Speed of Sound, Densities, and Isentropic Compressibilities of Liquid 1-Bromoalkanes at Temperatures from (243.15 to 423.15) K†

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This work presents the results of systematic research of the sound speed and density of liquid bromoalkanes on the saturation line within the temperature range from (243.15 to 423.15) K. These results have been used for the calculation of isentropic compressibility β_S, ratio of thermal capacities, and other thermophysical properties of the studied substances. Additionally, the Integral Constant of Dispersion Forces B for bromoalkanes has been estimated on the basis of the atom−atomic interactions model.

Introduction

Creation of the general theory of the liquid state is the important problem of modern physics. That is why experimental and theoretical investigations of equilibrium properties of organic liquids are fairly actual. As distinct from *n*-alkanes, the results of systematic studies of which have been generalized and presented in reference books and on the NIST WEBBOOK sites, studies of thermophysical properties of their halogen-substituted analogues are fragmentary and not consecutive.[1−4] For this reason, revealing of the general behavior of thermophysical properties of these extremely important organic liquids is not possible.

The previous article[5] reports experimental data of density for some 1-bromoalkanes. Here we present the results of further measurements of density and sound speed of 1-bromoalkanes including new homologues (1-bromononane, 1-bromodecane, and 1-bromoundecane).

Experimental Section

Materials. The materials used in this study [1-bromopropane (1-BrPr), 1-bromobutane (1-BrBu), 1-bromohexane (1-BrHx), 1-bromoheptane (1-BrHp), 1-bromooctane (1-BrOc), and 1-bromododecane (1-BrDod) (with a mole fraction purity of 0.999, 0.997, 0.994, 0.993, 0.992, and 0.992, respectively)] were supplied by Acros Organics. 1-Bromopentane (1-BrPe) with mole fraction purity 0.989 was obtained from Sigma-Aldrich. 1-Bromononane (1-BrNo), 1-bromodecane (1-BrDec), and 1-bromoundecane (1-BrUnd) (with mole fraction purity 0.979, 0.978, and 0.979, respectively) were supplied by Fluka.

Measurements. The speed of sound was measured by the pulse-phase echo ultrasonic method developed in the Molecular Acoustic Laboratory of Kursk State University.[6] The measurements were executed in nondisperse area at a frequency of (1 to 5) MHz on saturation line. For thermostatting of the sound speed measuring cell with a temperature stability of ± 0.01 K, KRIOVIST and VIS-T thermostats (Termex, Russia) were used. The temperature was controlled and measured with the 100 Ω platinum resistance thermometers by a digital thermometer bridge (Terkon, Termex Russia) on ITS-90. The temperature of studied liquids was measured with one of the thermometers placed in the measuring cell directly, with the uncertainty of ± 0.02 K.

The account of various uncertainty factors leads to the conclusion that the total uncertainty of sound speed does not surpass 0.1 %.

Table 1. Values of the Parameters of Equation 1 for Sound Speed u and Standard Deviation $\sigma(u)$ for Liquid 1-Bromoalkanes from (243.15 to 423.15) K

liquid	A_0 / m·s^{-1}	A_1 / m·s^{-1}	$A_2 \cdot 10^3$ / m·s^{-1}	$\sigma(u)$ / m·s^{-1}
1-bromopropane	2113.38	−4.36290	1.5961	0.15
1-bromobutane	2131.10	−4.20129	1.4597	0.37
1-bromopentane	2200.47	−4.37484	1.7793	0.22
1-bromohexane	2224.33	−4.34326	1.8104	0.32
1-bromoheptane	2261.16	−4.37313	1.8888	0.39
1-bromooctane	2293.74	−4.40304	1.9409	0.27
1-bromononane	2322.42	−4.42191	1.9936	0.33
1-bromodecane	2347.64	−4.46154	2.0881	0.10
1-bromoundecane	2375.51	−4.48286	2.1001	0.29
1-bromododecane	2413.68	−4.62141	2.3119	0.37

