

# INFLUENCE OF TEMPERATURE ON ULTRASONIC VELOCITY MEASUREMENTS OF ETHANOL+WATER+1-PROPANOL MIXTURES

J. M. Resa<sup>1\*</sup>, C. González<sup>1</sup>, J. M. Goenaga<sup>1</sup> and M. Iglesias<sup>2</sup>

<sup>1</sup>Departamento de Ingeniería Química, Universidad del País Vasco, Apto. 450, Vitoria, Spain

<sup>2</sup>Departament d'Enginyeria Química, Escola Tècnica Superior d'Enginyeria Química, Universitat Rovira i Virgili, Avinguda Paisos Catalans 26, Campus Sescelades, 43007 Tarragona, Spain

The ultrasonic velocity of the ternary mixtures ethanol+water+1-propanol at the range 288.15–323.15 K and atmospheric pressure, has been measured over the whole concentration range. The corresponding change of isentropic compressibility was computed from the experimental data. The results were fitted by means of a temperature dependent equation, such parameters being gathered. The obtained experimental values indicate varying extent of interstitial accommodation among unlike molecules as a function of steric hindrance attending to 1-propanol composition as key component and as a function of hydrogen bond and temperature attending to ethanol composition as key component.

**Keywords:** derived magnitude, estimation, ethanol+water+1-propanol, isentropic compressibilities, mixtures, ultrasonic velocities

## Introduction

Knowledge of thermodynamic properties and phase equilibria of ethanol, water and the different flavour components in distilled alcoholic beverages is of practical interest to the food industry since industrial procedures applied are close related on their temperature and pressure dependence in order to obtain a quality final product. Thermodynamics studies provide the additional advantage of an interesting trend of analysis of microscale interaction for understanding macroscale behaviour of gases and liquids on mixing. In accordance to that, a considerable effort has been developed in the field of thermodynamic properties although a great scarce of data is observed in open literature for mixtures enclosed into commercial alcoholic beverages, this fact is described in [1].

## Experimental

### Materials

All chemical solvents used in the preparation of samples were of Panreac quality with richness better than

99.5 mol%. The pure components were stored in sun light protected form and constant humidity and temperature. In order to reduce fraction molar errors, the vapour space into the vessels was minimized during samples preparation. Mixtures were prepared by mass using a Salter ER-182A balance, the whole composition range of the ternary mixture being covered. The accuracy in molar fractions was obtained as higher than  $\pm 5 \cdot 10^{-4}$ .

### Methods

The ultrasonic velocities were measured with an Anton Paar DSA-48 device with a precision of  $\pm 1 \text{ m s}^{-1}$ . Calibration of the apparatus was performed periodically, in accordance with technical specifications, using Millipore quality water (resistivity, 18.2 M $\Omega$  cm) and ambient air. Maximum deviation in the calculation of changes of isentropic compressibility for these mixtures have been estimated better than 1 TPa<sup>-1</sup>. The values of the pure components, as well as, open literature data are enclosed into Table 1. More details about techniques and procedure in our

**Table 1** Comparison of experimental speed of sound ( $\text{m s}^{-1}$ ) with literature data for chemicals at the studied temperatures in K

Component	Molecular mass	288.15	293.15	298.15	303.15	308.15	313.15	318.15	323.15	lit. (298.15)
Ethanol	46.1	1178.2	1160.3	1143.1	1126.2	1109.4	1092.7	1075.9	1058.8	1143 <sup>a</sup>
Water	18.0	1466.4	1482.5	1496.9	1509.5	1520.3	1529.3	1536.6	1542.1	1497 <sup>a</sup>
1-propanol	60.096	1238.8	1221.0	1203.6	1186.6	1169.7	1152.9	1136.0	1118.9	n. a.

