Thermodynamic State Surface and Cycle Analysis for Refrigerant 114*

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An equation of state for Refrigerant 114 (CClF_2 CClF_2) was formulated based on available *PVT* measurements, taking the thermodynamic consistency into consideration. The developed equation of state is expressed by the reduced Helmholtz function as a function of reduced temperature and reduced density. Derived isochoric specific heat capacity, isobaric specific heat capacity, speed of sound and vapor pressure were critically examined. The developed equation is effective for a range of temperatures 200 to 550 K and of pressures up to 20 MPa, which corresponds to the density range up to $1\,750\text{ kg/m}^3$. With the aid of the proposed equation, the cycle analysis for the heat pump, refrigeration and Rankine cycle was carried out, and it became clear that Refrigerant 114 is a suitable working fluid especially for high-temperature heat pump systems.

Key Words: Equation of State, Heat Pump Cycle, Refrigerant 114, Refrigeration Cycle, Rankine Cycle, Thermodynamics, Thermodynamic State Surface, Thermophysical Properties

1. Introduction

In order to achieve efficient use of energy, there has been an increasing interest in residential heat pump systems for high-temperature, industrial heat pump systems and organic Rankine cycle systems with a small temperature differences by using low-boiling-point refrigerants as working fluids. For the purpose of designing these systems, however, it is very important to obtain precise information on the thermophysical properties of the fluid concerned so as to select an optimum working fluid for these cycles. In most cases, information on the potential substances is very limited. 1,2-dichlorotetrafluoroethane (CClF_2 CClF_2), commonly known as Refrigerant 114 (R 114),

is considered as one of the potential substances for such purposes. In the present paper, we propose an equation of state for R 114 for a wide range of temperatures 200 to 550 K and of pressures up to 20 MPa. The theoretical cycle analysis for heat pump systems, refrigeration and organic Rankine cycle systems with the aid of the proposed equation is also reported.

2. Formulation of Equation of State

2.1 Basic idea for formulating equation of state

In the case of formulating the equation of state for SF_6 ⁽¹⁾, we proposed that attention should be paid not only to representing the available experimental *PVT* data within tolerable deviations, but also to fulfilling thermodynamic consistency among the properties calculated from the equation of state. In the present paper, the items to be considered in regard to the thermodynamic consistency were increased from two for SF_6 to five of R 114, and it was aimed to formulate a precise equation of state for R 114.

2.2 Survey of experimental investigations for thermodynamic properties

Six experimental studies of the *PVT* properties of R 114, which are shown in Table 1, have been

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Table 1 Available experimental investigations on *PVT* properties for Refrigerant 114

First Author	Year	Temperature		Pressure		Density		Sample Purity %	No. of Data Points	Ref.
		Range K	Uncertainty mK	Range MPa	Uncertainty %	Range kg/m ³	Uncertainty %			
McHarness	1951	298-444	-	0.1- 2.1	-	7- 167	-	-	17	2
Martin	1960	391-486	-	1.7- 10.9	-	131- 936	-	-	47	3
Geller	1975	323-433	-	0.9- 20.6	0.1	1134-1376	0.1	99.99	24	4
Harada	1981	283-398	50	0.4-102.4	0.2MPa	1017-1707	0.1	-	104	5
Wilson	1981	310-507	20	0.7- 10.5	0.1	6-1419	0.2	99.99	99	6
Hasegawa	1985	373-433	10	1.3- 9.1	2.0kPa	98- 984	0.09	99.94	43	7

investigated for a wide range of temperatures 287 to 507 K and of pressures up to 102 MPa. Figure 1 shows these 334 experimental data on a temperature-density diagram. For the available experimental investigations of the saturated vapor pressure, P_s , McHarness⁽²⁾ reported 8 data in a range of temperatures 195 to 413 K, Martin⁽³⁾ measured 14 data in a range 195 to 417 K, Wilson et al.⁽⁶⁾ reported 33 data in a range 291 to 415 K and Hasegawa et al.⁽⁷⁾ reported 36 data in a range 283 to 418 K. The uncertainties of these experimental data were as large as those of the *PVT* data listed in Table 1. For the isobaric specific heat capacity, c_p , Ernst⁽⁸⁾ measured with a 99.9% pure sample for a range of temperatures 293 to 353 K and of pressures up to 0.6 MPa with an uncertainty of 0.1 %, whereas Ponomareva⁽⁹⁾ measured c_p for a range of temperatures 224 to 424 K and of pressures 1 to 20 MPa with an error of about 0.5%. Concerning the available experimental data for velocity of sound, w , Takagi et al.⁽¹⁰⁾ reported w measurements for a range of temperatures 293 to 323 K and pressures up to 50 MPa with an uncertainty of about 0.3%. For saturated liquid density, ρ' , Okada et al.⁽¹¹⁾ reported ρ' measurements for temperatures 203 to 363 K with an uncertainty of 0.1%. Higashi et al.⁽¹²⁾ measured the vapor-liquid coexistence curve in the critical region and proposed the critical parameters.

