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Surface tension measurement of low GWP refrigerant mixtures HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32

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ABSTRACT

The study presents the surface tension measurement data of HFO-1123 and HFO-1234ze(E) with global warming potential (GWP) of less than 1 and of binary mixtures HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32. The capillary constant and surface tension of the fluids are measured across temperatures in a range of 267 K to 307 K with a differential capillary rise with a propagated uncertainty of $\pm 0.23 \text{ mN m}^{-1}$. Among the selected predicting methods, the correlations of DiNicola et al. (2018) exhibit the best agreement to the measured surface tension of both HFO-1123 and HFO-1234ze(E). In order to predict the surface tension HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32, empirical correlations are proposed based on the measured data.

1. INTRODUCTION

Worldwide attention focused on HFOs (hydro-fluoro-olefins), HFO-1123 and HFO-1234ze(E) with GWP_{100} (global warming potential of 100-year time horizon) that is less than 1 (Myhre et al., 2013) as replacements for R410A and R134a in the field of air conditioning and refrigeration engineering. However, the use of the HFO alone in the aforementioned heat pump systems lacks adequate capacity and/or chemical stability. In order to compensate for these shortcomings, mixing with HFC-32 is considered as a solution. Given the significance, the thermophysical and transport properties of above refrigerants are vigorously investigated in recent years. Higashi and Akasaka (2016) reported precisely measured critical parameters, vapor-liquid coexistence curve, and saturation pressure among others for HFO-1123 and HFO-1123/HFC-32. Based on the measurement data, Helmholtz type EOS (equation of state) and mixing parameters are proposed. Additionally, with respect to HFO-1234ze(E) and HFO-1234ze(E)/HFC-32, the EOS and the mixing parameters are provided in REFPROP9.1 (Lemmon et al., 2013). Nevertheless, the data of transport properties are not adequately reported. The surface tension plays an important role in boiling/condensation heat transfer and is essential in heat pump systems. In order to provide the sufficient data bank for phase change process prediction, the present study presents the surface tension measurement data of HFC-32, HFO-1123, and HFO-1234ze(E), and their binary mixtures HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32 at various mass fractions.

2. MEASUREMENT METHOD

2.1 Measurement setup

Fig. 1 shows a measurement setup developed based on the capillary rise method. The surface tension is measured as the capillary elevation in small diameter tubes immersed in a liquid. Two capillaries with inner radii of $r_1 = 0.4222 \pm 0.0009 \text{ mm}$ and $r_2 = 0.7526 \pm 0.0009 \text{ mm}$ are vertically set by a supporting brace in a pressure vessel (A) composed of a glass tube with 17 mm ID and 25 mm OD. The capillary radii were precisely measured with mercury slugs (Okada et al., 1995). The capillaries and pressure vessel are cleansed by using an alkaline aqueous solution and an ultrasound bath. The pressure vessel filled with a sample refrigerant is placed in a thermostatic bath (B), and the temperature is

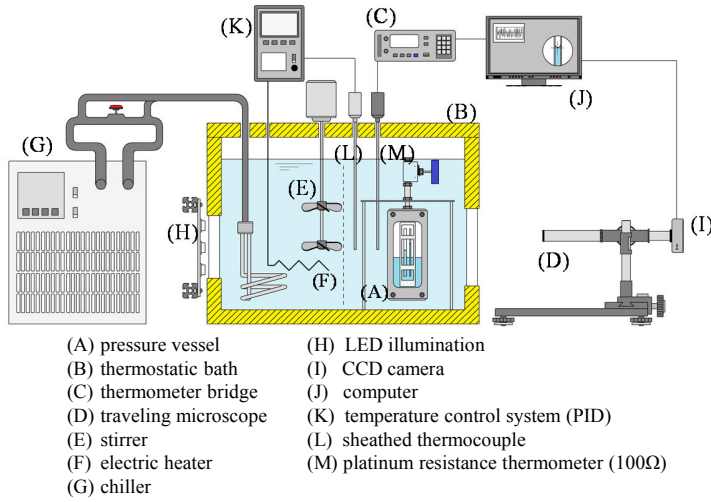


Figure 1: measurement setup

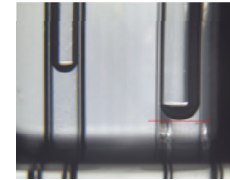
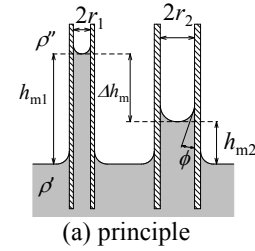


