Acoustic velocities in petroleum fluids: Measurement and prediction

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ABSTRACT

Acoustic velocity or speed of sound is a thermodynamic property which may be used to estimate thermophysical properties needed for hydrocarbon production and processing. In this paper, experimental data is reported on the speed of sound in n-octane, a binary mixture of (n-octane+ n-hexadecane), a binary mixture of (n-decane+n-hexadecane) and a ternary mixture of (n-octane+ n-decane+n-hexadecane) at temperature of (293 to 393) K and at pressures up to 100 MPa. In addition a predictive method for the speed of sound in liquid hydrocarbons is presented and validated by comparison with both the present data and the literature data. The model is based on the extended principle of corresponding states with two reference fluids, chosen as n-C_{18} and n-C_{28} in which the parameters may be estimated from the knowledge of molecular weight. The method can be used for both pure liquid n-alkanes and their mixtures from propane to very heavy hydrocarbons ($\sim C_{50}$) at pressures of (0.1 to 150) MPa and at temperatures from (200 to 400) K. The method has been evaluated by comparison with over 2000 data points for pure normal alkanes (from C_3 to C_{36}), as well as for binary and ternary mixtures, petroleum fractions and crude oils, and is found to predict speed of sound in these fluids within $\pm 2\%$.

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1. Introduction

Knowledge of thermodynamic and physical properties of hydrocarbons and especially complex hydrocarbon mixtures, such as crude oils and petroleum fractions, is vital in petroleum production and processing. Generally these properties are calculated through PVT relations and generalized correlations based on the principles of corresponding states. Such methods require knowledge of the critical constants and acentric factor which are not usually available for heavy hydrocarbons or their mixtures. Estimation methods for such properties applied to heavy hydrocarbons ($> C_{20}$) usually lead to significant errors with great impact on calculated properties needed for design and operation of processing equipment (Riazi, 2005).

It has been shown that acoustic velocity data can be used to generate and predict physical and thermodynamic properties of fluids and their mixtures (Shabani et al., 1998; Dayton and Goodwin, 1999; Estrada-Alexanders and Trusler, 1997). Colgate et al. (1991, 1992) and Ball and Goodwin (2002) have reported data on the speed of sound in some crude oils and have shown how bubble points may be determined from such data. Equations of state have been developed based on the speed of sound which may be used for thermodynamic property calculations (Riazi and Mansoori, 1993a; Peleties et al., 2010). Thus it appears that the speed of sound is a quantity that may be used for estimation of thermophysical properties of heavy compounds and complex fluid mixtures in which such properties are not available.

A number of researchers have measured and reported speed of sound in hydrocarbon systems at high pressures (Dutour et al., 2000, 2001, 2002a, 2002b). Queimada et al. (2006) have proposed a corresponding states approach for the speed of sound in long-chain hydrocarbons using a reduced speed of sound based on the ideal-gas equation of state.

In this paper, new experimentally measured data on the speed of sound in liquids at high pressures for several binary and ternary hydrocarbon systems are reported. In the second part of this work, we propose a new predictive method for estimation of the speed of sound in heavy liquid hydrocarbons and their mixtures based on the corresponding states principle (CSP) using parameters which can be estimated reliably from available data. The CSP approach is a widely used technique for predicting thermodynamic properties of pure as well as fluid mixtures. Standard methods are also suggested

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for the estimation of the input parameters required for the proposed method.

2. Experimental measurements

The experimental system was based on the design reported by Peleties et al. (2010); however, the present apparatus operates at a lower frequency of 4 MHz to permit measurements on liquids with viscosities up to approximately 150 mPa s. Measurements can also be made on liquid mixtures with dissolved gases and or dense compressed gases. The working ranges of the apparatus are pressures from (0.1 to 100) MPa and temperatures from (283 to 473) K and it was installed and commissioned by the manufacturer (Imperial College, UK). Fig. 1 illustrates a schematic of flow diagram

showing ultrasonic, temperature control, pressure generation and automation sub-systems.

As shown in Figs. 2 and 3, the ultrasonic cell was mounted within a stainless-steel pressure vessel with an internal volume of approximately 100 cm^3. The pressure vessel was suspended within a temperature-controlled oil-bath (Fluke model 6040) where the temperature was controlled with a stability and uniformity of better than $\pm$ 0.01 K. The temperature was measured by means of a secondary-standard platinum resistance thermometer (Fluke model 5615) which was inserted into a thermowell in the wall of the pressure vessel. This thermometer was calibrated by the manufacturer with an expanded uncertainty of better than 0.02 K in the working range of the instrument.

Pressure of the fluid was measured with an expanded uncertainty of 0.25 kPa by means of a pressure transducer (Paroscientific model

PG : Pressure gauge
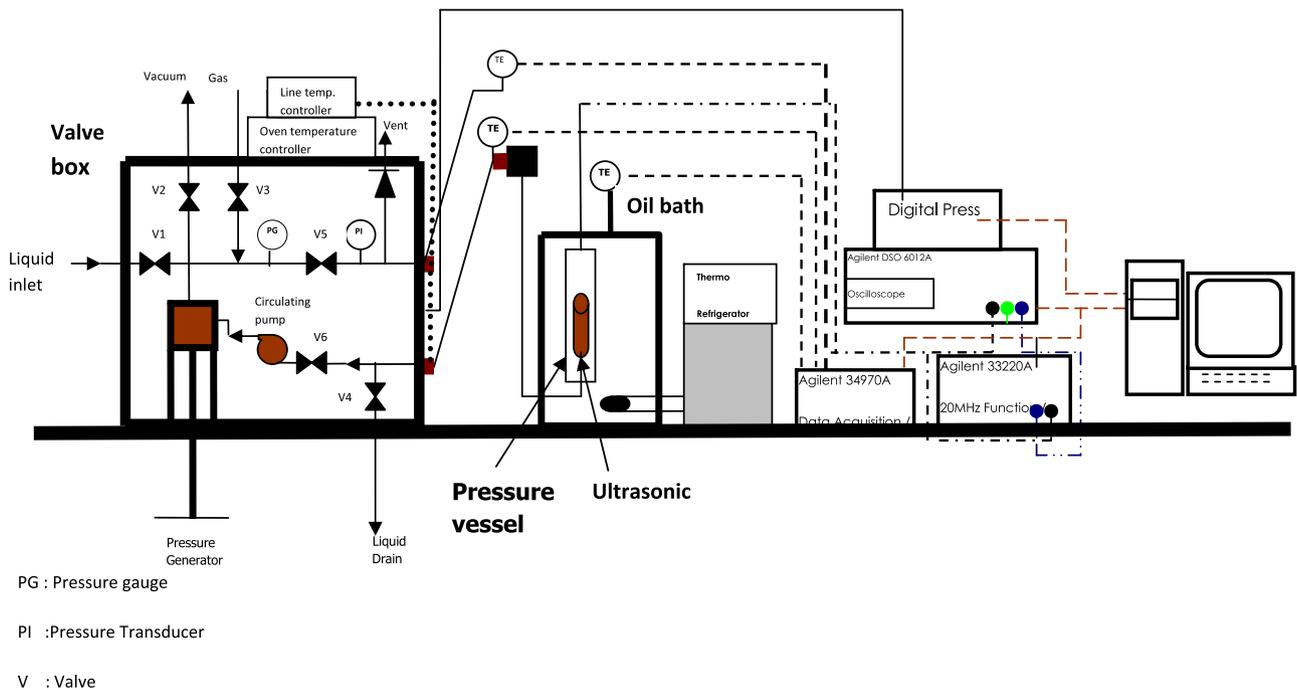
PI   :Pressure Transducer

