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ABSTRACT

PPR78 (Predictive-Peng-Robinson78) is a well-established cubic equation of state in which, the temperature-dependent binary interaction parameters (bips) between two molecules i and j [$k_{ij}(T)$] are predicted by a group-contribution method. In such a model, two group-binary-interaction parameters (G-bips), noted A_{kl} and B_{kl}, are required per (k,l) pair of groups. The purpose of this article is to demonstrate that the inclusion of enthalpy and heat capacity of mixing data in the process of optimisation of the G-bips leads to a remarkable overall reduction of the error on such properties, from 841.51% to 51.42%, while keeping constant the accuracy in the prediction of VLE data, 8.6% (without inclusion of mixing properties) to 8.8% (with inclusion of mixing thermal properties). This new version of the PPR78 model (with the new sets of A_{kl} and B_{kl} G-bips) has thus been called *Enhanced*-PPR78 (*E*-PPR78) model. From 2013 to 2017, this model was continuously developed and it is currently based on 40 groups. The many G-bips were fitted over 131,207 vapour liquid equilibrium data and 33,629 mixing properties data (h^M and c_P^M) belonging to 1301 binary systems. After almost 18 years from the initial development of the PPR78 model, this paper summarizes for the first time the overall accuracy of the *E*-PPR78 model.

1. Introduction

After eight years since the release of the *Enhanced*-Predictive-PR78 equation of state (*E*-PPR78), this paper details the reason which drove our research group to develop this model, from the modification of the original PPR78.

The *predictive* character of PPR78 relies on its capability of calculating thermodynamic properties of multicomponent systems knowing: (*i*) pure-component characteristics (i.e., acentric factor, critical temperature and pressure, decomposition in elementary groups from the molecular structure) and (*ii*) group-binary-interaction parameters (G-bips), which allow the predictive determination of the temperature-dependent binary interaction parameters (k_{ij}) between 2 components i and j by a group-contribution method. In the PPR78 model, two G-bips, noted A_{kl} and B_{kl}, are required per (k,l) pair of groups and this paper is devoted to highlight the importance of properly selecting the type of experimental data underlying the process of optimisation of such G-bips.

The theoretical development of PPR78 was presented for the first time in 2004 [1]. In the years that followed, prior to 2015, the model was optimised over phase equilibrium data (liquid and gas phase compositions, critical pressures and compositions, azeotropic pressures and compositions) of binary mixtures [2–13], resulting in a model counting 21 groups (CH_3, CH_2, CH, C, aromatic carbon atom groups, cyclic carbon atom groups, double-bonded carbon atom groups, CO_2, N_2, H_2S, -SH, H_2O, H_2). For these 21 groups, it was necessary to determine the values of 420 G-bips (210 A_{kl} and 210 B_{kl}). Among the mostly applied predictive cubic equations of state (EoS), PPR78 proved to be one of the most accurate in modelling phase-equilibrium properties of multicomponent fluids containing non-associating compounds explaining why it is available in commercial process simulators like ProSimPlus, Simulis® Thermodynamics and PRO/II. However, as stated in 2001 by Agarwal et al. [14], "*if you are modelling a system with significant excess enthalpies using a Gibbs free-energy-based EoS and you have a good VLE fit, this does not automatically ensure you a good overall model (from an energy balance point of view)*". With the aim to evaluate

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the applicability of the PPR78 model to the calculation of enthalpy changes and heat capacities so that it can be used to perform energy and exergy balances, Qian et al. published in 2013 [15] extended results about the capability of PPR78 to predict enthalpy and heat capacity changes of mixing. It was concluded that without being excessively high, the deviations between calculated and experimental data were not as accurate as expected so that there was room for improvement. In response to such an outcome, the 420 G-bips of the model were then re-optimised over both experimental VLE and property changes on mixing (h^M and c_P^M) data. Despite the impressive gain in accuracy, the resulting parameters and the corresponding deviations remained unpublished until the writing of this paper. They however can be found in the thesis by Qian [16]. This increase in accuracy explains why, since 2015, published PPR78's G-bips have been optimised over, *simultaneously*, enthalpy and heat capacity of mixing in addition to phase equilibrium data [17–21].

This paper intends to report on the improved predictive capability of the version of PPR78, the G-bips of which were optimised over such an extended set of data (h^M, c_P^M, VLE) and named *Enhanced*-PPR78 (*E*-PPR78). After recalling the mathematical features of the PPR78 model (which formulation is equivalent to the one of the *E*-PPR78 model) and of the optimisation framework, we present the results obtained in 2013 with the 21 first groups and the corresponding 420 G-bips optimised over VLE data, enthalpy and heat capacity of mixing properties. A comparison of this re-optimised model (*E*-PPR78) with the predictive capability of the original PPR78 model is also reported. From 2013 to 2017, 19 new groups were added so that the *E*-PPR78 model includes currently 40 groups. For the first time, this paper presents the average modelling errors on the ensemble of data (more than 150,000 data points) considered to develop such a model.

2. From PPR78 to *E*-PPR78

2.1. *PPR78 and* E-*PPR78, a unique mathematical formulation*

The PPR78 model is a predictive version of the equation of state published in 1978 by Peng and Robinson [22], which we are used to refer to with the label PR78. As detailed below, such an EoS is made predictive through the development of a group contribution method to predict the temperature-dependent bips that appear in the classical Van der Waals one-fluid mixing rules.

The PR78 EoS can be written:

$$P = \frac{RT}{v-b} - \frac{a}{v(v+b)+b(v-b)} \tag{1}$$

When applied to the i-th pure component, a in eq. (1) corresponds to its specific cohesive parameter, a_i, and b to its co-volume, b_i, which are in turn calculated with the generalized functions reported in the following:

$$\begin{cases} R = 8.314472\ J\cdot mol^{-1}\cdot K^{-1} \\ X = \left[1+\sqrt[3]{4-2\sqrt{2}}+\sqrt[3]{4+2\sqrt{2}}\right]^{-1} \approx 0.253076587 \\ b_i = \Omega_b \frac{RT_{c,i}}{P_{c,i}} with: \Omega_b = \frac{X}{X+3} \approx 0.0777960739 \\ a_i(T) = a_{c,i}\alpha_i(T) with \begin{cases} a_{c,i} = \Omega_a \frac{R^2T_{c,i}^2}{P_{c,i}} \ and \\ \Omega_a = \frac{8(5X+1)}{49-37X} \approx 0.457235529 \\ \alpha_i(T) = \left[1+m_i\left(1-\sqrt{\frac{T}{T_{c,i}}}\right)\right]^2 \end{cases} \\ if\ \omega_i < 0.491\ then\ m_i = 0.37464 + 1.54226\omega_i - 0.26992\omega_i^2 \\ if\ \omega_i > 0.491\ then\ m_i = 0.379642 + 1.48503\omega_i \\ -0.164423\omega_i^2 + 0.016666\omega_i^3 \end{cases} \tag{2}$$

The application of this EoS to a mixture requires the selection of mixing rules for calculating its cohesive and co-volume parameters, a and b. Classical Van der Waals one-fluid mixing rules are used in the PR78 model:

$$\begin{cases} a(T,z) = \sum_{i=1}^{N}\sum_{j=1}^{N} z_i z_j \sqrt{a_i(T)a_j(T)}\left(1-k_{ij}(T)\right) \\ b(z) = \sum_{i=1}^{N} z_i b_i \end{cases} \tag{3}$$

Where N is the number of compounds in the mixture and $\mathbf{z} = (z_1, ..., z_N)$, the mole fraction vector. The temperature-dependent $k_{ij}(T)$ parameter is the so–called binary interaction parameter characterizing the molecular interactions between molecules i and j. The most accurate application of the original PR78 model requires the empirical optimization of the $k_{ij}(T)$ parameter over, at least, vapour-liquid equilibrium experimental data.

In order to render predictive the PR78 EoS, the temperature-dependent $k_{ij}(T)$ parameter was expressed as:

$$k_{ij}(T) = \frac{E_{ij}(T) - (\delta_i - \delta_j)^2}{2\delta_i\delta_j} \ with\ \delta_i = \frac{\sqrt{a_i}}{b_i} \tag{4}$$

and a group contribution method was developed to predict $E_{ij}(T)$:

$$E_{ij}(T) = -\frac{1}{2}\left[\sum_{k=1}^{N_g}\sum_{l=1}^{N_g}(\alpha_{ik}-\alpha_{jk})(\alpha_{il}-\alpha_{jl})A_{kl}\left(\frac{298.15}{T/K}\right)^{\left(\frac{B_{kl}}{A_{kl}}-1\right)}\right] \tag{5}$$

In Eq. (4), a_i and b_i values are calculated from Eq. (2). A_{kl} and B_{kl} are the G-bips; they are constant real numbers (like the A_{ij} parameters of the UNIFAC matrix), the numerical values of which were determined by correlating thousands of fluid-phase equilibria experimental data. In addition, they are symmetric, $A_{kl} = A_{lk}$ and $B_{kl} = B_{lk}$ (where k and l are two different groups), and $A_{kk} = B_{kk} = 0$. In Eq. (5), N_g is the number of different groups defined by the method and α_{ik} is the fraction of molecule i occupied by group k (occurrence of group k in molecule i divided by the total number of groups present in molecule i). The inclusion in the PR78 equation of state of this predictive expression for k_{ij} [Eqs. (4)-(5)] results in the *Predictive*-PR78 (PPR78).

