Erratum: Flux-Based Modeling of Heat and Mass Transfer in Multicomponent Systems [Phys. Fluids 34, 033113 (2022)]

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In the Article "Flux-Based Modeling of Heat and Mass Transfer in Multicomponent Systems" by A.N. Beris, S. Jariwala and N.J. Wagner, that was published in Physics of Fluids, 34, 033113 (2022)[1], there was an erratum in the second integral of Eq. (14) due to an erroneous application of Eq. (A3) of the appendix A of that paper. The correct Eq. (14) should read as follows

$$
\begin{aligned}
[F,H]_a =& -\int_\Omega \sum_{i=1}^{n} \begin{pmatrix} \dfrac{\delta F}{\delta \rho_i} \nabla_\alpha \left(\left(\rho_i \dfrac{\delta H}{\delta \hat{u}_\alpha^{(i)}} \right) - \dfrac{\rho_i}{\rho} \left(\sum_{k=1}^{n} \rho_k \dfrac{\delta H}{\delta \hat{u}_\alpha^{(k)}} \right) \right) \\ - \dfrac{\delta H}{\delta \rho_i} \nabla_\alpha \left(\left(\rho_i \dfrac{\delta F}{\delta \hat{u}_\alpha^{(i)}} \right) - \dfrac{\rho_i}{\rho} \left(\sum_{k=1}^{n} \rho_k \dfrac{\delta F}{\delta \hat{u}_\alpha^{(k)}} \right) \right) \end{pmatrix} \mathrm{d}^3 x \\
& - \int_\Omega \sum_{i=1}^{n} \begin{pmatrix} \dfrac{\delta F}{\delta s} \nabla_\alpha \left(\left(s_i \dfrac{\delta H}{\delta \hat{u}_\alpha^{(i)}} \right) - \dfrac{s_i}{\rho} \left(\sum_{k=1}^{n} \rho_k \dfrac{\delta H}{\delta \hat{u}_\alpha^{(k)}} \right) \right) \\ - \dfrac{\delta H}{\delta s} \nabla_\alpha \left(\left(s_i \dfrac{\delta F}{\delta \hat{u}_\alpha^{(i)}} \right) - \dfrac{s_i}{\rho} \left(\sum_{k=1}^{n} \rho_k \dfrac{\delta F}{\delta \hat{u}_\alpha^{(k)}} \right) \right) \end{pmatrix} \mathrm{d}^3 x - \int_\Omega \begin{pmatrix} \dfrac{\delta F}{\delta s} \nabla_\alpha \left(s \dfrac{\delta H}{\delta \hat{u}_\alpha^{(0)}} \right) \\ - \dfrac{\delta H}{\delta s} \nabla_\alpha \left(s \dfrac{\delta F}{\delta \hat{u}_\alpha^{(0)}} \right) \end{pmatrix} \mathrm{d}^3 x
\end{aligned},
\tag{1}
$$

where s_i is the partial entropy density per unit of mass of component i. Note that this is connected to the most often used partial molar entropy, $S_i \equiv \partial S / \partial N_i \big|_{p,T,N_j,j\neq i}$, evaluated from $S \equiv \sum_{i=1}^{n} N_i S_i \Rightarrow s = S/V = \sum_{i=1}^{n} N_i/V \, S_i = \sum_{i=1}^{n} n_i S_i \equiv \sum_{i=1}^{n} s_i$ through $s_i \equiv S_i n_i = S_i \frac{1}{M_i} \rho_i$ where N_i, n_i, ρ_i are the number of moles, the mole number density and mass density of component *i*. Note that only when this error is corrected, the (correct) equation for the time evolution for the entropy density, Eq. (29) of paper[1], can be derived.

However, due to this erratum, the following additional term should appear in the right-hand-side of: Second equation in Eqs. (31), (32), (34), and (37) and in Eq. (35):

$$-\left(s_i - \frac{\rho_i}{\rho} s \right) \nabla_\alpha T \ . \tag{2}$$

This term first appears in Eq. (31) as a result of the application of the procedure that generates the governing equations following the bracket formalism indicated before, in the beginning of section II, around Eqs. (1) and (2) of the manuscript[1] (for the general case) and then repeated again around Eqs. (19) and (20) of the manuscript[1] for the particular flux transport application discussed in this work. More specifically, the new term emerges from a comparison of a suitably trasformed Eq. (1) through differentiation by parts, (considering that fully integrated terms leave no contributions as all fields are assumed in the model derivation to decay to zero at infinity) so that it only involves Volterra derivatives of the functional F outside any derivative operations (to allow for a direct comparison against Eq. (20) of the manuscript[1]) as

