

Experimental determination of sound velocity in liquefied propane-butane gas mixture (LPG)

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Abstract

In this article, experimentally determined values of sound velocity in LPG in a temperature range ($-23\dots+55$) °C are presented. The measurements were taken using two industrial LPG samples in a standard five liter pressurized gas tank. The propagation time of an ultrasonic signal with the frequency of 0.7 MHz was measured. To increase the accuracy of measurement, a pulse-phase method was applied in our measurement system. The propagation time of the acoustical signal through the LPG was measured by filling the measurement interval with pulses of 100 MHz frequency. The propagation of ultrasonic signal through the walls of the tank was eliminated by applying a special method. Measurements showed that the amplitude of the signal, which has propagated through the LPG, has attenuated by about 30% in the selected temperature range. The curve of sound velocity for LPG, with 71.74 % of propane, is below the curve for LPG with 27.80 % propane. At the temperature of 20.0 °C, sound velocities in the samples are 810.16 m/s (sample with 71.74 % of propane) and 840.31 m/s (sample with 27.80 % of propane). The sound velocity in the previously mentioned temperature range is close to linear dependence versus temperature. The sound velocity differences in industrial LPG samples with different propane-butane ratios can be used to develop a method to determine the composition of LPG, in which this ratio is unknown.

Keywords: velocity of sound, ultrasound velocity, propane, butane, fluid mixture, liquid mixture, liquefied petroleum gas, LPG, thermophysical properties

Introduction

The measurement of parameters of multi-component fluid mixtures is important when applying these mixtures in various industrial processes, for example, when designing devices for the oil and gas industry. Thermophysical and chemical properties are the most important parameters.

One of the multi-component fluid classes is LPG (liquefied petroleum gas). The industrial LPG is composed of (in various percentages): ethane (C_2H_6), propane (C_3H_8), propene (C_3H_6), isobutane (C_4H_{10}), n-butane (C_4H_{10}), *cis*-but-2-ene (C_4H_8), 1,3-butadiene (C_4H_6) and isopentane (C_5H_{12}). Small traces of these gases are also present: methane (CH_4), but-1-ene (C_4H_8), isobutylene (C_4H_8), *trans*-but-2-ene (C_4H_8), n-pentane (C_5H_{12}) and pentane (C_5H_{12}).

An acoustic method using the ultrasound time delay measurement technique has been developed to determine thermophysical parameters of fluid mixtures [1]. The propagation velocity of ultrasonic waves in n-hexane was measured by a pulse transmission-reflection instrument operating at 3 MHz [2]. This frequency constitutes an acceptable compromise between lower frequencies, which give clearer signals at a reduced accuracy, and higher frequencies, which provide a better accuracy of measurements, but with a greater attenuation of the waves.

The thermophysical properties of pure propane and butane (the compositional elements of LPG), including the dependence of sound velocity on temperature, are described in [1, 3-14].

The sound velocity in hydrocarbon mixtures has been measured previously over a wide range of pressures and temperatures by several authors [5-14]. In the results

provided by these authors were some inconsistencies. This justified our present measurements to some extent.

Also it proved unsuccessful to find data on a sound velocity, measured in real industrial LPG mixtures. Because of this, we will present our experimental measurements of sound velocity in a temperature range, which is most widely encountered in practical applications.

Measurements were performed using two industrial LPG mixtures with different percentages of liquefied propane and butane. Since the sound velocity proved to be different in LPG with various component percentages, it can be used to determine the composition of LPG, in which the propane-butane ratio is unknown.

Knowing the exact composition is important, because when practically using LPG during different seasons of the year, the propane-butane ratio determines the steaming rate.

The combustion speed and caloric content of LPG is determined by this ratio as well.

Theoretical part

At the frequencies of a few MHz, the ultrasound speed u and the speed of sound C_0 extrapolated to zero frequency can be matched in the case of hydrocarbons in the liquid state [2]. It follows from the assumption that the usual thermodynamic relations which relate the speed of sound to thermodynamic properties of the fluid can also be used for the ultrasound speed [2]:

$$u = \sqrt{\left(\frac{\partial P}{\partial p}\right)_S} = \sqrt{\frac{1}{\rho\kappa_S}}, \quad (1)$$

where

$$\kappa_S = \kappa_T - \frac{T\alpha_p^2}{\rho C_p}, \quad (2)$$

where ρ , α_p , κ_s , κ_T , and C_p are, respectively, the density, the isobaric thermal expansivity, the isentropic and isothermal compressibilities, and the isobaric heat capacity.

