Int J Thermophys (2008) 29:423–433
DOI 10.1007/s10765-007-0360-2

Simplified Gradient Theory Modeling of the Surface Tension for Binary Mixtures

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Abstract In this work, the gradient theory was combined with the volume translation Peng-Robinson and Soave Redlich-Kwong equations of state (VTPR and VTSRK EOSs) and the influence parameter correlation to predict the surface tension of binary mixtures. The density profiles of mixtures across the interface were assumed to be linearly distributed to simplify the gradient theory model. The only two inputs of the theory are the Helmholtz free-energy density of the homogeneous fluid and the influence parameter of the inhomogeneous fluid. The VTPR and VTSRK equations of state were applied to determine the Helmholtz free-energy density and the bulk properties. The influence parameter of the inhomogeneous fluid was calculated from a correlation published previously (Lin et al. Fluid Phase Equilib 254:75, 2007). The only adjustable coefficient of the simplified gradient theory was set equal to zero, which made the theory predictive. The surface tension predicted by this model shows good agreement with experimental data for binary non-polar and polar mixtures.

Keywords Equation of state · Halogenated hydrocarbon · Mixtures · Simplified gradient theory · Surface tension

1 Introduction

Surface tension is one of the basic thermophysical properties and plays an important role in many industrial processes. The liquid–vapor surface tension influences and

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controls many processes in chemical or reservoir engineering applications, especially for analyzing the heat transfer through heat-exchanging surfaces with bubbles or fluid drops.

The parachor method [1–5], the corresponding states principle [6–8], perturbation theory [9,10], density functional theory [11–15], and gradient theory can be used to calculate the surface tension of heterogeneous fluids. Among these theories, the gradient theory is currently a very popular method for predicting surface tension and has satisfactory precision for pure fluids and mixtures. The density gradient and the heterogeneity in the surface region are taken into account in this theory. The only two inputs of the gradient theory are the Helmholtz free-energy density of the homogeneous fluid and the influence parameter of the inhomogeneous fluid. The Helmholtz free-energy density can be calculated by many thermodynamic models such as, for example, cubic equations of state (EOSs). The influence parameter can be computed from the molecular theoretical definition or a semi-empirical expression, although the influence parameter calculated from the molecular theoretical definition is too complex and has poor performance in predicting the surface tension [16].

Since its origination near the end of the 19th century, gradient theory was quickly developed. Cahn and Hilliard [17] rediscovered this theory. Carey et al. [18–22] were the first to combine gradient theory with cubic EOSs to predict the surface tension of pure fluids and mixtures. In this work, the density profiles of binary mixtures across the interface were assumed to be linearly distributed to simplify the gradient theory in predicting the surface tension of binary mixtures. The only adjustable coefficient of the influence parameter of the mixtures was set equal to zero, which makes the whole theory predictive. The Helmholtz free-energy density of the homogeneous fluid was calculated by the volume translation Peng-Robinson (VTPR) [23] and volume translation Soave Redlich-Kwong VTSRK [24] EOSs. The surface tension of non-polar and polar binary mixtures predicted by the simplified gradient theory showed good agreement with the experimental data.

2 Gradient Theory

Considering a planar interface between the two bulk phases of a pure fluid whose z-axis is normal to the interface, the number density $\rho_i(\mathrm{z})$ of component i at position z in an N-component, inhomogeneous fluid between two bulk phases at equilibrium is determined according to gradient theory:

$$\sum_j \frac{\mathrm{d}}{\mathrm{d}z}\left(\kappa_{ij}\frac{\mathrm{d}\rho_j}{\mathrm{d}z}\right) - \frac{1}{2}\sum_k\sum_j \frac{\partial\kappa_{kj}}{\partial\rho_i}\frac{\mathrm{d}\rho_k}{\mathrm{d}z}\frac{\mathrm{d}\rho_j}{\mathrm{d}z} = \frac{\partial\varpi}{\partial\rho_i} \tag{1}$$

where $\varpi(\rho) = f_0(\rho) - \sum_i \rho_i\mu_{i\mathrm{s}}$, $\mu_{i\mathrm{s}}$, is the chemical potential of component i of the bulk phases, $f_0(\rho)$ is the Helmholtz free-energy density of the homogeneous fluid, and κ is the influence parameter.