2.3 Formulation of equation of state

In the process of formulating the equation of state, not only the goodness of fit to the experimental *PVT* data but also the completeness of thermodynamic consistency were carefully considered. Thermodynamic consistency was examined in the light of the following items:

- (1) behavior of isobars on the T - ρ diagram,
- (2) behavior of isobars on the c_p - T diagram,
- (3) behavior of isobars on the c_p - T diagram,
- (4) behavior of isobars on the w - T diagram, and
- (5) behavior of vapor pressure values calculated by the formulated equation of state.

The same functional form as the extended B-W-R equation for SF₆ was adopted. Since the second virial coefficients govern the behavior of the equation of state to be formulated in a lower density region, the

Fig. 1 Distribution of the available experimental *PVT*-property data for Refrigerant 114

correlation of second virial coefficients for R 114 proposed by Hasegawa⁽¹³⁾, which was devised based on the data by Bottomley⁽¹⁴⁾, was included in the equation. The coefficients of the equation of state except those corresponding to the second virial coefficients were determined by the least square fitting based on the selected *PVT* data, taking the above-mentioned items (1)-(5) into consideration. The basic *PVT* property data used for determination of the final form of the equation of state numbered 1041: 16 data by McHarness⁽²⁾, 28 data by Martin⁽³⁾, 10 data by Geller et al.⁽⁴⁾, 98 data by Wilson et al.⁽⁶⁾, 43 data by Hasegawa et al.⁽⁷⁾, 23 data of the vapor-liquid coexistence curve by Higashi et al.⁽¹²⁾ and 823 graphically-generated data in the region where only a few experimental data were available. The coefficients of the equation of state thus determined were adjusted so that the critical temperature calculated agreed with the experimental data by Higashi et al.⁽¹²⁾ within the uncertainty of the experimental data. The critical temperature, T_c , the critical density, ρ_c and the critical pressure, P_c , were calculated to be 418.77 K, 626.0 kg/m³ and 3.2558 MPa, respectively.

The equation of state was converted into a reduced form of specific Helmholtz function, f_r , and is expressed by Eq. (1).

$$\begin{aligned}
f_r = & f\rho_c/P_c = a_{00} + a_{01}T_r + a_{02}\ln T_r \\
& + a_{03}T_r\ln T_r + a_{04}T_r^2 \\
& + (RT_c\rho_c/P)T_r\ln\rho_r \\
& + \sum_{i=1}^7 (a_{i,0}T_r + a_{i,1} + a_{i,2}/T_r^2)\rho_r^i \\
& + (a_{80}/T_r^2)[((2 + \beta\rho_r^2)\exp(-\beta\rho_r^2) \\
& - 2)/(-2\beta)] \\
& + (a_{81}/T_r^2)[((3/\beta + 3\rho_r^2 + \beta\rho_r^4)\exp(-\beta\rho_r^2) \\
& - 3/\beta)/(-2\beta)] \\
& + a_{90}T_r^2\rho_r^4 + a_{91}T_r^2\rho_r^5 \quad (1)
\end{aligned}$$

where $T_r = T/T_c$, $\rho_r = \rho/\rho_c$ and R denotes the gas constant. The numerical constants of Eq.(1) are given in Table 2. The correlation of the specific heat capacity, C_p^0 , at the ideal gas state, which was essential to derive the reduced specific Helmholtz function, was formulated based on the data by Wilson cited in the Reference (15). The numerical constants a_{00} and a_{01} in Eq.(1) were so adjusted that the specific enthalpy, h , and the specific entropy, s , for the saturated liquid at 273.15 K became $h=200$ kJ/kg and $s=1$ kJ/(kg·K), respectively.

2.4 Examination of developed equation of state

Table 3 shows the comparison of the derived critical parameters with the experimental data by Higashi et al.⁽¹²⁾ The percentage deviation of the derived ρ_c value from the experimental data by Higashi et al. is about 8.7%, while the derived P_c value is almost equal to the experimental data.