V   : Valve

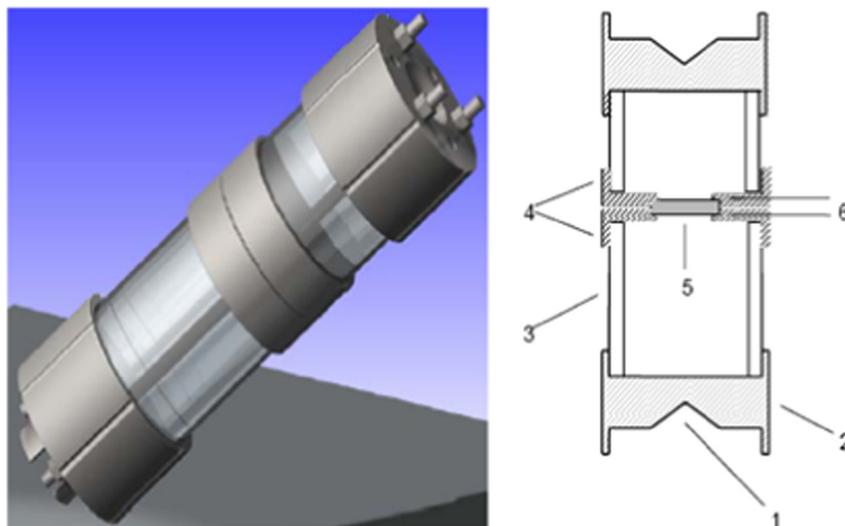Fig. 1. Flow diagram of speed of sound experimental set up.

Fig. 2. Ultrasonic cell picture and diagram on right shows: (1) conical cavity; (2) reflector; (3) quartz spacer tube; (4) stainless steel clamping ring; (5) piezoceramic transducer; (6) lead wires.
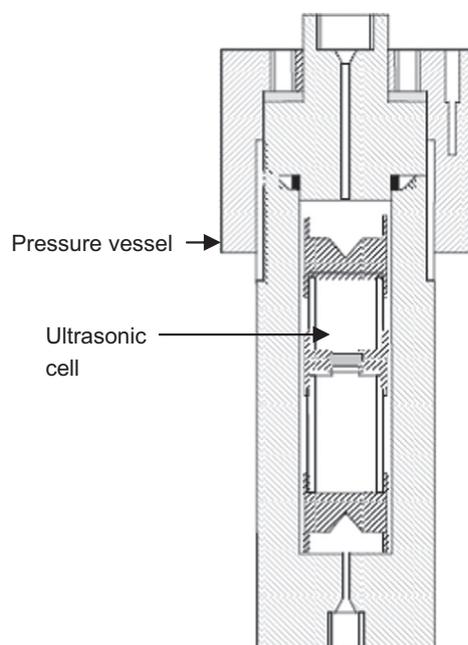
Fig. 3. Pressure vessel with ultrasonic cell.

Table 1
Experimental measurement of speed of sound for n-octane (n-C_8).

T (K)	P (MPa)	c (m s^{-1})
298.1	0.1	1172.7
298.1	10.0	1239.5
298.1	20.0	1299.6
298.1	30.0	1354.4
298.1	40.0	1404.8
298.1	50.0	1451.9
298.1	60.0	1495.9
298.1	70.0	1537.4
298.1	80.0	1576.3
298.1	90.0	1613.9
298.1	100.0	1649.7
323.1	0.1	1070.1
323.1	10.0	1145.1
323.1	20.0	1211.6
323.1	30.0	1271.1
323.1	40.0	1325.3
323.1	50.0	1375.5
323.1	60.0	1422.3
323.1	70.0	1466.2
323.1	80.0	1507.7
323.1	90.0	1546.9
323.1	100.0	1584.4
343.1	0.1	988.7
343.1	10.0	1072.8
343.1	20.0	1144.6
343.1	30.0	1208.2
343.1	40.0	1265.6
343.1	50.0	1318.1
343.1	60.0	1367.1
343.1	70.0	1412.8
343.1	90.0	1496.8
343.1	100.0	1535.5
362.2	0.1	912.0
362.2	10.0	1003.8
362.2	20.0	1080.8
362.2	30.0	1148.2
362.2	40.0	1208.9
362.2	50.0	1264.6
362.2	60.0	1314.9
362.2	70.0	1362.4
362.2	80.0	1407.1
362.2	90.0	1449.2
362.2	100.1	1489.4
383.2	10.0	935.9
383.2	20.0	1019.5
383.2	30.0	1090.5
383.2	40.0	1155.1
383.2	50.0	1213.0
383.2	60.0	1266.0
383.2	70.0	1315.2
383.2	80.0	1361.2
383.2	90.0	1404.7
383.2	100.1	1446.2

Literature data as given by: Wang and Nur (1991) 293 K, 0.1 MPa, 1192.3 m s^{-1}; 298 K, 0.1 MPa, 1173 m s^{-1}; 303 K, 0.1 MPa, 1151 m s^{-1}; 313 K, 0.1 MPa, 1109 m s^{-1}; 333 K, 0.1 MPa, 1008 m s^{-1}; 363 K, 0.1 MPa, 909 m s^{-1}.

KR415-101) located within the pressure generation sub-system. A manual syringe pump with a displacement of 30 cm^3 was used to raise and adjust the pressure. The apparatus was calibrated using speed of sound for n-octane at atmospheric pressure and various temperatures. The calibration procedure was through determination of $\Delta L = L1 - L2$, where $L1$ and $L2$ are two different path lengths between transducer and the two reflectors. The value of ΔL as determined from measured data was 20.037 mm compared to the actual value of 20 mm. However, in each new round of experiments the instrument was recalibrated using n-octane at $T = 298.15$ K and $P = 0.1$ MPa and the speed of sound under these conditions was taken as 1172.0 m s^{-1} as reported by Peleties et al. (2010). Uncertainty for the measurements was about 1 m s^{-1}.

Measurements of speed of sound were carried out at various temperatures and pressures for n-Octane, n-Decane and n-Hexadecane, for a binary mixtures of n-Octane and n-Hexadecane, and for several ternary mixtures of n-Octane, n-Decane and n-Hexadecane. n-Octane was supplied by Merck with purity of $\geq 99\%$. The densities of pure liquids were measured by means of a vibrating-tube densimeter (Anton Paar model DMA 4500) previously calibrated in air and pure degassed water. The measured densities at $T = 293.15$ K and $P = 0.1$ MPa were 703 kg m^{-3} for n-octane, 729 kg m^{-3} for n-decane, and 773 kg m^{-3} for n-hexadecane. Literature data for these data are: 703.1, 730.2 and 772.9 kg m^{-3}, for nC_8, nC_{10} and nC_{16}, respectively (Riazi, 2005).

Mixtures were prepared by mixing precisely-measured volumes of pure components together at the ambient temperature of (298 ± 1) K. The amount of each component was then computed from the measured densities leading to mole fractions with an overall expanded uncertainty of 0.003. Measured acoustic velocities for pure hydrocarbons and mixtures are given in Tables 1–7.

