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A Novel Equilibrium Equation for Binary Mixtures

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ABSTRACT

In the thermodynamic of non-ideal solutions, there are two valuable approaches for linking the saturated vapor and liquid phases in an equilibrium system, i.e. the γ-φ and φ-φ approaches. Nevertheless, these methods are complex and need huge calculation procedures. Therefore, developing novel equations, with a simple format, is necessary for decreasing the size of the calculation procedure or increasing the accuracy of the predicted values for the required thermodynamic parameters. This study aims to develop a simple and accurate (with less than 5% mean absolute error for several cases of validation) relation among the thermodynamic parameters of two phases of a pure or binary mixture vapor-liquid equilibrium system. The format of this equation is very simple, which improves its applicability for solving various thermodynamic problems in vapor-liquid equilibrium systems. For this purpose, the compressibility factors of each phase that are suitable combinations of the PVT parameters of the system, are chosen for relating the thermodynamic parameters of these phases. Therefore, obtaining a relation between the compressibility factors of the saturated liquid and vapor phases relates the PVT parameters of these phases to each other. Although the idea of this theory-based equation has been raised from the investigation of equilibrium PVT values of pure substances, this equation has also been validated using the experimental PVTxy data of several binary mixtures. Comparing the predictions of the obtained equilibrium equation with the experimental data of pure and binary mixtures approves the accuracy of the predictions of the obtained equation.

1. Introduction

The ideal gas thermodynamic parameters, i.e. the temperature (T), pressure (P) and molar volume (v) have been related to each other using the ideal gas law (Pv=RT, where R is the universal gas constant), which is the equation of state of a hypothetical ideal gas and has a good approximation of the behavior of many gases under many conditions, although it has several limitations [1]. For the identification of the non-ideal behavior of gases, different empirical [2-6] or theoretical equations have been developed for obtaining this PVT relation based on its compressibility factor

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(z_v). Virial equation [7] is one of these theoretical equations that presents this factor as a power polynomial function of P or v. This compressibility factor is a dimensionless quantity, indicating how much a real fluid deviates from an ideal gas. Vander Waals [8], Redlich-Kwong [9], Soave-Redlich-Kwong [10] and Peng-Robinson [11] equations are other equations of state for non-ideal gases, which are based on a similar theory and are modifications of the ideal gas law and take into account both the volume of the gas molecules and the attractive forces acting between them. In the liquid state, Wilson [12], Van laar [13], Margules [14], UNIQUAC [15], UNIFAC [16], and NRTL [17] models present the non-ideal behavior of this single phase, which may lead to calculating the liquid compressibility factor (z_L). All of these equations are equations of state and calculate PVT relations for a single liquid or gas phase. In addition, some numerical studies for obtaining accurate equations with simple formats for predicting z_L [5, 6].

In vapor-liquid equilibria, we have two saturated phases with the same temperature and pressure and the fugacity of these phases is equal to each other. In ideal conditions for these two phases, Raoult [18], Dalton [19] and Henry [20] laws present acceptable relations between the thermodynamic parameters of these phases. The γ-φ and φ-φ approaches [21] are well-known methods for relating these two phases in non-ideal cases. These equations calculate the thermodynamic parameters of these phases, such as temperature (T), pressure (P), molar volume (v), or molar fractions of components in each one of the phases of an equilibrium system of mixtures (x or y). In addition, some numerical investigations have occurred in this field of thermodynamic science [22].

Measuring the values of the thermodynamic parameters are complex process requiring specific equipment and experiences with notable fix and operating costs. In addition, the measured values for these parameters have ignorable differences in different experimental studies [22-39], which improves the importance of the predictor equations. These methods are applicable for calculating each one of the PVTxy variables, separately. Nevertheless, these methods are complex and require a huge volume of calculations. In addition, they have limited ranges of application, for example, the Peng-Robinson equation is more suitable for hydrocarbon systems at low or medium pressures [21]. Therefore, obtaining suitable models for both of these phases in a certain equilibrium system, and solving some of these equations using numerical methods, is a usual method for predicting unknown thermodynamic parameters of a vapor-liquid equilibrium system.

