SURFACE TENSION OF LOW GWP REFRIGERANTS R1234ZE(Z) AND R1233zd(E)

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ABSTRACT

R1234ze(Z) and R1233zd(E) are very recently under consideration as a potential candidate to replace R245fa that is widely used in organic Rankine cycles and high-temperature industrial heat pumps. The capillary constant and surface tension of saturated R1234ze(Z) and R1233zd(E) were measured across the temperature range from 274 K to 350 K using a measuring apparatus based on the differential capillary rise method. The measurement uncertainty in the surface tension was typically 0.3 mN m^{-1}. The experimental surface tension was well represented by a van der Waals type equation expressing the temperature dependence as, $\sigma = 59.90$ $(1 - T_r)^{1.237}$ for R245fa , $\sigma = 56.57 (1 - T_r)^{1.220}$ for R1234ze(Z), and $\sigma = 61.95 (1 - T_r)^{1.277}$ for R1233zd(E), where, σ and T_r are the surface tension in mNm^{-1} and the reduced temperature, $T_r = T /T_{crit}$. The critical temperatures T_{crit} are given as 427.01 K for R245fa, 423.27 K for R1234ze(Z) by Higashi et al. (2015), and 438.75 K for R1233zd(E) by Hulse et al. (2012). The constants and exponents are empirically determined by the least square mean method. The above correlations represent the experimental surface tension within ±0.13, ±0.21 mN m^{-1}, and ±0.05 mN m^{-1} for R245fa, R1234ze(Z), and R1233zd(E), respectively.

1. INTRODUCTION

Ozone depletion and global warming caused by refrigerants are currently a worldwide concern. Brown et al. (2010) compared the estimated thermodynamic properties for eight hydro-fluoro-olefins (HFOs). Among these substances, the GWP$_{100}$ of R1225ye(E), R1234yf, R1234ze(E), R1234ze(Z) and R1243zf were reported as less than 1 in IPCC 5AR (Myhre et al., 2013). The newly developed blowing agent R1233zd(E) was recently suggested as the refrigerant or working fluid (Hulse et al., 2012) alternatives to R245fa that is widely used in organic Rankine cycles. On the basis of the second law analysis, Cavallini et al. (2014) evaluated the performance potential of hydro-fluoro-olefins (HFOs), R1234yf, R1234ze(E), and R1234ze(Z). Meanwhile, very comprehensive research has been conducted by McLinden et al. (2014). A set of 1200 low GWP refrigerants with critical temperatures between 300 K and 400 K was assessed, considering flammability, thermal stability, and toxicity for air conditioners. Nevertheless, the debate on the final candidate of low GWP refrigerant from a long-term prospective is still controversial.

The physical property data are important to build models to evaluate the efficiency and capacity of air conditioning and heat pump systems in which the candidate refrigerants are used. The surface tension is the one of dominant parameters that affects the heat transfer coefficient during condensation and evaporation. For R1234ze(E) and R1234yf, the surface tension data have been provided by Tanaka and Higashi (2013), Takahashi et al. (2009), and Zhao et al. (2014). For R1233zd(E), only three points of surface tension data are reported by Hulse et al. (2012). The measurement data of R1234ze(Z) are not yet available in the open literature. Surface tension measurements were thus made for R1234ze(Z) and R1233zd(E) in this study.
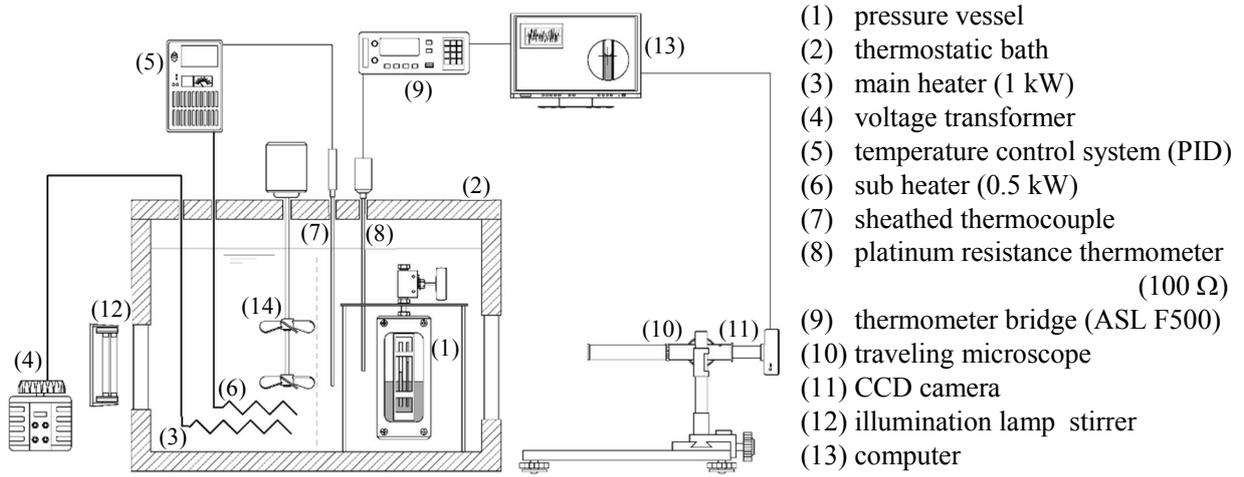
(1) pressure vessel
(2) thermostatic bath
(3) main heater (1 kW)
(4) voltage transformer
(5) temperature control system (PID)
(6) sub heater (0.5 kW)
(7) sheathed thermocouple
(8) platinum resistance thermometer (100 Ω)
(9) thermometer bridge (ASL F500)
(10) traveling microscope
(11) CCD camera
(12) illumination lamp stirrer
(13) computer