Table 1 gives the measured speeds of sound in n-Octane at temperatures from 298 K to 363 K and at pressures up to 100 MPa. Literature data are also given in this table which differ with reported data by 0.1–0.3%. Experimentally measured speeds of sound for n-Decane are given in Table 2 while for n-Hexadecane, results are given in Table 3. In both these cases measurements were restricted to the ambient pressure of 0.1 MPa. In addition to pure hydrocarbons, measurements were carried out for four binary mixtures of n-Octane and n-Hexadecane and three ternary mixtures of (n-Octane + n-Decane + n-Hexadecane) from 298 K to 383 K at pressures up to 100 MPa; the results are given in Tables 4 to 7. These data are used to evaluate the proposed predictive model described in the next section.

3. Predictive method for estimation of speed of sound in petroleum fluids

Speed of sound, c, is a thermodynamic property which is defined as the rate of propagation of small disturbances in a medium and may be considered an isentropic process. With use of

Table 2
Experimental measurement of speed of sound for *n*-decane (*n*-C$_{10}$).

T (K)	P (kPa)	c (m s^{-1})
293.1	106.2	1254.3
293.1	105.0	1254.2
298.1	111.3	1234.4
303.1	107.0	1214.5*
313.1	105.7	1175.3
333.1	104.0	1098.4
353.1	108.4	1023.8
373.2	107.0	951.1
393.2	103.0	880.2

Literature data for *n*-C$_{10}$ at 303.15 K, 0.1 Mpa is 1215.2 m s^{-1} (Dubey and Sharma, 2008).

Table 3
Experimental measurement of speed of sound for *n*-hexadecane (*n*-C$_{16}$).

T (K)	P (MPa)	c (m s^{-1})
298.1	0.1	1338.5
298.1	10.0	1392.2
298.1	20.0	1441.8
298.1	30.0	1488.1
323.1	0.1	1246.6
323.1	10.0	1305.0
323.1	20.1	1359.2
323.1	30.0	1408.9
323.1	40.0	1455.3
323.1	50.0	1490.1
323.1	60.0	1488.8
323.1	70.0	1487.6
323.1	80.0	1492.1
323.1	90.3	1517.2
323.1	99.3	1531.7
343.1	0.1	1176.1
343.1	10.0	1239.1
343.1	20.0	1296.7
343.1	30.0	1349.1
343.1	40.0	1397.9
343.1	50.0	1443.4
343.1	60.0	1482.1
343.1	70.0	1481.7
343.1	80.0	1483.0
343.1	90.2	1483.8
343.1	100.0	1485.4
363.2	0.1	1108.2
363.2	10.0	1176.3
363.2	20.0	1237.3
363.2	30.1	1292.7
363.2	40.1	1343.8
363.2	50.0	1391.1
363.2	60.1	1435.9
363.2	70.0	1475.0
363.2	80.1	1474.8
363.2	90.1	1474.3
363.2	100.3	1475.0
383.2	0.1	1043.1
383.2	10.0	1116.0
383.2	20.0	1181.0
383.2	30.0	1239.7
383.2	40.0	1292.7
383.2	50.1	1342.4
383.2	60.1	1388.5
383.2	70.0	1431.7
383.2	80.0	1472.6
383.2	90.0	1472.5
383.2	100.1	1472.4

Literature data as given by Khasanshin et al. (2008) are: 1338.8 m s^{-1} at 298 K, 0.1 MPa and 1442.8 m s^{-1} at 298 K, 20 MPa.
Literature data as given by Ball and Trusler (2001) are: 1248.11 m s^{-1} at 323 K, 0.1 MPa; 1363.27 m s^{-1} at 323.15 K, 20.956 MPa; and1411.64 m s^{-1} at 323.15 K, 30.642 MPa.

Table 4
Experimental measurement of speed of sound for binary mixtures of *n*-octane and *n*-hexadecane (C$_8$+C$_{16}$) at *T* from (298–383) K and *P* from (0.1–100) MPa.

T (K)	P (MPa)	c (m s^{-1})
x_1 (mol frac), *n*-C$_8$=0.310, x_2 (mol frac), *n*-C$_{16}$=0.690		
298.1	0.1	1306.3
298.1	10.0	1362.0
298.1	20.0	1413.9
298.1	30.1	1462.1
298.1	40.1	1506.5
298.1	50.0	1548.0
323.1	0.1	1213.7
323.1	10.1	1275.1
323.1	20.0	1330.9
323.1	30.0	1382.5
323.1	40.0	1430.1
323.1	50.1	1475.1
323.1	60.0	1516.6
323.1	70.1	1556.7
323.1	80.1	1585.1
323.1	90.1	1592.2
323.1	100.1	1597.3
343.1	0.1	1142.2
343.1	10.0	1208.1
343.1	20.0	1267.8
343.1	30.0	1322.2
343.1	40.1	1372.6
343.1	50.0	1419.2
343.1	60.0	1463.0
343.1	70.1	1504.5
343.1	80.0	1543.4
343.1	90.0	1572.1
343.1	100.1	1577.2
363.2	0.1	1072.4
363.2	10.0	1143.9
363.2	20.0	1207.5
363.2	30.0	1265.0
363.2	40.0	1317.7
363.2	50.0	1366.5
363.2	60.0	1416.7
363.2	70.0	1490.4
363.2	80.0	1514.8
363.2	90.1	1538.3
363.2	100.0	1547.3
x_1 (mol frac), *n*-C$_8$=0.545, x_2 (mol frac), *n*-C$_{16}$=0.455		
298.1	0.1	1271.8
298.1	10.0	1330.0
298.1	20.1	1385.8
298.1	30.0	1433.2
298.1	40.0	1479.2
298.1	50.0	1522.2
298.1	60.1	1563.1
298.1	70.1	1601.6
298.1	80.0	1638.2
323.1	0.1	1180.9
323.1	10.0	1245.0
323.1	20.1	1303.2
323.1	30.0	1356.3
323.1	40.1	1405.7
323.1	50.0	1451.4
323.1	60.0	1494.4
323.1	70.0	1535.1
323.1	80.0	1573.5
323.1	90.1	1610.5
323.1	100.1	1645.7
343.1	0.1	1106.5
343.1	10.0	1175.9
343.1	20.0	1238.2
343.1	30.1	1294.8
343.1	40.1	1346.5
343.1	50.0	1394.5
343.1	60.0	1439.4
343.1	70.1	1481.9
343.1	80.0	1521.8
343.1	90.0	1559.5
343.1	100.0	1595.9
363.2	0.1	1032.9

Table 4 (*continued*)