Assuming that the density dependence of the influence parameter can be neglected, Eq. 1 becomes

$$\sum_j \kappa_{ij} \frac{\mathrm{d}^2 \rho_j}{\mathrm{d}z^2} = \mu_i(\rho_1, \rho_2, \ldots, \rho_2) - \mu_{is} \tag{2}$$

where $\mu_i(\rho_1, \rho_2, \ldots, \rho_2)$ is the chemical potential of the interface. The boundary conditions for a planar interface are $\rho(z \to +\infty) = \rho_\mathrm{L}$, $\rho(z \to -\infty) = \rho_\mathrm{V}$, where ρ_L and ρ_V are the saturation liquid and vapor number densities of the bulk phases.

Multiplying $\mathrm{d}\rho_i/\mathrm{d}z$ on both sides of Eq. 2, summing over i, and integrating, the result can be rewritten as

$$\sum_i \sum_j \frac{1}{2} \kappa_{ij} \frac{\mathrm{d}\rho_i}{\mathrm{d}z} \frac{\mathrm{d}\rho_j}{\mathrm{d}z} = \varpi(\rho) - \varpi_\mathrm{s} \tag{3}$$

where $\varpi_\mathrm{s} = -p_\mathrm{s}$ and p_s is the saturated pressure of the bulk phases.

The surface tension γ can be calculated by the following formula:

$$\gamma = \int_{-\infty}^{+\infty} \sum_i \sum_j \kappa_{ij} \frac{\mathrm{d}\rho_i}{\mathrm{d}z} \frac{\mathrm{d}\rho_j}{\mathrm{d}z} \mathrm{d}z = \int_{-\infty}^{+\infty} 2 \left[f_0(\rho) - \sum_i \rho_i \mu_{is} + p_\mathrm{s} \right] \mathrm{d}z \tag{4}$$

where κ_{ii} and κ_{jj} are the influence parameters of the pure i and j components, The cross-influence parameter κ_{ij} can be calculated by the following formula:

$$\kappa_{ij} = (1 - \eta)\sqrt{\kappa_{ii}\kappa_{jj}} \tag{5}$$

Here, η is the binary interaction parameter. In this work,

$$\eta = 0 \tag{6}$$

to make the gradient theory predictive.

The independent variable ρ is difficult to determine. The simplified gradient theory assumes that the number densities of each component of the mixture near the interface are linearly distributed relative to the distance from the interface [25,26]:

$$\frac{\mathrm{d}\rho_i(z)}{\mathrm{d}z} = \text{constant} \tag{7}$$

From Eq. 7, the density profile of either component of the binary mixture (component 1 or 2) is a monotonic function of the spatial variable z across the interface. Here, component 2 is taken as the reference and the surface tension γ can be computed via Eqs. 3 and 4:

$$\gamma = \int_{\rho_{2\mathrm{V}}}^{\rho_{2\mathrm{L}}} \sqrt{2\kappa \left[f_0(\rho) - \sum_i \mu_{is}\rho_i + p_\mathrm{s} \right]} \mathrm{d}\rho_2 \tag{8}$$

where $\rho_{2\mathrm{V}}$ and $\rho_{2\mathrm{L}}$ are the saturation vapor and liquid number densities of reference component 2, respectively.

From the definition of the Helmholtz free-energy density of the homogeneous fluid, $f_0(\rho)$ can be written as

$$f_0(\rho) = \rho\mu(\rho) - p(\rho) \tag{9}$$

Therefore, the surface tension can be rewritten from Eqs. 8 and 9 as [16,27]

$$\gamma = \int_{\rho_{2\mathrm{V}}}^{\rho_{2\mathrm{L}}} \sqrt{2\kappa \left\{ \sum_i \rho_i [\mu_i(\rho) - \mu_{i\mathrm{s}}] - [p(\rho) - p_\mathrm{s}] \right\} \mathrm{d}\rho_2} \tag{10}$$

where κ is the influence parameter of the binary mixture which can be defined from Eqs. 4–6 as

$$\kappa = \kappa_{11} \left(\frac{\mathrm{d}\rho_1}{\mathrm{d}\rho_2} \right)^2 + 2\sqrt{\kappa_{11}\kappa_{22}} \left(\frac{\mathrm{d}\rho_1}{\mathrm{d}\rho_2} \right) + \kappa_{22} \tag{11}$$