<sup>a</sup> 7, n.a. – not available

\* Author for correspondence: iqpredij@vc.ehu.es

**Table 2** Ultrasonic velocities, isentropic compressibilities and change of isentropic compressibilities for ternary mixture at range of 288.15–323.15 K

$x_1$	$x_2$	$u/\text{m s}^{-1}$	$\kappa_s/\text{TPa}^{-1}$	$\delta\kappa_s/\text{TPa}^{-1}$	$x_1$	$x_2$	$u/\text{m s}^{-1}$	$\kappa_s/\text{TPa}^{-1}$	$\delta\kappa_s/\text{TPa}^{-1}$
323.15 K									
0.9049	0.0452	1085.9	1102.3	-24.6	0.3003	0.2987	1180.4	884.6	-4.2
0.0501	0.9004	1568.8	426.1	-66.3	0.3006	0.3983	1231.0	796.7	-32.5
0.0514	0.0480	1053.9	1165.6	161.8	0.2975	0.5029	1283.8	716.5	-49.5
0.7990	0.1021	1091.3	1082.3	4.7	0.2993	0.6012	1333.5	650.7	-56.5
0.7027	0.0970	1088.8	1086.7	19.7	0.2010	0.1027	1082.1	1094.5	102.2
0.6889	0.2004	1128.6	994.3	-10.2	0.1997	0.2021	1131.4	980.7	48.3
0.5987	0.3005	1167.2	912.2	-18.0	0.1994	0.3028	1185.1	874.4	2.4
0.6018	0.0978	1087.0	1089.4	37.3	0.1999	0.3990	1239.5	782.3	-32.1
0.4989	0.4009	1213.5	826.9	-28.9	0.2002	0.4995	1296.9	698.6	-55.6
0.4973	0.3031	1173.0	901.0	-13.2	0.1990	0.6006	1356.3	624.4	-69.0
0.4981	0.2020	1131.6	985.7	10.8	0.1995	0.7008	1416.5	558.5	-74.8
0.5003	0.0982	1087.1	1087.8	50.3	0.0988	0.0960	1083.1	1090.9	109.2
0.3994	0.1010	1084.4	1093.0	71.5	0.0984	0.2008	1139.2	964.9	46.0
0.4008	0.1979	1131.2	984.7	21.1	0.1005	0.2991	1191.2	862.8	2.6
0.4001	0.3006	1167.9	909.4	7.5	0.1007	0.3994	1250.3	764.9	-35.1
0.3989	0.3989	1220.0	814.0	-28.7	0.1002	0.4987	1308.2	682.3	-58.2
0.3991	0.5004	1269.0	738.0	-44.0	0.1001	0.5989	1378.8	599.4	-81.0
0.2991	0.1003	1085.5	1088.5	80.9	0.0994	0.7016	1440.9	535.2	-83.4
0.3006	0.1987	1132.1	981.5	32.7	0.1006	0.7993	1512.5	473.1	-87.1
318.15 K									
0.9049	0.0452	1103.0	1062.2	-23.7	0.3003	0.2987	1197.3	855.1	-7.0
0.0501	0.9004	1575.6	421.2	-69.2	0.3006	0.3983	1247.5	771.5	-34.7
0.0514	0.0480	1069.9	1124.2	154.8	0.2975	0.5029	1300.2	694.9	-52.1
0.7990	0.1021	1108.3	1043.3	3.6	0.2993	0.6012	1349.4	632.2	-59.8
0.7027	0.0970	1106.0	1047.2	17.6	0.2010	0.1027	1098.3	1056.1	97.4
0.6989	0.2004	1145.9	958.9	-12.1	0.1997	0.2021	1148.1	947.0	44.2
0.5987	0.3005	1184.5	880.7	-20.6	0.1994	0.3028	1201.6	845.9	-0.3
0.6018	0.0978	1104.1	1049.8	34.3	0.1999	0.3990	1255.7	758.1	-34.1
0.4989	0.4009	1230.4	799.9	-31.6	0.2002	0.4995	1313.0	678.0	-57.8