As explained by Jaubert and Privat [23], the expression for the $k_{ij}(T)$ reported in Eq. (4) can be equivalently derived by combining the PR EoS with the well-known *EoS*/g^E Huron–Vidal mixing rules [24]:

$$\begin{cases} \frac{a(T,z)}{b(z)} = \sum_{i=1}^{N} z_i \frac{a_i(T)}{b_i} - \frac{g^E(T,z)}{C_{EoS}} \\ b(z) = \sum_{i=1}^{N} z_i b_i \\ C_{EoS} = \frac{\sqrt{2}}{2}\ln\left(1+\sqrt{2}\right)\ for\ the\ PR\ EoS \end{cases} \tag{6}$$

and a Van Laar type activity coefficient (g^E) model

$$\frac{g^E_{VanLaar}(T,z)}{C_{EoS}} = \frac{1}{2}\frac{\sum_{i=1}^{N}\sum_{j=1}^{N} z_i z_j b_i b_j E_{ij}(T)}{\sum_{j=1}^{N} z_j b_j} \tag{7}$$

In the end, Eq. (5) can thus be seen as an equation aimed at predicting the $E_{ij}(T)$ parameter of a Van Laar type g^E model by group-contribution.

For completeness, we also recall that a further theoretical development of the model has been derived in the year 2010, with the recognition that it is possible to rigorously deduce the k_{ij} of any EoS, knowing those of the PR EoS [25]. The group–interaction parameters (A_{kl} and B_{kl}) initially developed for the PR EoS can be thus used to predict the k_{ij} of any other cubic EoS, including volume-translated equations [26,27], combined with any alpha function [28–30].

The final advancement of PPR78, which resulted in the so-called *Enhanced*-PPR78, concerned the improvement of its capability of calculating the enthalpy and heat capacity changes on mixing. Such a result has been obtained with the sole modification of the set of experimental data used to optimize the group-binary-interaction parameters A_{kl} and B_{kl} of the model. This dataset, which was originally composed of only vapour-liquid equilibrium data, has been extended to enthalpy and heat capacity changes-due-to-mixing data. The mathematical formulation of the *E*-PPR78 model is indeed equivalent to the PPR78 one and the two models differ uniquely in the G-bips (A_{kl} and B_{kl}) numerical values that were optimized on different types of experimental data. The specific optimization framework of the G-bips and corresponding results are detailed in section 2.3.

2.2. Optimizing A_{kl} and B_{kl} G-bips over mixing enthalpy and mixing heat capacity in addition to VLE data

One of the main open questions which arise when optimizing EoS parameters concerns the selection of the experimental data at the base of the optimization process: what type of data should be selected in order to ensure a precise and reliable determination of phase equilibrium and derived thermodynamic properties like enthalpies and heat capacities? The answer is not straightforward because the equations of state can predict a whole set of thermodynamic properties from the same set of optimized parameters.

In the case of pure fluids, the inclusion of vaporization enthalpies and liquid heat capacities in addition to saturation pressures and liquid densities in the optimization of model parameters leads to a small increase of accuracy in predicting enthalpies and heat capacity data and to a negligible negative impact on saturation pressures and liquid densities [31,32]. In this publication, we will show that, in case of mixtures, the conclusions are totally different. Indeed, the optimization of bips over h^M and c_P^M in addition to VLE data makes it possible to drastically improve the correlation of derived properties without deteriorating the VLE data. This is the basis for the development of the *E*-PPR78 model.

To better understand such a result, we preliminarily discuss how temperature-dependent bips (like those used in the *E*-PPR78 model) do affect the calculation of mixing enthalpy, heat capacity and phase equilibrium data. As emphasized in the work of Qian et al. [15], molar enthalpy and molar heat capacity changes on mixing in a N-component mixture are straightforwardly connected to the corresponding T,v-residual properties by:

$$h^M(T,v,\boldsymbol{z}) = h^{res}(T,v,\boldsymbol{z}) - \sum_{i=1}^{N} z_i h_i^{res}(T,v_i) \tag{8}$$

$$c_P^M(T,v,\boldsymbol{z}) = c_P^{res}(T,v,\boldsymbol{z}) - \sum_{i=1}^{N} z_i c_{P,i}^{res}(T,v_i) \tag{9}$$

where $m^{res}(T,v,\boldsymbol{z})$ and $m_i^{res}(T,v_i)$ are the T,v-residual property ($m = h$ or c_P) of, respectively, the mixture and of the i-th pure component, which can be both calculated from an EoS; v_i denotes the molar volume of pure i at the same temperature T and pressure $P_{mix} = P(T,v,\boldsymbol{z})$ as the mixture and is obtained basically from the resolution of the EoS for pure component i at fixed T and P_{mix}, i.e., by solving: $P(T,v_i,z_i=1) = P_{mix}$. Note that by "T,v-residual property", it is here meant the departure function between the real property and the corresponding perfect-gas property having the same temperature, composition and molar volume as the real mixture. The PPR78 equation of state leads to:

$$h^{res}(T,v,\boldsymbol{z}) = \frac{R\cdot T\cdot b_m(\boldsymbol{z})}{v-b_m(\boldsymbol{z})} - \frac{a_m(T,\boldsymbol{z})\cdot v}{v[v+b_m(\boldsymbol{z})]+b_m(\boldsymbol{z})[v-b_m(\boldsymbol{z})]} - \frac{1}{2\sqrt{2}\cdot b_m(\boldsymbol{z})}\cdot\left(a_m(T,\boldsymbol{z}) - T\cdot\frac{\mathbf{da_m(T,z)}}{\mathbf{dT}}\right)\cdot\ln\left[\frac{v+\left(1+\sqrt{2}\right)b_m(\boldsymbol{z})}{v+\left(1-\sqrt{2}\right)b_m(\boldsymbol{z})}\right] \tag{10}$$

and,

$$c_P^{res}(T,v,\boldsymbol{z}) = c_v^{res}(T,v,\boldsymbol{z}) - R + T(\kappa_T\cdot v)(\beta\cdot P)^2$$
$$\text{with}\begin{cases}\kappa_T\cdot v = \left[\frac{RT}{[v-b_m(\boldsymbol{z})]^2} - \frac{2\cdot a_m(T,\boldsymbol{z})[v+b_m(\boldsymbol{z})]}{[v(v+b_m(\boldsymbol{z}))+b_m(\boldsymbol{z})(v-b_m(\boldsymbol{z}))]^2}\right]^{-1}\\ \beta\cdot P = \frac{R}{v-b_m(\boldsymbol{z})} - \frac{\mathbf{da_m(T,z)}}{\mathbf{dT}}\cdot\frac{1}{v[v+b_m(\boldsymbol{z})]+b_m(\boldsymbol{z})[v-b_m(\boldsymbol{z})]}\\ c_v^{res} = \frac{T}{2\sqrt{2}\cdot b_m(\boldsymbol{z})}\cdot\frac{\mathbf{d^2a_m(T,z)}}{\mathbf{dT^2}}\cdot\ln\left[\frac{v+\left(1+\sqrt{2}\right)b_m(\boldsymbol{z})}{v+\left(1-\sqrt{2}\right)b_m(\boldsymbol{z})}\right]\end{cases} \tag{11}$$

Pure component properties can be derived from equations above, considering that:

$$h_i^{res}(T,v_i) = h^{res}(T,v_i,z_i=1) \tag{12}$$

$$c_{P,i}^{res}(T,v_i) = c_P^{res}(T,v_i,z_i=1) \tag{13}$$

Knowing that $a_m(T,\boldsymbol{z}) = \sum_{i=1}^{N}\sum_{j=1}^{N} z_i z_j\sqrt{a_i a_j}(1-k_{ij}(T))$ (Van der Waals mixing rule for the energy parameter), the first and the second temperature derivatives of $a_m(T,\boldsymbol{z})$ are:

$$\left(\frac{\mathbf{da_m(T,z)}}{\mathbf{dT}}\right)_{\mathbf{z}} = \sum_{i=1}^{N}\sum_{j=1}^{N} z_i z_j\left[\left[1-k_{ij}(T)\right]\frac{a_j\frac{da_i}{dT}+a_i\frac{da_j}{dT}}{2\sqrt{a_i a_j}} - \sqrt{a_i a_j}\frac{\mathbf{dk_{ij}(T)}}{\mathbf{dT}}\right] \tag{14}$$

$$\left(\frac{\mathbf{d^2a_m(T,z)}}{\mathbf{dT^2}}\right)_{\mathbf{z}} = \sum_{i=1}^{N}\sum_{j=1}^{N} z_i z_j\left[\begin{array}{l}\left[1-k_{ij}(T)\right]\left[\frac{a_j\frac{d^2a_i}{dT^2}+2\frac{da_i}{dT}\frac{da_j}{dT}+a_i\frac{d^2a_j}{dT^2}}{2\sqrt{a_i a_j}} - \frac{\left(a_i\frac{da_j}{dT}+a_j\frac{da_i}{dT}\right)^2}{4\left(a_i a_j\right)^{1.5}}\right]\\ -\frac{\mathbf{dk_{ij}(T)}}{\mathbf{dT}}\frac{a_j\frac{da_i}{dT}+a_i\frac{da_j}{dT}}{\sqrt{a_i a_j}} - \sqrt{a_i a_j}\frac{\mathbf{d^2k_{ij}(T)}}{\mathbf{dT^2}}\end{array}\right] \tag{15}$$

