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G. Artico

National Research Council Institute of Refrigeration

S. Bobbo

National Research Council Institute of Refrigeration

L. Fedele

National Research Council Institute of Refrigeration

R. Camporese

National Research Council Institute of Refrigeration

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REPRESENTATION OF THE BOUNDARY CONDITIONS FOR REFRIGERANT MIXTURES WITH THE LEMMON-JACOBSEN EQUATION OF STATE

*Artico Gianpiero, Bobbo Sergio, Fedele Laura, Camporese Roberto
National Research Council, Institute of Refrigeration.
Corso Stati Uniti 4 I-35127 Padua, Italy; Tel.: +39.049.8295736; Fax: +39.049.8295728
E-Mail: gianpiero.artico@itef.pd.cnr.it *Author for Correspondence

ABSTRACT

Vapour-liquid equilibria (VLE) data are within the most important experimental data to correctly represent the mixtures' behaviour at the saturation boundaries. In the last years, the VLE of several new binary mixtures have been studied by our group in order to find possible alternatives for the traditional refrigerants. The data have been correlated with the Lemmon-Jacobsen model, which is the base for the Refprop 6.0 database, by regressing the interaction parameters used in the model mixing rules. The results obtained and a comparison with the calculation performed with the Refprop 6.0, where ideal parameters are adopted for the mixtures here considered, show the importance to define the interaction parameters on the base of experimental data to properly represent the boundary conditions. The influence of a proper choice of the interaction parameters in the calculation of the performance (COP and volumetric refrigerating capacity) of the mixtures as working fluids in ideal refrigerating cycles is shown also.

NOMENCLATURE

| | |
|---|--|
| <p><i>A</i>: Dimensional Helmholtz energy <i>a</i>: Adimensional Helmholtz energy <i>AAD</i>: Absolute Average Deviation <i>CFC</i>: Chlorofluorocarbons <i>COP</i>: Coefficient of Performance <i>C_p</i>: Isobaric specific heat <i>L-J</i>: correlative L-J model <i>H</i>: Enthalpy <i>HFC</i>: Hydrofluorocarbons <i>HC</i>: Hydrocarbons <i>k_i, k_v, F_{ij}, β, γ</i>: Interaction parameters <i>N_k, d_k, t_k</i>: Coefficients of excess Helmholtz energy</p> | <p><i>M_k, i_k, j_k, ζ_k, l_k</i>: Coefficients of pure fluid equation <i>P</i>: Pressure <i>R</i>: Universal gas constant <i>RP6</i>: REFPROP 6.0 <i>S</i>: Entropy <i>T</i>: Temperature <i>VE</i>: Volumetric Efficiency <i>x, y</i>: liquid and vapour mole fraction <i>w</i>: mass fraction <i>ρ</i>: Density of saturated vapour <i>τ</i>: Reduced temperature <i>δ</i>: Reduced volume</p> |
|---|--|

INTRODUCTION

Several systems of hydrocarbons and hydro fluorocarbons have been studied in the last years by our group, in order to increase the knowledge on the thermodynamic properties of working fluids that might be considered as substitutes for CFCs in refrigeration, air-conditioning and heat pump applications. In particular, vapor-liquid equilibrium measurements for binary mixtures have been performed.

As it is well known, the experimental data must be correlated through suitable thermodynamic models to calculate the main properties of these new systems. In the last years, the database REFPROP has become more and more popular in the field of HVAC&R, and is now considered as a sort of standard for the calculation of the thermodynamic and transport properties of the new refrigerants. The last version of this software (REFPROP 6.0) [1] is based on a Free Helmholtz Energy mixture model developed by Lemmon and Jacobsen [2,3,4], which allows the calculation of the thermodynamic properties by simple partial derivation with a greater accuracy than with the classic cubic EoSs. The model can work either in predictive or in correlative mode using up to five interaction parameters. The best results are obviously achieved in correlative mode, but for several mixtures the parameters implemented in the REFPROP database are not obtained on the base of experimental data. This is the case for almost all the systems we have studied in the last years. This reduces considerably the reliability of the calculations

and can mislead on both the selection of proper alternatives to the traditional fluids and the design of the refrigerating plants components when these mixtures are considered.