Figure 2: Differential capillary rise height

temperature is measured with a 100 Ω platinum resistance thermometer (C), ASL model F500, calibrated relative to ITS-90. The uncertainty in temperature is estimated as within ±5 mK. At a steady state, the capillary rise difference between the two capillary tubes is measured by using a digital traveling microscope (L) with a tolerance of 0.01 mm.

2.2 Data reduction of differential capillary rise method

Fig. 2 illustrates the differential capillary-rise-height. Specifically, as shown in Fig. 2, the height difference of the bottom of the meniscus in each capillary tubes, Δh_m is measured with the traveling microscope (L) and a CCD camera (M) shown in Fig. 1. Fig. 2 (b) shows the CCD image displayed on a monitor (G), shown in Fig. 1 to determine the position of the meniscus bottom. As shown in Fig. 2, the contact angle ϕ is approximated as zero by assuming the semispherical meniscus. In order to obtain the actual differential capillary rise height, the measured differential height Δh_m at the bottom of the meniscus in each capillary tube is corrected by a methodology proposed by Rayleigh (1916).

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

The capillary constant a^2 is determined only from the capillary radii, r_1 and r_2 , and the differential capillary-rise-heights are as follows:

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

where g and g_n denote the local gravitational acceleration of 9.79585 m s⁻² at Nagasaki, Japan and normal gravitational acceleration of 9.80665 m s⁻², respectively. The contact angle ϕ of refrigerants in the capillary tube that is well cleansed is assumed as zero. The surface tension σ is determined from the capillary constant and orthobaric liquid and vapor densities ρ' and ρ'' , respectively, as follows:

$$\sigma = \frac{g \Delta h_c (\rho' - \rho'')}{2(1/r_1 - 1/r_2) \cos \phi} \approx \frac{g \Delta h_c (\rho' - \rho'')}{2(1/r_1 - 1/r_2)} = \frac{a^2 (\rho' - \rho'') g_n}{2} \quad (3)$$

With respect to the binary mixture HFO-1123/HFC-32, the following measurement procedure is introduced. The total charge amount, m_{total} , and the composition, \bar{X} , is determined from each mass of charge in the pressure vessel as follows:

$$m_{\text{total}} = m_{\text{HFO-1123}} + m_{\text{HFC-32}}, \quad \bar{X}_{\text{HFO-1123}} = m_{\text{HFO-1123}} / m_{\text{total}} \quad (4)$$

The charge composition is checked with the completely vaporized sample in a large chamber by a TCD gas chromatograph. The vapour quality in the vessel, x , is determined at a condition of satisfying the total volume is always equal to the internal volume of the pressure vessel.

$$V_{\text{total}} = m_{\text{total}} x / \rho_V(\bar{X}, T, x) + m_{\text{total}} (1-x) / \rho_L(\bar{X}, T, x) \quad (5)$$

The internal volume excluding some parts such as capillary tubes is 21.63 mL. The vapor quality typically ranges

from 0.04 to 0.12 during the measurement. The mass fractions of vapor and liquid phases can be calculated at that quality by REFPROP 9.1 (Lemmon et al. 2016). Therefore, the vapor and liquid densities are calculated as a function of charge composition, temperature, and quality. The liquid phase mass fraction changes within ± 0.01 from the charged mass fraction. The above vapor and liquid densities, ρ_v and ρ_L , are substituted into the orthobaric densities, ρ' and ρ'' , in Eq. (3), and thus the surface tension is obtained for the binary mixture HFO-1123/HFC-32.

