Speeds of Sound in Methanol at Temperatures from 233.33 to 353.21 K at Pressures up to 20 MPa

Christian W. Scholz¹ · Roland Span¹

Received: 14 January 2021 / Accepted: 4 February 2021 / Published online: 8 March 2021
© The Author(s) 2021

Abstract

We report experimental speeds of sound in methanol. Measurements were conducted at temperatures from 233 to 353 K with pressures up to 20 MPa using the double-path length pulse-echo technique. The relative expanded combined uncertainty ($k=2$) in measurement was estimated to vary from 0.012 to 0.014%, considering contributions from temperature, pressure, path length calibration, pulse timing, and purity of the sample. Experimental speeds of sound gained in the scope of this work were compared with the equation of state by de Reuck and Craven, as well as with further data from literature.

Keywords Methanol · Pulse-echo technique · Speed of sound

1 Introduction

The power-to-methanol technology is considered more and more important with increasing focus on the challenge of storing energy from volatile renewable sources. In such processes, hydrogen descending from electrolysis operated with excess renewable energy is converted to methanol with CO₂ captured from, e.g., industrial combustion gases [1–3]. At times of insufficient supply of power from renewable sources, stored liquid methanol can be used as fuel to cover the excess demand of electricity. Design and engineering of such processes employ thermophysical property data gained from fundamental equations of state. The currently used fundamental equation of state of methanol was developed by de Reuck and Craven [4]. Since some shortcomings of this equation of state are well known, Thol and Span [5] currently work on a new equation of state. Since such approaches are empirical models, their accuracy is directly related to the quality of the employed experimental data.

✉ Roland Span
r.span@thermo.rub.de

¹ Faculty of Mechanical Engineering, Thermodynamics, Ruhr University Bochum, Universitätsstraße 150, 44801 Bochum, Germany

Even though several experimental (p , c , T) data sets of methanol are available in the literature, only very few authors provide comprehensive data sets over larger ranges of temperature and especially pressure (see Table 1). In addition, not all of the available data are consistent, which necessitated further speed of sound measurements to clarify the existing data situation. Temperatures of the isotherms studied in this work have been selected partly to allow for direct comparison with other data sets. In a (p , T) diagram, Fig. 1 shows the state points of speed of sound measurements carried out in the scope of this work as well as of further data available from literature.

2 Experimental Section

2.1 Apparatus Description

Speed of sound measurements in methanol were conducted employing the double-path length pulse-echo technique with an apparatus according to the design of Meier and Kabelac [58]. The experimental set-up has been used for speed of sound measurements of several liquids in our previous works [59–62]; thus, it is only explained in brief at this point. A detailed description with illustrations of the measuring cell and a schematic drawing of the entire measuring system can be taken from the work of Gedanitz et al. [63]. The actual acoustic sensor consists of an x-cut quartz crystal as transducer, located axially off-center between two polished stainless steel reflectors, resulting in two different path lengths ($L_1=20$ mm and $L_2=30$ mm). If the piezoelectric disc transducer is excited by a tone burst (here a 30-cycle sinusoidal burst at 8 MHz) of a waveform generator (Agilent, model 3322A), ultrasonic pulses propagate into the surrounding fluid and are reflected at both ends of the measuring cell. The returning wave fronts excite the transducer and the resulting electronic signals are captured by a digital oscilloscope (Agilent, model MS6032A). These signals are referred to as echoes. Due to the unequal path lengths, the second echo is detected delayed by the time difference Δt_{echo} . The speed of sound can be determined according to Eq. (1), incorporating a correction τ according to Harris [64] to account for diffraction:

$$c = \frac{2(L_2 - L_1)}{\Delta t_{\text{echo}} + \tau}. \quad (1)$$

The waveform of the two resulting echoes is averaged from 16 consecutive pulses and a bandpass filter based on a Fast Fourier Transform with a bandwidth of 1.6 MHz is applied, to enhance the signal-to-noise ratio. Based on these processed data, the time difference Δt_{echo} is computed according to the algorithm described by Dubberke et al. [65].

The acoustic sensor was housed in a stainless steel pressure vessel and allows for speed of sound measurements up to 20 MPa. Two vibrating-quartz crystal pressure transducers (Paroscientific, models 1000-500A and 1000-6 K) and a differential pressure indicator (Rosemount, model 3051) are utilized to measure the pressure at