In this work, the interaction parameters for the Lemmon-Jacobsen (L-J) mixture model have been regressed on the base of the experimental data for four selected mixtures, representative of different families of mixtures in terms of molecular interactions. The corresponding boundary conditions have been compared with those given by the REFPROP 6.0 database. Moreover, the influence of the interaction parameters on the calculation of the ideal refrigerating cycles performance (COP and volumetric refrigerating capacity) is shown.

THE LEMMON-JACOBSEN MIXTURE MODEL

The L-J mixture model introduced in the last years a new approach in the application of equation of state to fluid mixtures. It is based on a generalized Helmholtz energy equation expressed as a sum of the ideal contribution of the pure compounds and an excess contribution to take into account the effects of mixing. This model is particularly suitable for mixtures of hydrocarbons and hydrofluorocarbons. The advantage of this kind of approach is that the function describing the excess Helmholtz energy is the same for all the mixtures, and relatively simple scaling factors are used to determine its magnitude for a particular system. Once the Helmholtz potential for the mixture is known as a function of density, temperature and composition, all the others thermodynamic properties can be calculated by partial differentiation.

The mixture Helmholtz energy can be written as the sum of two terms:

$$a_m(\delta, \tau, x) = a_m^i(\delta, \tau, x) + a^E(\delta, \tau, x), \quad (1)$$

a_m^i is the Helmholtz energy of the ideal mixture formed by the pure fluids, assuming no interactions between the different species in the mixture. It is calculated as follows:

$$a_m^i = \sum_i x_i a_i \quad (2)$$

where a_i is the contribution of the real pure fluid. It can be calculated through equations of state providing the Helmholtz energy as function of temperature and density (e.g. Helmholtz energy fundamental equations, MBWR or ECS).

The excess Helmholtz energy term a^E represents the contribution of the interaction forces arising from the mixing of the pure fluids in the real mixture and is calculated using the following equations [3,4]:

$$a^E(\delta, \tau, x) = \sum_{i=1}^{n-1} \sum_{j=i+1}^n x_i y_j F_{ij} \sum_{k=1}^{10} N_k \delta^{d_k} \tau^{t_k} \quad (3)$$

δ and τ are the reduced density and temperature respectively and are defined as

$$\delta = \frac{\rho}{\rho_{red}}, \quad \tau = \frac{T_{red}}{T}. \quad (4,5)$$

where the reducing parameters ρ_{red} and T_{red} are calculated through the following equations:

$$\rho_{red} = \left[\sum_{i=1}^n \frac{x_i}{\rho_{c_i}} + \sum_{i=1}^{n-1} \sum_{j=i+1}^n x_i x_j (k_v - 1) \right]^{-1} \quad (6)$$

$$T_{red} = \sum_{i=1}^n x_i T_{c_i} + \sum_{i=1}^{n-1} \sum_{j=i+1}^n x_i \beta x_j \gamma (k_t - 1) \quad (7)$$

N_k , d_k and t_k in equation (3) are general parameters, valid for every mixture, and characterize the generalized Helmholtz function. They have been calculated through a global fitting of various sets of experimental data regarding several binary mixtures of hydrocarbons and refrigerants [3].

Up to five interaction parameters (k_t , k_v , F_{ij} , β , γ) are used to adapt the generalized form of the model to the specific mixture considered and must be calculated through experimental data.

The F_{ij} parameter is necessary to adjust the excess Helmholtz function a^E . If it is set to zero the excess contribution (3) becomes null, and the mixture is modelled by eq. (1) as an ideal mixture of real fluids.

The parameters k_t and k_v are used to define the shapes of the reducing parameters ρ_{red} and T_{red} in equations (6) and (7). The strongest influence is observed for a variation of the parameter k_t : for values greater than one, a negative pressure azeotrope can be modelled, while values smaller than one are observed for positive deviations from the Raoult's law. The influence of the k_t parameter is illustrated in figure 1.

A variation of k_v has a much smaller influence: anyway, for values greater than one, the dew-bubble curves are moved to slightly higher saturation pressures, while for values below one a lower pressure is achieved.

To get a good representation of the VLE data for mixtures with big deviations from the Raoult's law, it is necessary also to introduce the parameters β and γ . For ideal mixtures they can be neglected, i.e. they are set to one in equations (6) and (7).