Experimental measurements

Object of measurements

The object for selected investigation was two industrial LPG samples in a standard five liter pressurized gas tank. The composition of these liquefied hydrocarbon gas samples are presented in Table 1. The parameters of these samples are presented in Table 2.

Table 1. Composition of industrial LPG samples

Component name	Concentration of sample Nr. 1, %	Concentration of sample Nr. 2, %
methane	0.00	0.00
ethane	0.72	1.47
propane	68.62	18.00
propene	3.12	9.80
isobutane	6.37	13.40
n-butane	18.80	53.64
but-1-ene	0.00	0.00
isobutylene	0.00	0.00
<i>trans</i> -but-2-ene	0.00	0.00
<i>cis</i> -but-2-ene	0.59	0.94
1,3-butadiene	0.46	0.51
isopentane	1.32	2.24
n-pentane	0.00	0.00
pentane	0.00	0.00

Table 2. Parameters of industrial LPG samples

Parameter name	Sample Nr. 1, value	Sample Nr. 2, value
pressure of gas at 45 °C, MPa	1.229	0.868
density at 20 °C, kg/m ³	510.8	552.2
density at 15 °C, kg/m ³	517.6	559.2

Ultrasound speed measurements

To measure the speed of sound in these liquids, a device was designed in our laboratory. The electro-acoustic part of this device was composed of two piezoceramic transducers mounted on the most curved sections of the opposite sides of the tank.

The propagation time of a signal with the frequency of 0.7 MHz was measured. To increase the accuracy of measurement, a pulse-phase method was implemented in our measurement system. The propagation time of the acoustical signal through LPG was measured by filling the measurement interval with pulses of 100 MHz frequency. The propagation of ultrasonic signal through the walls of the tank was eliminated by applying a special method.

Preliminary measurements of sound velocity showed that the amplitude of the signal, which has propagated through LPG, was attenuated by about 30 % in the selected temperature range. The attenuation of the signal's

amplitude is proportional to the increase in temperature. The amplitude of this signal was monitored using an oscilloscope. This enabled us to evaluate changes of amplitude of that signal. During the whole experiment, amplitude of the received signal was always kept constant.

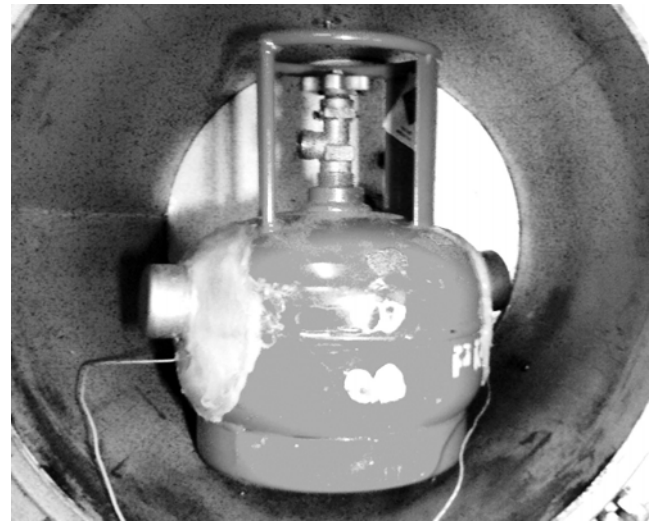


Fig.1. Electro-acoustic part of ultrasonic speed measurement system

The temperature was measured using a mercury-based thermometer with a scale of 0.1 °C. The temperature throughout the whole volume of LPG was uniform when the readings of the thermometer were stable and the propagation time of ultrasonic signal was constant. At that time the readings were recorded.

Results of experimental measurements

During the experiment, the temperature of the LPG was changed in selected intervals. Between these intervals, signal propagation times through LPG in the measurement tank were recorded. This data is presented in Table 3 and Fig. 2. One can see from Fig. 2 that the curve of propagation time for LPG, with 71.74 % of propane, is above the curve for LPG with 27.80 % propane.

