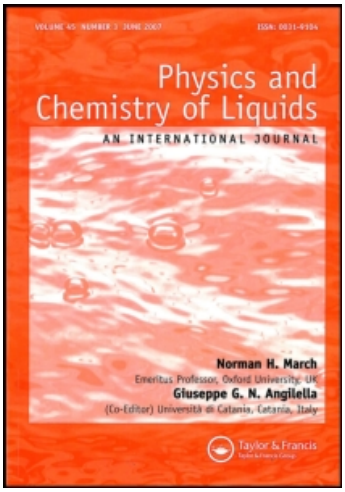


**Physics and Chemistry of Liquids**
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646857

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**To cite this Article** Choudary, N. V. , Kudchadker, A. P. and Naidu, P. R.(1986) 'Ultrasonic Behaviour of 1,1,2,2-Tetrachloroethane With An Alkane', Physics and Chemistry of Liquids, 16: 2, 137 — 146
**To link to this Article: DOI:** 10.1080/00319108608078511
**URL:** http://dx.doi.org/10.1080/00319108608078511

*Phys. Chem. Liq.*, 1986, Vol. 16, pp. 137-146
0031-9104/86/1602-0137$18.50/0

Printed in the United Kingdom

# Ultrasonic Behaviour of 1,1,2,2-Tetrachloroethane With An Alkane

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(*Received October 6, 1985*)



Experimental data for sound velocity and isentropic compressibility in binary mixtures of 1,1,2,2-tetrachloroethane with *n*-hexane, *n*-heptane, *n*-octane and *n*-nonane are reported at 303.15 and 313.15 K. Sound velocities have been analysed in terms of free length theory and collision factor theory. All the systems exhibited negative $\Delta K_s$ values.

## 1 INTRODUCTION

In continuation to our systematic study on excess thermodynamic properties and ultrasonic behaviour of binary mixtures of haloethane with a series of alkanes, alcohols and ketones,[1-9] we report here ultrasonic sound velocity and isentropic compressibility for the mixtures of 1,1,2,2-tetrachloroethane (hereafter referred to merely as TCE), with a series of *n*-alkanes at 303.15 and 313.15 K. The alkanes include *n*-hexane, *n*-heptane, *n*-octane and *n*-nonane. Sound velocity data have been analysed in terms of free length theory (FLT) of Jacobson,[10,11]

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extended to binary mixtures by Kaulgud[12–15] and collision factor theory (CFT) developed by Schaaffs[16,17] extended to binary mixtures by Nutsch-Kuhnkies[18] to test their applicability to the present systems. The molar sound velocity[19] ($R$), molar compressibility[20] ($W$), and van der Waals constant[21] ($b$) which give information on molecular interactions have also been computed.

## 2 EXPERIMENTAL SECTION

TCE and $n$-alkanes were purified by the methods described earlier.[9] The purity of the samples was checked by comparing the measured density of the samples with those reported in the literature.[22,23] Isentropic compressibilities were calculated from precise densities and sound speed data determined at 303.15 and 313.15 K. Density was computed from measured excess volume data reported elsewhere,[9] using the relation

$$\rho = \frac{x_1 M_1 + x_2 M_2}{V_m^i + V^E} \tag{1}$$

where $x_i$ and $M_i$ represent mole fraction and composition of component $i$ respectively. $V_m^i$ represents molar volume of ideal mixture and $V^E$ is the excess volume. Ultrasonic sound velocity was measured with a single crystal interferometer[24] at a frequency of 2 MHz and was accurate to $\pm 0.1\%$ leading to an accuracy of $\pm 1$ TPa$^{-1}$ in isentropic compressibility, $K_s$. All the measurements were made at a constant temperature employing a thermostat that could be maintained to $\pm 0.01$ K.