The values of parameters k_t , k_v , β , γ are often close to one; when they are set to one, the model reduces to ideal mixtures of real fluids, without interaction between different species.

REPRESENTATION OF THE BOUNDARY CONDITIONS WITH THE L-J MODEL

The systems studied at the CNR-ITC laboratory in the last few years [5,6,7,8,9,10] can be classified in three different families on the base of the shapes of the saturation boundaries and the molecular interactions between the different species. For each family, a representative mixture has been selected to calculate the interaction parameters for the L-J model and exemplify the thermodynamic behavior.

- HFC + HFC: these systems show an almost ideal behavior, with small deviations from Raoult's law. Only three parameters (k_t , k_v , and F_{ij}) are required to obtain a good representation of saturation boundaries. As it is shown in table 1 for the mixture HFC-143a+HFC-236fa [5], the values obtained are close to the ideal mixture parameters ($k_t=1$, $k_v=1$ and $F_{ij}=0$).
- HFC + HC: the deviation from the Raoult's law is strongly positive, often with the formation of an azeotrope. As exemplified in table 1 for the mixture HFC134a+HC-600a [6], all the five parameters need to be used. The k_t parameter is far below one, as expected for a system with positive deviation.
- HFC + dimethyl ether (RE170): this system shows a negative deviation from the Raoult's law that can be attributed to hydrogen bonding. This phenomenon is due to the proton donor property of hydrogen in molecules containing fluorine atoms such as refrigerants and the proton acceptor group of Dimethyl ether. Hydrogen bonding could play an important role in the solubility of refrigerants in polyester and polyether oils, which also have proton acceptor groups. Regression of experimental data shows a strong influence of hydrogen bonding in the k_t parameter and negligible effects on other parameters. The optimal value for k_t depends on temperature, though the dependence is almost linear as exemplified in figure 2 for the mixture HFC-236fa+Dimethyl ether [7,8,9].
- to highlight the different behavior of RE170 when mixed with a fluid not giving H-bonding, the interaction parameters calculated for the mixture RE170 + R600a [10] are reported in table 1 also. This mixture shows a positive deviation from the Raoult's law and a behavior strictly related with the case of HFC+HC mixtures.

In table 1, AAD P% and AAD y are the absolute average deviations for pressure, in percentage, and the vapour composition, in mole fraction, and have been calculated from the following equations:

$$AAD P = \frac{100}{N_p} \sum_{i=1}^{N_p} \left[\left| \frac{P_{cal} - P_{exp}}{P_{exp}} \right| \right]_i \quad (8)$$

$$AAD y = \frac{1}{N_p - 2} \sum_{i=2}^{N_p-1} \left(\left| y_{cal} - y_{exp} \right| \right)_i \quad (9)$$

where P_{calc} and y_{calc} are the values calculated through the L-J model, P_{exp} and y_{exp} are the experimental data.

The pure fluids properties for R143a, R236fa, R134a and R600a have been calculated by using the subroutines of REFPROP 6.0. The properties for RE170 have been calculated by the equation proposed by [11].

| MIXTURE | ISOTHERMS [°C] | k_t | k_v | F_{ij} | β | γ | AAD P [%] | AAD y |
|--------------------|-------------------|---------|---------|----------|---------|----------|--------------|--------|
| HFC-143a+HFC-236fa | 10,25,40 | 1.02116 | 1.03074 | 0.10563 | - | - | 0.21 | 0.0013 |
| HFC-134a+HC-600a | 20,30 | 0.80081 | 1.01227 | -0.29878 | 1.00837 | 0.98328 | 0.38 | 0.0026 |
| RE170+HFC-236fa | 10,30,50 | *** | 0.87517 | 1.43064 | 1.27312 | 0.94465 | 0.46 | 0.0076 |
| RE170-HC-600a | 7,17,27,37,47 | 0.99388 | 0.98723 | -1.44348 | 0.50201 | 1.40811 | 0.25 | 0.0026 |

$$*** K_t = 1.0531 + 0.005 \cdot T \text{ [}^\circ\text{C]}$$

Table 1: Interactions parameters for the selected mixtures and deviations for pressure and vapour composition

EVALUATION OF CYCLES PERFORMANCE

The L-J equation obtained for the Helmholtz energy is a fundamental equation which allows the direct calculation of all the thermodynamic properties by proper differentiation. This means that if the potential A is not well represented, errors can be generated in calculating other properties such as enthalpy, entropy and density.