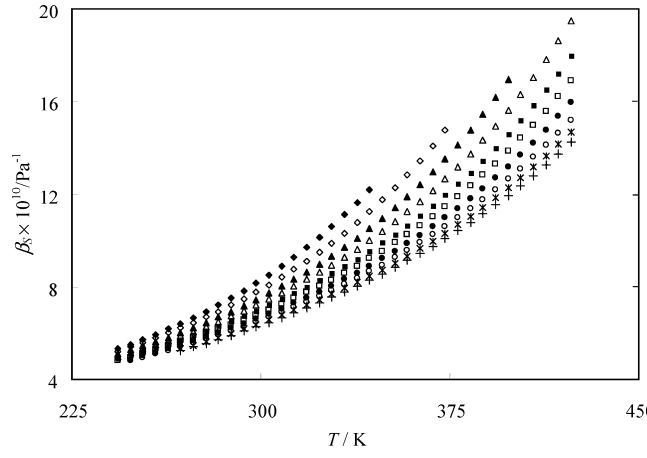

Figure 1. Temperature dependence of isentropic compressibility (β_S) of liquid 1-bromoalkanes. ♦, 1-bromopropane; ◊, 1-bromobutane; ▲, 1-bromopentane; Δ, 1-bromohexane; ■, 1-bromoheptane; □, 1-bromooctane; ●, 1-bromononane; ○, 1-bromodecane; *, 1-bromoundecane; +, 1-bromododecane.

† Part of the special section "2008 European Conference on Thermophysical Properties".

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10.1021/je900227j CCC: $40.75
Published on Web 05/01/2009

Table 2. Experimental Sound Speed u Values for Liquid 1-Bromoalkanes at Various Temperatures

T/K	u/m·s^{-1}	T/K	u/m·s^{-1}	T/K	u/m·s^{-1}	T/K	u/m·s^{-1}	T/K	u/m·s^{-1}
1-bromopropane		1-bromobutane		1-bromopentane		1-bromohexane		1-bromoheptane	
243.76	1144.8	243.38	1195.3	243.73	1240.0	243.60	1273.8	243.74	1308.4
263.38	1075.0	273.26	1091.0	273.32	1137.5	273.31	1172.9	273.33	1207.2
273.31	1040.1	323.22	926.0	303.22	1037.1	303.20	1073.6	303.19	1108.7
283.26	1005.5	343.22	861.2	323.17	972.4	323.20	1009.3	323.14	1045.1
293.22	971.2	363.23	797.6	353.18	877.6	353.16	916.0	353.14	952.9
298.21	954.1	383.31	735.2	383.23	785.5	383.19	826.4	383.17	863.5
323.21	870.2			403.28	725.5	403.25	767.5	423.22	748.1
343.21	804.2			423.48	666.6	423.23	710.0		
363.23	739.4								
383.47	674.9								
1-bromooctane		1-bromononane		1-bromodecane		1-bromoundecane		1-bromododecane	
243.61	1336.3	253.46	1329.9						
273.28	1235.4	273.29	1262.7	323.20	1123.8	273.42	1307.0	273.32	1323.5
303.16	1137.2	303.15	1165.1	353.19	1032.2	303.22	1209.0	303.21	1224.8
323.02	1073.8	323.14	1101.1	383.22	944.5	323.19	1145.6	323.18	1161.2
352.91	981.9	353.10	1009.9	403.23	888.2	353.14	1054.4	353.15	1069.7
382.88	892.9	383.05	921.5	423.27	833.2	383.14	966.6	383.17	983.0
402.55	835.5	402.98	864.4			403.13	909.7	403.19	926.4
423.04	778.7	422.72	808.8			423.13	854.5	423.14	871.7