Using the experimental propagation time data, expressions to calculate propagation time at any temperature were extrapolated.

For LPG with 71.74 % propane, the propagation time $t_{71.74}$ (μ s) is calculated using expression:

$$t_{71.74} = 0.0162T^2 + 1.5939T + 238.96, \quad (3)$$

where T is the temperature of LPG in the Celsius scale.

For LPG with 27.80 % propane, the propagation time $t_{27.80}$ (μ s) is calculated using expression:

$$t_{27.80} = 0.0124T^2 + 1.4544T + 233.32. \quad (4)$$

For sound velocity calculation when the propagation time is known, a widely used method was applied [15]. The sound velocity in water was used as the standard for determining the path length of ultrasonic signals. The measurement tank was filled with distilled water, in which the sound velocity is known [16-19]. The selected temperature for water was 20.0 °C. At this temperature the sound velocity is 1482 m/s. In this case, the propagation

time in water was 151.6 μs. The sound velocity in LPG was calculated using expression:

$$u_{LPG} = \frac{u_w t_w}{t_{LPG}}, \quad (5)$$

where u_w , t_w and t_{LPG} , are, respectively, the sound velocity in water, the signal propagation time in water and the signal propagation time in LPG.

To calculate the sound velocity in LPG, the extrapolated expressions (3 and 4) for calculating propagation time at given temperatures were used. The calculated velocities at these temperatures are presented in Table 3 and Fig. 3.

Using these calculated sound velocities, expressions for calculating sound velocity at a given temperature were extrapolated.

For LPG with 71.74 % propane, the ultrasound velocity $u_{71.74}$ (m/s) is calculated using expression:

$$u_{71.74} = 0.0004T^3 - 0.0224T^2 - 6.1989T + 940.04, \quad (6)$$

where T is the temperature of LPG in the degrees of Celsius.

For LPG with 27.80 % propane, ultrasound velocity $u_{27.80}$ (m/s) is calculated using expression:

$$u_{27.80} = 0.0003T^3 - 0.0131T^2 - 5.9648T + 962.74. \quad (7)$$

Table 3. Thermophysical parameters of industrial LPG samples

71.74% Propane						27.80% Propane					
T, °C	t _{LPG} , μs	u _{LPG} , m/s	T, °C	t _{LPG} , μs	u _{LPG} , m/s	T, °C	t _{LPG} , μs	u _{LPG} , m/s	T, °C	t _{LPG} , μs	u _{LPG} , m/s
-25.3	208.2	1075.0	14.5	266.7	846.3	-18.7	210.0	1067.5	24.7	276.6	811.6
-25.2	208.4	1074.6	15.0	267.6	843.0	-18.5	210.3	1066.5	25.0	277.7	809.8
-24.4	209.0	1071.3	15.3	268.1	841.0	-18.0	210.5	1064.0	25.8	279.7	805.0
-23.8	209.5	1068.8	16.0	269.4	836.4	-17.6	210.6	1062.0	26.2	280.5	802.6
-21.8	211.4	1060.2	17.0	271.8	829.8	-17.3	211.0	1060.4	26.7	281.6	799.6
-21.4	211.9	1058.4	17.4	272.8	827.2	-16.9	211.7	1058.4	27.1	281.5	797.2
-20.8	212.6	1055.7	17.6	273.0	825.9	-16.5	212.0	1056.3	28.0	283.6	791.8
-20.6	212.8	1054.8	20.5	278.8	806.9	-14.2	215.5	1044.2	30.0	287.0	779.8
-12.5	222.0	1014.0	21.0	280.1	803.6	-10.6	219.9	1024.5	31.1	290.5	773.3
-11.4	223.5	1008.0	23.5	285.0	787.3	-10.5	219.7	1024.0	31.7	291.5	769.7
-11.0	224.0	1005.7	24.5	287.0	780.8	-9.6	221.0	1018.9	33.0	295.0	762.1
-10.6	224.5	1003.5	24.8	288.0	778.9	-7.5	224.0	1007.0	33.5	295.3	759.1
-5.1	231.2	971.5	25.5	289.4	774.4	-6.4	226.0	1000.7	34.7	297.5	752.1
-5.0	231.5	970.9	26.0	291.0	771.1	-2.5	230.5	977.8	37.0	304.0	738.8
-3.4	233.5	961.3	26.5	292.0	767.9	-2.1	231.2	975.5	37.5	306.0	735.9
-3.1	234.1	959.4	27.5	294.0	761.5	2.0	237.0	950.9	38.0	307.0	733.0
-2.8	234.8	957.6	28.0	295.5	758.3	2.4	237.8	948.5	39.0	309.5	727.3
-1.9	236.0	952.0	28.5	297.0	755.1	2.8	238.2	946.0	41.0	313.0	716.0
-1.1	237.1	947.1	29.8	300.0	746.8	5.3	241.7	930.8	44.0	320.5	699.2
-0.8	237.8	945.2	30.5	301.6	742.4	5.6	242.2	929.0	47.0	330.0	682.7
-0.6	238.5	944.0	31.5	304.0	736.0	6.0	242.8	926.5	50.0	338.0	666.6
0.4	239.5	937.7	32.0	305.5	732.9	6.3	243.0	924.7			
2.0	242.1	927.6	33.0	308.2	726.6	7.6	245.0	916.7			
2.4	243.0	925.0	33.5	309.0	723.5	7.7	245.3	916.1			
2.8	243.8	922.5	34.0	310.2	720.4	8.0	246.0	914.2			
3.3	244.8	919.3	34.5	311.5	717.3	9.0	247.4	908.1			
4.7	246.8	910.3	35.0	313.0	714.2	9.1	248.0	907.5			
6.0	248.8	901.9	35.3	314.0	712.3	10.0	249.0	901.9			
6.8	250.4	896.7	35.8	315.0	709.2	10.0	248.3	901.9			
7.3	251.4	893.5	36.5	317.4	704.9	10.5	250.0	898.8			
8.6	253.8	885.0	37.5	320.0	698.8	13.5	255.0	880.3			
9.3	255.4	880.4	37.8	321.0	697.0	13.6	255.2	879.7			
9.6	256.0	878.5	39.5	325.8	686.7	15.6	258.6	867.4			
10.2	257.4	874.5	40.5	329.0	680.6	18.5	263.8	849.5			
10.6	258.3	871.9	42.0	334.0	671.7	18.5	263.9	849.5			
11.0	259.5	869.3	43.0	337.4	665.8	19.5	265.9	843.4			
11.7	260.9	864.7	44.0	340.6	659.9	19.7	266.2	842.1			
12.6	263.0	858.8	45.0	344.4	654.1	20.0	266.6	840.3			
13.5	264.6	852.9	47.0	352.0	642.5	22.2	270.6	826.9			
13.8	265.3	850.9	49.0	360.0	631.2	22.6	271.3	824.4			
14.1	265.9	848.9	50.5	366.0	622.8	23.2	274.1	820.8			

