科学通报 2017 年 第 62 卷 第 23 期：2691 ~ 2697
论 文

R1234ze(Z)在 243.152~373.150 K 内的饱和蒸汽压实验测量及拟合

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2016-05-19 收稿, 2016-07-06 修回, 2016-07-06 接受, 2017-04-14 网络版发表
国家自然科学基金(51376188)和航天低温推进剂技术国家重点实验室开放基金(SKLTSCP1406)资助

摘要 顺式-四氟丙烯(*cis*-1,3,3,3-Tetrafluoropropene, R1234ze(Z))作为环境友好(温室效应潜能值GWP<1, 臭氧消耗潜能值ODP=0), 热力性能优异的新型工质在制冷、热泵及有机朗肯循环等领域受到越来越多的关注. 本文采用两套高精度汽液相平衡装置测量了R1234ze(Z)在243.152~373.150 K内的63组饱和蒸汽压数据, 本文的实验数据与Wagner方程拟合偏差均小于0.001 MPa, 与REFPROP对比最大绝对偏差为0.0016 MPa, 最大相对偏差为1.07%. 结合文献数据采用Antoine方程、Wagner方程以及亥姆霍兹方程进行拟合, 并对计算值和实验值的偏差进行分析, 发现亥姆霍兹方程中起主导作用的是第2项, 并由此建立对比温度和对比压力的近似线性关系, 采用Wagner方程和最优项数为5项的亥姆霍兹方程拟合的平均绝对相对偏差分别为0.3105%和0.2813%, 后者较前者拟合精度提高11.4%.

关键词 顺式-四氟丙烯, 饱和蒸汽压, 偏差, Wagner 方程, 亥姆霍兹方程

R1234ze(Z)是近年来新开发的制冷剂，其大气寿命仅为10 d，而温室效应潜能值(global warming potential, GWP)不超过1[1]，因此R1234ze(Z)作为环境友好的替代工质在制冷、热泵及有机朗肯循环等领域受到越来越多的关注. Brown等人[2]根据基团贡献法预测了R1234ze(Z)的临界温度，临界压力以及偏心因子，其数值分别为426.8 K, 3970 kPa和0.333. Zhang等人[3]评估了含R1234ze(Z)非共沸混合制冷剂应用于制冷和高温热泵时的系统性能. 他们发现，在所有的丙烯氟化物中，R1234ze(Z)具有最高的性能系数COP，且与R114 (1, 2-二氯四氟乙烷)容积制冷量相当，尽管排气温度比R114高. Hihara[4]对R1234ze(Z)的基础热物性做了大量研究，提供了R1234ze(Z)与一些材料的相溶性，以及它的燃烧特性和毒性，除此之外，也提供了临界压力和临界温度，其值分别为3533 kPa和423.27 K，与Brown等人估计的值有一定差距. Fedele等人[5]在238.13~372.61 K温度范围内测量了R1234ze(Z)的饱和蒸汽压，确定其标准沸点和偏心因子分别为282.73 K和0.3257，但是其在273 K仅测量了8个数据，且其温度和压力的测量不确定度较大，分别为0.03 K和<1 kPa，难以满足国际物性数据库中高精度物性测量需求. Higashi等人[6]测量了R1234ze(Z)的*pvT*特性，给出了与Hihara一样的临界参数，这一参数被最新的REFPROP[7]采用. Katsuyuki[8]在高温区300.00~400.00 K内也测量了R1234ze(Z)较少的几个饱和蒸汽压.

引用格式: 卓可凡, 赵延兴, 董学强, 等. R1234ze(Z)在 243.152~373.150 K 内的饱和蒸汽压实验测量及拟合. 科学通报, 2017, 62: 2691–2697
Zhuo K F, Zhao Y X, Dong X Q, et al. Saturation pressure measurement and correlation of *cis*-1,3,3,3-Tetrafluoropropene at temperatures ranging from 243.152 to 373.150 K (in Chinese). Chin Sci Bull, 2017, 62: 2691–2697, doi: 10.1360/N972016-00615