T (K)	P (MPa)	c (m s^{-1})
363.2	10.0	1109.1
363.2	20.0	1176.0
363.2	30.1	1236.0
363.2	40.1	1290.5
363.2	50.1	1340.6
363.2	60.0	1387.3
363.2	70.0	1431.4
363.2	80.0	1472.9
363.2	90.0	1511.9
363.2	100.0	1549.4

x_1 (mol frac), n-C$_8$=0.729, x_2 (mol frac), n-C$_{16}$=0.271

T (K)	P (MPa)	c (m s^{-1})
298.1	0.1	1239.9
298.1	10.0	1299.0
298.1	20.0	1359.7
298.1	30.1	1409.9
298.1	40.0	1453.5
298.1	49.9	1497.5
298.1	60.0	1539.5
298.1	70.1	1579.3
298.1	80.0	1616.5
298.1	90.0	1652.4
298.1	100.0	1687.0
323.1	0.1	1140.4
323.1	10.0	1208.4
323.1	20.0	1269.0
323.1	30.0	1324.4
323.1	40.0	1375.5
323.1	50.0	1422.8
323.1	60.0	1467.0
323.1	70.0	1508.9
323.1	80.0	1548.3
323.1	90.0	1586.0
323.1	100.0	1621.9
343.1	0.1	1065.0
343.1	10.0	1140.3
343.1	20.0	1204.1
343.1	30.0	1263.0
343.1	40.0	1318.7
343.1	50.0	1366.4
343.1	60.0	1412.8
343.1	70.0	1456.5
343.1	80.0	1497.3
343.1	90.1	1536.4
343.1	100.0	1573.2
363.2	0.1	992.5
363.2	10.0	1067.7
363.2	20.0	1142.0
363.2	30.0	1204.4
363.2	40.0	1261.1
363.2	50.0	1312.9
363.2	60.0	1361.2
363.2	70.0	1406.4
363.2	80.0	1449.1
363.2	90.0	1489.2
363.2	100.2	1528.1
383.2	0.1	920.4
383.2	10.0	1008.7
383.2	20.0	1081.3
383.2	30.1	1149.4
383.2	40.1	1209.0
383.2	50.0	1262.9
383.2	60.1	1313.0
383.2	70.0	1359.8
383.2	80.0	1403.5
383.2	90.2	1445.6
383.2	100.1	1484.2

x_1 (mol frac), n-C$_8$=0.878, x_2 (mol frac), n-C$_{16}$=0.122

T (K)	P (MPa)	c (m s^{-1})
298.1	0.1	1205.8
298.1	10.0	1269.1
298.1	20.0	1327.2
298.1	30.0	1391.7
298.1	40.0	1433.2
298.1	50.0	1474.6
298.1	60.0	1517.4

Table 4 (*continued*)

T (K)	P (MPa)	c (m s^{-1})
298.1	70.0	1557.9
298.1	80.0	1596.2
298.1	90.0	1630.3
298.1	100.0	1667.8
323.1	0.1	1107.8
323.1	10.0	1179.8
323.1	20.0	1241.8
323.1	30.0	1300.8
323.1	40.0	1358.7
323.1	50.0	1401.2
323.1	60.0	1444.4
323.1	70.0	1487.3
323.1	80.0	1527.8
323.1	90.0	1566.0
323.1	100.1	1602.9
343.1	0.1	1027.9
343.1	10.0	1105.8
343.1	20.0	1174.4
343.1	30.0	1235.6
343.1	40.0	1291.2
343.1	50.0	1345.3
343.1	60.0	1391.9
343.1	70.1	1434.5
343.1	80.0	1476.1
343.1	90.0	1516.0
343.1	100.2	1554.6
363.2	10.0	1036.3
363.2	20.1	1110.4
363.2	30.1	1176.8
363.2	40.0	1234.8
363.2	50.0	1288.1
363.2	60.0	1337.9
363.2	70.1	1384.3
363.2	80.0	1429.7
363.2	90.0	1469.0
384.2	100.0	1507.8
384.2	0.1	875.2
384.2	10.0	968.8
384.2	20.0	1048.4
384.2	30.1	1118.5
384.2	40.0	1179.4
384.2	50.0	1235.3
384.2	60.0	1286.8
384.2	70.0	1334.8
384.2	80.0	1379.8
384.2	90.0	1421.9
384.2	100.1	1462.2

thermodynamic relations the speed of sound in a fluid can be given in terms of PVT relations:

$$c^2 = -\left(\frac{\gamma}{M}\right) V_m^2 \left(\frac{\partial P}{\partial V_m}\right)_T \tag{1}$$

where γ is the heat capacity ratio ($\gamma = C_P/C_V$). All quantities in the above relation: V_m, γ and $(\partial P/\partial V_m)_T$ can be calculated from a PVT relation (e.g., a cubic EOS), in the case of γ, the isobaric molar heat capacity of the perfect gas is required. Fig. 4 shows calculated speed of sound for three hydrocarbons versus temperature and at atmospheric pressure. However, such equations of state show poor performance for the density and compressibility of liquid systems, especially for heavy hydrocarbons where the input parameters cannot be estimated accurately (Shabani et al., 1998). A modified cubic equation of state in which the co-volume parameter b was related to molar refraction R_m instead of acentric factor has been proposed for heavy hydrocarbons (Riazi and Mansoori, 1993b). The modified equation of state was shown to provide a good description of the density of hydrocarbon liquids, both pure and mixed, including long-chain alkanes up to n-Tetracontane (C$_{40}$). In this EOS, molar refraction defined below was used as the third

Table 5
Experimental measurement of speed of sound for ternary mixtures (n-C_8+n-C_{10}+n-C_{16}) for composition (vol. fraction) of 0.3 n-C_8, 0.35 n-C_{10} and 0.35 n-C_{16} (0.382C_8, 0.371C_{10}, 0.247C_{16} mole fraction) at T=(298–383) K, and P=(0.1–100) MPa.

T (K)	P (MPa)	c (m s^{-1})
298.1	0.1	1252.4
298.1	10.0	1311.9
298.1	20.0	1370.2
298.1	30.0	1419.6
298.1	40.0	1464.3
298.1	50.0	1508.2
298.1	60.0	1549.3
298.1	70.0	1588.5
298.1	80.0	1625.9
298.1	90.0	1661.4
298.1	100.0	1695.6
323.1	10.0	1222.3
323.1	20.0	1281.9
323.1	30.0	1338.9
323.1	40.0	1388.2
323.1	50.0	1433.5
323.1	60.0	1477.4
323.1	70.0	1518.5
323.1	80.0	1557.7
323.1	90.0	1594.9
323.1	100.0	1630.5
343.1	10.0	1153.4
343.1	20.0	1217.5
343.1	30.0	1275.5
343.1	40.0	1328.2
343.1	50.0	1377.3
343.1	60.0	1423.1
343.1	70.0	1466.0
343.1	80.0	1506.8
343.1	90.1	1545.5
343.1	100.2	1582.5
363.2	10.0	1087.0
363.2	20.0	1155.8
363.2	30.0	1217.3
363.2	40.0	1272.8
363.2	50.0	1324.0
363.2	60.0	1371.8
363.2	70.0	1416.5
363.2	80.1	1458.7
363.2	90.0	1498.4
363.2	100.1	1536.4
383.2	10.0	1023.7
383.2	20.0	1097.3
383.2	30.0	1162.4
383.2	40.0	1220.7
383.2	50.0	1274.2
383.2	60.0	1323.7
383.2	70.0	1369.9
383.2	80.0	1413.6
383.2	90.0	1456.6
383.2	100.0	1492.9

Table 6
Experimental measurement of speed of sound for ternary mixtures (n-C_8+n-C_{10}+n-C_{16}) for composition (vol. fract.) of 0.1 n-C_8, 0.3 n-C_{10} and 0.6 n-C_{16} (0.146C_8, 0.366C_{10}, 0.487C_{16} mole fraction) at temperatures of 298 and 313 K, and pressures ranges of 0.1 to 100 MPa.

