



## Corrigendum to: “Entropy Scaling-Based Correlation for Estimating the Self-Diffusion Coefficients of Pure Fluids”

Aghilas Dehlouz, Jean-Noël Jaubert, Guillaume Galliero, Marc Bonnissel,  
Romain Privat

### ► To cite this version:

Aghilas Dehlouz, Jean-Noël Jaubert, Guillaume Galliero, Marc Bonnissel, Romain Privat. Corrigendum to: “Entropy Scaling-Based Correlation for Estimating the Self-Diffusion Coefficients of Pure Fluids”. Industrial and engineering chemistry research, 2024, 63 (34), pp.15314-15319. 10.1021/acs.iecr.4c02704 . hal-04683076

HAL Id: hal-04683076

<https://hal.univ-lorraine.fr/hal-04683076v1>

Submitted on 1 Sep 2024

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial - NoDerivatives 4.0 International License

## Corrigendum to: “Entropy Scaling-Based Correlation for Estimating the Self-Diffusion Coefficients of Pure Fluids”

Aghilas Dehlouz, Jean-Noël Jaubert,\* Guillaume Galliero, Marc Bonnissel, and Romain Privat\*

*Ind. Eng. Chem. Res.* 2022, 61, 37, 14033–14050. DOI: 10.1021/acs.iecr.2c01086



Cite This: *Ind. Eng. Chem. Res.* 2024, 63, 15314–15319



Read Online

ACCESS |

Metrics & More

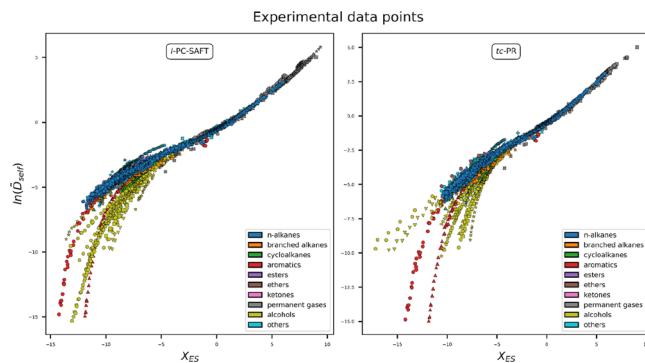
Article Recommendations

Supporting Information

The authors regret to report that they have identified two errors in the Fortran code that they wrote and was used to generate the results they published in 2022.<sup>1</sup>

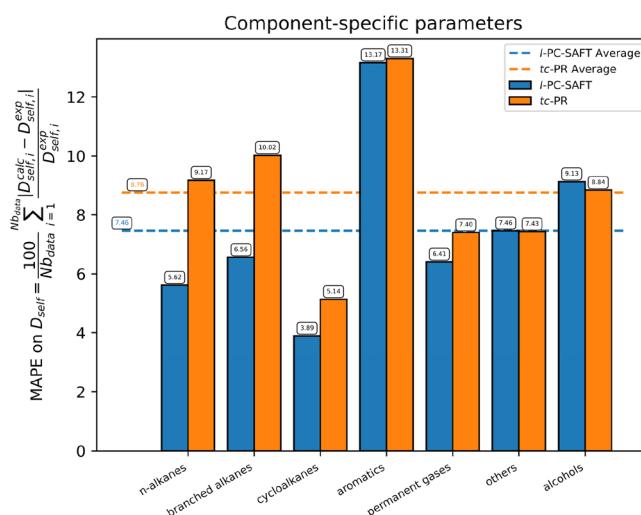
**Table 2. Universal Parameters for the Self-Diffusion Model in the Dilute Gas Region ( $X_{ES} \geq 0$ ) Associated with Two Different EoS (Used for Estimating Density and Residual Entropy), Namely, the *I*-PC-SAFT and *tc*-PR Models**

EoS	<i>I</i> -PC-SAFT	<i>tc</i> -PR
$b_{univ}$	0.6686	0.6621
$c_{univ}$	0.2861	0.3351
$d_{univ}$	-0.5451	-0.5969



**Figure 4.** Overview of all experimental data in the  $\ln(\tilde{D}_{self})$  vs  $X_{ES}$  plane. The thermodynamic properties involved in the definition of  $\tilde{D}_{self}$  and  $X_{ES}$  were estimated using the *I*-PC-SAFT EoS (left panel) and the *tc*-PR EoS (right panel). Each color corresponds to a specific chemical family, and each symbol corresponds to a specific compound.