Table 2 Continued

$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_s/TPa^{-1}$	$\delta\kappa_s/TPa^{-1}$	$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_s/TPa^{-1}$	$\delta\kappa_s/TPa^{-1}$
0.4973	0.3031	1190.0	870.4	-15.7	0.1990	0.6006	1372.1	607.1	-71.7
0.4981	0.2020	1148.6	951.2	8.2	0.1995	0.7008	1431.6	544.3	-78.4
0.5003	0.0982	1104.1	1048.3	46.7	0.0988	0.0960	1099.0	1053.3	104.6
0.3994	0.1010	1101.1	1053.8	67.3	0.0984	0.2008	1155.2	933.0	43.2
0.4008	0.1979	1148.0	950.5	18.3	0.1005	0.2991	1207.1	835.5	0.6
0.4001	0.3006	1184.7	878.7	4.3	0.1007	0.3994	1266.5	741.6	-37.0
0.3989	0.3989	1237.5	787.8	-31.3	0.1002	0.4987	1324.2	662.6	-60.2
0.3991	0.5004	1285.4	715.4	-46.7	0.1001	0.5989	1393.7	583.8	-82.7
0.2991	0.1003	1102.1	1049.7	76.4	0.0994	0.7016	1455.3	522.4	-86.3
0.3006	0.1987	1148.6	948.1	29.8	0.1006	0.7993	1525.8	463.1	-90.9
313.15 K									
0.0949	0.0452	1119.7	1024.9	-22.8	0.3003	0.2987	1213.8	827.5	-9.8
0.0501	0.9004	1581.5	416.9	-72.7	0.3006	0.3983	1263.6	748.0	-37.2
0.0514	0.0480	1086.0	1084.5	147.5	0.2975	0.5029	1316.2	674.7	-55.1
0.7990	0.1021	1125.2	1006.4	2.1	0.2993	0.6012	1364.9	615.0	-63.5
0.7027	0.0970	1122.8	1010.2	15.5	0.2010	0.1027	1114.5	1019.8	-92.3
0.6989	0.2004	1162.9	925.8	-14.2	0.1997	0.2021	1164.5	915.3	40.2
0.5987	0.3005	1201.5	851.3	-23.3	0.1994	0.3028	1217.7	819.1	-3.1
0.6018	0.0978	1121.0	1012.7	31.4	0.1999	0.3990	1271.8	735.2	-36.6
0.4989	0.4009	1247.1	774.4	-34.8	0.2002	0.4995	1328.9	658.6	-60.6
0.4973	0.3031	1206.8	841.7	-18.6	0.1990	0.6006	1387.0	591.3	-74.7
0.4981	0.2020	1165.4	918.9	5.5	0.1995	0.7008	1446.3	530.8	-82.6
0.5003	0.0982	1120.9	1011.4	43.3	0.0988	0.0960	1115.4	1016.8	98.9
0.3994	0.1010	1117.6	1017.2	63.4	0.0984	0.2008	1170.8	903.3	40.5
0.4008	0.1979	1164.7	918.4	15.3	0.1005	0.2991	1222.9	809.8	-1.8
0.4001	0.3006	1201.5	849.7	0.6	0.1007	0.3994	1282.0	720.1	-38.9
0.3989	0.3989	1254.0	763.1	-34.3	0.1002	0.4987	1339.8	644.0	-62.7
0.3991	0.5004	1301.6	694.1	-50.1	0.1001	0.5989	1408.3	569.1	-85.1
0.2991	0.1003	1118.6	1013.1	71.8	0.0994	0.7016	1469.2	510.4	-89.8
0.3006	0.1987	1164.8	916.8	26.9	0.1006	0.7993	1538.3	453.9	-95.2