## 3 RESULTS AND DISCUSSION

### (a) Sound velocities

Theoretical aspects of FLT and CFT have been discussed in detail in earlier publications.[2,4] Procedures for the calculation of $R$, $W$ and $b$ have also been described earlier.[7] The surface area ($Y$) and collision factor ($S$) of the pure components used in FLT and CFT were calculated using the experimental sound velocity and density. Critical temperature, surface tension and the ratio of specific heats, which were used in the calculations of molar volume at absolute zero ($V_0$) and the average molecular radius ($r_m$), were taken from the literature.[22–26] The values of molar volume ($V_m$), molar volume at absolute zero ($V_0$),

TABLE I

Values of $V_m$, $V_0$, $V_a$, $Y$, $S$ and $r_m$ of the pure liquid components at 303.15 K

| Component | $V_m$ | $V_0$ | $V_a$ | $L_f$ | $Y$ | $S$ | $r_m$ |
|---|---|---|---|---|---|---|---|
| | $cm^3\ mol^{-1}$ | | | A° | | | A° |
| 1,1,2,2-tetrachloroethane | 106.334 | 88.460 | 17.874 | 0.4433 | 80.645 | 1.4227 | 2.758 |
| *n*-hexane | 132.454 | 100.807 | 31.647 | 0.7429 | 85.199 | 1.4601 | 2.871 |
| *n*-heptane | 148.411 | 115.914 | 32.497 | 0.6918 | 93.948 | 1.4861 | 3.017 |
| *n*-octane | 164.478 | 130.889 | 33.589 | 0.6596 | 101.854 | 1.4951 | 3.151 |
| *n*-nonane | 180.755 | 145.943 | 34.812 | 0.6311 | 110.327 | 1.5132 | 3.275 |

TABLE II

Values of $L_f^M$, $U$ exptl. $U$ Theo (FLT), $U$ Theo (CFT), $\Delta U\%$ (FLT) and $\Delta U\%$ (CFT), $R$, $W$ and $b$ for 1,1,2,2-tetrachloroethane with $n$-alkanes at 303.15 K

| | | U/m sec$^{-1}$ | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | Theoretical | | | | | | |
| $x_1$ | $L_f^M$ A° | Exptl. | FLT | CFT | $\Delta U\%$ (FLT) | $\Delta U\%$ (CFT) | $R$ | $W$ | $b$ cm$^3$·mol$^{-1}$ |
| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| | | | | TCE + $n$-hexane | | | | | |
| 0.0000 | 0.7429 | 1053 | 1053 | 1053 | 0.00 | 0.00 | 1348 | 2441 | 238.9 |
| 0.1515 | 0.6978 | 1045 | 1032 | 1063 | 1.24 | 1.77 | 1303 | 2418 | 234.8 |
| 0.2567 | 0.6661 | 1042 | 1026 | 1071 | 1.58 | 2.81 | 1274 | 2399 | 231.9 |
| 0.3451 | 0.6395 | 1042 | 1025 | 1077 | 1.71 | 3.46 | 1250 | 2383 | 229.6 |
| 0.4350 | 0.6123 | 1044 | 1027 | 1084 | 1.64 | 3.93 | 1227 | 2366 | 227.1 |
| 0.6141 | 0.5580 | 1058 | 1044 | 1099 | 1.34 | 3.92 | 1184 | 2333 | 222.3 |
| 0.7446 | 0.5190 | 1075 | 1065 | 1110 | 0.92 | 3.28 | 1156 | 2308 | 218.7 |
| 0.8090 | 0.4998 | 1087 | 1079 | 1116 | 0.75 | 2.65 | 1143 | 2300 | 217.0 |
| 0.9021 | 0.4722 | 1106 | 1103 | 1124 | 0.32 | 1.63 | 1125 | 2285 | 214.5 |
| 1.0000 | 0.4433 | 1133 | 1133 | 1133 | 0.00 | 0.00 | 1109 | 2271 | 211.8 |
| | | | Average % deviation | | 1.19 | 2.93 | | | |
| | | | | TCE + $n$-heptane | | | | | |
| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| 0.0000 | 0.6918 | 1110 | 1110 | 1110 | 0.00 | 0.00 | 1537 | 2791 | 227.2 |
| 0.1935 | 0.6516 | 1082 | 1078 | 1111 | 0.38 | 2.66 | 1441 | 2688 | 264.6 |
| 0.3773 | 0.6095 | 1070 | 1063 | 1113 | 0.63 | 4.04 | 1357 | 2669 | 252.5 |
| 0.4738 | 0.5859 | 1067 | 1062 | 1115 | 0.48 | 4.51 | 1314 | 2539 | 246.2 |
| 0.5807 | 0.5587 | 1069 | 1065 | 1118 | 0.37 | 4.55 | 1269 | 2483 | 239.2 |
| 0.6799 | 0.5329 | 1076 | 1072 | 1120 | 0.35 | 4.13 | 1228 | 2431 | 232.7 |
| 0.7442 | 0.5154 | 1082 | 1080 | 1122 | 0.18 | 3.74 | 1203 | 2397 | 228.5 |
| 0.8310 | 0.4914 | 1096 | 1094 | 1126 | 0.22 | 2.71 | 1170 | 2354 | 222.9 |
| 0.8951 | 0.4732 | 1107 | 1107 | 1128 | 0.01 | 1.93 | 1146 | 2322 | 218.7 |
| 1.0000 | 0.4433 | 1133 | 1133 | 1133 | 0.00 | 0.00 | 1109 | 2271 | 211.8 |
| | | | Average % deviation | | 0.33 | 3.53 | | | |

