Speed of Sound and Isentropic Compressibility of Organic Solvents + Sunflower Oil Mixtures at 298.15 K

Cristina González*, José M. Resa, Juan Lanz, and M. Angeles Fanega

Chemical Engineering Department, Faculty of Pharmacy, University of the Basque Country, Vitoria, Spain

ABSTRACT: The speed of sound in binary liquid mixtures of organic solvents with sunflower oil was measured over the entire range of concentrations at 298.15 K. The solvents considered were: *n*-hexane, *n*-heptane, *n*-octane, *n*-nonane, ethanol, 1-propanol, 2-propanol, 1-butanol, ethyl acetate, propyl acetate, isopropyl acetate, butyl acetate, and vinyl acetate. The speed of sound data were used to calculate isentropic compressibilities and excess isentropic compressibilities. The excess isentropic compressibilities vs. concentrations were correlated using Redlich–Kister polynomials. Speed of sound determinations in the mixtures were predicted using two mixing rules proposed in the literature, and values were compared with experimental data. In all cases, SD between predicted and experimental data were less than 3.5%.

Paper no. J10155 in *JAOCS 79,* 543–548 (June 2002).

KEY WORDS: Isentropic compressibility, mixture properties, speed of sound, sunflower oil mixtures.

The lack of knowledge of physical properties of foods is often a limiting factor when new food processes are being designed. In some food operations, such as the analysis of alcoholic beverages, measuring the speed of sound is of special importance. Also, the speed of sound is a reliable property that allows a quick and easy determination of concentrations of solvents in the micelles that appear in oil technology processes, such as fractionation with solvents. Several properties of mixtures of organic solvents with edible oils have previously been determined by our group. Densities and excess molar volumes have been presented (1–4), as have refractive indices, speed of sound determinations, and isentropic compressibilities (5,6). Several studies (6–9) have been carried out on the use of speed of sound for analysis of vegetable oils and fats, but no data have been published on the influence of solvents on speed of sound determinations in sunflower oil mixtures. This paper presents speed of sound and isentropic compressibilities of mixtures of organic solvents with sunflower oil measured at 298.15 K. The solvents included in the study were: *n*-hexane, *n*-heptane, *n*-octane, *n*-nonane, ethanol, 1-propanol, 2-propanol, 1-butanol, ethyl acetate, propyl acetate, isopropyl acetate, butyl acetate, and vinyl acetate. Two mixing rules, Danusso (10) and Nomoto (10), have been used to predict speed of sound determinations of the mixtures, and these results were compared to experimental data. Although sunflower oil is a mixture of different compounds, it was considered a pseudo-pure compound for the calculations done in this work. Based on the results, this approximation was shown to be accurate.

*To whom correspondence should be addressed at Chemical Engineering Department, Faculty of Pharmacy, University of the Basque Country, P.O. Box 450, Vitoria, Spain. E-mail: iqpgoorc@vc.ehu.es

EXPERIMENTAL PROCEDURES

Commercially refined sunflower oil was supplied by Koipesol (Jaén, Spain). Hydrocarbons, alcohols, and acetates were analytical-grade reagents and were supplied by Fluka (Buchs, Switzerland). Samples of different solvent concentrations were prepared by weighing in a Salter ER-182A balance (Tokyo, Japan). The accuracy of the balance was $\pm 5 \times 10^{-4}$ g. Density of the samples was measured at different temperatures with an Anton Paar DMA-58 (Graz, Austria) densitometer with a precision of ±0.00001 g/cm^3. Speed of sound of the mixtures and pure solvents was measured in an Anton Paar DSA-48 sound velocity analyzer with a precision of ±1 m·s^{-1}.

Oil was analyzed to determine its composition and concentration. FA concentrations were determined by a Shimadzu GC-14B (Kyoto, Japan) gas chromatograph equipped with an FID. The chromatographic technique and the chemical procedure for the preparation of FA have been described previously (11). The concentration of FA in the sunflower oil and other properties of the oil were as follows: Composition (wt%): 8.4 palmitic, 3.9 stearic, 28.9 oleic, 58.3 linoleic, 0.5 linolenic; acid value: 0.07; saponification value: 190.3; iodine value: 125.4; PV: 13.4; water and volatiles: 0.010. The methods used to analyze these properties were standard Spanish procedures (12). From the FA concentrations, the average molar mass of this oil was calculated according to the following expression:

$$M_{\text{oil}} = 3\left(\sum_{i=1}^{N} x_i M_i\right) + M_{\text{CH–C–CH}} \qquad [1]$$

where x_i is the mole fraction, M_i the M.W. of each FA, N the number of FA found by analysis, and $M_{\text{CH–C–CH}}$ the molar mass of the glycerol molecule fraction. The calculated average molar mass in sunflower oil samples was 874.12 g·mol^{-1}. Variations in the concentrations of FA due to different samples affected mainly the mono- and polyunsaturated FA, with the change in molar mass being lower than ±1 g·mol^{-1}.