Figure 1. Experimental setup.

2.   MEASUREMENT

2.1 Measurement procedure and data reduction

Figure 1 shows the experimental apparatus for measuring the surface tension. The surface tension is measured as the capillary elevation in a small diameter tube immersed in a liquid. Two capillaries with inner radii of r_1 = 0.4222 ±0.0009 mm and r_2 = 0.7526 ±0.0009 mm are vertically set by a supporting brace in a pressure vessel (1) composed of a Pyrex glass tube that is 17 mm in inner diameter and 25 mm in outer diameter. The capillary radii were precisely measured with mercury slugs. These capillaries and pressure vessel are carefully cleansed by using an alkaline aqueous solution and an ultrasound bath in the preparatory procedure. The liquid of the test refrigerant is filled roughly to a half volume of the pressure vessels at room temperature. Then, the pressure vessel is placed in a thermostatic bath (2), and the temperature in the thermostatic bath is kept constant with a fluctuation within ±2 mK using a main heater (3) and a PID (5) controlled sub heater (6). The temperature is measured with a 100 Ω platinum resistance thermometer (8), and a resistance bridge ASL model F500 (9), calibrated against ITS-90. The uncertainty of the temperature measurement is estimated to be within ±5 mK. At steady state, the capillary rise difference between the two capillary tubes is measured using a digital traveling microscope (10) with a tolerance of 0.01 mm.

Figure 2 illustrates the differential capillary-rise-height method. Specifically, with the traveling microscope and a CCD camera (11), the height difference of the bottom of the meniscus in each capillary tube, Δh_m as shown in Figure 2, is measured. The differential height is read by six people to eliminate the individual reading variation. The two standard deviations of 12 times readings was 0.046 mm. This deviation is taken into account as the reading error in the differential height measurement. To obtain the actual differential capillary rise height, the measured differential height Δh_m at the bottom of the meniscus in each capillary tube has to be corrected by a methodology of Rayleigh (1916).

$$\Delta h_c = (h_{m1} - h_{m2}) + \frac{(r_1 - r_2)}{3} - 0.1288\left(\frac{r_1^2}{h_{m1}} - \frac{r_2^2}{h_{m2}}\right) + 0.1312\left(\frac{r_1^3}{h_{m1}^2} - \frac{r_2^3}{h_{m2}^2}\right) + \dots \approx \Delta h_m + \frac{(r_1 - r_2)}{3} \tag{1}$$

