

Speed of Sound of Hexane + 1-Chlorohexane, Hexane + 1-Iodohexane, and 1-Chlorohexane + 1-Iodohexane at Saturation Condition

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Speed of sound for the binary mixtures hexane + 1-chlorohexane, hexane + 1-iodohexane, and 1-chlorohexane + 1-iodohexane has been measured as a function of composition and temperature along the saturation line between 293.15 K and 373.15 K. Nonlinear behavior is observed with temperature and concentration. The experimental results are correlated with polynomial equations of the second power.

Introduction

Knowledge of the thermophysical properties of mixtures of organic liquids is of great importance in various fields of petrochemical industry and technology. In contrast to the mixtures of *n*-alkanes, which have been studied comprehensively, the mixtures of *n*-alkanes and their halogen substituents are less well understood. A literature search indicated the availability of only a few thermodynamic properties on the binary mixtures of *n*-alkanes with 1-chloroalkanes and 1-iodoalkanes. The excess molar volumes V^E of the binary mixtures pentane + 1-chloropropane, or 1-chlorobutane, or 1-chloropentane, or 1-chlorohexane, and octane + 1-chloropropane, or 1-chlorobutane, or 1-chloropentane, or 1-chlorohexane, at $T = 298.15$ K have been studied by Kovacs et al.;¹ those of heptane + 1-chloropropane, or 1-chlorobutane, or 1-chloropentane, or 1-chlorohexane, at $T = 298.15$ K have been studied by Kovacs et al.² The excess molar enthalpies H^E of the binary mixtures 1-chloropentane + nonane or decane and 1-chlorohexane + nonane or decane or undecane at $T = 298.15$ K have been studied by Nunez et al.³ The viscosities, densities, and refractive indexes of nine *n*-alkane mixtures with 1-chloroalkane at 298.15 K have been measured by Aucejo.⁴ To the best of our knowledge, speed of sound of mixtures of *n*-alkanes with alkyl chlorides and alkyl iodides has not been studied.

In this paper, measurements of the speed of sound of the binary mixtures (hexane + 1-chlorohexane), (hexane + 1-iodohexane), and (1-chlorohexane + 1-iodohexane) at saturation condition are reported as part of our ongoing research program of measuring the thermodynamic properties of binary liquid mixtures of *n*-alkanes and their halogen substituents.^{5–7}

Experimental Section

The materials used in this study, hexane, 1-chlorohexane, and 1-iodohexane (mole fraction > 0.99), were supplied from Sigma-Aldrich Ltd. All chemicals were partially degassed and dried over Fluka type 0.4 nm molecular sieves. The purity of the products was checked by gas chromatography (GC). The mole percent GLC purity data 99.7, 99.3, and 99.4 were shown respectively for hexane, 1-chlorohexane, and 1-iodohexane. The mixtures were prepared by mass, with a precision of $\pm 5 \times 10^{-5}$ g. The

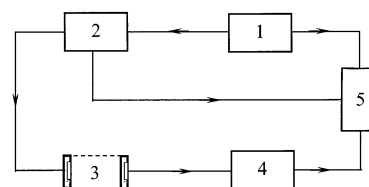


Figure 1. Block diagram of the equipment.

Table 1. Comparison of Experimental Speeds of Sound, u , with Literature Data for Pure Liquids at Different Temperatures

liquid	$u/\text{m}\cdot\text{s}^{-1}$		
	T/K	expt	lit.
hexane	293.15	1098.1	1099.8 ⁹
			1100 ¹⁰
	298.15	1076.0	1077 ¹¹
			1077.7 ¹²
1-chlorohexane	308.15	1031.8	1032 ¹⁰
	293.15	1219.1	1216.2 ¹³
1-iodohexane	293.15	1045.5	1045 ¹³