Table 2 Continued

$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_s/TPa^{-1}$	$\delta\kappa_s/TPa^{-1}$	$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_s/TPa^{-1}$	$\delta\kappa_s/TPa^{-1}$
308.15 K									
0.9049	0.0452	1136.1	989.8	-21.9	0.3003	0.2987	1230.3	801.0	-13.3
0.0501	0.9004	1586.4	413.2	-76.8	0.3006	0.3983	1279.7	725.4	-40.3
0.0514	0.0480	1102.0	1047.1	140.7	0.2975	0.5029	1331.7	655.7	-58.5
0.7990	0.1021	1142.0	971.6	0.6	0.2993	0.6012	1379.5	599.0	-67.4
0.7027	0.0970	1139.6	975.1	13.3	0.2010	0.1027	1130.8	984.0	87.0
0.6989	0.2004	1179.8	894.4	-16.5	0.1997	0.2021	1180.6	885.6	36.4
0.5987	0.3005	1218.3	823.5	-26.3	0.1994	0.3028	1233.7	793.8	-6.2
0.6018	0.0978	1137.7	977.5	28.4	0.1999	0.3990	1287.4	713.7	-39.3
0.4989	0.4009	1263.5	750.4	-38.2	0.2002	0.4995	1344.3	640.4	-63.6
0.4973	0.3031	1223.4	814.6	-21.5	0.1990	0.6006	1401.8	576.1	-78.4
0.4981	0.2020	1181.9	888.4	2.8	0.1995	0.7008	1460.7	518.1	-87.5
0.5003	0.0982	1137.6	976.4	39.9	0.0988	0.0960	1131.5	982.3	93.6
0.3994	0.1010	1133.9	982.4	59.6	0.0984	0.2008	1186.1	875.2	37.7
0.4008	0.1979	1181.1	888.0	12.2	0.1005	0.2991	1238.3	785.5	-4.2
0.4001	0.3006	1218.2	822.1	-3.4	0.1007	0.3994	1297.2	699.6	-41.1
0.3989	0.3989	1270.4	739.6	-37.8	0.1002	0.4987	1355.4	626.2	-66.0
0.3991	0.5004	1317.3	674.1	-53.7	0.1001	0.5989	1422.4	555.4	-87.9
0.2991	0.1003	1134.9	978.6	67.6	0.0994	0.7016	1482.8	498.9	-94.2
0.3006	0.1987	1180.9	886.9	23.8	0.1006	0.7993	1550.4	445.1	-100.4
303.15 K									
0.9049	0.0452	1151.9	957.5	-19.8	0.3003	0.2987	1246.5	776.2	-16.4
0.0501	0.9004	1590.2	410.2	-81.4	0.3006	0.3983	1295.5	704.2	-43.4
0.0514	0.0480	1117.9	1011.4	134.3	0.2975	0.5029	1347.4	637.3	-62.6
0.7990	0.1021	1158.8	938.4	-0.9	0.2993	0.6012	1394.2	583.6	-72.1
0.7027	0.0970	1156.5	941.5	11.1	0.2010	0.1027	1147.0	951.7	82.0
0.6989	0.2004	1196.8	864.5	-18.8	0.1997	0.2021	1196.6	857.3	32.6
0.5987	0.3005	1235.0	797.0	-29.4	0.1994	0.3028	1249.6	769.6	-9.5
0.6018	0.0978	1154.5	944.0	25.6	0.1999	0.3990	1303.0	693.2	-42.4

Table 2 Continued

$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_S/TPa^{-1}$	$\delta\kappa_S/TPa^{-1}$	$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_S/TPa^{-1}$	$\delta\kappa_S/TPa^{-1}$
0.4989	0.4009	1279.7	727.7	-41.8	0.2002	0.4995	1359.5	623.1	-67.1
0.4973	0.3031	1239.9	788.7	-24.7	0.1990	0.6006	1416.2	561.8	-82.6
0.4981	0.2020	1198.5	859.2	0.0	0.1995	0.7008	1474.8	506.0	-93.1
0.5003	0.0982	1154.3	943.0	36.6	0.0988	0.0960	1147.6	949.5	88.6
0.3994	0.1010	1150.3	949.3	55.9	0.0984	0.2008	1200.6	849.5	36.0
0.4008	0.1979	1197.6	858.9	9.1	0.1005	0.2991	1253.6	762.4	-6.9
0.4001	0.3006	1234.8	795.8	-7.5	0.1007	0.3994	1312.1	680.4	-43.5
0.3989	0.3989	1286.6	717.3	-41.4	0.1002	0.4987	1370.4	609.7	-69.4
0.3991	0.5004	1332.9	655.1	-57.8	0.1001	0.5989	1436.1	542.4	-91.3
0.2991	0.1003	1151.2	945.6	63.5	0.0994	0.7016	1496.1	488.0	-99.2
0.3006	0.1987	1197.1	858.4	20.5	0.1006	0.7993	1562.2	436.9	-106.3
298.15 K									
0.9049	0.0452	1165.9	929.5	-15.0	0.3003	0.2987	1262.6	752.6	-19.7
0.0501	0.9004	1593.2	407.7	-86.8	0.3006	0.3983	1311.3	683.8	-47.1
0.0514	0.0480	1133.9	977.4	128.3	0.2975	0.5029	1362.9	619.9	-67.3
0.7990	0.1021	1175.6	906.7	-2.4	0.2993	0.6012	1408.8	568.9	-77.6
0.7027	0.0970	1173.3	909.8	9.1	0.2010	0.1027	1163.3	920.1	77.2
0.6989	0.2004	1213.8	835.9	-21.3	0.1997	0.2021	1217.7	830.2	28.8
0.5987	0.3005	1251.7	771.7	-32.9	0.1994	0.3028	1265.5	746.5	-13.0
0.6018	0.0978	1171.3	912.1	22.9	0.1999	0.3990	1318.5	673.6	-46.0
0.4989	0.4009	1295.9	705.9	-45.8	0.2002	0.4995	1374.3	606.8	-71.0
0.4973	0.3031	1256.3	764.2	-28.1	0.1990	0.6006	1430.4	548.1	-87.5
0.4981	0.2020	1215.0	831.6	-2.8	0.1995	0.7008	1488.8	494.4	-99.7
0.5003	0.0982	1171.0	911.2	33.5	0.0988	0.0960	1163.2	919.0	84.6
0.3994	0.1010	1166.8	917.6	52.1	0.0984	0.2008	1215.5	824.3	33.6
0.4008	0.1979	1214.2	831.2	5.8	0.1005	0.2991	1268.9	740.3	-9.8
0.4001	0.3006	1251.3	770.8	-11.7	0.1007	0.3994	1327.1	661.9	-46.6