In this study, an equation with a simple format has been presented that relates the compressibility factors of the saturated liquid and vapor phases in an equilibrium condition in a single or double-component system. This equation relates the PVT parameters of these phases and can be used as an equilibrium equation or state equation simultaneously. The novelty of this equation is that this equation is the only format of equilibria equation that presents a very simple format relation between the z_L and z_v. This accurate equation (with its simple format) is applicable for obtaining PVT parameters of each phase of an equilibrium system of a pure substance or a binary mixture (without any numerical solution complexity). This equation is also suitable for obtaining the PVTxy parameters in high pressures, which is the γ-φ approach limitation. In this method, for calculating the non-ideal behavior of the liquid phase, we need to know the dependency of the liquid molar volume on the pressure for obtaining the liquid fugacity [21]. Attending to the low dependency of this quantity on the pressure variations in the liquid state, in the low and medium range of pressure, the molar volume of the liquid phase has been considered as a constant value. But, at high pressures, this dependency is very complex attending to the influence of the liquid's intra-molecular forces. Presenting a relation between z_L and z_v, attending to the temperature and pressure equivalences between the liquid and vapor phases in the equilibria ($z_L = \frac{Pv_L}{RT}$ and $z_V = \frac{Pv_V}{RT}$), relates the liquid molar volume to the gas phase molar volume, which has several well-known equations for calculation. Therefore, one of the applications of the presented equation in this study is improving the applicability γ-φ approach in high-pressure utilization.

2. Theory

Attending to the non-ideal behaviors of saturated liquid and vapor phases of pure substances, there is a nonlinear dependency between the empirical values of these phases' compressibility factors (i.e. z_L and z_V, for each one of the saturated liquid and vapor phases, respectively). Fig. 1 presents an example of the investigated pure cases in this study [23]. As shown in this Figure, this dependency can be presented as an exponential function.

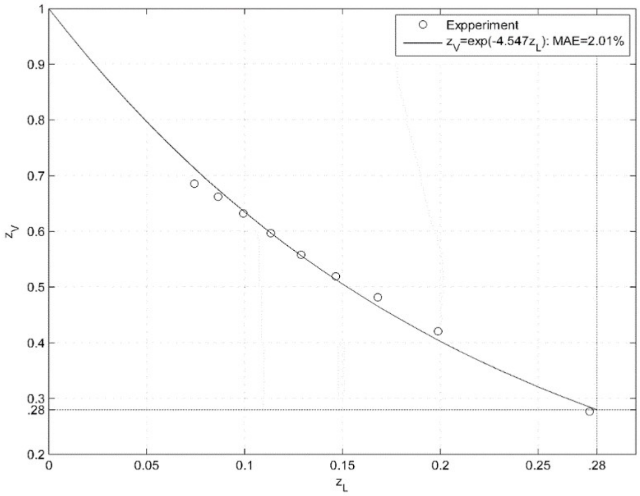

Fig. 1. Dependency of z_V to z_L in the CO_2 VLE system [23]

Therefore, the following equation format may be proposed as a relation between z_v and z_L for the VLE of a single-component system:

$$z_v = a\exp\left(bz_L\right) \tag{1}$$

where *a* and *b* are constant as or functions of the type of substance. Two boundary values of z_V and z_L are necessary for obtaining *a* and *b* in this relation. As shown in Fig. 1, the values of z_v and z_L converged to specific values in two boundary conditions as below:

$$at \quad z_L = 0 \ : \ z_v = 1 \ \Rightarrow \ a = 1 \tag{2}$$

$$at \quad z_L = z_c \ : \ z_v = z_c \ \Rightarrow \ b = \frac{Ln(z_c)}{z_c} \tag{3}$$

where z_c is the compressibility factor of the substance at its critical point. Therefore, equation (1), which relates the compressibility factors of two saturated phases of a VLE single (or double) component system, can be presented below:

$$z_v = \exp\left[\frac{\ln(z_c)}{z_c} z_L\right] \tag{4}$$

as previously described, one of the applications of the presented equation in this study is improving the γ-φ method applicability at high-pressure vapor-liquid equilibrium systems. In this method, through the procedure of obtaining the non-ideal behavior of the liquid phase, the following equation relates the fugacity of the liquid phase (f_L) to the liquid molar volume (v_L) [21]:

$$f_L = f^{sat}\exp(\int_{P^{sat}}^{P}\frac{v_L}{RT}dP) \tag{5}$$

Therefore, it is necessary to know the dependency of this molar volume on the pressure, especially at high-pressure ranges, where this dependency is not ignorable. Attending to the complexity of the liquid intra-molecular forces, there is no theoretical equation for this dependency. Attending to the temperature and pressure equivalence in vapor-liquid equilibria, the presented relation between z_L and z_v in equation (4), leads to a simple equation between v_L and v_v, as $\frac{Pv_L}{RT}=z_c\frac{\ln(\frac{Pv_v}{RT})}{\ln(z_c)}=z_c\ln(\frac{Pv_v}{RT}-z_c)$. It should be noted that several equations of gaseous state exist in different references [7-11] that present the v_v to P dependency. Therefore, using this equation gives a formula for calculating the liquid fugacity based on the vapor equation of states.

The simple format of this equation improves its applicability and makes it possible to develop a simple VLE equation for multi-component systems. In the next section, validating the accuracy of the predictions of this equation using a set of empirical VLE data approves its ability to relate PVT parameters of these saturated phases in a simple or double-component system.

3. Validation

Fig. 2 compares the model predictions and experimental data of saturated liquid and vapor phases for some pure substances [22-33]. Fig. 3 to Fig. 7 present similar validations for approving the accuracy of the predictions of this equation for some binary mixtures [23, 34-39]. Attending to the presented mean of absolute errors ($MAE=\left|z_V^{emperical}-z_V^{predicted}\right|/z_V^{emperical}$) in these Figures, it is clear that the presented equation has an acceptable ability to predict the values of z_V (or z_L) using the values of z_L (or z_V) for the pure substance or binary mixture in VLE systems.

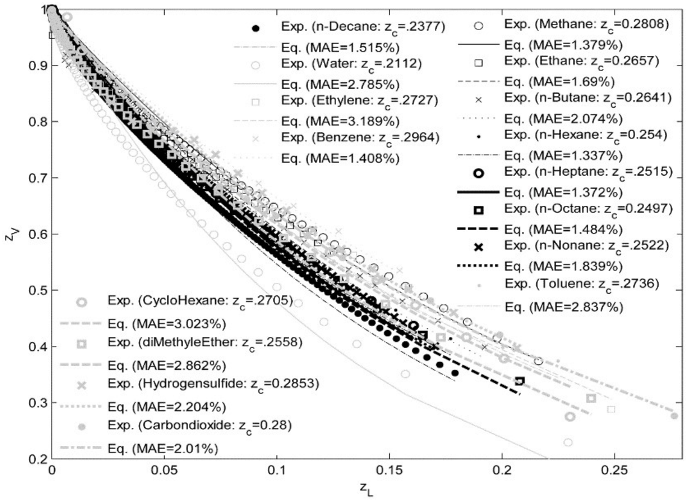

Fig. 2. Validation of the presented VLE equation using the experimental data of the saturated liquid and vapor phases of some pure substances [22-33]

These cases approve the presented relation between the PVT parameters of the saturated vapor and liquid phases of single or double-component systems. It is observed in Fig. 2 that the calculated MAE for the predicted values by the presented equation comparing with the experimental data of some pure substances are between 1.337% to 3.189% (with a mean value equal to 2.063%). These low values of the calculated mean absolute error for these cases approves the high accuracy of the presented equation for predicting the existed relation between z_L and z_V, for these pure substances.

As shown in Fig. 3, the calculated MAE for the predicted values by the presented equation comparing with the experimental data of different mole fractions of acetone (from x=0.514 to x=0.8) in n-pentane binary mixture are between 0.224% to 1.977% (with a mean value equal to 1.187%). These low values of the calculated mean absolute error for these cases approves the high accuracy of the presented equation for predicting the existed relation between z_L and z_V, for this binary mixture.