The capillary constant a^2 is determined only from the capillary radii r_1 and r_2, and the differential capillary-rise-height Δh_c,

$$a^2 = \frac{g \Delta h_c}{g_n (1/r_1 - 1/r_2)\cos\theta} \approx \frac{g \Delta h_c}{g_n (1/r_1 - 1/r_2)} \tag{2}$$

where g and g_n are the local gravitational acceleration 9.8001 m s^{-2} at Iwaki, Japan and the normal gravitational accelerations 9.80665 m s^{-2}, respectively. The contact angle θ of refrigerants in the capillary tube, as shown in Figure 2, is assumed zero. Hence, $\cos\theta$ is nearly equal to 1. The capillary constant is rewritten with the surface tension σ as (Heide, 1997),

$$a^2 = \frac{2\sigma}{g_n (\rho' - \rho'')} \tag{3}$$
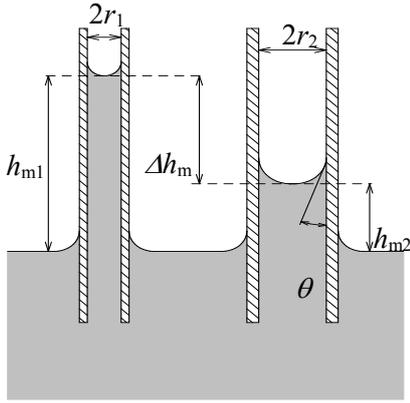
Figure 2. Capillary-rise-height

Table 1 Measurement uncertainties

measured parameter	equipment	uncertainty
temperature (ITS-90)	Platinum resistance thermometer and thermometer bridge, ASL F500	±5 mK
differential capillary-rise-height, Δh_m	digital traveling microscope, Nippon optical works Co., Ltd., NRM-D-2XZ	±0.046 mm (tolerance; 0.01 mm)
inner radii of the capillaries, r_1 and r_2	-	±0.0009 mm
orthobaric densities, ρ' and ρ''	-	±0.04% of the calculated values

$$\therefore \quad \sigma = \frac{g\Delta h_c(\rho'-\rho'')}{2(1/r_1-1/r_2)\cos\theta} \approx \frac{g\Delta h_c(\rho'-\rho'')}{2(1/r_1-1/r_2)} = \left[\frac{gr_1r_2}{2(r_2-r_1)}\Delta h_m - \frac{gr_1r_2}{6}\right](\rho'-\rho'') \tag{4}$$

The orthobaric densities, ρ' and ρ'', are calculated from measured temperature with REFPROP 9.1 (Lemmon *et al.*, 2013) for R245fa and R1233zd(E). For R1234ze(Z), a fluid file optimized by Akasaka *et al.* (2015) from the measurement data provided by Higashi *et al.* (2015) were used. The tested R1234ze(Z) and R1233zd(E) were supplied from Central Glass Co., Ltd., Japan. Preliminarily, they were well distilled, and the impurities were checked by a TCD gas chromatograph. These impurities were less than 0.1% by mole.

Table 1 lists the measurement uncertainties of the equipment mentioned above. The propagated uncertainty in surface tension is calculated by means of the square-root rule using the listed uncertainties. The reading error in the capillary rise height was most dominant in the propagated uncertainty. Typically, the propagated uncertainty is estimated to be within ±0.2 mN m^{-1}.

2.2 Validity assessment for the measurement method

The reproducibility of the above mentioned measurement methods were confirmed with R134a and R245fa. Figures 3 (a) and 3 (b) compare the surface tension between the present data and other data in literature for R134a and R245fa, respectively. For R134a, the measured surface tension provided by Chae *et al.* (1990), Heide (1997), Higashi *et al.* (1992), Higashi *et al.* (1997), and Zhu *et al.* (1993), and also the calculated surface tension by REFPROP 9.1 were compared in Figure 3 (a). The standard deviation of the present R134a data from the calculated surface tension was ±0.13 mN m^{-1}, which is within the uncertainty ±0.2 mN m^{-1}. For R245fa, Higashi *et al.* (2015) updated the critical parameters and some other thermodynamic properties, therefore the comparison with the calculated data was avoided. Instead, the data provided by Schmidt *et al.* (1996), Lin *et al.* (2003), Zhelezny *et al.* (2007), and Geller *et al.* (1999) were compared in Figure 3 (b). Although the data by Geller *et al.* (1999) deviate from the other data, the present surface tension data agree well with the most of reference data. As remarked here, it was confirmed that the measurement method is sufficiently reproducible.