TCE + *n*-octane

| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
|---|---|---|---|---|---|---|---|---|---|
| 0.0000 | 0.6596 | 1148 | 1148 | 1148 | 0.00 | 0.00 | 1722 | 3136 | 315.8 |
| 0.2011 | 0.6290 | 1116 | 1110 | 1141 | 0.53 | 2.23 | 1586 | 2962 | 294.9 |
| 0.3081 | 0.6095 | 1104 | 1097 | 1138 | 0.62 | 3.09 | 1518 | 2869 | 283.8 |
| 0.4071 | 0.5904 | 1096 | 1089 | 1136 | 0.68 | 3.64 | 1455 | 2782 | 273.5 |
| 0.5082 | 0.5692 | 1091 | 1084 | 1134 | 0.66 | 3.94 | 1392 | 2694 | 263.0 |
| 0.7005 | 0.5247 | 1092 | 1087 | 1132 | 0.46 | 3.65 | 1277 | 2527 | 243.0 |
| 0.7816 | 0.5041 | 1097 | 1094 | 1131 | 0.29 | 3.15 | 1230 | 2456 | 234.5 |
| 0.8478 | 0.4866 | 1104 | 1102 | 1131 | 0.19 | 2.49 | 1192 | 2399 | 227.6 |
| 0.9096 | 0.4696 | 1113 | 1112 | 1132 | 0.07 | 1.70 | 1157 | 2347 | 221.2 |
| 1.0000 | 0.4433 | 1133 | 1133 | 1133 | 0.00 | 0.00 | 1109 | 2271 | 211.8 |
| | | | | Average % deviation | 0.44 | 2.99 | | | |



