



Viscosity measurements for 2,3,3,3-tetrafluoroprop-1-ene (R1234yf) and trans-1,3,3,3-tetrafluoropropene (R1234ze(E))

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ABSTRACT

The refrigerants with high global warming potential (GWP) will be phased out, which has stimulated research to find replacement fluids. The chemical and thermophysical properties of 2,3,3,3-tetrafluoroprop-1-ene (R1234yf) and trans-1,3,3,3-tetrafluoropropene (R1234ze(E)) have made them the promising candidate. Accurate thermophysical properties will be essential to design and develop efficient processes that use these compounds. In this work, the viscosity of 2,3,3,3-tetrafluoroprop-1-ene (R1234yf) in the temperature of (243 to 363) K from saturated pressures up to 30 MPa was reported. With regard to R1234ze(E), measurements were performed at temperatures between (243 and 373) K from saturated pressures up to 30 MPa. The viscosity was measured using a vibrating-wire viscometer with a combined expanded uncertainty of about 2.0%. The scheme based on a hard-sphere model was used to correlate the experimental results.

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1. Introduction

Recently, the release of Mobile Air Conditioning (MAC) directive of EU, which bans the use of refrigerants with a GWP above 150 in new type of mobile air conditioning from 2011 in the EU market, has triggered the research and development of new refrigerants for mobile air conditioners. Hydrofluorocarbons (HFCs) having great GWP values will be shifting toward low GWP substances. Hydrofluoro-olefins (HFOs) such as 2,3,3,3-tetrafluoroprop-1-ene (R1234yf) and trans-1,3,3,3-tetrafluoropropene (R1234ze(E)) having low GWP values are expected as alternatives for HFCs. R1234yf offers a remarkably low global warming potential (GWP) of four relative to CO₂ for a 100 year time horizon [1]. Although it is slightly flammable, under normal conditions, it is a stable material with low toxicity. It has been assigned a safety classification of A2L according to ASHRAE Standard 34 [2]. R-1234ze(E) is also one of fluorinated propene isomers, and its GWP is also low (about six) [3]. It can be used as one component of refrigerant mixtures or used as a foam-blowing agent in applications requiring a low GWP.

The thermophysical properties of refrigerants are indispensable for optimum design of energy-conversion systems and selection of the refrigerant. However, to the author's knowledge, only a limited number of the thermophysical properties of R1234yf and R1234ze(E) could be found in the published literature. For the viscosity in the saturate and liquid phases of R1234yf and R1234ze(E),

only six various sets were found in literature with limited temperature and pressure range [4–8]. In the present work, the viscosity of R1234yf and R1234ze(E) from saturated pressure up to 30 MPa was measured using a vibrating-wire viscometer. These new experimental data, covering a temperature range of (243 to 363) K for R1234yf and (243 to 373) K for R1234ze(E), were used to develop correlations for viscosity based on the hard-sphere scheme.

2. Experimental

2.1. Chemicals

The toluene was supplied by Tianjin Fuyu Industry of Fine Chemicals Co., Ltd., China with a declared mass purity of 0.995. 1,1,1,2-tetrafluoroethane (R134a) was manufactured by Sinochem Modern Environmental Protection Chemicals (Xi'an) Co., Ltd., China with a declared mass purity of 0.999. 2,3,3,3-tetrafluoroprop-1-ene (R1234yf) and trans-1,3,3,3-tetrafluoropropene (R1234ze(E)) were supplied by Honeywell with declared mass purity of 0.999. Complete specification of chemical samples is listed in table 1. All refrigerants above were purified by using freeze–pump–thaw cycles with liquid nitrogen and a high vacuum pump to eliminate the effect of non-condensable gaseous impurities.

2.2. Apparatus

The apparatus consists of a vibrating-wire viscometer, formed from a wire-clamped at both ends, a pressure vessel, and magnets

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TABLE 1
Specification of chemical samples.

| Chemical name | Source | Initial mass fraction purity ^a | Purification method |
|---------------|---|---|---------------------|
| Toluene | Tianjin Fuyu Industry of Fine Chemicals Co., Ltd., China | 0.995 | No |
| 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 |

^a All the stated purities of the samples listed above were obtained by the certificates of merchandises, and no further purity measurements were performed after the purification.

that were described in detail previously [9,10]. However, tiny changes had been performed to improve the performance of the apparatus. As provided by predecessors, a centerless-ground rod gave better results than rough drawn wires [11,12]. The new instrument employed a centerless-ground tungsten wire with a nominal radius of $(50.00 \pm 0.32) \mu\text{m}$ and a nominal length of $L = 58 \text{ mm}$, supplied by Metal Cutting. The wire was clamped at one end between two stainless steel pieces with screws. The two clamps were separated from each other by a machinable glass ceramic tube. A pair of samarium-cobalt magnets was mounted parallelly onto a cage with a distance of 6 mm, and the length of the rectangular magnets is about $L_B = 40 \text{ mm}$, and the ratio $L/L_B = 1.45$ was sufficient to eliminate the third harmonic mode oscillation. The magnetic field exerted on the wire was estimated to be 0.4 T using Ansoft Maxwell v.10 software. The cell was assembled in a custom-made stainless steel vessel with a maximum design pressure of 70 MPa. The vibrating-wire was connected with external instrument by using electrical leads penetrating out of two holes on the top of the vessel body, which were hermetically sealed with glass-enclosed. A sinusoidal voltage was achieved by a function generator (model: 33220A, Agilent). The in-phase and quadrature voltages of the signal were detected by the lock-in amplifier (model: SR830, Stanford Research Systems) over the frequency range.