3. RESULTS AND DISCUSSION

3.1. Measurement results

Table 2 provides the measured capillary constants a^2 given by Eq. (2) and the surface tensions σ given by Eq. (4) for R245fa, R1234ze(Z), and R1233zd(E). The measured temperature T, calculated densities ρ' and ρ'', corrected capillary rise height Δh_c are listed, as well. Figures 4 (a) and 4 (b) plot the measured surface tension as a function of temperature for R1234ze(Z) and R1233zd(E), respectively. The circle symbols indicate the present data; the small vertical bars overlapped with the symbols indicate the measurement uncertainty in surface tension. The diamond symbols are the critical temperatures T_{crit}, which are measured by Higashi *et al.* (2015) for R1234ze(Z) and by Hulse *et al.* (2012) for R1233zd(E). The triangle symbols in Figure 4 (b) are data for R1233zd(E) surface tension measured by Hulse *et al.* (2012). As seen in Figure 4 (b), the present data exhibits an approximately 1.5 mN m^{-1} greater value than their data at a given temperature. For R1234ze(Z), no available measurement data could be found.
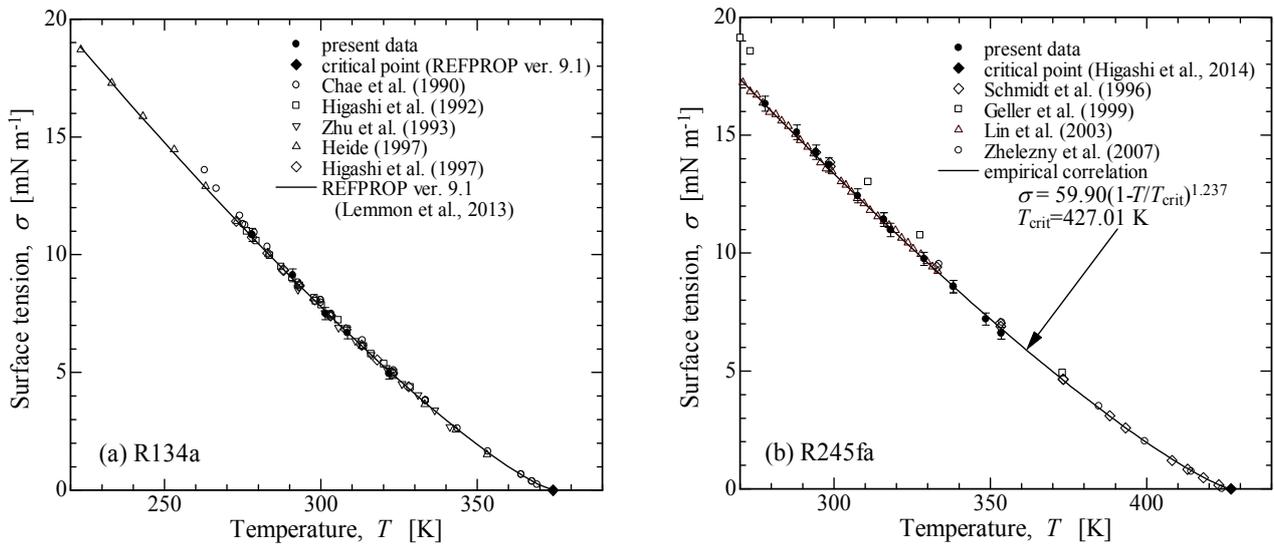
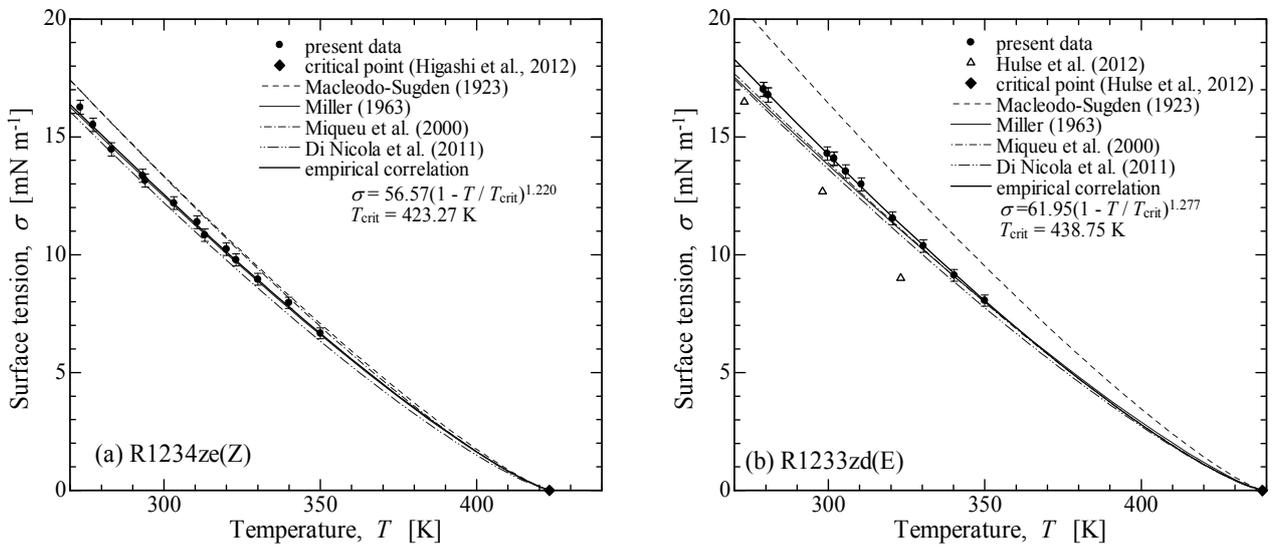Figure 3. Comparison between present data and existing data in literature.