RESULTS AND DISCUSSION

Experimental and literature densities, and speed of sound determinations for each of the solvents and for the sunflower oil

TABLE 1
Comparison of Experimental Data (exp) with Literature (lit) for Density (ρ) and Speed of Sound (*u*) of Solvents and Oil at 298.15 K[a]

	ρ (g·cm^{-3})		*u* (m·s^{-1})	
Compound	Exp	Lit	Exp	Lit
Sunflower oil	0.91545	0.920–0.925[b] (13)	1453.7	1456.0 (7)
n-Hexane	0.65485	0.65484 (14)	1076.9	1078 (15)
n-Heptane	0.67955	0.67946 (14)	1130.3	1130 (15)
n-Octane	0.69859	0.69862 (14)	1172.2	1173 (15)
n-Nonane	0.71398	0.71381 (14)	1206.3	1207 (15)
Methanol	0.78637	0.78637 (14)	1101.8	1102 (16)
Ethanol	0.78513	0.78509 (14)	1142.6	1145 (16)
1-Propanol	0.79952	0.79975 (14)	1205.6	1204 (16)
2-Propanol	0.78095	0.78126 (14)	1138.3	1141 (16)
1-Butanol	0.80614	0.80600 (14)	1241.4	1240 (16)
Ethyl acetate	0.8943	0.89455 (14)	1139.5	1140 (17)
				1148 (18)
Propyl acetate	0.88206	0.88303 (14)	1115.7	
Isopropyl acetate	0.86645	0.8702 (14)	1165.4	1172 (18)
Butyl acetate	0.87605	0.87636 (14)	1103.6	
Vinyl acetate	0.92565	0.92634 (14)	1190.5	1190 (18)

[a]Numbers in parentheses indicate literature references.
[b]At 293.15 K.

are listed in Table 1. Experimental densities were within ±0.001 g/cm^3 of literature values, and experimental speed of sound data were within ±3 m·s^{-1} of literature values.

Speed of sound, isentropic compressibilities, and excess isentropic compressibilities of the mixtures formed by hydrocarbons, alcohols, and acetates with sunflower oil at 298.15 K appear in Tables 2–4, respectively. From experimental speed of sound data, isentropic compressibilities (k_s) were calculated using the Laplace equation:

$$k_s = u^{-2}\rho^{-1} \quad [2]$$

where ρ and u are the density and the speed of sound of binary mixtures, respectively. The density of the mixtures was determined as previously reported in the literature (1,19). Variations in the isentropic compressibility (or excess isentropic compressibility, k_s^E) were evaluated from the equation:

$$k_s^E = k_s - x_1 k_{s1} - x_2 k_{s2} \quad [3]$$

where k_s, k_{s1}, and k_{s2} are the isentropic compressibilities of the mixture and the pure components 1 and 2, respectively, and x_1 and x_2 are the mole fractions of the organic solvent and the oil, respectively.

Variations in isentropic compressibility were calculated as a function of concentration using Redlich–Kister polynomials (20):

$$k_s^E = x_1 x_2 \sum_{k\geq 0} a_k (x_1 - x_2)^k \quad [4]$$

where a_k are the adjustable parameters obtained by regression analysis using a least squares method, including six adjustable parameters. Table 5 summarizes the values of the

TABLE 2
Experimental Speeds of Sound (*u*), Predicted Speeds of Sound [u_{Nomoto} (Eq. 6) and $u_{Danusso}$ (Eq. 5)], Isentropic Compressibilities (k_s), and Excess Isentropic Compressibilities (k_s^E) of Mixtures of Hydrocarbons with Sunflower Oil at 298.15 K

x_1	*u* (m·s^{-1})	u_{Nomoto} (m·s^{-1})	$u_{Danusso}$ (m·s^{-1})	k_s (TPa^{-1})	k_s^E (TPa^{-1})
			n-Hexane		
0.0537	1450.6	1450.8	1446.6	519.9	−39.6
0.0997	1447.6	1447.8	1439.9	523.1	−73.2
0.1791	1441.9	1441.9	1429.5	530.1	−129.8
0.3188	1428.7	1428.9	1400.2	543.5	−228.3
0.4138	1417.2	1417.4	1377.8	556.5	−291.4
0.4528	1411.4	1411.8	1367.7	563.3	−315.8
0.4931	1404.8	1405.4	1356.2	571.1	−340.3
0.5461	1395.0	1395.6	1339.2	582.9	−371.0
0.5983	1383.5	1384.3	1320.8	597.3	−398.4
0.7032	1352.2	1354.0	1276.5	639.1	−440.6
0.7995	1307.8	1311.4	1223.9	705.6	−451.2
0.9010	1226.7	1234.6	1153.4	856.5	−381.7
0.9651	1142.0	1148.9	1101.4	1075.9	−213.6
			n-Heptane		
0.0502	1450.9	1451.2	1448.0	519.6	−28.7
0.1073	1447.2	1447.6	1440.9	523.6	−61.0
0.2038	1440.2	1440.7	1427.3	531.3	−114.5
0.3158	1430.0	1430.8	1408.6	542.6	−174.2
0.4304	1416.5	1417.6	1385.1	558.2	−231.4
0.4602	1412.2	1413.5	1378.3	563.3	−245.1
0.4940	1407.0	1408.5	1369.8	569.6	−260.4
0.5570	1395.7	1397.7	1352.8	583.6	−286.3
0.6086	1384.9	1387.1	1336.9	597.4	−305.2
0.7047	1358.2	1361.5	1301.5	633.6	−330.1
0.7998	1319.1	1324.1	1257.5	692.7	−331.4
0.9076	1244.9	1252.5	1193.5	830.0	−262.4
0.9537	1196.1	1202.9	1161.5	945.8	−175.8
			n-Octane		
0.0502	1450.9	1451.3	1448.7	519.7	−23.2
0.1039	1447.7	1448.1	1442.8	523.2	−47.8
0.1954	1441.2	1441.9	1431.3	530.4	−88.8
0.2995	1432.3	1433.5	1416.4	540.5	−133.3
0.4335	1417.5	1419.2	1392.6	558.0	−186.3
0.4605	1413.9	1415.7	1387.0	562.4	−196.2
0.5127	1406.0	1408.3	1375.4	572.0	−213.9
0.5515	1399.4	1402.0	1366.0	580.4	−226.0
0.5945	1391.2	1394.1	1354.8	591.1	−237.9
0.6968	1366.2	1370.2	1322.9	625.2	−257.5
0.7986	1329.3	1335.0	1282.7	681.2	−255.1
0.9087	1264.4	1271.5	1226.5	800.4	−193.7
0.9531	1225.7	1231.5	1200.0	888.7	−128.8
			n-Nonane		
0.0456	1451.3	1451.7	1449.7	519.3	−17.5
0.0979	1448.1	1448.7	1444.4	522.8	−37.3
0.2006	1441.4	1442.1	1433.0	530.6	−75.4
0.2857	1434.4	1435.6	1422.1	538.7	−105.3
0.4066	1422.1	1424.1	1403.6	553.3	−144.6
0.4483	1417.0	1419.3	1396.4	559.6	−156.9
0.4999	1410.2	1412.7	1386.4	568.1	−171.4
0.5476	1402.6	1405.6	1376.4	577.8	−183.0
0.6111	1391.1	1394.7	1361.4	593.1	−196.0
0.6945	1371.9	1376.4	1338.5	620.0	−206.4
0.8003	1337.5	1343.6	1301.9	672.6	−201.0
0.9176	1276.4	1282.7	1249.4	785.9	−140.0
0.9577	1246.4	1250.8	1228.3	853.9	−89.9

