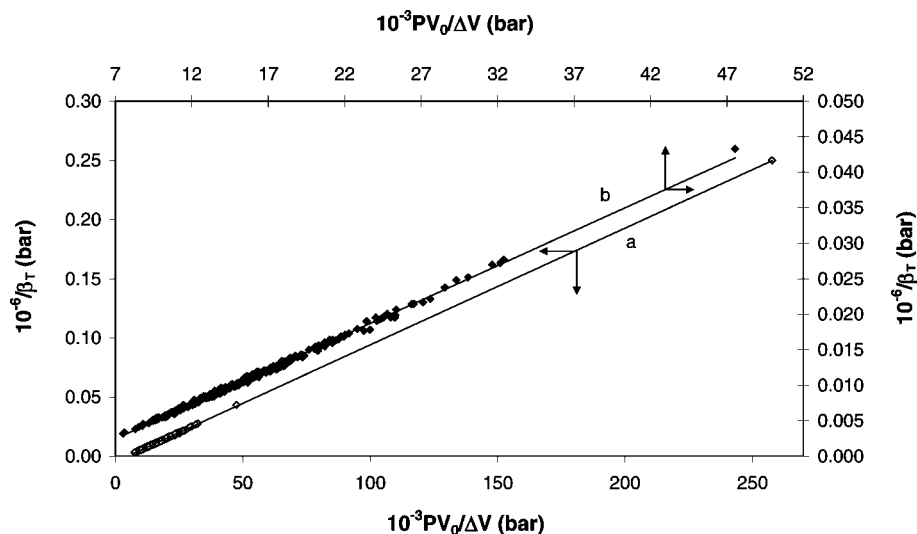
Figure 3. Relation between the tangent bulk modulus values at atmospheric pressure $(1/\beta_0)$ and secant bulk modulus values at 1000 bar $(PV_0/\Delta V)$ of all liquids in Table 2 (line a) and without mercury (line b).

TABLE 3: Experimental Values of the Tangent Bulk Modulus at Atmospheric Pressure $(1/\beta_0,$ bar) of Some Liquids and Unknown Values of the Second Bulk Modulus Calculated From Eq 11 at 1000 bar $(PV_0/\Delta V_{1\text{kbar}})$ and Coefficients (C,B) of the Tait Equation at 25°C until Other Temperatures Indicated

N^0	liquid	experimental data		calculated data		
		$1/\beta_0$, bar	ref.	$PV_0/\Delta V_{1\text{kbar}}$	C	B, bar
1	2-heptanone	10449	27	15202	0.0929	971
2	2-nonanone	11561	27	16342	0.0932	1078
3	2-decanone	12048	27	16841	0.0933	1125
4	1,1,2-trichloro-1,2,2-trifluoroethane	5637	28	10271	0.0885	499
5	carbon disulfide	10526	28	15281	0.0929	978
6	hexamethylphosphoric triamide	12658	29	17466	0.0934	1183
7	2-methyl-1-pentanol	11534	30	16314	0.0932	1075
8	3-methyl-2-pentanol	11416	30	16192	0.0932	1064
9	3-methyl-3-pentanol	10718	30	15478	0.0930	997
10	2-ethylhexanol (20)	10870	31	15633	0.0930	1011
11	propyl formate (20)	9434	32	14162	0.0924	872
12	n-propyl acetate(30)	8703	33	13413	0.0920	800
13	n-butyl acetate (30)	9542	33	14272	0.0925	882
14	n-pentyl acetate (30)	10438	33	15191	0.0929	970
15	i-pentyl acetate (30)	9452	33	14180	0.0924	874
16	ethyl propanoate (20)	9434	32	14162	0.0924	872
17	methyl butanoate (30)	8170	33	12867	0.0916	748
18	ethyl butanoate (30)	7974	33	12666	0.0914	729
19	1-chlorohexane	9930	30	14671	0.0927	920
20	pentanonitrile	10526	34	15281	0.0929	978
21	hexanonitrile	10799	34	15561	0.0930	1004
22	octanonitrile	11628	34	16410	0.0932	1084
23	2-propylamine (20)	5945	35	10587	0.0890	529
24	2-butylamine (20)	7962	35	12654	0.0914	728
25	diethylamine (20)	6605	35	11263	0.0900	594
26	di-n-propylamine (20)	8110	35	12805	0.0915	742
27	di-n-butylamine (20)	9390	35	14116	0.0924	868
28	triethylamine	7252	36	11926	0.0907	658
29	m-toluidine (20)	19608	32	24587	0.0931	1826
30	pyrrole (20)	15337	35	20211	0.0936	1435
31	pyrrolidine (20)	12285	35	17083	0.0934	1147
32	piperidine (20)	10905	35	15669	0.0930	1015
33	2-aminoethanol (20)	23474	35	28549	0.0923	2167
34	morpholine (20)	16260	35	21157	0.0935	1521

