J. Chem. Eng. Data **2003,** *48,* 1418−1421

Saturated Liquid Viscosity of Cyclopentane and Isopentane

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In this paper, the saturated liquid viscosity of cyclopentane and isopentane was measured through the capillary flow method, over the temperature range from 253.15 K to 353.15 K. The results were correlated as a function of temperature. The viscometer was calibrated using water, ethanol, acetone, and ethyl acetate.

Introduction

The development of natural working fluids with zero ozone depletion potential (ODP) and low global warming potential (GWP) is required for environment-friendly refrigerants and blowing agents. As a blowing agent, cyclopentane has been confirmed as a long-term alternative to R11. The mixture of isopentane and pentane was also considered as one of the potential alternatives of R11. Effective use of these natural working fluids requires that the thermodynamic and transport properties be accurately measured, but few data are available for the transport properties of cyclopentane and isopentane.

In this paper, the viscosity of these two natural working fluids was measured with a capillary viscometer at temperatures from 253.15 K up to 353.15 K along the saturation line. To reduce systematic uncertainty, the viscometer was calibrated using water, ethanol, acetone, and ethyl acetate. The results were correlated as a function of temperature.

Instrumentation

The instrument used in this experiment and previous work[1,2] is shown in Figure 1.

Three valves as shown in Figure 1 are necessary for the experiment. Before the experiment, a measured amount of liquid is injected into the pressure vessel through valve 3. During the experiment, valve 3 is always closed. Prior to each measurement, the liquid must be forced through the capillary to the upper volume above mark 2. When the height of the liquid surface in the capillary is raised, valve 1 is closed and valve 2 is open. The vapor in the capillary is connected with the atmosphere. When the saturation pressure in the pressure vessel is higher than 1 atm, the higher pressure in the pressure vessel can force the liquid into the capillary. When the saturation pressure in the pressure vessel is less than 1 atm, some instrument should be used to draw the liquid into the upper volume through the capillary. During each measurement, valve 2 is closed and valve 1 is opened. So the vapors in and out of the capillary are connected. The liquid can flow down owing to the effect of the weight. After the experiment, valve 3 is used to let out the measured liquid.

The viscometer was installed in a thermostat bath. The temperature of the bath can be varied from 243 K to 393 K. The temperature instability in the bath is less than ±5 mK in 1 h.

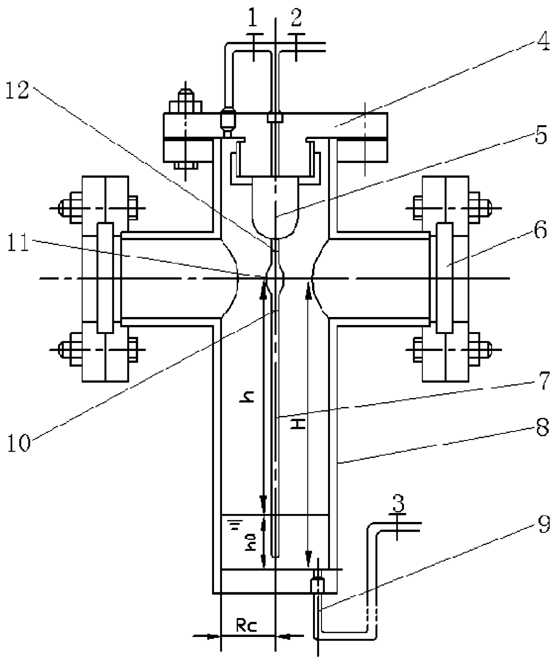

Figure 1. Instrument schematic: 1, valve 1; 2, valve 2; 3, valve 3; 4, flange plate; 5, liquid store; 6, observation window; 7, capillary; 8, pressure vessel; 9, liquid injection and discharging pipe; 10, mark 1; 11, liquid volume; 12, mark 2.