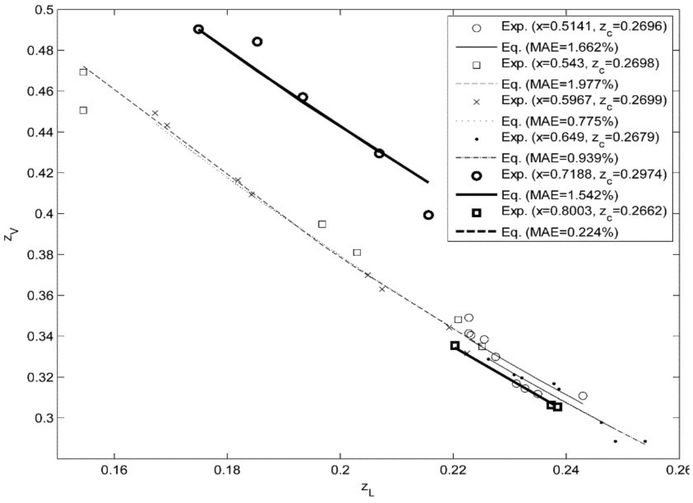

Fig. 3. Validation of the presented VLE equation using the experimental data of the saturated liquid and vapor phases of acetone(x) – n-pentane mixture [34]

As shown in Fig. 4, the calculated MAE for the predicted values by the presented equation comparing with the experimental data of different mole fractions of carbon dioxide (from x=0.063 to x=0.901) in hydrogen sulfide binary mixture are between 1.276% to 3.157% (with a mean value equal to 2.006%). These low values of the calculated mean absolute error for these cases approves the high accuracy of the presented equation for predicting the existed relation between z_L and z_V, for this binary mixture.

Fig. 5 shown the calculated MAE for the predicted values by the presented equation compared with the experimental data of different mole fractions of n-butane (from x=0.182 to x=0.818) in hydrogen sulfide binary mixture are between 3.48% to 6.368% (with a mean value equal to 4.824%). These values of the calculated mean absolute error for these cases approves the acceptable accuracy of the presented equation for predicting the existed relation between z_L and z_V, for this binary mixture.

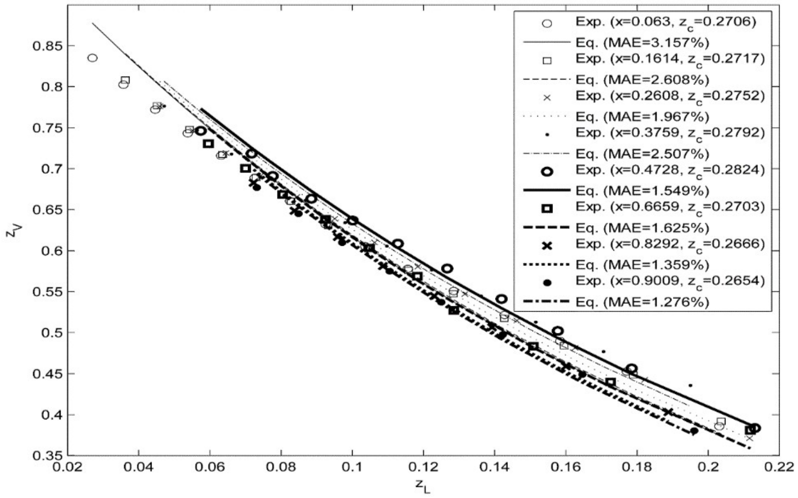

Fig. 4. Validation of the presented VLE equation using the experimental data of the saturated liquid and vapor phases of carbon dioxide(x) – hydrogen sulfide mixture [23]