TABLE 3
Experimental Speeds of Sound (u), Predicted Speeds of Sound [u_{Nomoto} (Eq. 6) and $u_{Danusso}$ (Eq. 5)], Isentropic Compressibilities (k_s), and Excess Isentropic Compressibilities (k_s^E) of Mixtures of Alcohols with Sunflower Oil at 298.15 K

x_1	u (m·s^{-1})	u_{Nomoto} (m·s^{-1})	$u_{Danusso}$ (m·s^{-1})	k_s (TPa^{-1})	k_s^E(TPa^{-1})
			Methanol		
0.0520	1452.7	1453.1	1452.6	517.6	−26.4
0.1054	1451.2	1452.1	1451.0	518.9	−53.5
0.1549	1450.4	1451.0	1449.0	519.6	−79.0
0.2068	1448.6	1449.7	1447.1	521.2	—
0.2619	1447.2	1448.2	1444.2	522.3	−133.1
0.2999	1445.1	1447.1	1442.5	524.1	−151.4
0.3577	1442.8	1445.0	1439.1	526.2	−180.1
0.3994	1440.0	1443.4	1436.6	528.6	−199.7
0.4534	1437.7	1440.8	1432.6	530.8	−226.2
0.5053	1432.9	1437.9	1428.6	535.2	−249.3
			Ethanol		
0.0632	1452.4	1452.6	1451.7	517.9	−27.6
0.0913	1451.3	1451.9	1450.7	518.8	−39.5
0.1999	1447.7	1448.9	1446.1	522.1	−86.1
0.3193	1442.4	1444.6	1439.3	527.0	−136.1
0.4242	1436.2	1439.5	1431.4	532.7	−178.5
0.4687	1433.0	1436.7	1427.4	535.7	−195.9
0.5130	1429.5	1433.6	1422.7	539.1	−212.9
0.5540	1425.5	1430.2	1417.6	542.9	−227.9
0.5924	1421.4	1426.6	1412.3	546.9	−241.5
0.6993	1405.8	1412.3	1391.7	562.5	−275.0
			1-Propanol		
0.0301	1453.0	1453.4	1453.2	517.4	−9.4
0.0939	1451.1	1451.9	1450.9	519.1	−29.7
0.1869	1448.4	1449.3	1447.3	521.7	−59.2
0.2905	1444.2	1445.7	1442.2	525.7	−90.9
0.4061	1437.9	1440.5	1434.9	531.7	−124.8
0.4482	1435.4	1438.1	1431.6	534.2	−136.8
0.5006	1431.4	1434.7	1426.8	538.1	−151.0
0.5492	1427.1	1431.0	1421.7	542.3	−163.5
0.6158	1420.2	1424.5	1413.1	549.4	−179.4
0.6929	1409.3	1414.4	1399.8	560.6	−194.7
0.8075	1382.6	1389.3	1368.7	589.9	−204.9
0.9050	1335.2	1343.5	1318.6	647.7	−180.7
0.9646	1274.3	1280.8	1261.0	736.1	−112.9
			2-Propanol		
0.0504	1452.1	1452.5	1451.7	518.1	−22.1
0.0986	1450.2	1451.0	1449.3	519.9	−43.1
0.1877	1446.2	1447.8	1444.8	523.7	−81.3
0.3337	1438.4	1440.8	1433.7	531.0	−143.0
0.4034	1433.5	1436.4	1427.1	535.7	−171.2
0.4466	1430.0	1433.2	1422.3	539.1	−188.1
0.4802	1427.0	1430.4	1418.1	542.0	−201.1
0.5461	1419.9	1424.0	1408.8	549.1	−225.1
0.6059	1412.2	1416.6	1398.2	556.9	−245.5
0.6974	1395.6	1401.1	1377.0	574.3	−271.3
0.8108	1361.6	1368.0	1334.7	612.4	−286.8
			1-Butanol		
0.0425	1452.7	1453.0	1452.7	517.7	−11.1
0.0917	1451.5	1451.8	1451.0	518.8	−24.0
0.2045	1447.7	1448.6	1446.6	522.5	−52.9
0.3054	1443.4	1444.9	1441.7	526.8	−77.7
0.4377	1436.0	1438.4	1433.1	534.1	−108.5
0.4503	1435.1	1437.7	1431.9	535.0	−111.3
0.4888	1432.1	1435.2	1428.9	538.1	−119.3
0.5693	1425.2	1428.8	1420.5	545.1	−135.5
0.6188	1419.8	1423.9	1414.1	550.8	−144.1
0.7015	1408.2	1413.1	1400.6	563.3	−155.5
0.7928	1388.9	1394.5	1378.5	585.0	−160.0
0.8937	1349.7	1356.1	1337.3	633.4	−140.7
0.9575	1301.3	1305.9	1290.8	702.3	−90.2