- The first error was a mistake in reading the experimental data file, which led to only one experimental data point out of 2 being considered. Consequently, the deviations reported in 2022, between experimental and calculated self-diffusion coefficients only considered half of the available data points. **Table S1 of the Supporting Information**, attached to this corrigendum, provides detailed information on the data considered in this study.
- The second error was in the equation used to calculate the  $T_v$ -residual entropy from the *translated-consistent Peng–Robinson* (*tc*-PR)<sup>2</sup> cubic equation of state (EoS).



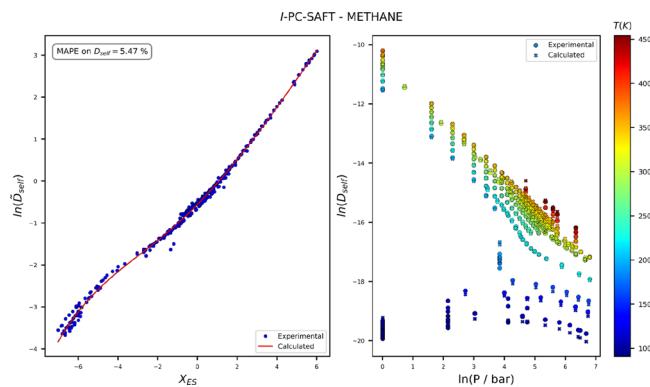
**Figure 5.** Correlation of self-diffusion coefficients using the model given by eq C4 and parametrization strategy 1 (component-specific parameters). Summary of the MAPEs obtained using either the *I*-PC-SAFT or the *tc*-PR EoS for estimating density and residual entropy.

The authors sincerely apologize for the inconvenience caused and, in order to clarify the second error, have decided to recall below the correct equation to be used to calculate the  $T_v$ -residual entropy from the *tc*-PR EoS.

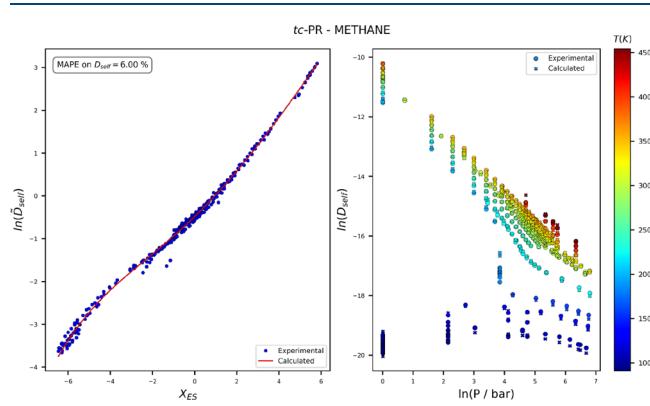
For a pure component, the *tc*-PR equation writes:

Published: August 16, 2024

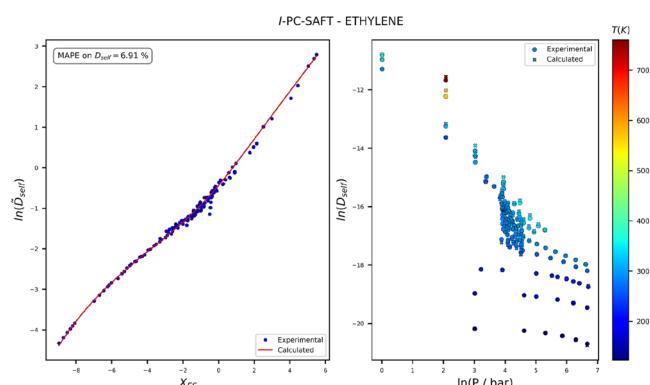




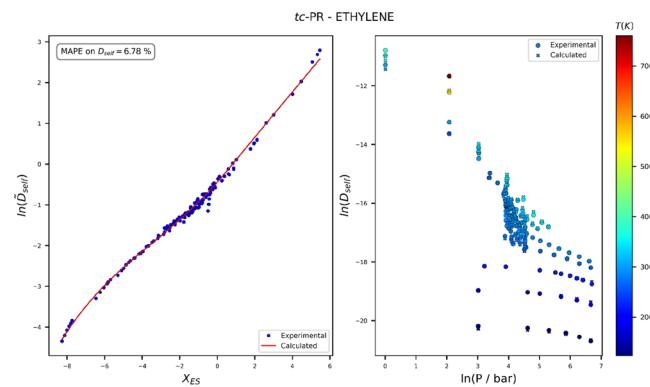
**Figure 6.** Application of the entropy scaling (ES)-based model combining eq C4 and parametrization strategy 1 to methane using the I-PC-SAFT EoS to estimate density and residual entropy. Left panel: ( $X_{\text{ES}}$ ,  $Y_{\text{ES}}$ ) plane. Right panel: self-diffusion coefficient vs pressure (log-log scale).