As shown in our previous studies, temperature-dependent binary interaction parameters $k_{ij}(T)$ play an important role in the phase equilibrium calculations. Equations (10)-(11) and (14)-(15) show the mathematical relation between the first derivative of $k_{ij}(T)$ with temperature (dk_{ij}/dT) and mixing enthalpy, as well as the dependence of mixing heat capacity with dk_{ij}/dT and the second derivative d^2k_{ij}/dT^2.

In order to give an illustration of these relations, *Fig. 1*-a shows three calculated $h^M - x$ curves for the system (benzene(1) + isooctane(2)) at T = 298.15 K and under P = 1.00 bar, with the Peng-Robinson EoS and Van der Waals mixing rules (see Eqs. (4)-(5)), together with experimental data points. The k_{ij} chosen here for the 3 curves is the value returned by the PPR78 model, that is: $k_{ij} = 0.0029$. $h^M - x$ curves are calculated with three different dk_{ij}/dT values, resulting from the use of different couples of parameters A_{kl}, B_{kl} giving the same $k_{ij}(T)$; they are: $\frac{dk_{ij}}{dT} = -1.7\cdot10^{-4}$ (in red), $\frac{dk_{ij}}{dT} = -5.7\cdot10^{-5}$ (in green) and $dk_{ij}/dT = 0.0$ (in blue), among which $\frac{dk_{ij}}{dT} = -1.7\cdot10^{-4}$ (in red) is the value obtained from the PPR78 model, and $\frac{dk_{ij}}{dT} = -5.7\cdot10^{-5}$ (in green) is the value that best describes these experimental h^M data points. *Fig. 1*-a emphasises that the influence of dk_{ij}/dT on h^M is very significant, despite of its small magnitude.

Similar illustration (see *Fig. 1*-b) and explanation apply to the influence of d^2k_{ij}/dT^2 on c_P^M. We must indicate that $k_{ij} = 0.0029$ and $\frac{dk_{ij}}{dT} = -1.7\cdot10^{-4}$ taken here are those obtained from the PPR78 model and only the influence of d^2k_{ij}/dT^2 on c_P^M has been checked (i.e., different couples of parameters A_{kl}, B_{kl} can provide the same $k_{ij}(T)$ and the same dk_{ij}/dT but different d^2k_{ij}/dT^2). It is obvious that the c_P^M values change a lot as d^2k_{ij}/dT^2 varies from

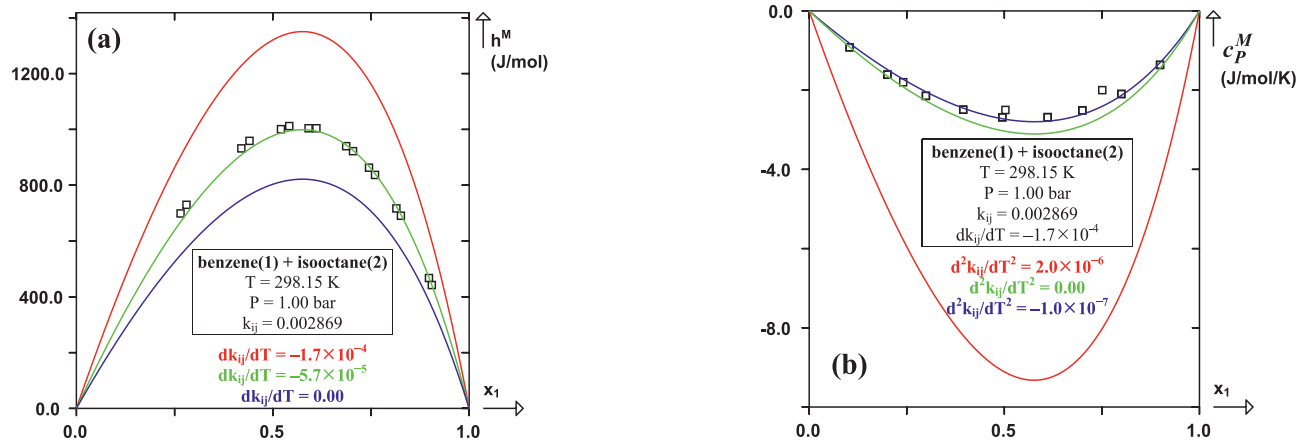

Fig. 1. h^M-x and c_P^M-x curves for the binary system: (benzene(1) + isooctane(2)) at T = 298.15 K and under P = 1 bar. (*) experimental h^M and c_P^M points. Solid line: calculated curves. (a) Calculated h^M curves with the Peng-Robinson EoS and Van der Waals mixing rules, using $k_{ij} = 0.0029$ (PPR78) and three different dk_{ij}/dT values: $dk_{ij}/dT = -1.7\times10^{-4}$ (PPR78), $dk_{ij}/dT = -5.7\times10^{-5}$ (best fit) and $dk_{ij}/dT = 0.00$. (b) Calculated c_P^M curves with the Peng-Robinson EoS and Van der Waals mixing rules, using $k_{ij} = 0.0029$, $dk_{ij}/dT = -1.7\times10^{-4}$ (PPR78), and three different d^2k_{ij}/dT^2 values: $d^2k_{ij}/dT^2 = 2.0\times10^{-6}$ (PPR78), $d^2k_{ij}/dT^2 = 0.00$ and $d^2k_{ij}/dT^2 = -1.0\times10^{-7}$ (best fit).

$-1.0\cdot10^{-7}$ to $2.0\cdot10^{-6}$ passing through 0.0 and the value that best fits the experimental c_P^M points is: $\frac{d^2k_{ij}}{dT^2} = -1.0\cdot10^{-7}$ (in blue).

The first conclusion is that, at a given temperature, there exists a broad range of couples of parameters (A_{kl}, B_{kl}) which allow for the determination of equal $k_{ij}(T)$ but very different dk_{ij}/dT and d^2k_{ij}/dT^2. As a direct consequence, the correlation of enthalpy (that depends on dk_{ij}/dT) and heat capacity (that depends on both dk_{ij}/dT and d^2k_{ij}/dT^2) data at a given temperature can be thus improved without affecting the correlation of isothermal VLE data (that depends on k_{ij} only at given temperature). By analysing the sensitivity of the $k_{ij}(T)$ function with respect to the (A_{kl}, B_{kl}) parameters, we observed that k_{ij} values are strongly influenced by the A_{kl} G-bips but depend weakly on B_{kl} while the influence of the B_{kl} parameters is more visible on the derived properties (h^M and c_P^M). By overstating the point, it should be possible thus to control the k_{ij} values through the A_{kl} parameter to reproduce VLE data accurately and to control k_{ij} derivatives through the B_{kl} parameters, enabling to reproduce h^M and c_P^M data accurately. As a key conclusion, it should be possible therefore to find A_{kl} and B_{kl} parameters that lead to k_{ij}, dk_{ij}/dT and d^2k_{ij}/dT^2 able to accurately and simultaneously correlate VLE data, h^M data and c_P^M data.

It follows that the optimization of A_{kl} and B_{kl} over only phase equilibrium properties (as it is for A_{kl} and B_{kl} of the original PPR78 model) allows the determination of A_{kl} and B_{kl} which optimize the value of $k_{ij}(T)$ and not the one of dk_{ij}/dT and d^2k_{ij}/dT^2. As a consequence, although the original PPR78 model offers a good prediction of binary interaction parameters (and thus of VLE data), the estimations of their first and second derivatives (dk_{ij}/dT and d^2k_{ij}/dT^2) and thus of h^M and c_P^M data remain uncertain. This is because enthalpies and heat capacities (which depend on dk_{ij}/dT and d^2k_{ij}/dT^2) are not included in the optimization process. For this reason, it is important to re-fit A_{kl} and B_{kl} by using VLE, h^M and c_P^M data, so as to establish a group contribution aimed at accurately estimating the $k_{ij}(T)$, dk_{ij}/dT and d^2k_{ij}/dT^2 functions.

In the following section 2.3, we will quantify how the A_{kl} and B_{kl} values, simultaneously optimized over VLE, h^M and c_P^M data, affect the accuracy of each set of properties.