, TCE + *n*-nonane

| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
|---|---|---|---|---|---|---|---|---|---|
| 0.0000 | 0.6311 | 1187 | 1187 | 1187 | 0.00 | 0.00 | 1914 | 3490 | 354.6 |
| 0.1853 | 0.6103 | 1153 | 1149 | 1174 | 0.40 | 1.81 | 1754 | 3262 | 328.2 |
| 0.3160 | 0.5919 | 1134 | 1128 | 1166 | 0.52 | 2.77 | 1644 | 3104 | 309.5 |
| 0.4111 | 0.5767 | 1123 | 1117 | 1160 | 0.55 | 3.27 | 1565 | 2956 | 295.9 |
| 0.5396 | 0.5534 | 1112 | 1108 | 1152 | 0.45 | 3.65 | 1461 | 2849 | 277.5 |
| 0.7035 | 0.5191 | 1106 | 1104 | 1144 | 0.20 | 3.47 | 1331 | 2599 | 254.1 |
| 0.8000 | 0.4965 | 1107 | 1108 | 1140 | 0.09 | 3.00 | 1256 | 2516 | 240.3 |
| 0.8502 | 0.4839 | 1111 | 1112 | 1138 | 0.09 | 2.39 | 1219 | 2433 | 233.2 |
| 0.9153 | 0.4670 | 1116 | 1118 | 1135 | 0.24 | 1.76 | 1170 | 2368 | 223.9 |
| 1.0000 | 0.4433 | 1133 | 1133 | 1133 | 0.00 | 0.00 | 1109 | 2271 | 211.8 |
| | | | | Average % deviation | 0.36 | 2.77 | | | |

## TABLE III

Volume fraction $\phi_1$, of 1,1,2,2-tetrachloroethane, density $\rho$, speed of sound $U$, isentropic compressibility $K_s$, calculated from Eq. 2 and $K_s$ calculated from Eq. 3, for 1,1,2,2-tetrachloroethane + $n$-alkanes at 303.15 and 313.15 K.

| $\phi_1$ | $\rho$/g cm$^{-3}$ | U/ms$^{-1}$ | $K_s$/TPa$^{-1}$ | $\Delta K_s$/TPa$^{-1}$ | $\phi_1$ | $\rho$/g cm$^{-3}$ | U/ms$^{-1}$ | $K_s$/TPa$^{-1}$ | $\Delta K_s$/TPa$^{-1}$ |
|---|---|---|---|---|---|---|---|---|---|
| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| | | 303.15 K | | | | | 313.15 K | | |
| | | | | TCE + $n$-hexane | | | | | |
| 0.0000 | 0.65064 | 1053 | 1386 | 0 | 0.0000 | 0.64058 | 1012 | 1524 | 0 |
| 0.1254 | 0.76742 | 1045 | 1193 | −81 | 0.1381 | 0.76933 | 995 | 1313 | −73 |
| 0.2171 | 0.85291 | 1042 | 1080 | −112 | 0.2296 | 0.85459 | 992 | 1189 | −106 |
| 0.2973 | 0.92764 | 1042 | 993 | −128 | 0.2790 | 0.90064 | 993 | 1126 | −120 |
| 0.3820 | 1.00656 | 1044 | 911 | −134 | 0.3737 | 0.98868 | 998 | 1016 | −135 |
| 0.5609 | 1.17312 | 1058 | 762 | −124 | 0.4634 | 1.07194 | 1006 | 922 | −140 |
| 0.7006 | 1.30262 | 1075 | 664 | −97 | 0.5959 | 1.19464 | 1024 | 798 | −131 |
| 0.7727 | 1.36931 | 1087 | 618 | −79 | 0.7088 | 1.29870 | 1044 | 706 | −110 |
| 0.8809 | 1.46906 | 1106 | 556 | −44 | 0.7771 | 1.36161 | 1057 | 657 | −91 |
| 1.0000 | 1.57851 | 1133 | 494 | 0 | 1.0000 | 1.56619 | 1102 | 526 | 0 |
| | | | | TCE + $n$-heptane | | | | | |
| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| 0.0000 | 0.67522 | 1110 | 1202 | 0 | 0.0000 | 0.66678 | 1070 | 1310 | 0 |
| 0.1467 | 0.80713 | 1082 | 1058 | −40 | 0.1244 | 0.73716 | 1044 | 1179 | −33 |
| 0.2672 | 0.91577 | 1070 | 954 | −59 | 0.2239 | 0.77821 | 1032 | 1082 | −52 |
| 0.3921 | 1.02859 | 1067 | 854 | −70 | 0.2874 | 0.92477 | 1028 | 1023 | −62 |
| 0.4981 | 1.12437 | 1069 | 778 | −71 | 0.3663 | 0.99577 | 1029 | 948 | −75 |
| 0.6035 | 1.21965 | 1076 | 708 | −67 | 0.4562 | 1.07674 | 1032 | 872 | −80 |
| 0.6758 | 1.28519 | 1082 | 665 | −59 | 0.5977 | 1.20433 | 1044 | 766 | −75 |
| 0.7789 | 1.37854 | 1096 | 604 | −47 | 0.6974 | 1.29413 | 1049 | 702 | −61 |
| 0.8594 | 1.45151 | 1107 | 562 | −32 | 0.8545 | 1.43657 | 1071 | 607 | −33 |
| 1.0000 | 1.57851 | 1133 | 494 | 0 | 1.0000 | 1.56619 | 1102 | 526 | 0 |