2.3 Sample Refrigerant and Thermodynamic Properties

The sample refrigerants of HFO-1123 and HFC-32 are provided by Asahi Glass Co., Ltd. The nominal purity of the samples is considered to exceed 99.5 mol% (impurities: acids < 0.0001 mol%, $H_2O < 0.002$ mol%, others < 0.5 mol%). Based on Higashi and Akasaka (2016), the critical temperatures of HFO-1123 and HFO-1234ze(E) at which the liquid-vapor interface disappears are 331.7 K (Higashi and Akasaka, 2016) and 382.5 K (Lemmon et al., 2013), respectively. The temperatures of the binary mixtures dependent the composition is given as follows:

$$T_{\text{crit}} = \bar{\theta}_1 T_{\text{crit},1} + \bar{\theta}_2 T_{\text{crit},2} + 2\bar{\theta}_1 \bar{\theta}_2 \Delta_T$$

$$\text{where, } \bar{\theta}_i = \frac{\tilde{x}_i V_{\text{crit},i}^{2/3}}{\sum \tilde{x}_j V_{\text{crit},j}^{2/3}} \quad (6)$$

$$\Delta_T = \begin{cases} -5.5 & \text{for HFO-1123/HFC-32} & \text{Higashi and Akasaka (2016)} \\ -4.1 & \text{for HFO-1234ze(E)/HFC-32} & \text{Kobayashi et al. (2010)} \end{cases}$$

The density data are also extremely important to determine the surface tension in the differential capillary rise height method as specified in Eq. (4). In the study, the liquid and vapor densities are evaluated with EOS of HFO-1123 developed by Higashi and Akasaka (2016). With respect to the mixture, the vapor quality is also considered to determine mass fraction difference in vapor and liquid phases. Furthermore, in order to predict the mutual interaction effects in HFO-1123/HFC-32, a KW0 mixture model with default parameters provided by REFPROP9.1 is used. There is a paucity of studies on the density data, and thus the uncertainty in calculated density is relatively high. With respect to HFO-1234ze(E)/HFC-32, the mixing parameters provided by REFPROP 9.1 provides are used (KW2 model with $\beta_1=1.0034$, $\gamma_1=0.97786$, $\beta_v=1.0059$, $\gamma_v=0.98271$, and $F_{ij}=-0.62542$).

2.4 Validation check with HFC-32

Fig. 3 plots the surface tension measurement results of HFC-32 and the deviation of the results from the calculated surface tension by REFPROP 9.1 as a function of temperature. The various measurement data in extant studies are also plotted for comparison purposes. The present data for series 1, 2, and 3 exhibit a high degree of overlap and are measured with the other sample re-charged after the cleansing in different days. The data agree with the calculated and referential surface tension data within $\pm 0.2 \text{ mN}\cdot\text{m}^{-1}$. The measurement uncertainty (2k) is typically $\pm 0.23 \text{ mN}\cdot\text{m}^{-1}$ (Taylor, 1982), and the deviation is lower than the uncertainty. Overall, the validation of repeatability and reproducibility is demonstrated with the maximum authorized value of HFC-32.