In this work, the VTPR [23] and VTSRK [24] EOSs were used to calculate the thermodynamic properties of the bulk phases and the Helmholtz free-energy density because of their good performance in predicting the vapor pressure, the liquid and vapor densities, and the vapor–liquid equilibrium. The van der Waals mixing rule was used when calculating the vapor–liquid equilibrium properties:

$$a_\mathrm{m} = \sum_{i=1}^{2} \sum_{j=1}^{2} x_i x_j a_{ij} \tag{12}$$

$$a_{ij} = (1 - k_{ij}) a_{ii}^{1/2} a_{jj}^{1/2} \tag{13}$$

$$b_\mathrm{m} = \sum_{i=1}^{2} x_i b_i \tag{14}$$

$$c_\mathrm{m} = \sum_{i=1}^{2} x_i c_i \tag{15}$$

where x_i is the mole fraction of component i. a_{ii} is the cohesive energy, and b_i is the volumetric parameter of component i for EOSs. k_{ij} is the binary interaction coefficient which is used to represent the vapor–liquid equilibrium.

The influence parameter of pure fluids can be calculated from the correlation [27]:

$$\ln\left(\frac{\kappa}{ab^{2/3}}N_{\mathrm{A}}^{8/3}\right)=\kappa_0+\kappa_1\ln t+\kappa_2(\ln t)^2 \tag{16}$$

where κ_0, κ_1, and κ_2 are the fitting coefficients, $t=1-T/T_{\mathrm{c}}$ is the reduced temperature, a and b are the parameters of the EOS, and $N_{\mathrm{A}}=6.0221415\times10^{23}\,\mathrm{mol}^{-1}$ is the Avogadro constant.

A generalized correlation of the influence parameters for nonpolar and polar fluids (including inorganic fluids, alkanes, halogenated hydrocarbons, olefins, cyclic olefins, alkines, aromatics, etc.) was developed by correlating κ_0, κ_1, and κ_2 as functions of the critical compressibility factor, Z_{c}, the acentric factor, ω, and the reduced dipole moment, ϑ_{r} [27]:

VTPR EOS

$$\kappa_0=-2.985+4.332Z_{\mathrm{c}}+10.859Z_{\mathrm{c}}^2-1.990\omega+1.798\omega^2-5.436\times10^{-6}\,(\vartheta_{\mathrm{r}})^2 \tag{17}$$

$$\kappa_1=-0.965+1.405Z_{\mathrm{c}}+2.764Z_{\mathrm{c}}^2-0.963\omega+1.346\omega^2-1.110\times10^{-6}\,(\vartheta_{\mathrm{r}})^2 \tag{18}$$

$$\kappa_2=-0.121+0.156Z_{\mathrm{c}}+0.540Z_{\mathrm{c}}^2-0.123\omega+0.090\omega^2+0.219\times10^{-6}\,(\vartheta_{\mathrm{r}})^2 \tag{19}$$

VTSRK EOS

$$\kappa_0=-3.471+4.927Z_{\mathrm{c}}+13.085Z_{\mathrm{c}}^2-2.067\omega+1.891\omega^2-4.600\times10^{-6}\,(\vartheta_{\mathrm{r}})^2 \tag{20}$$

$$\kappa_1=-1.690+2.311Z_{\mathrm{c}}+5.644Z_{\mathrm{c}}^2-1.027\omega+1.424\omega^2-1.403\times10^{-6}\,(\vartheta_{\mathrm{r}})^2 \tag{21}$$

$$\kappa_2=-0.318+0.299Z_{\mathrm{c}}+1.710Z_{\mathrm{c}}^2-0.174\omega+0.157\omega^2+0.077\times10^{-6}\,(\vartheta_{\mathrm{r}})^2 \tag{22}$$

where $\vartheta_{\mathrm{r}}=\vartheta^2p_{\mathrm{c}}/(1.01325T_{\mathrm{c}}^2)$ with ϑ as the dipole moment in Debye ($1\,\mathrm{D}=3.33564\times10^{-30}\,\mathrm{C\cdot m}$).

