

2016

The Viscosity Characteristics for the Mixed Refrigerant HFO-1234yf + HFC-152a

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Yang, Zhangzhang; Wang, Xuehui; Fang, Yibo; Han, Xiaohong; Qiao, Xiaogang; and Chen, Guangming, "The Viscosity Characteristics for the Mixed Refrigerant HFO-1234yf + HFC-152a" (2016). *International Refrigeration and Air Conditioning Conference*. Paper 1650.
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The viscosity characteristics for the mixed refrigerant HFO-1234yf + HFC-152a

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ABSTRACT

Since HFC-134a with high global warming potential (GWP) would make the problem of global warming serious, it will be phased out and substituted by environmental friendly refrigerants. Environmental benign refrigerants such as HFO-1234yf and HFC-152a are regarded as good candidates to substitute HFC-134a and the mixture of HFO-1234yf + HFC-152a is a promising alternative refrigerant. Before the actual application of alternative refrigerants, thermophysical properties of mixed refrigerants need to be carefully investigated. In this paper, the viscosity characteristics for the mixed refrigerant HFO-1234yf + HFC-152a were measured and the experimental results were correlated.

1. INTRODUCTION

The ozone depletion is one of the most important environmental problems around the world since the refrigerants of chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) were extensively used. Due to this, hydrofluorocarbons (HFCs) with zero ozone depletion potential (ODP) were regarded as one of the most suitable substitutions for CFCs and HCFCs. However, the high global warming potential (GWP) of HFCs has increased the effects of global warming. At present, refrigerants with zero ODP and low GWP are promising.

As a preferable alternative to replace CFC-12 in mobile air conditioning systems (MACs), HFC-134a used to be a commonly used refrigerant with zero ODP. However, HFC-134a was only a temporary substitution since its high GWP of 1430 and long atmospheric lifetime of 14 years (Calm and Hourahan, 2007) would make the problem of global warming serious. For the sake of environment, HFC-134a will be phased out and the substitution of it is imperative.

In recent years, environmental benign refrigerants such as HFO-1234yf and HFC-152a (Akram *et al.*, 2013; Cabello *et al.*, 2015; Im *et al.*, 2014; Jarall, 2012; Tillner-Roth, 1995; Tveit *et al.*, 2013) have aroused much concerns in the chemical and air conditioning industry and are regarded as good candidates to substitute HFC-134a because of low GWP and zero ODP. HFO-1234yf, with zero ODP, the GWP of 4, and a very short atmospheric lifetime of 11 days (Minor and Spatz, 2008), shows similar thermophysical properties to HFC-134a. (Calm, 2008) However, it exhibits a smaller volumetric cooling capacity and COP than those of HFC-134a. (Minor *et al.*, 2010) In order to make better use of HFO-1234yf, some HFO-1234yf + HFCs or HFO-1234yf + HCs binary mixtures (Akasaka *et al.*, 2013; Chen *et al.*, 2015a; Chen *et al.*, 2015b; Hu *et al.*, 2014a; Hu *et al.*, 2014b) were proposed as alternative refrigerants. On the other hand, HFC-152a with low GWP of 140 and a short atmospheric lifetime of 1.5 years (Calm and Hourahan, 2007) was considered as a replacement of HFC-134a and has been selected as component in refrigerant mixtures for many years. Hence, the mixture of HFO-1234yf + HFC-152a is a promising alternative refrigerant.

Before the actual application of alternative refrigerants in the refrigeration and air conditioning systems, thermophysical properties of mixed refrigerants need to be carefully investigated. The isothermal vapor liquid equilibrium of the mixture HFO-1234yf + HFC-152a has been studied (Hu *et al.*, 2014b). However, viscosity characteristics of the mixture HFO-1234yf + HFC-152a, one of the major concerns in the study of the thermophysical properties of alternative refrigerants, has not been studied yet.

Since the knowledge of viscosity characteristics has significant impact on heat transfer and pressure drop in the flow, and viscosity data with high accuracy are of considerable value in the calculation of heat transfer and fluid flow, in this work, the measurement of liquid viscosity of the mixture HFO-1234yf + HFC-152a was carried out with a new type of gravitational capillary viscometer developed in our previous work. The experimental system was validated with pure refrigerant and mixed refrigerant. It is adequate for repeated liquid viscosity measurement of mixed refrigerants under high pressure. The liquid viscosity data of the mixture HFO-1234yf + HFC-152a (0.81 + 0.19, by mole fraction) in the phase equilibrium state from 278.15 K to 333.15 K were given, and three most commonly used viscosity models based on the Andrade equation were used to correlate the experimental results. The correlation results were compared with the experimental data, and the model with higher accuracy was recommended for the mixture.

