Simultaneous optimization of saturation equations for two hydrocarbons and four hydrofluoroolefins refrigerants

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Abstract. This research exhibits a different simultaneous optimization for saturation equations of six pure refrigerants from Hydrocarbon (HCs) and Hydrofluoroolefins (HFOs). The study objective is to model two different group of saturation equations in accordance with the same structural forms, one for n-butane (R600) and isobutane (R600a), and other for 2,3,3,3-tetrafluoropropan (R1234yf), trans-1,3,3,3-tetrafluoropropene (R1234ze(E)), cis-1,3,3,3-tetrafluoropropene (R1234ze(Z)), and trans-1-chloro-3,3,3-trifluoropropene (R1233zd(E)) refrigerants. Genetic algorithm with weighted least square regression was assigned to carry out the research objectives. The modeling equations for both groups are valid from triple points up to temperature nearby the critical points. The uncertainties respecting to each equation of R600, R600a, R1234yf, R1234ze(E), R1234ze(Z), and R1233zd(E) refrigerants are estimated to be 0.05%, 0.75%, 0.10%, 0.35%, 0.57%, and 0.22% in saturated liquid density; 0.30%, 2.68%, 0.86%, 1.48%, 4.13%, and 1.23% in saturated vapor density; and 0.73%, 2.15%, 0.08%, 0.47%, 0.97%, and 2.85% in vapor-pressures, respectively. The new ancillary equations can be used to calculate the saturation properties for engineering interests.

1. Introduction

Hydrocarbons (HCs) and hydrofluoroolefins (HFOs) have notable interest in scientific and industrial sectors. Regarding to the global warming issues, HCs and HFOs refrigerants are considered as an environment substance due to their zero-ozone depleting potential (ODP) and very small global warming potential (GWP). Saturation properties, as an important thermodynamic property of refrigerants are required for the precision of calculation of the cycle simulation and optimal design of the thermal systems such refrigerator, heat pump, and power systems. Saturation properties have also been widely used in calculating the non-dimensional parameters of heat and mass transfer analysis such as Bond number and Weber number in boiling and convection heat transfer calculation, Reynolds number in flow characteristic calculation, friction factor in pressures drop calculation and so on. On the other hand, saturation properties can be used for evaluating performance of equation of state (EOS) such as Helmholtz energy EOS and cubic EOS. The most reliable and accurate thermodynamic properties play important role in finding prospective candidate working fluids for engineering interests. Accurate analysis result for system design contributes in improvement of system efficiency and energy saving. High performance systems with ozone-friendly and negligible GWP refrigerant are expected to
improve the global warming environmental issue. Therefore, it is very important to develop reliable saturation equations by applying different simultaneous optimization to get the same structural form equations for the four HFOs refrigerants consisting of R1234yf, R1234ze(E), R1234ze(Z), and R1233zd(E), and other two HCs refrigerants consisting of R600, R600a.

2. Data preparation and model development

2.1. Data selection for modelling

Experimental data are very important for property modeling of saturation equations. Assessment of all experimental input data of the HCs and HFOs were carefully observed through the existing experimental data to find the reliable data set for simultaneous optimizations. All data selections were prepared in the same standard unit system and scale. Data with older temperatures standard than ITS-90 were converted into ITS-90. All experimental input data of the HCs and HFOs refrigerants are summarized in table 1.

| Fluids         | Sources        | Selected Points | Fluids         | Sources        | Selected Points |
|----------------|----------------|-----------------|----------------|----------------|----------------|
| R1234yf        | Tanaka[1]      | 11              | R600a          | Lim[27]        | 8              |
|                | Zhao[2]        | - 10            |                | Lim[28]        | 4              |
| R1234Ze(E)     | Gong[3]        | 10              |                | Martinez[29]   | 9              |
|                | Mark[4]        | 26              |                | Morris[30]     | 2              |
|                | Nicolas[5]     | 78              |                | Sage[31]       | 8              |
|                | Tanaka[6]      | 18              |                | Waxman[32]     | 16             |
|                | Yin[7]         | 15              |                | Weber[33]      | 13             |
|                | Gao[8]         | 12              |                | Kayukawa[34]   | 8              |
|                | Higashi[9]     | - 7             |                | Liu[35]        | 86             |
|                |                | 14              |                | Glos[36]       | 20             |
| R1234Ze(Z)     | Tanaka[1]      | 22              |                | Sliwinski[37]  | - 10           |
|                | Fedele[10]     | 64              |                | McClune[38]    | 11             |
|                | Higashi[11]    | 19              |                | Haynes[39]     | 12             |
|                |                | 9 10            |                | Orrit[40]      | 36             |
| R1233zd(E)     | Hulse[12]      | 16              |                | Benolie[41]    | 9              |
|                | Raabe[13]      | - 8             |                |                |                |
|                | Cui[14]        | - 12 12         |                |                |                |
| R1233zd(E)     | Kondou[15]     | - 10 9          |                | Flebbe[42]     | 12             |
|                | Kondou[16]     | - - 7           |                | Kratzke[43]    | 12             |
| R600a          | Beattie[17]    | 5 5             |                | Magee[44]      | - 100          |
|                | Connolly[18]   | 6 -             |                | Sako[45]       | 22             |
|                | Miyamoto[19]   | 25 -            |                | Holldorff[46]  | 20             |
|                | Higashi[20]    | 5 9 11          |                | Glos[36]       | 20 23         |
|                | Gilliland[21]  | 5 -             |                | Machin[47]     | 224            |
|                | Hipkin[22]     | 5 -             |                | Sliwinski[37]  | - 9            |
|                | Hirata[23]     | 5 -             |                | Orrit[39]      | - 50           |
|                | Kahre[24]      | - 5             |                | Olds[48]       | 4              |
|                | Lee[25]        | 8 -             |                | Haynes[39]     | - 12           |
|                | Leu[26]        | 5 -             |                | Kay[49]        | 19 19         |