Operating Principles

The capillary viscometer is a classic method for measuring liquid viscosity. According to the Hagen−Poiseuille principle, the fluid volume flow through the capillary is related to the pressure difference between the two ends of the capillary, the radius and length of the capillary, and the viscosity of the fluid by

$$\frac{Q}{t} = \frac{\pi R^4 \Delta P}{8\eta L} \tag{1}$$

where Q is the volume of fluid flowing through the capillary for a given time t, R is the capillary radius, ΔP is the pressure difference between the two ends of the capillary, η is the fluid viscosity, and L is the capillary length. The length and the radius of the capillary are 120 mm and 0.177 mm in this work. The effect of surface tension can be ignored because it is very small for our instrument.[2] To include the kinetic energy correction and the vapor buoy-

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10.1021/je0202174 CCC: $25.00

ancy effect,[3] eq 1 can be rewritten in the following form:

$$\eta = \frac{\pi(\rho_L - \rho_V)ghR^4}{8QL}t - \frac{\xi\rho_L Q}{8\pi Lt} \tag{2}$$

where ρ_L is the saturated liquid density, ρ_V is the saturated vapor density of the sample, g is the acceleration of gravity, h is the height difference between the fluid surface in the cell and the fluid surface in the capillary, and ξ is the kinetic energy factor, $\xi = 0.037\mathrm{Re}^{0.5}$, where Re is the Reynolds number.[3] The vapor pressure and vapor density affect the flow of the liquid through the capillary in a sealed instrument, and the effect increases with temperature. The effective driving force is $(\rho_L - \rho_V)gh$ in eq 2, while $\rho_V gh$ presents the vapor buoyancy. Equation 2 can be rewritten in the following form:

$$\nu = kAt - B/t \tag{3}$$

where

$$k = \frac{\rho_L - \rho_V}{\rho_L} \tag{4}$$

$$A = \frac{\pi ghR^4}{8QL} \tag{5}$$

$$B = \frac{\xi Q}{8\pi L} \tag{6}$$

where ν is the kinematic viscosity. A and B are nearly temperature independent. k is a correction coefficient which relates to the densities of the liquid and vapor samples. It generally decreases as the temperature increases.

To ensure that the Reynolds number in the capillary is small (Re < 2000) and to keep the flow time sufficiently long to reduce measurement uncertainty, the capillary radius must be small. The Reynolds number is defined as

$$\mathrm{Re} = \frac{2Ru}{\nu} \tag{7}$$

and Q can be calculated as

$$Q = \pi R^2 ut \tag{8}$$

where u is the velocity of the fluid in the capillary. So eq 3 can be rearranged as

$$4ghkR^3 - \mathrm{Re}^2\nu^2\xi R - 16\mathrm{Re}\nu^2 L = 0 \tag{9}$$

From the numerical computation of eq 9, it is shown that the capillary radius R of the instrument must be less than 0.2 mm. A capillary with $R = 0.177$ mm was used in this work. The value of the radius was obtained by calibration with mercury.

According to eq 5, A is proportional to h, which is related to the volume of the test liquid. However, A cannot be treated as a constant because it varies with temperature and mass variations caused by vapor venting. Sun et al.[2] proposed a method to modify A at different temperatures by using the following equation:

$$A = \frac{A_1}{h_1}(H - h_0) = \frac{A_2}{h_2}(H - h_0) \tag{10}$$

where A_1 and A_2 are the calibration results using two different liquid volumes, V_1 (200 cm^3) and V_2 (250 cm^3); h_1 and h_2 can be obtained from V_1, V_2, and the radius of the pressure vessel R_c (see Figure 1) using

$$A_1/A_2 = h_1/h_2 \tag{11}$$

$$h_1 - h_2 = \frac{V_1 - V_2}{\pi R_c^{\,2}} \tag{12}$$

H is a constant of the instrument that can be determined from

$$H = h_1 + \frac{V_1}{\pi R_c^{\,2}} = h_2 + \frac{V_2}{\pi R_c^{\,2}} \tag{13}$$

h_0 is the liquid surface height in the cell that can be determined from

$$m = \rho_L V_L + \rho_V V_V \tag{14}$$

$$V = V_L + V_V \tag{15}$$

$$h_0 = \frac{V_L}{\pi R_c^{\,2}} \tag{16}$$

where m is the experimental fluid mass, V is the cell volume, V_L and V_V are the volumes of saturated liquid and saturated vapor, and ρ_L and ρ_V are the saturated liquid and saturated vapor densities.