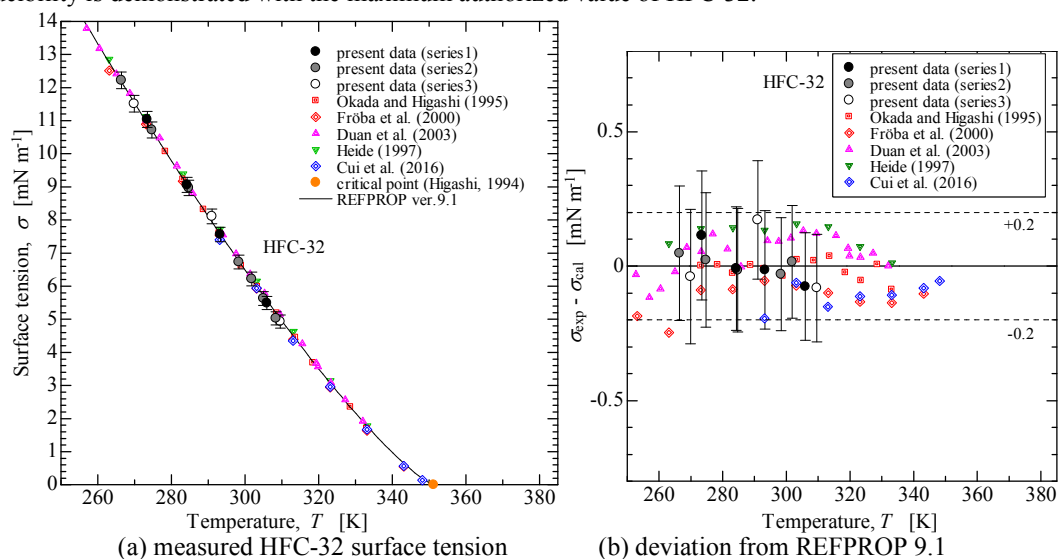


Figure 3: Measured surface tension data and proposed empirical correlation for HFC-32

3. MEASUREMENT RESULTS AND CORRELATION

3.1 Measurement results of HFO-1123 and HFO-1234ze(E)

Fig. 4 plots the surface tension measurement results as a function of saturation temperature for HFO-1123 and HFO-1234ze(E) alone. The circled symbols and the appended vertical bars denote the present data and the measurement uncertainty in surface tension that is estimated as typically $\pm 0.23 \text{ mN}\cdot\text{m}^{-2}$. The surface tension of HFO-1123 is lower than that of HFC-32. Conversely, the surface tension of HFO-1234ze(E) exceeds that of HFC-32 at temperatures above 250 K. With respect to HFO-1123, the data of series 1 to 3 overlap within $\pm 0.2 \text{ mN}\cdot\text{m}^{-2}$. The measured surface tension exhibits a gradual curve asymptotic to the critical point. The present HFO-1234ze(E) data exhibits a slight deviation to the data obtained by Zhao et al., (2017) at lower temperatures. However, it exhibits good agreement with data obtained by Tanaka and Higashi (2013) and Zhao et al. (2014) within uncertainty. With the exception of Tanaka and Higashi (2013), the measured data exhibit a slightly lower surface tension than that calculated by REFPROP 9.1. Based on the present measurement data, the empirical correlations of Van der Waals type are proposed as follows:

$$\sigma_{\text{HFO-1123}} = 61.02 \times 10^{-3} (1 - T/T_{\text{crit}})^{1.301} [\text{N m}^{-1}], \quad T_{\text{crit}} = 331.7 [\text{K}] \quad (7)$$

$$\sigma_{\text{HFO-1234ze(E)}} = 58.07 \times 10^{-3} (1 - T/T_{\text{crit}})^{1.265} [\text{N m}^{-1}], \quad T_{\text{crit}} = 382.5 [\text{K}] \quad (8)$$

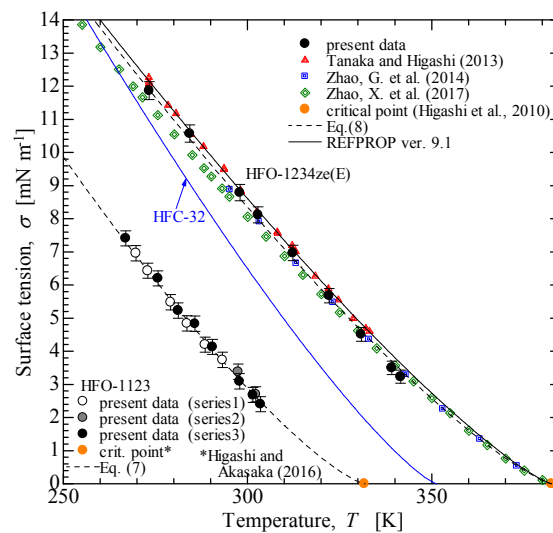


Figure 4: Measured surface tension data and proposed empirical correlation for HFO-1123 and HFO-1234ze(E)