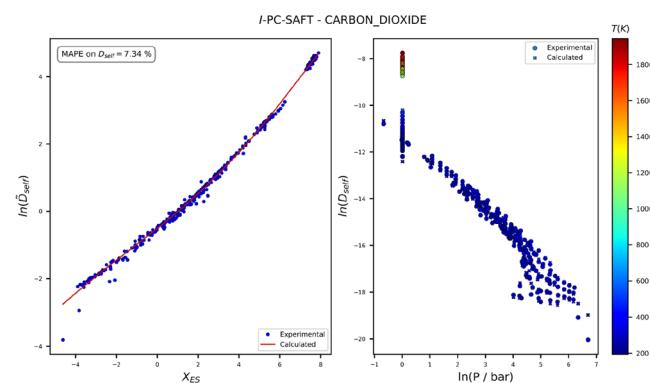
**Figure 7.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to methane using the tc-PR EoS (for more details, refer to the caption of Figure 6).



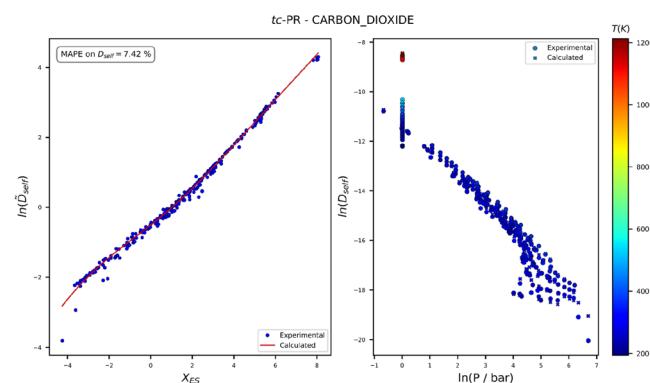
**Figure 8.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to ethylene using the I-PC-SAFT EoS (for more details, refer to the caption of Figure 6).



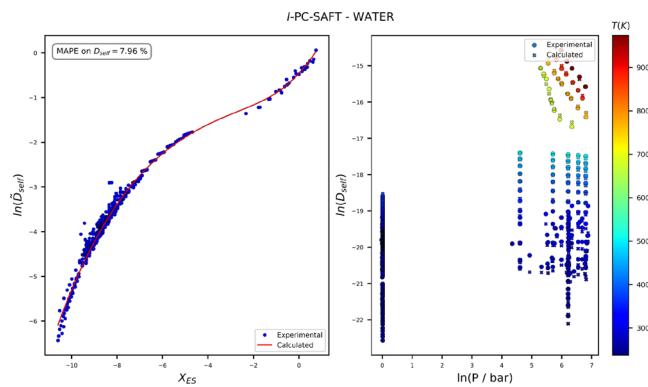
**Figure 9.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to ethylene using the tc-PR EoS (for more details, refer to the caption of Figure 6).



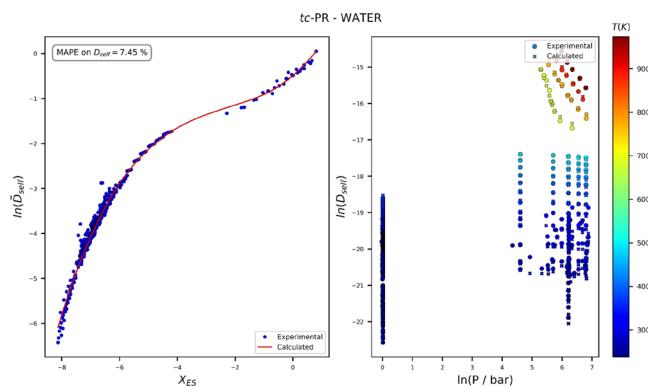
**Figure 10.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to  $\text{CO}_2$  using the I-PC-SAFT EoS (for more details, refer to the caption of Figure 6).