Ps: Vapor Pressure [MPa]; ρ': Saturated Liquid Density [kg.m⁻³]; ρ'' : Saturated Liquid Density [kg.m⁻³]
2.2. Modelling frameworks

Different simultaneous optimization for two of HCs and other four of HFOs refrigerants are conducted for the same structural form of saturation equations by using genetic algorithm with weighted least square regression method. The optimization criterion of this method is called as fitness, which is sum of weighted least squares for the mathematical model was fitted. The main paradigm of simultaneous optimization is shown in figure 1. Normally, the modeling process start with trial structural forms of the equation to find better structure for each of studied fluids. Not only functional form that has the influence on model quality, but weighting factor has significant effects also. In the case, advancement by iteration of weighting factors may be assigned to accomplish desired deviations of the models from the experimental input data.

![Figure 1. Main paradigm optimization [50].](image)

3. Saturation equations

Saturation equations of two HCs and four HFOs refrigerants were differently simultaneous established. The equations of the HFOs consist of 3 terms and have the same functional form. The equations of the HCs consist of 6, 5, and 3 terms for saturated liquid density, saturated vapor density, and vapor pressure equations, respectively. Each equation group has the same functional form with the general form of ancillary equations as given in Eqs.(1)-(3). The numerical coefficients and functional constants for each refrigerant are listed in table 2.

\[
\ln \left( \frac{\rho}{\rho_c} \right) = \sum_i A_i \left( \frac{T}{T_c} \right)^{t_i} \\
\frac{\rho}{\rho_c} - 1 = \sum_i B_i \left( \frac{T}{T_c} \right)^{t_i} \\
\ln \left( \frac{\rho}{\rho_c} \right) = \sum_i C_i \left( \frac{T}{T_c} \right)^{t_i}
\]  

(1) 

(2) 

(3)
Table 2. Numerical coefficients and structural forms of equations.

| Fluids | HC | R600 | R600a | HFO | R1234yf | R1234ze(E) | R1234ze(Z) | R1233zd(E) |
|--------|----|------|-------|-----|---------|------------|------------|-----------|
|        | ![Equation](eq1) | ![Equation](eq2) | ![Equation](eq3) |     |
| i      | t_i | A_i  | A_i   | t_i | A_i    | A_i        | A_i        | A_i       |
| 1      | 1.11 | -18.104 | -17.849 | 0.766 | -0.6315 | -0.6132    | -0.6995    | -0.4697   |
| 2      | 1.20 | 12.4259 | 12.257  | 1.016 | -6.0585 | -6.2635    | -6.2247    | -6.3603   |
| 3      | 3.50 | -3.3681 | -3.366  | 4.146 | -3.6479 | -4.6392    | -4.2351    | -5.0158   |
| ![Equation](eq2) |     | ![Equation](eq2) |     | ![Equation](eq2) |     |
| i      | t_i | B_i  | B_i   | t_i | B_i    | B_i        | B_i        | B_i       |
| 1      | 0.15 | 4.3635 | -0.4270 | 0.15 | 0.1110 | 0.1507     | 0.0504     | 0.0200    |
| 2      | 0.25 | -15.855 | 1.8797  | 0.4  | 2.2920 | 2.2616     | 2.4272     | 2.4555    |
| 3      | 0.41 | 38.155  | -0.0164 | 1.641 | 0.4620 | 0.4374     | 0.4759     | 0.3373    |
| ![Equation](eq3) |     | ![Equation](eq3) |     | ![Equation](eq3) |     |
| i      | t_i | C_i  | C_i   | t_i | C_i    | C_i        | C_i        | C_i       |
| 1      | 0.11 | -0.13369 | -0.1323 | 0.37 | -2.8147 | -2.8108    | -3.0410    | -2.7704   |
| 2      | 0.537 | -4.55516 | -4.5684 | 1.37 | -9.6775 | -10.1414   | -9.8409    | -10.073   |
| 3      | 2.025 | -11.8311 | -11.546 | 4.73 | -51.7094 | -52.4962   | -58.979    | -53.1901  |
| 4      | 5    | -31.6358 | -31.172  | -    | -      | -          | -          | -         |
| 5      | 9.375 | -61.0603 | -62.210  | -    | -      | -          | -          | -         |
| Main Parameters |
| $P_c$ | 3.796 | 3.628 | 3.3822 | 3.632 | 3.533 | 3.573 |
| $T_c$ | 425.3 | 407.79 | 367.85 | 382.51 | 423.27 | 439.6 |
| $\rho_c$ | 225.3 | 229 | 475.55 | 486 | 470.62 | 480.23 |
| $T_e$ | 134.9 | 113.73 | 220 | 168.62 | 273 | 195.15 |