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define. In the proposed method, its determination is not necessary. The difference in the relations in eqs 1 and 2 works to give the simple eq 4

$$(m_{1,p} - m_{2,p})/d_p = \Delta v_{(3+11),\text{HL}} - \Delta v_{(3+11),\text{Water}} \quad (4)$$

To find the dependencies of the HL density (d_p) on pressure (P) by means of the relation in eq 4, we should have the experimental data for dependence ($m_p - P$) when loading the reservoirs (3, 11) with HL and water. From the relation in eq 3, it follows that $\Delta v_{p(3+11),\text{HL}} = [V_0(d_p - d_0)/d_p]_{\text{HL}}$ and $\Delta v_{p(3+11),\text{Water}} = (V_0 - V_{p,\text{Water}}) = [V_0(d_p - d_0)/d_p]_{\text{Water}}$.

At the equal initial volumes (V_0) of water and HL in reservoirs (3+11), we have

$$\Delta m_p/d_p = \Delta v_{p(3+11),\text{HL}} - \Delta v_{p(3+11),\text{Water}} = V_0(d_p - d_0)/d_p -$$
$$(V_0 - V_p)_{\text{Water}} \quad (5)$$

where d_0 and d_p are densities of HL under atmospheric and elevated pressures, respectively.

Taking into account that $V_0 d_{0,\text{HL}} = M_{0,\text{HL}}$, we obtain the main relation defined below

$$(\Delta m_p + M_{0,\text{HL}}) = V_{p,\text{Water}} \times d_{p,\text{HL}} \quad (6)$$

where $M_{0,\text{HL}}$ is the mass of HL in reservoirs (3 + 11) with the known volume V_0 under atmospheric pressure.

Data on the compressibility of water for broad temperature and pressure ranges are determined with high precision in refs 17 and 18, and the values $d_{p,\text{HL}}$ are calculated from eq 6.

The next measurements (the third cycle) are performed with liquid (S) under consideration, which is placed in volumes (3+11) of the device. Similar comparison gives us the relation in eq 7

$$(m_{\text{HL/HL}} - m_{\text{HL/S}})/d_p = \Delta v_{p(3+11),\text{HL}} - \Delta v_{p(3+11),\text{S}} =$$
$$M_{\text{HL}}[(d_p - d_0)/d_p d_0]_{\text{HL}} - M_{\text{S}}[(d_p - d_0)/d_p d_0]_{\text{S}} \quad (7)$$

where M_{HL} and M_{S} are the masses of the HL and liquid (S) under tests with equal initial volumes under atmospheric pressure and a chosen temperature and d_0 and d_p are the densities of the hydraulic liquid (HL) and investigated liquid (S) at normal and elevated pressures.

The values $d_{p,\text{S}}$ in eq 7 can be calculated from experimental data ($m_{\text{HL/HL}} - P$) and ($m_{\text{HL/S}} - P$) and the presently known data of the density ($d_{p,\text{HL}}$) of the hydraulic liquid from eq 6. It is necessary to mention that the proposed method takes away the necessity to determine the compressibility of the liquid in fixed volumes ($\Delta v_{1,p}$, $\Delta v_{2,p}$, and $\Delta v_{\text{cap.},p}$) and that of the vessel ($\Delta v_{\text{def.},p}$).

Results and Discussion

Table 1 presents an example of the experimental data on the determination of the compressibility ($\Delta V/V_0 = F(P)$) for toluene at 20 °C. High sensitivity and reproducibility of the experimental data (Figure 2) is provided by a heavy load of hydraulic liquid [(3000–7000) ± 1 mg], resulting in receiver 9. The error of the determination of compressibility values does not exceed 0.5% and can be reduced by an increasing level of control of temperature, mass, and pressure.

The comparison of the toluene data ($\Delta V/V_0 - P$) from Table 1 with analyzed data from 24 papers[11] on toluene compressibility produces a straight line with a slope = 1.00304 and $r = 0.99998$.