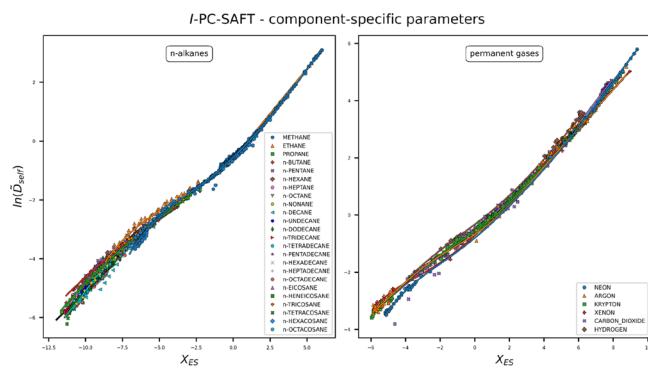
**Figure 11.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to  $\text{CO}_2$  using the tc-PR EoS (for more details, refer to the caption of Figure 6).



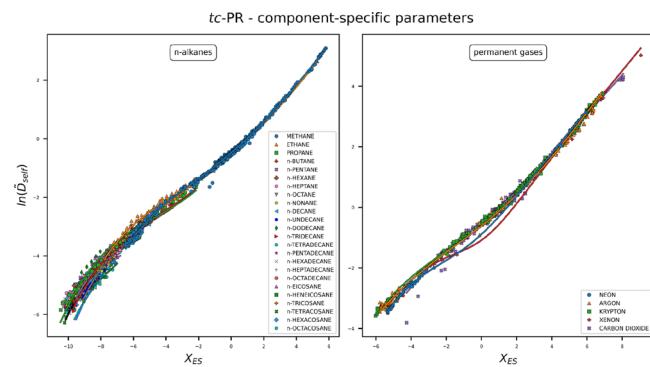
**Figure 12.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to water using the *I*-PC-SAFT EoS (for more details, refer to the caption of Figure 6).



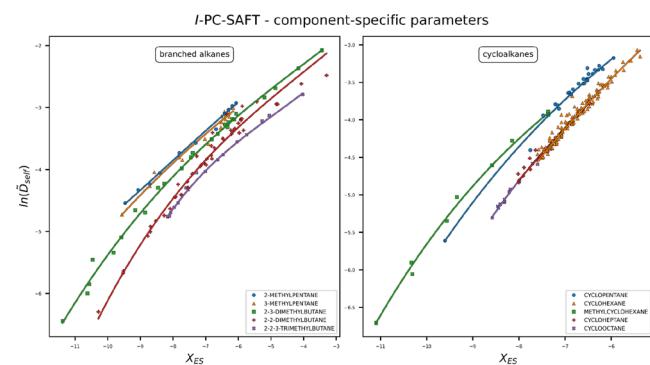
**Figure 13.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to water using the *tc*-PR EoS (for more details, refer to the caption of Figure 6).



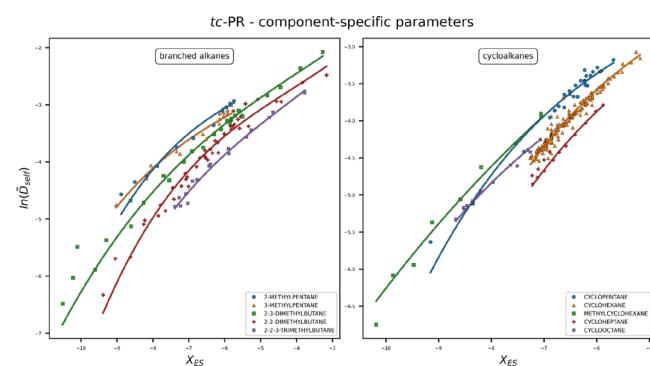
**Figure 14.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to *n*-alkanes and permanent gases using the *I*-PC-SAFT EoS. Points represent experimental data; solid line represents model data.