2. EXPERIMENT

2.1 Samples

The basic information of the experimental samples is given in Table 1. They were not further purified before used in this paper.

Table 1: Experimental samples.

Sample	Chemical Name	CAS No.	Purity (mass fraction)
HCFC-22	chlorodifluorom-ethane	75-45-6	99.90 %
HFO-1234yf	2,3,3,3-Tetrafluoropropylene	745-12-1	99.50 %
HFC-134a	1,1,1,2-tetrafluoroethane	811-97-2	99.95 %
HFC-152a	1,1-difluoroethane	75-37-6	99.8%

2.2 Experimental System

The liquid viscosity experimental system used in this paper, which is shown in Figure 1, consists of a gravitational capillary viscometer made of glass (the structure of which is shown in Figure 2), a pressure vessel with sight glasses, a thermostatic bath system, and a measurement system. The major instruments used in the measurement system are listed in Table 2.

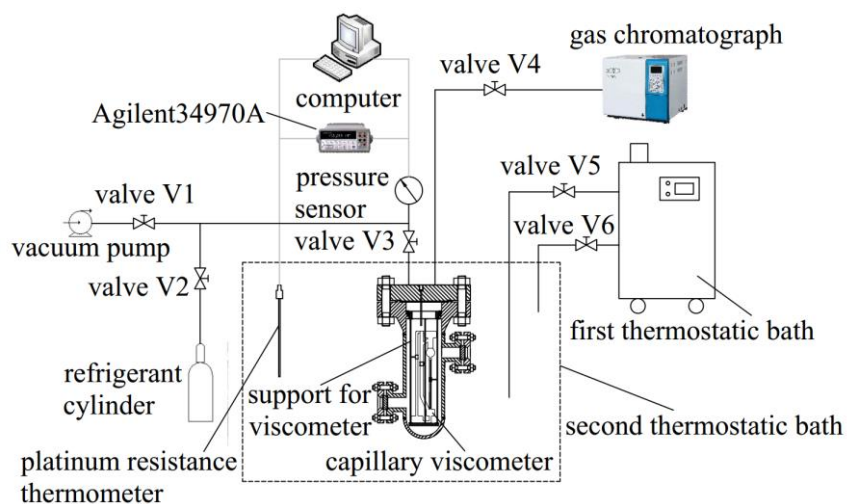


Figure 1: Schematic of viscosity experimental system.



Figure 2: Photo of gravitational capillary viscometer.

Table 2: Major equipment in the measurement system.

Equipment	Model	Range	Precision
platinum resistance thermometer	WZPB-I	/	0.001 K
pressure sensor	PMP4010	0~3.5 MPa	0.04 % F.S.
mechanical stopwatch	M504	0~15 min	0.1 s
first thermostatic bath	RTS-40T	- 40 °C ~ 95 °C	0.01 °C
gas chromatograph	GC-1690T	/	0.3 %
electronic balance	BL-5000S	0~5000 g	0.01 g

2.3 Experimental Procedure

Prior to the experiment, the capillary viscometer and pressure vessel were cleaned with concentrated sulfuric acid and potassium dichromate and rinsed with distilled water and acetone. After assembly, the vessel was evacuated and the liquid sample was then charged into the viscometer. After 30 ~ 60 minutes of thermal equilibrium, the pressure vessel was rotated in counterclockwise for 360° to return the sample in the viscometer to the upper reservoir. The liquid flowed down through the capillary due to gravity. The efflux time t through the capillary was measured by mechanical stopwatch operated manually. And then, the viscosity of the sample liquid was calculated by a modified Hagen-Poiseuille equation (Wu *et al.*, 2003) with the efflux time.

The modified Hagen-Poiseuille equation (Wu *et al.*, 2003) was shown as follows

$$v = k\alpha t - \beta/t \quad (1)$$

$$k = (\rho_L - \rho_v) / \rho_L \quad (2)$$

$$\eta = v\rho_L \quad (3)$$

where α and β are constants of equation only related to apparatus and are to be calibrated.

Considering the consistency of the efflux time, at least five sets of efflux times should be measured. Then the average value for efflux time was obtained.