During experiments, some vapor must be vented and the mass in the cell changes. Since the viscosity at a fixed temperature must be constant, the volume of vented vapor can be estimated by solving eqs 17−20 assuming that the vented volumes δV are constant.[3]

$$m_2 = m_1 - \sum_i^n \rho_i \delta V \tag{17}$$

$$\nu_1 = A_1 t_1 - B_1/t_1 \tag{18}$$

$$\nu_2 = A_2 t_2 - B_2/t_2 \tag{19}$$

$$\nu_1 = \nu_2 \tag{20}$$

where m_1 and m_2 are the mass of the fluid for the two measurements at the same temperature, n is the number of venting times between these two measurements, ρ_i is the fluid density, ν_1 and ν_2 are two measured viscosities of the fluid for the first time and the second time, t_1 and t_2 are two measured times for the first time and the second time, and A_1, B_1, A_2, and B_2 are the constants with m_1 and m_2, respectively.

The best experimental sequence is to begin with high temperatures and then reduce the temperature. But limited by the measurement apparatus,[3] the experimental temperature was increased from 293.15 K to 353.15 K. Later, the temperature was decreased from 293.15 K to 253.15 K in this work.

Calibration

To determine the values of A and B, the liquids water, ethanol, acetone, and ethyl acetate were used to calibrate the instrument. The samples were provided by Beijing Chemical Co., with a statement that the purity of liquid water is 18.1 MΩ·cm at 25 °C and the mass purities are 99.7%, 99.5%, and 99.5% for ethanol, acetone, and ethyl acetate, respectively. The samples were used without further purification. The calibrating volumes were 200 cm^3

Table 1. Calibration Results of the Instrument

V_l/cm^3	H/cm	h/cm	Q/cm^3	A/m^2·s^{-2}	B/m^2
200	16.738	11.069	2.772	1.273×10^{-9}	1.850×10^{-7}
250	16.738	9.652	2.772	1.110×10^{-9}	1.889×10^{-7}

Table 2. Verification of the Instrument Calibration Results at 297.15 K

V_l cm^3	liquid	ν_{ref} mm^2·s^{-1}	ν_{exp} mm^2·s^{-1}	$100(\nu_{exp} - \nu_{ref})/\nu_{ref}$
200	water	0.9161	0.9258	1.07
	ethanol	1.4311	1.4293	−0.13
	acetone	0.3989	0.3976	−0.32
	ethyl acetate	0.4815	0.4810	−0.10
250	water	0.9161	0.9203	0.46
	ethanol	1.4311	1.4407	0.67
	acetone	0.3989	0.3934	−1.18
	ethyl acetate	0.4815	0.4818	0.06

and 250 cm^3, and the calibrating temperature was 297.15 K. From calibration with these nonrelative liquids, the value of k can be set to 1.0.

The constant B embodies the kinetic energy correction. As the viscosity increases, the flow time will also increase, so that the second term in eq 3 will become much smaller than the first term. For the calibrating liquids with high viscosities, which makes ϵ (defined by eq 21) less than 0.1%, the effect of kinetic energy can be ignored. As discussed before, A can be obtained from eq 3. With the kinetic energy factor ξ, eq 5 and eq 6, B can be calculated as follows:

$$\epsilon = \frac{B/t}{kAt} \tag{21}$$

The calibration results are listed in Table 1. The verification of the calibration results is listed in Table 2. The reference data of viscosities were cited from Beaton and Hewitt.[4]