2.3. The early optimisation of the Enhanced-PPR78 model

2.3.1. Methodology

Between the years 2004 and 2013, A_{kl} and B_{kl} G-bips of the PPR78 model have been optimised for a total of 21 groups minimising the objective function depicted in Eq. (16), which includes only phase equilibrium data of binary systems (liquid and gas phase compositions, indicated with subscripts "b" for bubble and "d" for dew, critical compositions (mole fractions z_c) and pressures (P_c), azeotropic compositions (mole fractions z_{az}) and pressures (P_{az})):

$$F_{obj,PPR78}(\%) = \frac{f_b(\%)\cdot n_b + f_d(\%)\cdot n_d + f_{z_c}(\%)\cdot n_c + f_{P_c}(\%)\cdot n_c + f_{z_{az}}(\%)\cdot n_{az} + f_{P_{az}}(\%)\cdot n_{az}}{n_b + n_d + 2n_c + 2n_{az}} \tag{16}$$

where:

$$f_b(\%) = \frac{100}{2n_b}\sum_{i=1}^{n_b}\left(\frac{|\Delta x|}{x_{1,\exp}} + \frac{|\Delta x|}{x_{2,\exp}}\right)_i \text{ with } |\Delta x| = \left|x_{1,\exp} - x_{1,cal}\right| = \left|x_{2,\exp} - x_{2,cal}\right| \tag{17}$$

$$f_d(\%) = \frac{100}{2n_d}\sum_{i=1}^{n_d}\left(\frac{|\Delta y|}{y_{1,\exp}} + \frac{|\Delta y|}{y_{2,\exp}}\right)_i \text{ with } |\Delta y| = \left|y_{1,\exp} - y_{1,cal}\right| = \left|y_{2,\exp} - y_{2,cal}\right| \tag{18}$$

$$f_{z_c}(\%) = \frac{100}{2n_c}\sum_{i=1}^{n_c}\left(\frac{|\Delta z_c|}{(z_c)_{1,\exp}} + \frac{|\Delta z_c|}{(z_c)_{2,\exp}}\right)_i \text{ with } |\Delta z_c| = \left|(z_c)_{1,\exp} - (z_c)_{1,cal}\right| = \left|(z_c)_{2,\exp} - (z_c)_{2,cal}\right| \tag{19}$$

$$f_{P_c}(\%) = \frac{100}{n_c}\sum_{i=1}^{n_c}\left(\frac{|\Delta P_c|}{P_{c,\exp}}\right)_i \text{ with } |\Delta P_c| = \left|P_{c,\exp} - P_{c,cal}\right| \tag{20}$$

$$f_{z_{az}}(\%) = \frac{100}{2n_{az}}\sum_{i=1}^{n_{az}}\left(\frac{|\Delta z_{az}|}{(z_{az})_{1,\exp}} + \frac{|\Delta z_{az}|}{(z_{az})_{2,\exp}}\right)_i \text{ with } |\Delta z_{az}| = \left|(z_{az})_{1,\exp} - (z_{az})_{1,cal}\right| = \left|(z_{az})_{2,\exp} - (z_{az})_{2,cal}\right| \tag{21}$$

$$f_{P_{az}}(\%) = \frac{100}{n_c}\sum_{i=1}^{n_{az}}\left(\frac{|\Delta P_{az}|}{P_{az,\exp}}\right)_i \text{ with } |\Delta P_{az}| = \left|P_{az,\exp} - P_{az,cal}\right| \tag{22}$$

During his Ph.D. thesis, the 210 A_{kl} and 210 B_{kl} G-bips of the PPR78 model have been re-optimised by Qian [16] by minimising the objective function given by Eq. (23), which also includes mixing properties (enthalpy and heat capacity). A totally new set of G-bips (A_{kl} and B_{kl}) was thus obtained and the *E*-PPR78 model was born.

$$F_{obj.E-PPR78}(\%) = \frac{f_b(\%)\cdot n_b + f_d(\%)\cdot n_d + f_{z_c}(\%)\cdot n_c + f_{P_c}(\%)\cdot n_c + f_{z_{az}}(\%)\cdot n_{az} + f_{P_{az}}(\%)\cdot n_{az} + f_{h^M}(\%)\cdot n_{h^M} + f_{c_P^M}(\%)\cdot n_{c_P^M}}{n_b + n_d + 2n_c + +2n_{az} + n_{h^M} + n_{c_P^M}} \quad (23)$$

where:

$$f_{h^M}(\%) = \frac{100}{n_{h^M}}\sum_{i=1}^{n_{h^M}}\left(\frac{\Delta h^M}{h^M_{\exp}}\right)_i \text{ with } \Delta h^M = \left|h^M_{\exp} - h^M_{cal}\right| \quad (24)$$

$$f_{c_P^M}(\%) = \frac{100}{n_{c_P^M}}\sum_{i=1}^{n_{c_P^M}}\left(\frac{\Delta c_P^M}{c^M_{P,\exp}}\right)_i \text{ with } \Delta c_P^M = \left|c^M_{P,\exp} - c^M_{P,cal}\right| \quad (25)$$

The pure fluid physical properties (T_c, P_c and ω) used in the optimization originate from two sources. It has been made reference to Poling et al. [33] for alkanes, aromatics, naphthenes, alkenes, CO_2, N_2, H_2S, H_2 and H_2O. Moreover, as some mercaptans and alkenes were missing in Poling's book, the DIPPR database was chosen instead. As recently discussed by Chiko and Polishuk [34], the disagreements between (T_c, P_c and ω) from different sources, which may exist for very heavy compounds, may substantially influence the results of PPR78. This is because the critical pressures of very heavy components are very small so that a slight absolute difference between two values (let us say 0.5 bar) may lead to an important relative difference (around 10%). In the cases of compounds absent in both the book of Poling et al. and DIPPR, the only solution is to estimate the critical properties from a group-contribution method and the reader has to be aware that its accuracy on the estimated P_c can significantly affect the results. The list of the pure fluids considered to develop the *E*-PPR78 model can be found in Table S1 of the Supporting Information.

The fluid-phase-equilibrium data used at the basis of the optimization process of PPR78 are reported in publications [1–5,7–12]. All bibliographic sources of data used to optimize *E*-PPR78 model (including its extension to 40 groups) can be either found in Qian's thesis [16] or in some of our previous publications [17–21]; they are also reported in Table S3 of the Supporting Information and at the end of this paper. In order to quantify the accuracy with which the h^M and c_P^M data are correlated, we also defined the error function on h^M and c_P^M data ($f_{h^M+c_P^M}(\%)$) as:

$$f_{h^M+c_P^M}(\%) = \frac{f_{h^M}(\%)\cdot n_{h^M} + f_{c_P^M}(\%)\cdot n_{c_P^M}}{n_{h^M} + n_{c_P^M}} \quad (26)$$

Similarly, the error function on VLE data only, noted $f_{VLE}(\%)$ in Table 1 is expressed by Eq. (16) and the overall calculated error function on h^M, c_P^M and VLE data, noted $f_{tot}(\%)$ in Table 1 is given by Eq. (23).

2.3.2. Results

In order to show a comparison between the PPR78 and *E*-PPR78 EoS versions and thus to demonstrate the gain in accuracy underlying the introduction of mixing enthalpies and heat capacities in the optimization process, Table 1 presents the error functions presented in equations (16)-(26) obtained by the application of the PPR78 and *E*-PPR78 models to the binary data used to optimise the A_{kl} and B_{kl} G-bips relative to 21 first groups that allow to define 10 classes of molecules: Alkanes (groups: G1-G6), Aromatics (groups: G7-G9), Naphthenes (groups: G10-G11), CO_2 (group G12), N_2 (group G13), H_2S (group G14), Mercaptans (group G15), H_2O (group G16), Alkenes (groups G17-G20), H_2 (group G21).

It is specified that the optimal A_{kl} and B_{kl} G-bips suitable for the *E*-PPR78 model and determined according to the optimisation method presented in section 2.3.1 (minimization of Eq. (23)) are reported in Table S4 of the Supporting Information. Such parameters coincide with the ones published in our previous papers devoted to the *E*-PPR78 model [17–21]. We however decided to take advantage of the publication of this article to partially complete the G-bips matrix given in Table S4. We indeed found enough experimental data to determine the missing interactions between 3 pairs of groups: [G12(CO_2) and G15(–SH)], [G13(N_2) and G15(SH)] and [G3($CH_{alkanes}$) and G20($CH_{cycloalkenic}$/-$C_{cycloalkenic}$)].