The pressure and density deviations of the available experimental PVT data from Eq.(1) are shown in Figs. 2 and 3, respectively. On the 17 data by McHarness⁽²⁾, all but 2 of the pressure deviations are within $\pm 1\%$, whereas all but 1 of the density deviations are within $\pm 2\%$. On the 47 data by Martin⁽³⁾, all but 12 of the pressure deviations are within $\pm 2\%$, while all but 5 of the density deviations are within $\pm 5\%$. The pressure deviations of 24 data by Geller et al.⁽⁴⁾ are rather significant since those exist in the high-density region, but the density deviations of all are within $\pm 1\%$. On the 98 data by Wilson et al.⁽⁶⁾, all but 6 of the pressure deviations are within $\pm 2\%$, whereas all but 1 of the density deviations are within $\pm 2\%$. On the 43 data by Hasegawa et al.⁽⁷⁾, all but 1 of the pressure deviations are within $\pm 1\%$, while all but 4 of the density deviations are within $\pm 2\%$. The pressure deviations of 104 data by Harada et al.⁽⁵⁾ are large

Table 2 Numerical constants of coefficients in Eq.(1)

$a_{00}=1.123739637E+01$	$a_{50}=6.564302804E+00$
$a_{01}=-4.655306956E+01$	$a_{51}=1.787402678E+00$
$a_{02}=-9.750440622E+00$	$a_{52}=-3.986758103E+00$
$a_{03}=-5.083237604E+01$	$a_{60}=-1.557527334E+00$
$a_{04}=-9.612538273E+00$	$a_{61}=-2.003690341E+00$
$a_{10}=2.338335359E+00$	$a_{62}=6.734896923E-01$
$a_{11}=-5.166292516E+00$	$a_{70}=2.019287184E-01$
$a_{12}=-2.444485160E+00$	$a_{71}=2.668554529E-01$
$a_{20}=-2.185511094E+01$	$a_{72}=-2.490004621E-02$
$a_{21}=3.185269715E+01$	$a_{80}=-3.727082046E+00$
$a_{22}=-7.286049436E+00$	$a_{81}=-1.760703242E+01$
$a_{30}=3.909377906E+01$	$a_{90}=-1.187456330E+00$
$a_{31}=-4.934989250E+01$	$a_{91}=4.686555066E-01$
$a_{32}=1.213145412E+01$	$\beta=4.5497E-01$
$a_{40}=-2.161234339E+01$	$P_c=3.2558E+06$ Pa
$a_{41}=1.824700944E+01$	$\rho_c=6.260E+02$ kg/m ³
$a_{42}=4.815782530E+00$	$T_c=4.1877E+02$ K
$R=4.86440E+01$ J/(kg·K)	

Table 3 Comparison of the critical parameters for Refrigerant 114 calculated by Eq.(1) with the experimental data⁽¹²⁾

	Equation(1)	Experimental Data
T_c (K)	418.77	418.78 $\pm$ 0.02
ρ_c (kg/m ³)	626.0	576 $\pm$ 3
P_c (MPa)	3.2558	3.252 $\pm$ 0.004

Fig. 2 Pressure-deviation plots of the experimental PVT data from Eq.(1)

Fig. 3 Density-deviation plots of the experimental PVT data from Eq.(1)

since they exist in the high-density and high-pressure regions, but the density deviations for pressures up to 70 MPa are within $\pm 1\%$.

Based upon the reduced specific Helmholtz function given in Eq.(1), pressure, the isochoric and isobaric specific heat capacities, and the velocity of sound can be calculated by Eqs.(2) to (5), respectively.

$$P = \rho^2 (\partial f / \partial \rho)_T \quad (2)$$

$$c_v = -T (\partial^2 f / \partial T^2)_\rho \quad (3)$$

$$c_p = c_v + T / \rho^2 \{ (\partial P / \partial T)_\rho^2 / (\partial P / \partial \rho)_T \} \quad (4)$$

$$w = \sqrt{(c_p / c_v) (\partial P / \partial \rho)_T} \quad (5)$$

The temperature dependence of these thermodynamic properties thus calculated by Eqs.(2) to (5) are shown in Figs. 4 to 6. Figure 4 represents the behavior of isochoric specific heat capacity. The comparisons with the experimental data were impossible since no measured data for R 114 were reported. However, it seems that the proposed equation of state represents this property reasonably well, except in the high-density region. Although the pressure dependence of

this property due to the proposed equation of state may be less accurate at lower temperatures in the liquid region, the c_v values calculated in this region might be good enough to be used for the purpose of industrial applications. Figure 5 shows the behavior of the isobaric specific heat capacity. The experimental data at 0.1 and 0.2 MPa by Ernst⁽⁸⁾ and those along 1, 2, 3, 246, 5, 10 and 20 MPa by Ponomareva⁽⁹⁾ are also shown in Fig. 5. The isobaric specific heat capacity derived from the proposed equation of state satisfactorily represents the behavior of the experimental data. Figure 6 shows the behavior of the velocity of sound, including the experimental data at 1 and 10 MPa by Takagi et al.⁽¹⁰⁾ Although the derived velocity of sound is lower than the experimental data by about 4%, the proposed equation of state represents the trends of the experimental data and their pressure dependence reasonably well.