与此同时，一些学者也研究了R1234ze(Z)在冷凝和蒸发条件下的传热特性及摩擦压降．根据Kondou等人[9]的研究，R1234ze(Z)的压降大概是R1234ze(E) (*trans*-1,3,3,3-Tetrafluoropropene)以及传统制冷剂R134a的3倍，但是其换热系数却是它们的2.6倍．Longo等人[10,11]给出了类似的结论，他们发现，R1234ze(Z)的换热系数比现在常用的制冷剂大得多，但是其摩擦压降与R600a类似．Fukuda等人[12]、Petr和Raabe[13]也暗示了R1234ze(Z)在高温热泵和ORC系统的优越性．

综上所述，R1234ze(Z)是一种非常有潜力的低GWP、低可燃性，高换热系数的环境友好制冷剂，但是目前对其热物性的研究还未完善，权威的物性数据库REFPROP也还未收录其在273 K以下的饱和蒸汽压数据．因此本文的主要工作为提供R1234ze(Z)在273 K以下以及部分高温区下的饱和蒸汽压数据，结合文献中已有数据，采用包括亥姆霍兹方程在内的多个方程进行拟合，并对计算值和实验值的偏差进行分析，给定合适的饱和蒸汽压方程．

1 实验装置与测量

本文采用两套不同的实验装置进行R1234ze(Z)的饱和蒸汽压测量，在低温区采用汽液单循环法，实验装置的介绍见文献[14]，高温区采用静态法，实验装置的介绍见文献[15]．本文只对两套实验装置的温度和压力测量系统简单介绍．

1.1 汽液单循环法

温度测量系统主要包括Pt25铂电阻温度计和测温电桥．铂电阻温度计采用云南仪表厂生产的一等标准套管铂电阻，型号为WZPB-11，出厂编号6135，由中国计量科学院根据JJG350-94标准套管铂电阻温度计检定规程检定，标准测量不确定度为2.5 mK，按照1990年国际温标分度．温度采集使用加拿大Guildline公司的6622A型自动测温电桥，出厂编号69876，由中国计量科学院检定，测量不确定度为0.02 ppm，实验中温度波动度小于±3 mK，温度测量的综合不确定度小于±5 mK．

压力测量采用美国Mensor公司生产的6000型数字压力传感器，拥有两个量程，分别为0~3和0~6 MPa，测量不确定度为0.02% FS，对应两个量程分别为0.00060和0.0012 MPa．该数字压力传感器自带线路可以直接连接计算机的RS232接口，并通过自带软件直接显示测得的压力值，分辨率为0.00001 MPa，无需额外仪表和计算机硬件，实验中压力波动度小于±0.0001 MPa，采用0~3 MPa的量程，压力测量的合成标准不确定度小于±0.0007 MPa．

1.2 静态法

温度测量系统主要包括Pt25铂电阻温度计和超级精密电阻测温仪．该温度计测量不确定度同上．该套系统以FLUKE 1594A超级精密电阻测温仪为测温设备，电阻比准确度达到0.24 ppm．电阻测温仪配有标准的计算机通讯接口，通过数据线与计算机显示，由计算机LABVIEW程序显示和保存测温仪测得的温度数据，温度测量的综合不确定度小于±5 mK．

压力测量系统主要由压力传感器和压差变送器组成．平衡釜内的高温工质进入环境易冷凝，因此其内部压力不能直接测量，而是通过其与平衡氮气的压差得到．压力传感器采用美国MENSOR公司生产的6100型数字压力传感器，用来测量氮气稳压瓶内的压力，量程为0~1.5和0~3 MPa，测量不确定度为0.01% FS．压差变送器采用美国GE公司生产的UNIK 5000型压差变送器，用来测量氮气稳压瓶和平衡釜内的压力差，温度测量范围为–40~125℃，量程为0~40 kPa，测量误差在±0.04%以内．综合考虑压力传感器和压差变送器以及测量过程中的压力波动，压力测量的合成标准不确定度小于±0.0005 MPa．

1.3 实验材料

R1234ze(Z)由北京宇极科技发展有限公司提供，其纯度不小于0.995 (体积分数)．在测定饱和蒸汽压前采用气相色谱仪进行纯度分析，未发现明显杂质，样品未再做提纯处理．

2 结果与讨论

2.1 实验数据

在243.152~373.150 K内，测量了63组R1234ze(Z)的饱和蒸汽压实验数据，其中28组用低温实验台测量，另外35组用高温实验台测量，如图1，表1和2所示．在重叠区域(288~297 K)，二者具有高度的一致性，从一个方面说明本实验测量的准确性．图1(b)给出了本文实验数据与文献中实验数据对比，所有