Table 2 Continued

$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_S/TPa^{-1}$	$\delta\kappa_S/TPa^{-1}$	$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_S/TPa^{-1}$	$\delta\kappa_S/TPa^{-1}$
0.3989	0.3989	1302.5	696.2	-45.4	0.1002	0.4987	1385.6	593.6	-73.5
0.3991	0.5004	1348.3	637.1	-62.3	0.1001	0.5989	1449.7	530.0	-95.5
0.2991	0.1003	1167.6	914.1	59.4	0.0994	0.7016	1509.3	477.6	-105.0
0.3006	0.1987	1213.2	831.1	17.2	0.1006	0.7993	1573.6	429.1	-113.1
293.15 K									
0.9049	0.0452	1181.0	901.0	-11.8	0.3003	0.2987	1279.0	729.6	-23.6
0.0501	0.9004	1595.0	405.9	-92.9	0.3006	0.3983	1327.6	663.8	-51.6
0.0514	0.0480	1150.0	944.9	122.9	0.2975	0.5029	1378.3	603.1	-72.3
0.7990	0.1021	1192.8	875.9	-4.2	0.2993	0.6012	1424.2	554.1	-84.3
0.7027	0.0970	1190.5	878.9	7.0	0.2010	0.1027	1180.0	889.3	72.3
0.6989	0.2004	1231.1	808.2	-24.1	0.1997	0.2021	1229.2	803.8	24.6
0.5987	0.3005	1268.8	747.1	-36.7	0.1994	0.3028	1282.0	723.7	-17.3
0.6018	0.0978	1188.5	881.1	20.1	0.1999	0.3990	1334.3	654.5	-50.1
0.4989	0.4009	1312.4	684.7	-50.6	0.2002	0.4995	1389.5	590.8	-75.7
0.4973	0.3031	1273.1	740.3	-32.0	0.1990	0.6006	1444.9	534.8	-93.3
0.4981	0.2020	1231.7	804.9	-5.7	0.1995	0.7008	1503.5	482.8	-107.4
0.5003	0.0982	1187.9	880.7	30.5	0.0988	0.0960	1178.6	890.3	81.5
0.3994	0.1010	1183.5	886.8	48.3	0.0984	0.2008	1230.3	800.3	31.3
0.4008	0.1979	1231.0	804.2	2.3	0.1005	0.2991	1284.5	718.8	-13.2
0.4001	0.3006	1268.2	746.5	-16.4	0.1007	0.3994	1342.4	643.8	-50.2
0.3989	0.3989	1318.8	675.7	-49.9	0.1002	0.4987	1401.1	577.9	-78.4
0.3991	0.5004	1364.0	619.5	-67.6	0.1001	0.5989	1463.4	517.8	-100.5
0.2991	0.1003	1184.1	883.9	55.7	0.0994	0.7016	1523.0	467.3	-112.1
0.3006	0.1987	1229.8	804.6	13.6	0.1006	0.7993	1585.5	421.2	-121.2
288.15 K									
0.9049	0.0452	1197.1	871.5	-10.4	0.3003	0.2987	1295.3	707.2	-27.8
0.0501	0.9004	1595.6	404.6	-99.9	0.3006	0.3983	1343.8	644.2	-56.8
0.0514	0.0480	1165.4	914.1	118.3	0.2975	0.5029	1393.3	587.0	-77.9
0.7990	0.1021	1210.1	846.0	-5.9	0.2993	0.6012	1439.4	539.6	-91.9