TABLE 4
Experimental Speeds of Sound (u), Predicted Speeds of Sound [u_{Nomoto} (Eq. 6) and $u_{Danusso}$ (Eq. 5)], Isentropic Compressibilities (k_s), and Excess Isentropic (k_s^E) Compressibilities of Mixtures of Acetates with Sunflower Oil at 298.15 K

x_1	u (m·s^{-1})	u_{Nomoto} (m·s^{-1})	$u_{Danusso}$ (m·s^{-1})	k_s (TPa^{-1})	k_s^E) (TPa^{-1})
			Ethyl acetate		
0.0473	1451.6	1452.3	1451.6	518.2	−14.4
0.1084	1448.7	1449.8	1448.2	520.3	−33.2
0.2089	1442.6	1445.0	1441.7	524.9	−63.1
0.3130	1434.8	1438.8	1433.7	530.9	−92.7
0.4391	1422.5	1428.8	1420.4	540.5	−126.4
0.4705	1418.7	1425.7	1416.5	543.5	−134.1
0.5334	1410.0	1418.5	1407.4	550.5	−148.6
0.5742	1403.4	1413.0	1400.4	555.9	−157.2
0.6095	1396.9	1407.5	1393.7	561.3	−163.9
0.7198	1369.9	1384.1	1365.7	584.5	−178.4
0.8329	1323.1	1341.8	1318.7	628.5	−173.2
0.9160	1261.3	1281.6	1258.5	694.6	−135.6
0.9748	1188.4	1199.2	1186.0	787.1	−63.2
			Propyl acetate		
0.0452	1451.7	1452.2	1451.6	518.2	−12.6
0.1028	1448.8	1449.8	1448.2	520.4	−28.7
0.2187	1441.8	1443.8	1440.2	525.8	−60.2
0.3371	1432.6	1436.1	1430.1	533.1	−90.6
0.4056	1426.1	1430.4	1422.7	538.3	−107.1
0.4517	1421.1	1426.0	1417.1	542.4	−117.7
0.5306	1410.6	1416.9	1405.7	551.1	−134.1
0.5632	1405.6	1412.5	1400.1	555.2	−140.3
0.6057	1398.0	1405.9	1392.1	561.8	−147.3
0.7025	1376.1	1386.1	1368.7	581.1	−158.7
0.8144	1336.4	1349.0	1327.6	619.0	−156.4
0.8842	1296.4	1310.0	1287.9	661.0	−136.6
0.9674	1215.3	1224.6	1211.5	760.9	−63.1
			Isopropyl acetate		
0.0623	1450.2	1450.9	1449.4	519.4	−23.9
0.1031	1447.8	1448.6	1446.0	521.2	−39.7
0.2335	1437.9	1440.2	1433.6	529.0	−88.1
0.3416	1427.6	1431.1	1420.6	537.3	−126.4
0.4259	1417.5	1422.1	1408.2	545.6	−154.5
0.4473	1414.6	1419.5	1404.6	548.1	−161.3
0.5012	1406.3	1412.1	1394.7	555.1	−177.4
0.5626	1395.2	1402.1	1381.7	564.8	−194.2
0.6120	1384.5	1392.4	1369.3	574.3	−206.0
0.6994	1360.5	1370.2	1341.9	596.6	−221.4
0.8117	1312.4	1324.9	1290.7	645.2	−221.2
0.9286	1217.3	1230.6	1200.1	761.2	−155.6
0.9568	1180.2	1191.2	1167.6	815.4	−113.6
			Butyl acetate		
0.0513	1451.5	1451.9	1451.2	518.4	−12.9
0.0893	1449.6	1450.2	1448.9	519.9	−22.4
0.2072	1442.8	1444.2	1440.9	525.2	−51.0
0.3371	1433.1	1435.5	1429.7	533.1	−80.7
0.4013	1427.2	1430.2	1422.9	537.9	−94.4
0.4488	1422.2	1425.6	1417.1	542.0	−104.0
0.4831	1418.1	1421.9	1412.5	545.5	−110.5
0.5294	1412.0	1416.3	1405.7	550.7	−118.6
0.5982	1401.2	1406.3	1393.5	560.0	−129.2
0.6903	1382.1	1388.5	1372.8	577.2	−138.6
0.8128	1342.6	1350.7	1331.4	615.2	−136.0
0.9151	1283.3	1291.8	1273.9	680.1	−100.6
0.9702	1230.4	1235.6	1225.6	747.4	−49.3
			Vinyl acetate		
0.0490	1451.5	1452.2	1452.2	518.2	−15.2
0.1007	1448.8	1450.0	1449.4	520.1	−31.5
0.1998	1442.8	1445.3	1443.0	524.4	−62.0
0.2956	1435.6	1439.6	1435.4	529.5	−90.6
0.3951	1426.4	1432.0	1425.7	536.3	−118.7
0.4506	1420.0	1426.9	1418.9	541.1	−133.5
0.5059	1412.6	1420.8	1411.2	546.7	−147.3
0.5414	1407.2	1416.3	1405.6	550.8	−155.7
0.5901	1398.5	1409.1	1396.6	557.6	−166.1
0.6940	1373.9	1388.3	1371.5	577.4	−182.8
0.8342	1315.5	1336.1	1312.8	628.9	−180.6
0.9153	1250.0	1273.0	1249.3	695.2	−142.7
0.9752	1166.4	1180.9	1167.5	796.0	−63.0