possible error in the mole fraction is estimated to be less than 1×10^{-4} . All molar quantities are based on the IUPAC relative atomic mass table (IUPAC, 1986). The ultrasonic speed was measured along the saturation line with a pulse-phase echo ultrasonic device constructed by the authors,⁸ to a precision of $\pm 1 \text{ m}\cdot\text{s}^{-1}$. A block diagram of the equipment is shown in Figure 1. The slave sweep of the oscilloscope 5 and the two-channel pulse generator (G5-4B, Russia) 2 was triggered by the oscillator 1. A rectangular pulse was transmitted from the first channel of the generator 2 to the emitter. After passing through the investigated medium, the ultrasonic pulse was transformed by the receiving piezoceramic into an electrical pulse, which was then amplified by the wide-band amplifier 4 and sent to the Y-plates of the oscilloscope.

The G5-4B generator was capable of delaying the second-channel pulse relative to the first-channel pulse, the delay time being determined by the frequency meter with the accuracy 0.005 μs .

The delayed second-channel pulse was transmitted to the cathode of the oscilloscope cathode-ray tube, where it functioned as a floating time marker. In the sound velocity measurements the marker coincided with one of the peaks of the high-frequency component of the pulse observed on

Table 2. Speed of Sound of Hexane (1) + 1-Chlorohexane (2) from 293.15 K to 373.15 K

Hexane (1) + 1-Chlorohexane (2)									
T/K	$u/m\cdot s^{-1}$								
	0	0.1	0.15	0.2	0.3	0.5	0.7	0.9	1
293.15	1098.1	1111.3	1121.1	1129.1	1135.3	1159.3	1179.6	1206.7	1219.1
298.15	1076.0	1089.2	1098.9	1106.9	1114.9	1138.3	1159.2	1186.4	1200.1
303.15	1054.3	1067.2	1076.7	1084.8	1094.3	1117.4	1139.0	1166.1	1181.9
308.15	1031.8	1045.2	1054.6	1062.8	1073.5	1096.6	1118.9	1146.1	1165.2
313.15	1009.9	1023.2	1032.6	1041.0	1052.6	1075.9	1098.8	1126.1	1143.7
318.15	987.7	1001.3	1010.6	1019.3	1031.5	1055.4	1078.9	1106.4	1128.1
323.15	965.7	979.5	988.8	997.7	1010.2	1035.1	1059.2	1086.7	1106.0
328.15	943.7	957.5	967.1	976.3	988.7	1014.9	1039.5	1067.3	1087.0
333.15	921.3	935.9	945.5	955.0	967.1	994.8	1020.0	1048.0	1068.8
338.15	899.7	914.3	923.9	933.8	949.0	974.8	1000.5	1028.9	1050.2
343.15	877.8	892.6	902.5	912.7	928.2	955.0	981.2	1009.8	1031.7
348.15	855.9	871.1	881.1	891.7	908.7	935.4	962.1	991.0	1013.3
353.15	834.2	849.5	859.9	870.9	888.7	915.9	943.0	972.2	995.0
358.15	812.1	828.0	838.7	850.2	868.8	896.5	924.0	953.7	976.9
363.15	790.3	806.6	817.6	829.6	849.1	877.3	905.2	935.2	958.9
368.15	768.4	785.2	796.6	809.1	829.5	858.2	886.5	917.0	940.6
373.15	746.6	763.9	775.7	788.8	810.1	839.3	867.9	898.9	922.9