Table 1 Review of speed of sound measurements in methanol

Author	Year	Points	T (K)	p (MPa)	Phase ^a
Abraham et al. [8]	2000	3	303–313	0.101325	Liquid
Abraham et al. [9]	1997	3	303–313	0.101325	Liquid
Allam and Lee [10]	1996	2	298	0.101325	Liquid
Aminabhavi et al. [11]	1993	3	298–308	0.101325	Liquid
Aminabhavi and Banerjee [12]	1998	1	298	0.101325	Liquid
Aminabhavi and Patil [13]	1999	1	298	0.101325	Liquid
Arce et al. [14]	2004	1	283–334	0.101325	Liquid
Arce et al. [15]	2006	1	298–314	0.101325	Liquid
Artemchenko [16]	1963	6	283–333	0.101325	Liquid
Bahadur and Deenadayalu [17]	2011	4	298–303	0.101325	Liquid
Bakshi et al. [18]	1996	1	280–361	0.101325	Liquid
Bohidar [19]	1988	9	293	0.1–83	Liquid
Carnevale and Litovitz [20]	1955	5	303	0.098–196	Liquid
Casas et al. [21]	2001	1	298	0.101325	Liquid
Chauhan et al. [22]	1995	3	298–318	0.101325	liquid
Danusso and Fadigati [23]	1953	4	293–323	0.101325	liquid
Dávila et al. [7]	2016	42	253–353	0.1–30	liquid
Dávila and Trusler [24]	2009	56	298–343	0.091–60	liquid
Eden and Richardson [25]	1960	3	293	0.1–98	Liquid
Emery and Gasse [26]	1979	1	273	0.101325	Liquid
Fort and Moore [27]	1965	1	298	0.101325	Liquid
Freyer et al. [28]	1929	6	273–323	0.101325	Liquid
Gadaibaev and Shakhparonov [29]	1973	4	181–213	0.101325	Liquid
George et al. [30]	2002	2	308–319	0.101325	liquid
Gill et al. [31]	1993	1	298	0.101325	Liquid
Golik et al. [32]	1958	4	293–323	0.101325	Liquid
Gonzalez et al. [33]	2004	3	293–303	0.101325	Liquid
Govindarajan et al. [34]	2003	5	293–313	0.101325	Liquid
Iglesias et al. [35]	1998	1	298	0.101325	Liquid
Iglesias et al. [36]	1998	1	298	0.101325	Liquid
Iloukhani and Rostami [37]	2003	1	303	0.101325	Liquid
Jatkar [38]	1939	2	370–408	0.101325	Liquid
Javed [6]	2019	63	220–500	0.1–124	liquid
Kumar et al. [39]	1981	1	298	0.101325	Liquid
Marino et al. [40]	2000	1	298	0.101325	Liquid
Moses Ezhil Raj et al. [41]	2009	5	303–323	0.101325	liquid
Ogawa and Murakami [42]	1987	1	298	0.101325	Liquid
Ohmori et al. [43]	2001	32	513–540	4–50	SCR
Orge et al. [44]	1997	1	298	0.101325	Liquid
Parthasarathy et al. [45]	1953	2	300	0.101325	Liquid
Pereira et al. [46]	2002	5	293–333	0.101325	Liquid
Pereira et al. [47]	2003	5	293–333	0.101325	Liquid

Table 1 (continued)

Author	Year	Points	T (K)	p (MPa)	Phase ^a
Povey [48]	2003	1	293	0.101325	Liquid
Rabinovich et al. [49]	1957	6	283–333	0.101325	Liquid
Rama Rao [50]	1940	6	273–323	0.101325	Liquid
Rao et al. [51]	2007	4	303–318	0.101325	Liquid
Resa et al. [52]	2002	1	298	0.101325	Liquid
Rodriguez et al. [53]	2001	3	293–313	0.101325	Liquid
Scholz and Span (<i>this work</i>)	2021	65	233–353	0.15–20	Liquid
Sun et al. [54]	1988	105	274–332	0.1–275	Liquid
Wilson and Bradley [55]	1964	48	273–323	0.1–96	Liquid
Yu et al. [56]	2011	2	298–303	0.1009	Liquid
Zafarani-Moattar and Shekaari [57]	2005	5	298–318	0.101325	Liquid

^aSCR denotes supercritical region

Fig. 1 Selection of experimental (p , c , T) data of methanol from literature and (p , T) state points investigated in the present work shown in a (p , T) phase diagram. $\circ$, this work; ∇ , Javed et al. [6]; $\diamond$, Davila et al. [7]; $\times$, further data from literature; —, phase boundary and $\star$, critical point as calculated with the equation of state of de Reuck and Craven [4] (Color figure online)

a standard uncertainty of $u(p) = 0.0024$ MPa. The entire pressure vessel is immersed into a calibration bath thermostat (Fluke, model 7060), realizing speed of sound measurements at temperatures from 233 to 353 K. The temperature is measured with a long-stem $25\ \Omega$ standard platinum resistance thermometer (SPRT, Rosemount Aerospace, model 162CE), calibrated on ITS-90 and read with a direct current thermometry bridge (Isotech, model TTI-2), stating a standard uncertainty in temperature measurement of $u(T) = 0.004$ K. With measurements over the stated temperature and pressure range, thermal expansion and pressure deformation can affect the actual path length, which is considered according to Eq. (2):

$$\Delta L(T, p) = \Delta L_0 \left[1 + \alpha(T - T_0) - \frac{\beta}{3}(p - p_0) \right], \quad (2)$$

where ΔL_0 is the path length difference at the reference temperature $T_0 = 293.15$ K and the reference pressure $p_0 = 0.1$ MPa. The mean value of the isobaric expansivity α at reference pressure p_0 was received from values for the differential thermal expansion of stainless steel 1.4571 according to Eq. (3):

$$10^6 \alpha / \text{K}^{-1} = \sum_0^4 \frac{a_i}{(i+1)} \cdot \left(\frac{T - T_0}{T_0} \right)^i. \quad (3)$$