T (K)	P (MPa)	c (m s^{-1})
298.1	0.1	1291.0
298.1	10.0	1349.4
298.1	20.1	1400.5
298.1	30.0	1449.0
298.1	40.0	1494.1
298.1	50.0	1536.6
298.1	60.0	1576.5
298.1	70.0	1614.8
323.1	0.1	1197.8
323.1	10.0	1260.4
323.1	20.0	1317.3
323.1	30.0	1369.5
323.1	40.0	1418.0
323.1	50.0	1463.3
323.1	60.1	1506.1
323.1	70.0	1545.9
323.1	80.0	1584.2
323.1	90.0	1620.4
323.1	100.1	1656.0
363.2	0.1	1051.8
363.2	10.0	1127.4
363.2	20.0	1192.7
363.2	30.0	1251.4
363.2	40.0	1304.8
363.2	50.0	1354.4
363.2	60.0	1400.6
363.2	70.1	1444.2
363.2	80.0	1485.0
363.2	90.0	1524.2
363.2	100.2	1562.6
383.2	0.1	986.1
383.2	10.0	1064.4
383.2	20.1	1135.2
383.2	30.0	1196.8
383.2	40.1	1253.1
383.2	50.0	1304.4
383.2	60.1	1352.5
383.2	70.1	1397.5
383.2	80.0	1439.7
383.2	90.0	1479.7
383.2	100.0	1519.3

parameter:

$$R_m = V_m I \tag{2}$$

where I depends upon the refractive index n as follows:

$$I = \frac{(n^2 - 1)}{(n^2 + 2)} \tag{3}$$

Experimentally and theoretically, R_m depends weakly on temperature and pressure (Hirschfelder et al., 1964). Values of R_m at reference conditions of T=293.15 K and P=101.325 kPa are available for hydrocarbons up to C_{40} (Riazi and Mansoori, 1993b). For components that are not liquid under these conditions, R_m was evaluated for the saturated liquid at its normal boiling point. A dimensionless reduced molar refraction was then defined by

dividing R_m of a compound to its value for methane:

$$r = \frac{R_m}{(6.987 \text{ cm}^3 \text{ mol}^{-1})} \tag{4}$$

Then an equation of state can be expressed in a dimensionless form for molar volume $V_m(P_c/RT_c)$ as a universal function of reduced temperature $T_r = T/T_c$, reduced pressure $P_r = P/P_c$ and reduced molar refraction r. Parameter r has been also used as a third parameter for the prediction of the viscosity of heavy hydrocarbons (Riazi et al., 2005). On this basis, a three-parameter corresponding-states model is proposed for the reduced speed of sound in heavy hydrocarbon liquids as follows:

$$c_r = f^0(T_r, P_r) + r^{-1}f^1(T_r, P_r) \tag{5}$$

where c_r is a reduced speed of sound, c/sf, in which sf is a scaling factor; f^0 and f^1 are universal functions of T_r and P_r to be determined from data on two reference compounds. The scaling factor was used in lieu of critical data for the speed of sound. It was calculated using Eq. (1), assuming a constant value for γ and using PR EOS for calculation of $(V_m)^2_{T_c}(\partial P/\partial V_m)_{T_c}$ term at critical temperature. To have standard values for sf of heavy hydrocarbons the following relation was developed based on calculated data for

Table 7
Experimental measurement of speed of sound for ternary mixtures (n-C_8+n-C_{10}+n-C_{16}) for composition (vol. fract.) of 0.2 n-C_8, 0.4 n-C_{10} and 0.4 n-C_{16} (0.265C_8, 0.442C_{10}, 0.0.294C_{16} mole fraction) at T=(298–383) K, and P=(0.1–100) MPa.

T/K	P (MPa)	c (m s^{-1})
298.1	0.1	1263.4
298.1	10.0	1321.9
298.1	20.0	1380.3
298.1	30.0	1426.0
298.1	40.0	1472.6
298.1	50.0	1515.9
298.1	60.0	1556.9
298.1	70.0	1595.5
298.1	80.0	1632.5
298.1	90.0	1667.7
298.1	100.1	1702.2
323.1	10.0	1232.5
323.1	20.0	1291.7
323.1	30.0	1345.4
323.1	40.0	1395.3
323.1	50.0	1441.5
323.1	60.0	1485.0
323.1	70.0	1525.9
323.1	80.0	1564.7
323.1	90.0	1601.7
323.1	100.0	1637.0
343.1	10.0	1165.4
343.1	20.0	1227.6
343.1	30.0	1284.4
343.1	40.0	1337.0
343.1	50.0	1385.4
343.1	60.0	1430.8
343.1	70.0	1473.7
343.1	80.0	1513.9
343.1	90.0	1552.2
343.1	100.0	1588.5
363.2	10.0	1098.5
363.2	20.0	1166.2
363.2	30.0	1226.7
363.2	40.0	1281.7
363.2	50.0	1332.3
363.2	60.0	1379.6
363.2	70.0	1424.0
363.2	80.0	1465.8
363.2	90.0	1505.2
363.2	100.0	1542.8
384.2	10.0	1032.2
384.2	20.0	1104.8
384.2	30.0	1169.2
384.2	40.0	1227.1
384.2	50.0	1280.0
384.2	60.0	1329.2
384.2	70.0	1375.1
384.2	80.0	1418.4
384.2	90.0	1458.7
384.2	100.1	1497.6

Fig. 4. Calculated speed of sound from a cubic EOS for pure liquid hydrocarbons at atmospheric pressure.
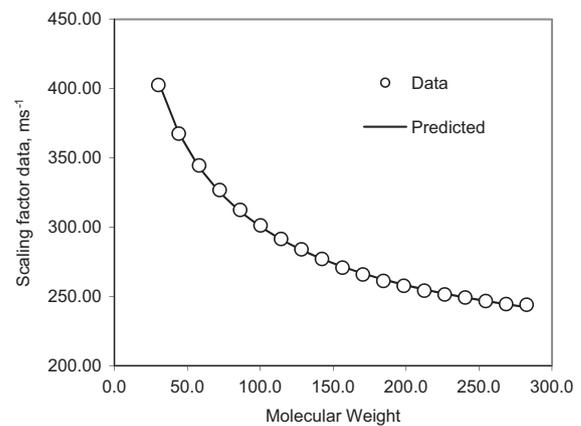
Fig. 5. Estimation of scaling factor for n-alkanes from Eq. (6).

n-alkanes from C_1 to C_{20}:

$$sf = 200 + \exp(6.9745 + 0.5945\, M^{0.3}) \tag{6}$$

where M is the molecular weight of hydrocarbon. This relation is valid for n-alkane hydrocarbons and reproduces original data with AAD of 0.28%. Values of scaling factor calculated from Eq. (6) and a comparison with the original data (calculated from PR EOS) is shown in Fig. 5. Critical properties for pure hydrocarbons used in this study are given in Table 8. Experimental data on the speed of sound used in this work was obtained from open literature for liquid hydrocarbon systems as given in Table 9.

