A multi-component drop evaporation model based on analytical solution of Stefan-Maxwell equations

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Abstract

A novel evaporation model for multi-component spherical drop has been developed by analytically solving the Stefan-Maxwell equations under spherical symmetry assumptions. The evaporation rate predicted by the new model are compared with the predictions obtained by previous models based on Fick's law approximation, under steady-state isothermal conditions for a wide range of gas and drop temperature and compositions. The effect of non-isothermal conditions are considered in a simplified way, through the effect of temperature on the reference value of gas density and mass diffusion coefficients. The Fick's law based models are found to generally under-predict the total evaporation rate, particularly at higher evaporation rate conditions.

1 Nomenclature

\mathcal{A}		coefficient matrix	-		
в		source term vector	-		
\mathbf{C}_0		constant of integration vector	-		
c		molar density	$kmol/m^3$		
D		species diffusion coefficient	m^2/s		
I		unit matrix	-		
m_{ev}	ט	evaporation rate	kg/s		
M_{rr}	ı	molar mass	kg/kmol		
\mathbf{n}		mass flux vector	kg/m^2s		
Ν		molar flux vector	$kmol/m^2s$		
N_{ei}	,	molar evaporation rate	kmol/s		
N_r		molar flux vector radial component	$kmol/m^2s$		
r		radial coordinate	m		
R		universal gas constant	J/kmolK		
R_0		drop radius	m		
Т		temperature	Κ		
y		molar fraction	-		
Greek symbols					
	ε	mass evaporation rate fraction	-		
	ζ	non-dimensional <i>coordinate</i>	-		
	ν	molar evaporation rate fraction	-		
	ρ	mass density	kg/m^3		
	φ	species diffusion coefficient ratio	-		
	χ	mass fraction	-		
	Ψ	molar fraction vector	-		

Subscripts

ev	eva	poration		
ref	refe	rence		
s	dro	p surface		
v	vapour			
∞	aml	pient conditions		
Superscripts				
F		Fick model		
j, k, n	, p	indexes		
m		mixture		
T		total		
^		non-dimensional		

2 Introduction

In many applied fields, like spray combustion, spray painting, aerosol for medical applications, etc., the evaporation of a liquid drop floating in a gaseous atmosphere is a phenomenon of paramount importance. Since the early work of Maxwell on this subject [1], back in 1877, a relatively vast literature has become available, reporting the valuable findings that helped to increase our understanding of the complex phenomena involved. During the evaporation of a liquid drop different simultaneous mechanisms of heat (conduction, convection and possibly radiation) and mass (convection and diffusion) transfer, between the drop surface and the surroundings, influence and drive the drop evaporation (see [2] for a detailed description).

A wide literature is available on the modelling of the above mentioned phenomena, (see for example [3], [4] for a review), particularly for a single component drop, and such models are often used to simulate evaporating spray, as part of CFD methodologies, although detailed models based on single drop analysis have to be simplified to be CPU efficient.

A far less amount of literature is available on the more complex problem of evaporation from a multi-component droplet, since in this case the simultaneous diffusive-convective mass transfer from the drop to the gas cannot be simply modelled and/or experimentally studied. The Stefan flow combined with the differential diffusion of each component into the gas-vapour mixture renders the problem much more complex than that relative to single component drops. A typical approach to this problem is to model the diffusive mass flux of each component by the well-known Fick's law, which however would exactly hold only for binary diffusion (i.e. single component vapour diffusing in a gas) [5].

The available studies on multi-component diffusion from spherical drops are based on the simplified extension of single component models (namely the Fuchs' model [6] for evaporation in a still gas and the Abramzon and Sirignano extension [7] to Re>0), with different simplifying conditions. A typical simplification is to consider the vapour mixture as a single component, defining an average diffusive coefficient into the gaseous atmosphere, that can be done in different ways. For example, [8] proposes to evaluate the diffusion coefficient of the mean vapour mixture in air according to the Wilke formula [9], which takes into account the physical properties of each species.