Hexane (1) + 1-Iodohexane (2)									
T/K	$u/m\cdot s^{-1}$ at $x_2 =$								
	0	0.05	0.1	0.2	0.3	0.5	0.7	0.9	1
293.15	1098.1	1086.8	1075.4	1061.6	1049.3	1046.2	1044.4	1042.2	1045.5
298.15	1076.0	1064.7	1054.3	1041.6	1030.6	1028.8	1028.7	1027.3	1031.3
303.15	1054.3	1042.7	1033.4	1021.7	1011.9	1011.5	1013.1	1012.4	1017.2
308.15	1031.8	1020.7	1012.5	1001.8	993.4	994.3	997.5	997.7	1003.2
313.15	1009.9	998.9	991.6	982.0	974.9	977.2	982.0	983.0	989.2
318.15	987.7	977.1	970.8	962.2	956.6	960.2	966.6	968.5	975.4
323.15	965.7	955.5	950.1	942.5	938.3	943.3	951.3	954.0	961.6
328.15	943.7	933.9	929.4	922.9	920.2	926.5	936.1	939.6	947.9
333.15	921.3	912.5	908.7	903.3	902.2	909.8	921.0	925.4	934.2
338.15	899.7	891.1	888.1	883.8	884.2	893.2	905.9	911.2	920.7
343.15	877.8	869.8	867.6	864.4	866.4	876.7	891.0	897.1	907.2
348.15	855.9	848.6	847.1	845.0	848.6	860.3	876.1	883.1	893.8
353.15	834.2	827.5	826.7	825.6	830.9	844.0	861.3	869.2	880.5
358.15	812.1	806.6	806.4	806.3	813.4	827.8	846.6	855.4	867.3
363.15	790.3	785.5	786.1	787.1	796.0	811.7	832.0	841.7	854.1
368.15	768.4	764.7	765.8	767.9	778.6	795.8	817.5	828.1	841.0
373.15	746.6	744.0	745.6	748.8	761.4	779.9	803.0	814.6	828.0

1-Chlorohexane (1) + 1-Iodohexane (2)							
T/K	$u/m\cdot s^{-1}$ at $x_2 =$						
	0	0.1	0.3	0.5	0.7	0.9	1
293.15	1219.1	1190.9	1144.7	1114.2	1082.2	1058.2	1045.5
298.15	1200.1	1172.6	1127.7	1097.5	1066.4	1043.3	1031.3
303.15	1181.9	1154.3	1110.7	1080.8	1050.8	1028.5	1017.2
308.15	1165.2	1136.2	1093.7	1064.4	1035.3	1013.8	1003.2
313.15	1143.7	1118.0	1076.8	1048.1	1019.8	999.3	989.2
318.15	1128.1	1100.0	1059.9	1031.9	1004.6	984.8	975.4
323.15	1106.0	1082.0	1043.1	1015.9	989.4	970.5	961.6
328.15	1087.0	1064.1	1026.3	1000.0	974.5	956.4	947.9
333.15	1068.8	1046.2	1009.5	984.3	959.6	942.3	934.2
338.15	1050.2	1028.4	992.8	968.7	944.8	928.4	920.7
343.15	1031.7	1010.6	976.1	953.3	930.2	914.6	907.2
348.15	1013.3	992.9	959.5	938.0	915.7	901.0	893.8
353.15	995.0	975.3	942.9	922.9	901.4	887.4	880.5
358.15	976.9	957.7	926.4	907.9	887.2	874.0	867.3
363.15	958.9	940.2	909.8	893.0	873.1	860.8	854.1
368.15	940.6	922.7	893.4	878.4	859.1	847.6	841.0
373.15	922.9	905.4	877.0	863.8	845.3	834.6	828.0

the oscilloscope screen. This helped to eliminate systematic error due to variation of the pulse shape. For the given procedure the sound propagation velocity in the investigated medium is determined by the simple relation

$$c = \frac{2L}{\tau_2 - \tau_1} \quad (1)$$

where L is the acoustic path and τ_2 and τ_1 are the total delay time of the second and the first pulses, respectively.

The measuring of speed of sound was fulfilled at the frequencies 2 and 5 MHz. A dispersion was not observed. The investigated liquids were located in a stainless steel autoclave in which were the measuring cell and platinum resistance thermometer. The measuring cell 3 consisted of

