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Surface tension and liquid viscosity measurement for binary mixtures of R134a with R1234yf and R1234ze(E)

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ABSTRACT

The surface tension and liquid kinematic viscosity of two binary refrigerant mixtures 1,1,1,2-tetrafluoroethane (R134a) (1) +2,3,3,3-tetrafluoroprop-1-ene (R1234yf) (2) and R134a (1) + trans-1,3,3,3-tetrafluoroprop-1-ene (R1234ze(E)) (2) were measured in the temperature range from 293 K up to the liquid–vapor critical point with the surface light scattering (SLS) method. The experimental data were correlated as a function of temperature and mole fraction of the pure components. For surface tension, the average absolute deviations are 0.019 $mN \cdot m^{-1}$ for R134a + R1234yf and R134a + R1234ze(E). For liquid viscosity, the average absolute deviations are 0.96% and 1.16% for R134a + R1234yf and R134a + R1234ze(E), respectively.

1. Introduction

Recently, 2,3,3,3-Tetrafluoroprop-1-ene (R1234yf, $CF_3CF{=}CH_2$,754-121-1), with zero ODP and low GWP (100 year GWP = 4) [1], has been proposed as a drop-in replacement for 1,1,1,2-tetrafluoroethane (R134a) [2,3]. Trans-1, 3, 3,3-Tetrafluoroprop-1-ene (R1234ze(E), $CHF{=}CHCF_3$, 29118-24-9) is also a promising candidate with low GWP (100-year GWP = 6) and could be mixed with other refrigerants. However, their main drawbacks, flammability and minor cooling capacity could hinder their application. In order to overcome these limitations related to R1234yf and R1234ze(E), the binary mixtures of R1234yf and R1234ze(E) with R134a have been developed [4]. Lee et al. [5] found R134a + R1234yf mixture with 10–11% R134a is virtually non-flammable and azeotropic. Its coefficient of performance (COP), capacity and discharge temperature are similar to those of R134a. R450A (42% R134a + 58% R1234ze(E) in mass fraction), with non-flammability, a zero ODP and a GWP of 547, was developed by Honeywell as a replacement of R134a. Additionally, R450A was listed in the Significant New Alternatives Policy (SNAP) program by America Environmental Protection Agency. Mota-Babiloni [6] carried out an experiment in a vapor compression plant and proved that R450A was a good candidate to replace R134a.

Thermophysical properties of the refrigerant are basic data and used for designing condenser and evaporator in a refrigerator. Raabe et al. [7] conducted an experiment on the vapor–liquid equilibrium (VLE) property of blend R134a + R1234ze(E) at a temperature range from 273 K to 330 K by molecular simulation method. Kamiaka et al. [8] performed the VLE properties of the binary mixture R134a + R1234yf at mass fractions of R1234yf from 25% to 80% over the temperature range from 273 K to 333 K. Chen et al. [9] measured the PVT*x* properties in the gas phase for binary mixture R134a + R1234yf in the range of temperature from 298.58 K to 403.24 K, pressure range from 567.5 to 3171.2 kPa. Surface tension and viscosity are two important thermophysical properties influencing the heat transfer, flow and phase change characteristic of the refrigerant. Unfortunately, there are no surface tension and viscosity data of mixture refrigerants R134a + R1234yf and R134a + R1234ze(E) in the literature.

In this paper, the surface tension and liquid kinematic viscosity of binary mixtures of R134a + R1234yf and R134a + R1234ze(E) were conducted by surface light scattering (SLS) method. The experimental surface tension and viscosity data were correlated as a function of temperature and mole fraction of the pure components.

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2. Experimental section

2.1. Material

R134a was manufactured by Sinochem Modern Environmental Protection Chemicals (Xi'an) Co., Ltd., China with a declared mass purity of 0.999. R1234yf and R1234z were supplied by Honeywell with declared mass purity of 0.999. The complete specifications for the three refrigerants are given in Table 1.

In this work, the pure refrigerants were firstly purified by freeze-pump-thaw cycles, and then the mixtures were prepared by the following process. Firstly, a known quality of R1234yf or R1234ze(E) with a lower saturation pressure was introduced into the cell by heating the cylinder with a hot-air generator for at least 20 min. Secondly, R134a with a higher saturation pressure was introduced into the cell by the same way. The mass of the residual refrigerant in the connection pipe was less than 0.1 g. The weights of refrigerants and cylinders were precisely measured by a balance (METTLER TOLEDO ME3002) with a resolution of 0.01 g. Since the total mass of the refrigerants in the experimental cell was about 75 g, the uncertainty of the composition of the mixture was estimated to be better than 0.2% in mole fraction. Finally, the R134a (1) + R1234yf (2) mixtures were prepared with mole factions x_1 of 0.3196, 0.6018 and 0.8070. R134a (1) + R1234ze(E) (2) mixtures were prepared with mole faction x_1 of 0.4446.

2.2. Surface light scattering

In the SLS technique, fluctuations are analyzed on the surface of a liquid or, in general, on the boundary layers between two different phases. For the case of low viscosity, as is relevant in the present experiment of a binary refrigerant mixture, the amplitude of the surface fluctuations decreases with time as a damped oscillation. The surface tension σ and kinematic viscosity of the liquid phase $\nu'(=\eta'/\rho')$ under saturation conditions were determined by means of an exact numerical solution of the dispersion relation for surface waves. A more detailed description of the SLS method can be found in Refs. [10–13]. This method has already been successfully applied to the surface tension and viscosity measurement for refrigerants, ionic liquids and so on [14–19].