While the coefficients a_i for $i \geq 1$ are values given by Meier [66] according to Table 2, a_0 and the path length difference ΔL_0 given in Eq. (2) are determined from calibration measurements with high-purity water. Here, the calibration remains the same as presented in our previous work [60], where measurements at $T = 274.15$, 278.15, 283.15, 293.15, 303.15, 313.15, 323.15, 333.15, 343.15, and 353.15 K and ambient pressure were conducted. The calibration was evaluated by comparison with the IAPWS-95 equation of state by Wagner and Pruß [67] with the experimental speeds of sound being reported within the uncertainty of the equation of state of 0.005%.

In Eq. (2), β describes the mean compressibility of the cell at temperature T and is determined from material properties [68] according to Eq. (4):

$$\beta = (1 - 2\nu)/E = (1 - 2\nu) \left[\sum_0^1 b_i (T/\text{K})^i \right]^{-1}, \quad (4)$$

where E is Young's modulus and $\nu = 0.3$ is Poisson's ratio with the employed coefficients b_i given in Table 2.

2.2 Experimental Material

The samples used in the present work are specified in Table 3. High-purity water with a purity of 99.9997 mol% was employed for the calibration. The purity of the used methanol was found to be 99.9300 mol%, investigated by gas chromatographical analysis, as stated by the supplier. Additionally, a mass fraction of water of less than 0.001%, known from coulometric Karl Fischer titration, was stated. In order to avoid contact with the humidity of ambient air, the methanol sample was transferred

Table 2 Coefficients of Eq. (3) for the mean thermal expansivity α and coefficients of Eq. (4) for the mean isothermal compressibility β of the stainless steel 1.4571 cell

i	0	1	2	3	4
a_i	8.36800	5.08059	−3.74480	1.86720	−0.34148
b_i (GPa)	219.720	−0.080	−	−	−

Table 3 Description of the used chemical samples water and methanol

Chemical name	CAS number	Source	Purity/ (mole frac- tion)	Purification method	Analytical method ^a
Water	7732-18	Sigma-Aldrich	0.999997	Freeze-thaw	
Methanol	67-56-1	VWR Chemicals	0.999300	Freeze-thaw	GC, KF ^b

^aGC is gas chromatography and KF is Karl Fischer titration

^bH₂O < 0.0010 mass %

to a stainless steel bottle within the dry inert argon atmosphere of a glove box that was manufactured in-house. Prior to use, the sample was degassed by repeated freeze-pump-thaw cycles, as described in our previous works [59–62].

2.3 Experimental Procedure

The experimental procedure applied to conduct the speed of sound measurements in the scope of this work was adapted from previous works [59, 60]. The stainless steel bottle containing the sample of methanol was attached to the evacuated speed of sound apparatus and carefully heated. Simultaneously, the temperature of the bath thermostat was set to the lowest possible temperature of 233 K, thus taking advantage of the pressure and temperature gradient during the filling process. A connected hand pump was utilized to increase the pressure up to 20 MPa and the system was left to equilibrate prior to the first measurement. Measurements were subsequently carried out along isotherms with decreasing pressure. From isotherm to isotherm, the temperature of the measuring cell was increased overnight.

2.4 Uncertainty Analysis

The combined expanded uncertainty in speed of sound was estimated according to our previous studies [59, 60], where the approach is explained in detail, by developing the following term with consideration of Eqs. (1) and (2):

$$c(T, p) = c_0 \frac{(\Delta t_0 + \tau_0)}{(\Delta t + \tau)} \left[1 + \alpha(T - T_0) - \frac{\beta}{3}(p - p_0) \right]. \quad (5)$$