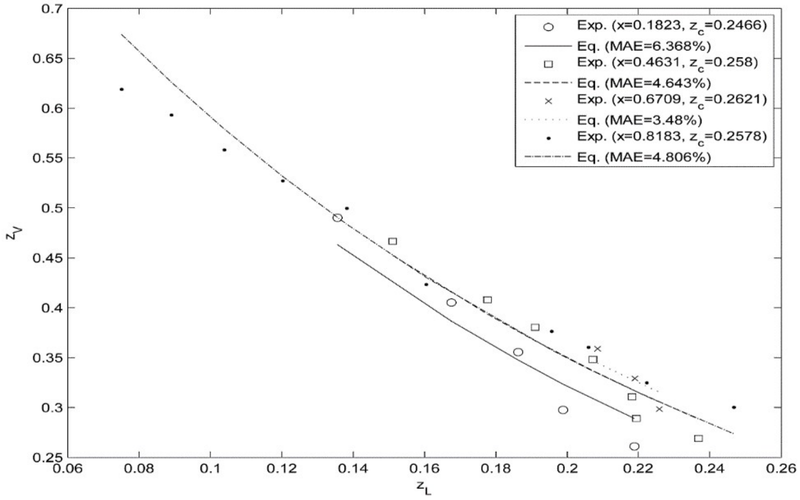

Fig. 5. Validation of the presented VLE equation using the experimental data of the saturated liquid and vapor phases of n-butane(x) – n-octane mixture [37]

As shown in Fig. 6(a), the calculated MAE for the predicted values by the presented equation comparing with the experimental data of different mole fractions of propane (from x=0.066 to x=0.848) in neopentane binary mixture are between 1.328% to 16.26% (with a mean value equal to 6.168%). These values of the calculated mean absolute error for these cases approves the acceptable accuracy of the presented equation for predicting the existed relation between z_L and z_V, for this binary mixture. It should be noted that increasing the concentration of propane in this binary mixture decreases the accuracy of the predictions of the presented equation.

Fig. 6(b) compares the predictions of the presented equation with the experimental data of different mole fractions of propane (from x=0.144 to x=0.922) in n-hexane binary mixture are between 1.069% to 2.078% (with a mean value equal to 1.504%). These low values of the calculated mean absolute error for these cases approves the high accuracy of the presented equation. Fig. 6(c) compares the predictions of the presented equation with the experimental data of different mole fractions of propane (from x=0.214 to x=0.959) in n-octane binary mixture are between 2.569% to 10.26% (with a mean value equal to 5.276%). These values of the calculated mean absolute error for these cases approves the acceptable accuracy of the presented equation for predicting the existed relation between z_L and z_V, for this binary mixture.