$$\begin{aligned}
[F,H]_a = & -\int_\Omega \sum_{i=1}^{n} \left(\begin{array}{l} \dfrac{\delta F}{\delta \rho_i} \nabla_\alpha \left(\left(\rho_i \dfrac{\delta H}{\delta \widehat{u}_\alpha^{(i)}} \right) - \dfrac{\rho_i}{\rho} \left(\displaystyle\sum_{k=1}^{n} \rho_k \dfrac{\delta H}{\delta \widehat{u}_\alpha^{(k)}} \right) \right) \\ + \left(\left(\rho_i \dfrac{\delta F}{\delta \widehat{u}_\alpha^{(i)}} \right) - \dfrac{\rho_i}{\rho} \left(\displaystyle\sum_{k=1}^{n} \rho_k \dfrac{\delta F}{\delta \widehat{u}_\alpha^{(k)}} \right) \right) \nabla_\alpha \dfrac{\delta H}{\delta \rho_i} \end{array} \right) \mathrm{d}^3 x \\
& -\int_\Omega \sum_{i=1}^{n} \left(\begin{array}{l} \dfrac{\delta F}{\delta s} \nabla_\alpha \left(\left(s_i \dfrac{\delta H}{\delta \widehat{u}_\alpha^{(i)}} \right) - \dfrac{s_i}{\rho} \left(\displaystyle\sum_{k=1}^{n} \rho_k \dfrac{\delta H}{\delta \widehat{u}_\alpha^{(k)}} \right) \right) \\ + \left(\left(s_i \dfrac{\delta F}{\delta \widehat{u}_\alpha^{(i)}} \right) - \dfrac{s_i}{\rho} \left(\displaystyle\sum_{k=1}^{n} \rho_k \dfrac{\delta F}{\delta \widehat{u}_\alpha^{(k)}} \right) \right) \nabla_\alpha \dfrac{\delta H}{\delta s} \end{array} \right) \mathrm{d}^3 x - \int_\Omega \left(\begin{array}{l} \dfrac{\delta F}{\delta s} \nabla_\alpha \left(s \dfrac{\delta H}{\delta \widehat{u}_\alpha^{(0)}} \right) \\ + \left(s \dfrac{\delta F}{\delta \widehat{u}_\alpha^{(0)}} \right) \nabla_\alpha \dfrac{\delta H}{\delta s} \end{array} \right) \mathrm{d}^3 x \ .
\end{aligned} \tag{3}$$

The extra term indicated in Eq. (2) is then obtained as the weight of $\delta F / \delta \widehat{u}_\alpha^{(i)}$ in Eq. (3), where an exchange of the dummy indices involved in the double summation, $k \leftrightarrow i$, has been performed.

Correction of the above error requires consideration of a more general expression for the gradient of the chemical potential $\underline{\nabla} \mu_j$ (obtained from a general Equilibrium Thermodynamics reference, such as refs.[2,3]), also allowing for temperature variations and replacing the old Eq. (43), as

$$\begin{aligned}
\underline{\nabla} \mu_j &= R_G T \left(\underline{\nabla} \ln(x_j) + \underline{\nabla} \ln p \right) + \frac{\partial \mu_j}{\partial T} \underline{\nabla} T = \frac{R_G T}{x_j} \left(\underline{\nabla} x_j + x_j \underline{\nabla} \ln p \right) - S_j \underline{\nabla} T \\
&= \frac{R_G T}{x_j} \left(\underline{\nabla} x_j + x_j \underline{\nabla} \ln p \right) - \frac{1}{\rho_j} M_j s_j \underline{\nabla} T \quad \text{ideal gas}
\end{aligned} \ , \tag{4}$$

where S_j is the partial molar entropy for component j, leading to the following corrected Eq. (44) that replaces the old Eq. (44) as

$$\begin{aligned}\frac{\rho_i}{\rho}\sum_{j=1}^{n}\rho_j\nabla_\alpha\left(\frac{\mu_i}{M_i}-\frac{\mu_j}{M_j}\right) &= x_i c\nabla_\alpha\mu_i-\frac{\rho_i}{\rho}c\sum_{j=1}^{n}x_j\nabla_\alpha\mu_j \\ &= cR_GT\left(\underline{\nabla}x_i+x_i\underline{\nabla}\ln p-\frac{\rho_i}{\rho}\sum_{j=1}^{n}\left(\underline{\nabla}x_j+x_j\underline{\nabla}\ln p\right)\right)-s_i\underline{\nabla}T+\frac{\rho_i}{\rho}\sum_{j=1}^{n}\left(s_j\underline{\nabla}T\right) \\ &= cR_GT\left(\underline{\nabla}x_i+x_i\underline{\nabla}\ln p-\frac{\rho_i}{\rho}\underline{\nabla}\ln p\right)-\left(s_i-\frac{\rho_i}{\rho}s\right)\underline{\nabla}T \\ &= cR_GT\underline{\mathrm{d}}^{(i)}-\left(s_i-\frac{\rho_i}{\rho}s\right)\underline{\nabla}T,\quad \text{ideal gas}\end{aligned}. \tag{5}$$