For each considered type of VLE data reported in Table 1 (for example, liquid phase composition data), we present in the same table: 1) the total number of considered experimental data points ($n_{datapoints}$) and 2) the number of data (for liquid phase composition: n_b) for which a deviation between calculated and experimental value could be calculated, i.e., that results to be not-"out-of-model" from the application of the original or enhanced version of the PPR78 model (for liquid phase composition: $n_{b\ PPR78}$ and $n_{b\ E-PPR78}$). To fix the ideas and understand in which occasion an experimental data point is declared "out of model" (i.e., not calculable by the model), let us take an example by considering a model which does not predict the existence of a homogeneous azeotrope that is however experimentally observed. In such a case, the deviations between calculated and experimental azeotropic pressure and composition cannot be evaluated and the corresponding experimental data point is said to be "out of model" (for more examples, the reader is referred to [35–37]).

To simplify the comparison between the PPR78 and *E*-PPR78 models, Figs. 2 and 3 graphically show the error functions on VLE (Eq. (16)) and mixing properties functions (Eq. (26)) reported in Table 1.

2.3.3. Analysis

Numerical results reported in the previous section reveal how the accuracy of the model on VLE calculations and on mixing properties evolves when including mixing properties in the optimization process. In spite of the simultaneous correlation of VLE, h^M and c_P^M data during the development of the *E*-PPR78 model, the error function over all the VLE data (10 families of molecules) with such a model ($f_{VLE_{E-PPR78}} = 8.77$ %) is exactly the same (less than 0.2% difference) than that obtained with the original PPR78 model ($f_{VLE_{PPR78}} = 8.62$ %) where only VLE data were considered for model optimisation. In other words, the inclusion of h^M and c_P^M data in addition to fluid-phase equilibrium data during the optimisation of the G-bips does not deteriorate the accuracy in predicting VLE properties. This is an extremely significant and not-easily anticipable result. Another key result requires discussion: although the accuracy on VLE data remains the same, the inclusion of h^M and c_P^M in the optimization process makes drastically decrease the deviations on both mixing properties data. Table1 shows that the error functions on h^M and c_P^M (Eqs. (24)-(25)) are remarkably improved; they are divided by a factor 11 and 55 respectively. We can thus conclude to the absolute necessity to include h^M and c_P^M data in the fitting procedure if we want to catch, in addition to VLE data, such properties with the PR EoS. Such results confirm our insight regarding the role and influence of the A_{kl} and B_{kl} parameters on the reproduction of VLE and energetic property data, respectively (see section 2.2)

The major achievements occur for h^M data points in the liquid single-phase region and for all c_P^M data points (available only in the liquid single-phase region). The improvements in h^M in the gaseous single-phase region and those referring to the two-phase region are less significant since such data were already accurately correlated by the PPR78 model.

Although the simultaneous inclusion of h^M, c_P^M and VLE data has been carried out during the fitting procedure aimed at determining the G-bips of the *E*-PPR78 model, the error functions on h^M and c_P^M are still much higher than that of VLE, which can be explained by the reasons as follows:

(1) The set of experimental h^M and c_P^M data reported in the literatures are sometimes discordant and scattered.
(2) Many liquid mixtures (especially those that contain H_2) slightly deviate from an ideal behaviour so that calculated

Table 1
Error functions by class of compounds obtained with PPR78 model and *E*-PPR78 model. A total of 135,186 data points from 1051 binary systems were considered.

Type of data		**Alkanes** (G1-G6)	**Aromatics** (G7-G9)	**Naphthenes** (G10-G11)	**CO_2** (G12)	**N_2** (G13)	**H_2S** (G14)	**Mercaptans** (G15)	**H_2O** (G16)	**Alkenes** (G17-G20)	**H_2** (G21)	**Total** (G1-G21)
Liquid phase composition	$n_{datapoints}$	9205	6883	5051	8956	4362	2351	775	4986	9123	5534	57226
	$n_{b\ PPR78}$	9084	6700	4902	8661	4305	2220	751	4693	8924	5517	55757
	$n_{b\ E\text{-}PPR78}$	9094	6701	4905	8641	4296	2231	753	4677	8911	5514	55723
	$f_{b\ PPR78}$ (%), eq. (17)	3.77	5.99	3.92	7.46	9.80	10.20	9.93	28.28	8.95	8.68	8.81
	$f_{b\ E\text{-}PPR78}$ (%), eq. (17)	4.00	6.23	4.10	7.85	9.74	10.01	10.76	28.34	9.30	8.83	9.02
Gas phase composition	$n_{datapoints}$	6995	4864	3438	7268	4680	1651	364	3875	6268	3905	43308
	$n_{d\ PPR78}$	6894	4752	3309	6785	4631	1558	339	3437	6048	3835	41588
	$n_{d\ E\text{-}PPR78}$	6898	4753	3314	6752	4630	1562	345	3431	6033	3833	41551
	$f_{d\ PPR78}$ (%), eq. (18)	5.94	6.35	4.53	8.72	7.23	7.71	8.37	16.48	10.83	10.30	8.54
	$f_{d\ E\text{-}PPR78}$ (%), eq. (18)	5.94	6.51	4.63	8.65	7.30	7.71	9.51	16.52	11.19	10.29	8.63
Critical point composition	$n_{datapoints}$	159	146	347	274	97	67	0	159	143	114	1506
	$n_{c\ PPR78}$	159	146	345	263	97	62	0	99	142	104	1417
	$n_{c\ E\text{-}PPR78}$	158	146	347	262	96	62	0	100	142	103	1416
	$f_{zc\ PPR78}$ (%), eq. (19)	5.99	6.53	2.84	7.44	8.99	8.92	0.00	18.88	10.29	5.95	7.21
	$f_{zc\ E\text{-}PPR78}$ (%), eq. (19)	6.17	5.00	4.42	6.75	8.94	9.49	0.00	18.49	10.84	5.83	7.37
Critical point pressure	$n_{datapoints}$	159	146	347	274	97	67	0	159	143	114	1506
	$n_{c\ PPR78}$	159	146	345	263	97	62	0	99	142	104	1417
	$n_{c\ E\text{-}PPR78}$	158	146	347	262	96	62	0	100	142	103	1416
	$f_{Pc\ PPR78}$ (%), eq. (20)	4.18	4.97	1.51	3.28	6.33	3.02	0.00	21.33	3.49	10.11	5.11
	$f_{Pc\ E\text{-}PPR78}$(%), eq. (20)	4.05	4.26	1.59	3.21	5.66	3.72	0.00	21.32	3.39	9.30	4.94
Azeotropic point composition	$n_{datapoints}$	0	0	0	0	0	0	58	0	0	0	58
	$n_{az\ PPR78}$	0	0	0	0	0	0	51	0	0	0	51
	$n_{az\ E\text{-}PPR78}$	0	0	0	0	0	0	52	0	0	0	52
	$f_{zAz\ PPR78}$ (%), eq. (21)	0.00	0.00	0.00	0.00	0.00	0.00	20.80	0.00	0.00	0.00	20.80
	$f_{zAz\ E\text{-}PPR78}$ (%), eq. (21)	0.00	0.00	0.00	0.00	0.00	0.00	13.45	0.00	0.00	0.00	13.45
Azeotropic point pressure	$n_{datapoints}$	0	0	0	0	0	0	58	0	0	0	58
	$n_{az\ PPR78}$	0	0	0	0	0	0	51	0	0	0	51
	$n_{az\ E\text{-}PPR78}$	0	0	0	0	0	0	52	0	0	0	52
	$f_{PAz\ PPR78}$ (%), eq. (22)	0.00	0.00	0.00	0.00	0.00	0.00	2.02	0.00	0.00	0.00	2.02
	$f_{PAz\ E\text{-}PPR78}$ (%), eq. (22)	0.00	0.00	0.00	0.00	0.00	0.00	1.16	0.00	0.00	0.00	1.16
All VLE data	$n_{datapoints}$	16518	12039	9183	16772	9236	4136	1255	9179	15677	9667	103662
	$n_{VLE\ PPR78}$	16296	11744	8901	15972	9130	3902	1192	8328	15256	9560	100281
	$n_{VLE\ E\text{-}PPR78}$	16308	11746	8913	15917	9118	3917	1202	8308	15228	9553	100210
	$f_{VLE\ PPR78}$ (%), eq. (16)	4.71	6.13	4.01	7.93	8.45	9.07	9.61	23.21	9.66	9.31	**8.62**
	$f_{VLE\ E\text{-}PPR78}$ (%), eq. (16)	4.84	6.30	4.21	8.10	8.45	8.99	10.10	23.26	10.01	9.39	**8.77**
Mixing enthalpy	$n_{datapoints}$	3405	5544	8368	4952	900	39	246	1631	3621	567	29273
	$f{h^M}_{PPR78}$ (%), eq. (24)	628.7	86.3	1481.6	65.2	13.5	56.4	362.5	81.4	235.2	21.4	561.6
	$f{h^M}_{E\text{-}PPR78}$(%), eq. (24)	48.8	34.2	65.0	45.8	11.6	54.3	27.5	81.4	40.0	21.4	49.0
Mixing heat capacity	$n_{datapoints}$	312	869	1070	0	0	0	0	0	0	0	2251
	$fc_p{}^M{}_{PPR78}$ (%), eq. (25)	327.75	3115.04	6802.34	0.00	0.00	0.00	0.00	0.00	0.00	0.00	4481.44
	$fc_p{}^M{}_{E\text{-}PPR78}$ (%), eq. (25)	25.73	133.83	56.81	0.00	0.00	0.00	0.00	0.00	0.00	0.00	82.24
All mixing properties	$n_{datapoints}$	3717	6413	9438	4952	900	39	246	1631	3621	567	31524
	$f{h^M}+c_p{}^M{}_{PPR78}$ (%), eq.(26)	603.44	496.69	2084.79	65.24	13.47	56.42	362.50	81.40	235.19	21.40	**841.51**
	$f{h^M}+c_p{}^M{}_{E\text{-}PPR78}$ (%), eq.(26)	46.84	47.70	64.10	45.78	11.60	54.28	27.54	81.37	40.03	21.40	**51.42**
All data	$\boldsymbol{n_{datapoints}}$	*20235*	*18452*	*18621*	*21724*	*10136*	*4175*	*1501*	*10810*	*19298*	*10234*	*135186*
	$\boldsymbol{n_{tot\ PPR78}}$	*20013*	*18157*	*18339*	*20924*	*10030*	*3941*	*1438*	*9959*	*18877*	*10127*	*131805*
	$\boldsymbol{n_{tot\ E\text{-}PPR78}}$	*20025*	*18159*	*18351*	*20869*	*10018*	*3956*	*1448*	*9939*	*18849*	*10120*	*131734*
	$\boldsymbol{f_{tot\ PPR78}}$ **(%)**, eq. (23)	***115.92***	***179.40***	***1074.86***	***21.49***	***8.90***	***9.54***	***69.98***	***32.74***	***52.92***	***9.99***	***207.83***
	$\boldsymbol{f_{tot\ E\text{-}PPR78}}$ **(%)**, eq. (23)	***12.63***	***20.92***	***35.01***	***17.04***	***8.73***	***9.44***	***13.06***	***32.79***	***15.77***	***10.06***	***18.98***