The use of only VLE data to regress the interaction parameters in the Helmholtz energy equations, allows a good representation of all the thermodynamic properties in the region close to the saturation boundaries. Far from the saturation boundaries, even if the errors become a little greater, they should maintain a good consistency due to the theoretical conception of the model. Anyway, the representation of the thermodynamic properties of the mixture should be much improved with respect to the application of ideal mixture interaction parameters (eg. as in the case of REFPROP 6.0).

For these reasons, different results should be expected in calculating theoretical cycles performance such as the coefficient of performance (COP) and the volumetric refrigerating effect (VE) with the predictive mode (i.e. using REFPROP 6.0) or with the correlative mode (i.e. applying the L-J model with interaction parameters regressed on the base of experimental VLE data).

A single stage theoretical cycle has been considered in order to verify the influence of the model in calculating thermodynamic efficiency parameters such as COP and volumetric refrigerating capacity. The operative conditions of the cycle are summarized in table 3.

| | |
|------------------------------|-----|
| Evaporating Temperature [°C] | -15 |
| Condensing Temperature [°C] | 30 |
| Superheating [°C] | 10 |
| Sub cooling [°C] | 0 |
| Isentropic Efficiency | 1 |

Table 3: Operative conditions of the theoretical cycle

All the mixtures previously considered have been studied as a working fluid in this cycle. A comparison of the results obtained applying the L-J model in the correlative and the predictive mode is reported in Tables 4 and 5. Table 4 reports the condensing and the evaporating pressures and the compressor suction density. In Table 5 the compressor work W , the refrigerating effect ΔH_{ev} , the COP and the volumetric refrigerating effect VE are reported. The refrigerating effect ΔH_{ev} has been calculated as the enthalpy variation between the evaporator inlet and the saturated vapor at evaporating pressure; the COP has been calculated as the ratio between the refrigerating effect ΔH_{ev} and the compressor work W ; finally, the volumetric refrigerating effect VE has been calculated as the ratio between the refrigerating effect ΔH_{ev} and the compressor suction density.

The deviations $\Delta\%$ express the percentage difference between the results obtained with REFPROP 6.0 (L-J in the predictive mode) and those obtained with the L-J in the correlative mode.

Three different compositions have been considered for each mixture.

The results clearly show a big difference in the condensing and evaporating pressures calculated in the two different modes. Considering the good representation of this properties in the correlative mode, as shown in Table 1, this means that the predictive mode (i.e. REFPROP 6.0) is absolutely not able to correctly represent the saturation boundaries for the mixtures here considered. A big difference in suction densities is shown also. As expected, the lowest deviations (anyway between 3 and 5% for saturation pressures and between 3 and 6% for suction densities) are achieved for the almost ideal mixture R143a+R236fa. The deviations obtained for the other mixtures are much higher (up to -60% for both the saturation pressures and the suction density).

As refer the performance of the theoretical cycle, not big differences result in calculating the COP by the predictive mode and the correlative mode. This is not surprising since the deviations in the compressor work and the refrigerating effect are of the same order and the effect on the ratio is small. As a matter of fact, considering the relative deviations for the refrigerating effect ΔH_{ev} and the compressor work W , it is evident that they are not negligible, especially for mixtures with a big deviation from ideality.

On the contrary, the volumetric efficiency is strongly affected by the model considered, cause of the large deviations of the vapor density at the compressor suction. This means that calculating the thermodynamic properties with the predictive mode can strongly mislead in dimensioning the compressor.