Results and Analysis

The viscosities of cyclopentane and isopentane were measured along the saturation line. The samples of cyclopentane and isopentane were provided by Beijing Eastern Acrylic Chemical Science and Technology Ltd. The mass purity of the cyclopentane sample is 99.79%, with the following impurities: butane, 0.01%; isopentane, 0.07%; pentane, 0.13%. The mass purity of the isopentane sample is 99.42%, with the following impurities: butane, 0.14%; pentane, 0.15%; cyclopentane, 0.3%. The mass purity data for the cyclopentane and isopentane samples are obtained from the chromatographic detection report by the provider. These samples were used without further purification. The uncertainty of the temperature measurements was within ±10 mK, and the thermal gradient of the bath used in the experiment was less than ±5 mK. The temperature range was 253.15 K to 353.15 K. For cyclopentane, the liquid densities were provided by Beaton[4] when the temperature was larger than 323.15 K; the others were from McLinden.[5] The vapor densities of cyclopentane were calculated with the Virial equation from ref 6. For isopentane, the liquid densities and vapor densities were provided by Beaton[4] when the temperature was larger than 303.15 K; the others were from McLinden.[5]

The experimental results were correlated as a function of temperature as the following equation:

$$\log(\eta/\mathrm{Pa{\cdot}s}) = D + E/(T/\mathrm{K}) + F(T/\mathrm{K}) + G(T/\mathrm{K})^2 \tag{22}$$

where the coefficients D, E, F, and G were determined by fit. For cyclopentane, $D = 2.1864$, $E = -2.8217 \times 10^2$, $F = -2.2977 \times 10^{-2}$, and $G = 2.5054 \times 10^{-5}$. For isopentane, $D = 1.5372$, $E = -3.0600 \times 10^2$, $F = -1.9453 \times 10^{-2}$, and $G = 1.8493 \times 10^{-5}$. The temperature range of the correlation eq 22 is from 253.15 K to 353.15 K.

Table 3. Saturated Liquid Viscosity of Cyclopentane

T K	ρ_l kg·m^{-3} [a]	ρ_v kg·m^{-3} [b]	η_{exp} mPa·s	$100(\eta_{exp} - \eta_{cal})/\eta_{cal}$
253.15	784.64	0.0881	0.7268	0.18
258.15	779.53	0.1127	0.6786	0.05
263.15	774.59	0.1443	0.6347	0.01
268.15	769.77	0.1848	0.5930	−0.27
273.15	765.12	0.2368	0.5567	−0.17
278.15	760.20	0.3036	0.5224	−0.26
283.15	755.32	0.3894	0.4922	−0.08
288.15	750.27	0.4999	0.4646	0.15
293.15	745.02	0.6421	0.4382	0.14
298.15	738.63	0.8255	0.4148	0.31
303.15	731.97	1.062	0.3923	0.27
308.15	725.15	1.368	0.3714	0.14
313.15	718.32	1.764	0.3521	−0.03
318.15	711.59	2.279	0.3350	−0.04
323.15	705.11	2.950	0.3190	−0.15
328.15	699.08	3.827	0.3048	−0.07
333.15	693.40	4.980	0.2912	−0.25
338.15	688.44	6.509	0.2793	−0.19
343.15	684.25	8.554	0.2690	0.04
348.15	680.96	11.33	0.2590	0.06
353.15	678.71	15.20	0.2502	0.18

[a] References 4 and 5. [b] Reference 6.

Table 4. Saturated Liquid Viscosity of Isopentane

T K	ρ_l kg·m^{-3} [a]	ρ_v kg·m^{-3} [a]	η_{exp} mPa·s	$100(\eta_{exp} - \eta_{cal})/\eta_{cal}$
253.15	658.32	0.4655	0.3893	−0.36
258.15	653.55	0.5889	0.3661	0.04
263.15	648.73	0.7372	0.3439	0.25
268.15	643.87	0.9137	0.3201	−0.11
273.15	639.01	1.122	0.3023	0.43
278.15	634.15	1.366	0.2859	0.01
283.15	629.35	1.650	0.2703	0.21
288.15	624.63	1.979	0.2547	0.31
293.15	620.05	2.356	0.2399	−0.10
298.15	615.69	2.788	0.2289	−0.37
303.15	610.87	3.278	0.2144	−0.12
308.15	605.63	3.833	0.2023	−0.30
313.15	600.05	4.459	0.1910	−0.25
318.15	594.23	5.162	0.1813	−0.58
323.15	588.24	5.949	0.1724	−0.08
328.15	582.18	6.827	0.1611	0.50
333.15	576.13	7.803	0.1543	−0.37
338.15	570.18	8.886	0.1480	0.49
343.15	564.41	10.09	0.1411	1.05
348.15	558.92	11.41	0.1332	0.87
353.15	553.79	12.87	0.1287	−1.51

[a] References 4 and 5.

