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Viscosities and densities of different alcohols (1-propanol, 2-propanol, 1-pentanol and 2-pentanol) at high pressures



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1. Introduction

Viscosity is a thermophysical property of fluids that plays a key role in a wide range of industrial applications, many of which occurs at high pressure conditions. Therefore, viscosity values provide a valuable information in order to characterize the behavior of the concerned fluids at those pressures.

Unfortunately, high pressure viscosity determination is not a trivial task as there are quite limited experimental techniques available with low uncertainties. But in the last few years, experimental equipment has been successfully developed in our laboratory [1,2], allowing to determine accurate viscosities at high pressure.

Alcohols are important for many industrial processes; they are used as solvents or in the synthesis of other compounds. One of the main applications, which is increasing the interest of these compounds, is related with second-generation biofuels since those alcohols can be produced in biorefineries from non-food biomass and can be mixed with gasolines or diesel to substituted other oxygenated compounds. A performant use of these biofuels implies an

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ABSTRACT

High pressure viscosity data are necessary to complement fluid characterization, but their accurate determination is always a challenge due to the lack of these data. This work focuses on characterizing four alcohols (1-propanol, 2-propanol, 1-pentanol and 2-pentanol) through viscosity measurements using two different equipment: a falling body viscometer and a vibrating wire viscometer. Both techniques can measure up to 100 MPa, with relative expanded uncertainties (k = 2) of 3.5% and 1.5%, respectively. Since both viscometers need, as an input data, density of the measured compounds, a vibrating tube Anton Paar DMA-HPM densimeter is used to determine their densities in the same ranges of pressure and temperature with an expanded uncertainty (k = 2) of 0.70 kg·m⁻³. Finally, the experimental data are fitted to Tammann-Tait equation for densities and VFT model for viscosities obtaining standard deviations within the uncertainty of the measurements.

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accurate knowledge of properties such as density and viscosity in wide ranges of temperature and pressure.

High pressure viscosities of 1-butanol and 2-butanol were already provided for this purpose in former works [3,4] using a vibrating wire viscometer technique. The aim of the present work is to extend this knowledge, reporting new measurements for other alcohols such as 1-propanol, 2-propanol, 1-pentanol and 2-pentanol, from 0.1 MPa to 100 MPa and four isotherms from 293.15 K to 353.15 K for both propanols and six isotherms from 293.15 K to 373.15 K for both pentanols. In addition, densities for these compounds were measured, from 0.1 MPa to 100 MPa and at six different isotherms from 293.15 K to 373.15 K, using an Anton Paar vibrating tube densimeter which was automated in our laboratory [5].

2. Experimental section

2.1. Materials

The alcohols were purchased from Sigma-Aldrich and Fluka with the highest purity available, chromatography quality reagents with a purity > 0.995 (GC) for 1-propanol; >0.998 for 2-propanol; >0.99 for 1-pentanol and finally > 0.98 for 2-pentanol. Additionally, their water content was determined with a Mettler Toledo C20



coulometric Karl Fischer titrator. Molecular sieves were used in order to control and reduce their water. The details of the compounds are listed in Table 1.

2.2. Experimental techniques

The three experimental techniques used in this work were extensively described in previous papers [1-5], nevertheless, a brief explanation of the main features of them is presented in the next sections.

2.2.1. Vibrating tube densimeter

Densities were obtained using a vibrating tube densimeter (Anton Paar DMA-HPM) calibrated with water and vacuum, following the procedure previously described in Ref. [5]. The calibration is checked with toluene regularly obtaining deviations lower than the uncertainty of the measurements. This rig is able to measure density in the range of (0–3000) kg·m⁻³ with a resolution of 10^{-2} kg·m⁻³. The apparatus is fully automated using the Agilent VEE Pro software to control the system and the data acquisition. Uncertainties calculations were performed following "The guide to the expression of uncertainty in measurement JCGM100: 2008" [6] whose procedure was explained in Ref. [5], obtaining an expanded uncertainty (k = 2) <0.70 kg·m⁻³.

2.2.2. Falling body viscometer

Viscosities for 1-propanol and 2-propanol were obtained by means of a falling body viscometer whose principle of operation is based on measuring the fall time of a cylindrical ferromagnetic stainless-steel body passing between two fixed coils through a vertical tube. The cell, originally developed by "Groupe de Haute Pression, Laboratoire des Fluides Complexes" of the University of Pau [7], was implemented in our laboratory. All the experimental setup was entirely developed by our research group and described in Refs. [1,8]. This apparatus is able to work in wide ranges of pressure (0.1–140 MPa) and temperature (from 253.15 K to 523.15 K), providing good measurements as shown in other papers [2,9–11]. Temperature of the system is measured by four Pt100 probes, with an expanded uncertainty U(T) = 20 mK. Moreover, pressure is determined by means of a digital Druck DPI 104 manometer with a relative expanded uncertainty $U_r(p) = 0.02\%$. Both devices were calibrated in TERMOCAL laboratory using primary standards traceable to national standards and those values correspond to the calibration uncertainty.

This experimental technique requires a calibration procedure which is based on the use of well-known viscosity reference fluids at the same (p, T) conditions and viscosity range in which the viscometer is used [12,13]. Specifically, the calibration was performed at p = (0.1-100) MPa and T = (293.15-393.15) K using dodecane [1,14] as reference fluid, the equipment is periodically calibrated and its drift is much lower than its uncertainty.

Uncertainty calculations were undertaken following the guide JCGM 100:2008 [6], and all the details can be found in previous works [2,8]. The viscosity uncertainty was evaluated at the limits of the calibration range (0.630 mPa·s for the lowest viscosity and

Table 1	
Material	description.

4.929 mPa·s for the highest one). Taking into account these viscosity limits, the relative expanded uncertainty (0.95 confidence level) $U_{\rm r}(\eta)$ varies from 2.5% for the highest viscosity value up to 3.5% for the lowest value.

2.2.3. Vibrating wire viscometer

There is also a vibrating-wire viscometer available in our laboratory that can be used to accurately measure dynamic viscosities up to 35 mPa·s, at working temperatures from 288.15 K to 423.15 K and pressures up to 140 MPa [1,3]. The technique uses a goldplatted tungsten wire anchored at both ends [15,16] as sensor. The frequency of vibration of the wire due to the constant magnetic field and the circulation of a sinusoidal current through the wire is related to the viscosity and density of the fluid. The sensor is placed into a pressure vessel, and it is under an external constant magnetic field. The pressure vessel is immersed in a high precision thermostatic bath and the fluid temperature is measured using two platinum resistance thermometers (PRT) whose expanded uncertainty is U(T) = 20 mK. A Druck DPI 104 transducer is used for pressure measurements with relative expanded uncertainties (k = 2) of $U_{\rm r}(p)$ = 0.02%. Both temperature and pressure sensors were also calibrated in our laboratory using primary standards traceable to national standards. Finally, the radius of the tungsten wire was calibrated using toluene as reference hydrocarbon [17] and the viscosity measurements were checked with dodecane [14]. The calibration is periodically checked to assure that the drift between calibrations is below the uncertainty. The relative expanded uncertainty of the viscosity measurements is estimated better than 1.5% for a cover factor k = 2 (0.95 level of confidence). The performance of this equipment was proven in previous papers [1,3,4].