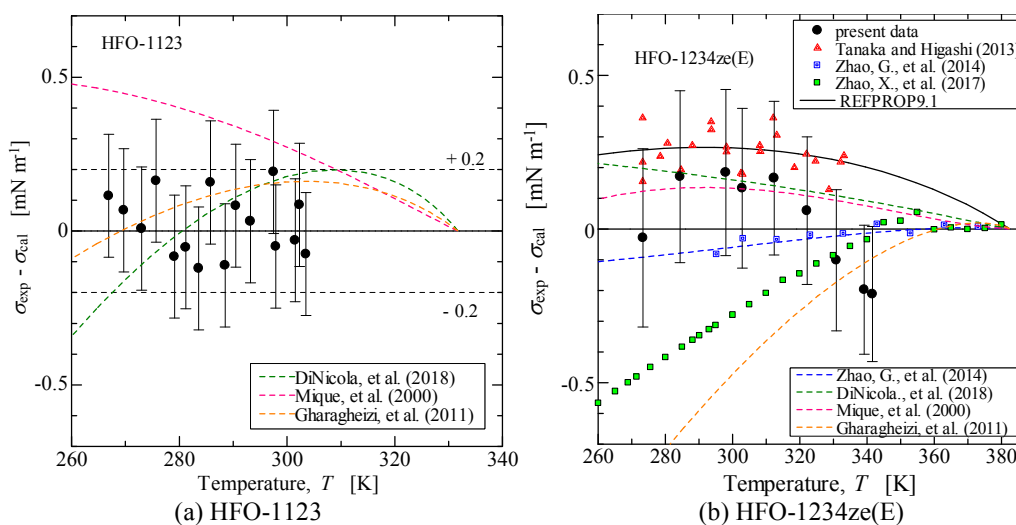


Figure 5: Deviation from the proposed empirical correlations Eqs. (7) and (8)

The calculated surface tension by Eqs. (7) and (8) are plotted as the dashed line in Fig. 4. The deviation of measurement data and prediction results from the calculated value are plotted in Figs. 5 (a) and 5(b). When compared to Fig. 5 (a) for HFO-1123, the empirical correlation agrees with the data within $\pm 0.2 \text{ mN}\cdot\text{m}^{-2}$ in the temperature range from 266 K to 304 K. The prediction of DiNicola et al. (2018) and Gharagheizi et al. (2011) agree with the present data almost within the measurement uncertainty. As shown in Fig. (8), the proposed correlation agrees well with the present data and that reported by Zhao et al. (2014). The REFPROP9.1 and the measured data obtained by Tanaka and Higashi (2013) yield higher value relative to the other data. The predictions of Miqueu et al. (2000) and DiNicola (2018) indicate a similar trend that exhibits a high degree of overlap with the present data. The prediction of Gharagheizi et al. (2011) appears to underestimate the surface tension for HFO-1234ze(E).

3.2 Measurement results of HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32

Fig. 6 plots the surface tension measurement results for the binary mixture HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32 at various charged mass fractions. All the measured surface tension data are within the value of HFC-32 and HFO-1123 or HFO-1234ze(E) by itself. Fig. 6(a) shows the predicted surface tension by the correlation of Heide (1997) and DiNicola (2017). The correlation obtained by Heide (1997) is not available at temperatures beyond the critical point that one of the component reaches. The correlation obtained by DiNicola (2017) indicates precise agreement except at a mass fraction of 0.19/0.81. In Fig. 6(b), the calculated surface tension by REFPROP is plotted for HFO-1234ze(E)/HFC-32. Furthermore, REFPROP 9.1 indicates a precise agreement at HFC-32 richer mass fractions although it slightly overestimates the same at HFO-1234ze(E) richer mass fractions because it positively deviates from the data of HFO-1234ze(E) by itself. Nevertheless, the qualitative agreement in the temperature dependency between REFPROP and present data are observed especially at temperatures from 270 K to 270 K in which the surface tension band between HFC-32 and HFO-1234ze(E) by itself becomes narrow.