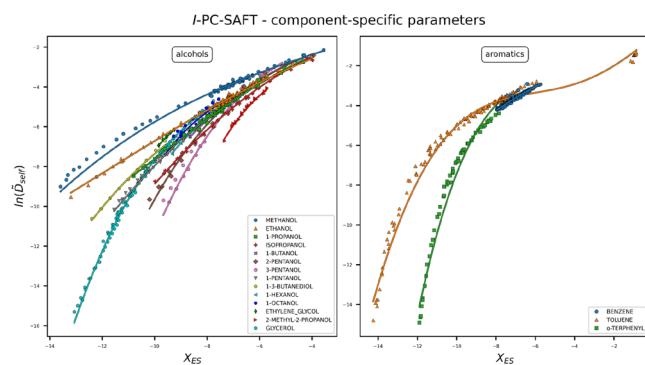
**Figure 15.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to *n*-alkanes and permanent gases using the *tc*-PR EoS. Points represent experimental data; solid line represents model data.



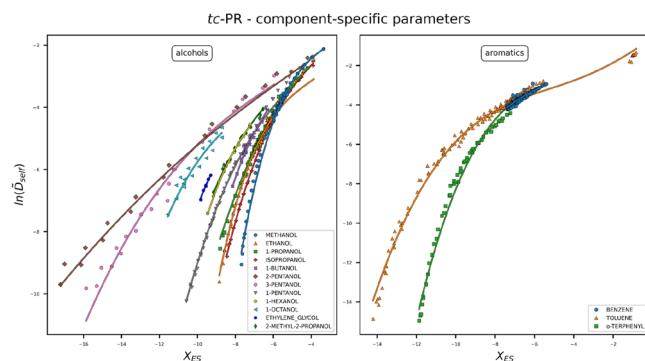
**Figure 16.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to branched alkanes and cycloalkanes using the *I*-PC-SAFT EoS. Points represent experimental data; solid line represents model data.



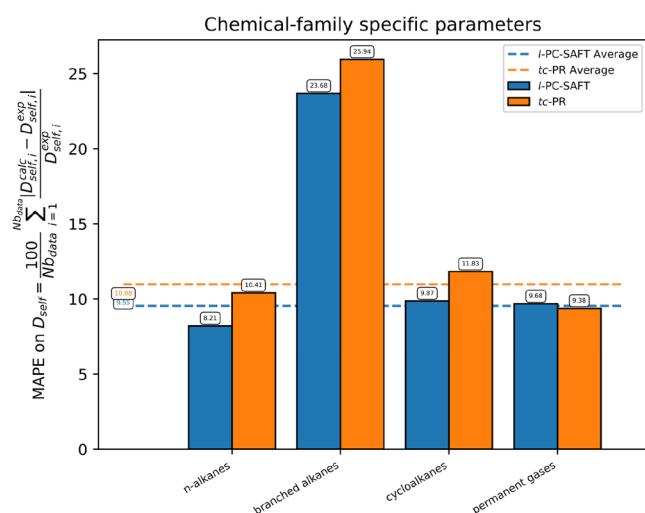
**Figure 17.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to branched alkanes and cycloalkanes using the *tc*-PR EoS. Points represent experimental data; solid line represents model data.



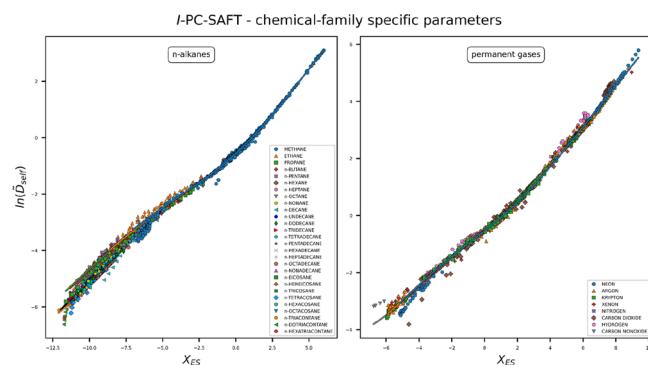
**Figure 18.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to *n*-alcohols and aromatic compounds using the *I*-PC-SAFT EoS. Points represent experimental data; solid line represents model data.



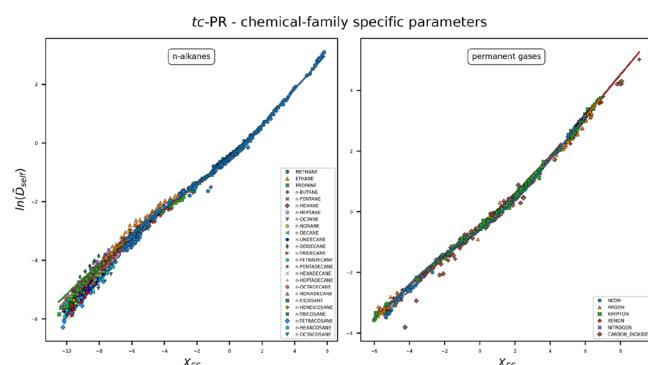
**Figure 19.** Application of the ES-based model combining eq C4 and parametrization strategy 1 to *n*-alcohols and aromatic compounds using the *tc*-PR EoS. Points represent experimental data; solid line represents model data.