A thermostatic bath (model: 7037, Fluke) was used to maintain the constant temperature of the apparatus. The temperature stability and uniformity of the bath were better than $\pm 2 \text{ mK}$. The temperature of the thermostatic bath was measured using a 100Ω platinum resistance thermometer connected to a DMM (model: 3458A, Agilent). The thermometer has been calibrated over the experimental temperature range against a 25Ω standard platinum-resistance (model: CST6601, No. 68804, Beijing ConST Instruments Technology Inc.) connected to a precision thermometry bridge (model: F700B, ASL). The combined expanded uncertainty

of temperature with level of confidence 0.95 ($k = 2$) is $U_c(T) = 11.2 \text{ mK}$.

The pressure was generated with a manual piston pump (model: 50-6-15, HIP) and measured by a high pressure transducer (model: P3 MB, HBM) with a pressure range up to 100 MPa. A nanovolt meter (model: 34420A, Agilent) with 7 1/2 digits resolution was employed for the transformation of the pressure transducer measurement signal. The combined expanded uncertainty of pressure $U_c(p) = 0.117 \text{ MPa}$ ($k = 2$). A diagram of the experimental system is shown in figure 1.

2.3. Calibration and validation

The internal damping, Δ_0 , and the radius of tungsten wire, R , were determined by calibration. The vacuum measurement was used to obtain the internal damping, while the wire radius was experimentally determined in toluene at $T_{\text{ref}} = 298.15 \text{ K}$ and $p_{\text{ref}} = 0.1 \text{ MPa}$. The reference datum was taken to be $\rho_{\text{ref}} = 862.24 \text{ kg} \cdot \text{m}^{-3}$ and $\eta_{\text{ref}} = 0.5542 \text{ mPa} \cdot \text{s}$ [13,14]. The final determined values of R and Δ_0 were $49.60 \mu\text{m}$ and $1.96 \cdot 10^{-5}$, respectively. So with density of the fluid known, its viscosity could be obtained by fitting the experimental complex voltages to working equations.

This work carried out the viscosity measurement for toluene over the temperature range from (273 to 363) K and at pressures up to 30 MPa, listed in table 2. When comparing the experimental results with the calculated results using REFPROP Ver. 9.0 [20], it can be observed that the relative deviation, shown in figure 2, is within the range of (–1.0 to 0.2)%. A same procedure of uncertainty analysis of the measurements was performed as our previous work [9]. By taking into account the uncertainties of temperature, pressure, repeatability of measurement, regression procedure, and the density of fluid, the combined expanded uncertainty of viscosity with level of confidence of 0.95 ($k = 2$) is better than $\pm 2.0\%$.

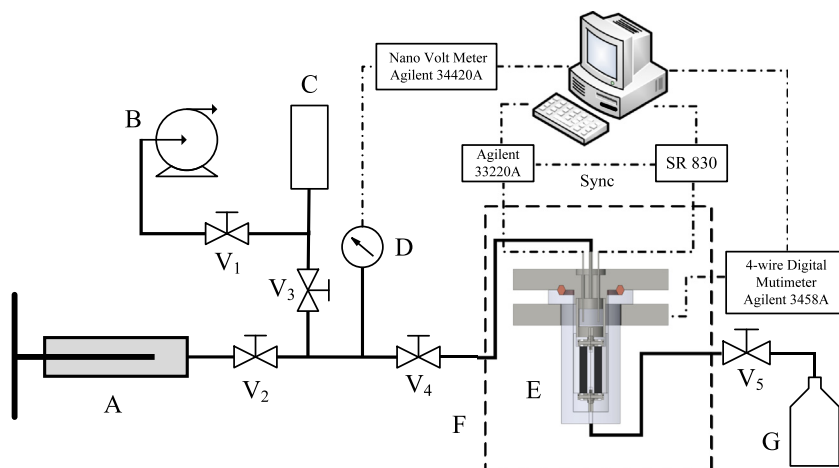


FIGURE 1. Schematic diagram of experimental system: (A) Manual piston pump; (B) Vacuum pump; (C) Sample container; (D) Pressure transducer; (E) Vibrating wire viscometer; (F) Thermostatic bath; (G) Wasting recycle; (V1–V5) Valves.

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