The square of the standard uncertainty of the speed of sound is then described by Eq. (6):

$$\begin{aligned} u^2(c) &= \left[\left(\frac{\partial c}{\partial c_0} \right) u(c_0) \right]^2 + \left[\left(\frac{\partial c}{\partial \Delta t_0} \right) u(\Delta t_0) \right]^2 + \left[\left(\frac{\partial c}{\partial \Delta t} \right) u(\Delta t) \right]^2 + \left[\left(\frac{\partial c}{\partial \alpha} \right) u(\alpha) \right]^2 + \left[\left(\frac{\partial c}{\partial \beta} \right) u(\beta) \right]^2 \\ &= \left[\left(\frac{c}{c_0} \right) u(c_0) \right]^2 + \left[\left(\frac{c}{\Delta t_0} \right) u(\Delta t_0) \right]^2 + \left[\left(\frac{c}{\Delta t} \right) u(\Delta t) \right]^2 + \left[\left(\frac{c_0 \Delta t_0 (T - T_0)}{\Delta t} \right) u(\alpha) \right]^2 + \left[\left(\frac{-c_0 \Delta t_0 (p - p_0)}{3 \Delta t} \right) u(\beta) \right]^2. \end{aligned} \quad (6)$$

Further contributions resulting from the measurement of temperature and pressure as well as from the presence of impurities are considered within the determination of the expanded combined uncertainty $U(c)$ according to Eq. (7) with the expansion factor $k=2$:

$$U(c) = k \sqrt{u^2(c) + \left[(\partial c / \partial p)_T u(p) \right]^2 + \left[(\partial c / \partial T)_p u(T) \right]^2 + u^2(c(w))}. \quad (7)$$

The respective derivatives were determined by sensitivity analysis, employing the equation of state of de Reuck and Craven [4]. The impact of potential impurities was investigated using mixture models, implemented in TREND 4.0 [69]. Water and ethanol were investigated as the impurities with the largest influence. The result for the combined expanded uncertainty $U(c)$ for the speed of sound measurement in methanol at $T=293.20$ K and $p=5$ MPa is exemplified in Table 4 and was estimated to be $0.134 \text{ m}\cdot\text{s}^{-1}$. Among all conducted measurements, $U(c)$ varies between 0.126 and $0.176 \text{ m}\cdot\text{s}^{-1}$, while the relative expanded combined uncertainty remains almost constant with values from 0.012 to 0.014% .

3 Results and Discussion

The speed of sound was measured at seven or eight state points on each of nine isotherms, leading to a (p, c, T) data set comprising 65 measuring points (see Table 5). Absolute experimental speeds of sound are plotted versus pressure in Fig. 2, clarifying internal consistency throughout the entire (p, T) region and showing a decreasing speed of sound with increasing temperature. In the present work, speeds of

Table 4 Uncertainty budget for speed of sound measurements conducted in methanol, exemplary at one state point where T is temperature, p is pressure, c is sound speed, Δt is time difference, α is the mean isobaric expansivity, β is the mean isothermal compressibility, w is mass fraction, and subscript 0 refers to calibration measurements in pure water. Sensitivity coefficients associated with temperature and pressure were estimated from the equation of state of de Reuck and Craven [4]

Quantity	Value	Standard uncertainty	Sensitivity coefficient	Uncertainty contribution
c_0	$1482.52 \text{ m}\cdot\text{s}^{-1}$	$0.07 \text{ m}\cdot\text{s}^{-1}$	0.774	$0.057 \text{ m}\cdot\text{s}^{-1}$
Δt_0	$13.32385 \mu\text{s}$	$0.00017 \mu\text{s}$	$86 \text{ m}\cdot\text{s}^{-1}\cdot\mu\text{s}^{-1}$	$0.015 \text{ m}\cdot\text{s}^{-1}$
Δt	$17.11560 \mu\text{s}$	$0.00022 \mu\text{s}$	$67 \text{ m}\cdot\text{s}^{-1}\cdot\mu\text{s}^{-1}$	$0.015 \text{ m}\cdot\text{s}^{-1}$
α	$8.37 \times 10^{-6} \text{ K}^{-1}$	$4.18 \times 10^{-7} \text{ K}^{-1}$	$4.59 \times 10^{-1} \text{ m}\cdot\text{s}^{-1}\cdot\text{K}$	$0.002 \times 10^{-4} \text{ m}\cdot\text{s}^{-1}$
β	$2.04 \times 10^{-6} \text{ MPa}^{-1}$	$1.02 \times 10^{-7} \text{ MPa}^{-1}$	$1.88 \times 10^3 \text{ m}\cdot\text{s}^{-1}\cdot\text{MPa}$	$0.019 \times 10^{-2} \text{ m}\cdot\text{s}^{-1}$
T	293.202 K	0.004 K	$3.14 \text{ m}\cdot\text{s}^{-1}\cdot\text{K}^{-1}$	$0.013 \text{ m}\cdot\text{s}^{-1}$
p	5.0166 MPa	0.0024 MPa	$6.02 \text{ m}\cdot\text{s}^{-1}\cdot\text{MPa}^{-1}$	$0.014 \text{ m}\cdot\text{s}^{-1}$
$c(w)$	$1147.612 \text{ m}\cdot\text{s}^{-1}$	$0.036 \text{ m}\cdot\text{s}^{-1}$	1	$0.021 \text{ m}\cdot\text{s}^{-1}$
Combined expanded uncertainty ($k=2$):				$0.134 \text{ m}\cdot\text{s}^{-1}$