3 Results and Discussion

3.1 Density Profile

The density profiles of mixtures across the interface were assumed to be linearly distributed and the derivatives of number density in Eq. 11 can be obtained from Eq. 7 as follows:

$$\frac{\mathrm{d}\rho_1}{\mathrm{d}\rho_2} = \frac{\rho_{1\mathrm{L}} - \rho_{1\mathrm{V}}}{\rho_{2\mathrm{L}} - \rho_{2\mathrm{V}}} \tag{23}$$

where $\rho_{1\mathrm{V}}$, $\rho_{1\mathrm{L}}$, $\rho_{2\mathrm{V}}$, and $\rho_{2\mathrm{L}}$ can be calculated from the following equations:

$$\rho_{1\mathrm{L}} = x_1 \rho_\mathrm{L}, \quad \rho_{1\mathrm{V}} = y_1 \rho_\mathrm{V} \tag{24}$$

$$\rho_{2\mathrm{L}} = x_2 \rho_\mathrm{L}, \quad \rho_{2\mathrm{V}} = y_2 \rho_\mathrm{V} \tag{25}$$

and x and y are the mole fractions of the liquid and vapor bulk phases, respectively. ρ_L and ρ_V can be determined by the EOSs from the vapor–liquid equilibrium calculations.

3.2 Surface Tension of Binary Polar Mixtures

The surface tension of binary mixtures then can be calculated from Eqs. 10 and 11 and Eqs. 23–25 with the VTPR and VTSRK EOSs while the influence parameter correlation is from Eqs. 16–22. Table 1 lists the average absolute deviations $\bar{\delta}_\mathrm{a}$ between the experimental data of some polar mixtures and the values calculated by the simplified gradient theory with the VTPR and VTSRK EOSs, and the table also includes the results calculated using the parachor method [5] and the corresponding-states method [6] for comparison. The binary interaction coefficient k_{ij}, temperature range, number of experimental data points N, and references for experimental surface tension data are also listed in Table 1. The binary interaction coefficient k_{ij} was determined from the vapor–liquid equilibrium experimental data and the EOSs. The results in Table 1 show that the simplified gradient theory combined with the VTPR and VTSRK EOSs can be successfully applied to calculate the surface tension of halogenated hydrocarbon mixtures. The average absolute deviation $\bar{\delta}_\mathrm{a}$ can be defined as

$$\bar{\delta}_\mathrm{a} = \frac{1}{N} \sum_{i=1}^{N} \left| \gamma_{\mathrm{exp},i} - \gamma_{\mathrm{cal},i} \right| \tag{26}$$

where γ_exp and γ_cal, respectively, are the experimental and calculated values and N is the number of data points.

The average absolute deviations are $0.23\,\mathrm{mN \cdot m^{-1}}$ and $0.26\,\mathrm{mN \cdot m^{-1}}$ calculated by the gradient theory with the VTPR and VTSRK EOSs, respectively. Our model gives better results than the other methods for polar fluid mixtures, especially for the

Table 1 Average absolute deviations for the surface tension between the experimental data and the values calculated by different models