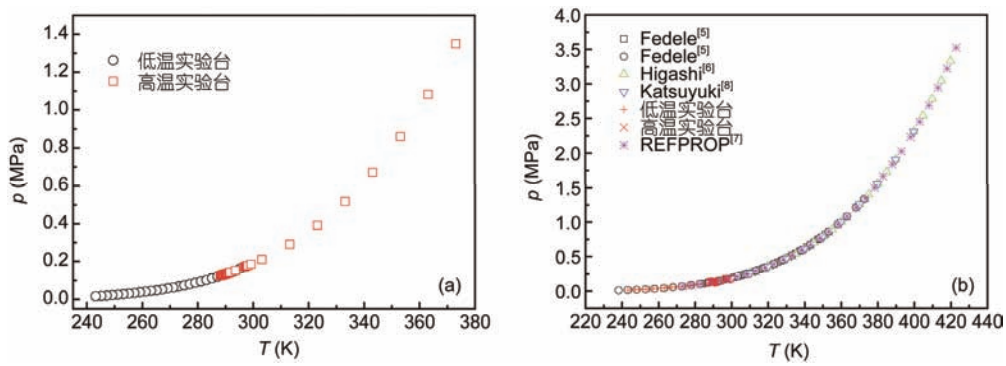

图 1 (网络版彩色)R1234ze(Z)的饱和蒸汽压. (a) 本文实验数据; (b) 本文实验数据与文献中实验数据对比

Figure 1 (Color online) The experimental saturation pressure of R1234ze(Z). (a) The data in present work; (b) the data from different literatures

表 1 由低温实验台测量的R1234ze(Z)饱和蒸汽压实验数据

Table 1 The experimental saturation pressure of R1234ze(Z) obtained by low temperature setup

T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)
243.152	0.0161	257.150	0.0338	271.152	0.0627	285.150	0.1111
245.148	0.0180	259.150	0.0372	273.150	0.0684	287.150	0.1199
247.148	0.0202	261.150	0.0410	275.150	0.0745	289.150	0.1292
249.153	0.0225	263.150	0.0450	277.150	0.0809	291.150	0.1391
251.153	0.0250	265.150	0.0483	279.150	0.0879	293.150	0.1497
253.150	0.0278	267.154	0.0529	281.150	0.0949	295.150	0.1608
255.155	0.0307	269.152	0.0576	283.150	0.1027	297.150	0.1726

表 2 由高温实验台测量的R1234ze(Z)的饱和蒸汽压实验数据

Table 2 The experimental saturation pressure of R1234ze(Z) obtained by high temperature setup

T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)	T (K)	p (MPa)
288.072	0.1243	289.682	0.1318	291.479	0.1409	303.150	0.2106
288.245	0.1251	289.757	0.1323	291.582	0.1420	313.150	0.2908
288.348	0.1254	289.978	0.1327	293.587	0.1527	323.150	0.3918
288.523	0.1264	290.062	0.1338	296.758	0.1701	333.150	0.5174
288.651	0.1268	290.256	0.1347	297.141	0.1726	343.150	0.6713
288.949	0.1282	290.506	0.1355	297.473	0.1747	353.150	0.8593
289.077	0.1291	290.569	0.1360	297.722	0.1758	363.150	1.0821
289.187	0.1296	290.765	0.1374	297.994	0.1768	373.150	1.3491
289.299	0.1303	291.132	0.1390	299.208	0.1853		

数据显示了较好的一致性.

2.2 数学模型

为了评估实验数据的准确性, 并与文献中数据进行对比, 本文采用多种方程对温度压力进行拟合. Antoine方程, Wagner方程以及亥姆霍兹方程形式如下:

Antoine方程

$$\ln p = A - B/(T + C - 273.15), \tag{1}$$

Wagner方程[16]

$$\ln p_r = (a_1\tau + a_2\tau^{1.5} + a_3\tau^3 + a_4\tau^6)/T_r, \tag{2}$$

式中, 其中临界压力和临界温度均采用文献[4]中的数据.

亥姆霍兹方程[17]

$$\ln(p_r) = \sum_{i=1}^{21} a_i (1 - T_r)^{i/2} \Big/ T_r, \tag{3}$$

式(1)~(3)中，p为压力，T为温度，$p_r=p/p_c$，$\tau=1-T_r$，$T_r=T/T_c$, A, B, C和a_i为拟合参数.