Table 5 Speeds of sound c_{exp} of methanol and expanded uncertainties $U(c)$ ($k=2$) at temperatures T and pressures p^a

T / K	p / MPa	$c_{\text{exp}} / \text{m}\cdot\text{s}^{-1}$	$U(c)/\text{m}\cdot\text{s}^{-1}$	T / K	p / MPa	$c_{\text{exp}} / \text{m}\cdot\text{s}^{-1}$	$U(c)/\text{m}\cdot\text{s}^{-1}$
233.249	0.155	1335.959	0.166	300.199	10.015	1154.058	0.135
233.249	0.509	1337.570	0.166	300.199	15.023	1181.108	0.137
233.249	2.011	1344.397	0.167	300.198	20.012	1206.709	0.139
233.249	5.019	1357.677	0.169	313.202	0.155	1053.364	0.128
233.249	10.027	1379.331	0.171	313.202	0.509	1055.703	0.128
233.249	15.025	1400.133	0.173	313.202	2.009	1065.503	0.129
233.249	19.991	1420.070	0.176	313.202	5.012	1084.413	0.130
253.218	0.155	1260.037	0.151	313.202	10.021	1114.569	0.132
253.218	0.507	1261.797	0.151	313.202	15.020	1142.949	0.135
253.218	2.011	1269.270	0.152	313.202	20.053	1169.991	0.137
253.218	5.026	1283.832	0.153	333.204	0.156	989.182	0.126
253.219	10.024	1307.299	0.155	333.204	0.508	991.750	0.126
253.218	15.027	1329.873	0.157	333.204	2.009	1002.559	0.127
253.218	20.016	1351.505	0.160	333.204	5.017	1023.307	0.128
273.205	0.155	1187.716	0.140	333.204	10.010	1055.986	0.131
273.205	0.506	1189.615	0.140	333.204	15.028	1086.748	0.133
273.205	2.017	1197.773	0.141	333.204	20.030	1115.589	0.136
273.205	5.012	1213.683	0.142	350.206	0.254	935.924	0.126
273.205	10.013	1239.253	0.144	350.205	0.504	937.922	0.126
273.205	15.019	1263.648	0.147	350.205	1.005	941.877	0.126
273.205	20.064	1287.210	0.149	350.205	2.007	949.700	0.127
293.203	0.155	1119.236	0.132	350.206	5.013	972.170	0.128
293.203	0.509	1121.340	0.132	350.205	10.021	1007.528	0.131
293.202	2.009	1130.279	0.133	350.205	15.020	1040.294	0.134
293.202	5.017	1147.612	0.134	350.206	19.988	1070.761	0.137
293.202	10.013	1175.297	0.137	353.203	0.254	926.000	0.126
293.202	15.013	1201.627	0.139	353.203	0.509	928.054	0.126
293.203	20.049	1226.850	0.141	353.203	2.007	940.019	0.127
300.199	0.155	1096.280	0.130	353.202	5.011	962.872	0.128
300.199	0.505	1098.470	0.131	353.203	10.017	998.583	0.131
300.199	1.007	1101.570	0.131	353.203	15.026	1031.849	0.134
300.199	2.248	1109.144	0.131	353.203	20.010	1062.759	0.137
300.199	5.018	1125.549	0.133				

^aStandard uncertainties are $u(T)=0.004 \text{ K}$, $u(p)=0.0024 \text{ MPa}$

sound in methanol vary from $926 \text{ m}\cdot\text{s}^{-1}$ ($T=353 \text{ K}$ and $p=0.25 \text{ MPa}$) to $1420 \text{ m}\cdot\text{s}^{-1}$ ($T=233 \text{ K}$ and $p=20 \text{ MPa}$).

Relative deviations of the experimental speeds of sound in methanol from values calculated with the equation of state of de Reuck and Craven [4] as reference (zero line) are plotted versus pressure in Fig. 3. Relative deviations of the entire data set fall well within the uncertainty of the equation of state, stated by the authors to be

Fig. 2 Results of the speed of sound measurements in methanol. Absolute speeds of sound c_{exp} are plotted versus pressure. $\circ$, 233 K; $\square$, 253 K; ∇ , 273 K; $\star$, 293 K; $\triangle$, 300 K; $\times$, 313 K; $+$, 333 K; T , 350 K; $\diamond$, 353 K. Lines are shown only to guide the eye (Color figure online)

Fig. 3 Results of the speed of sound measurements in methanol. Relative deviations of the experimental speeds of sound c_{exp} from values c_{EOS} calculated with the equation of state of de Reuck and Craven [4] are plotted versus pressure. $\circ$, 233 K; $\square$, 253 K; ∇ , 273 K; $\star$, 293 K; $\triangle$, 300 K; $\times$, 313 K; $+$, 333 K; T , 350 K; $\diamond$, 353 (Color figure online)

2%. However, a trend can be observed along each isotherm with the relative deviations from the lowest to the highest pressure decreasing between 0.27 and 0.42%. Altogether, relative deviations vary from -0.66% at $T=353$ K and $p=15.03$ MPa to 0.35% at $T=253$ K and $p=0.16$ MPa.