Based on data available in Table 9, hydrocarbon compounds n-C_{18} and n-C_{28} were chosen as two reference fluids for use in Eq. (5), because they represent heavy compounds in our data bank and have wider temperature and pressure ranges with larger data as shown in Table 9. The linear relation between c_r and $1/r$ for
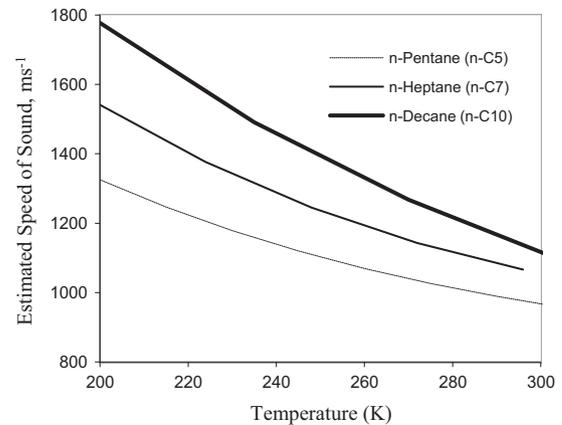
hydrocarbons from C_{18} to C_{36} at T_r=0.5 and P_r=10 is demonstrated in Fig. 6. In order to increase accuracy of proposed method for lighter hydrocarbons, data for C_3 was also used in addition to n-C_{18} and n-C_{28} for development of the corresponding states relation in the form of Eq. (5). The resulting equation is in the following form:

$$c_r = (9.971 - 10.087\, T_r + 0.0005 P_r) + (-9.5 + 12.536\, T_r + 0.244 P_r)/r$$

$$c_r = \frac{c}{sf}, \quad T_r = \frac{T}{T_c}, \quad P_r = \frac{P}{P_c} \tag{7}$$

The standard values of d_{20}, I, T_c and P_c for heavy hydrocarbons were calculated from the following relations specifically proposed for heavy n-alkanes as given in the ASTM Manual (Riazi, 2005).

$$\frac{d_{20}}{d_{ref}} = 0.859 - \exp(99.01379 - 85.744 M^{0.01}) \tag{8}$$

$$I = 0.2833 - \exp(87.6593 - 86.62167 M^{0.01} \tag{9}$$

$$\frac{T_c}{K} = \frac{\left(1070 - \exp(6.98291 - 0.02013 M^{2/3}\right)}{1.15 - \exp(-0.41966 - 0.02436 M^{0.58})} \tag{10}$$

$$\frac{P_c}{MPa} = 0.1\exp(4.65757 - 0.13423 M^{0.5}) \tag{11}$$

In the above relations d_{20} is the density of hydrocarbon at 20 °C and d_{ref} is the density of water at 4 °C ($=1\,\text{kg L}^{-1}$). M is the molecular weight and for n-alkanes it can be calculated from

Table 8
Properties of pure hydrocarbons used for evaluation purposes.

| No.	Hydrocarbon	Carbon number	MW	T_c (K)	P_c (bar)	r	sf (m s^{-1})
1	Propane	C_3	44.1	369.8	42.48	2.259	367.9
2	n-Butane	C_4	58.1	425.1	37.96	2.951	343.1
3	n-Hexane	C_6	86.2	507.4	30.31	4.246	311.2
4	n-Heptane	C_7	100.2	540.3	27.49	4.905	300.1
5	n-Octane	C_8	114.2	569.2	25.10	5.556	291.1
6	n-Decane	C_{10}	142.3	618.2	21.25	6.890	277.0
7	n-Hexadecane	C_{16}	226.4	720.6	13.98	10.864	251.9
8	n-Octadecane	C_{18}	254.5	745.3	12.38	12.189	246.6
9	n-Nonadecane	C_{19}	268.5	756.4	11.68	12.852	244.3
10	n-Tricosane	C_{23}	324.6	794.3	9.39	15.502	236.8
11	n-Tetracosane	C_{24}	338.6	802.4	8.91	16.165	235.2
12	n-Octacosane	C_{28}	394.7	830.5	7.32	18.816	230.0
13	n-Hexatriacontane	C_{36}	507.0	871.8	5.13	24.160	222.7
Overall		C_3–C_{36}	44–507	369–872	5–42	2.3–24.1	222–368

Calculated from Eqs. (6) and (8)–(11).

Table 9
Evaluation of proposed method for pure hydrocarbons.

No.	Carbon number	No. of data	T range (K)	P range (MPa)	C range (m s^{-1})	T_r range	P_r range	C_r range	% Abs. Dev. Avg.	% Abs. Dev. Max.	Ref.
1	C_3	240	200–340	0.04–60	863–1582-	0.54–0.92	0.01–14.15	1.58–4.58	3.65	24.1	a*
2	C_4	250	200–375	0.02–60	618–1718	0.47–0.88	0.005–15.81	1.64–5.01	2.88	13.1	a
3	C_6	16	263–341	0.1	892–1224	0.52–0.67	0.03	287–3.93	3.35	7.1	b
4	C_7	15	265–368	01	820–1270	0.490.68	0.04	2.73–4.23	4.38	6.4	b
5	C_8	16	265–392	0.1	776–1314	0.46–0.69	0.04	2.67–4.51	4.46	8.5	b
6	C_{10}	14	268–391	0.1	859–1342	0.43–0.63	0.05	2.78–4.34	5.38	8.5	b
7	C_{16}	13	287–394	0.1	967–1354	0.40–0.55	0.07	3.84–5.37	5.57	9.8	b
8	C_{18}	141	313–383	0.1–149.6	1067–1804	0.42–0.52	0.08–120.78	3.87–6.59	1.21	4.1	c
9	C_{19}	134	313–383	0.1–149.6	1244–1811	0.41–0.51	0.09–128.02	3.96–6.57	1.18	3.9	c
10	C_{23}	112	333–393	0.1–149.6	1080–1790	0.42–0.50	0.11–159.35	4.57–7.55	1.25	4.3	d
11	C_{24}	105	333–393	0.1–149.6	1082–1807	0.41–0.49	0.1–167.80	4.63–7.61	1.24	4.2	d
12	C_{28}	108	353–403	0.1–149.6	1084–1788	0.42–0.49	0.14–204.3	4.71–7.77	1.26	2.4	e
13	C_{36}	74	363–403	0.1–141.3	1118–1755	0.41–0.46	0.19–275.3	5.55–7.88	1.20	5.3	e
Measured data in this work (Tables 1–3)											
14	C_8	53	298–383	0.1–100	921–1725	0.52–0.67	0.04–39.8.	3.17–5.96	2.66	7.01	Table 1
15	C_{10}	8	293–393	0.11	880–1254	0.47–0.64	0.05	3.10–4.84	4.14	5.0	Table 2
16	C_{16}	48	298–393	0.1–100	1010–1339	0.41–0.55	0.08	4.01–5.31	2.73	10.57	Table 3
Overall	C_3 –C_{36}	1303	200–403	0.02–149.6	618–1811	0.4–0.92	0.005–275.3	1.58–7.88	2.26	24	