Table 3. Values of the Parameters of Equation 3 for Density ρ and Standard Deviation $\sigma(\rho)$ for Liquid 1-Bromoalkanes from (243.15 to 423.15) K

liquid	A_0 / kg·m^{-3}	A_1 / kg·m^{-3}	A_2·10^3 / kg·m^{-3}	A_3·10^6 / kg·m^{-3}	$\sigma(\rho)$ / kg·m^{-3}
1-bromopropane	2954.08	−14.740	50.469	−64.187	0.15
1-bromobutane	1333.03	1.8533	−9.9389	9.9428	0.16
1-bromopentane	1349.85	0.63417	−4.9056	3.9934	0.17
1-bromohexane	1504.09	−1.1780	0.38417	−0.77016	0.20
1-bromoheptane	1490.15	−1.5892	2.1526	−2.8206	0.12
1-bromooctane	1392.80	−0.92809	−0.070002	−0.14485	0.13
1-bromononane	1409.08	−1.4308	1.7356	−2.0668	0.10
1-bromodecane	1370.39	−1.2595	1.1955	−1.3834	0.08
1-bromoundecane	1383.82	−1.6233	2.4011	−2.5700	0.10
1-bromododecane	1376.72	−1.6890	2.6883	−2.8971	0.26

Density measurements were carried out using a quartz pycnometer within the temperature range from (243.15 to 423.15) K at atmospheric pressure. The mass of the pycnometer was measured using an analytical balance with the precision ± 3·10^{-4} g. The pycnometer was calibrated with bidistillated water. The position of the pycnometer liquid level was recorded by a traveling microscope with the precision ± 0.01 mm. Thermostating of the pycnometer was carried out by the thermostats mentioned above. The account of push out force and thermal expansion of the pycnometer provide the estimated uncertainty of the density data for studied liquids within 0.01 %. Estimation has shown that distinction between the experimental density values for studied liquids at atmospheric pressure and on the saturation line within the considered temperature range does not surpass 0.01 %.

Results and Discussion

The set of experimental sound speed values has been approximated by the second polynomial degree

$$u/(\mathrm{m{\cdot}s^{-1}}) = A_0 + A_1(T/\mathrm{K}) + A_2(T/\mathrm{K})^2 \qquad (1)$$

where coefficients A_0, A_1, and A_2 are the adjustable parameters. The values of standard deviation $\sigma(u)$ were defined with the following equation

$$\sigma(u) = \left[\sum_{i=1}^{n} (u_{\mathrm{obs}} - u_{\mathrm{cal}})^2/(n-p)\right]^{1/2} \qquad (2)$$

Here u_{obs} and u_{cal} are observed and calculated by eq 1 quantities of sound speed; n is a total number of experimental points; and p is the number of estimated parameters. The values of the parameters A_0, A_1, and A_2 and standard deviation $\sigma(u)$ are listed in Table 1 for all studied liquids. The experimental sound speed values for liquid 1-bromoalkanes within the considered temperature range are listed in Table 2. Part of these data was presented in ref 7.

Table 4. Experimental Density Values ρ for Liquid 1-Bromononane, 1-Bromodecane, and 1-Bromoundecane at Various Temperatures

	ρ/kg·m^{-3}		
T/K	1-BrNo	1-BrDec	1-BrUnd
253.15	1124.52		
253.40		1105.66	
263.20	1115.20		
263.30		1096.34	
273.15	1105.50		
273.25		1087.32	1067.23
283.15	1096.09		
283.20		1078.06	1058.34
293.15	1086.58	1069.01	
293.20			1049.49
298.15	1082.20	1064.47	1045.03
323.05	1058.34		
323.15		1041.72	
323.20			1023.21
343.15	1038.92	1023.01	
343.20			1005.75
363.15	1019.42	1004.33	
363.20			987.93
383.15	999.36	985.41	
383.20			969.56
393.15	989.17		
403.25		966.27	951.27
413.15	968.48		
423.30		946.49	

The set of experimental density values has been approximated by the third polynomial degree that provides a better fit of density vs temperature for studied 1-bromoalkanes.

$$\rho/(\mathrm{kg{\cdot}m^{-3}}) = A_0 + A_1(T/\mathrm{K}) + A_2(T/\mathrm{K})^2 + A_3(T/\mathrm{K})^3 \qquad (3)$$

The values of the parameters A_0, A_1, A_2, and A_3 of eq 3 and standard deviation $\sigma(\rho)$ calculated by eq 2 for all studied liquids are listed in Table 3.