Again, Fig. 4 shows the comparison of the speed of sound measured in the scope of this work with the equation of de Reuck and Craven [4] as reference (zero line). However, the comparison focuses on the individual temperatures and can thus consider further data sets from literature. The data set by Davila et al. [7] is in very good agreement with our reported data and falls within the experimental uncertainty

Fig. 4 Results of the speed of sound measurements of methanol. Relative deviations of the experimental speeds of sound c_{exp} from values c_{EOS} calculated with the equation of state by de Reuck and Crave [4] (zero line) are plotted versus pressure. $\times$, this work; $\diamond$, Davila et al. [7]; ∇ , Javed et al. [6]; $\square$, Sun et al. [54]; $\circ$, Wilson and Bradley [55] (Color figure online)

at temperatures between 273 and 333 K. At 253 and 353 K both data sets differ by up to 0.04%, slightly exceeding the claimed combined experimental uncertainty. Speeds of sound, gained in the scope of this work, are also in good agreement with the data set by Javed et al. [6] at 350 K, especially at higher pressures, where deviations are within the experimental uncertainty. Comparison with data from Javed et al. [6] at lower temperatures shows a systematical downward offset at 300 K and a systematical upward offset at 244 and 253 K, respectively (see Fig. 4). Further data by Sun et al. [54] and Wilson and Bradley [55] show a upward offset at overlapping temperatures from 273 to 313 or 333 K and at comparable pressures (see Fig. 4).

4 Conclusion

Speeds of sound in high-purity methanol were measured at temperatures from 233 to 353 K and at pressures up to 20 MPa with the overall relative expanded combined uncertainty ($k=2$) ranging from 0.012 to 0.014%. The new data deviate from the equation of state of de Reuck and Craven [4] by up to 0.66% but agree with the data set of Davila et al. [7] within 0.014% at temperatures between 273 and 333 K

and within 0.040% beyond this temperature range. The measurements conducted by Javed et al. [6] are confirmed partly, especially at higher temperatures. On other isotherms significant deviations to the data of Javed et al. [6] are observed.

Acknowledgements The authors thank Dr. Monika Thol of RUB for helpful discussions regarding the present measurements.

Funding Open Access funding enabled and organized by Projekt DEAL.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

References

1. M. Nielsen, E. Alberico, W. Baumann, H.-J. Drexler, H. Junge, S. Gladiali, M. Beller, *Nature* (2013). <https://doi.org/10.1038/nature11891>
2. M.J. Bos, S.R.A. Kersten, D.W.F. Brilman, *Appl. Energy* (2020). <https://doi.org/10.1016/j.apenergy.2020.114672>
3. I. González-Aparicio, Z. Kapetaki, E. Tzimas, *Appl. Energy* (2018). <https://doi.org/10.1016/j.apenergy.2018.03.114>
4. K.M.d. Reuck, R.J.B. Craven, S. Angus, B. Armstrong, R.J. Craven, W. Wagner, *Methanol - International Thermodynamic Tables of Fluid State Vol. 12* (Blackwell Scientific Publications, London, 1993)
5. M. Thol, R. Span, *Int. J. Thermophys.* (to be submitted 2021)
6. M.A. Javed, E. Baumhögger, J. Vrabec, *J. Chem. Eng. Data* (2019). <https://doi.org/10.1021/acs.jced.8b00938>
7. M.J. Dávila, H. Gedanitz, R. Span, *J. Chem. Thermodyn.* (2016). <https://doi.org/10.1016/j.jct.2015.10.006>
8. R. Abraham, M. Abdulkadhar, C.V. Asokan, *J. Chem. Thermodyn.* (2000). <https://doi.org/10.1006/jcht.1999.0608>
9. R. Abraham, M. Abdulkadhar, C.V. Asokan, *Acoust. Lett.* **20**, 236–245 (1997)
10. D.S. Allam, W.H. Lee, *J. Chem. Soc. A* (1966). <https://doi.org/10.1039/J19660000005>
11. T.M. Aminabhavi, M.I. Aralaguppi, S.B. Harogopad, R.H. Balundgi, *J. Chem. Eng. Data* (1993). <https://doi.org/10.1021/jc00009a008>
12. T.M. Aminabhavi, K. Banerjee, *J. Chem. Eng. Data* **43**, 1096 (1998)
13. T.M. Aminabhavi, V.B. Patil, K. Banerjee, *J. Chem. Eng. Data* (1999). <https://doi.org/10.1021/jc990120q>
14. A. Arce, O. Rodríguez, A. Soto, *Fluid Phase Equilib.* (2004) doi:<https://doi.org/10.1016/j.fluid.2004.05.008>
15. A. Arce, E. Rodil, A. Soto, *J. Solution Chem.* (2006). <https://doi.org/10.1007/s10953-006-8939-y>
16. A.I. Artemchenko, *Zh. Fiz. Khim* **37**, 3 (1963)
17. I. Bahadur, N. Deenadayalu, *J. Solution Chem.* (2011). <https://doi.org/10.1007/s10953-011-9740-0>
18. M.S. Bakshi, J. Singh, H. Kaur, S.T. Ahmad, G. Kaur, *J. Chem. Eng. Data* (1996). <https://doi.org/10.1021/jc960180l>
19. H.B. Bohidar, *J. Appl. Phys.* (1988). <https://doi.org/10.1063/1.341780>
20. E.H. Carnevale, T.A. Litovitz, *J. Acoust. Soc. Am.* (1955). <https://doi.org/10.1121/1.1907959>
21. L.M. Casás, G. Marino, M. Iglesias, B. Orge, J. Tojo, L. Mosteiro, *J. Chem. Thermodyn.* (2001). <https://doi.org/10.1006/jcht.2001.0878>
22. M.S. Chauhan, K.C. Sharma, S. Gupta, *Acoust. Lett.* **18**, 233–240 (1995)
23. F. Danusso, E. Fagigati, *Atti Accad. Naz. Lincei* **14**, 81 (1953)