2.4 Uncertainty Analysis

In the experimental system, considering the uncertainties from the platinum resistance thermometer, pressure sensor, and the digital multimeter, the expanded uncertainty of temperature is 0.011 K and expanded pressure uncertainty is

1.4 kPa for the system. Taking both of the uncertainty of mechanical stopwatch and the error brought by manual operation into consideration, 3.0 s was taken as the maximum error of efflux time. The minimum of efflux time in this paper was 189.4 s, thus, the maximum uncertainty of time is 1.58 %, which is considering as the expanded uncertainty of dynamic viscosity since it is determined by the uncertainty of time.

2.5 Calibration of Viscometer

The viscometer constants α and β in Equation (1) were determined by calibration with HCFC-22. The saturated liquid viscosity data of HCFC-22 were obtained from NIST REFPROP 9.0 (Lemmon *et al.*, 2010), the overall average absolute deviation of which was 1.09 %. The constants α and β were then obtained by regression analysis with the data in Table 3. The calibration results were that $\alpha = 0.0006305 \text{ mm}^2/\text{s}^2$ and $\beta = 0.3125 \text{ mm}^2$. Thus, the dynamic viscosity η can be calculated with Equations (1) to (3) with the values of α and β . The reliability of the experimental apparatus has been validated with HFO-1234yf and the binary mixture HFC-22 + HFC-134a (0.7 + 0.3, by mole fraction) in previous work.

2.6 Experimental Results

The liquid viscosity of the mixture HFO-1234yf + HFC-152a (0.81 + 0.19, by mole fraction) in equilibrium state was measured in the temperature range of 278.15 to 333.15 K. Dealing with the similar process with literature (Laesecke *et al.*, 2001), the mole fractions of HFO-1234yf and HFC-152a in the liquid phase were considered to be the same as the total components in the experiment. Hence, the vapor density and liquid density of the mixture could be calculated with the measured temperature and pressure data with PR EoS at the condition of $x_{\text{HFC-152a}} : x_{\text{HFO-1234yf}} = 0.19 : 0.81$. The experimental results were shown in Table 4 and Figure 3.

Table 3: Experimental data obtained by this work and liquid viscosity data of HCFC-22 from REFPROP 9.0.

T^a (K)	p^a (MPa)	ρ_L (kg·m ⁻³)	ρ_V (kg·m ⁻³)	k	t^a (s)	η_{ref} (μPa·s)
278.15	0.5841	1264.3	24.791	0.9804	263.7	204.53
283.19	0.6818	1246.6	28.852	0.9769	254.2	193.63
288.30	0.7928	1228.0	33.505	0.9727	248.5	183.15
293.23	0.9121	1209.6	38.560	0.9681	234.3	173.54
298.28	1.0476	1190.1	44.390	0.9627	229.1	164.15
303.18	1.1928	1170.6	50.746	0.9567	221.7	155.42
308.18	1.3558	1149.9	58.039	0.9495	212.0	146.87
313.24	1.5369	1128.1	66.352	0.9412	207.9	138.53
318.22	1.7321	1105.7	75.602	0.9316	204.2	130.59
323.08	1.9396	1082.7	85.790	0.9208	199.1	123.07
328.11	2.1731	1057.4	97.792	0.9075	194.2	115.47
333.12	2.4259	1030.5	111.510	0.8918	189.4	108.04

^a Standard uncertainties u are $u(T) = 0.011 \text{ K}$, $u(p) = 1.4 \text{ kPa}$, $u(t) = 1.58\%$.

Table 4: Experimental results of the mixture HFO-1234yf + HFC-152a (0.81 + 0.19, by mole fraction).

T/K	p/MPa	ρ_L	ρ_V	k	t	η
278.12	0.3793	1121.74	18.89	0.9832	280.1	193.52
283.17	0.4455	1102.71	22.14	0.9799	272.5	184.39
288.18	0.5192	1084.06	25.8	0.9762	262.9	174.13
293.13	0.6003	1064.77	29.91	0.9719	254.8	164.95
298.13	0.6912	1044.49	34.60	0.9669	245.2	154.80
303.11	0.7924	1022.00	39.95	0.9609	242.0	148.52
308.05	0.9055	999.71	45.90	0.9541	239.2	142.54
313.26	1.0348	973.91	53.04	0.9455	232.4	133.62
318.20	1.1706	948.94	60.82	0.9359	224.2	124.22
323.18	1.3214	921.26	69.70	0.9243	216.8	115.07
328.15	1.4839	892.02	79.91	0.9104	210.1	106.25
333.15	1.6659	861.35	91.67	0.8936	206.3	98.81