Mixtures	$k_{ij,\text{VTPR}}$	$k_{ij,\text{VTSRK}}$	T (K)	N	$\bar{\delta}_{\text{a}}$ (mN · m^{-1})				Ref.
					VTPR	VTSRK	Parachor [5]	Corr. States [6]	
R32/134a	−0.0016	−0.0068	223.15–333.53	173	0.08	0.11	4.42	0.15	[28,29]
R32/125	0.0023	−0.0041	223.15–333.41	158	0.11	0.09	2.86	0.45	[28,30,31]
R143a/134a	−0.0016	−0.0089	223.15–333.15	123	0.14	0.21	2.29	0.35	[28,32]
R143a/227ea	0	0	252.56–333.42	121	0.12	0.14	0.78	0.22	[33]
R32/227ea	0.0143	0.0116	252.66–333.55	206	0.19	0.33	2.15	0.61	[34]
R125/134a	0.0014	−0.0058	223.15–333.15	21	0.42	0.49	0.86	0.34	[28]
R125/143a	0.0018	−0.0060	223.15–333.15	25	0.12	0.07	1.59	0.05	[28,30]
R134a/152a	−0.0004	−0.0069	223.15–333.15	21	0.38	0.42	3.59	0.26	[28]
R125/152a	−0.0118	−0.0221	223.15–333.15	21	0.63	0.68	2.54	0.33	[28]
R22/115	0.0485	0.0485	273.19–343.16	16	0.14	0.07	0.22	1.09	[35]
Average					0.23	0.26	2.13	0.38	

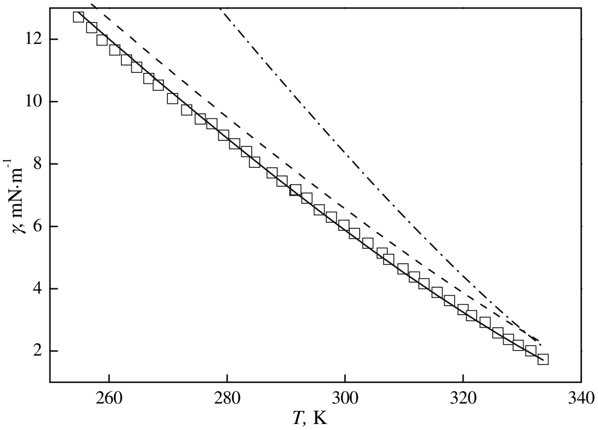

Fig. 1 Surface tension for R32/227ea versus temperature. (□) Experimental data (mass fraction 40.64%/59.36%) [34]; (——) simplified gradient theory prediction values combined with VTPR EOS; (— · — · —) values calculated by the parachor model [5]; (– – – –) values calculated by the corresponding-states model [6]

strongly polar R22/115 mixture. But for the R134a/152a and R125/134a mixtures, the deviations are larger than for the other mixtures and the experimental data are all less than the calculated values. This may be partly due to a systematic error in the experimental measurements [16]. The k_{ij} for R143a/227ea was set equal to zero because there were no vapor–liquid equilibrium data available. Figure 1 shows the experimental surface tension data of R32/227ea and the values calculated by the three models. The results in Fig. 1 show that the simplified gradient theory combined with a volume translation EOS can represent the experiment data well. Figures 2 and 3 show the relationship between the surface tension and the temperature for R32/125 and R143a/134a, also including the values calculated by the present model, and the parachor and corresponding-states results.

3.3 Other Systems

The gradient theory and the simplified model can also be applied to other binary mixtures. Table 2 lists the average absolute deviations between the experimental surface tension data and the values calculated by the different models. The results in Table 2 show that the surface tensions calculated by the simplified model compare very well with the experimental data. The interaction coefficient k_{ij} of CH_4/Kr was set equal to zero because there was no vapor–liquid equilibrium data available.

3.4 Discussion

The gradient theory combined with the volume translation EOSs can represent the surface tension of binary mixtures well when the linear density profile was assumed. There are no adjustable parameters whereas other simplified gradient-theory models