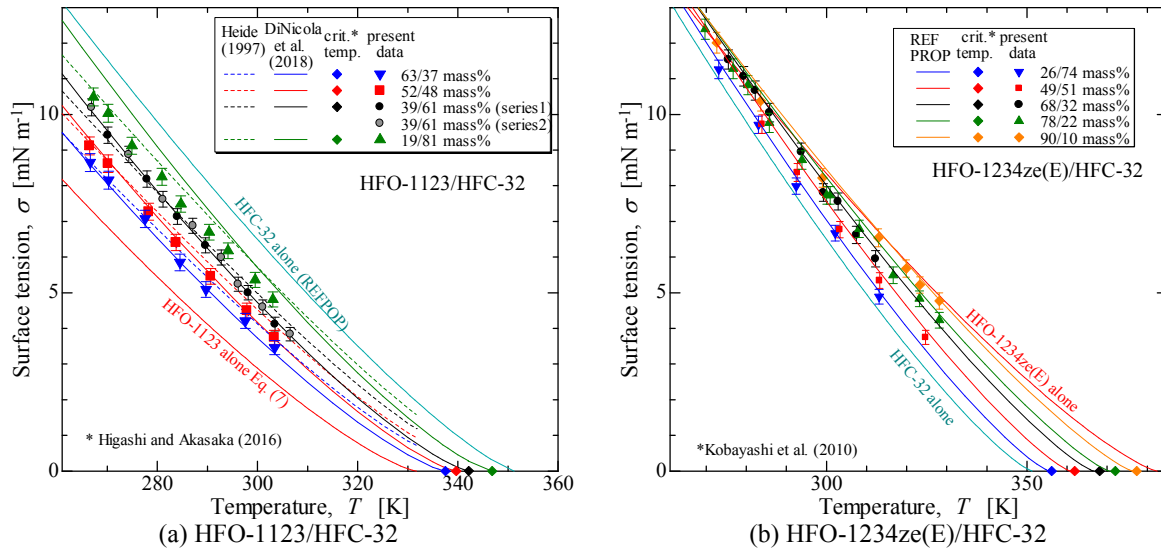


Figure 6: Measured surface tension data and available predicting correlations for binary mixtures

3.3 Empirical correlation for HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32

Based on the measured surface tension data, the following empirical correlation that is available for the entire mass fraction range and temperatures from 265 to the critical point is proposed for the tested binary mixtures.

$$\sigma = \sigma_0 \left(1 - T/T_{\text{crit}}\right)^c$$

where,

$$\sigma_0 = \sigma_{0,1} \bar{\theta}_1 + \sigma_{0,2} \bar{\theta}_2 + \Delta_{\sigma_0} \bar{\theta}_1 \bar{\theta}_2, \quad c = c_2 + (c_1 - c_2) \bar{\theta}_1, \quad \bar{\theta}_i = \frac{\tilde{x}_i V_{\text{crit},i}^{2/3}}{\sum_j \tilde{x}_j V_{\text{crit},j}^{2/3}} \quad (9)$$

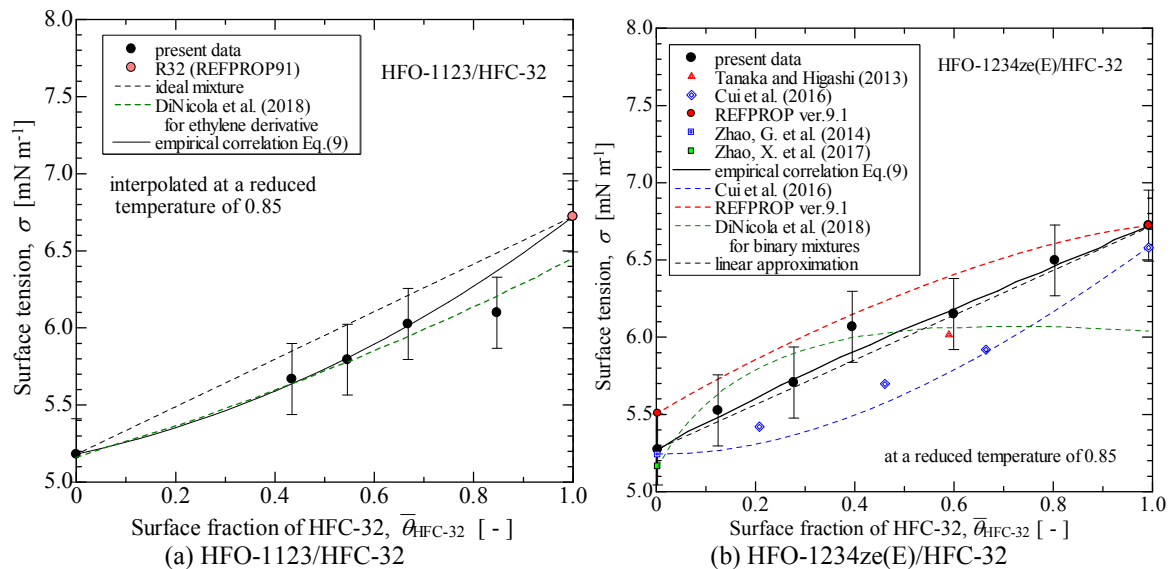
$$\Delta_{\sigma_0} = \begin{cases} -4.1 \times 10^{-3} & \text{for HFO-1123/HFC-32} \\ +3.0 \times 10^{-3} & \text{for HFO-1234ze(E)/HFC-32} \end{cases}$$