Figure 4. Temperature dependence of low GWP refrigerants, R1234ze(Z) and R1233zd(E).

Table 2 Measured capillary constant and surface tension.

T	ρ'	ρ''	Δh_c	a^2	σ
K	kg m^{-3}	kg m^{-3}	mm	mm^2	mN m^{-1}
R134a					
278.107	1278.21	17.11	1.824	1.75	10.84
290.997	1233.18	25.98	1.605	1.54	9.13
301.449	1194.09	35.70	1.374	1.32	7.50
308.490	1166.12	43.84	1.263	1.22	6.68
321.750	1108.79	63.74	1.006	0.97	4.95
R245fa					
294.308	1348.91	7.45	2.258	2.28	14.28
278.025	1391.56	3.97	2.497	2.51	16.33
288.137	1365.31	5.93	2.360	2.38	15.12
298.354	1337.99	8.61	2.193	2.22	13.74
307.578	1312.53	11.79	2.027	2.06	12.43
318.120	1282.32	16.50	1.841	1.88	10.98
328.848	1250.14	22.71	1.687	1.73	9.76
348.592	1186.04	39.08	1.333	1.39	7.20
315.924	1288.72	15.41	1.903	1.94	11.42
338.220	1220.60	29.58	1.528	1.58	8.58
353.491	1168.90	44.40	1.247	1.31	6.61
R1234ze(Z)					
294.00	1231.44	7.67	2.28	2.19	13.15

T	ρ'	ρ''	Δh_c	a^2	σ
310.61	1187.64	13.09	2.06	1.98	11.38
319.97	1161.56	17.29	1.90	1.83	10.23
330.01	1132.27	22.96	1.71	1.65	8.95
339.94	1101.73	30.01	1.58	1.52	7.96
350.03	1068.67	38.99	1.37	1.32	6.66
273.22	1282.53	3.58	2.70	2.60	16.26
277.34	1272.71	4.20	2.59	2.50	15.51
283.24	1258.37	5.24	2.45	2.46	14.47
293.20	1233.48	7.46	2.31	2.33	13.35
303.19	1207.58	10.38	2.16	2.18	12.19
313.05	1180.96	14.09	1.97	2.00	10.83
323.05	1152.75	18.89	1.83	1.87	9.78
R1233zd(E)					
299.64	1259.20	7.56	2.42	2.33	14.29
310.54	1232.22	10.75	2.26	2.17	12.99
320.56	1206.51	14.53	2.05	1.98	11.54
330.37	1180.36	19.19	1.90	1.82	10.38
340.23	1152.92	25.01	1.72	1.65	9.13
349.98	1124.41	32.11	1.56	1.51	8.05
305.58	1244.61	9.19	2.32	2.24	13.53
280.72	1303.97	3.82	2.74	2.64	16.78
301.79	1253.94	8.12	2.40	2.31	14.08
279.26	1307.32	3.61	2.77	2.67	17.02