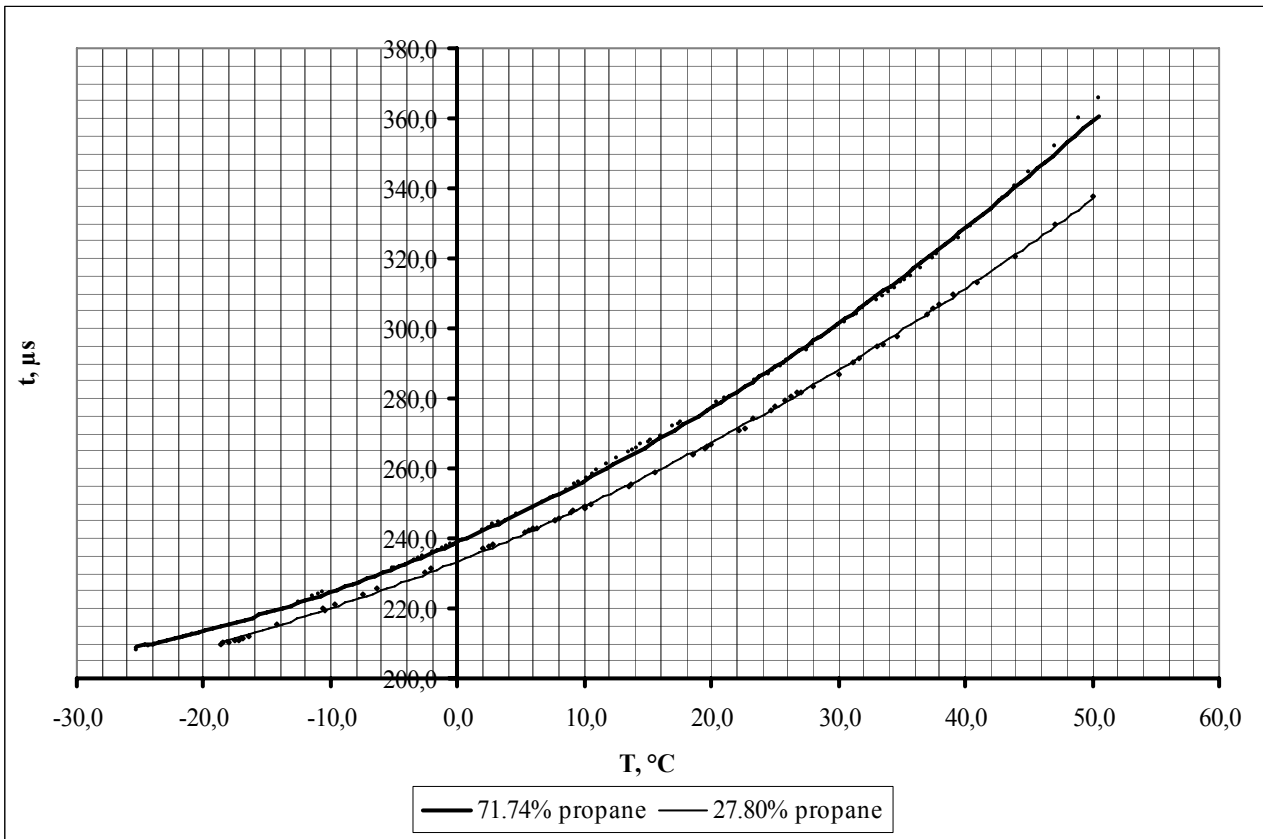


Fig.2. Sound propagation time in LPG

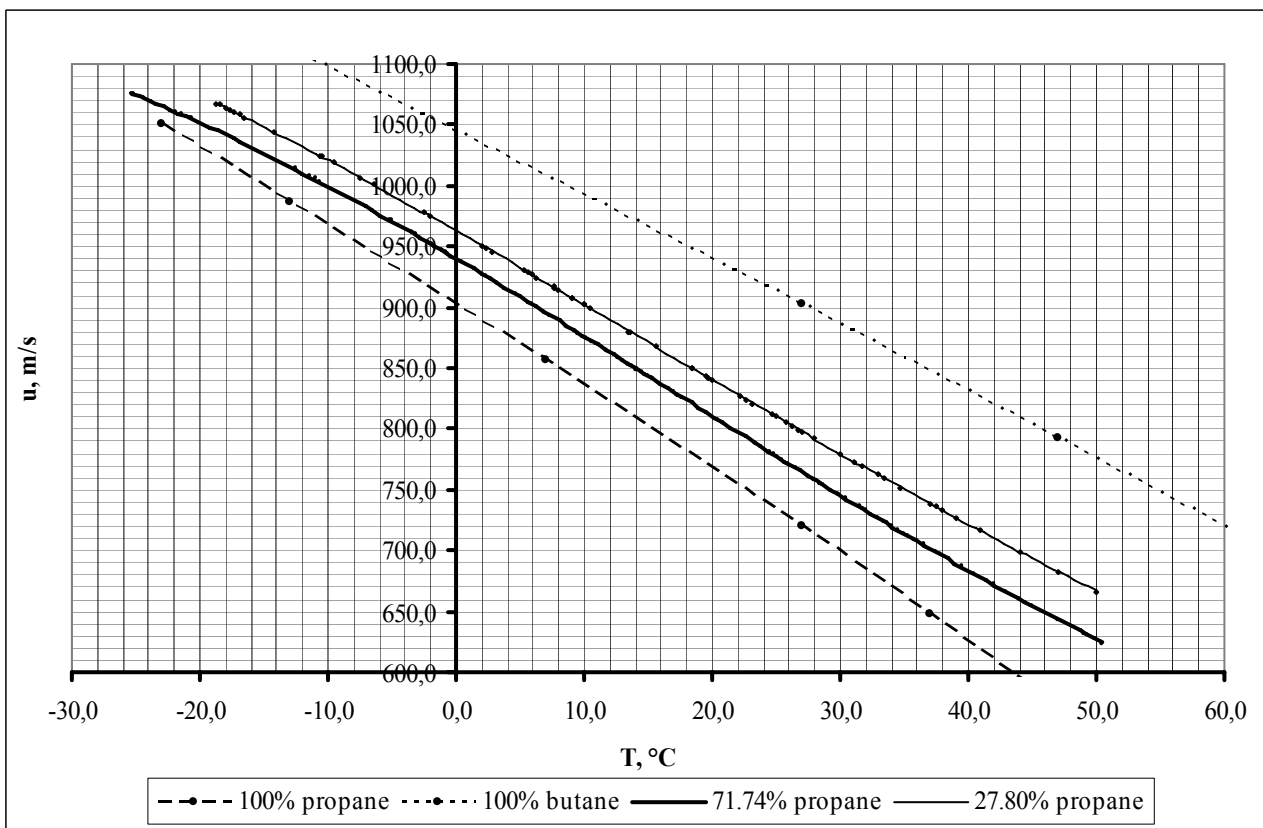


Fig.3. Sound velocity in liquefied gases

In Fig. 3 the sound velocities in pure propane [1] and pure butane [1] (at the pressure of 1.6 MPa) are added for comparison.

Fig. 3 shows that the sound velocity of pure liquefied propane is lowest at a selected temperature. As the content of butane in LPG increases, the sound velocity decreases. For example, at the temperature of 20.0 °C, the difference of sound velocity between pure liquefied propane and liquefied butane is about 172 m/s. At the same temperature, the difference of sound velocity between our two LPG samples is about 30 m/s.

The sound velocity in LPG is decreasing non-linearly with increasing temperature.

Conclusions

This experiment proved, that there are significant differences in sound velocities in LPG samples with different propane-butane ratios. These differences in sound velocity can be used to develop a method to determine the composition of LPG, in which propane-butane ratio is unknown.

For example, non-invasive ultrasonic measurement devices for industrial applications can be designed to measure the composition of LPGs. This measurement process requires additional measurement of temperatures. In non-standard processes the pressure of LPG must also be evaluated.