Antoine方程是一种较为简单的三参数方程，工程上常用来拟合饱和蒸汽压力，Wagner方程实质上是亥姆霍兹饱和蒸汽压方程的一种，两种方程应用都较为广泛，气液物性估算手册[18]中给出了大量物质的Antoine方程和Wagner方程的拟合参数. 亥姆霍兹方程是一种相当准确的物性方程，目前一些热力学软件中主要采用这种方程计算流体热物性，本文基于进化优化算法(evolutionary optimization method)从亥姆霍兹方程中选择最优组合项对实验数据拟合，具体算法可参见文献[19]. 在3种方程的拟合过程中，目标函数为压力的平均绝对偏差最小，即

$$\mathrm{OF}=\min\frac{1}{N}\sum_{i}^{N}\mathrm{abs}\left(p_{\mathrm{exp}}-p_{\mathrm{cal}}\right)\Big/p_{\mathrm{exp}}. \tag{4}$$

式中，p_{exp}为实验压力，p_{cal}为计算压力，N为实验点个数.

2.3 偏差分析

本文首先对测量的实验数据进行拟合，并与REFPROP数据值对比，其中REFPROP仅有273 K以上的数据，实验数据通过不同方程拟合的绝对偏差和相对偏差如图2所示. 从图中可以发现，本文的实验数据与Wagner方程拟合偏差均小于0.001 MPa; Antoine方程拟合在250 K以下偏差超过1%，但总体拟合效果也可令人满意. REFPROP与本实验数据相对偏差除两个点外都在1%以内. Antoine方程，Wagner方程以及亥姆霍兹型方程的拟合参数和平均绝对相对偏差如表3所示.

结合文献数据，本文应用Wagner方程和亥姆霍兹方程对R1234ze(Z)的饱和蒸汽压进行拟合，实验数据共157个. 图3给出了采用Wagner方程对本文及文献数据的拟合偏差，不难发现，在较高温度下(320 K以上)，不同文献的数据都有着非常好的拟合效果; 但在280 K以下，相对偏差较大. 这是因为在低温下，R1234ze(Z)的饱和蒸汽压很低(T=243.152 K，p=0.0161 MPa)，尽管绝对偏差不大(从图2(a)中可以看出，280 K以下，绝对偏差均小于0.001 MPa)，由于分母较小，导致较大的相对值. 表3还显示，对Helmholtz方程限定项数时，最优项库并不是包含关系，也就是项数为3时的最优项库并不完全包含于项数为4, 5, 6, 7的项库中. 但是在本文的优化中，不同项数的项库中总包含第2项，即$a_2(1-T_r)/T_r$项，暗示了其起主导

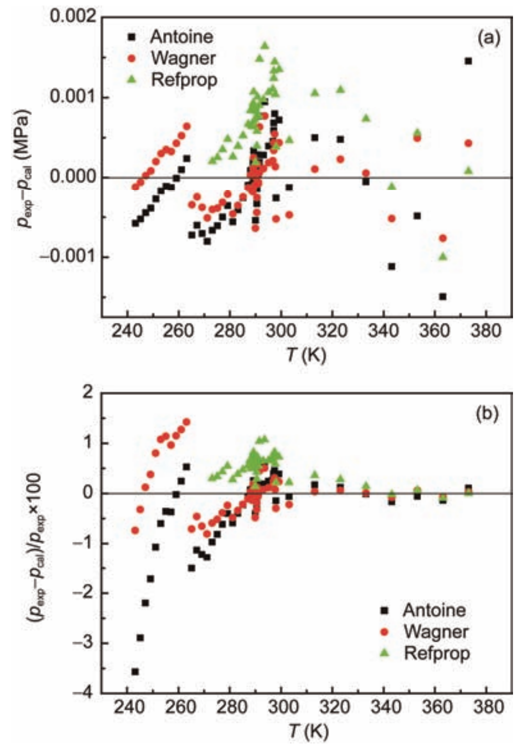

图 2 (网络版彩色)本文实验数据通过不同方程的拟合偏差. (a) 绝对偏差; (b) 相对偏差

Figure 2 (Color online) The deviations of pressures calculated by Antoine equation, Wagner equation and Refprop. (a) The absolute deviation; (b) the relative deviation