* a: Niepmann (1984), b: Wang and Nur (1991), c: Dutour et al. (2000), d: Dutour et al. (2001), e: Dutour et al. (2002a, 2002b).
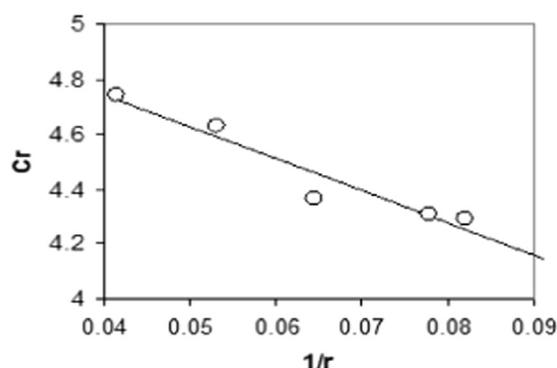
Fig. 6. The relation between reduced speed of sound and reduced molar refraction for n-alkanes from C_{18} to C_{36} at $T_r=0.5$ and $P_r=10$.
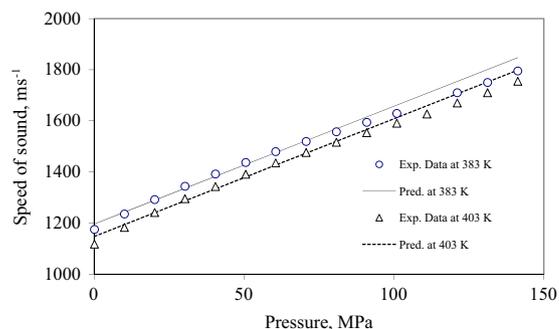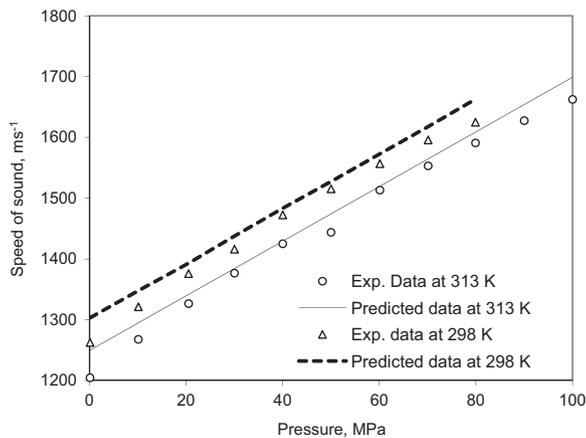

Fig. 7. Prediction of speed of sound for n-C_{36} at 383 and 403 K.

Table 10
Evaluation of proposed method for defined hydrocarbon mixtures.

No.	Mixture	x_1 range	MW range	T range (K)	P range (MPa)	C range (m s^{-1})	No. of points	% Deviation Avg.	% Deviation Max.	Reference
1	Hexane+hexadecane	0.2–0.8	114–198	298–348	0.1–70	1004–1530	345	1.38	6.0	a*
2	Methylcyclohexane+hexane	0.0–0.9	86–97	298	0.1	1080–1203	11	2.26	5	b
3	Methylcyclohexane+dodecane	0–1.0	98–170	298	0.1	1217–1284	10	1.96	5.0	b
4	$C_{17}H_{36}+C_{16}H_{32}+C_{16}H_{26}$	0.4+0.35N+0.25A	229.2	293.303	0.1–60	1400–1775	10	1.5	2.6	c
5	$C_{13}H_{28}+C_{13}H_{26}+C_{13}H_{20}$	0.4P+0.35N+0.25A	181	293–373	0.1–150	1080–1800	279	1.79	5.0	c
6	C_8–C_{16}	0.31–0.88	114–226	298–383	0.1–100	915–1745	190	1.9	6.1	Table 4 (this work)
7	C_8–C_{10}–C_{16}	0.368+0.359+0.239	152.2	298–383	0.1–100	1102.6–1749	51	1.8	4.9	Table 5 (this work)
8	C_8–C_{10}–C_{16}	0.147+0.366+0.487	179.1	298–383	0.1–100	1299–1673	14	1.88	5	Table 6 (this work)
9	C_8–C_{10}–C_{16}	0.245+0.410+0.273	159.4	298–383	0.1–100	1116.1–1752.5	51	1.74	4.7	Table 7 (this work)
Overall	Binary and ternary	0.0–1.0	86–229	298–383	0.1–150	1004–1800	1403	1.97	6.0	

* References: a: Wang et al. (1990), b: Daridon et al. (1998), c: Baragi et al. (2006).

Fig. 8. Prediction of speed of sound for a ternary mixture of C_8–C_{10}–C_{16} with composition of (0.265C_8, 0.442C_{10}, 0.294C_{16} mole fraction)—Data are taken from Table 7.

carbon number N_c as:

$$M = 14.026N_c + 2.016 \qquad (12)$$

Table 9 also shows results of evaluation of the proposed method for more than 1303 data points for pure hydrocarbons from C_3 to C_{36} and pressures from 0.02 to 150 MPa. The average deviation (%AAD) between predicted and experimental data is about 2.26%. As Eq. (7) was developed based on data for these two compounds, the method can also be applied to heavier hydrocarbons considering the linear relation between C_r and $1/r$ which is demonstrated in Fig. 6. Graphical evaluation of Eq. (7) for n-C_{36} at different temperatures and pressure up to 140 MPa is presented in Fig. 7. As shown in this figure for pressures from 0.1 to 100 MPa (1 to 1000 bar) predicted values of speed of sound are so close to the actual experimental values and deviations begin at extremely high pressures above 1000 bar. As the main intended application of proposed method is for reservoir fluids and under reservoir conditions (well below 100 MPa) it can be considered a reliable method. Most of corresponding states correlations are developed based on very limited data while they show good performance once applied to a wide range of conditions and compounds (API, 1997). Similarly it is expected that the proposed CSP relation for the speed of sound (Eq. (7)), to be applicable to a wide range of compounds as demonstrated in the next section.

4. Application to mixtures and evaluations

Corresponding state relation as given by Eq. (7) can be directly applied to mixtures if the composition is known. In such application, T_c, P_c, sf, and r should be calculated for the mixture using Kay's mixing rule as the simplest mixing rule for calculation of these pseudo-properties.

$$\theta_{mix} = \sum x_i \theta_i \qquad (13)$$

where x_i is mole fraction of component i and θ is a parameter such as T_c or P_c. However, since in this work we have studied n-alkane mixtures it was found that it is simpler (with same degree of accuracy) to calculate M for the mixture and to use it in Eqs. (8) to (11) for calculation of all input parameters needed for Eq. (7). This method can also be applied to mixtures which are mainly paraffinic but also contain non-paraffinic compounds.

For a defined mixture (known composition, x_i) the molecular weight (M_{mix}) is calculated as:

$$M_{mix} = \sum_i x_i M_i \qquad (14)$$

where x_i is mole fraction of component i in the mixture. For unknown mixtures such as petroleum fractions and crude oils the whole mixture molecular weight can be calculated by the methods suggested in the ASTM Manual (Riazi, 2005).

Summary of evaluation of the proposed methods for prediction of speed of sound in some binary and ternary systems is given in Table 10 and for some 1403 data points %AAD is 1.97%. In this table for systems 2, 3, 4 and 5 the mixtures contain some non-paraffinic hydrocarbons including naphthenes and aromatics. For systems 6, 7, 8 and 9 data have been reported from the measurements in this work. For both ternary and binary systems the errors are within the range of $\pm 2\%$ as shown in Table 10.

Fig. 8 shows evaluation of the method for a defined ternary mixture. For this mixture data are given in Table 7 and evaluated as System #9 in Table 10. Application of the method to undefined petroleum products and crude oil is shown in Table 11. Experimental data reported in this table were measured by Wang et al. (1990). For a relatively light petroleum fraction with API gravity of 43, the evaluation is shown graphically in Fig. 9. For an undefined wider boiling range and heavier mixture (a crude oil) with API gravity of 23 evaluation of proposed method is shown in Fig. 10. The new developed correlation was tested for seven crude oils in which the only available data were density at 15 °C. Specific gravity (taken the same as density at 15.5 °C) was used as the sole parameter to calculate molecular weight assuming the whole
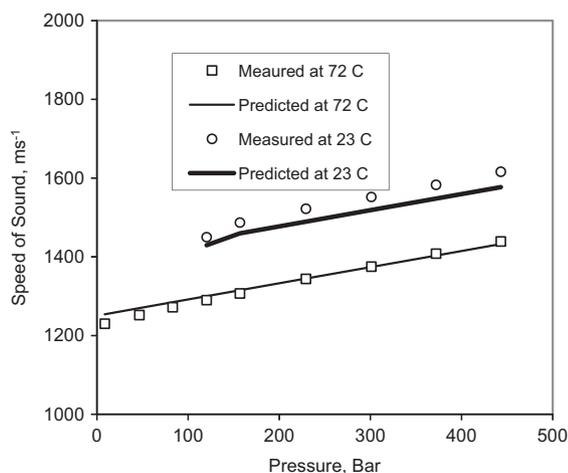
Table 11
Evaluation of proposed method for undefined mixtures (crude oils).