3. Results and discussion

3.1. Density measurements

Density measurements of 1-propanol, 2-propanol, 1-pentanol and 2-pentanol were performed, by means of an Anton Paar vibrating tube densimeter, from 0.1 MPa to 100 MPa and at six different isotherms from 293.15 K to 373.15 K. These experimental data results are detailed in Tables 2, 3, 4 and 5, respectively.

Additionally, in order to extend the applicability of the measurements, the experimental data were fitted to a modified Tammann–Tait equation (Eq. (1)) and the fitting parameters and the standard deviation obtained are reported in Table 6.

$$\rho(T,p) = \frac{A_0 + A_1 T + A_2 T^2}{1 - C \ln \left(\frac{B_0 + B_1 T + B_2 T^2 + p/MPa}{B_0 + B_1 T + B_2 T^2 + 0.1}\right)}$$
(1)

As can be observed from Table 6, standard deviations of Tammann-Tait fitting are lower than the experimental uncertainty proving the validity of this fitting equation. In order to visualize the fittings, the residuals of density for the four alcohols are plotted in Fig. 1.

Chemical name	CAS	Source	Mass fraction purity ^a	Water content ^b	Purification method
1-Propanol	71-23-8	Sigma-Aldrich	≥ 0.995	203 ppm	none
2-Propanol	67-63-0	Sigma-Aldrich	≥ 0.998	189 ppm	none
1-Pentanol	71-41-0	Sigma-Aldrich	≥ 0.99	< 100 ppm	none
2-Pentanol	6032-29-7	Fluka	\ge 0.98	< 100 ppm	none

^a Stated by the supplier and checked by gas chromatography.

^b Coulometric Karl Fischer titrator.

	$ ho/kg \cdot m^{-3}$ T/K					
p/MPa	293.15	313.15	333.15	353.15	363.15	373.15
0.100	803.82	787.71	770.48	752.34	742.69	
1.00	804.49	788.34	771.36	753.29	743.78	733.68
5.00	807.55	791.73	775.18	757.69	748.49	738.76
10.00	811.23	795.74	779.76	762.79	753.93	744.61
20.00	818.03	803.31	788.12	772.09	763.80	755.09
30.00	824.38	810.19	795.63	780.40	772.56	764.42
40.00	830.27	816.62	802.64	788.04	780.53	772.73
50.00	835.88	822.61	809.09	795.03	787.81	780.38
60.00	841.11	828.23	815.11	801.49	794.61	787.43
70.00	846.08	833.55	820.77	807.64	800.94	793.98
80.00	850.87	838.60	826.18	813.38	806.88	800.19
100.00	859.75	847.99	836.12	823.93	817.80	811.47

^a Expanded uncertainties (k = 2): U(T) = 0.02 K; $U_r(p) = 0.0002$ and $U(\rho) = 0.70$ kg·m⁻³

Table 3

Experimental densities, ρ , for 2-propanol at different conditions of temperature, T, and pressure, p.^{a.}

$ ho/{ m kg} \cdot { m m}^{-3}$						
	T/K					
p/MPa	293.15	313.15	333.15	353.15	363.15	373.15
0.100	786.48	769.18	750.16	729.33	717.94	705.85
1.00	787.14	769.93	751.15	730.48	719.19	707.29
5.00	790.53	773.74	755.60	735.65	724.88	713.53
10.00	794.53	778.26	760.74	741.60	731.31	720.53
20.00	801.97	786.52	770.01	752.21	742.72	732.88
30.00	808.72	793.98	778.31	761.53	752.65	743.48
40.00	815.00	800.82	785.93	769.93	761.52	752.85
50.00	820.89	807.23	792.89	777.58	769.57	761.29
60.00	826.43	813.16	799.27	784.59	777.00	769.04
70.00	831.62	818.75	805.33	791.18	783.80	776.14
80.00	836.57	824.06	811.02	797.28	790.20	782.84
100.00	845.80	833.84	821.47	808.47	801.75	794.87

^a Expanded uncertainties (*k* = 2): *U*(*T*) = 0.02 K; $U_r(p) = 0.0002$ and $U(\rho) = 0.70$ kg·m⁻³.

Table 4

Experimental densities, ρ , for 1-pentanol at different conditions of temperature, *T*, and pressure, *p*.^{a.}

ρ/kg⋅m [−]						
	T/K					
p/MPa	293.15	313.15	333.15	353.15	363.15	373.15
0.100	814.80	800.07	784.39	768.24	759.67	750.66
1.00	815.34	800.61	785.18	769.03	760.55	751.66
5.00	818.05	803.62	788.55	772.80	764.57	756.00
10.00	821.30	807.17	792.57	777.24	769.27	760.99
20.00	827.46	813.90	799.93	785.42	777.89	770.10
30.00	833.18	820.12	806.71	792.81	785.69	778.29
40.00	838.53	825.90	813.02	799.67	792.78	785.75
50.00	843.60	831.34	818.87	805.99	799.36	792.61
60.00	848.45	836.52	824.41	811.92	805.54	798.99
70.00	853.01	841.40	829.56	817.51	811.29	804.93
80.00	857.43	846.05	834.51	822.74	816.78	810.60
100.00	865.59	854.71	843.70	832.47	826.79	820.90

^a Expanded uncertainties (k = 2): U(T) = 0.02 K; $U_{t}(p) = 0.0002$ and $U(\rho) = 0.70$ kg·m⁻³.

3.2. Viscosity measurements

Viscosity measurements are presented in Table 7 and Table 8 for 1-propanol and 2-propanol, respectively. Both set of measurements were carried out, using a falling body viscometer, at pressures from 0.1 MPa to 100 MPa and temperatures of 293.15 K, 313.15 K, 333.15 K and 353.15 K.

Blank cells of Table 8 correspond to viscosities which are outside calibration limits (0.630 mPa·s and 4.929 mPa·s). That is the reason why viscosities of 2-propanol at 293.15 K at 100 MPa and 353.15 K (0.1 and 5 MPa) are not given. However, the viscosity value of 0.610 mPa·s, which corresponds to 2-propanol at 353.15 K and 10 MPa, is slightly below the lowest limit of 0.630 mPa·s, showing a relative deviation of 3.1% from this limit. Considering that the falling body equipment has an expanded uncertainty (k = 2) of 3.5% for low viscosities, that value is within calibration domains since it is compatible with the uncertainty of the apparatus.