TABLE 5
Redlich–Kister Fitting Parameters[a] at 298.15 K for the k_s^E vs. Mole Fraction Data

Compound	Parameters of k_s^E vs. mole fraction curves						
	a_0	a_1	a_2	a_3	a_4	a_5	σ^b
n-Hexane	–1386.1	1231.4	–673.3	287.4	–1868.1	1911.9	0.16
n-Heptane	–1054.6	890.9	–544.7	325.3	–1080.3	1004.5	0.061
n-Octane	–840.5	682.7	–420.3	240.8	–697.2	641.3	0.078
n-Nonane	–686.6	529.2	–327.4	205.7	–464.5	399.7	0.092
Methanol	–987.2	874.4	–616.1	227.2	—[c]	—	0.23
Ethanol	–830.4	756.8	–753.1	731.4	–362.5	—	0.36
1-Propanol	410.1	–354.0	195.7	–62.3	494.2	–537.1	3.02
2-Propanol	–834.8	737.4	–596.8	657.9	–920.6	547.7	0.44
1-Butanol	–609.7	573.2	–226.2	–61.9	–1168.9	1359.8	1.53
Ethyl acetate	–566.7	490.5	–261.1	42.2	–658.2	746.3	0.89
Propyl acetate	–513.9	420.5	–251.2	137.8	–492.3	475.3	0.35
Isopropyl acetate	–711.4	596.9	–359.4	187.9	–757.8	751.1	0.45
Butyl acetate	–455.6	367.5	–226.0	121.4	–379.4	359.8	0.28
Vinyl acetate	–576.0	509.9	–293.1	135.7	–594.1	726.8	0.75

[a]Coefficients a_0 through a_5 as defined in Equation 4.
[b]Indicates SD.
[c]Not determined.

coefficients a_k, together with the SD, σ, of the differences between experimental values and those calculated from Equation 4. The coefficients, a_k, in Equation 4 were used to calculate the excess isentropic compressibilities, which are shown in Figures 1–3. In Figure 1, the excess isentropic compressibilities of the mixtures as a function of the hydrocarbon mole fraction at 298.15 K can be observed. Figure 2 shows the excess isentropic compressibilities of the mixtures of alcohols with sunflower oil as a function of the alcohol mole fraction. In this figure, lines are truncated because methanol, ethanol, and 2-propanol were not miscible with oil in all the concentration ranges. Properties were measured only for the soluble mixtures. In Figure 3, the excess isentropic compressibilities of mixtures of acetate and sunflower oil are shown. All systems had negative values for k_s^E over the entire concentration range. The negative values may be attributed to the effects of dispersion and dipolar interaction between unlike molecules. The observed negative k_s^E indicates that interaction of self-associated species of esters was predominant.

Experimental data for speed of sound measurements of the mixtures (u) were compared with values determined by two mixing rules (10): the model of Danusso (Eq. 5) and the equation of Nomoto (Eq. 6):

$$u = \frac{1}{\rho}\left[\frac{1}{M}\left(\frac{n_1 M_1}{\rho_1^2 u_1^2} + \frac{n_2 M_2}{\rho_2^2 u_2^2}\right)\right]^{-1/2} \quad [5]$$

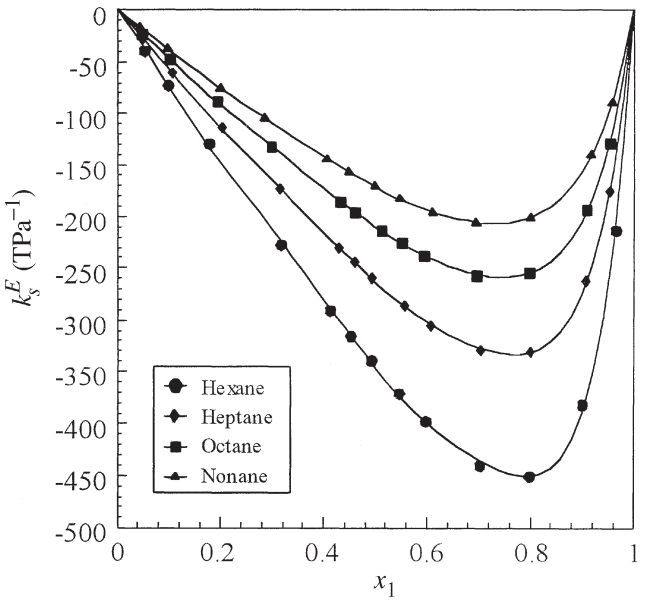

FIG. 1. Excess isentropic compressibilities of mixtures of hydrocarbons and sunflower oil in terms of the mole fraction of the hydrocarbons. See text for details.