3.2 Comparison with estimation methods

The present measurement data are compared with the estimation methods below with the units $N\,m^{-2}$ and MPa.

i) Estimation based on the parachor, Macleodo(1923) and Sugden (1924) with modification by Fishtine (1963):

$$\sigma = ([P]\rho_b')^4 \left(\frac{1-T_r}{1-T_{br}}\right)^{4n} \times 10^{-3}, \quad 4n = 1.22 \text{ (recommended between 1.0 and 1.24)} \tag{5}$$

where ρ_b' is the molar liquid density at the normal boiling point in $mol\,cm^{-3}$; $T_r \equiv T/T_{crit}$ is the reduced temperature; while $T_{br} \equiv T_b/T_{crit}$ is the reduced normal boiling point. The normal boiling points T_b and the critical temperatures are 283.40 K and 423.27 K for R1234ze(Z), and 291.47 K and 438.75 K for R1233zd(E) (Akasaka *et al.*, 2015; Hulse *et al.*, 2012). [P] is a temperature independent parameter characteristic of a substance, called parachor. The values of parachor [P] are respectively 180.1 and 209.2 for R1234ze(Z) and R1233zd(E).

ii) Estimation based on the principle of corresponding states, Miller (1963):

$$\sigma = (10 \times P_{crit})^{2/3} T_{crit}^{1/3} (1-T_r)^{11/9} (Q \times 10^{-3}), \quad Q = 0.1196 \left[1 + \frac{T_{br} \ln(P_{crit}/1.01325)}{1-T_{br}}\right] - 0.279 \tag{6}$$

where the critical pressures P_{crit} of R1234ze(Z) and R1233zd(E) are respectively 3.533 MPa and 3.573 MPa (Akasaka *et al.*, 2015; Hulse *et al.*, 2012).

iii) Estimation based on the extended scaled equation, Miqueu *et al.* (2000):

$$\sigma = kT_{crit}\left(\frac{N_A}{V_{crit}}\right)^{2/3}(4.35+4.14\omega)t^{1.26}(1+0.19t^{0.5}-0.25t), \quad t \equiv 1 - T/T_{crit} \tag{7}$$

where k, N_A, V_{crit}, and ω are the Boltzmann constant in $cm^2 kg\,s^{-2}K^{-1}$, Avogadro number, critical volume in $cm^3 mol^{-1}$, and acentric factor, respectively. ω of R1234ze(Z) and R1233zd(E) are respectively 0.3247 and 0.3050 (Akasaka *et al.*, 2014; Lemmon *et al.* 2013)

iv) Estimation based on the extended scaled equation, Di Nicola *et al.* (2011):

$$\sigma = 0.658(10 \times P_{crit})^{0.618} T_{crit}^{0.340}(1+\omega)^{0.770}(1-T_r)^{1.262} \times 10^{-3} \tag{8}$$

where P_{crit} and T_{crit} are the critical pressure in MPa and critical temperature in K. $T_r \equiv T/T_{crit}$ is the reduced temperature. ω is the acentric factor.

The estimated surface tension by the selected methods are plotted in Figures 4 (a) and 4 (b) with lines. As seen, the estimated value by the method of Macleodo-Sugden (Macleodo, 1923; Sugden, 1924) is notably greater than that of the other estimations and present data. Overall, the estimation method of Miller (1963), Miqueu *et al.* (2000), and Di Nicola *et al.* (2011) agreed with the measurement results within $\pm 0.43\,mN\,m^{-1}$. In the case of R1234ze(Z), the estimated values by Miqueu *et al.* (2000) slightly deviate from those by Miller (1963) and Di Nicola *et al.* (2011). The value of present data seems to agree with the estimations by Miller (1963) and Di Nicola *et al.* (2011) rather than that by Miqueu *et al.* (2000). In contrast, for R1233zd(E), the present data are somewhat greater than the estimated values and are close to the estimations by Miqueu *et al.* (2000) rather than those by Miller (1963) and Di Nicola *et al.* (2011).