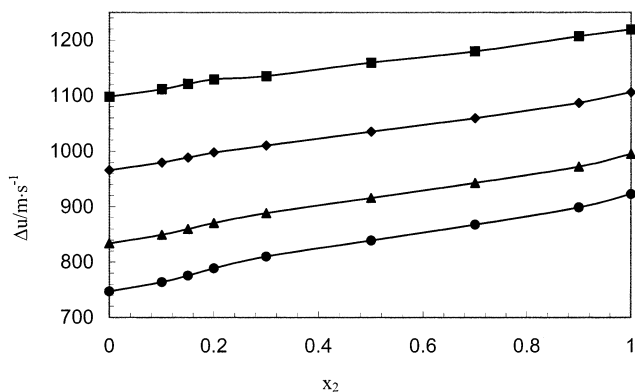


Figure 2. Speed of sound for *n*-hexane (1) + 1-chlorohexane (2): (■) 293.15 K; (◆) 323.15 K; (▲) 353.15 K; (●) 373.15 K.

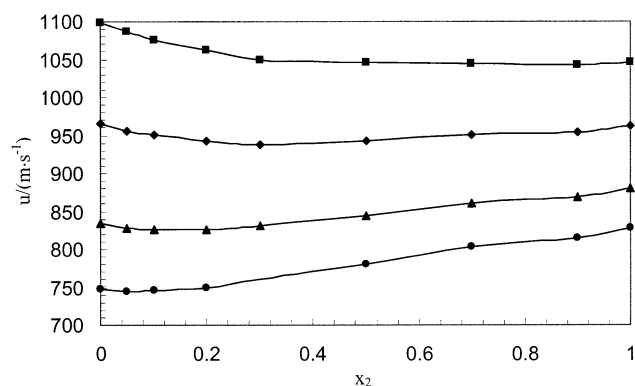


Figure 3. Speed of sound for *n*-hexane (1) + 1-iodohexane (2): (■) 293.15 K; (◆) 323.15 K; (▲) 353.15 K; (●) 373.15 K.

two transducer piezoceramics of 20 mm in diameter located at the fixed distance 21.234 ± 0.001 mm. The temperature change of the acoustic path was taken into account.

The speed of sound measuring cell was thermostated with a temperature stability of $\pm 10^{-2}$ K. The apparatus calibration was performed periodically. Double-distilled water, benzene, nonane, and octane were used for calibrating the speed of sound cell. Experimental values of sound speed for the pure liquids at different temperatures were compared with those found in the literature and were in fairly good agreement, as shown in Table 1.

Results and Discussion

The speeds of sound of the binary mixtures (*n*-hexane + 1-chlorohexane), (*n*-hexane + 1-iodohexane), and (1-chlorohexane + 1-iodohexane) are given in Table 2 respectively from 293.15 K to 373.15 K. The speeds of sound of these mixtures at 293.15 K, 323.15 K, 353.15 K, and 373.15 K are presented also as a function of mole fraction of 1-chlorohexane and 1-iodohexane in Figures 2–4. The speed of sound, u , values are fitted by the method of least squares using the polynomial

$$u = A_0 + A_1 T + A_2 T^2 \quad (2)$$

Here T is absolute temperature and A_0 , A_1 , and A_2 are the adjustable parameters. The parameters are presented in Table 3 along with the standard deviation σ , defined by

$$\sigma = \left[\sum_{i=1}^n (u_{\text{obs}} - u_{\text{cal}})^2 / (n - p) \right]^{1/2} \quad (3)$$

where u_{obs} and u_{cal} are the observed and calculated quanti-

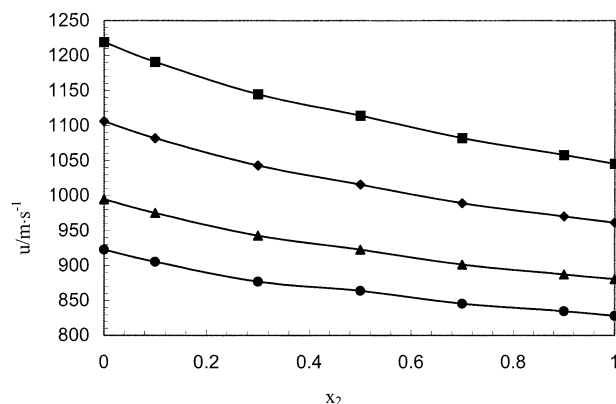


Figure 4. Speed of sound for 1-chlorohexane (1) + 1-iodohexane (2): (■) 293.15 K; (◆) 323.15 K; (▲) 353.15 K; (●) 373.15 K.