Additionally, viscosities of 1-pentanol and 2-pentanol were measured from 0.1 MPa to 100 MPa and at six isotherms between

$ ho/{ m kg} \cdot { m m}^{-3}$						
	T/K					
p/MPa	293.15	313.15	333.15	353.15	363.15	373.15
0.100	809.33	792.87	774.59	755.37	745.19	734.74
1.00	809.88	793.43	775.50	756.37	746.31	735.84
5.00	812.76	796.69	779.31	760.74	751.02	740.96
10.00	816.23	800.56	783.77	765.78	756.44	746.73
20.00	822.70	807.78	791.89	774.96	766.20	757.15
30.00	828.68	814.40	799.24	783.15	774.89	766.40
40.00	834.28	820.55	806.04	790.65	782.75	774.67
50.00	839.57	826.30	812.28	797.50	789.93	782.15
60.00	844.58	831.66	818.12	803.88	796.58	789.13
70.00	849.26	836.77	823.61	809.87	802.82	795.55
80.00	853.79	841.62	828.83	815.47	808.64	801.67
100.00	862.17	850.56	838.43	825.75	819.29	812.66

^a Expanded uncertainties (k = 2): U(T) = 0.02 K; $U_r(p) = 0.0002$ and $U(\rho) = 0.70$ kg·m^{-3.}

Table 6

Fitting parameters of modi	ed Tammann-Tait equation	(Eq. (1)) and standard	1 deviations (σ) fo	or density correlations.
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Parameters	1-Propanol	2-Propanol	1-Pentanol	2-Pentanol
$A_0/\text{kg}\cdot\text{m}^{-3}$	915.90	810.94	918.34	898.95
$A_1/\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-1}$	0.01206	0.63763	-0.00554	0.18778
$A_2/\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-2}$	-0.00134	-0.00246	-0.00119	-0.00168
B ₀ /MPa	332.61	322.52	317.96	414.27
$B_1/MPa \cdot K^{-1}$	-1.0627	-1.0648	-0.89401	-1.4938
$B_2/MPa K^{-2}$	0.000816	0.000808	0.000534	0.001367
С	0.08803	0.08630	0.08608	0.08440
$\sigma/\text{kg}\cdot\text{m}^{-3}$	0.12	0.17	0.11	0.07

293.15 K and 373.15 K. These measurements were performed by means of a vibrating wire viscometer. The experimental data are summarized in Table 9 and Table 10 for 1-pentanol and 2-pentanol, respectively, including the uncertainty of the measured data.

Finally, experimental viscosities were correlated using a modified VFT model, (Eq. (2)) successfully used by Comuñas et al. [18], Paredes et al. [19] and in a previous work [2].

$$\eta(p,T) = A \cdot exp\left(\frac{B}{T-C}\right) \cdot \left(\frac{p/MPa + E(T)}{0.1 + E(T)}\right)^{F}$$
(2)

Being $E(T) = E_0 + E_1 \cdot T + E_2 \cdot T^2$. The results of fitting parameters and standard deviations are presented in Table 11.

The results of the standard deviations are very good for 1propanol and 2-propanol, 0.005 mPa·s and 0.010 mPa·s, respectively, being within the uncertainty of the measurements, however, the values obtained for 1-pentanol (0.070 mPa·s) and 2pentanol (0.080 mPa·s) are worse proving the limitation of the model to describe the behaviour of these fluids due to their wide range of viscosity variation. These differences in the fittings are shown graphically in Fig. 2 where the residuals of viscosity are plotted as a function of pressure for the different temperatures.

4. Discussion and conclusions

In order to visualize and discuss the density behaviour of these measured alcohols, the experimental data and the fitting equation are depicted in Figs. 3 and 4.

As can be directly observed from the experimental data, density follows the next sequence: 1-pentanol > 2-pentanol > 1-pro panol > 2-propanol at the same conditions and the alcohol group position has a direct influence in density, being denser those alcohols with the position 1-alcohol, it was also observed for 1-butanol [20] and 2-butanol [21]. This behaviour is related to a higher self-association due to the hydrogen bonding in primary alcohols.

As expected, density increases with increasing pressure and decreasing temperature. The growth of density, due to the effect of increasing the pressure from 0.1 MPa to 100 MPa, is quite similar for all the studied alcohols ranging from 7% (at T = 293.15 K) up to 10.1% (at T = 363.15 K) for 1-propanol, 7.5% (at T = 293.15 K) up to 12.6% (at T = 373.15 K) for 2-propanol, 6.2% (at T = 293.15 K) up to 9.4% (at T = 373.15 K) for 1-pentanol and 6.5% (at T = 293.15 K) up to 10.6% (at T = 373.15 K) for 2-pentanol. Moreover, this increase is larger at higher temperatures, and 2-propanol is the alcohol with the highest effect due to the increasing pressure.

As regards the effect of increasing temperature, it is compared the decrease in density when temperature is changed from 293.15 K to 373.15 K. The lowering of density varies for the studied alcohols as follows: from 5.6% (at p = 100 MPa) up to 8.8% (at p = 1 MPa) for 1-propanol, from 6% (at p = 100 MPa) up to 10.3% (at p = 0.1 MPa) for 2-propanol, from 5.2% (at p = 100 MPa) up to 7.9% (at p = 0.1 MPa) for 1-pentanol and from 5.7% (at p = 100 MPa) up to 9.2% (at p = 0.1 MPa) for 2-pentanol. It can be concluded that the highest effect is observed at the lowest pressure, being similar for all the alcohols, however, 2-propanol is slightly more sensitive to these changes.

As commented in Section 3.1, the standard deviations of the density fitting are lower than the uncertainty of the measurements showing the goodness of that equation to represent the density as a function of pressure and temperature as is also observed in Fig. 1. These standard deviations range from 0.07 kg·m⁻³ for 2-pentanol up to 0.17 kg·m⁻³ for 2-propanol, being the expanded uncertainty (k = 2) of 0.70 kg·m⁻³.

In addition, the derived properties, isobaric thermal expansivity (α_p) and isothermal compressibility (κ_T) have been obtained

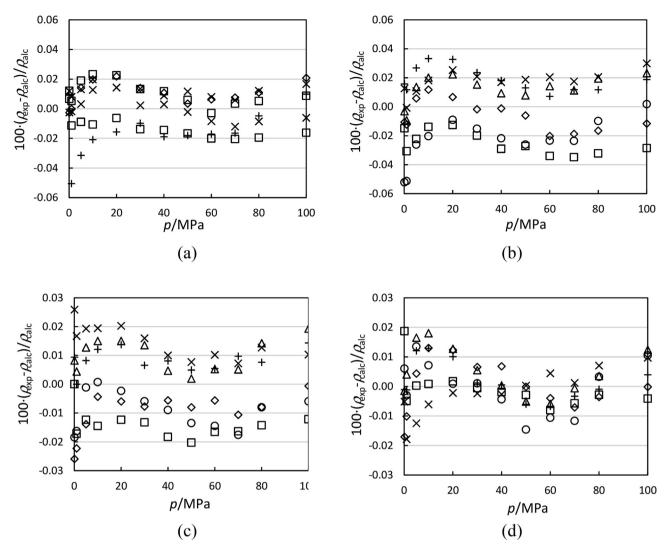


Fig. 1. Relative deviations of experimental density measurements and calculated values using Tamman-Tait equation for (a) 1-propanol, (b) 2-propanol, (c) 1-pentanol and (d) 2-pentanol at temperatures: (×) 293.15 K; (□) 313.15 K; (◊) 333.15 K; (□) 373.15 K; (○) 373.15 K.