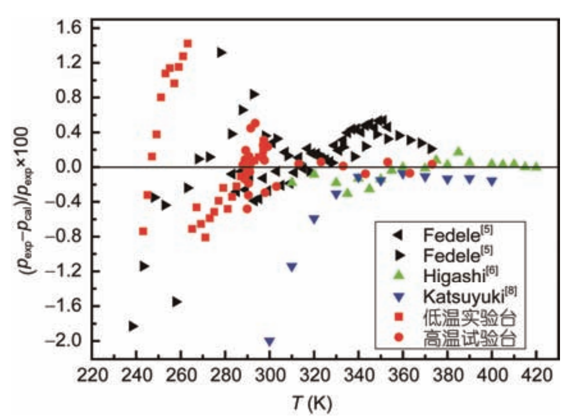

图 3 (网络版彩色)通过Wagner方程拟合不同文献实验数据偏差

Figure 3 (Color online) The relative deviation of pressure from literatures and data calculated by Wagner equation

作用，方程可改写为

$$\ln(p_r)\ \Theta\ a_2\left(1-T_r\right)\big/T_r=a_2\left(\frac{1}{T_r}-1\right). \tag{5}$$

将实验数据$\ln(p_r)$与$(1/T_r-1)$作图，如图4所示，所有的实验数据几乎都落在一条直线上，其标准偏差仅为

表 3 Helmholtz方程, Wagner方程和Antoine方程拟合参数

Table 3 The regressed parameters in Helmholtz equation, Wagner equation and Antoine equation

项数		最优项库及拟合参数						
3	i	1	2	9				
	a_i	−0.0988	−6.8639	−5.0867				
4	i	2	3	4	21			
	a_i	−7.8924	3.3578	−3.5514	−205.54			
5	i	2	3	7	20	21		
	a_i	−7.586	1.298	−5.333	4011	−5838		
6	i	2	3	5	19	20	21	
	a_i	−7.6792	1.7980	−2.4976	−15209	46831	−36302	
7	i	2	5	12	13	15	16	17
	a_i	−7.3871	7.3430	−27998	102726	−471871	731570	−344020
Wagner		a_1	a_2	a_3	a_4			
		−7.6351	1.5169	−3.5972	2.9746			
Antoine		A	B	C				
		3.33632	1117.76	248.567				

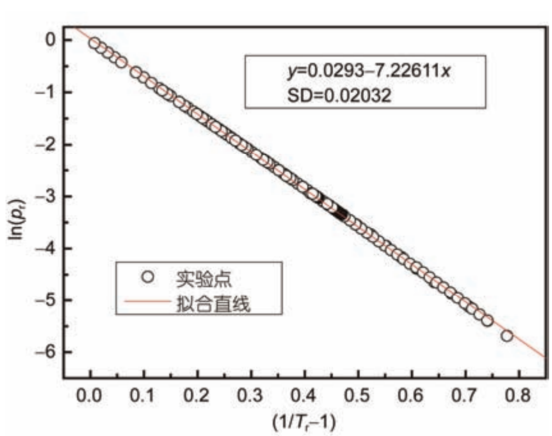

图 4 (网络版彩色)对比温度与对比压力导数的近似线性关系

Figure 4 (Color online) The reduced pressure in relation to the inverse reduced temperature

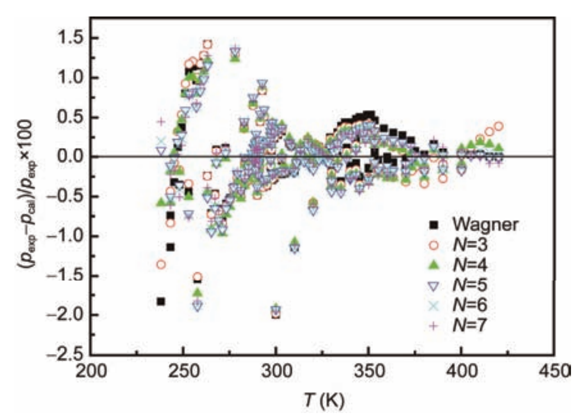

图 5 (网络版彩色)亥姆霍兹方程和Wagner方程与文献数据的压力相对偏差

Figure 5 (Color online) The relative deviation of pressure from different literatures and data calculated by Wagner equation and Helmholtz equation (N=3, 4, 5, 6, 7)

0.02032, 因此仅用一个参数也能较好地拟合并预测纯质的饱和蒸汽压力.