Table 1: Parameters in the empirical correlation Eq. (9)

	molar mass	$V_{crit,j}$	$T_{crit,j}$	$\sigma_{0,j}$	c_j
	[g mol ⁻¹]	[cm ³ mol ⁻¹]	[K]	[N m ⁻¹]	[-]
HFC-32	52.024	123	351.3	71.47×10^{-3}	1.246
HFO-1123	82.025	167	331.7	61.02×10^{-3}	1.301
HFO-1234ze(E)	114.04	235	382.5	58.07×10^{-3}	1.265

where, σ_0 and c denote the coefficient and exponent, respectively, of the van der Waals equation for binary mixtures obtained from the components, and $\Delta_{\sigma 0}$ denotes the only adjustable parameters for binary mixtures. The parameters are specified in Table 1 for HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32. The empirical correlation expresses the composition dependence with surface fraction $\bar{\theta}$ and is the same as the crucial locus. Thus, at surface fractions of 0 and 1, the correlation results in the surface tension of the components HFC-32 and HFO-1123 or HFO-1234ze(E) alone. The adjustable parameter is determined as the deviation from linear variation relative to surface fraction. Figure 7 shows the surface tension variation at a reduced temperature of 0.85. The surface tension of HFO-1123/HFC-32 is slightly lower than the assumed linear variation. The prediction obtained by DiNicola et al. (2018) agree well at HFC-32 surface fractions below 0.7 although it slightly deviates at 1.0. The present data at surface fraction 0.85 exhibit trends that evidently differ from the gradual variation and deviates from the empirical correlation. The present surface tension data of HFO-1234ze(E)/HFC-32 varies almost linearly. Specifically, REFPROP9.1 exhibits a positive deviation to the data at surface fraction 0.0. Conversely, the prediction and measurement data of Cui et al. (2016) exhibit a negative deviation. The negative deviation is also observed at a surface fraction of 1.0.

Fig. 8 confirms the deviation of present data from the empirical correlation Eq. (9). The empirical correlation agree with the measured data within ± 0.35 mN·m⁻¹ at entire mass fraction range for HFO-1123/HFC-32/HFO-1234ze(E)/HFC-32.