The results of measurements and the relative deviations of experimental data from the correlation eq 22 are shown in Table 3 and Table 4. The standard deviation and the maximum relative deviation of the experimentally measured results from the correlation eq 22 for cyclopentane are 0.14% and 0.31%. The standard deviation and the maximum relative deviation of the experimentally measured results from the correlation eq 22 for isopentane are 0.53% and 1.22%.

Figure 2 shows deviations between the experimental data from Assael and Dalaouti,[7] TRC,[9] and eq 22 for cyclopentane. The experimental data of Assael and Dalaouti[7] and TRC[9] are the viscosity of cyclopentane at atmospheric pressure, which is close to the results of saturated

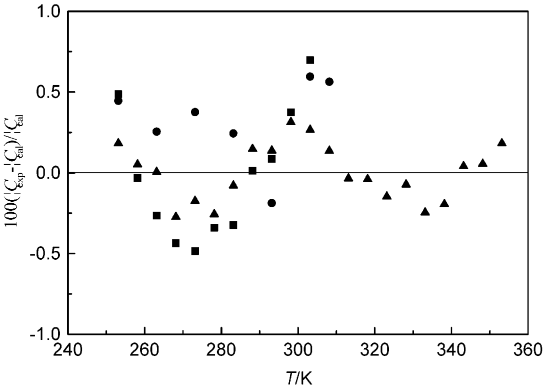

Figure 2. The relative deviations of experimental data from the correlation eq 22 (cyclopentane): ▲, this work; ●, Assael and Dalaouti (ref 7); ■, TRC (ref 9).

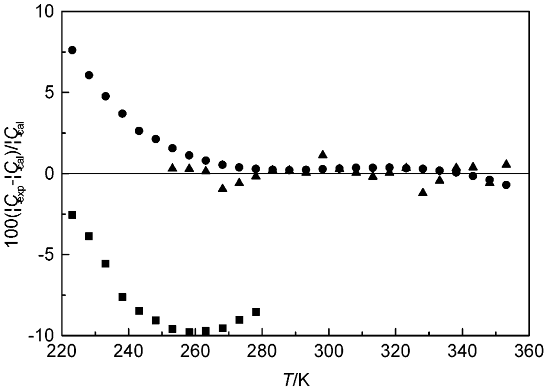

Figure 3. The relative deviations of the data from the correlation eq 22 (isopentane): ▲, this work; ●, Huber et al. (ref 8); ■, TRC (ref 9).

liquid. The maximum relative deviation of the above data is less than ±1%.

Figure 3 shows the deviations between the data from Huber et al.,[8] TRC,[9] and eq 22 for isopentane. The maximum relative deviations of the estimated viscosities from Huber et al.[8] shown in Figure 3 are less than ±2% in the temperature range of the correlation eq 22, but the maximum relative deviations are positive and less than ±8% from 223.15 K to 253.15 K. The data of TRC[9] from 223.15 K to 278.15 K are the viscosity of isopentane at atmospheric pressure. The maximum relative deviations of the viscosities from TRC[9] shown in Figure 3 are negative and less than ±10%.

In this work, the venting volumes were about 4 cm^3 under the ambient temperature. Duplicate measurements were made at each temperature to avoid occasional error. According to numerical computation, the maximum uncertainty caused by vapor venting approaches 1%. So the corrections are necessary in this work.

Conclusion

A method was used for measuring the viscosity of saturated liquids with a capillary viscometer. The viscosities of cyclopentane and isopentane along the saturation line were measured over the temperature range from 253.15 K to 353.15 K. The uncertainty of the results is estimated to be less than ±2% for cyclopentane. The experimental data of isopentane have some deviations with other data.

Acknowledgment

We acknowledge Dr. Wang Xin for his assistance to the work.

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Received for review December 2, 2002. Accepted August 11, 2003. This project was supported by the National Natural Science Foundation of China (Grant No. 59706012).

JE0202174