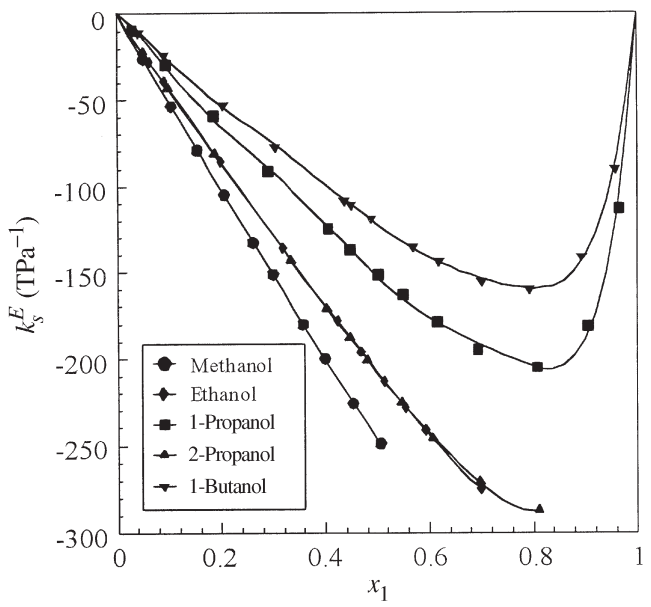

FIG. 2. Excess isentropic compressibilities of mixtures of alcohols and sunflower oil in terms of the mole fraction of the alcohols.

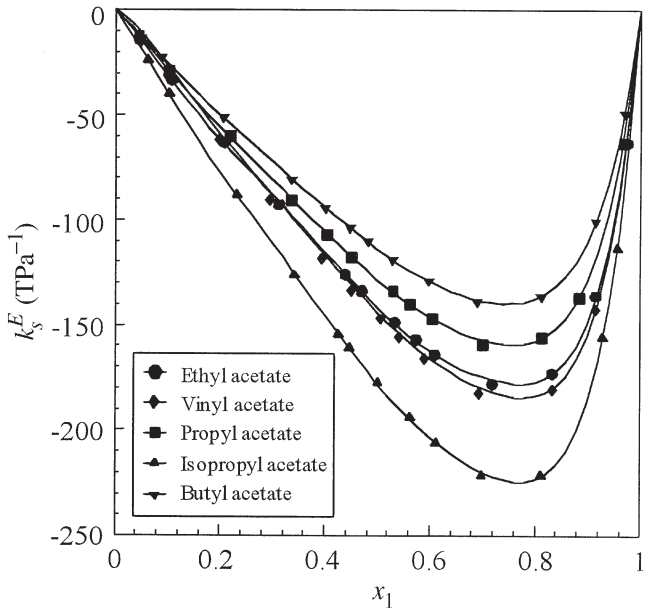

FIG. 3. Excess isentropic compressibilities of mixtures of acetates and sunflower oil in terms of the mole fraction of the acetates.

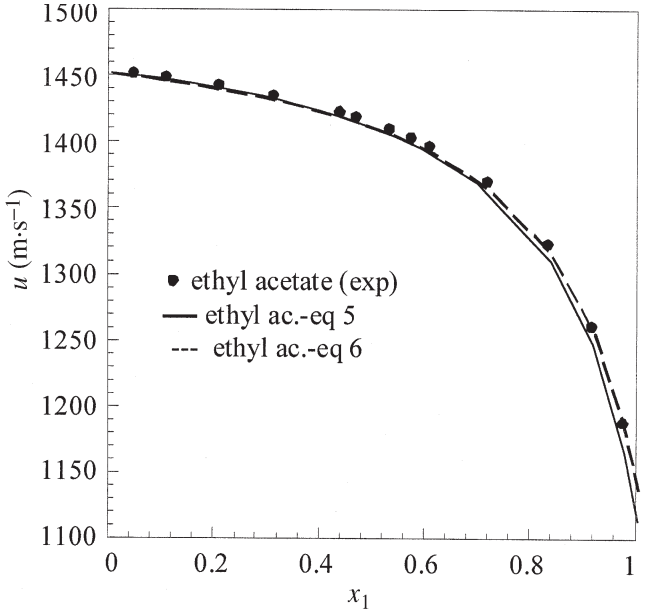

FIG. 4. Speed of sound of mixtures of ethyl acetate and sunflower oil in terms of the mole fraction of the acetate. Symbols indicate experimental values and lines indicate values predicted by Equations 5 and 6. See text for details.

$$u = \left(\frac{n_1 R_1 + n_2 R_2}{n_1 u_1 + n_2 u_2} \right)^3 \quad [6]$$

Here,

$$M = n_1 M_1 + n_2 M_2 \quad [7]$$

and

$$R = (n_1 V_1 + n_2 V_2) u^{1/3} \quad [8]$$

where V_1 and V_2 are the molar volumes of the pure components.