**Figure 7:** Variation in surface tension against surface fraction at a reduced temperature of 0.85

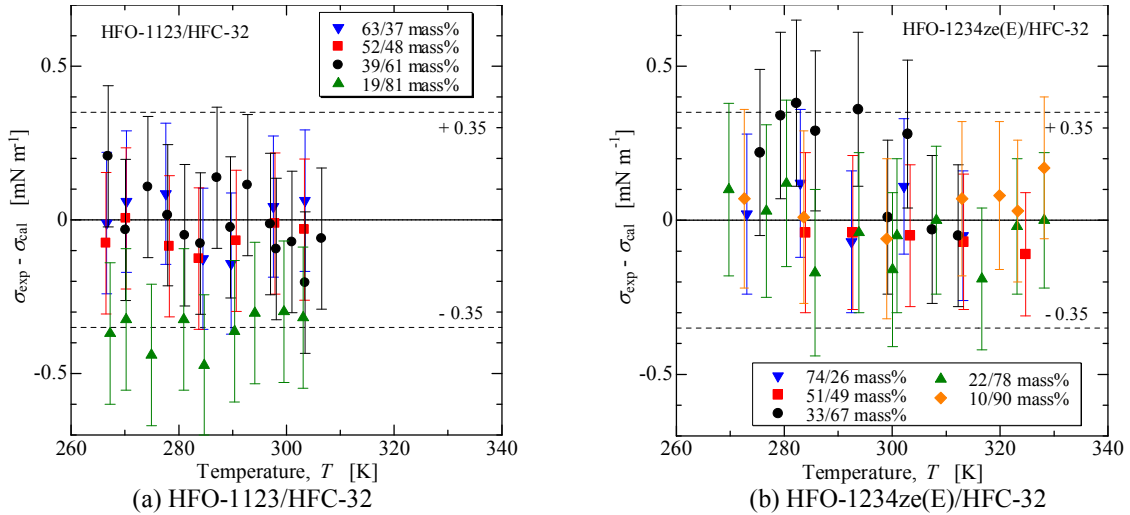


Figure 8: Deviation from the empirical correlation Eq.(9) for mixtures

4. CONCLUSIONS

Given the differential capillary rise method, the surface tension of HFO-1123 and HFO-1234ze(E) alone and of the binary mixtures HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32 are measured across temperatures of 265 K to 345 K with an uncertainty within $\pm 0.23 \text{ mN}\cdot\text{m}^{-1}$. Based on the measured data, the following empirical correlations are proposed.

- With respect to HFO-1123 and HFO-1234ze(E) alone,

$$\sigma_{\text{HFO-1123}} = 61.02 \times 10^{-3} (1 - T/T_{\text{crit}})^{1.301} [\text{N m}^{-1}], \quad T_{\text{crit}} = 331.7 [\text{K}]$$

$$\sigma_{\text{HFO-1234ze(E)}} = 58.07 \times 10^{-3} (1 - T/T_{\text{crit}})^{1.265} [\text{N m}^{-1}], \quad T_{\text{crit}} = 382.5 [\text{K}]$$

- With respect to the binary mixtures HFO-1123/HFC-32 and HFO-1234ze(E)/HFC-32,

$$\sigma = \sigma_0 (1 - T/T_{\text{crit}})^c \quad \text{where, } \sigma_0 = \sigma_{0,1} \bar{\theta}_1 + \sigma_{0,2} \bar{\theta}_2 + \Delta_{\sigma_0} \bar{\theta}_1 \bar{\theta}_2, \quad c = c_2 + (c_1 - c_2) \bar{\theta}_1,$$

$$\text{and } \Delta_{\sigma_0} = \begin{cases} -4.1 \times 10^{-3} & \text{for HFO-1123/HFC-32} \\ +3.0 \times 10^{-3} & \text{for HFO-1234ze(E)/HFC-32} \end{cases}$$

These correlations agree well with the measured data within $\pm 0.35 \text{ mN}\cdot\text{m}^{-1}$.

NOMENCLATURE

a^2	:capillary constant (m^2)	\bar{X}	:charged/nominal mass fraction (-)
g	:local gravitational acceleration (m s^{-2})	Δh_c	:corrected differential height (m)
g_n	:normal gravitational acceleration (m s^{-2})	Δh_m	:measured differential height (m)
M	:mass (kg)	ϕ	:contact angle (rad)
P	:pressure (MPa)	$\bar{\theta}$: charged/nominal surface fraction (-)
r_1	:smaller radius of capillary (m)	ρ'	:orthobaric vapour density (kg m^{-3})
r_2	:larger radius of capillary (m)	ρ''	:orthobaric liquid density (kg m^{-3})
T	:temperature (K)	ρ_L	:liquid density of mixture (kg m^{-3})
V	:molar volume ($\text{cm}^3 \text{mol}^{-1}$)	ρ_V	:vapour density of mixture (kg m^{-3})
x	:vapour quality (-)	σ	:surface tension (N m^{-1})
\tilde{x}	:charged/nominal mole fraction (-)		

Subscript

HFO-1123 :HFO-1123

HFC-32 :HFC-32

total :total

cal :calculate

HFO-1123/HFC-32 :HFO-1123/HFC-32 exp :measured
crit :critical point

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