Table 2 Continued

$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_s/TPa^{-1}$	$\delta\kappa_s/TPa^{-1}$	$x_1$	$x_2$	$u/m\ s^{-1}$	$\kappa_s/TPa^{-1}$	$\delta\kappa_s/TPa^{-1}$
0.7027	0.0970	1207.5	849.0	5.0	0.2010	0.1027	1196.6	859.4	67.4
0.6989	0.2004	1248.4	781.1	-27.2	0.1997	0.2021	1245.7	777.8	19.9
0.5987	0.3005	1285.7	723.2	-40.9	0.1994	0.3028	1298.6	701.2	-22.3
0.6018	0.0978	1205.5	851.1	17.4	0.1999	0.3990	1350.7	635.1	-55.6
0.4989	0.4009	1328.9	664.0	-55.8	0.2002	0.4995	1404.4	575.2	-81.2
0.4973	0.3031	1289.9	716.8	-36.3	0.1990	0.6006	1459.4	521.6	-100.2
0.4981	0.2020	1248.3	778.8	-8.9	0.1995	0.7008	1518.9	470.7	-116.9
0.5003	0.0982	1204.7	850.9	27.5	0.0988	0.0960	1193.5	862.7	78.6
0.3994	0.1010	1200.2	857.0	44.6	0.0984	0.2008	1245.1	776.7	28.4
0.4008	0.1979	1247.9	777.8	-1.6	0.1005	0.2991	1300.0	697.7	-17.2
0.4001	0.3006	1285.3	722.5	-21.7	0.1007	0.3994	1357.5	626.0	-54.7
0.3989	0.3989	1335.1	655.5	-55.1	0.1002	0.4987	1416.7	562.3	-84.4
0.3991	0.5004	1379.7	602.1	-73.8	0.1001	0.5989	1476.9	505.9	-106.5
0.2991	0.1003	1200.2	855.1	52.5	0.0994	0.7016	1539.5	455.2	-122.1
0.3006	0.1987	1246.4	778.5	9.3	0.1006	0.7993	1598.0	413.0	-131.1

**Table 3** Parameters of Eq. (3) in the range 288.15–323.15 K

Ethanol+Water		
$B_{00} = -5621.928$	$B_{01} = 32.53$	$B_{02} = -0.048$
$B_{10} = 2240.750$	$B_{11} = -11.282$	$B_{12} = 0.015$
$B_{20} = -5654.858$	$B_{21} = 29.884$	$B_{22} = -0.042$
$B_{30} = 18652.408$	$B_{31} = -110.576$	$B_{32} = 0.168$
$B_{40} = 18652.408$	$B_{41} = -110.576$	$B_{42} = 0.168$
Water+1-propanol		
$B_{00} = -4402.688$	$B_{01} = 23.338$	$B_{02} = -0.027$
$B_{10} = 3027.912$	$B_{11} = -18.507$	$B_{12} = 0.026$
$B_{20} = 2369.873$	$B_{21} = -14.187$	$B_{22} = 0.023$
$B_{30} = 17979.324$	$B_{31} = -104.762$	$B_{32} = 0.157$
$B_{40} = -26361.265$	$B_{41} = 152.481$	$B_{42} = -0.229$
Ethanol+1-propanol		
$B_{00} = -109.503$	$B_{01} = 0.930$	$B_{02} = -0.002$
$B_{10} = -247.501$	$B_{11} = 1.649$	$B_{12} = -0.002$
$B_{20} = -781.068$	$B_{21} = 4.994$	$B_{22} = -0.008$
$B_{30} = 775.317$	$B_{31} = -4.984$	$B_{32} = 0.008$
$B_{40} = 1884.772$	$B_{41} = -12.088$	$B_{42} = 0.019$