3.3 Empirical correlation

Empirical correlations are developed for R1234ze(Z), R1233ze(Z), and also for R245fa, because the critical parameters of R245fa were recently updated by Higashi *et al.* (2015). The solid lines in Figures 3(b), 4 (a), and 4 (b) denote the empirical correlation, which has the following functional form of the van der Waals equation.

$$\sigma = \sigma_0 (1-T/T_{crit})^n \tag{9}$$

where σ, T, and T_{crit} are the surface tension in $N\,m^{-1}$, temperature in K, and the critical temperature in K, respectively. The coefficient σ_0 and the exponent n are optimized to fit the measured surface tension and the provided critical temperature by the least square mean method. This exponent n should range from 1.2 to 1.3 for most non-polar fluids (Reid *et al.*, 1987). According to the scaling law (Widom, 1974), this exponent is given as 1.28 in three dimensions; however, the number 1.26 is most authorized nowadays (Moldover, 1985). The correlations empirically obtained from the measurement data are summarized as,

$$\sigma = 59.90(1-T/427.01)^{1.237}[mN\,m^{-1}] \quad \text{for R245fa} \tag{10}$$

$$\sigma = 56.57(1-T/423.27)^{1.220}[mN\,m^{-1}] \quad \text{for R1234ze(Z)} \tag{11}$$

$$\sigma = 61.95(1-T/438.75)^{1.277}[mN\,m^{-1}] \quad \text{for R1233zd(E)} \tag{12}$$
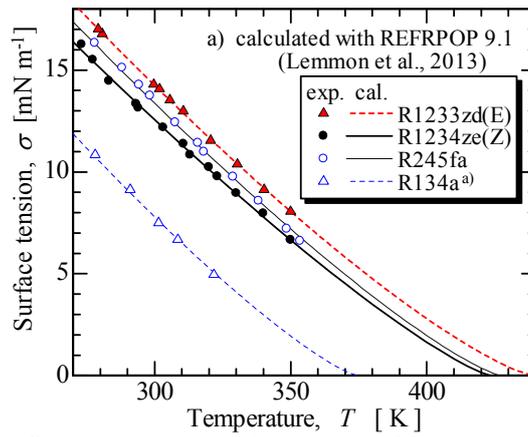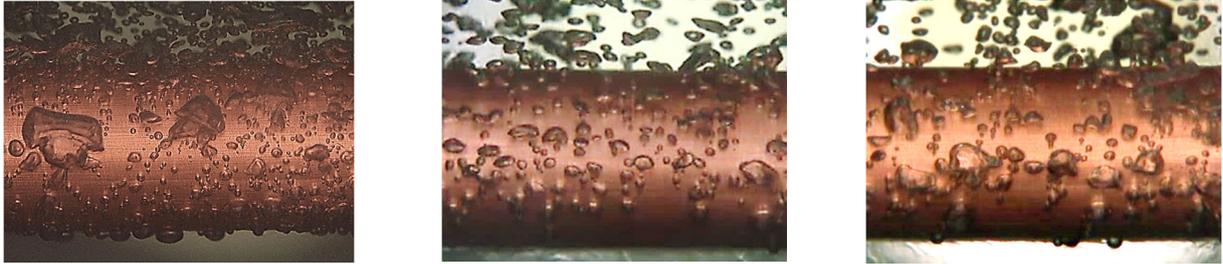
Figure 5. Comparison on surface tension between refrigerants

(a) R1234ze(Z) at 20 ºC and 20 kWm^{-2}, (b) R245fa at 20 ºC and 20 kWm^{-2}, (c) R1233zd(E) at 20 ºC and 20 kWm^{-2}, σ = 13.4 mNm^{-1}, α = 2.46 kWm^{-2}K^{-1}. σ = 14.1 mNm^{-1}, α = 2.00 kWm^{-2}K^{-1}. σ = 15.1 mNm^{-1}, α = 1.66 kWm^{-2}K^{-1}.
Figure 6. Effects of surface tension on pool boiling on a horizontal tube of 19.12 mm OD.

The exponents in the empirical correlations Eqs. (10) to (12) are close to the suggested number in previous studies. Figure 5 compares surface tension between refrigerants. The symbols denote the present experimental data; while, the lines denote the calculated surface tension by the empirical correlations for R245fa, R1234ze(Z), and R1233zd(E), and REFPROP 9.1 (Lemmon *et al*., 2013) for R134a. The calculated surface tension is highly overlapped with the experimental surface tension. The developed empirical correlations represent the measured surface tension within ±0.13 mN m^{-1} and are applicable for temperatures from 270 K to 360 K.