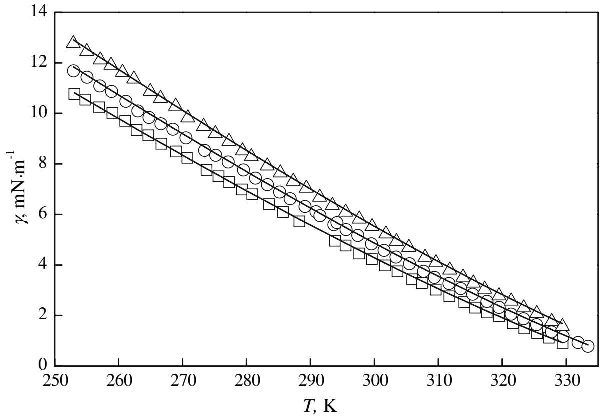

Fig. 2 Surface tension for R32/125 versus temperature. (□) mass fraction 24.95%/75.05% [31]; (○) mass fraction 44.83%/55.17% [31]; (△) mass fraction 66.75%/33.25% [31]; (——) simplified gradient theory prediction values combined with VTPR EOS

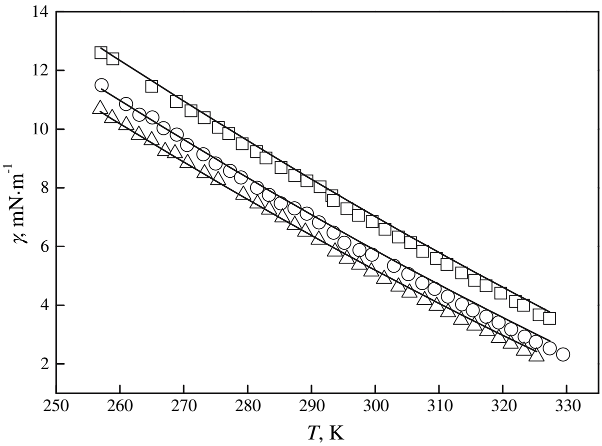

Fig. 3 Surface tension for R143a/134a versus temperature. (□) mass fraction 26.88%/73.12% [32]; (○) mass fraction 57.14%/42.86% [32]; (△) mass fraction 75.76%/24.24% [32]; (——) simplified gradient theory prediction values combined with VTSRK EOS

[25,26] all have one or more adjustable parameters fitted from the experimental surface tension data. In this work, the reasons that the model made the simplified gradient theory predictive are as follows. Firstly, the VTPR and VTSRK EOSs can represent the saturated liquid and vapor densities very well, and these densities have a direct relationship with the surface tension calculation. Secondly, the binary interaction coefficient k_{ij} was fitted from the experimental vapor–liquid equilibrium data and the EOSs to closely represent the bulk thermophysical properties. Thirdly, the influence parameters of pure fluids were determined from experimental surface tension data of pure fluids and the gradient theory which was generalized as a function of the thermophysical

Table 2 Average absolute deviations for the surface tension between the experimental data and calculated values by different models

Mixtures	$k_{ij,\mathrm{VTPR}}$	$k_{ij,\mathrm{VTSRK}}$	T (K)	N	$\bar{\delta}_\mathrm{a}$ $(\mathrm{mN \cdot m^{-1}})$				Ref.
					VTPR	VTSRK	Parachor [5]	Corr. States [6]	
CH_4/Kr	0	0	125.18–188.84	85	0.32	0.40	0.34	0.27	[36,37]
Ar/N_2	−0.0098	−0.0069	83.82–85.4	40	0.40	0.35	0.20	0.28	[38,39]
N_2/CO	0.0172	0.0185	83.82	10	0.12	0.28	0.41	0.14	[38]
Ar/O_2	0.0147	0.0279	64.3–88	66	0.26	0.17	0.53	0.14	[40,41]
R10/Toluene	0	0	293.15–443.25	116	0.23	0.34	1.23	0.32	[42–45]
Average					0.27	0.31	0.54	0.23	

properties. The gradient theory with this correlation can reliably predict the surface tension of pure non-polar and polar fluids [27].

4 Conclusions

The simplified gradient theory was combined with the VTPR and VTSRK EOSs to represent the surface tension of binary mixtures. The density profiles of mixtures were assumed to distribute linearly across the interface. The only adjustable coefficient in the cross-influence parameter was set equal to zero to make the theory predictive. The calculated values compared well with the experimental data.

Acknowledgments This work was supported by the National Natural Science Foundation of China (No. 50636020) and the Doctoral Special Start-up Fund (No. TGQD05-015) of the National Institute of Metrology (NIM) in China.

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