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References

1. **Younglove B., Ely J.** Thermophysical properties of fluids. II. Methane, Ethane, Propane, Isobutane and Normal Butane. *J. Phys. Chem. Ref. Data.* 1987. Vol. 16. No. 4. P. 577-797.
2. **Daridon J., Lagourette B., Grolier J.-P.** Experimental measurements of the speed of sound in n-hexane from 293 to 373 K and up to 150 MPa. *International Journal of Thermophysics.* 1998. Vol. 19. No.1. P. 145-159.
3. **Niepmann R.** Thermodynamic properties of propane and n-butane. II: Speeds of sound in the liquid up to 60 MPa. *Journal of chemical thermodynamics.* ISSN 0021-9614. CODEN JCTDAF. 1984. Vol. 16. No. 9. P. 851-860.
4. **Heydemann P., Houck J.** Self-consistent ultrasonic method for determination of the equation of state of liquids at very high pressures. *J. Appl. Phys.*, 1969. No.40. P. 609-613.
5. **Marczak W., Dzida M.** Determination of the thermodynamic properties of 1-propanol and 1-hexanol from speed of sound measurements under elevated pressures. *High Temp.s High Pressures.* 2000. No. 32. P. 283-292.
6. **Dzida M., Ernst S.** Speed of sound in propan-1-ol + heptane mixtures under elevated pressures. *J. Chem. Eng.* 2003. No. 48. P. 1453-1457.
7. **Sun T. F., Bominaar S. A., Ten Seldam C.A., Biswas S.N.** Evaluation of the thermophysical properties of toluene and n-heptane from 180 to 320 K and up to 260 MPa from speed-of-sound data. *Ber. Bunsen-Ges. Phys. Chem.* 1991. No.95. P. 696-704.
8. **Haynes W. M., Hiza M. J.** Measurements of the orthobaric liquid densities of methane, ethane, propane, isobutane, and normal butane. *The Journal of Chemical Thermodynamics.* 1977. Vol. 9. No. 2. P. 179-187.
9. **Lago S., Giuliano Albo P., Madonna Ripa D.** Speed-of-sound measurements in n-nonane at temperatures between 293.15 and 393.15 K at pressures up to 100 MPa. *International Journal of Thermophysics.* 2006. Vol.27, No.4. P 1083-1094.
10. **Avsec J., Watanabe K.** An approach to calculate thermodynamic properties of mixtures including propane, n-butane, and isobutane. *International Journal of Thermophysics.* 2005. Vol. 25. No. 6. P. 1769-1780.
11. **Miyamoto H., Watanabe K.** Thermodynamic property model for fluid-phase n-butane. 2001. *International Journal of Thermophysics.* Vol. 22. No. 2. P. 459-475.
12. **Miyamoto H., Watanabe K.** Helmholtz – type equation of state for hydrocarbon refrigerant mixtures of propane / n-butane, propane / isobutane, n-butane / isobutane and propane / n-butane / isobutane. *International Journal of Thermophysics.* 2003. Vol. 24. No. 4. P. 1007-1031.
13. **Lemmon E., Jacobsen R.** A generalized model for the thermodynamic properties of mixtures. *International Journal of Thermophysics.* 1999. Vol. 20. No.3. P. 825-835.
14. **Vasserman A. et al.** A Thermophysical property databank for technically important gases and liquids. *International Journal of Thermophysics.* 2001. Vol. 22. No. 2. P. 477-485.
15. **Marczak W.** Water as a standard in the measurements of speed of sound in liquid. *J. Acoust. Soc. Am.* 1997. Vol. 102. P. 2776-2779.
16. **Del Grosso V., Mader C.** Speed of sound in pure water. *J. Acoust. Soc. Am.* 1972. Vol. 52. P. 1442-1446.
17. **Wilson W. D.** Speed of sound in distilled water as a function of temperature and pressure. *J. Acoust. Soc. Am.* 1959. Vol. 31. P. 1067-1072.
18. **Greenspan M., Tschieg C. E.** Tables of the speed of sound in water. *J. Acoust. Soc. Am.* 1959. Vol. 31. P. 75-76.
19. **Petrauskas A.** Non-invasive ultrasonic level measurement technology. ISSN:1392-2114 Ultragarsas (Ultrasound). Kaunas: Technologija. 2006. Vol. 61, No.4. P.29-33.

A. Petrauskas

Eksperimentiniai garso greičio suskystintose naftos dujose (LPG) matavimo rezultatai

Reziumė

Išmatuotos garso greičio vertės suskystintuose propano-butano dujų mišiniuose praktikoje naudojamame nuo –23 °C iki +55 °C temperatūrų intervale. Garso greičio matavimai atlikti neinvaziniu faziniu akustiniu matavimo metodu standartiniame 5 l. talpos balione 700 kHz dažnio srityje. Garso greitis minėtame temperatūrų intervale kinta 1,71 karto. Esant aplinkos temperatūrai 20 °C, garso greitis suskystintose dujose, turinčiose 71,74 % propano, yra 810,16 m/s, o suskystintose dujose, turinčiose 27,80 % propano, - 840,31 m/s. Garso greičio kitimas minėtame temperatūrų intervale yra artimas tiesinei priklausomybei. Pažymėtina, kad šio kitimo temperatūrinė priklausomybė suskystintose naftos dujose yra kur kas didesnė negu jų tankio kitimo priklausomybė. Praktiniai matavimo rezultatai gali būti naudingi suskystintų dujų sudėjų įvairiose talpose nustatant neinvaziniu akustiniu matavimo metodu, taip pat automatizuojant technologinius procesus suskystintų dujų gamybos procese.

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