Experimental dynamic viscosities, η (mPa·s), for 1-propanol at different conditions of temperature, *T*, and pressure, *p*.^a

η/mPa·s	T/K			
p/MPa	293.15	313.15	333.15	353.15
0.100	2.203	1.388	0.924	0.638
5.00	2.293	1.439	0.964	0.669
10.00	2.388	1.500	1.006	0.700
20.00	2.578	1.618	1.087	0.760
30.00	2.771	1.737	1.166	0.821
40.00	2.97	1.861	1.245	0.877
60.00	3.37	2.110	1.406	0.994
80.00	3.79	2.373	1.575	1.117
100.00	4.24	2.650	1.755	1.234

^a Expanded uncertainties (k = 2): U(T) = 0.02 K; $U_r(p) = 0.0002$; and $U_r(\eta) = 0.035$.

through the derivation of the modified Tammann-Tait equation using the following equations:

$$\begin{aligned} \alpha_{p}(T,p) &= \frac{\sum_{i=1}^{n} (i)A_{i}T^{i-1}}{\sum_{i=0}^{n}A_{i}T^{i}} - \frac{C[0.1-p]}{[B(T)+p][B(T)+0.1]} \\ &\times \frac{\sum_{j=1}^{m} (j)B_{j}T^{j-1}}{\left[1 - Cln\left[\frac{B(T)+p}{B(T)+0.1}\right]\right]} \end{aligned} \tag{3}$$

$$\kappa_T(T,p) = \frac{C}{\left[B(T) + p\right] \left[1 - Cln\left[\frac{B(T) + p}{B(T) + 0.1}\right]\right]}$$
(4)

where $B(T) = B_0 + B_1 \cdot T + B_2 \cdot T^2$. The calculated values are summarized in Tables S1 and S2 as supplementary data and they are plotted in Figs. 5 and 6 for the isobaric thermal expansivity and isothermal compressibility respectively.

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Table 8

Experimental dynamic viscositie	s, η (mPa·s), for 2-propanol at different	conditions of temperature, <i>T</i> , and pressure, <i>p</i> . ^a
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η/mPa∙s									
	<u>Т/К</u>	T/K							
p/MPa	293.15	313.15	333.15	353.15					
0.100	2.436	1.361	0.831						
5.00	2.554	1.427	0.881						
10.00	2.692	1.498	0.932	0.610					
20.00	2.96	1.646	1.026	0.676					
30.00	3.23	1.799	1.118	0.740					
40.00	3.50	1.954	1.211	0.806					
60.00	4.06	2.279	1.393	0.933					
80.00	4.64	2.615	1.586	1.067					
100.00		2.98	1.794	1.202					

^a Expanded uncertainties (k = 2): U(T) = 0.02 K; $U_r(p) = 0.0002$; and $U_r(\eta) = 0.035$.

Table 9

Experimental dynamic viscosities, η (mPa·s), for 1-pentanol at different conditions of temperature, *T*, and pressure, *p*.^a

η/mPa∙s						
	T/K					
p/MPa	293.15	313.15	333.15	353.15	363.15	373.15
0.100	4.046	2.346	1.449	0.941	0.778	0.718
1.00	4.052	2.375	1.463	0.965	0.792	0.745
5.00	4.263	2.469	1.538	0.999	0.826	0.783
10.00	4.455	2.555	1.591	1.053	0.864	0.850
20.00	4.878	2.865	1.790	1.171	0.945	0.901
30.00	5.369	3.055	1.899	1.268	1.046	0.980
40.00	5.625	3.298	2.036	1.374	1.153	1.067
50.00	6.181	3.594	2.173	1.464	1.209	1.171
60.00	6.83	3.865	2.387	1.576	1.311	1.219
70.00	7.37	4.175	2.507	1.682	1.398	1.306
80.00	7.90	4.402	2.664	1.779	1.482	1.385
100.00	9.50	5.133	3.052	2.020	1.714	1.524

^a Expanded uncertainties (k = 2): U(T) = 0.02 K; $U_r(p) = 0.0002$; and $U_r(\eta) = 0.015$.

Table 10

Experimental dynamic viscosities, η (mPa·s), for 2-pentanol at different conditions of temperature, *T*, and pressure, *p*.^{a.}

η/mPa∙s						
	T/K					
p/MPa	293.15	313.15	333.15	353.15	363.15	373.15
0.100	4.217	1.985	1.088	0.680	0.551	0.461
1.00	4.299	2.021	1.107	0.682	0.562	0.468
5.00	4.588	2.133	1.166	0.727	0.593	0.496
10.00	4.787	2.256	1.251	0.777	0.627	0.529
20.00	5.449	2.573	1.384	0.875	0.706	0.591
30.00	6.113	2.824	1.544	0.968	0.795	0.654
40.00	6.85	3.131	1.700	1.052	0.867	0.720
50.00	7.43	3.534	1.847	1.149	0.941	0.781
60.00	8.21	3.753	1.981	1.235	1.010	0.843
70.00	9.21	4.071	2.125	1.298	1.069	0.907
80.00	10.28	4.407	2.352	1.410	1.150	0.967
100.00	12.86	5.199	2.711	1.614	1.319	1.075

^a Expanded uncertainties (k = 2): U(T) = 0.02 K; $U_r(p) = 0.0002$; and $U_r(\eta) = 0.015$

Table 11

Fitting parameters of Eq. (2)	and standard deviations	σ for the viscosity correlation.
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Parameters	1-Propanol	2-Propanol	1-PENTZANOL	2-Pentanol	
A/mPa·s	0.0024	0.0024	0.0038	0.0080	
B/K	1872.9	1872.9 1511.6 1616.1		915.07	
C/K	18.432	74.203	61.359	147.19	
E ₀ /MPa	0.9954	0.7705	0.0655	-0.0615	
$E_1/MPa\cdot K^{-1}$	1.0130	0.8243	2.3540	0.9345	
$E_2/MPa \cdot K^{-2}$	-0.0015	-0.0013	-0.0027	0.0013	
F	1.4057	1.3293	4.2201	4.7696	
σ/mPa⋅s	0.005	0.010	0.070	0.080	

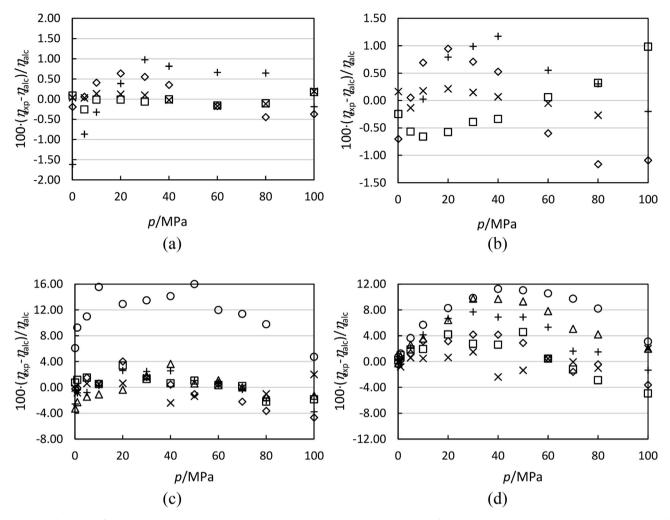


Fig. 2. Relative deviations of experimental viscosity measurements and calculated values using VFT equation for (a) 1-propanol, (b) 2-propanol, (c) 1-pentanol and (d) 2-pentanol at temperatures: (×) 293.15 K; (□) 313.15 K; (◊) 333.15 K; (+) 353.15 K; (△) 363.15 K; (○) 373.15 K.