Table 3. Values of the Parameters of Eq 1 and Standard Deviation for Mixtures from 293.15 K to 373.15 K

x_2	A_1	A_2	A_3	$\sigma/\text{m}\cdot\text{s}^{-1}$
Hexane (1) + 1-Iodohehexane (2)				
0	2438	-4.707	4.690×10^{-4}	0.447
0.05	2543	-5.506	1.832×10^{-3}	0.126
0.1	2405	-4.862	1.110×10^{-3}	0.125
0.2	2336	-4.691	1.172×10^{-3}	0.093
0.3	2323	-4.930	1.998×10^{-3}	0.124
0.5	2245	-4.684	2.034×10^{-3}	0.072
0.7	2117	-4.164	1.720×10^{-3}	0.091
0.9	2084	-4.110	1.897×10^{-3}	0.085
1	2014	-3.762	1.566×10^{-3}	0.084
Hexane (1) + 1-Chlorohexane (2)				
0	2438	-4.707	4.690×10^{-4}	0.447
0.1	2500	-5.045	1.054×10^{-3}	0.156
0.15	2589	-5.548	1.874×10^{-3}	0.126
0.2	2648	-5.908	2.483×10^{-3}	0.111
0.3	2620	-5.833	2.633×10^{-3}	0.150
0.5	2642	-5.890	2.834×10^{-3}	0.131
0.7	2581	-5.472	2.365×10^{-3}	0.124
0.9	2663	-5.850	3.005×10^{-3}	0.155
1	2458	-4.621	1.356×10^{-3}	0.148
1-Chlorohexane (1) + 1-Iodohehexane (2)				
0	2458	-4.621	1.356×10^{-3}	0.148
0.1	2369	-4.374	1.208×10^{-3}	0.112
0.3	2217	-3.903	8.344×10^{-4}	0.097
0.5	2354	-5.095	2.949×10^{-3}	0.101
0.7	2236	-4.703	2.613×10^{-3}	0.138
0.9	2158	-4.500	2.558×10^{-3}	0.112
1	2014	-3.762	1.566×10^{-3}	0.084

ties as defined earlier, n is the total number of experimental points, and p is the number of parameters.

The changing of the sound speed by mixing, Δu , from a mole fraction was calculated by

$$\Delta u = u - (x_1 u_1 + x_2 u_2) \quad (4)$$

where x_1 and x_2 are the mole fractions and u_1 , u_2 , and u are the sound speeds of the pure components and the mixture, respectively. The experimental values of Δu at various temperatures are also reported in Table 4. The uncertainty in the changes Δu was estimated to be better than $\pm 2 \text{ m}\cdot\text{s}^{-1}$. The changing of the sound speed by mixing, Δu , was fitted by a Redlich–Kister type equation¹⁴

$$\Delta u/\text{m}\cdot\text{s}^{-1} = x_1(1 - x_1) \sum_{j=0}^k B_j (2x_2 - 1)^j \quad (5)$$

The coefficient B_j in eq 5 was estimated by the least-squares fit method. The values of the parameters B_j and the standard deviation $\sigma(\Delta u)$ are given in Table 5 at the

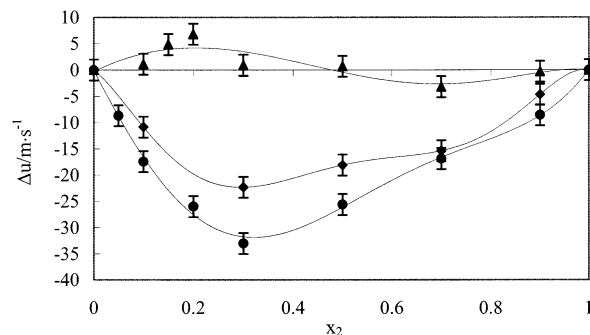
Table 4. Values of Δu for the Binary Mixtures at Various Temperatures