Table 4 presents the experimental values of density for liquid 1-bromononane, 1-bromodecane, and 1-bromoundecane. The

Table 5. Heat Capacity Ratio Values γ for Liquid 1-Bromoalkanes at Various Temperatures

	γ							
T/K	1-BrPr	1-BrBu	1-BrPe	1-BrHx	1-BrHp	1-BrOc	1-BrNo	1-BrDec
253.15	1.353	1.278	1.227	1.268	1.244	1.229	1.228	1.221
263.15	1.341	1.294	1.239	1.267	1.242	1.230	1.224	1.219
273.15	1.346	1.306	1.250	1.266	1.240	1.229	1.221	1.216
283.15	1.366	1.316	1.259	1.264	1.238	1.228	1.218	1.213
293.15	1.404	1.321	1.266	1.262	1.237	1.227	1.216	1.210
298.15	1.431	1.323	1.269	1.261	1.236	1.226	1.214	1.208
303.15		1.323	1.272	1.259	1.236	1.225	1.213	1.207
313.15		1.322	1.276	1.257	1.235	1.222	1.210	1.204
323.15		1.316	1.278	1.253	1.234	1.220	1.208	1.201
333.15			1.278	1.250	1.233	1.217	1.206	1.198
343.15			1.277	1.246	1.233	1.213	1.204	1.195
353.15			1.274	1.242	1.233	1.209	1.202	1.192
363.15			1.269	1.237	1.233	1.205	1.200	1.189
373.15			1.263	1.233	1.233	1.201	1.198	1.186
383.15			1.255	1.228	1.234	1.196	1.197	1.184
393.15			1.246	1.223	1.235	1.191	1.196	1.181

Table 6. Thermal Expansion Coefficient Values α_P for Liquid 1-Bromoalkanes at Various Temperatures

	$\alpha_P \cdot 10^3/K^{-1}$									
T/K	1-BrPr	1-BrBu	1-BrPe	1-BrHx	1-BrHp	1-BrOc	1-BrNo	1-BrDec	1-BrUnd	1-BrDod
243.15	1.106				0.876					
253.15	1.080	0.955	0.858	0.929	0.883	0.861	0.844	0.832		
263.15	1.081	0.999	0.895	0.941	0.891	0.872	0.849	0.837		
273.15	1.109	1.039	0.930	0.954	0.901	0.883	0.855	0.842	0.831	0.822
283.15	1.167	1.075	0.965	0.968	0.913	0.894	0.862	0.849	0.833	0.824
293.15	1.255	1.108	0.998	0.982	0.926	0.906	0.871	0.856	0.837	0.827
298.15	1.312	1.123	1.014	0.989	0.934	0.912	0.875	0.860	0.839	0.829
303.15		1.137	1.030	0.997	0.942	0.918	0.880	0.864	0.842	0.832
313.15		1.163	1.062	1.013	0.959	0.930	0.891	0.873	0.848	0.839
323.15		1.184	1.092	1.029	0.978	0.943	0.904	0.884	0.857	0.847
333.15			1.121	1.047	0.999	0.956	0.918	0.895	0.867	0.858
343.15			1.148	1.065	1.022	0.969	0.933	0.907	0.878	0.870
353.15			1.175	1.084	1.048	0.983	0.950	0.920	0.892	0.885
363.15			1.200	1.104	1.075	0.997	0.969	0.934	0.907	0.901
373.15			1.223	1.125	1.105	1.011	0.990	0.950	0.924	0.920
383.15			1.245	1.147	1.138	1.026	1.012	0.967	0.944	0.941
393.15			1.266	1.170	1.173	1.042	1.036	0.985	0.965	0.965