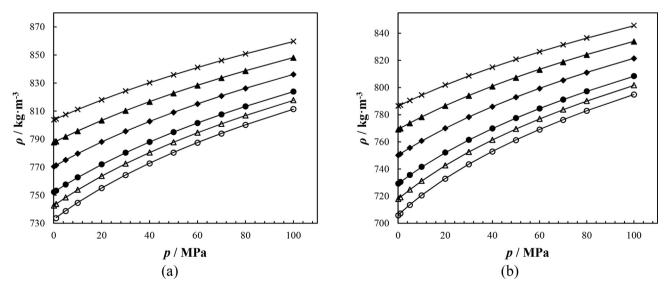


Fig. 3. Experimental densities as function of pressure for (a) 1-propanol and (b) 2-propanol at temperatures: (\times) 293.15 K; (\bigstar) 313.15 K; (\bigstar) 333.15 K; (\bigstar) 353.15 K; (\bigstar) 363.15 K; (\circlearrowright) 373.15 K; (\bigstar) 373.15 K. Lines represent the calculated values using modified Tammann-Tait equation with parameters given in Table 6.

It can be observed that the isobaric thermal expansivity diminishes with increasing pressure for the alcohols studied. The highest reduction is obtained at the highest temperature for all, being close to 51% for 2-propanol. On the other hand, α_p increases with increasing temperature at a given pressure, being the highest increase at 0.1 MPa (close to 66% for 2-propanol). This behaviour

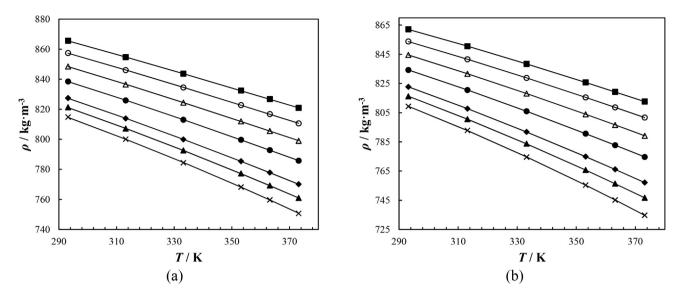


Fig. 4. Experimental densities as function of temperature for (a) 1-pentanol and (b) 2-pentanol at pressures: (×) 0.1 MPa; (▲) 10 MPa; (▲) 20 MPa; (▲) 40 MPa; (△) 60 MPa; (○) 80 MPa and (■) 100 MPa. Lines represent the calculated values using modified Tammann-Tait equation with parameters given in Table 6.

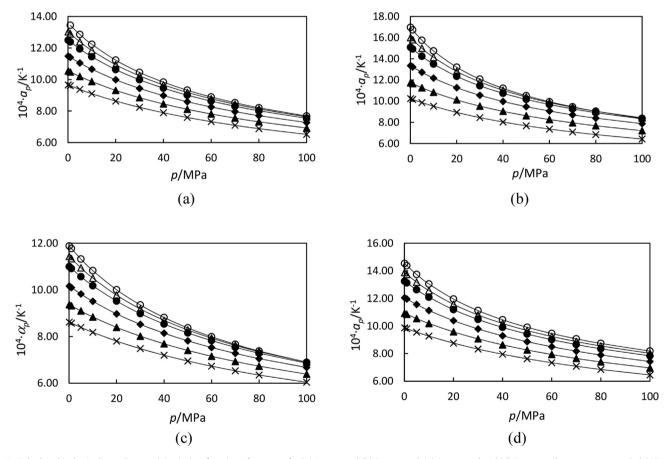


Fig. 5. Calculated isobaric thermal expansivity (α_p) as function of pressure for (a) 1-propanol (b) 2-propanol, (c) 1-pentanol and (d) 2-pentanol at temperatures: (×) 293.15 K; (\bigstar) 313.15 K; (\bigstar) 333.15 K; (\bigstar) 353.15 K; (\bigtriangleup) 363.15 K; (\circlearrowright) 373.15 K.

indicates that, at higher pressures, due to the decreasing of free intermolecular spaces, the alcohols also decrease their capacity to expand, in contrast with the effect of increasing temperature, where the association due to the hydrogen bonding decrease, increasing the free volume and the capacity to expand of the alcohols. Similarly, the isothermal compressibility lessens with increasing pressure for the studied alcohols at all temperatures, being the highest reduction at 373.15 K, whereas it grows with increasing temperature and the largest increase occurs at 0.1 MPa. 2-Propanol is the alcohol more sensitive to these effects so κ_T is increased by 111% for the change in temperature from 293.15 K

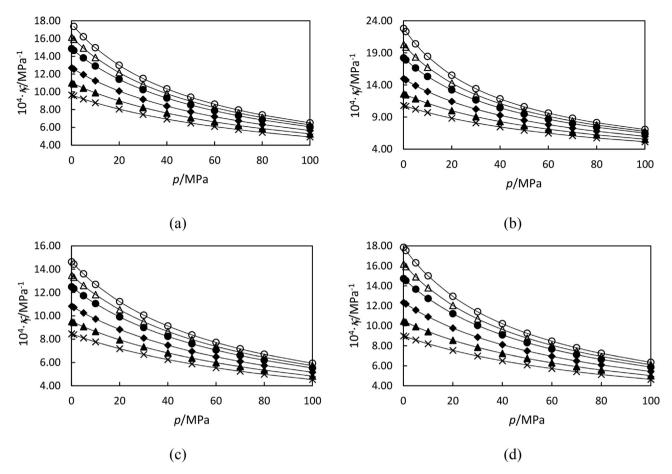


Fig. 6. Calculated isothermal compressibility (κ_T) as function of pressure for (a) 1-propanol (b) 2-propanol, (c) 1-pentanol and (d) 2-pentanol at temperatures: (×) 293.15 K; (\bigstar) 313.15 K; (\bigstar) 333.15 K; (\bigstar) 353.15 K; (\bigtriangleup) 353.15 K; (\circlearrowright) 353.15 K; (\bigstar) 353.15 K; (\circlearrowright) 353.15 K; (\circlearrowright) 353.15 K; (\bigstar) 353

to 373.15 K at 0.1 MPa and is decreased by 69 % for the change in pressure from 0.1 MPa to 100 MPa at 373.15 K. As pointed out previously, the capacity of these alcohols to contract is reduced when temperature is decreased or pressure is increased, because of the reduction the intermolecular free space.