The results of the comparison of predicted values with experimental data appear in Tables 2–4, in which the speed of sound measurements predicted by Equations 5 and 6 are shown. The standard average deviations (δu) and the percentage of deviation (ε) between experimental data and calculated values appear in Table 6. The model proposed by Danusso (Eq. 5) predicts speed of sound values with deviations less than 0.9% for acetate mixtures (Fig. 4). However, deviations on the order of 3.5% were found for hydrocarbon mixtures (Fig. 5). Since Danusso's model assumes additivity of the

TABLE 6
Average SD [δu (m·s^{-1})] and Percentage of Deviation (ε) for Predicted Speed of Sound with Respect to Experimental Data for Binary Mixtures at 298.15 K

	Equation 5 (Danusso)		Equation 6 (Nomoto)	
Solvent	δu (m·s^{-1})	ε (%)	δu (m·s^{-1})	ε (%)
n-Hexane	50.9	3.31	3.1	0.15
n-Heptane	38.4	2.51	3.5	0.19
n-Octane	29.4	1.91	3.5	0.21
n-Nonane	22.5	1.46	3.5	0.22
Methanol	3.0	0.18	2.4	0.14
Ethanol	6.8	0.38	3.7	0.22
1-Propanol	8.2	0.45	4.3	0.26
2-Propanol	12.0	0.65	3.6	0.23
1-Butanol	6.3	0.36	3.6	0.22
Ethyl acetate	2.6	10.6	0.17	0.65
Propyl acetate	5.2	7.5	0.32	0.47
Isopropyl acetate	12.6	7.6	0.82	0.49
Butyl acetate	6.3	4.8	0.39	0.30
Vinyl acetate	1.4	11.6	0.09	0.70

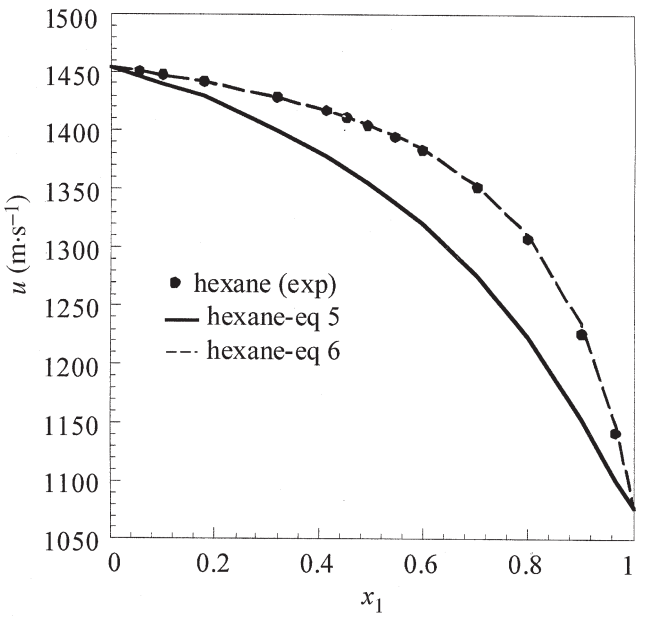

FIG. 5. Speed of sound of mixtures of hexane and sunflower oil in terms of the mole fraction of the hydrocarbon. Symbols indicate experimental values and lines indicate values predicted by Equations 5 and 6. See text for details.

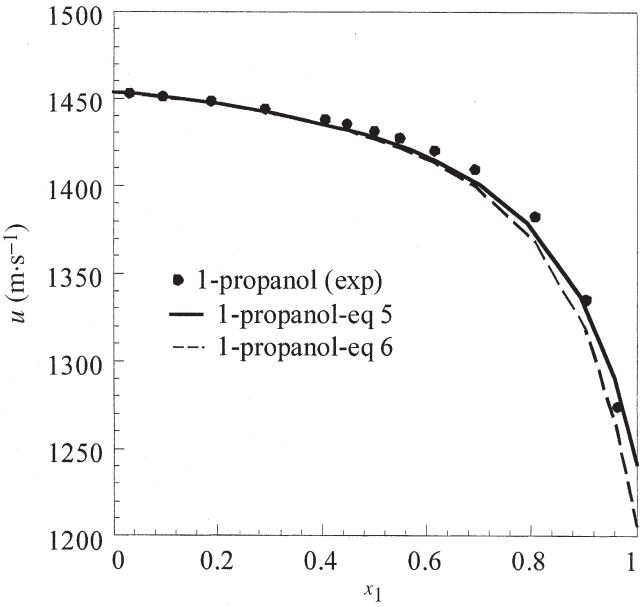

FIG. 6. Speed of sound of mixtures of 1-propanol and sunflower oil in terms of the mole fraction of the alcohol. Symbols indicate experimental values and lines indicate values predicted by Equations 5 and 6. See text for details.

molar volume, this equation more accurately predicts values for mixtures that show ideal behavior (no excess volume). This fact explains the highest deviations found for the mixtures with high excess volumes, such as hydrocarbons (10), while mixtures with low excess volumes (acetates) also satisfy this model (Eq. 5). In the alcohol–oil mixtures, both predictions show similar results (Fig. 6), and the best agreement is for the higher-M.W. alcohols (Table 4). The deviations corresponding to the mixture rules proposed by Nomoto (Eq. 6) are lower than 0.7% in all the studied mixtures.