The considered cycle for the mixture R134a + R600a at the equimolar composition is exemplified in Figure 4 for both the predictive mode (REFPROP 6.0) and the correlative mode (L-J model with interaction parameters regressed on the base of experimental VLE data). The diagram shows a big difference in the saturation boundaries shape and the cycle position: lower values for enthalpy and higher value for pressure are achieved in correlative mode.

| MIX | x_1 | w_1 | P_{ev} [kPa] | | | P_{cond} [kPa] | | | $\rho_{suction}$ [kg/m ³] | | |
|--------------|-------|-------|----------------|-------|-------------|------------------|--------|-------------|---------------------------------------|-------|-------------|
| | | | RP6 | L-J | $\Delta \%$ | RP6 | L-J | $\Delta \%$ | RP6 | L-J | $\Delta \%$ |
| R143a-R236fa | 0.2 | 0.12 | 77.3 | 74.7 | 3.36 | 458.8 | 437.7 | 4.59 | 4.90 | 4.73 | 3.39 |
| | 0.5 | 0.36 | 133.7 | 125.9 | 5.83 | 694.9 | 662.0 | 4.73 | 7.21 | 6.80 | 5.73 |
| | 0.8 | 0.69 | 235.3 | 224.5 | 4.59 | 1046.9 | 1010.5 | 3.48 | 10.74 | 10.15 | 5.45 |
| R134a-R600a | 0.2 | 0.31 | 97.6 | 127.5 | -30.6 | 453.7 | 596.8 | -31.5 | 3.03 | 3.93 | -29.5 |
| | 0.5 | 0.64 | 115.4 | 182.4 | -58.1 | 544.0 | 787.8 | -44.8 | 4.31 | 6.84 | -58.7 |
| | 0.8 | 0.88 | 140.4 | 197.5 | -40.6 | 663.4 | 870.4 | -31.2 | 6.14 | 8.75 | -42.5 |
| DME-R600a | 0.2 | 0.17 | 96.9 | 109.8 | -13.3 | 442.7 | 490.9 | -10.8 | 2.51 | 2.83 | -12.8 |
| | 0.5 | 0.44 | 112.6 | 136.8 | -21.5 | 511.4 | 589.0 | -15.1 | 2.72 | 3.31 | -21.6 |
| | 0.8 | 0.76 | 133.4 | 152.0 | -13.9 | 599.3 | 656.5 | -9.54 | 3.02 | 1.47 | 51.1 |
| DME-R236fa | 0.2 | 0.07 | 65.4 | 56.5 | 13.6 | 367.1 | 325.8 | 11.2 | 3.93 | 3.39 | 13.6 |
| | 0.5 | 0.23 | 86.3 | 66.4 | 23.1 | 452.2 | 373.2 | 17.4 | 3.93 | 3.01 | 23.4 |
| | 0.8 | 0.55 | 118.2 | 97.2 | 17.7 | 566.2 | 498.4 | 11.9 | 3.68 | 2.99 | 18.8 |

Table 4: Comparison of theoretical cycle thermodynamic parameters for different mixtures