The deviation of the experimental saturated vapor pressure data from the correlation by Hasegawa et al.⁽¹⁶⁾ is shown in Fig. 7. The departure of the calculated values by the proposed equation of state is

Fig. 4 c_v - T diagram calculated with the aid of Eq.(1)

Fig. 6 w - T diagram calculated with the aid of Eq.(1)

Fig. 5 c_p - T diagram calculated with the aid of Eq.(1)

See Figure 1 for key.

Fig. 7 Comparison of the experimental saturated vapor pressure data with Eq.(1)

also shown in Fig. 7. The derived values from Eq. (1) agree well with the correlation by Hasegawa et al. within $\pm 1\%$, and they agree with most of the experimental data within the same order of magnitude.

2.5 Comparison with other equations of state

The equations of state proposed by Wilson et al.⁽⁶⁾ and by Kashiwagi et al.⁽¹⁷⁾ cover a range of temperatures and pressures as wide as the present equation of state. With respect to PVT properties, these two equations reproduce the experimental data as well as Eq. (1). With regard to the isobaric specific heat capacity, the equation by Wilson et al. reproduces the behavior of the experimental values for temperatures above around 270 K, although its accuracy at lower temperatures below 270 K in the liquid region is reduced. The equation by Kashiwagi et al., however, represents their behavior reasonably well. The maximum isobaric specific heat capacity values along the isobars on a c_p - T diagram calculated by these two equations of state are lower than the derived values from Eq. (1). The velocity of sound calculated using the equation by Wilson et al. is greater than the experimental data by Takagi et al. by about 10 to 20% in the high-density liquid region. The equation by Kashiwagi et al. reproduces the velocity of sound in the liquid region below 400 K with reduced accuracy, and its calculated pressure dependence seems less reliable. Simply because of the fact that no experimental isochoric specific heat capacity was available, it was impossible to examine the reliability of these equations of state in detail. However, these two equations of state reduce their accuracy in representing the isochoric specific heat capacity at lower temperatures in the liquid region similarly to the case of the present equation of state.

For saturated vapor pressures, the deviations of the calculated values by these equations of state from the correlation proposed by Hasegawa et al. are shown in Fig. 7. The equation by Wilson et al. does not reproduce the behavior of the measured vapor pressure data, whereas the equation by Kashiwagi et al. reproduces its behavior as well as Eq. (1).

2.6 Effective region of the proposed equation of state

The proposed equation of state, Eq. (1), reproduces the experimental data of various thermodynamic properties satisfactorily, as mentioned in section 2.4, and the thermodynamic consistency has been confirmed as shown in Figs. 4 to 7. It is not so easy to specify clearly the effective range of the present equation of the state, but, taking into consideration the range of available experimental data and the behavior of the thermodynamic properties calculated, it seems reasonable to conclude that the present equa-

tion of state might be effective for a range of temperatures 200 to 550 K and of pressures up to 20 MPa which corresponds to the density range of 0 to 1750 kg/m³.

3. Consideration of Cycle Efficiency

With the aid of the proposed equation of state for R 114, the cycle analysis of the theoretical refrigeration cycle and organic Rankine cycle is discussed exclusively in the light of the thermodynamic properties of working fluids.

3.1 Refrigeration cycle

A theoretical vapor-compression refrigeration cycle and heat pump cycle using a compressor and an expansion valve were discussed under the assumed conditions including condensing temperature, T_{con} , and evaporating temperature, T_{eva} , given in Table 4. Concerning the common conditions, the degree of super-cooling was assigned as 5 K, and the state at the compressor outlet was assumed to that of dry saturated vapor. The condition of cycle 1 in Table 4 assumes a basic refrigeration system, cycle 2 assumes a heat pump system for colder climate districts and the condition of cycle 3 assumes an industrial heat pump system for obtaining high-temperature output using waste heat. The thermodynamic processes of the respective cycles were composed of isentropic compression - isobaric condensation - isenthalpic expansion - isobaric evaporation. The analysis for these cycles in Table 4 were carried out so as to satisfy the above conditions with the aid of the present equation of state, Eq. (1). The results including compressor inlet pressure, P_1 , outlet pressure, P_2 , compression ratio, P_2/P_1 , degree of superheating at the evaporator outlet, ΔT , refrigeration effect for the evaporator (for cycle 1) or heat of rejection at the condenser (for cycle 2, 3), Q , and the theoretical coefficient of performance, COP, are summarized in Table 5. The theoretical coefficient of performance of the refrigeration cycle, the cycle 1, is almost equal to our previous results in the case of using R 12 or R 22, that is 4.89 and 4.84⁽¹⁸⁾, respectively, which are the most conventional working fluids for refrigerators. However, some problems in the cycle design exist because P_1 becomes less than the atmospheric pressure when R 114 is selected as the working fluid. Comparing the cycle 2 and cycle 3 for heat pump systems, it becomes clear