The measurements of the density at 20, 30, 40, and 50 °C and pressures up to 1000 bar were performed for 1,4-dioxane,

acetonitrile, toluene, ethyl acetate, chlorobenzene, and *n*-hexane, and coefficients C and B of Tait (eq 8)[19–21] were calculated (Table 2).

$$\Delta V/V_0 = C \ln[(B + P)/B] \quad (8)$$

The polynomial function ($\Delta V/V_0$ vs P) can satisfactorily describe the compression isothermal curve; however, its derivatives in boundary points P_{min} and P_{max} can be strongly distorted. The unreliable results especially appear with extrapolation out of the borders of the measurements, where a false maximum for this dependence is often predicted. Careful examinations of the errors of the measurement of compressibility were performed earlier.[17,21–23] The change of volume in a small pressure range ($P \sim 5$ bar) can be used for determination of the isothermal coefficient of the liquid compressibility (β_{T}) with an error of about ±0.5% due to the curvature (eq 8) However, in this pressure range, the value $\Delta V/V_0$ is only about 5×10^{-4}, which requires the measurements of $\Delta V/V_0$ to within ±2–3 ppm to be among-means ± 0.5%. Due to the same reason, the temperature control must be within ±1·10^{-3} °C. The small pressures (ranges up to 100–300 bar) increase the error in the values of the coefficients of eq 8.[21,22] It is necessary to have a large number of measurements for smoothing of the dependencies ($\Delta V/V_0 = F(P)$) due to fluctuation of the experimental errors. The method presented here meets the optimal conditions for determination of the values of β_0 and C and B (Figure 2 and Table 2).

It was suggested[21,22] that the best linear correlation for the volumes of the liquid in the pressure ranges up to 1000 bar is (eq 9)

$$V_0 P/(V_0 - V_p) = 1/\beta_0 + kP \quad (9)$$

where k is the coefficient of proportionality for a given liquid.

Considerable errors in the estimation of the $V_0 P/(V_0 - V_p)$ values were shown[21] to be observed at low pressures (up to 100–200 bar) because of low accuracy of the measurements of the $P/(V_0 - V_p)$ value. It was proposed[21] that the relation in eq 9 should be simple and reliable for description of the experimental data on the compressibility of organic liquids in the interval of 200–1000 bar and for their verification at 0–200 bar.

We found that in contrast to eq 9, which is assigned for an individual liquid, a distinct linear dependence, eq 10, between the tangent bulk modulus value at atmospheric pressure [$K_0 = 1/\beta_0 = -V_0(\partial P/\partial V)_T$] and the secant bulk modulus value at 1000 bar ($-1000 V_0/\Delta V_{\text{1kbar}}$) is observed for different compounds (Table 2, Figure 3). This dependence covers liquids of different classes and in a wide temperature range.

$$1/\beta_0 = (-4559 \pm 22.9) +$$
$$(0.9865 \pm 0.0010)(1000 V_0/\Delta V_{\text{1kbar}})$$
$$r = 0.9999; \ n = 272 \quad (10)$$

When mercury, which differs drastically in compressibility from that of other liquids, was excluded from consideration, the correlation parameters remained almost unchanged

$$1/\beta_0 = (-4386 \pm 51.2) +$$
$$(0.9759 \pm 0.0030)(1000 V_0/\Delta V_{\text{1kbar}})$$
$$r = 0.9993; \ n = 271 \quad (11)$$

The discovered dependencies (eqs 10 and 11) for different classes of compounds (cyclic, linear, and branched alkanes and alkenes, alcohols, aldehydes, ketones, ethers, esters, nitriles, halogen- and nitro-derivatives, glycols, glycerol, and even mercury) in a wide temperature range enable one to draw the

conclusion about the realization of the rule of noncrossing of the compression curvatures. The slope near to unity corresponding to that initial degree of compressibility of liquids is in accord with the change of compressibility at elevated pressures, and relation eq 9 for all liquids (Table 2) generates near to parallel lines.

Since volume changes at 1000 bar are rather large and can be measured with high accuracy, the method based on eq 11 for checking and predicting β_0 values appears to be reliable for different compounds in a wide temperature range. However, more often, there is another problem: calculating the coefficients of the Tait equation from the known value β_0. In this case, eq 11 allows calculation of the value $\Delta V/V_0$ at a pressure of 1 kbar. Then two relations, $\beta_0 = C/B$ and $\Delta V/V_0 = C \ln[(B + P)/B]$, allow us to calculate unknown coefficients C and B (Table 3) and $V-P$ curves in this pressure interval.

Conclusion

We have proposed a simple and precise method for the determination of the compressibility of liquids in a range of temperatures by measuring the differences in the weights of hydraulic oil injected into the high-pressure device. The detected clear relation between the tangent and secant bulk moduli for various liquids at different temperatures ("noncrossing rule") has revealed the means of calculation of the Tait equation coefficients from the values of compressibility at ambient pressure.

Supporting Information Available: Additional experimental data and values. This material is available free of charge via the Internet at http://pubs.acs.org.

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