3.4 Effects of surface tension on pool boiling heat transfer

For bubble departure diameter D_b (i.e., equilibrium breakoff diameter) of nucleate pool boiling, Fritz (1935) introduced a model: $D_b \propto \{2\sigma/[g(\rho'-\rho'')]\}^{1/2}$. This is dictated by the balance between buoyancy force and surface tension force under the thermodynamic equilibrium. Essentially, what this model expresses is that the bubble diameter is proportional to the square of the capillary constant, as in Eq. (3). This model gives larger bubble diameter D_b for the refrigerants exhibiting a larger capillary constant. Figure 6 is a picture of bubble formation on a horizontal tube with outer diameter of 19. 12 mm at a saturation temperature of 20 °C and a heat flux of 20 kWm^{-2}. The detailed experimental procedure is stated in Nagata *et al*. (2015). In Figures 6(a), 6(b), and 6(c), the surface tension is calculated with Eqs. (10) to (12) and the heat transfer coefficient α are noted in the captions for R1234ze(Z), R245fa, R1233zd(E). The layout was set by the order of the surface tension. As seen in Figure 6, the bubble size on the tube surface becomes greater for the refrigerants exhibiting greater surface tension. This indicates the exact same tendency as Fritz derived. The bubble behaviour has a dominant effect on the heat transfer coefficient. That is, the smaller bubble with denser nucleation site and more frequent bubble departure exhibits higher heat transfer coefficient. The obtained heat transfer coefficients, 2.46 kWm^{-2}K^{-1}.for R1234ze(Z), 2.00 kWm^{-2}K^{-1} for R245fa, and 1.66 kWm^{-2}K^{-1} for R1233zd(E) confirm this.

4. CONCLUSION

The surface tension data of low GWP refrigerants R1234ze(Z) and R1233zd(E) have been measured at temperatures from 270 K to 360 K by the differential capillary rise method. The measurement method was validated with conventional refrigerants R134a and R245a. The estimation method of Miller (1963), Miqueu *et al*. (2000), and Di Nicola *et al*. (2011) agreed with the measurement results within ±0.43 mN m^{-1}. Empirical correlations with the function form of van der Waals equation were developed as,

$$\sigma = 59.90(1-T/427.01)^{1.237} \text{ for R245fa,}$$
$$\sigma = 56.57(1-T/423.27)^{1.220} \text{ for R1234ze(Z),}$$
$$\text{and } \sigma = 61.95(1-T/438.75)^{1.277} \text{ for R1233zd(E),}$$

where σ and T are the surface tension in mN m^{-1} and the temperature in K, respectively. The correlation represents the measured surface tension values within ± 0.13 mN m^{-1}. The effect of surface tension on bubble behaviour in pool boiling was visually confirmed. The larger bubble diameter were observed in the refrigerants that exhibit greater surface tensions.

ACKNOWLEDGMENTS

NOMENCLATURES

D_b	bubble departure diameter	[m]	$\bar{\varepsilon}$	bias	[N m^{-1}]
N_A	Avogadro number	[-]	σ	surface tension	[N m^{-1}]
P	pressure	[MPa]	σ_0	substance dependent parameter	[N m^{-1}]
$[P]$	parachor	[-]	ρ'	orthobaric liquid density	[kg m^{-3}]
T	temperature	[K]	ρ''	orthobaric vapor density	[kg m^{-3}]
V	molar volume	[cm^{3}mol^{-1}]			
a^2	capillary constant	[m^2]	**subscripts**		
g	local gravitational acceleration	[m s^{-2}]	1	capillary tube of the smaller diameter	
g_n	normal gravitational acceleration	[m s^{-2}]	2	capillary tube of the larger diameter	
h	capillary rise height	[m]	b	normal boiling point	
k	Boltzmann constant	[cm^2 kg s^{-2}K^{-1}]	c	corrected	
r	radius	[m]	cal	calculation	
t	reduced temperature	[-]	crit	critical point	
α	heat transfer coefficient	[Wm^{-2}K^{-1}]	m	measured	
ω	acentric factor	[-]	meas	measurement	
θ	contact angle	[rad]	r	reduced parameter	

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