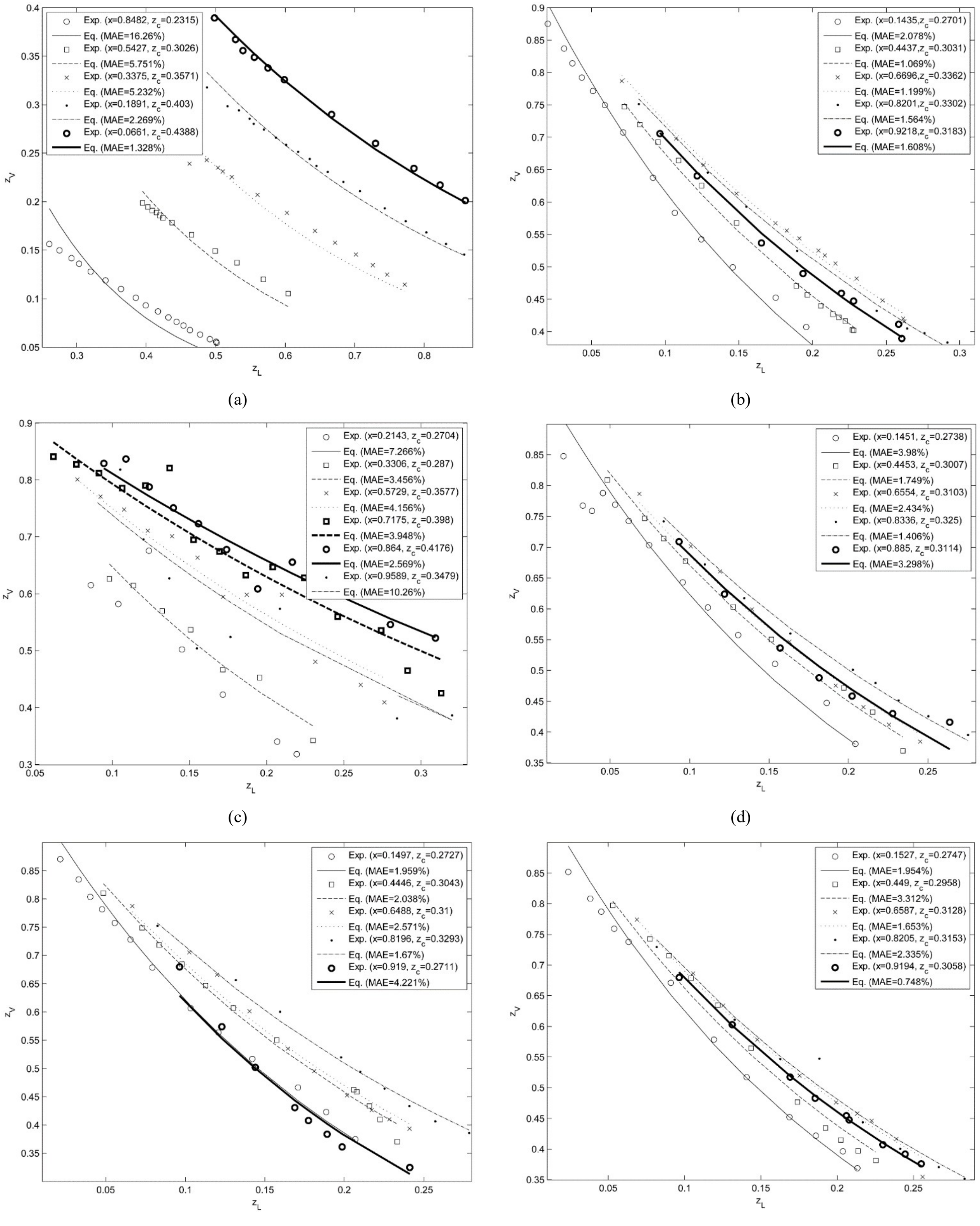

Fig. 6. Validation of the presented VLE equation using the experimental data of the saturated liquid and vapor phases of propane (x) – (a) neopentane, (b) n-hexane, (c) n-octane, (d) 3-methylebutane, (e) 2-methylepentane and (f) 2-2-dimethyl butane mixtures [35-37]

Fig. 6(d) compares the predictions of the presented equation with the experimental data of different mole fractions of propane (from x=0.145 to x=0.885) in 3-methylebutane binary mixture are between 1.406% to 3.98% (with a mean value equal to 2.573%). These low values of the calculated mean absolute error for these cases approves the high accuracy of the presented equation. The section (e) of this Figure compares the predictions of the presented equation with the experimental data of different mole fractions of propane (from x=0.15 to x=0.919) in 2-methylepentane binary mixture are between 1.67% to 4.221% (with a mean value equal to 2.478%). These low values of the calculated mean absolute error for these cases approves the high accuracy of the presented equation for predicting the existed relation between z_L and z_V, for this binary mixture. Fig. 6(f) compares the predictions of the presented equation with the experimental data of different mole fractions of Propane (from x=0.153 to x=0.919) in 2-2-dimethyl butane binary mixture are between 0.748% to 3.312% (with a mean value equal to 2%), which approves the high accuracy of the equation.

As shown in Fig. 7, the calculated mean value of the MAE for the predicted values by the presented equation comparing with the experimental data of different mole fractions of perfluoromethylcyclohexane (x) in n-hexane, 3-methylpentme, 2-methylpentane, 2,3-dimethylbutane, and 2,2-dimethylbutane binary mixtures are 2.44%, 1.718%, 2.008%, 2.523% and 1.591%, respectively. These low values of the calculated mean absolute error for these cases approves the high accuracy of the presented equation for predicting the existed relation between z_L and z_V, for these binary mixtures.

4. Conclusion

In this study, a VLE equation of state, with a simple structural format, was presented for single or double-component systems. This equation related the compressibility factors of these phases to each other. The simple format of this equation was suitable for calculating unknown values of PVT parameters of each one of these phases in VLE systems. This equation decreases the volume of calculations in the bubble or dew point calculations in non-ideal single or double-component VLE systems. The predictions of this equation were validated and approved using the experimental data for some cases of pure substances or binary mixtures. One of the valuable applications of the developed equation in this study was improving the γ-φ method for calculating PVTxy parameters for vapor-liquid equilibrium systems at high pressures.

Conflict of interest

The authors declare that there is no conflict of interest regarding the publication of this article.