TCE + *n*-octane

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| 0.0000 | 0.62450 | 1148 | 1093 | 0 | 0.0000 | 0.68639 | 1108 | 1187 | 0 |
| 0.1400 | 0.81684 | 1116 | 983 | −26 | 0.1479 | 0.81546 | 1074 | 1063 | −26 |
| 0.2235 | 0.89028 | 1104 | 922 | −37 | 0.2278 | 0.88530 | 1064 | 998 | −37 |
| 0.3074 | 0.96404 | 1096 | 864 | −45 | 0.2770 | 0.92830 | 1058 | 962 | −42 |
| 0.4005 | 1.04609 | 1091 | 803 | −50 | 0.4170 | 1.05100 | 1052 | 860 | −51 |
| 0.6019 | 1.22428 | 1092 | 685 | −47 | 0.4998 | 1.12378 | 1052 | 804 | −53 |
| 0.6982 | 1.30966 | 1097 | 635 | −40 | 0.6008 | 1.21268 | 1055 | 741 | −49 |
| 0.7827 | 1.38468 | 1104 | 593 | −31 | 0.7098 | 1.30893 | 1062 | 677 | −41 |
| 0.8668 | 1.45955 | 1113 | 553 | −21 | 0.8452 | 1.42870 | 1076 | 605 | −23 |
| 1.0000 | 1.57851 | 1133 | 494 | 0 | 1.0000 | 1.56619 | 1102 | 526 | 0 |

TCE + *n*-nonane

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| 0.0000 | 0.70958 | 1187 | 1000 | 0 | 0.0000 | 0.70212 | 1148 | 1081 | 0 |
| 0.1181 | 0.81092 | 1153 | 930 | −12 | 0.1375 | 0.81919 | 1109 | 993 | −12 |
| 0.2138 | 0.89320 | 1134 | 871 | −21 | 0.2283 | 0.89688 | 1093 | 933 | −21 |
| 0.2912 | 0.95999 | 1123 | 826 | −27 | 0.2608 | 0.92474 | 1088 | 914 | −22 |
| 0.4082 | 1.06127 | 1112 | 762 | −31 | 0.4157 | 1.05804 | 1075 | 818 | −32 |
| 0.5827 | 1.21302 | 1106 | 674 | −31 | 0.4897 | 1.12181 | 1073 | 774 | −35 |
| 0.7019 | 1.31703 | 1107 | 616 | −25 | 0.5775 | 1.19777 | 1071 | 728 | −32 |
| 0.7696 | 1.37630 | 1111 | 591 | −22 | 0.7277 | 1.32806 | 1074 | 653 | −24 |
| 0.8641 | 1.45908 | 1116 | 550 | −13 | 0.7658 | 1.36128 | 1076 | 635 | −21 |
| 1.0000 | 1.57851 | 1133 | 494 | 0 | 1.0000 | 1.56619 | 1102 | 526 | 0 |

available volume ($V_a$), free length ($L_f$), surface area ($Y$), collision factor ($S$) and the average radius of the molecules ($r_m$) of the pure components are given in Table I. The free lengths of the binary mixtures and sound velocity data predicted in terms of FLT and CFT are given in Table II along with experimental results in columns 2 to 5. The percentage deviation between the experimental sound velocity data and those of the FLT and CFT values are given in columns 6 and 7 along with average percentage deviation. Values of $R$, $W$ and $b$ are also given in columns 8-10 of Table II.