* $P_c$: Critical Pressure [MPa]; $T_c$: Critical Temperatures[K]; $\rho_c$: Critical Density [kg.m$^{-3}$]; $T_e$: triple point temperature[K].

where $A_i$, $B_i$, $C_i$ and $t_i$ are numerical coefficients and predicted variable, respectively.

4. Assessment and discussion

The deviations of experimental data of vapor-pressures from the vapor pressure equation of HFOs and HC refrigerants are shown in figure 2. The experimental vapor-pressure data of R1234yf are represented within ±0.2%, which are corresponding to the temperatures nearby triple point up to the critical point. Most of experimental data of R1234ze(E) can be reproduced with deviations less than 0.5%, except some data points at lower temperatures. The experimental data of R1234ze(Z) can be accurately represented with deviation less than 1% in the temperature range from 280 K up to near critical points, except some data at lower temperatures (±1.5%). The deviation of data used for R1233zd(E) can be represented within ±2.5%, except 4 data points at temperatures around 300 K. The data for R600 can be exhibited with deviations less than 0.5% from lower temperatures up to higher temperatures, while the data from Magee et al. [44] and Machin et al. [47] can be represented within ±1.6%. Most of experimental data of R600a can be reproduced with deviations less than 0.5%, except some data points of Higashi et al. [20] and Weber et al. [33]. All data calculated from existing EOS are also plotted for the HC and HFO refrigerants.

For saturated liquid density equations, the deviations of measurement data from the equations of HFOs and HC refrigerants are illustrated in figure 3. The experimental data of R1234yf can be...
accurately represented within ±0.05% in the temperature range between 280 K up to 350 K, while region nearby critical point has a maximum deviation of 0.27%. The data of existing models of R1234yf are also shown in the figure 3. Most of the deviations of data used for R1234ze(E) can be reproduced with good agreement within ±0.06%, except some data points nearby the critical temperatures have deviations higher than 0.4%. Some data points of R1234ze(Z) have deviations within ±0.06% and other data points have (±0.1%). Three data points at higher temperatures have deviations higher than 1%. The data calculated from EOS model of Akasaka et al. [51] have good deviations less than 0.5%. All the experimental data used in modeling of R1233zd(E) can be represented with deviations less than ±0.5%. The saturated liquid density of R600 refrigerants can be accurately reproduced with deviations of 0.0001% up to 0.11%, while the data of R600a can be represented with deviations less than 0.1%.

For the saturated vapor density equations, the experimental data of saturated vapor density from the equations of HFOs and HCs refrigerants are plotted in figure 4. The experimental data of R1234yf can be reproduced within ±0.3% in the temperature range between 293 K up to temperatures 358 K, while region nearby critical point has a maximum deviation of 1.79%. The existing models of R1234yf are also plotted. Most of the experimental data of R1234ze(E) can be represented with deviations within ±0.9%, except some data points have deviations higher than ±2% up to ±3.4%. The experimental data of R1234ze(Z) have deviations within ±0.6% and some data points at higher temperatures are off-scale from the criteria. The data calculated from model of Akasaka et al. [51] have good deviations. Most of experimental data of Cui et al. [14] and Kondou et al. [15-16] used in model of R1233zd(E) can be represented with deviations within ±0.3%, except the data of Raabe et al. [13]. The saturated liquid density of R600a refrigerants can be accurately reproduced with deviations less than ±0.5%, except the data of Sage et al. [31] and some data point nearby the critical temperatures. On the other hand, some experimental data of R600 can be represented with deviations less than 0.5%, except data of Kay et al. [49].