Finally, an intensive literature search for these four pure alcohols was carried out. Most of the available density data are reported at atmospheric pressure and only the most significant ones were used in the comparison. Since some literature data are at different conditions, the comparison was performed using the modified Tammann-Tait correlation (with the parameters given in Table 6), through the statistical parameters: AAD (absolute average deviation), MD (absolute maximum deviation), Bias (average deviation) and RMS (root mean square). The results of the comparison can be found in Tables 12–15.

As can be seen from Tables 12 to 15, there are scarce density data in wide ranges of pressure and temperature like those presented in this work and, in general, there is a good agreement between our data and literature values.

Regarding the experimental viscosity data, in Figs. 7 and 8, the viscosity behaviour as a function of temperature and pressure is depicted. As expected, viscosity decreases with increasing temperature and decreasing pressure. As remarked by Sülzner et al. [57], the association in alcohols decreases with temperature, therefore the highest slope is observed at the lowest temperature.

Viscosities of pentanols are higher than propanols in the whole range studied, however the comparison between propanols or pentanols shows a different behavior depending on the temperature or pressure range. In this way, the viscosity of 2-propanol is lower than the viscosity of 1-propanol at 353.15 K and the whole pressure range, at 333.15 K and pressures up to 60 MPa and at 313.15 K and pressures up to 10 MPa. Furthermore, the viscosity of 1-pentanol is lower than the viscosity of 2-pentanol at 293.15 K and the whole pressure range, at 313.15 K and pressures of 80 MPa and 100 MPa.

Concerning the effect of increasing pressure from 0.1 MPa to 100 MPa in the viscosity, the average increase on viscosity is around 92% for 1-propanol, 117% for 2-propanol and 119% for 1-pentanol. In the case of 2-pentanol, this alcohol exhibits a higher influence than the other studied alcohols, thus, the increase varies from 209% at 293.15 K and 133% at 373.15 K.

On the other hand, when the temperature is increased, the viscosity is reduced. In order to compare the four alcohols, the reduction of viscosity is evaluated for an increment of temperature from 293.15 K to 353.15 K. These average reductions are by 71% for 1-propanol, 77% for both 2-propanol and 1-pentanol, and 85% for 2-pentanol.

In addition, the experimental viscosities were fitted to VFT model (Eq. (2)), the standard deviations are 0.005 mPa·s for 1-propanol and 0.010 mPa·s for 2-propanol, which are lower than the uncertainty of the falling body viscometer. However, the standard deviations are 0.070 mPa·s for 1-pentanol and 0.080 mPa·s for 2-pentanol. These alcohols were measured in a higher temperature range and, also, these values of viscosities vary in a wide range from 0.46 mPa·s up to 12.86 mPa·s for 2-pentanol and the model works worse for them.

Finally, the reliability of the measured viscosities was checked by comparison with the existing literature data. Most of the

Comparison between density literature data and our correlation for 1-propanol. Statistical parameters: AAD (absolute average deviation), MD (absolute maximum deviation), Bias (average deviation) and RMS (root mean square). Temperature and pressure ranges and number of experimental points (NP) of the literature data are included.

Reference	Year	T/K	p/MPa	NP	AAD%	MD%	Bias%	RMS/kg m ⁻³
Pan et al. [22]	2019	298.15-313.15	0.1	4	0.03	0.05	0.03	0.24
Shirazi et al. [23]	2019	293.15-323.15	0.1	7	0.05	0.06	0.05	0.39
Ramos-Estrada et al. [24]	2018	278.15-343.15	0.1	14	0.02	0.06	0.01	0.20
Iglesias-Silva et al. [25]	2016	283.15-363.15	0.1	17	0.02	0.03	0.00	0.16
Lu et al. [26]	2015	293.15-323.15	0.1	7	0.04	0.05	0.04	0.33
Wang et al. [27]	2015	293.15-323.15	0.1	6	0.02	0.03	-0.02	0.18
Pang et al. [28]	2007	293.15-333.15	0.1	9	0.14	0.30	-0.14	1.26
Cano-Gómez et al. [29]	2012	293.15-328.15	0.1	8	0.02	0.03	0.02	0.19
Mokhtarani et al. [30]	2009	283.15-333.15	0.1	11	0.02	0.03	0.02	0.18
Baylaucq et al. [31]	2009	293.15-353.15	0.1-100	24	0.12	0.19	-0.09	2.85
Watson et al. [32]	2006	293.15-353.15	0.1-65	98	0.03	0.10	-0.02	0.34
Kubota et al. [33]	1987	283.15-328.15	0.1-102	28	0.10	0.37	0.06	1.10
Alaoui et al. [34]	2014	293.15-353.15	0.1-100	44	0.03	0.08	-0.02	0.27
Nills et al. [35]	2019	293.15-313.15	0.1-35	24	0.02	0.07	-0.01	0.20
Dávila et al. [36]	2012	278.15-358.15	0.1-60	126	0.10	0.24	0.10	1.02
Zéberg-Mikkelsen et al. [37]	2005	303.15-333.15	0.1-30	28	0.02	0.03	-0.02	0.17
Abdulagatova et al. [38]	2008	298.15-373.15	0.11-39.56	36	0.06	0.19	0.04	0.62
Gil-Hernández et al. [39]	2006	298.15	0.2-19.7	18	0.07	0.16	-0.07	0.61
Ormanoudis et al. [40]	1991	298.15	0.1-33.88	10	0.06	0.08	-0.06	0.49
Papaioannou et al. [41]	1993	298.15	0.1-33.9	10	0.06	0.08	-0.07	0.60
Zúñiga-Moreno et al. [42]	2002	313.15-362.77	0.5-25	156	0.07	0.12	0.07	0.58
Yaginuma et al. [43]	1998	313.15	1–9.8	11	0.04	0.08	0.02	0.37

Table 13

Comparison between density literature data and our correlation for 2-propanol. Statistical parameters: AAD (absolute average deviation), MD (absolute maximum deviation), Bias (average deviation) and RMS (root mean square). Temperature and pressure ranges and number of experimental points (NP) of the literature data are included.