The results given in columns 3-7 of Table II reveal that the FLT sound velocities are in excellent agreement with the experimental results in all the systems. The maximum average percentage deviation in any system does not exceed 0.44% (except in case of *n*-hexane). The maximum individual deviation is 1.71%. However, the sound velocities estimated by CFT are not in good agreement with the experimental results. The average percentage deviation varies from 2.77 to 3.53. The maximum individual deviation in case of CFT is 4.55%. Venkateswarlu *et al.*[27] have also reported similar observations in their recent publication.

The data included in columns 8 to 10 of Table II show that the values of $R$, $W$ and $b$ exhibit linear relationship with mole fraction in all the systems studied as expected. This in turn shows the absence of strong interactions between unlike molecules.

### (b) Isentropic compressibility

Isentropic compressibility ($K_s$) was calculated using the relation

$$K_s = U^{-2}\rho^{-1} \tag{2}$$

where $U$ and $\rho$ denote the sound velocity and density. The deviation in $K_s$ from ideal behaviour, assumed to be additive in terms of volume fraction, is estimated using the equation

$$\Delta K_s = K_s - \phi K_{s1} - (1 - \phi)K_{s2} \tag{3}$$

where $K_{s1}$ and $K_{s2}$ are the isentropic compressibilities of the pure components 1 and 2 respectively. $\phi$ is the volume fraction of TCE.

Experimental data for $\rho$, $U$, $K_s$ and $\Delta K_s$ at 303.15 and 313.15 K are included in Table III. The dependence of $\Delta K_s$ on volume fraction is expressed by an empirical equation of the form

$$\Delta K_s = \phi(1 - k)\left\{\sum_{j=0}^{n-1} b_j(1 - 2\phi)^j\right\} \tag{4}$$

TABLE IV

Standard deviation $\sigma$ ($\Delta K_s$) and values of parameters $b_j$ for the systems of TCE + *n*-alkanes at 303.15 and 313.15 K

| System | $b_0$ | $b_1$ | $b_2$ | $b_3$ | $\sigma(\Delta K_s)$/ TPa$^{-1}$ |
|---|---|---|---|---|---|
| | | Temp. 303.15 K | | | |
| TCE + *n*-hexane | −521.0 | 166.9 | −103.6 | 78.1 | 0.6 |
| TCE + *n*-heptane | −284.0 | 36.1 | −16.8 | — | 0.7 |
| TCE + *n*-octane | −201.8 | 30.7 | 7.2 | −13.5 | 0.3 |
| TCE + *n*-nonane | −130.1 | 4.9 | 28.2 | — | 0.7 |
| | | Temp. 313.15 K | | | |
| TCE + *n*-hexane | −558.0 | 76.5 | −19.7 | −27.1 | 0.4 |
| TCE + *n*-heptane | −315.0 | 23.8 | 66.7 | — | 1.7 |
| TCE + *n*-octane | −210.4 | 11.1 | 39.1 | 23.8 | 0.5 |
| TCE + *n*-nonane | −134.9 | −0.4 | 64.9 | — | 0.9 |



The values of $b_j$ calculated by the method of least squares are given in Table IV along with the standard deviation $\sigma$ ($\Delta K_s$).

The results included in Table III show that $\Delta K_s$ is negative over the whole range of composition in all the mixtures at both the temperatures. Such an observation is not common in nonpolar systems. The negative values of $\Delta K_s$ for the four systems fall in the order: *n*-hexane > *n*-heptane > *n*-octane > *n*-nonane. The temperature coefficient of $\Delta K_s$ is negative in all the systems.

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