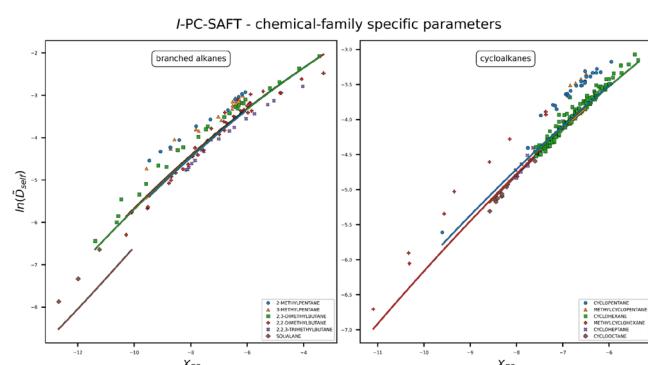
**Figure 20.** Prediction of self-diffusion coefficients using the model given by eq C4 and parametrization strategy 2 (with chemical-family specific parameters). Summary of the MAPEs obtained using either the *I*-PC-SAFT or the *tc*-PR EoS for estimating density and residual entropy.



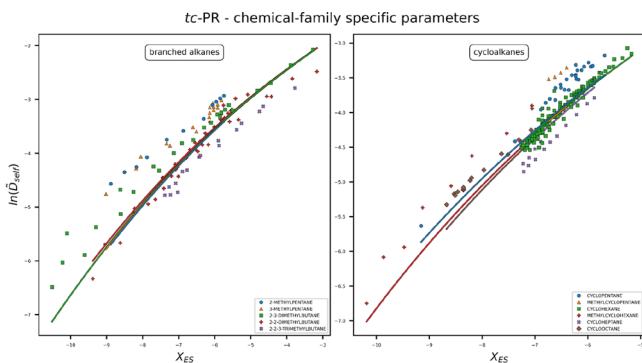
**Figure 21.** Application of the ES-based model combining eq C4 and parametrization strategy 2 to *n*-alkanes and permanent gases using the *I*-PC-SAFT EoS. Points represent experimental data; solid line represents model data.



**Figure 22.** Application of the ES-based model combining eq C4 and parametrization strategy 2 to *n*-alkanes and permanent gases using the *tc*-PR EoS. Points represent experimental data; solid line represents model data.



**Figure 23.** Application of the ES-based model combining eq C4 and parametrization strategy 2 to aromatic compounds and ketones using the *I*-PC-SAFT EoS. Points represent experimental data; solid line represents model data.



**Figure 24.** Application of the ES-based model combining eq C4 and parametrization strategy 2 to aromatic compounds and ketones using the *tc*-PR EoS. Points represent experimental data; solid line represent model data.

$$P(T, v) = \frac{RT}{v + c_i - b_i} - \frac{a_i(T)}{(v + c_i)(v + c_i + b_i) + b_i(v + c_i - b_i)}$$

$a_i(T) = a_{c,i} \cdot \alpha_i(T_r); \quad T_r = \frac{T}{T_{c,i,\text{exp}}}$   
 $\alpha_i(T_r) = T_r^{N_i(M_i-1)} \exp[L_i(1 - T_r^{M_i N_i})]$   
 $c_i = v_{i,\text{liq}}^{\text{sat,u-PR}}(T_r = 0.8) - v_{i,\text{liq,exp}}^{\text{sat}}(T_r = 0.8)$

with:

$$\eta_c = [1 + \sqrt[3]{4 - 2\sqrt{2}} + \sqrt[3]{4 + 2\sqrt{2}}]^{-1} \approx 0.25308$$

$$a_{c,i} = \frac{40\eta_c + 8}{49 - 37\eta_c} \frac{R^2 T_{c,i,\text{exp}}^2}{P_{c,i,\text{exp}}} \approx 0.45724 \frac{R^2 T_{c,i,\text{exp}}^2}{P_{c,i,\text{exp}}}$$

$$b_i = \frac{\eta_c}{\eta_c + 3} \frac{R T_{c,i,\text{exp}}}{P_{c,i,\text{exp}}} \approx 0.07780 \frac{R T_{c,i,\text{exp}}}{P_{c,i,\text{exp}}}$$
(C1)