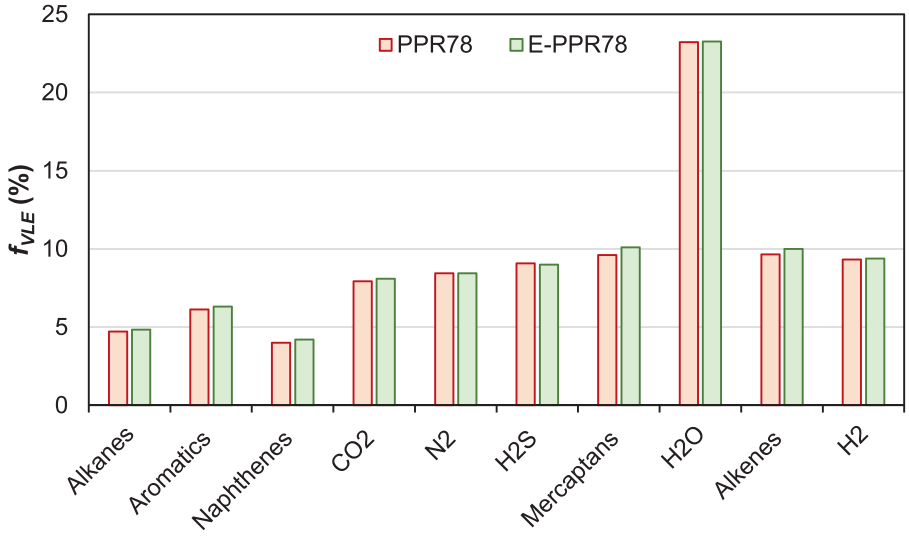

Fig. 2. Histograms of error functions on VLE data, for each of the 10 considered classes of molecules, for the PPR78 and the E-PPR78 model.

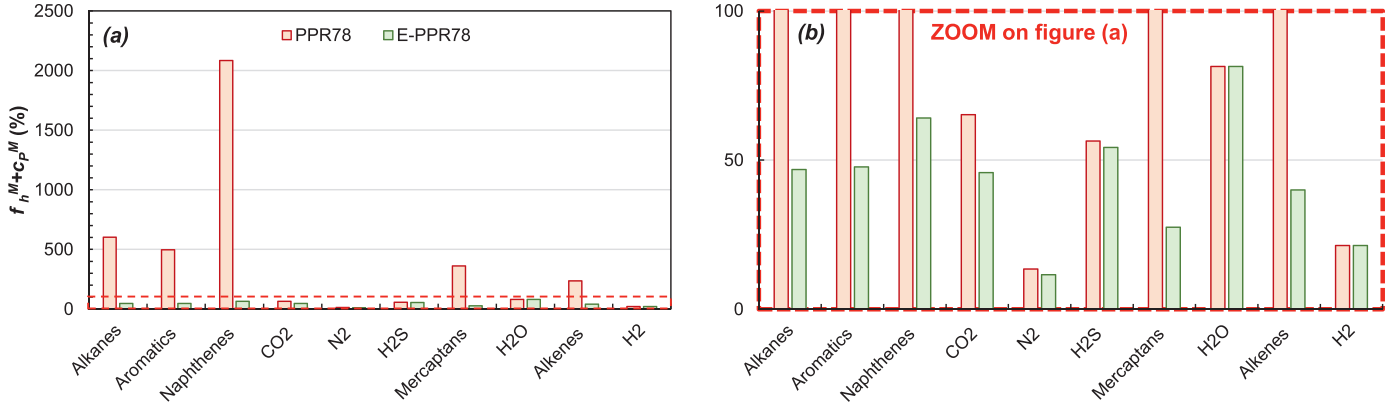

Fig. 3. Histograms of overall error functions on mixing enthalpy and heat capacity data, for each of the 10 considered classes of molecules, for the PPR78 and the E-PPR78 model. Fig. (b) is the zoom of the part of Fig. (a) in a dotted red square.

and experimental values of h^M and c_P^M are close to zero. Due to the definition of relative errors functions on h^M and c_P^M where absolute errors are standardized by experimental values (see Eqs. (24)-(25)), such small magnitudes inevitably increase these error functions.

(3) Binary systems in the gaseous single-phase region at low temperature and under one atmosphere show small endothermic mixing effect which is not easy to be well predicted by the PR EoS.

Considering that 1051 different binary mixtures have been investigated for the definition of 21 groups and determination of 420 G-bips (210 A_{kl} and 210 B_{kl}) for both the PPR78 and the *E*-PPR78 models, we present only some examples to illustrate the modification of model accuracy when including h^M and c_P^M data in the optimization process. Taking for example the binary mixture benzene (1) + isooctane (2), we plotted in Fig. 4 the predictions of VLE, h^M and c_P^M, returned by the original PPR78 model (dashed line) and its enhanced version *E*-PPR78 (solid line). As expected, with *E*-PPR78, the accuracy of predicted $P-xy$ curves at six different temperatures is retained (Fig. 4-a) and the predicted h^M-x and c_P^M-x curves in the liquid singe-phase region are in much better agreement with experimental data (see Fig. 4-b,c). Fig. 4.d shows that in the temperature range where experimental data are available (between 298 and 348 K), the PPR78 and *E*-PPR78 models return similar k_{ij} values so that the 2 models correlate with the same accuracy the VLE data. At the same time, the slopes of the curves k_{ij} vs. T are totally different. A much steeper curve is obtained with PPR78 in comparison to *E*-PPR78 so that dk_{ij}/dT take strongly different values with both models explaining why the h^M values returned by the 2 models are so different. Fig. 4.e highlights that the slopes of the curves dk_{ij}/dT vs. T calculated with the 2 models are even much different than those of the curves k_{ij} vs. T. The immediate consequence is the calculation of totally different d^2k_{ij}/dT^2 values by both models leading to a huge difference in the c_P^M values returned by both models.

After our examination over all the mixtures, we have found that by using the parameters (A_{kl}, B_{kl}) determined for the *E*-PPR78 model, both dk_{ij}/dT and d^2k_{ij}/dT^2 for most of the binary mixtures at low and moderate temperatures become less temperature-dependent showing a magnitude less significant, as observed for the (benzene (1) + isooctane (2)) system. Counter examples can however be found.

The key conclusion of this paper is that the *E*-PPR78 model can give a better estimation of the dk_{ij}/dT and d^2k_{ij}/dT^2 functions than the original PPR78 model explaining why better predictions of h^M and c_P^M are obtained. The good prediction of VLE data is retained because the k_{ij} values returned by both models are similar.

Table 2
Error functions by class of compounds obtained with *E*-PPR78.