Table 7. Energy of Intermolecular Force Values $|E_P|$ for Liquid 1-Bromopropane, 1-Bromobutane, 1-Bromopentane, and 1-Bromohexane at Various Temperatures

	$\|E_P\|$/kJ·mol^{-1}							
	1-bromopropane		1-bromobutane		1-bromopentane		1-bromohexane	
T/K	(6)	(7)	(6)	(7)	(6)	(7)	(6)	(7)
253.15	29.6	31.9	33.9	37.9	37.9	43.7	46.1	50.0
263.15	29.1	31.2	34.2	37.1	38.4	42.9	45.9	49.0
273.15	28.9	30.6	34.4	36.4	38.7	42.1	45.6	48.1
283.15	28.9	29.9	34.4	35.6	38.8	41.3	45.3	47.2
293.15	29.2	29.2	34.2	34.9	38.8	40.5	44.9	46.3
298.15	29.4	28.8	34.0	34.5	38.8	40.1	44.7	45.8
303.15			33.8	34.1	38.7	39.7	44.4	45.4
313.15			33.4	33.3	38.5	38.9	43.9	44.5
323.15			32.8	32.5	38.1	38.1	43.3	43.6
333.15					37.6	37.2	42.7	42.7
343.15					37.1	36.4	42.0	41.8
353.15					36.4	35.6	41.3	40.9
363.15					35.6	34.7	40.5	40.0
373.15					34.8	33.9	39.6	39.1
383.15					33.8	33.1	38.7	38.3
393.15					32.8	32.3	37.8	37.4

Table 8. Energy of Intermolecular Force Values $|E_P|$ for Liquid 1-Bromoheptane, 1-Bromooctane, 1-Bromononane, and 1-Bromodecane at Various Temperatures

	$\|E_P\|$/kJ·mol^{-1}							
	1-bromoheptane		1-bromooctane		1-bromononane		1-bromodecane	
T/K	(6)	(7)	(6)	(7)	(6)	(7)	(6)	(7)
253.15	51.3	55.8	57.1	61.8	62.8	67.5	68.7	73.6
263.15	51.0	54.8	57.0	60.8	62.5	66.4	68.4	72.4
273.15	50.7	53.9	56.8	59.7	62.1	65.3	68.0	71.2
283.15	50.4	52.9	56.4	58.7	61.6	64.2	67.5	70.0
293.15	50.0	51.9	56.0	57.6	61.1	63.1	67.0	68.8
298.15	49.8	51.5	55.8	57.1	60.9	62.5	66.7	68.2
303.15	49.6	51.0	55.5	56.6	60.6	62.0	66.4	67.7
313.15	49.2	50.0	54.9	55.5	60.1	60.9	65.8	66.5
323.15	48.8	49.1	54.3	54.5	59.5	59.8	65.1	65.3
333.15	48.3	48.1	53.5	53.5	58.8	58.7	64.4	64.2
343.15	47.8	47.1	52.7	52.5	58.2	57.6	63.6	63.0
353.15	47.3	46.2	51.9	51.5	57.5	56.6	62.8	61.9
363.15	46.7	45.2	50.9	50.4	56.8	55.5	62.0	60.8
373.15	46.1	44.2	49.9	49.4	56.0	54.4	61.1	59.6
383.15	45.5	43.2	48.9	48.4	55.3	53.3	60.2	58.5
393.15	44.9	42.3	47.8	47.5	54.5	52.2	59.2	57.4

experimental density values for other studied 1-bromoalkanes were presented in refs 5 and 7.

Experimental values of density, sound speed, and available values of isobaric heat capacity[8] have been used for the calculation of the isentropic compressibility, the ratio of heat capacities c_P and c_V, and the energy of intermolecular forces for studied bromoalkanes.