Figure 2. Deviations of $P_s$ from new equation. (▲) Tanaka et al.[1]; (×) Gong et al.[3]; (⁎) Mark et al.[4]; (■) Nicolas et al.[5]; (+) Tanaka et al.[6]; (○) Yin et al.[7]; Fedele et al.[10]; (●) Higashi et al.[11]; (×,⁎) Hulse et al. [12]; (♦) Beattie et al.[17]; (×) Miyamoto et al.[19]; (●) Higashi et al.[20]; (●) Hipkin et al.[22]; (●) Hirata et al.[23]; (●) Lee et al.[25]; (●) Lim et al.[27]; (●) Lim et al.[28]; (●) Martinez et al.[29]; (●) Morris et al.[30]; (+) Sage et al.[31]; (●) Waxman et al.[32]; (▲) Weber et al.[33]; (○) Kuyukawa et al.[34]; (+) Liu et al.[35]; (■) Glos et al.[36]; (+) Fieble et al.[42]; (→) Kratze et al.[43]; (●) Magee et al.[44]; (+) Hollorolf et al.[46]; (●) Machin et al.[47]; (○) Kay et al.[49]; (→) Akasaka et al. [51]; (―) Miyamoto et al.[52]; (…) Sarin and Astina [53]; (→) Richter et al.[54]; (→) Akasaka et al.[55]; (→) Mondejar et al.[56]; (→) McLinden et al.[57]; (→) Thol et al.[58]; (●) Miyamoto et al. [59].
Figure 3. Deviations of $\rho'$ from the new equations. (♦) Zhao et al.[2]; (♦) Gong et al.[3]; (•,▲) Tanaka et al.[6]; (●) Higashi et al.[9]; (●) Higashi et al.[11]; (●) Hulse et al.[12]; (×) Rabbe et al.[13]; (▲) Cui et al.[14]; (●) Kondou et al.[15]; (●) Kondou et al.[16]; (○) Hulse et al.[20]; (─) Kahre[24]; (▲) Morris et al.[30]; (×) Sage et al.[31]; (+) Kayukawa et al.[34]; (-,×) Glos et al.[36]; (●,◊) Sliwinski et al.[37]; (●,★) Haynes et al.[39]; (+,○) Orrit et al.[40]; (●) Benoleil et al.[41]; (●,•) McClune et al.[38]; (●) Olds et al.[48]; (─,--) Akasaka et al.[51]; (─) Miyamoto et al.[52]; (-----) Sarin and Astina[53]; (----) Richter et al.[54]; (-----) Akasaka et al.[55]; (－,----) Mondejar et al.[56]; (-----) McLinden et al.[57]; (-----) Thol et al.[58]; (-----) Miyamoto et al.[59]; (-----) Younglove and McLinden[60].

Figure 4. Deviations of $\rho''$ from the new equations. (▲) Zhao et al.[2]; (×) Higashi et al.[9]; (●) Higashi et al.[11]; (○) Hulse et al.[12]; (×) Rabbe et al.[13]; (▲) Cui et al.[14]; (●) Kondou et al.[15]; (●) Kondou et al.[16]; (○) Higashi et al.[20]; (●) Waxman et al.[32]; (▲,□) Glos et al.[36]; (●,◊) Sliwinski et al.[37]; (●) Olds et al.[48]; (-----) Akasaka et al.[51]; (-----) Miyamoto et al.[52]; (-----) Sarin and Astina[53]; (-----) Richter et al.[54]; (-----) Akasaka et al.[55]; (-----) Mondejar et al.[56]; (-----) McLinden et al.[57]; (-----) Thol et al.[58]; (-----) Miyamoto et al.[59]; (-----) Younglove and McLinden[60].
5. Conclusion
New saturation equations for saturation thermodynamic properties of two HCs (R600 and R600a) and four HFOs (R1234yf, R1234ze(E), R1234ze(Z), and R1233zd(E)) refrigerants that are valid for triple point temperatures up to temperatures nearby critical points have been simultaneously developed. The uncertainties respecting to different equations of R600, R600a, R1234yf, R1234ze(E), R1234ze(Z), and R1233zd(E) refrigerants are estimated to be 0.05%, 0.75%, 0.10%, 0.35%, 0.57%, and 0.22% in saturated liquid density; 0.30%, 2.68%, 0.86%, 1.48%, 4.13%, and 1.23% in saturated vapor density; and 0.73%, 2.15%, 0.08%, 0.47%, 0.97%, and 2.85% in vapor-pressures, respectively. The reliable and accurate experimental saturation property data of the HFOs and HCs are satisfactorily reproduced, except saturated vapor density data of R1233ze(E) and vapor-pressures data of R1233zd(E) that have lower accuracy.

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