Type of data		Alkanes (G1-G6)	Aromatics (G7-G9)	Naphthenes (G10-G11)	CO_2 (G12)	N_2 (G13)	H_2S (G14)	Mercaptans (G15)	H_2O (G16)	Alkenes (G17-G20)	H_2 (G21)	Freons (G22-G27)	CO (G28)	He (G29)	Ar (G30)	SO_2 (G31)	O_2 (G32)	NO (G33)	COS (G34)	NH_3 (G35)	NO_2 (G36)	N_2O (G37)	Alkynes (G38-G40)	Total (G1-G40)
Liquid phase comp.	$n_{datapoints}$	9205	6883	5051	8956	4362	2351	775	4986	9123	5534	2214	1831	2599	2434	468	1770	157	184	2035	76	314	1015	72323
	n_b	9094	6701	4905	8641	4296	2231	753	4677	8911	5514	1993	1807	2327	2331	467	1713	157	180	1971	76	309	966	70020
	f_b(%)	4.00	6.23	4.10	7.85	9.74	10.01	10.76	28.34	9.30	8.83	11.52	8.95	12.30	9.82	8.97	6.76	12.57	12.29	5.40	2.27	9.23	8.82	9.07
Gas phase comp.	$n_{datapoints}$	6995	4864	3438	7268	4680	1651	364	3875	6268	3905	1939	1408	2662	1838	287	1402	140	22	1405	1	364	428	55204
	n_d	6898	4753	3314	6752	4630	1562	345	3431	6033	3833	1766	1350	2369	1730	272	1343	137	22	1354	1	359	418	52672
	f_d(%)	5.94	6.51	4.63	8.65	7.30	7.71	9.51	16.52	11.19	10.29	6.77	9.26	13.66	12.27	10.01	5.82	7.26	2.68	10.93	27.95	6.25	7.97	8.89
Critical point comp.	$n_{datapoints}$	159	146	347	274	97	67	0	159	143	114	92	34	31	23	6	8	2	8	16	0	20	4	1750
	n_c	158	146	347	262	96	62	0	100	142	103	88	34	23	17	5	8	2	8	11	0	20	4	1636
	f_{z_c}(%)	6.17	5.00	4.42	6.75	8.94	9.49	0.00	18.49	10.84	5.83	16.77	19.50	21.70	112.96	1.44	8.85	10.33	15.86	14.51	0.00	16.34	2.10	9.61
Critical point pressure	$n_{datapoints}$	159	146	347	274	97	67	0	159	143	114	92	34	31	23	6	8	2	8	16	0	20	4	1750
	n_c	158	146	347	262	96	62	0	100	142	103	88	34	23	17	5	8	2	8	11	0	20	4	1636
	f_{P_c}(%)	4.05	4.26	1.59	3.21	5.66	3.72	0.00	21.32	3.39	9.30	3.19	10.84	43.26	63.59	7.04	7.45	33.01	2.03	9.02	0.00	6.94	0.44	6.20
Azeotropic point comp.	$n_{datapoints}$	0	0	0	0	0	0	58	0	0	0	0	0	0	0	20	0	0	4	0	0	0	8	90
	n_{az}	0	0	0	0	0	0	52	0	0	0	0	0	0	0	20	0	0	4	0	0	0	8	84
	$f_{z_{az}}$(%)	0.00	0.00	0.00	0.00	0.00	0.00	13.45	0.00	0.00	0.00	0.00	0.00	0.00	0.00	15.34	0.00	0.00	5.36	0.00	0.00	0.00	8.07	13.00
Azeotropic point pressure	$n_{datapoints}$	0	0	0	0	0	0	58	0	0	0	0	0	0	0	20	0	0	4	0	0	0	8	90
	n_{az}	0	0	0	0	0	0	52	0	0	0	0	0	0	0	20	0	0	4	0	0	0	8	84
	$f_{P_{az}}$(%)	0.00	0.00	0.00	0.00	0.00	0.00	1.16	0.00	0.00	0.00	0.00	0.00	0.00	0.00	8.23	0.00	0.00	0.96	0.00	0.00	0.00	2.47	2.96
All VLE data	$n_{datapoints}$	16518	12039	9183	16772	9236	4136	1255	9179	15677	9667	4337	3307	5323	4318	807	3188	301	230	3472	77	718	1467	131207
	n_{VLE}	16308	11746	8913	15917	9118	3917	1202	8308	15228	9553	3935	3225	4742	4095	789	3072	298	226	3347	77	708	1408	126132
	$\boldsymbol{f_{VLE}}$ **(%)**	**4.84**	**6.30**	**4.21**	**8.10**	**8.45**	**8.99**	**10.10**	**23.26**	**10.01**	**9.39**	**9.32**	**9.21**	**13.17**	**11.51**	**9.41**	**6.35**	**10.25**	**10.80**	**7.68**	**2.60**	**7.86**	**8.49**	**8.97**
Mixing enthalpies	$n_{datapoints}$	3405	5544	8368	4952	900	39	246	1631	3621	567	0	44	1	878	0	56	0	0	252	0	104	770	31378
	f_{hM}(%)	48.77	34.20	65.03	45.78	11.60	54.28	246	81.37	40.03	21.40	0.00	9.21	7.97	45.67	0.00	11.74	0.00	0.00	27.21	0.00	104.95	23.38	48.21
	ΔT_h(K)	**0.171**	**0.205**	**0.193**	**1.075**	**0.775**	**1.265**	**0.183**	**6.540**	**0.293**	**2.060**	**0.000**	**0.081**	**0.032**	**2.042**	**0.000**	**0.114**	**0.000**	**0.000**	**1.858**	**0.000**	**0.284**	**0.41**	**0.795**
Mixing heat capacities	$n_{datapoints}$	312	869	1070	0	0	0	27.54	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2251
	$f_{c_P^M}$(%)	25.73	133.83	56.81	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	82.24
All mixing properties	$n_{datapoints}$	3717	6413	9438	4952	900	39	0	1631	3621	567	0	44	1	878	0	56	0	0	252	0	104	770	33629
	$f_{h_M+c_P^M}$(%)	**46.84**	**47.70**	**64.10**	**45.78**	**11.60**	**54.28**	0.00	**81.37**	**40.03**	**21.40**	0.00	**9.21**	**7.97**	**45.67**	0.00	**11.74**	0.00	0.00	**27.21**	0.00	**104.95**	**23.38**	**50.49**
All data	$\boldsymbol{n_{datapoints}}$	*20235*	*18452*	*18621*	*21724*	*10136*	*4175*	1501	*10810*	19298	*10234*	*4337*	*3351*	*5324*	*5196*	*807*	*3244*	*301*	*230*	*3724*	*77*	*822*	2237	164836
	$\boldsymbol{n_{tot}}$	*20025*	*18159*	*18351*	*20869*	*10018*	*3956*	1448	*9939*	18849	*10120*	*3935*	*3269*	*4743*	*4973*	*789*	*3128*	*298*	*226*	*3599*	*77*	*812*	2178	159761
	$\boldsymbol{f_{tot}}$ **(%)**	***12.63***	***20.92***	***35.01***	***17.04***	***8.73***	***9.44***	***13.06***	***32.79***	***15.77***	***10.06***	***9.32***	***9.21***	***13.17***	***17.54***	***9.41***	***6.45***	***10.25***	***10.80***	***9.05***	***2.60***	***20.29***	**13.75**	***17.71***