Such a model is said to be *translated*<sup>3,4</sup> because a substance-dependent volume-translation parameter,  $c_b$ , is introduced, in order that the translated EoS exactly reproduces the experimental saturated liquid volume at a reduced temperature of 0.8, [ $v_{i,\text{liq,exp}}^{\text{sat}}(T_r = 0.8)$ ]:

$$c_i = v_{i,\text{liq}}^{\text{sat},\text{u-CEoS}}(T_r = 0.8) - v_{i,\text{liq,exp}}^{\text{sat}}(T_r = 0.8) \quad (\text{C2})$$

where  $v_{\text{iliq}}^{\text{sat-u-CEoS}}(T_r = 0.8)$  is the molar volume calculated with the original (untranslated) EoS at  $T_r = 0.8$ . In turn, the term “consistent” stands for the fact that the *tc*-PR EoS relies on an  $\alpha$ -function that passes the consistency test proposed by Le Guennec et al.<sup>5,6</sup>

Starting from eq C1, the mathematical expression of the T<sub>v</sub>-residual entropy can be straightforwardly determined by

$$\begin{aligned}
 s_{\text{TV-res}} &= \left[ \frac{\partial}{\partial T} \int_{\infty}^v \left( P(T, v) - \frac{RT}{v} \right) dv \right]_v \\
 &= R \ln \left( \frac{v + c_i - b_i}{v} \right) - \frac{1}{b_i(r_1 - r_2)} \ln \left( \frac{v + c_i - b_i r_1}{v + c_i - b_i r_2} \right) \cdot \underbrace{a_{c_i}}_{\text{omitted}} \cdot \frac{\partial a_i}{\partial T} \\
 &\quad \text{in 2022}
 \end{aligned}$$

Unfortunately, in our paper from 2022, the  $a_{c,i}$  quantity that appears in eq C3 was involuntarily omitted in our software, leading to erroneous values of  $s_{\text{Ty-res}}$ .

Despite these regrettable errors, the general conclusions and observations reported in our original article remain valid. Regardless of the used parametrization strategy (use of component-specific parameters or use of chemical-family specific parameters), the overall mean deviations, calculated correctly and reported in this corrigendum, are slightly different from those published in 2022.

Clearly, the numerical values of the coefficients to be used in the developed correlations when the *I*-PC-SAFT<sup>7</sup> or the *tc*-PR EoS is considered must be updated. They are reported in the revised **Supporting Information** file associated with this correction.

**Corrigendum to the Supporting Information:** All fitted coefficients related to the *I*-PC-SAFT and *tc*-PR EoS-based model are now updated in the new version of the **Supporting Information** that was constructed with the correct values of  $s_{T\text{v-res}}$  (for the case of the *tc*-PR EoS) and considering the correct number of data points (for both EoS).

**Corrigendum to Table 2:** The universal parameters to be used to model the dilute gas region were kept unchanged for the *I*-PC-SAFT EoS, as the addition of many new experimental data points in the fitting procedure resulted in coefficients that were almost identical to those obtained in the original article. On the other hand, the parameters to be used with the *tc*-PR-based model had to be updated after correcting the  $T_v$ -residual entropy values returned by such an EoS.

**Corrigendum to the overall MAPEs obtained using the I-PC-SAFT EoS:** In the original article, the overall MAPEs were 7.45% and 9.31% when component-specific and chemical-family specific parameters were used, respectively. They are now 7.46% and 9.55%, respectively, after considering the complete experimental datasets available for each component.

**Corrigendum to the overall MAPEs obtained using the tc-PR EoS:** In the original article, the overall MAPEs were 7.86% and 9.41% when component-specific and chemical-family specific parameters were used, respectively. They are now 8.76% and 10.98%, respectively, after using the correct values of the  $T_v$ -residual entropy and the whole experimental data points available for each component.

**Corrigendum to Figures 4–24:** After taking into account all the experimental data and correcting the  $T_v$ -residual entropy values returned by the  $tc$ -PR EoS, the figures presenting the results obtained with the  $I$ -PC-SAFT and  $tc$ -PR EoS are updated as follows.