**Table 4** Parameters of Eq. (6) in the range 288.15–323.15 K and  $\delta$  in accordance to Eq. (7)

$C_{00} = 3364.918$	$C_{01} = 5.113$	$C_{02} = -0.002$	$C_{03} = 0.000$	
$C_{10} = 1820.884$	$C_{11} = 0.245$	$C_{12} = -0.015$	$C_{13} = 0.000$	
$C_{20} = -9865.899$	$C_{21} = -15.144$	$C_{22} = 0.006$	$C_{23} = 0.000$	$\sigma = 34.15$

laboratory could be obtained from previously published works [2].

#### Data procedure

The changes of isentropic compressibilities are presented in Table 2 and were computed from the Eq. (1):

$$\delta Q = Q - \sum_{i=1}^N x_i Q_i \quad (1)$$

In this equation,  $\delta Q$  means the variation of a magnitude  $Q$  ( $\kappa_s$ , isentropic compressibilities calculated by the Laplace-Newton equation from density and ultrasonic velocity),

$$\kappa_s = \frac{1}{\rho v^2} \quad (2)$$

being  $\rho$  density and  $v$  velocity of sound.

$Q_i$  is the pure solvent magnitude,  $x_i$  is the mole fraction, and  $N$  is the number of components into the mixtures.

A Redlich-Kister [3] type equation was used to correlate the derived properties of the binary mixtures, Table 3, by the unweighted least squares method, all experimental points weighting equally:

$$\delta Q_{ij} = x_i x_j \sum_{p=0}^m B_p (x_i - x_j)^p \quad (3)$$

where  $\delta Q_{ij}$  stands for the derived magnitude,  $B_p$  are the fitting parameters and  $M$  is the degree of the polynomial, determined applying the F-test due to Bevington [4]. The  $B_p$  parameters were computed using a non-linear optimization algorithm due to Marquardt [5]. The ternary derived magnitudes were fitted to the equation:

$$\delta Q_{123} = \delta Q_{12} + \delta Q_{13} + \delta Q_{23} + \Delta_{123} \quad (4)$$

where the binary magnitudes  $\delta Q_{ij}$  have been correlated to Eq. (3) and  $\Delta_{123}$  is the ternary contribution fitted by means of a modified Cibulka equation [6]:

$$\Delta_{123} = x_1 x_2 x_3 RT (C_0 + C_1 x_1 + C_2 x_2) \quad (5)$$

where  $x_i$  is the molar fraction,  $R$ , the universal constant for gases and  $T$ , the temperature in Kelvin degrees. The  $B_i$  parameters are temperature dependent as follows:

$$C_i = \sum_{j=0}^3 C_{ij} T^j \quad (6)$$

The  $C_{ij}$  parameters were computed and enclosed with their root means square deviations in Table 4. The root mean square deviations presented were computed using the Eq. (7), where  $z$  is the value of the derived magnitude, and  $n_{\text{DAT}}$  is the number of experimental data:



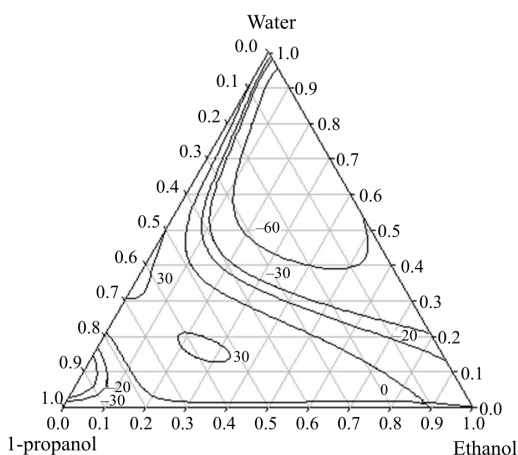
$$\sigma = \left( \frac{\sum_{i=1}^{n_{\text{DAT}}} (z_{\text{exp}} - z_{\text{pred}})^2}{n_{\text{DAT}}} \right)^{1/2} \quad (7)$$