Using the new Eq. (44), Eq. (5), and the known value for the parameter α_{i0} as provided by the old Eq. (39), we get a new Eq. (45) as

$$\frac{1}{cR_GT}\frac{\rho_i}{\rho}\sum_{j=1}^{n}\rho_j\nabla_\alpha\left(\frac{\mu_i}{M_i}-\frac{\mu_j}{M_j}\right)=\sum_{j=1}^{n}\frac{x_ix_j}{D_{ij}}\left(v_\alpha^{(j)}-v_\alpha^{(i)}\right)+\frac{1}{cT}\nabla_\alpha T\frac{\alpha_{i0}\lambda}{sR_GT}-\frac{1}{cR_GT}\left(s_i-\frac{\rho_i}{\rho}s\right)\underline{\nabla}T. \tag{6}$$

If we then use the first line in old Eq. (37), that we have shown above to be equivalent to Eq. (38) derived from the kinetic theory (Eq. (7.6-12) from ref[4]) to eliminate the dependence on the first temperature gradient in Eq. (6) it yields a new Eq. (46):

$$\frac{1}{cR_GT}\frac{\rho_i}{\rho}\sum_{j=1}^{n}\rho_j\nabla_\alpha\left(\frac{\mu_i}{M_i}-\frac{\mu_j}{M_j}\right)=\sum_{j=1}^{n}\frac{x_ix_j}{D_{ij}}\left(v_\alpha^{(j)}-v_\alpha^{(i)}\right)-\frac{\alpha_{i0}\lambda}{cR_Gs^2T^2}\left(\sum_{i=1}^{n}\left(\alpha_{i0}v_\alpha^{(i)}\right)+\alpha_{00}\frac{q_\alpha}{sT}\right)-\frac{1}{cR_GT}\left(s_i-\frac{\rho_i}{\rho}s\right)\underline{\nabla}T, \tag{7}$$

or, using old Eq. (39) to eliminate λ and after multiplying both sides by cR_GT we find as new Eq. (47):

$$\frac{\rho_i}{\rho}\sum_{j=1}^{n}\rho_j\nabla_\alpha\left(\frac{\mu_i}{M_i}-\frac{\mu_j}{M_j}\right)=cR_GT\sum_{j=1}^{n}\frac{x_ix_j}{D_{ij}}\left(v_\alpha^{(j)}-v_\alpha^{(i)}\right)-\frac{\alpha_{i0}}{\alpha_{00}}\left(\sum_{j=1}^{n}\left(\alpha_{j0}v_\alpha^{(j)}\right)\right)-\alpha_{i0}\frac{q_\alpha}{sT}-\left(s_i-\frac{\rho_i}{\rho}s\right)\underline{\nabla}T\quad. \tag{8}$$

Finally, this can be written explicitly in terms of velocity differences by using the condition given by old Eq. (16) as new Eq. (48):

$$\frac{\rho_i}{\rho}\sum_{j=1}^{n}\rho_j\nabla_\alpha\left(\frac{\mu_i}{M_i}-\frac{\mu_j}{M_j}\right)=cR_GT\sum_{j=1}^{n}\frac{x_ix_j}{D_{ij}}\left(v_\alpha^{(j)}-v_\alpha^{(i)}\right)-\frac{\alpha_{i0}}{\alpha_{00}}\left(\sum_{j=1}^{n}\left(\alpha_{j0}\left(v_\alpha^{(j)}-v_\alpha^{(i)}\right)\right)\right)-\alpha_{i0}\frac{q_\alpha}{sT}-\left(s_i-\frac{\rho_i}{\rho}s\right)\underline{\nabla}T \tag{9}$$

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All other equations and statements in the paper[1] remain unchanged.

References

[1]Beris, A. N., Jariwala, S. and Wagner, N.J., "Flux-Based Modeling of Heat and Mass Transfer in Multicomponent Systems," *Physics of Fluids* **34**, 033113 (2022).

[2]Kestin, J., "A Course in Thermodynamics," Vols 1, 2, Revised Printing, Hemisphere Publishing Co., New York: 1979

[3]Kondepudi, D. and Prigogine, I., "Modern Thermodynamics," Wiley, Chichester: 1998.

[4]Hirschfelder, J. O., Curtiss, C. F. and Bird, R. B., "Molecular Theory of Gases and Liquids," Corrected Printing with Notes Added, John Wiley & Sons, New York: 1964.