Hexane (1) + 1-Iodoheptane (2)							
<i>T</i> /K	$\Delta u/\text{m}\cdot\text{s}^{-1}$ at $x_2 =$						
	0.050	0.100	0.200	0.300	0.500	0.700	0.900
293.15	-8.6	-17.4	-25.9	-33.0	-25.6	-16.8	-8.5
298.15	-9.1	-17.2	-25.4	-31.9	-24.8	-16.0	-8.4
303.15	-9.7	-17.1	-25.1	-31.2	-24.2	-15.2	-8.5
308.15	-9.6	-16.4	-24.2	-29.8	-23.2	-14.2	-8.3
313.15	-9.9	-16.2	-23.7	-28.7	-22.3	-13.4	-8.2
318.15	-9.9	-15.6	-23.0	-27.4	-21.3	-12.4	-8.1
323.15	-9.9	-15.1	-22.3	-26.1	-20.3	-11.5	-8.0
328.15	-10.0	-14.7	-21.6	-24.7	-19.3	-10.5	-7.8
333.15	-9.4	-13.8	-20.5	-22.9	-17.9	-9.3	-7.5
338.15	-9.6	-13.7	-20.1	-21.8	-17.0	-8.5	-7.4
343.15	-9.4	-13.1	-19.2	-20.2	-15.8	-7.3	-7.1
348.15	-9.1	-12.5	-18.4	-18.6	-14.5	-6.3	-6.9
353.15	-9.0	-12.1	-17.8	-17.1	-13.3	-5.3	-6.6
358.15	-8.2	-11.2	-16.8	-15.2	-11.9	-4.1	-6.3
363.15	-7.9	-10.5	-15.9	-13.4	-10.5	-2.9	-6.0
368.15	-7.3	-9.8	-15.0	-11.5	-8.9	-1.7	-5.6
373.15	-6.6	-9.1	-14.0	-9.6	-7.4	-0.5	-5.2

Hexane (1) + 1-Chlorohexane (2)							
<i>T</i> /K	$u/\text{m}\cdot\text{s}^{-1}$ at $x_2 =$						
	0.100	0.150	0.200	0.300	0.500	0.700	0.900
293.15	1.1	4.9	6.8	0.9	0.7	-3.2	-0.3
298.15	0.8	4.3	6.1	1.7	0.3	-3.7	-1.3
303.15	0.1	3.3	5.0	1.7	-0.7	-4.6	-3.0
308.15	0.1	2.8	4.3	1.7	-1.9	-6.3	-5.8
313.15	-0.1	2.6	4.3	2.6	-0.9	-4.8	-4.2
318.15	-0.4	1.8	3.5	1.7	-2.5	-7.1	-7.7
323.15	-0.2	2.1	3.9	2.4	-0.8	-4.7	-5.3
328.15	-0.5	1.9	3.9	2.0	-0.5	-4.5	-5.4
333.15	-0.1	2.1	4.2	1.6	-0.3	-4.5	-6.0
338.15	-0.5	1.6	4.0	4.2	-0.2	-4.5	-6.3
343.15	-0.6	1.6	4.1	4.2	0.3	-4.3	-6.5
348.15	-0.5	1.6	4.3	5.6	0.8	-4.0	-6.6
353.15	-0.8	1.6	4.5	6.3	1.3	-3.8	-6.7
358.15	-0.6	1.9	5.1	7.3	2.0	-3.5	-6.7
363.15	-0.6	2.0	5.6	8.2	2.7	-3.1	-6.8
368.15	-0.4	2.4	6.3	9.4	3.7	-2.4	-6.4
373.15	-0.3	2.7	6.9	10.6	4.5	-2.1	-6.4