The SLS apparatus is already employed in our former investigation for the surface tension and viscosity measurement of the pure R1234yf and R1234ze(E) [18]. The experimental system includes a diode-pumped solid state laser (Spectra-Physics Excelsior, 300 mW) with a wavelength of $\lambda_0 = 532$ nm, a digital correlator with a single-tau structure (ALV-LinCorr) for the computation of the pseudo cross-correlation function, optical path and sample cell (stainless steel and equipped with quartz windows; Inner diameter: 70 mm; Volume: 160 cm^3). The temperature of the sample cell is regulated through resistance heating and measured by two calibrated 100 Ω platinum resistance probes with a total uncertainty of ±0.03 K. A more detailed description of the SLS apparatus can be found in Ref. [18].

3. Results and discussions

3.1. Results

The surface tension and liquid kinematic viscosity of the mixtures of R134a (1) + R1234yf (2) at 3 mol fractions x_1 of 0.3196, 0.6018 and 0.8070 were measured over the temperature range from 293 K to 363 K, and the experimental data were showed in Table 2. The surface tension and liquid kinematic viscosity of the mixtures of R134a (1) + R1234ze(E) (2) with x_1 of 0.4446 were measured over the temperature range from 293 K to 369 K, and the experimental data were showed in Table 3. The saturated liquid, vapor densities and vapor viscosity of mixtures were calculated from the NIST refprop9.0 [20]. For the non-azeotropic binary mixtures, the liquid and vapor component varies at different temperature. However, Kamiaka et al. [8] conducted a research on VLE properties for binary mixtures of R1234yf with R134a. The results showed that, the liquid fraction keeps unchanged at temperature from 273 K to 333 K, Thus, the liquid mole fractions of each pure component were assumed as a constant at the whole temperature range.

Table 2
Liquid Density ρ', Vapor Density ρ'', Dynamic Viscosity of the Vapor Phase η'', Kinematic Viscosity of the Liquid Phase ν', and Surface Tension σ of R134a (1) + R1234yf (2) with mole factions x_1 of 0.3196, 0.6018 and 0.8070 of R134a under Saturation Conditions[a] from (293–363) K by SLS.

T/K	ρ' [b]/kg·m^{-3}	ρ'' [b]/kg·m^{-3}	η'' [b]/μPa·s	ν'/mm^2·s^{-1}	σ/mN·m^{-1}
$x_1 = 0.3196$					
293.22	1142.5	31.2	11.19	0.1490	7.38
303.21	1105.3	41.9	11.57	0.1347	6.14
313.12	1065.8	55.3	11.99	0.1249	4.93
323.10	1022.2	72.7	12.62	0.1150	3.95
333.16	972.9	95.8	13.44	0.1066	2.87
343.19	915.5	126.9	14.57	0.0977	1.88
353.16	843.7	172.0	16.32	0.0870	1.00
358.15	798.1	204.0	17.67	0.0836	0.60
$x_1 = 0.6018$					
293.20	1174.3	29.9	11.36	0.1612	7.80
303.13	1137.2	40.0	11.74	0.1425	6.59
313.15	1096.9	53.2	12.13	0.1294	5.41
323.16	1052.9	70.1	12.71	0.1221	4.24
333.12	1004.0	92.2	13.46	0.1122	3.15
343.16	946.8	122.2	14.48	0.1001	2.11
353.19	876.6	164.9	16.03	0.0890	1.17
363.19	778.5	235.1	18.90	0.0795	0.37
$x_1 = 0.8070$					
293.15	1199.7	28.77	11.50	0.1617	8.25
303.15	1162.2	38.75	11.89	0.1470	6.99
313.14	1121.7	51.58	12.27	0.1370	5.76
323.15	1077.5	68.13	12.80	0.1258	4.55
333.15	1028.2	89.75	13.51	0.1133	3.41
343.14	971.5	118.73	14.46	0.1040	2.39
353.10	903.0	159.43	15.87	0.0922	1.31
363.16	808.9	225.11	18.39	0.0821	0.53

[a] Directly measured values for frequency and damping at a defined wave vector of surface waves were combined with literature data for η'', ρ' and ρ'' from Ref. [20] to derive ν' and σ by an exact numerical solution of the dispersion relation. The combined expanded uncertainties U_c are U_c (T) = 0.03 K, U_c (x) = 0.002. x, U_c (ν') = 0.02. ν' for $T_r < 0.95$ and 0.06. ν' for T_r close to 0.99, and U_c (σ) = 0.015. σ (level of confidence = 0.95).
[b] Calculated from NIST REFPROP9.0.[20].

Table 1
Specifications of chemical samples.

Chemical	Source	Purity (mass fraction)	Purification method
R134a	Sinochem Modern Environmental Protection Chemicals Co., Ltd., China	0.999	freeze-pump-thaw
R1234yf	Honeywell International Inc.	0.999	freeze-pump-thaw
R1234ze(E)	Honeywell International Inc.	0.999	freeze-pump-thaw