24. M.J. Dávila, J.P.M. Trusler, *J. Chem. Thermodyn.* (2009). <https://doi.org/10.1016/j.jct.2008.08.003>
25. H.F. Eden, Richardson E. G., *Acta Acust. united Acust.* **10**, 309 (1960)
26. J. Emery, S. Gasse, *Acta Acust. united Acust.* **43**, 205 (1979)
27. R.J. Fort, W.R. Moore, *Trans. Faraday Soc.* (1965). <https://doi.org/10.1039/TF9656102102>
28. E.B. Freyer, J.C. Hubbard, D.H. Andrews, *J. Am. Chem. Soc.* (1929). <https://doi.org/10.1021/ja01378a014>
29. S.V.S. Gadaibaev, M.I. Shakhparonov, *Moscow Univ. Chem. Bull.* **28**, 66 (1973)
30. J. George, N.V. Sastry, S.R. Patel, M.K. Valand, *J. Chem. Eng. Data* (2002). <https://doi.org/10.1021/je0102681>
31. D.S. Gill, J. Singh, R. Ludwig, M.D. Zeidler, *J. Chem. Soc., Faraday Trans.* (1993) doi:<https://doi.org/10.1039/FT9938903955>
32. A.Z. Golik, Y.I. Shimanskii, N.M. Kobiichuk, *Ukr. Fiz. Zh* **3**, 537 (1958)
33. B. González, A. Domínguez, J. Tojo, *J. Chem. Eng. Data* (2004). <https://doi.org/10.1021/je0342825>
34. S. Govindarajan, V. Kannappan, M.D. Naresh, K. Venkataboopathy, B. Lokanadam, *J. Mol. Liq.* (2003). [https://doi.org/10.1016/S0167-7322\(03\)00156-9](https://doi.org/10.1016/S0167-7322(03)00156-9)
35. M. Iglesias, B. Orge, M. Domínguez, J. Tojo, *Phys. Chem. Liq.* (1998). <https://doi.org/10.1080/00319109808032796>
36. M. Iglesias, B. Orge, M.M. Piñeiro, B.E. de Cominges, G. Marino, J. Tojo, *J. Chem. Eng. Data* (1998). <https://doi.org/10.1021/je980041z>
37. H. Iloukhani, Z. Rostami, *J. Solution Chem.* (2003). <https://doi.org/10.1023/A:1024524928767>
38. S.K.K. Jatkar, *J. Indian I. Sci. Sec. A-Sci.* **22**, 19 (1939)
39. A. Kumar, O. Prakash, S. Prakash, *J. Chem. Eng. Data* (1981). <https://doi.org/10.1021/je00023a021>
40. G. Marino, M. Iglesias, B. Orge, J. Tojo, M.M. Piñeiro, *J. Chem. Thermodyn.* (2000). <https://doi.org/10.1006/jcht.1999.0615>
41. A. Moses Ezhil Raj, L.B. Resmi, V.B. Jothy, M. Jayachandran, C. Sanjeeviraja, *Fluid Phase Equilib.* (2009) doi:<https://doi.org/10.1016/j.fluid.2009.04.009>
42. H. Ogawa, S. Murakami, *J. Solution Chem.* **16**, 315 (1987)
43. T. Ohmori, Y. Kimura, N. Hirota, M. Terazima, *Phys. Chem. Chem. Phys.* (2001). <https://doi.org/10.1039/b101773j>
44. B. Orge, M. Iglesias, A. Rodríguez, J.M. Canosa, J. Tojo, *Fluid Phase Equilib.* (1997). [https://doi.org/10.1016/S0378-3812\(97\)00031-9](https://doi.org/10.1016/S0378-3812(97)00031-9)
45. S. Parthasarathy, A.F. Chhapgar, H. Singh, *Nuovo Cimento* (1953). <https://doi.org/10.1007/BF02786196>
46. S.M. Pereira, M.A. Rivas, T.P. Iglesias, *J. Chem. Eng. Data* (2002). <https://doi.org/10.1021/je0200281>
47. S.M. Pereira, M.A. Rivas, J.L. Legido, T.P. Iglesias, *J. Chem. Thermodyn.* (2003). [https://doi.org/10.1016/S0021-9614\(02\)00311-7](https://doi.org/10.1016/S0021-9614(02)00311-7)
48. M.J.W. Povey, S.A. Hindle, J.D. Kennedy, Z. Stec, R.G. Taylor, *Phys. Chem. Chem. Phys.* (2003). <https://doi.org/10.1039/B206425A>
49. I.B. Rabinovich, V.G. Golov, N.A. Efimova, S.M. Rustamov, *Dokl. Akad. Nauk* **114**, 590 (1957)
50. M. Rama Rao, *Indian J. Phys.* **14**, 109 (1940)
51. G.V.R. Rao, P.B.S. Sri, A.V. Sarma, C. Rambabu, *Ind. J. Pure Appl. Phys.* **45**, 135 (2007)
52. J.M. Resa, C. González, M.A. Fanega, S. Ortiz de Landaluce, J. Lanz, *J. Food Eng.* (2002). [https://doi.org/10.1016/S0260-8774\(01\)00045-0](https://doi.org/10.1016/S0260-8774(01)00045-0)
53. A. Rodríguez, J. Canosa, J. Tojo, *J. Chem. Eng. Data* (2001). <https://doi.org/10.1021/je010148d>
54. T. Sun, S.N. Biswas, N.J. Trappeniers, A. ten Seldam, *J. Chem. Eng. Data* (1988). <https://doi.org/10.1021/je00054a002>
55. W. Wilson, D. Bradley, *J. Acoust. Soc. Am.* (1964). <https://doi.org/10.1121/1.1918956>
56. Z. Yu, H. Gao, H. Wang, L. Chen, *J. Chem. Eng. Data* (2011). <https://doi.org/10.1021/je2000606>
57. M.T. Zafarani-Moattar, H. Shekaari, *J. Chem. Eng. Data* (2005). <https://doi.org/10.1021/je050165t>
58. K. Meier, S. Kabelac, *Rev. Sci. Instrum.* (2006). <https://doi.org/10.1063/1.2400019>