| MIX | x_1 | w_1 | ΔH_{ev} [kJ/kg] | | | W [kJ/kg] | | | COP | | | VE [kJ/m ³] | | |
|--------------|-------|-------|-------------------------|-------|-------------|-------------|------|-------------|------|------|-------------|-------------------------|--------|-------------|
| | | | RP6 | L-J | $\Delta \%$ | RP6 | L-J | $\Delta \%$ | RP6 | L-J | $\Delta \%$ | RP6 | L-J | $\Delta \%$ |
| R143a-R236fa | 0.2 | 0.12 | 123.5 | 122.9 | 0.42 | 29.1 | 28.9 | 0.84 | 4.23 | 4.25 | -0.47 | 604.7 | 583.0 | 3.59 |
| | 0.5 | 0.36 | 134.9 | 136.2 | -0.99 | 31.6 | 31.8 | -0.78 | 4.26 | 4.27 | -0.23 | 974.2 | 926.9 | 4.86 |
| | 0.8 | 0.69 | 138.7 | 141.9 | -2.31 | 33.7 | 34.2 | -1.37 | 4.11 | 4.15 | -0.97 | 1496.7 | 1454.7 | 2.81 |
| R134a-R600a | 0.2 | 0.31 | 231.7 | 230.8 | 0.39 | 51.8 | 52.2 | -0.84 | 4.47 | 4.41 | 1.34 | 703.3 | 907.1 | -28.9 |
| | 0.5 | 0.64 | 194.9 | 174.1 | 10.7 | 43.6 | 40.4 | 7.24 | 4.46 | 4.30 | 3.59 | 840.0 | 1191.0 | -41.7 |
| | 0.8 | 0.88 | 165.6 | 148.4 | 10.4 | 37.3 | 34.8 | 6.69 | 4.43 | 4.25 | 4.06 | 1016.9 | 1299.1 | -27.7 |
| DME-R600a | 0.2 | 0.17 | 279.2 | 273.9 | 1.89 | 61.8 | 61.0 | 1.25 | 4.51 | 4.48 | 0.67 | 700.1 | 775.6 | -10.7 |
| | 0.5 | 0.44 | 303.7 | 289.2 | 4.78 | 66.3 | 63.6 | 3.99 | 4.57 | 4.54 | 0.66 | 828.5 | 959.0 | -15.7 |
| | 0.8 | 0.76 | 329.5 | 317.0 | 3.80 | 71.1 | 68.9 | 3.36 | 4.62 | 4.60 | 0.43 | 994.5 | 1094.1 | -10.0 |
| DME-R236fa | 0.2 | 0.07 | 130.5 | 134.3 | -2.92 | 29.9 | 30.4 | -1.79 | 4.36 | 4.41 | -1.15 | 513.6 | 456.9 | 11.0 |
| | 0.5 | 0.23 | 171.3 | 182.7 | -6.67 | 38.2 | 40.2 | -5.09 | 4.47 | 4.54 | -1.57 | 673.9 | 551.5 | 18.1 |
| | 0.8 | 0.55 | 245.8 | 263.2 | -7.08 | 53.8 | 57.1 | -6.05 | 4.56 | 4.61 | -1.10 | 904.7 | 787.4 | 12.9 |

Table 5: Comparison of the theoretical cycles performance parameters for different mixtures

CONCLUSIONS

The results obtained with the present work clearly shows that the Lemmon-Jacobsen is a valid instrument to predict the thermodynamic properties of new refrigerants mixtures when interaction parameters based on experimental data are used. Anyway, when the model is used in predictive mode, as it happens in the REFPROP 6.0 database for the mixtures here considered, the results can be acceptable only for mixture with almost ideal behavior, while for mixtures with high deviations from the Raoult's law the results are completely misleading. This influences all the thermodynamic properties calculation and then also the calculation of the performance for refrigerating cycles, here demonstrated for theoretical cycles. In particular, the evaporating and the condensing pressures, the compressor suction density and the volumetric refrigerating effect are strongly affected by the interactions parameters used in the model.

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FIGURES

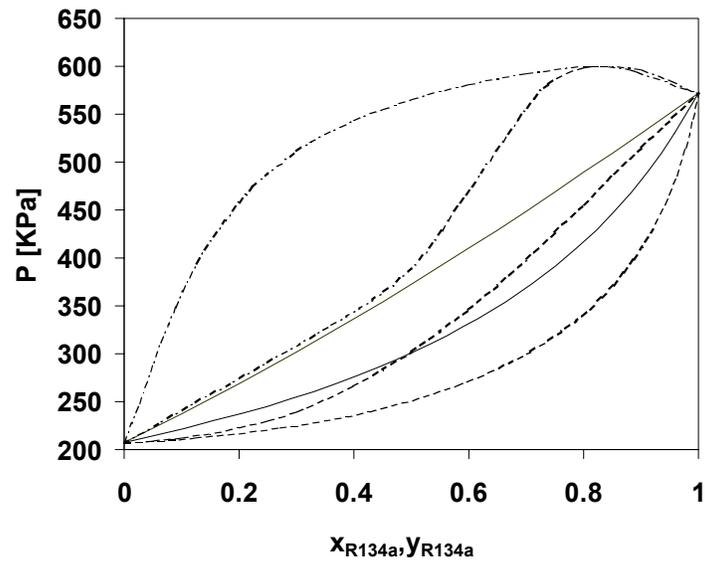


Figure 1: Effect of the interaction parameter k_t for the binary mixture R134a-Butane
 $k_t=1$ (—), $k_t=0.8$ (.....), $k_t=1.1$ (-----)