图5给出了亥姆霍兹方程截取不同最优项数的拟合偏差. 所有的偏差显示了几乎一致的趋势. 与上述平均绝对相对偏差的分析类似, 亥姆霍兹方程取得了与Wagner方程相当的拟合效果, 即使是仅截取3项. 由Antoine方程和Wagner方程拟合的压力平均绝对偏差分别为0.4751%和0.3105%, 由项库数为(3, 4, 5, 6, 7)的亥姆霍兹方程拟合的压力平均绝对偏差分别为0.3135%, 0.2997%, 0.2813%, 0.2815%和0.2809%. 由于亥姆霍兹方程最优项数超过5之后, 拟合的平均绝对相对偏差变化非常小, 因此本文推荐最优项数为5项的亥姆霍兹方程, 拟合精度较Wagner方程提高了11.4%, 其项库为(2, 3, 7, 20, 21).

3 结论

本文采用两套高精度汽液相平衡装置测量了R1234ze(Z)在243.152~373.150 K内的63组饱和蒸汽压数据, 两实验台在温度重叠区域具有高度一致性, 且与文献数据以及REFPROP符合良好; 结合文献中已有数据采用Antoine方程、Wagner方程以及亥姆霍

兹方程进行拟合，并对计算值和实验值的偏差进行分析发现普遍使用的Wagner方程具有较好的拟合效果；通过分析亥姆霍兹方程的最优项库发现起主导作用的是第2项，由此建立对比压力和对比温度的线性关系；亥姆霍兹方程最优项数超过3时均取得比Wagner方程更好的精度，但最优项数超过5之后，拟合效果提高不明显，因此本文推荐最优项数为5项的亥姆霍兹方程.

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Summary for "R1234ze(Z)在 243.152~373.150 K 内的饱和蒸汽压实验测量及拟合"

Saturation pressure measurement and correlation of *cis*-1,3,3,3-Tetrafluoropropene at temperatures ranging from 243.152 to 373.150 K

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R1234ze(Z) (*cis*-1,3,3,3-Tetrafluoropropene), with the 10 d atmospheric life time and <1 global warming potential (GWP) value, has been investigated in refrigeration, heat pump, organic rankine cycle and other fields in recent years. In the work, some fundamental information about R1234ze(Z) was provided. At the same time, many researchers had investigated the heat transfer characteristic and frictional pressure drop of R1234ze(Z) for both the condensation and evaporation. The heat transfer coefficients of R1234ze(Z) are much higher than those of all the other refrigerants now used in heat pump application while the frictional pressure drop of R1234ze(Z) was a little higher than that of the conventional refrigerants. In conclusion, R1234ze(Z) is a very potential low-GWP and low-flammable refrigerant with high heat transfer coefficient while its frictional pressure drop is higher than conventional refrigerants. In this work, the saturation pressures of *cis*-1,3,3,3-tetrafluoropropene were investigated using two different apparatuses at temperatures rangingfrom 243.152 K to 373.150 K. The uncertainties of the temperature and pressure were less than ± 5 mK and ± 0.0007 MPa for one setups, and were less than ± 5 mK and ± 0.0005 MPa for another. The experimental data were compared with Fedele, Higashi, Katsuyuki and Refprop 9.1. Good agreements were found among them with the maximum deviation of 0.0016 MPa, and the maximum relative deviation of 1.07% comparing to Refprop 9.1. Using Antoine equation, Wagner equation and Helmholtz equation, the expermental data were regressed, and the average absolute relative deviation of the pressure are satisfactory. The relative deviation of the pressure were higher at lower temperature than that at higher temperature, due to the very low pressure at lower temperature. In the end, the Helmholtz equation was analyzed with various number of terms to regress the experimental data. An approximate linear correlation between the reduced pressure and the inverse reduced temperature was found by analyzing the dominant factor in the terms of the Helmholtz equation. The Helmholtz equation of three terms were good enough to represent the saturated vapor pressure of R1234ze(Z), and to get the best result, five terms were recommended in Helmholtz equation. The average absolute relative deviations of the pressures calculated by Wagner equation and Helmholtz equation with five terms are 0.3105% and 0.2813%, respectively.

R1234ze(Z), saturation pressure, deviation, Wagner equation, Helmholtz equation

doi: 10.1360/N972016-00615