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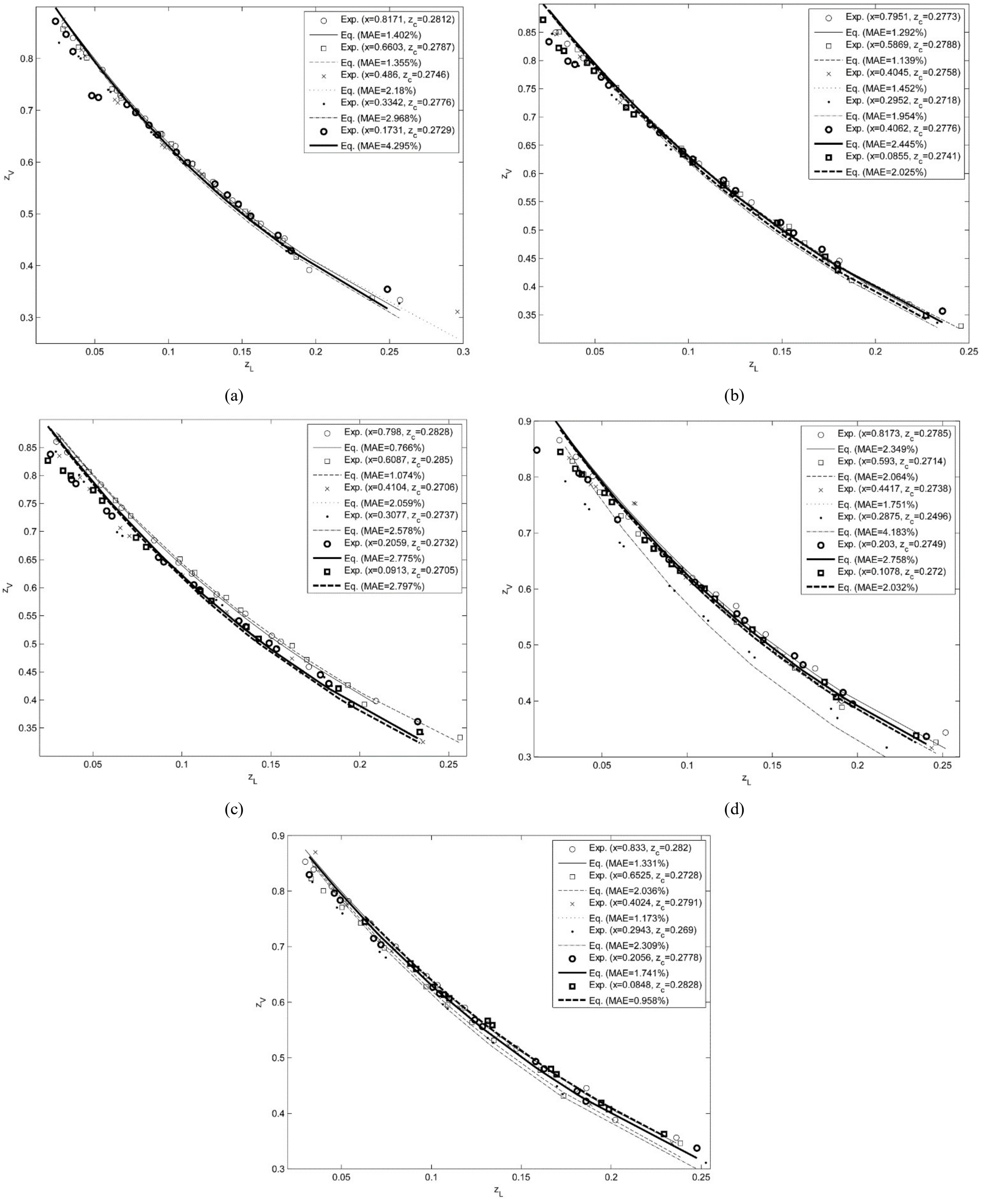

(a)

(b)

(c)

(d)

(e)

Fig. 7. Validation of the presented VLE equation using the experimental data of the saturated liquid and vapor phases of perfluoromethylcyclohexane (x) – (a) n-hexane, (b) 3-methylpentme, (c) 2-methylpentane, (d) 2,3-dimethylbutane, and (e) 2-2-dimethylbutane mixtures [38-39]

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