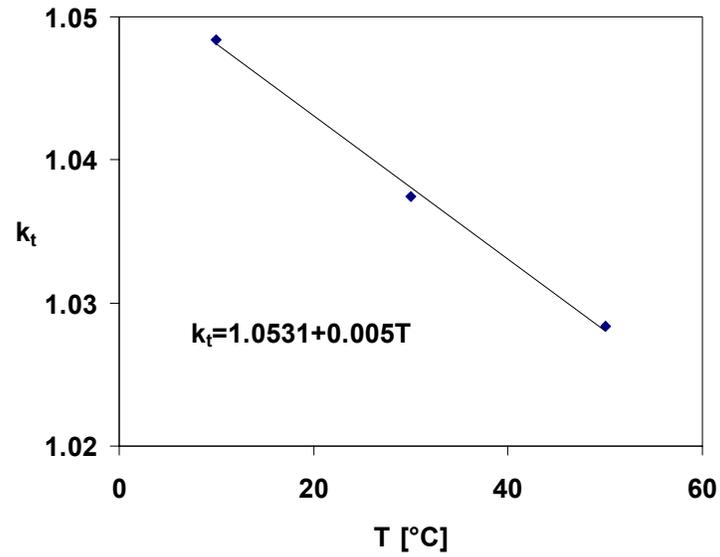


Figure 2: Effect of Hydrogen bonding on k_t interaction parameter

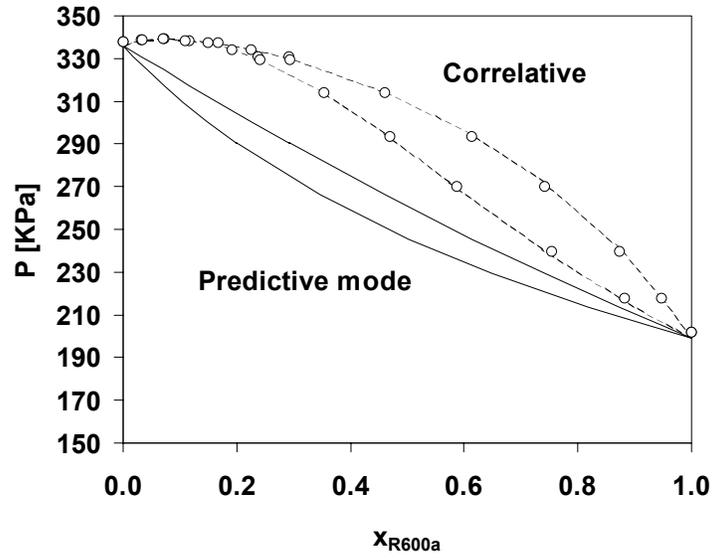


Figure 3: Comparison between L-J in predictive (—) and correlative (---) mode; (○) experimental data

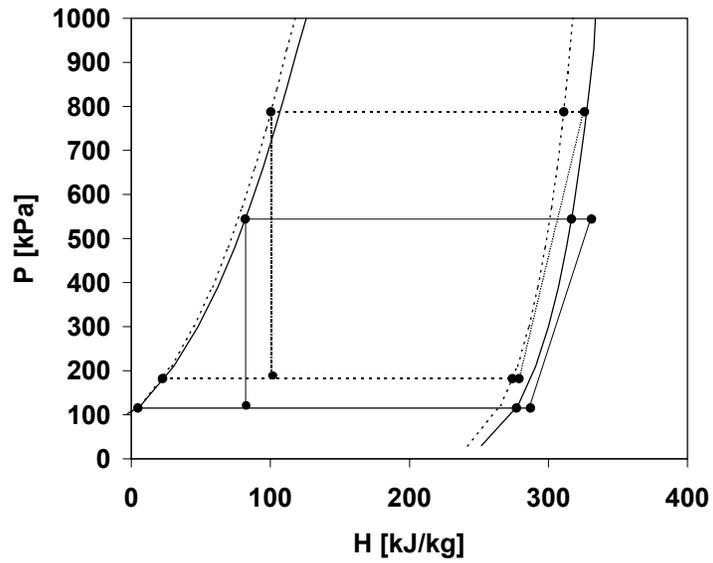


Figure 4: Studied cycle through the predictive mode (REFPROP 6.0) (—) and the correlative mode (---)