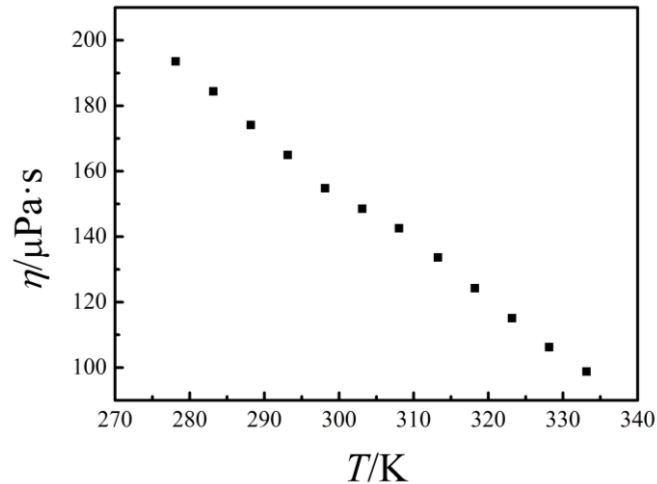


Figure 3: Experimental results of the mixture HFO-1234yf + HFC-152a (0.81 + 0.19, by mole fraction).

3. CORRELATION

There was not a theoretical method for liquid viscosity that was always applicable to all types of mixtures since the theory of liquid viscosity is quite complicated. (Herráez *et al.*, 2008; Mehrotra *et al.*, 1996) Due to this, numerous correlation and prediction models for liquid viscosity were proposed empirically or semi-empirically based on the Eyring's absolute rate theory. (Monnery *et al.*, 1995; Orbey and Sandler, 1993; Sagdeev *et al.*, 2014; Vogel and Weiss, 1981; Yaws *et al.*, 1994) Among various viscosity models, the Andrade equation (Andrade, 1930) is one of the most commonly used equations for the correlation of liquid viscosities. In order to improve the accuracy of correlation, Andrade equation was modified and introduced for calculation of liquid viscosity. In this paper, three viscosity models, which were basing on the Andrade equation, were used to correlate the viscosity data of the mixture HFO-1234yf + HFC-134a and the correlation results were compared.

One of the modified Andrade equation was expressed as follow (Sagdeev *et al.*, 2014)

$$\ln \eta = A_1 + B_1/T_r + C_1/T_r^2 \quad (4)$$

$$T_r = T / 100 \quad (5)$$

where A_1 , B_1 , and C_1 are correlating constants.

Another modification (Sagdeev *et al.*, 2013) showed the relationship between the viscosity and temperature as well as pressure

$$\ln \eta = \ln A_2 + B_2/T_r \quad (6)$$

$$\ln A_2 = a_1 + a_2P + a_3P^2 \quad (7)$$

$$B_2 = b_1 + b_2P + b_3P^2 \quad (8)$$

where a_1 , a_2 , a_3 , b_1 , b_2 , and b_3 are parameters to be correlated.

The temperature dependence of the viscosity can also be represented as the form of Equation (9) (Yaws *et al.*, 1994), which was available for the calculation of the liquid viscosity at any temperature between the melting and critical points of liquid.

$$\lg \eta = A_3 + B_3/T + C_3T + D_3T^2 \quad (9)$$

where A_3 , B_3 , C_3 , and D_3 are regression coefficients.

For the correlation of the experimental data in Table 4 with the three models introduced above, the least square method was applied for fitting an objective function which is shown below

$$OF = \frac{1}{N_p} \sum_{j=1}^{N_p} \left[(\eta_{cal} - \eta_{exp}) / \eta_{exp} \right]_j^2 \quad (10)$$

Table 5: Correlation parameters of viscosity models obtained by regression analysis.

model	A	B	C	D
$\ln \eta = A_1 + B_1/T_r + C_1/T_r^2$	-7.946	67.52	-85.68	/
	$a_1 = -19.78$	$b_1 = 59.42$		
$\ln \eta = \ln A_2 + B_2/T_r$	$a_2 = -38.27$	$b_2 = 131.2$	/	/
	$a_3 = -0.6428$	$b_3 = 7.784$		
$\lg \eta = A_3 + B_3/T + C_3T + D_3T^2$	1.061	36.14	0.01127	-2.638×10^{-5}

The correlation parameters of the three models were given in Table 5 and the correlation results were shown in Table 6. Figure 4 showed the deviation of the correlation results with the three models and Table 7 gave the values of the average absolute deviation (AAD) and the maximum absolute deviation (MAD) for the three models. From the results shown in Figure 4, Table 6, and Table 7, it can be seen that the experimental results agreed with the correlation results and the model with the form of Equations (9) had the best accuracy.

Table 6: Experimental values and correlation results with the viscosity models.