No.	Oil sample	MW	API gravity	T range (K)	P range (MPa)	C range (m s^{-1})	No. of points	% Deviation	
								Avg.	Max.
1	Oil F	243	34	298–380	0.1–44.3	1085–1599	38	2.01	5.0
2	Oil G	193	43	297–359	0.8–44.3	1088–1556	24	3.8	6.4
3	Oil H	130	57	296–363	0.1–44.3	991–1356	35	2.2	6.6
4	Oil I	128	62	296–382	0.1–44.3	904–1471	43	2.7	6.8
5	Oil J	290	23	296–345	0.8–44.3	1230–1616	16	1.89	3.9
Overall	Crude oil and products	128–290	23–62	296–382	0.1–44.3	904–1616	156	2.4	6.7

Data are taken from Wang et al. (1990).
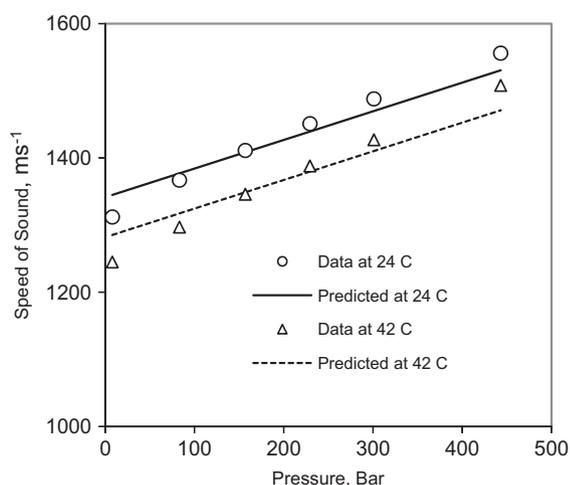
Fig. 9. Prediction of speed of sound for an undefined petroleum fraction with API Gravity of 43 at 297 K (24C) and 315 K (42C) (Oil G in Table 11).

Fig. 10. Prediction of speed of sound for a crude oil with API Gravity of 23 at 296 K (23C) and 345 K (72C). (Data from Table 11 for Oil J).

crude is like one single carbon number group as discussed by Riazi and Al-Sahaf (1996). Table 12 shows an average deviation (%AAD) between predicted and experimental data at ambient pressure of 1.3% and Fig. 11 presents the graphical comparison between the experimental and predicted values of speed of sound.

5. Sample calculation

To show how the proposed method can be applied to a defined mixture consider a mixture of 30% n-C_8, 35% n-C_{10} and 35% n-C_{16} (composition in vol%) and for this mixture it is required to estimate speed of sound at 40C and 79.9 MPa.

The system is composed of three components with n-C_8 (component 1), n-C_{10} (component 2) and n-C_{16} (component 3). The carbon numbers for these three n-alkane hydrocarbons are 8, 10 and 16, respectively, and through use of Eq. (12) we get: $M_1 = 114.2$, $M_2 = 142.3$ and $M_3 = 226.4$. From Eq. (8) densities of these compounds are calculated as: $d_1 = 0.7081$, $d_2 = 0.7352$ and $d_3 = 0.7776$. Using these values composition of the mixture is calculated in terms of mole fractions: $x_1 = 0.382$, $x_2 = 0.371$, $x_3 = 0.247$. From Eq. (14), $M_{mix} = 152.35$. Using this value for the mixture molar mass and through Eqs. (11)–(13) we calculate: $d_{20} = 0.7426$, $I = 0.2508$, $T_c = 633.5$ K, $P_c = 20.10$ bar. From Eq. (4), $R_m = 51.46$ and from Eq. (6), $r = 7.365$ (or $1/r = 0.136$). From Eq. (7) at $T_r = 0.494$ and $P_r = 40.28$ value of c_r is calculated as: $c_r = 5.891$. From Eq. (6), $sf = 272.9$ m s^{-1} and finally c is calculated as $c = sf \times c_r = 5.891 \times 272.9 = 1607.6$ m s^{-1}. Comparing this value with measured value of 1590.8 m s^{-1} gives an error of $+1.05\%$ which is within expected range of error.

6. Conclusions

In this paper experimental data on the velocity of sound for some hydrocarbons systems at various temperature, pressure and composition from atmospheric to 100 MPa were reported. In addition a simple relation based on the principles of corresponding states and with use of molar refraction has been proposed which can be used for prediction of speed of sound in liquid hydrocarbon systems for pressures up to 150 MPa. Although the method was developed based on two reference n-alkane hydrocarbon compounds but it is applicable to a wide range of pure compounds, defined and undefined mixtures. The proposed method was evaluated against some 1303 data points for pure hydrocarbons from C_3 to C_{36} and pressures up to 150 MPa, some binary and ternary mixtures (961 data points) as well as some petroleum fractions and crude oils (156 data points) and an overall average error of about 2% was obtained for over 2400 data points. The evaluations show that the proposed method can be used for very heavy hydrocarbons and heavy oil systems which may contain light gases. The main required parameter is mixture molar mass and some standard methods are provided for calculation of required parameters for the proposed method. The results could be used to estimate other thermophysical properties needed for heavy oil exploration and production.

Table 12
Evaluation of proposed method for crude oils with density data.

No.	Oil sample	MW (kg kmole^{-1})	Density@288 (K)	API Gravity	T (K)	P (MPa)	Calculated c (m s^{-1})	% Deviation
1	Crude 1	100.1	0.7363	60.68	295	0.10	1176.2	1.70
2	Crude 2	188.4	0.8238	40.26	296	0.10	1343.9	0.74
3	Crude 3	209.3	0.8364	37.68	295	0.10	1359.3	0.25
4	Crude 4	214.4	0.8392	37.11	295	0.10	1362.5	1.29
5	Crude 5	215.9	0.8400	36.95	295	0.10	1363.3	2.5
6	Crude 6	237.0	0.8506	34.85	295	0.10	1374.0	2.4
7	Crude 7	275.9	0.8670	31.71	295	0.10	1387.6	0.19
								ADD=1.3

Data were provided by Trusler (2010).
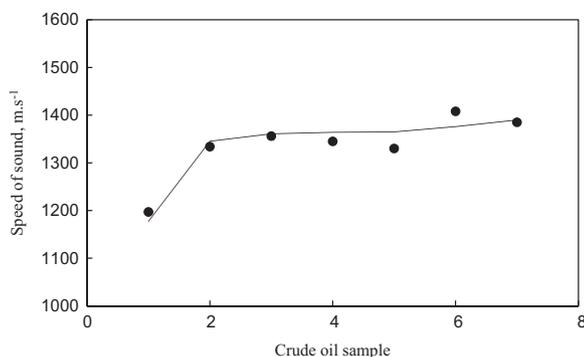
Fig. 11. Prediction of speed of sound for 7 crude oils with density at specific temperature as the only available data (from Table 12).

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