Table 4 Working conditions of the refrigeration cycles

No.	Cycle	T_{con} (K)	T_{eva} (K)
1	Refrigeration	303.15	258.15
2	Heat pump	323.15	263.15
3	Heat pump	393.15	333.15

Table 5 Cycle analysis of the refrigeration cycles using Refrigerant 114

No.	P_1 (MPa)	P_2 (MPa)	P_2/P_1	ΔT (K)	Q (kJ/kg)	COP
1	0.0467	0.249	5.34	6.94	108	4.91
2	0.0581	0.444	7.64	11.1	122	4.42
3	0.576	2.08	3.60	10.5	75.6	4.39

Table 6 Working conditions of organic Rankine cycles

No.	T_{eva} (K)	T_{con} (K)
1	373.15	302.15
2	393.15	302.15

Table 7 Cycle analysis of organic Rankine cycles using Refrigerant 114

No.	P_1 (MPa)	P_2 (MPa)	P_1/P_2	T_2 (K)	η (%)
1	1.42	0.241	5.87	317.96	15.0
2	2.08	0.241	8.60	320.08	17.2

Fig. 8 Coefficient of performance for heat pump cycle as a function of condensing and evaporating temperature

that the respective coefficients of performance are very similar to each other, although the temperature level increases by about 60 K from cycle 2 to 1. The calculated coefficients of performance for the heat pump cycle with varying condensing temperatures and evaporating temperatures are shown in Fig. 8. It is seen that the maximum COP is obtained under the condition that the condensing temperature is about 360 to 380 K. It also becomes clear that R 114 is a suitable working fluid, especially for high-temperature heat pump systems. The maximum coefficient of performance in the case of using R 114, as investigated by Enjo et al.⁽¹⁹⁾, was obtained under the condition that the condensing temperature was 373 K when the temperature difference between condensing and evaporating temperatures was 50 K. On the other hand, the maximum coefficient of performance with R 114 due to the present equation of state was obtained under the condition that the condensing temperature was 367 K.

3.2 Organic Rankine cycle

The geothermal power generation cycle driven by

Fig. 9 Cycle efficiency of organic Rankine cycle systems as a function of evaporating temperature ($T_{con} = 302.15$ K)

a turbine as organic Rankine cycle using R 114 was also investigated. The corresponding evaporating temperatures, T_{eva} , and condensing temperatures, T_{con} , are given in Table 6. The cycle process was assumed to be composed of isentropic expansion - isobaric condensation - isentropic expansion - isobaric evaporation, and the turbine inlet condition was chosen to be the dry saturated vapor. The analysis of the cycles given in Table 6 was carried out so as to satisfy the above conditions with the aid of the present equation of state. The results concerning the turbine inlet pressure, P_1 , turbine outlet pressure, P_2 , expansion ratio, P_1/P_2 , turbine outlet temperature, T_2 , and theoretical cycle efficiency, η , are summarized in Table 7. Moreover, taking into consideration the relation between the temperature levels and cycle efficiencies, the calculated cycle efficiencies are depicted in Fig. 9. It was thus confirmed that the maximum efficiency was obtained when T_{eva} was kept near the critical temperature.

4. Conclusions

In order to construct the thermodynamic state surface of Refrigerant 114, we have formulated the equation of state for a range of temperatures 200 to 550 K and of pressures up to 20 MPa, which corresponds to the density range up to 1750 kg/m³. The developed equation of state is expressed by the reduced Helmholtz function as a function of temperature and density. The derived *PVT* properties, isochoric and isobaric specific heat capacities, speed of sound, and saturated vapor pressure were critically examined. In addition, with the aid of the proposed equation, the thermodynamic cycle analysis for the heat pump cycle, refrigeration cycle and organic Rankine cycle was carried out, and it became clear that Refrigerant 114 is a suitable working fluid, especially for high-temperature heat pump systems.

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