T^u (K)	p^u (MPa)	η_{exp} ($\mu\text{Pa}\cdot\text{s}$)	$\ln \eta = A_1 + B_1/T_r + C_1/T_r^2$		$\ln \eta = \ln A_2 + B_2/T_r$		$\lg \eta = A_3 + B_3/T + C_3T + D_3T^2$	
			η_{cal} ($\mu\text{Pa}\cdot\text{s}$)	$\delta \eta^b$	η_{ca} ($\mu\text{Pa}\cdot\text{s}$)	$\delta \eta^b$	η_{ca} ($\mu\text{Pa}\cdot\text{s}$)	$\delta \eta^b$
278.16	0.3815	193.52	191.42	-1.08	194.86	0.70	192.68	-0.43
283.19	0.4494	184.39	183.66	-0.4	183.78	-0.33	183.94	-0.25
288.18	0.5261	174.13	175.36	0.71	174.48	0.20	175.14	0.58
293.12	0.6086	164.95	166.78	1.11	165.85	0.55	166.40	0.88
298.15	0.704	154.80	157.91	2.01	157.13	1.51	157.56	1.79
303.15	0.809	148.52	149.02	0.34	149.29	0.52	148.80	0.19
308.15	0.925	142.54	140.27	-1.59	143.46	0.65	140.19	-1.65
313.13	1.0573	133.62	131.22	-1.80	134.1	0.36	131.25	-1.78
318.14	1.2006	124.22	122.88	-1.08	125.5	1.03	122.94	-1.03
323.16	1.3567	115.07	114.78	-0.25	117.16	1.82	114.76	-0.27
328.11	1.5261	106.25	107.04	0.74	107.28	0.96	106.84	0.55
333.12	1.7103	98.81	99.629	0.83	100.29	1.50	99.13	0.32

^a Standard uncertainties u are $u(T) = 0.011$ K, $u(p) = 1.4$ kPa.

^b $\delta \eta = 100 (\eta_{cal} - \eta_{exp}) / \eta_{exp}$

Table 7: MAD and AAD of the viscosity models.

Model	MAD/%	AAD/%
$\ln \eta = A_1 + B_1/T_r + C_1/T_r^2$	2.01	0.99
$\ln \eta = \ln A_2 + B_2/T_r$	1.82	0.84
$\lg \eta = A_3 + B_3/T + C_3T + D_3T^2$	1.79	0.81

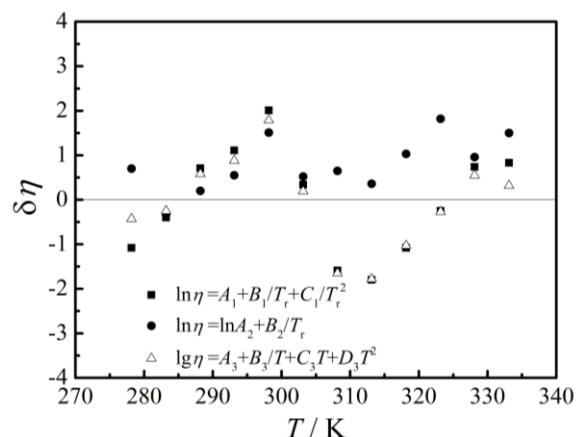


Figure 4: Deviations between correlation results and experimental data.

4. CONCLUSIONS

In this paper, a gravitational capillary viscometer was used for repeated measurement of the liquid viscosity. The gravitational capillary viscometer was firstly calibrated with HCFC-22, and then, the liquid viscosity data of the binary mixture HFO-1234yf + HFC-152a (0.81 + 0.19, by mole fraction) were given from 278.15 to 333.15 K. Three viscosity models based on the Andrade equation were used to correlate the experimental results and the correlation accuracies were compared. The comparison showed that the model with the form of Equations (9) had the best accuracy.

NOMENCLATURE

ν	kinematic viscosity	(mm ² /s)
η	dynamic viscosity	(μ Pa·s)
ρ	density	(kg/m ³)
t	efflux time	(s)
T	temperature	(K)
p	pressure	(MPa)
N_p	number	(-)

Subscript

ref	reference data
exp	experimental data
cal	calculated data
L	liquid phase
V	vapor phase

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ACKNOWLEDGEMENT

This work has been supported by the Nation Natural Science Foundation of China (Grant No. 51576171) and the Key Laboratory of Low-grade Energy Utilization Technologies and Systems (Chongqing University), Ministry of Education of China, Chongqing University, Chongqing 400044, China (No.LLEUTS-201510). The support provided for the completion of the present work are gratefully acknowledged.