59. C.W. Scholz, M. Richter, *Int. J. Thermophys.* (2021). <https://doi.org/10.1007/s10765-020-02778-6>
60. C.W. Scholz, Y. Sanchez-Vicente, T. Tananilgul, M. Thol, J.P.M. Trusler, M. Richter, *J. Chem. Eng. Data* (2020). <https://doi.org/10.1021/acs.jced.0c00299>
61. R. Wegge, M. Richter, R. Span, *J. Chem. Eng. Data* (2015). <https://doi.org/10.1021/je501065g>
62. R. Wegge, M. Richter, R. Span, *Fluid Phase Equilib.* (2016). <https://doi.org/10.1016/j.fluid.2015.10.034>
63. H. Gedanitz, M.J. Dávila, E. Baumhögger, R. Span, *J. Chem. Thermodyn.* (2010). <https://doi.org/10.1016/j.jct.2009.11.002>
64. G.R. Harris, *J. Acoust. Soc. Am.* (1981). <https://doi.org/10.1121/1.386671>
65. F.H. Dubberke, E. Baumhögger, J. Vrabec, *Rev. Sci. Instrum.* (2015). <https://doi.org/10.1063/1.4921478>
66. K. Meier, Dissertation, Helmut-Schmidt-Universität - Universität der Bundeswehr Hamburg, 2006, <https://www.hsu-hh.de/thermodynamik/wp-content/uploads/sites/741/2017/09/Habilitationsschrift-Meier.pdf>. Accessed 8 May 2020
67. W. Wagner, A. Pruß, *J. Phys. Chem. Ref. Data* (2002). <https://doi.org/10.1063/1.1461829>
68. Verein Deutscher Eisenhüttenleute, *Stahl-Eisen-Werkstoffblätter* (1997)
69. R. Span, R. Beckmüller, T. Eckermann, S. Herrig, S. Hielscher, A. Jäger, T. Neumann, S. Pohl, B. Semrau, M. Thol, *TREND. Thermodynamic Reference and Engineering Data* (Lehrstuhl für Thermodynamik: Ruhr-Universität Bochum, Bochum, 2019)

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.