For clarity, and to be on line with the captions of the listed figures, the main equations that are used to correlate self-diffusion coefficients are recalled below in eq C4:

$$\left\{ \begin{array}{l} \ln \left( \frac{D_{\text{self}}}{D_{\text{reference}}}_{\text{calc}} \right) = \left[ \left( \frac{a_1 + a_2 \tilde{s}_{\text{TV-res}} + a_3 \tilde{s}_{\text{TV-res}}^2}{1 + e^{cX_{\text{ES}}}} \right) + \left( \frac{b}{1 + e^{-cX_{\text{ES}}}} \right) \right] X_{\text{ES}} + d \\ \tilde{s}_{\text{TV-res}} = \frac{s_{\text{TV-res}}}{R} \quad \text{and} \quad D_{\text{self}}^{\text{reference}} = \rho_N^{-1/3} \sqrt{\frac{k_B T}{m_0}} \\ X_{\text{ES}} = \left( -\frac{\tilde{s}_{\text{TV-res}}}{\tilde{s}_{\text{TV-res}}^c} \right) - \ln \left( \frac{\tilde{s}_{\text{TV-res}}}{\tilde{s}_{\text{TV-res}}^c} \right); \quad Y_{\text{ES}} = \tilde{D}_{\text{self}} \end{array} \right. \quad (\text{C4})$$

In addition, in the captions of the figures shown in this Addition/Correction, the term “parametrization strategy 1” means that component-specific parameters are used to correlate the data.

whereas the term “parametrization strategy 2” means that chemical-family specific parameters are used.

## ASSOCIATED CONTENT

### SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.iecr.4c02704>.

Presentation of the experimental database used for the ES-based correlation development; parameters to be used with the *I*-PC-SAFT-based model; parameters to be used with the *tc*-PR-based model ([PDF](#))

Deviations between calculated and experimental self-diffusion coefficients for the experimental data points used in this study ([XLSX](#))

## ACKNOWLEDGMENTS

The authors would like to warmly thank Jarrell Richard Elliott, Professor at the University of Akron, for pointing out the error reported in this corrigendum.

## REFERENCES

- (1) Dehlouz, A.; Jaubert, J.-N.; Galliero, G.; Bonnissel, M.; Privat, R. Entropy Scaling-Based Correlation for Estimating the Self-Diffusion Coefficients of Pure Fluids. *Ind. Eng. Chem. Res.* **2022**, *61* (37), 14033–14050.
- (2) Le Guennec, Y.; Privat, R.; Jaubert, J.-N. Development of the Translated-Consistent *tc*-PR and *tc*-RK Cubic Equations of State for a Safe and Accurate Prediction of Volumetric, Energetic and Saturation Properties of Pure Compounds in the Sub- and Super-Critical Domains. *Fluid Phase Equilib.* **2016**, *429*, 301–312.
- (3) Jaubert, J.-N.; Privat, R.; Le Guennec, Y.; Coniglio, L. Note on the Properties Altered by Application of a Pénéloix-Type Volume Translation to an Equation of State. *Fluid Phase Equilib.* **2016**, *419*, 88–95.
- (4) Privat, R.; Jaubert, J.-N.; Le Guennec, Y. Incorporation of a Volume Translation in an Equation of State for Fluid Mixtures: Which Combining Rule? Which Effect on Properties of Mixing? *Fluid Phase Equilib.* **2016**, *427*, 414–420.
- (5) Le Guennec, Y.; Lasala, S.; Privat, R.; Jaubert, J.-N. A Consistency Test for  $\alpha$ -Functions of Cubic Equations of State. *Fluid Phase Equilib.* **2016**, *427*, 513–538.
- (6) Le Guennec, Y.; Privat, R.; Lasala, S.; Jaubert, J.-N. On the Imperative Need to Use a Consistent  $\alpha$ -Function for the Prediction of Pure-Compound Supercritical Properties with a Cubic Equation of State. *Fluid Phase Equilib.* **2017**, *445*, 45–53.
- (7) Moine, E.; Pina-Martinez, A.; Jaubert, J.-N.; Sirjean, B.; Privat, R. *I*-PC-SAFT: An Industrialized Version of the Volume-Translated PC-SAFT Equation of State for Pure Components, Resulting from Experience Acquired All through the Years on the Parameterization of SAFT-Type and Cubic Models. *Ind. Eng. Chem. Res.* **2019**, *58* (45), 20815–20827.