ACKNOWLEDGMENTS

The authors thank the Basque Government (Project PI-1998-91) and the University of the Basque Country (Project 00069.125-E-13813/2001) for financial support.

REFERENCES

1. González, C., J.M. Resa, A. Ruiz, and J.I Gutiérrez, Densities of Mixtures Containing *n*-Alkanes with Sunflower Seed Oil at Different Temperatures, *J. Chem. Eng. Data 41*:796–798 (1996).
2. González, C., J.M. Resa, A. Ruiz, and J.I Gutiérrez, Excess Molar Volumes of Mixtures of Hexane + Natural Oils from 298.15 K to 313.15 K, *Ibid. 42*:339–341 (1997).
3. González, C., M. Iglesias, J. Lanz, and J.M. Resa, Temperature Dependence of Excess Molar Volumes in (*n*-Alkane (C6–C9) or Alcohol (C2–C4)) + Olive Oil Mixtures, *Thermochim. Acta 328*:277–296 (1999).
4. González, C., J.M. Resa, and J. Lanz, Excess Volumes of Binary Mixtures That Contain Olive Oil with Alkyl and Vinyl Acetates, *J. Am. Oil Chem. Soc. 77*:985–990 (2000).
5. González, C., M. Iglesias, J. Lanz, G. Marino, B. Orge, and J.M. Resa, Temperature Influence on Refractive Indices and Isentropic Compressibilities of Alcohol (C2–C4) + Olive Oil Mixtures, *J. Food. Eng. 50*:29–40 (2001).
6. Resa, J.M., C. González, M.A. Fanega, S. Ortiz de Landaluce, and J. Lanz, Enthalpies of Mixing, Heat Capacities, and Viscosities of Alcohol (C1–C4) + Olive Oil Mixtures at 298.15 K, *Ibid. 51*:113–118 (2001).
7. Coupland, J.N., and J. McClements, Physical Properties of Liquid Edible Oils, *J. Am. Oil Chem. Soc. 74*:1559–1564 (1997).
8. Javanud, C., and R.R. Rahalkar, Velocity of Sound in Vegetable Oils, *Fat Sci. Technol. 90*:73–75 (1988).
9. McClements, J., and M.J.W. Povey, Ultrasonic Analysis of Edible Oils and Fats, *Ultrasonics 30*:383–388 (1992).
10. Nomoto, O., Empirical Formula for Sound Velocity in Liquid Mixtures, *J. Phys. Soc. Japan 13*:1528–1532 (1968).
11. González, C., Preparación y Caracterización de Catalizadores de Níquel y Platino Soportados y Comportamiento Cinético en la Hidrogenación de Aceite de Girasol, Universidad del País Vasco, Ph.D. Thesis, 1993.
12. AMV Ediciones, Producción, Análisis y Control de calidad de Grasas y Aceites Comestibles, AMV Ediciones, Madrid, 1987.
13. Karleskind, A. (ed.), *Edible Oils & Fats Manual: A Comprehensive Treatise,* Lavoisier, Paris, 1996.
14. Riddick, J.A., W.B. Bunger, and T.K. Sakano, *Organic Solvents: Physical Properties and Methods of Purification,* 4th edn., John Wiley & Sons, New York, 1986.
15. Rodriguez, A., J. Canosa, and J. Tojo, Binary Mixture Properties of Methyl-*tert*-butyl Ether with Hexane or Heptane or Octane or Nonane from 288.15 to 298.15 K, *J. Chem. Eng. Data 44*:666–671 (1999).
16. Aminabhavi, T.M., M.I. Aralaguppi, S.B. Harogoppad, and R.H. Balundgri, Densities, Viscosities, Refractive Indices, and Speeds of Sound for Methyl Acetoacetate + Aliphatic Alcohols (C_1–C_8), *Ibid. 38*:31–39 (1993).
17. Acosta, J., A. Arce, E. Rodil, and A. Soto, Densities, Speeds of Sound, Refractive Indices, and the Corresponding Changes of Mixing at 25°C and Atmospheric Pressure for Systems Composed of Ethyl Acetate, Hexane, and Acetone, *Ibid. 46*: 1176–1180 (2001).
18. Aralaguppi, M.I., C.V. Jadar, and T.M. Aminabhavi, Density, Viscosity, Refractive Index, and Speed of Sound in Binary Mixtures of 2-Chloroethanol with Methyl Acetate, Ethyl Acetate, *n*-Propyl Acetate, and *n*-Butyl Acetate, *Ibid. 44*:441–445 (1999).
19. González, C., J.M. Resa, J. Lanz, and M.A. Fanega, Excess Molar Volumes and Refractive Indices of Mixtures Formed by Acetates with Sunflower Oil at Different Temperatures, *J. Am. Oil Chem. Soc. 79*:539–542 (2002).
20. Redlich, O., and A.T. Kister, Thermodynamics of Nonelectrolytic Solutions. Algebraic Representation of Thermodynamic Properties and the Classification of Solutions, *Ind. Eng. Chem. 40*:845–848 (1948).

[Received November 12, 2001; accepted March 13, 2002]