Reference	Year	T/K	p/MPa	NP	AAD%	MD%	Bias%	$RMS/kg \cdot m^{-3}$
Pang et al. [28]	2007	293.15-333.15	0.1	9	0.09	0.15	0.08	0.76
Yang et al. [44]	2006	293.15-343.15	0.1	7	0.41	0.67	0.41	3.42
Bhuiyan et al. [45]	2008	303.15-323.15	0.1	5	0.15	0.20	0.15	1.18
Hoga et al. [46]	2011	293.15-308.15	0.1	4	0.16	0.17	0.16	1.25
Martin [47]	2001	298.15-313.15	0.1	4	0.17	0.19	0.17	1.30
Sadeghi et al. [48]	2011	288.15-313.15	0.1	6	0.18	0.20	0.18	1.41
Nourozieh et al. [49]	2013	323.15	1-10.02	10	0.03	0.05	0.03	0.27
Mendo-Sánchez et al. [50]	2020	291.26-349.91	2-50	114	0.18	0.80	0.17	1.73
Alaoui et al. [51]	2012	293.15-353.15	0.1-100	77	0.14	0.19	-0.14	1.16
Kubota et al. [33]	1987	283.15-348.15	0.1-104.9	28	0.11	0.20	0.11	1.01
Zúñiga-Moreno et al. [42]	2002	313.15-362.77	0.5-25	156	0.11	0.21	0.10	0.97
Yaginuma et al. [52]	1997	313.15	1-9.8	11	0.18	0.20	-0.18	1.37
Moha et al. [53]	1998	303.15-343.15	0.1-100	18	0.21	0.26	-0.21	1.66
Stringari et al. [54]	2009	280.21-375.01	0.12-8.3	110	0.14	0.30	-0.06	1.22

Table 14

Comparison between density literature data and our correlation for 1-pentanol. Statistical parameters: AAD (absolute average deviation), MD (absolute maximum deviation), Bias (average deviation) and RMS (root mean square). Temperature and pressure ranges and number of experimental points (NP) of the literature data are included.

Reference	Year	T/K	P/MPa	NP	AAD%	MD%	Bias%	RMS/kg m ⁻³
Iglesias-Silva et al. [25]	2015	283.15-363.15	0.1	17	0.02	0.05	-0.01	0.15
Wang et al. [27]	2015	298.15-323.15	0.1	6	0.05	0.06	-0.03	0.37
Cano-Gómez et al. [29]	2012	293.15-328.15	0.1	8	0.07	0.08	0.07	0.59
Estrada-Baltazar et al. [55]	2015	293.15-363.15	0.1	15	0.02	0.04	0.02	0.20
Vargas-Ibáñez et al. [56]	2018	288.15-338.15	0.1	11	0.02	0.06	-0.02	0.20
Sülzner et al. [57]	1997	298.15-373.15	50-80	6	0.84	1.14	0.84	7.15
Wappmann et al. [58]	1995	283.5-373.4	0.1-100	31	0.15	0.34	-0.06	1.40
Garg et al. [59]	1993	323.15-373.15	0.1-10	60	0.03	0.09	-0.03	0.31
Zúñiga-Moreno et al. [60]	2007	313.08-362.5	1-25	150	0.09	0.16	-0.09	0.71
Boned et al. [61]	2008	293.15-363.15	0.1-100	88	0.01	0.04	0.00	0.14
Pimentel-Rodas et al. [62]	2019	298.15-323.15	2-29.98	48	0.05	0.11	-0.01	0.50
Nills et al. [35]	2019	293.15-313.15	0.1-35	24	0.13	0.17	0.13	1.06

found literature data were measured at 0.1 MPa and only a few of them provide data at high pressures. In the same way than for densities, experimental data from the literature were compared with the values calculated using the VFT model (Eq. (2)) and the parameters of Table 11 for 1-propanol and 2-propanol since the model fits the experimental data within the uncertainty of the measurements. These comparison results are shown in Tables 16 and 17.

As can be observed from the values of the average absolute deviations, most of them are between the uncertainty of our measurements for 1-propanol, only three of the eighteen references shown in Table 16 give data at high pressures being the AAD within the uncertainty for these three set of data. Slightly worse results are observed for 2-propanol, however, there are also disagreements between literature values pointing out the difficulty of performing accurate viscosity measurements.

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Table 15

Comparison between density literature data and our correlation for 2-pentanol. Statistical parameters: AAD (absolute average deviation), MD (absolute maximum deviation), Bias (average deviation) and RMS (root mean square)). Temperature and pressure ranges and number of experimental points (NP) of the literature data are included.

Reference	Year	T/K	p/MPa	NP	AAD%	MD%	Bias%	$RMS/kg \cdot m^{-3}$
González et al. [63]	2004	293.15-303.15	0.1	3	0.04	0.04	0.04	0.33
Vargas-Ibáñez et al. [56]	2018	288.15-338.15	0.1	11	0.03	0.05	-0.03	0.27
Mendo-Sánchez et al. [50]	2020	291.26-349.91	2-50	114	0.13	0.60	-0.12	1.57
Zúñiga-Moreno et al. [60]	2017	313.08-362.5	1-25	150	0.07	0.17	-0.07	0.59
Wappmann et al. [58]	1995	283.5-373.4	10-100	31	0.16	0.97	0.00	2.06

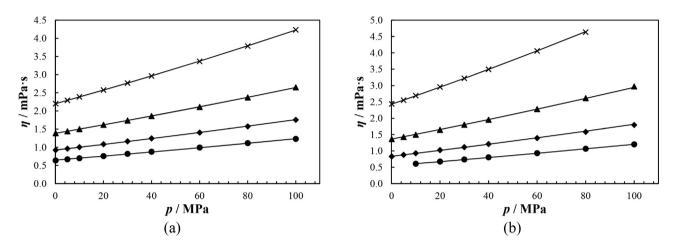


Fig. 7. Experimental viscosities as function of pressure for (a) 1-propanol and (b) 2-propanol at the isotherms: (×) 293.15 K; (▲) 313.15 K; (♦) 333.15 K and (●) 353.15 K. Lines represent the calculated values using modified VFT model with the parameters given in Table 11.

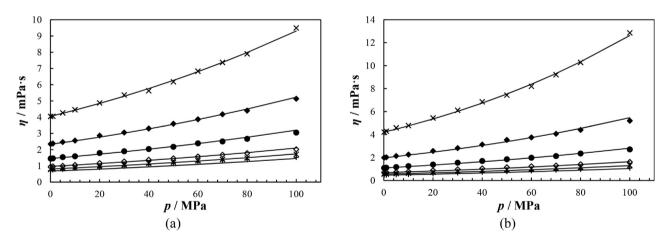


Fig. 8. Experimental viscosities as function of pressure for (a) 1-pentanol and (b) 2-pentanol at the isotherms: (×) 293.15 K; (◆) 313.15 K; (◆) 333.15 K; (◊) 353.15 K; (*) 363.15 K and (+) 373.15 K. Lines represent the calculated values using modified VFT model with the parameters given in Table 11.

For 1-pentanol and 2-pentanol, a direct comparison between our data and those found in the literature was performed through the average absolute deviation, instead of using the fitting equation due to the deviations obtained. Table 18 contains the results of the comparison including the temperature and pressure ranges of the literature data, the total number of the experimental points (NP) and the number of experimental points used for the comparison (CP). As can be seen, there is a limited amount of data at high pressure and the number of experimental points that can be compared is not significant. Most of the data at 0.1 MPa agree with our data within the uncertainty for 1-pentanol. In the case of 2pentanol, the comparison is limited to the data available at the same measuring conditions, and the average absolute deviation between data from Refs. [63,56] for three experimental points is 4.10% which confirms the dispersion of the data and the need of our new experimental values. In the case of the literature values given in Ref. [50], their experimental points are at different conditions than ours then this direct comparison was not possible.