## Results and conclusions

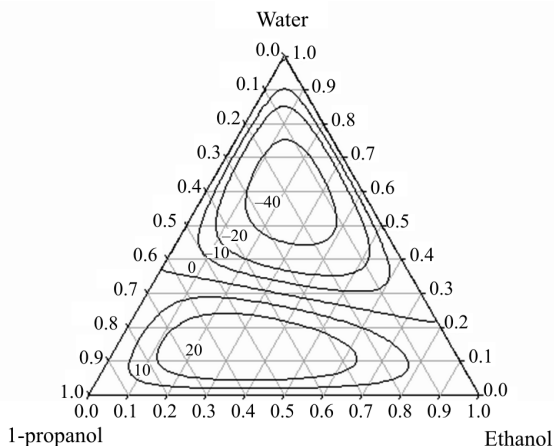
In Table 2, it can see measurements of ultrasonic velocity at range of temperature 323.15–288.15 K. Also isentropic compressibility has been calculated in same table, deviation of isentropic compressibility has been obtained in the same range of temperature.

To correlate this experimental data Cibulka equation [6] has been used. The parameter of this equation have been reported in Table 4. Standard deviation is very good for these experimental data.

The behavior of this system for deviation of isentropic compressibility is gathered in Fig. 1. In



**Fig. 1** Curves of changes of isentropic compressibility for ethanol+water+1-propanol 298.15 K



**Fig. 2** Curves of ternary contribution of the changes of isentropic compressibility for ethanol+water+1-propanol 298.15 K

Fig. 2 it can see ternary contribution, equation [5], of ethanol+water+1-propanol.

There are two clearly different regions this fact can see in Fig. 1, there is one contractive region near of pseudobinary ethanol+water, this binary has negative values of deviation of isentropic compressibility that is, it has contractive tendency. Second region is near of pseudobinary ethanol+1-propanol that is expansive region, the behavior in binary ethanol+1-propanol is according with trend of this ternary region. At last third binary 1-propanol+water have two different trends one contractive in  $x$  (1-propanol mole fraction) $>0.8$  where deviation of isentropic compressibility is negative and other one expansive  $x$  (1-propanol mole fraction) $<0.8$  where deviation of isentropic compressibility is positive. In general we can see that influence of ethanol+water system is very strong, this is because water have hydrogen bounds and it have strong unions between molecules.

In Fig. 2 can see two parts of ternary contribution one of them positive and the other negative. This ternary contribution around pure water till,  $x \approx 0.2$  (water mole fraction), in binary ethanol+water, and  $x \approx 0.35$  (water mole fraction), in binary 1-propanol+water, this region have a contractive contribution to system, we can see that, this contribution is around 60% of all trend. Other region ternary contribution is positive that is, expansive. This contribution is around 60% of total deviation property.

## Acknowledgements

The authors are grateful to Ministerio de Educación y Ciencia, for financial support in the project which reference is PPQ2002-00164.

## References

- 1 J. M. Resa, C. Gonzalez, J. M. Goenaga and M. Iglesias, *Phys. Chem, Liq.*, 43 (2005) 65.
- 2 C. Gonzalez, M. Iglesias, J. Lanz and J. M. Resa, *Thermochim. Acta*, 328 (1999) 277.
- 3 O. Redlich and A. T. Kister, *Ind. Eng. Chem.*, 40 (1948) 345.
- 4 P. Bevington, *Data Reduction and Error Analysis for the Physical Sciences*, McGraw-Hill: New York 1969.
- 5 D. W. Marquardt, *J. Soc. Ind. Appl. Math.*, 2 (1963) 431.
- 6 I. Cibulka, *Coll. Chem. Comm.*, 47 (1982) 1414.
- 7 A. Arce, A. Arce Jr., E. Rodil and A. Soto. *J. Chem. Eng. Data*, 45 (2000) 536.

DOI: 10.1007/s10973-006-7821-x