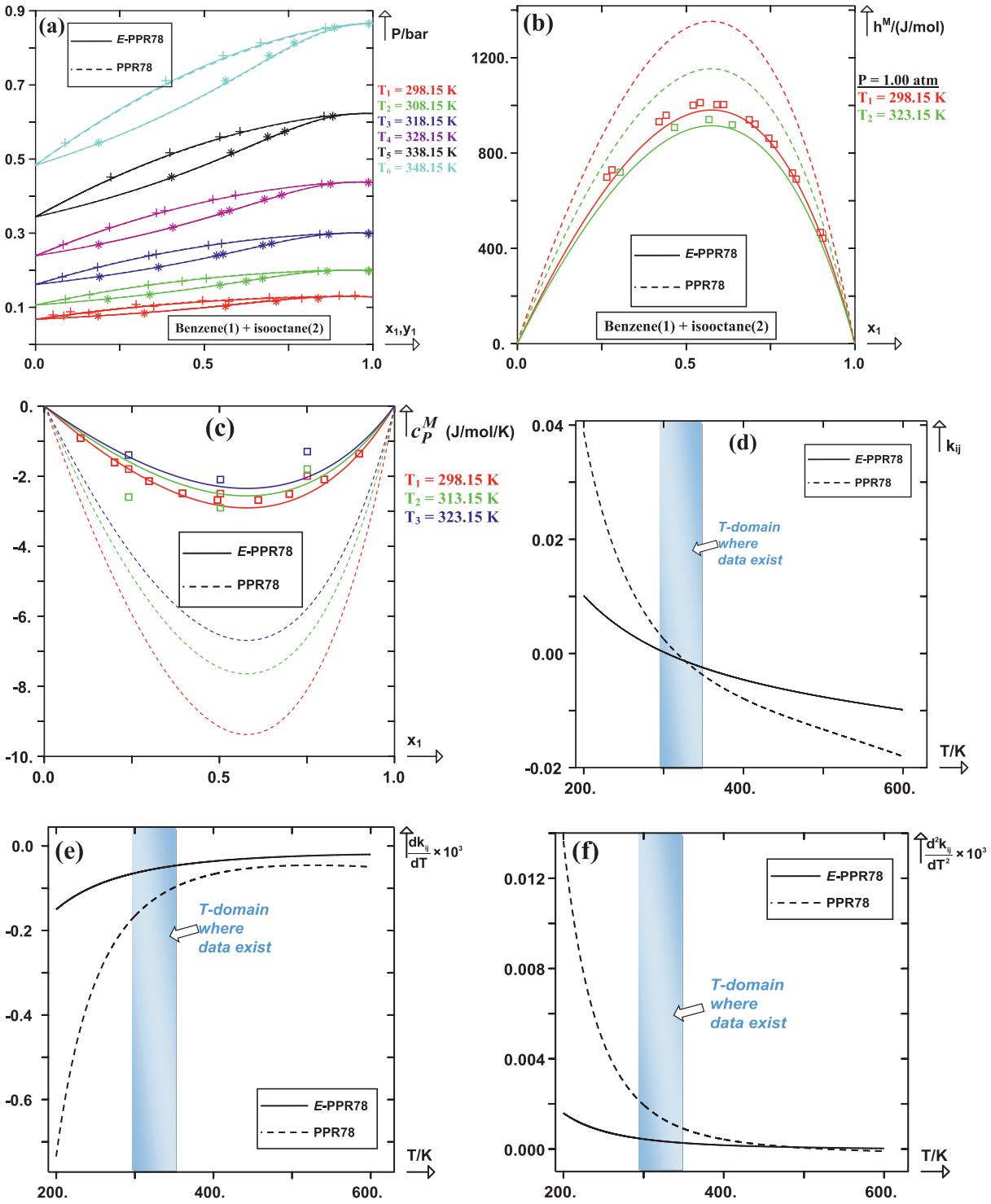

Fig. 4. Prediction of P-xy, h^M-x, c_P^M-x k_{ij}-T, dk_{ij}/dT-T and d^2k_{ij}/dT^2-T curves for the binary system: (benzene(1) + isooctane(2)). (+) experimental bubble points, (*) experimental dew points, (□) experimental h^M and c_P^M points. Solid line: predicted curves with the *E*-PPR78 model. Dashed line: predicted curves with the original PPR78 model. **(a)** Predicted P-xy curves at six different temperatures: $T_1 = 298.15$ K, $T_2 = 308.15$ K, $T_3 = 318.15$ K, $T_4 = 328.15$ K, $T_5 = 338.15$ K, $T_6 = 348.15$ K. **(b)** Predicted h^M-x curves at two different temperatures: $T_1 = 298.15$ K, $T_2 = 323.15$ K. **(c)** Predicted c_P^M-x curves at three different temperatures: $T_1 = 298.15$ K, $T_2 = 313.15$ K $T_3 = 323.15$ K. **(d)** k_{ij}-T curves calculated with the recent model and the original one. **(e)** dk_{ij}/dT-T curves calculated with the recent model and the original one. **(f)** d^2k_{ij}/dT^2-T curves calculated with the recent model and the original one.

2.4. The 40-GC-E-PPR78

Considering the positive results obtained when optimising the G-bips for 21 groups over, simultaneously, phase equilibrium, mixing enthalpies and heat capacities data, the *E*-PPR78 model has been furthermore developed by optimising G-bips for 19 more groups. The objective function which has been considered in the optimisation process is eq. (23) and the corresponding A_{kl} and B_{kl} G-bips are reported in Table S4 of the Supporting Information.

Table S1 in the Supporting Information presents the list of the 204 pure components involved in the development of the *E*-PPR78 model. For each binary system considered in the development of the *E*-PPR78 model, Table S2 contains the main features of collected experimental data, extracted from references [38–2068]. Bibliographic references are reported in Table S3.

Table 2 reports the values of the error functions stemming from equations (16)-(26), resulting from the application of the *E*-PPR78 model to all the binary systems used in the optimisation of the different G-bips between groups G1-G40. As expected, aqueous systems show the highest relative errors on VLE data while mixtures of hydrocarbons show lowest deviations. To provide a more explicit quantification of the deviations on mixing enthalpies, we also provide in Table 2 the average value of the temperature deviation (ΔT_h) corresponding to a deviation in mixing enthalpy:

$$\Delta T_h = \frac{1}{n_{h^M}} \sum_{i=1}^{n_{h^M}} \Delta T_i \ with \ \Delta T_i = \left| h^M_{exp} - h^M_{cal} \right| / c_P \tag{27}$$

where c_P is the molar heat capacity of the binary mixture, ΔT_i, is the temperature change for one experimental enthalpy data point. Indeed, as discussed in a previous publication [15], $f_{h^M}(\%)$ and ΔT_h criteria « *are strictly non-equivalent to discuss the capacity of a model to predict mixing enthalpy data. […] the* ΔT_h *criterion is definitively preferred because it provides a fair overview on the model accuracy to perform an energy balance*. » Indeed, as mentioned in section 2.3.3 relative error function ($f_{h^M}(\%)$) can be very high when enthalpies of mixing are very small, although the corresponding temperature change of the fluid (ΔT_h) due to such an error is very small.

Table S5 – Table S8 of the supporting information show the deviations between calculated and experimental data calculated with the *E*-PPR78 model on all types of data points following a further organisation of binary systems: error functions are reported grouping binary systems according to the type of each molecule of the mixture considered in the development of this model. Ten types of molecules are considered: alkanes, alkenes, alkynes, aromatics, naphthenes, freons, sulfur-containing compounds (mercaptans, H_2S, SO_2, COS), non-permanent gases (NH_3, CO_2, C_2H_6, C_2H_4, C_2H_2, NO, NO_2, N_2O), permanent gases (N_2, Ar, O_2, CO, He, CH_4, H_2), water.

The analysis of Table S5 – Table S8 shows that the highest deviations on VLE data are due to the presence of either water or aromatics in the mixture. As regards aromatics, this statement is valid for mixtures of aromatics and non-hydrocarbons. Also, it is possible to confirm the more reliable assessment of temperature deviations (Eq. (27)) due to inaccuracies on mixing enthalpies, rather than relative error function on mixing enthalpies (Eq. (24)), to evaluate the suitability of a model in accurately modelling energy balances. Indeed, naphthenes-naphthenes systems are characterized by an error function on mixing enthalpy equal to 276.50 % (very high) and $\Delta T_h = 0.136$ K (very low), while the opposite conclusion is for aromatics-water, for which the error function on mixing enthalpy is equal to 45.48 % (quite low) and $\Delta T_h = 10.097$ K (extremely high). The temperature deviation criterion is the one that better represents the quality and the accuracy of the model when applied to perform energy balances. These tables provide to the reader interested in using the *E*-PPR78 EoS a closer view on the errors made by the model in representing the data of binary systems.

3. Conclusion

This paper has shown the impact of including change-on-mixing derived property data (mixing enthalpies and mixing heat capacities) other than classically used VLE data, when optimising thermodynamic models. The key conclusion is that the inclusion of such data in such an optimisation methodology enables a significant improvement of mixing property prediction without deteriorating the accuracy in predicting VLE properties. Ten years ago, this observation led some of the authors of this paper to think about modifying the methodology at the basis of the development of the PPR78 EoS for identifying optimal bips between groups. This idea has led to the development of a model which is known today as *Enhanced*-PPR78 (*E*-PPR78). This paper explicitly invites model developers to consider mixing enthalpies and mixing heat capacities when parameterizing an EoS.

Finally, a complete overview on the accuracy of the state-of-the-art *E*-PPR78 model, based on 40 groups, is provided. Considering the predictive character of this model, the deviations are acceptably low for all the systems, except for water-hydrocarbons mixtures. Finally, this paper presents three never published (A_{kl}, B_{kl})-couples of G-bips, between group 12 (CO_2) and group 15 (-SH), group 13 (N_2) and group 15 (-SH), group 3 (-$CH_{alkanes}$) and group 20 (-$CH_{cycloalkenic}$/-$C_{cycloalkenic}$).

Eventually, it can be mentioned that these thoughts about EoS parameter estimation are taking part in a series of recent research works performed by our group and dealing with the general issue of parameterization [2069,2070] and accuracy assessment [2071] of cubic and SAFT EoS. In particular, regarding cubic models, proof was given that alpha functions involved in the attractive term also govern the prediction of VLE data in mixtures [28,29]. Through the present paper and the previous ones on similar topics [2072], it is desired to highlight the best practices for developing safe EoS, making it possible to define optimal models for pure components and mixtures, belonging indifferently to the SAFT or the cubic EoS family.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Jean-Noël Jaubert: Conceptualization, Writing – review & editing, Software. **Jun-Wei Qian:** Software. **Silvia Lasala:** Software, Writing – review & editing. **Romain Privat:** Conceptualization, Software, Writing – review & editing.

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Supplementary materials

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