As a summary, experimental density and viscosity measurements of four alcohols (1-propanol, 2-propanol, 1-pentanol and 2-pentanol) were obtained in a wide range of pressures (up to 100 MPa) and temperatures (most of them from 293.15 K to 373.15 K) using three different accurate experimental techniques. The experimental data were fitted to a modified Tammann-Tait equation for density as a function of pressure and temperature, obtaining very good results below the uncertainty of the equipment. In addition, the experimental viscosity data were successfully correlated using a modified VFT model for 1-propanol and 2-propanol.

Comparison between viscosity literature data and our correlation for 1-propanol. Statistical parameters: AAD (absolute average deviation), MD (absolute maximum deviation), Bias (average deviation) and RMS (root mean square). Temperature and pressure ranges and number of experimental points (NP) of the literature data are included.

Reference	Year	T/K	p/MPa	NP	AAD%	MD%	Bias%	RMS/mPa s
Pan et al. [22]	2019	298.15-313.15	0.1	4	1.39	2.61	1.39	0.024
Gharehzadeh et al. [23]	2019	293.15-323.15	0.1	7	0.81	1.09	-0.81	0.014
Ramos-Estrada et al. [24]	2018	293.15-343.15	0.1	11	3.27	4.57	3.27	0.041
Iglesias-Silva et al. [25]	2016	283.15-363.15	0.1	17	1.60	3.79	1.60	0.017
Lu et al. [26]	2015	293.15-323.15	0.1	7	1.35	1.75	1.35	0.023
Wang et al. [27]	2015	298.15-323.15	0.1	6	0.33	0.74	0.13	0.005
Hassein-bey-Larouci et al. [64]	2014	293.15-323.15	0.1	4	0.60	0.86	-0.60	0.010
Živković et al. [65]	2014	288.15-323.15	0.1	8	0.40	0.84	0.15	0.008
Pang et al. [28]	2007	293.15-333.15	0.1	9	0.81	1.95	0.81	0.011
Cano-Gómez et al. [29]	2012	293.15-328.15	0.1	8	2.38	3.35	2.38	0.034
Kurnia et al. [66]	2011	293.15-323.15	0.1	4	2.19	3.08	0.49	0.043
Ranjbar et al. [67]	2009	288.15-313.15	0.1	6	2.95	3.24	2.95	0.056
Kumagai et al. [68]	1998	273.15-333.15	0.1	4	1.10	2.19	1.10	0.043
Paez et al. [69]	1989	293.15-323.15	0.1	4	1.06	1.64	1.06	0.018
Papaioannou et al. [41]	1993	298.15	0.1-71.75	11	0.67	1.03	-0.44	0.017
Papaioannou et al. [70]	1995	298.15	0.1-59.8	12	0.55	0.96	-0.49	0.014
Baylaucq et al. [31]	2009	293.15-353.15	0.1-100	24	1.92	5.67	0.67	0.062

Table 17

Comparison between viscosity literature data and our correlation for 2-propanol. Statistical parameters: AAD (absolute average deviation), MD (absolute maximum deviation), Bias (average deviation) and RMS (root mean square). Temperature and pressure ranges and number of experimental points (NP) of the literature data are included.

Reference	Year	T/K	p/MPa	NP	AAD%	MD%	Bias%	RMS/mPa s
Pang et al. [28]	2007	293.15-333.15	0.1	9	1.67	3.24	1.67	0.021
Sovilj et al. [71]	1995	293.15-308.15	0.1	4	3.34	6.12	3.34	0.083
Yang et al. [44]	2006	293.15-343.15	0.1	7	4.50	7.65	4.50	0.053
Bhuiyan et al. [45]	2008	303.15-323.15	0.1	5	0.84	1.40	0.84	0.011
Kao et al. [72]	2011	293.15-323.15	0.1	4	2.77	3.88	2.77	0.046
Paez et al. [69]	1989	293.15-323.15	0.1	4	3.13	6.21	3.13	0.075
Hoga et al. [46]	2011	293.15-308.15	0.1	4	0.42	0.98	0.37	0.009
Contreras et al. [47]	2001	298.15-313.15	0.1	4	0.58	0.94	0.58	0.010
Sadeghi et al. [48]	2011	293.15-313.15	0.1	4	0.97	1.25	0.97	0.019
Mendo-Sánchez et al. [50]	2020	291.26-330.35	2-50	95	2.29	5.85	1.22	0.055
Moha-Ouchane et al. [53]	1998	303.15-343.15	0.1-100	18	4.66	7.21	4.66	0.101

Table 18

Comparison between viscosity literature data and our experimental data for 1-pentanol and 2-pentanol: the number of experimental points compared (CP) and average absolute deviation (AAD). Temperature and pressure ranges and number of experimental points (NP) of the literature data are included.

Reference	Year	T/K	p/MPa	NP	CP	AAD%
1-pentanol						
Iglesias-Silva et al. [25]	2016	283.15-363.15	0.1	17	5	0.70
Wang et al. [27]	2015	298.15-323.15	0.1	6	1	1.24
Cano-Gómez et al. [29]	2012	298.15-323.15	0.1	8	2	2.67
Weng et al. [73]	1999	303.15-3.23.15	0.1	3	1	0.63
Indraswati et al. [74]	2001	293.15-313.15	0.1	3	2	1.78
Al-Jimaz et al. [75]	2004	293.15-318.15	0.1	4	1	0.36
Estrada-Baltazar et al. [55]	2015	293.15-363.15	0.1	15	5	1.39
Vargas-Ibañez et al. [56]	2018	288.15-338.15	0.1	11	3	0.51
Pimentel-Rodas et al. [62]	2019	298.15-323.15	2-29.98	48	2	2.34
Sülzner et al. [57]	1997	298.15-373.15	50-120	9	2	7.74
2-pentanol						
González et al. [63]	2004	293.15-313.15	0.1	3	1	5.11
Vargas-Ibáñez et al. [56]	2018	288.15-338.15	0.1	11	3	1.91
Mendo-Sánchez et al. [50]	2020	291.26-330.35	2-50	95	0	_

CRediT authorship contribution statement

Declaration of Competing Interest

Alejandro Moreau: Investigation, Data curation, Writing – original draft. Manuel Sobrino: Investigation, Data curation. Johnny Zambrano: Investigation, Data curation. José J. Segovia: Conceptualization, Methodology, Formal analysis, Supervision. Miguel A. Villamañán: Conceptualization, Formal analysis, Supervision. M. Carmen Martín: Conceptualization, Supervision, Writing – review & editing, Project administration, Funding acquisition.

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.molliq.2021.117744.

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