1-Chlorohexane (1) + 1-Iodoheptane (2)					
<i>T</i> /K	$\Delta u/\text{m}\cdot\text{s}^{-1}$ at $x_2 =$				
	0.100	0.300	0.500	0.700	0.900
293.15	-10.8	-22.3	-18.1	-15.4	-4.7
298.15	-10.6	-21.8	-18.2	-15.5	-4.9
303.15	-11.1	-21.8	-18.8	-15.8	-5.2
308.15	-12.8	-22.9	-19.8	-16.5	-5.6
313.15	-10.3	-20.6	-18.4	-15.8	-5.4
318.15	-12.8	-22.4	-19.8	-16.6	-5.9
323.15	-9.6	-19.6	-17.9	-15.5	-5.5
328.15	-9.0	-19.0	-17.5	-15.1	-5.4
333.15	-9.1	-18.9	-17.2	-15.0	-5.4
338.15	-8.8	-18.6	-16.8	-14.8	-5.3
343.15	-8.7	-18.2	-16.2	-14.3	-5.1
348.15	-8.5	-17.9	-15.6	-13.9	-4.8
353.15	-8.3	-17.8	-14.9	-13.4	-4.6
358.15	-8.2	-17.6	-14.2	-13.0	-4.3
363.15	-8.2	-17.7	-13.5	-12.4	-3.8
368.15	-7.9	-17.3	-12.4	-11.8	-3.4
373.15	-8.0	-17.4	-11.7	-11.2	-2.9

temperatures 293.15, 333.15, and 373.15 K, respectively. Calculation of the values $\sigma(\Delta u)$ was carried out with a ratio of the type in eq 3. The values of sound speed changing upon mixing, Δu , as a function of components x_2 of researched mixtures at 293.15 K are plotted in Figure 5. As shown in Figure 5, the composition dependence, $\Delta u(x)$, for the binary mixture hexane + 1-chlorohexane is S-shaped, showing a small positive part in the region rich in hexane ($0 < x_2 < 0.5$). The curve of the dependence $\Delta u(x)$

**Figure 5.** Changing of the sound speed, Δu , as a function of x_2 , of hexane (1) + 1-chlorohexane (2) (\blacktriangle); 1-chlorohexane (1) + 1-iodohexane (2) (\blacklozenge); and hexane (1) + 1-iodohexane (2) (\bullet) at 293.15 K.**Table 5. Parameters B_j of Eq 5 and Standard Deviation, $\sigma(\Delta u)$**

<i>T</i> /K	B_0	B_1	B_2	B_3	$\sigma/\text{m}\cdot\text{s}^{-1}$
Hexane (1) + 1-Chlorohexane (2)					
293.15	-1.894	-40.313	25.149	23.413	4.542
333.15	-0.021	-42.446	-37.004	-10.611	2.460
373.15	26.257	-91.034	-80.300	74.756	3.521
Hexane (1) + 1-Iodoheptane (2)					
293.15	-105.746	102.870	-56.230	-75.386	2.371
333.15	-67.374	84.719	-75.508	-55.294	2.446
373.15	-20.119	63.763	-88.110	-41.745	4.538
1-Chlorohexane (1) + 1-Iodoheptane (2)					
293.15	-79.311	40.774	-24.512	3.348	3.504
333.15	-73.251	22.388	-20.357	5.167	2.257
373.15	55.530	37.831	-25.796	-3.555	4.528

for the mixture hexane + 1-iodohexane shows negative Δu values. The maximum of the deviation is in the region of $x_2 = 0.3$. For the mixture 1-chlorohexane + 1-iodohexane the speed of sound change is too negative, but it is not far less than that of the preceding mixture. It seems reasonable to say that the magnitude of the value of Δu is defined by a distinction between the properties of the pure components.

It is interesting to note that the speed of sound in the binary mixture *n*-hexane + 1-chlorohexane at 293 K does not depend on the composition of 1-chlorohexane in the range from 1 to 0.3 mole fraction 1-chlorohexane. This fact, from our point of view, deserves special attention. With an increase of temperature, this characteristic singularity disappears. More detail would be possible when other properties of mixtures are known and an adequate model of the liquid is chosen.

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