Isentropic compressibilities β_S were calculated from the Laplace equation

$$\beta_S = \frac{1}{\rho u^2} \tag{4}$$

where u is the speed of sound and ρ is the density of substances. The values of isentropic compressibility for 1-bromoalkanes as a function of temperature are plotted in Figure 1. As it follows

Table 9. Values of Integral Constant of Dispersion Forces *B* for Liquid Bromoalkanes

	B/J·m^6·kg^{-3}						
liquid	*B*	liquid	*B*	liquid	*B*	liquid	*B*
1-BrPr	0.1295	1-BrBu	0.1571	1-BrPe	0.1818	1-BrHx	0.2040
1-BrHp	0.2238	1-BrOc	0.2417	1-BrNo	0.2578	1-BrDec	0.2723
1-BrUnDec	0.2855	1-BrDoDec	0.2976	1-BrTriDec	0.3086	1-BrTetraDec	0.3187

from Figure 1, the values of isentropic compressibility decrease with an increase of bromoalkane number. It can be connected with an increase of the intermolecular interaction energy.

The heat capacity ratio $\gamma = (c_P/c_V)$ (Table 5) has been calculated by the known formula

$$\gamma = 1 + \frac{u^2 \alpha_P^2 T}{c_P} \tag{5}$$

where α_P is a thermal expansion coefficient and c_P is an isobaric thermal capacity. Because of a lack of literature data of isobaric thermal capacity, in Table 5 the values of heat capacity ratios for 1-bromoundecane and 1-bromododecane are missed.

Values of α_P for liquid bromoalkanes presented in Table 6 differ a little from the values obtained earlier[5] due to distinction of the used polynomial degree. They were calculated by differentiation eq 3.

The interaction energy $|E_P|$ has been estimated by means of the relation

$$|E_P| = \frac{u^2 \alpha_P T}{\gamma} - \frac{RT}{M^*} \tag{6}$$

obtained earlier.[9] The value $M^* = 2M$. It is connected with presence dimer association of bromoalkanes.[10]

Tables 7 and 8 present values of intermolecular energy for studied liquid bromoalkanes calculated by eqs 6 and 7, the latter considering only dispersion forces

$$|E_P| = B\rho^2 \tag{7}$$

Values of $|E_P|$ for liquid 1-bromoundecane and 1-bromododecane are missed because of a lack of literature data of isobaric thermal capacity for these substances.

The analysis of presented values of $|E_P|$ leads to the conclusion that in the liquid bromoalkanes, as well as in *n*-alkanes and chloroalkanes,[11] the energy of intermolecular forces is proportional to the square of its density. The discrepancies between the results from eqs 6 and 7 are observed at low temperatures and high densities, probably because of the influence of repulsive forces. It means that the prevailing role in the intermolecular energy belongs to dispersion attraction forces.

Estimation of *B* in eq 7 is fulfilled by the relation

$$B = B_0(M_0/M)^3(\xi^2 + (1 - \xi)^2 a + 2\xi(1 - \xi)a^{1/2}) \tag{8}$$

which was received on the assumption of the mechanism of atom−atomic interaction. In eq 8, B_0 and M_0 are the constant of attraction forces and molecular weight of the reference *n*-alkane, respectively;[9] *B* and *M* are the same values for the monohalogenated *n*-alkane corresponding to the reference *n*-alkane; and $\xi = (2n + 1)/(2n + 2)$ is the share of hydrogen atomic centers in the halogenated *n*-alkane molecule. The constant *a* is the ratio between the constant of the pair potential of dispersion forces for the halogen and hydrogen atoms ($a = 31.66$).

Values of *B* estimated by eq 8 are listed in Table. 9. They are used for the calculation of the energy of intermolecular forces for bromoalkanes.

Summary

Thus, we for the first time present the experimental data for the sound speed in liquid bromoalkanes along the saturation line. Together with the density data, they allow us to conclude that the intermolecular energy of liquid bromoalkanes is proportional to the square of its density. Values of the constants *B* of these forces can be estimated on the basis of the atom−atomic intermolecular interactions model.

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Received for review November 13, 2008. Accepted April 20, 2009. The authors express their sincere gratitude to the Russian Fund of Basic Researches for financial support.

JE900227J