A slightly more detailed numerical model accounts for the difference between the diffusion coefficient of each species, thus evaluating the evaporation rate of each component [10]. Another similar approach was suggested by [11] to account for differential diffusion of each component, namely the application of the single component model using an equivalent drop radius for each component, based on the volume composition of the real drop. Recently, [12] proposed an analytical model of multi-component drop evaporation accounting for the inter-species mass diffusion in the gaseous mixture and suggested a simpler model, based on the single-component analogy, with a new definition of the mean mass diffusion coefficient, which results were found to be in good agreement with the more accurate solution. Ebrahimian and Habchi [13] developed a multi-component drop evaporation model, which proposes a new expression to evaluate the Stefan velocity based on [14], implementing it in a numerical code to compare the predictions with previous models and with available experimental results. The model was used to investigate the effect on drop evaporation rate of various model assumptions, like the infinite thermal conductivity assumption [3], the physical property averaging [15], the effect of high pressure and temperature conditions [16] and of gravity and natural convection [17], [18], [19], [20].

All the previously mentioned models are based on the Fick's law approximation, although a more accurate way to cope with multi-component diffusion is through the so-called Stefan-Maxwell equations (see [5] for a comprehensive analysis), that can account for the mutual interaction among the mass fluxes of all the components. The major complexity of this approach comes from the fact that a system of *coupled* differential equations (one for each component) must be solved, together with continuity equations.

The next sections describe the mathematical formulation of a new model for drop evaporation based on the analytic solution of Stefan-Maxwell equations, under some simplifying hypotheses. A comparison with results obtained with previous models are reported. Finally the main conclusions arisen from the present investigation are briefly summarised.

3 Model equations

Steady multi-component diffusion can be correctly modelled by the Stefan-Maxwell (S-M) equations, that can be written, for a mixture of n + 1 species, neglecting Soret effect and diffusion due to pressure gradients and to external force, as [5]:

$$\nabla y^{(p)} = \sum_{k=0}^{n} \frac{1}{cD_{pk}} \left(y^{(p)} \mathbf{N}^{(k)} - y^{(k)} \mathbf{N}^{(p)} \right)$$
(1)

where c is the molar density, $D_{pk} = D_{kp}$ are the binary diffusion coefficient of p-component into k-component, $y^{(p)}$ is the molar fraction of p-component, $\mathbf{N}^{(p)}$ is the molar flux of the p-component, which is related to the mass flux by $\mathbf{n}^{(p)} = \mathbf{N}^{(p)} M m^{(p)}$ where $M m^{(p)}$ is the molar mass of the p-component.

Considering a multi-component spherical drop evaporating in a gaseous atmosphere, spherical symmetry assumption allows to retain only the radial component of the species molar fluxes $(N_r^{(k)})$. Assuming a still drop surface and a neglectful gas diffusion into the liquid drop, the gas flux $(N_r^{(0)})$ is necessarily nil everywhere, then:

$$N_r^{(k)} = \frac{N_{ev}^{(k)}}{4\pi r^2} \text{ for } k = 1...n$$
$$N_r^{(0)} = 0$$

where $N_{ev}^{(k)}$ is the molar evaporation rate of the k-component (again $m_{ev}^{(k)} = N_{ev}^{(k)} M m^{(k)}$, where $m_{ev}^{(k)}$ is the evaporation mass rate).

With the change of variable $\zeta = \frac{R_0}{r}$ and introducing the non-dimensional molar evaporation rate $\hat{N}_{ev}^{(k)} = \frac{N_{ev}^{(k)}}{4\pi R_0 c D_{ref}}$, where D_{ref} is a suitable reference value for the diffusion coefficients, equation (1) yields the ODE system:

$$\frac{dy^{(p)}}{d\zeta} = -\hat{N}_{ev}^{(T)} \sum_{k=0}^{n} \varphi^{pk} \left(y^{(p)} \nu^{(k)} - y^{(k)} \nu^{(p)} \right)$$
(2)

where $\varphi^{pk} = \frac{D_{ref}}{D_{pk}}$ and $\nu^{(k)} = \frac{N_{ev}^{(k)}}{\sum_{p=1}^{n} N_{ev}^{(p)}}$. Since $\nu^{(0)} = 0$ and $\sum_{k=0}^{n} y^{(k)} = 1$ and setting $\varphi^{pp} \equiv \varphi^{0p} \equiv \varphi^{p0}$ the system (2) can be written in matrix form as:

$$\frac{d}{d\zeta}\Psi = \mathcal{A}\Psi + \mathbf{B} \tag{3}$$

where

$$\mathcal{A} = \hat{N}_{ev}^{(T)} \begin{bmatrix} -\sum_{k=1}^{n} \varphi^{1k} \nu^{(k)} & \nu^{(1)} (\varphi^{12} - \varphi^{10}) & \dots & \nu^{(1)} (\varphi^{1n} - \varphi^{10}) \\ \nu^{(2)} (\varphi^{21} - \varphi^{20}) & -\sum_{k=1}^{n} \varphi^{2k} \nu^{(k)} & \dots & \nu^{(2)} (\varphi^{2n} - \varphi^{20}) \\ \dots & \dots & \dots & \dots \\ \nu^{(n)} (\varphi^{n1} - \varphi^{n0}) & \nu^{(n)} (\varphi^{n2} - \varphi^{n0}) & \dots & -\sum_{k=1}^{n} \varphi^{nk} \nu^{(k)} \end{bmatrix}$$

$$\Psi = \begin{bmatrix} y^{(1)} & \dots & y^{(n)} \end{bmatrix}^{T}$$

$$\mathbf{B} = \hat{N}_{ev}^{(T)} \begin{bmatrix} \nu^{(1)} \varphi^{10}, \nu^{(2)} \varphi^{20}, \dots, \nu^{(n)} \varphi^{n0} \end{bmatrix}^{T}$$
(4)

A simple solution of (3) can be found assuming a constant value of the molar density c. The constant density (molar or mass) assumption is common in the majority of drop evaporation models, and it was shown [21] that it may become influential for evaporation in high temperature environments.

The general solution takes then the form:

$$\Psi = e^{\mathcal{A}\zeta} \cdot \mathbf{C}_0 - \mathcal{A}^{-1}\mathbf{B}$$

and the vector \mathbf{C}_0 has to be determined from the B.C.:

$$\Psi (1) = \left[y_s^{(1)}, ..., y_s^{(n)} \right] = \Psi_s$$
$$\Psi (0) = \left[y_\infty^{(1)}, ..., y_\infty^{(n)} \right] = \Psi_\infty$$

and a simple manipulation yields:

$$e^{\mathcal{A}} \left(\mathcal{A} \Psi_{\infty} + \mathbf{B} \right) - \left(\mathcal{A} \Psi_{s} + \mathbf{B} \right) = 0$$
(5)

This is a set of n non linear algebraic equation for the n + 1 unknowns $\left(\nu^{(k)}, \widehat{N}_{ev}^{(T)}\right)$; the problem is then closed by the condition:

$$\sum_{k} \nu^{(k)} = 1$$

It must be noticed that equations (1) for the case of a binary mixture (i.e. n = 1, single component drop) yield the equation set:

$$\mathbf{N}^{(k)} = y^{(k)}\mathbf{N}^{(T)} - cD_v \nabla y^{(k)}$$

$$k = 0, 1$$
(6)

which is a form of the Fick's law (see [5], Tab 8.4.6-1, eq. D), that can be transformed, after some manipulations, into the "mass" form:

$$\mathbf{n}^{(k)} = \chi^{(k)} \mathbf{n}^{(T)} - \rho D_v \nabla \chi^{(k)}$$

$$k = 0, 1$$
(7)

where $D_v = D_{10} = D_{01}$. The two forms are a direct consequence of S-M equations and they are equivalent only for the case of single component drop (n = 1). For multi-component drop (n > 1) the approximate forms:

k

$$\mathbf{N}^{(k)} = y^{(k)} \mathbf{N}^{(T)} - c D^{(k,m)} \nabla y^{(k)}$$
(8)

$$\mathbf{n}^{(k)} = \chi^{(k)} \mathbf{n}^{(T)} - \rho D^{(k,m)} \nabla \chi^{(k)}$$
(9)

$$= 0, n$$

are often used in drop evaporation modelling (particularly eq. (9)), where $D^{(k,m)}$ is the diffusion coefficient of the k-component in the gas/vapour mixtures and can be evaluated from the binary diffusion coefficients (D_{pk}) by semiempirical laws [9], [22]. But it is important to notice that in that case (n > 1) the two forms (8) and (9) are not equivalent and they are expected to yield slightly different solutions. Simple analytical solutions of (8) and (9) can be found imposing the constancy of the molar (c) or the mass (ρ) densities respectively. These assumptions are not equivalent and then the two equations yield different solutions also for the case of single component drops, but it is clear that in such case the solution of the molar form (8) would yield exactly the same solution of the system (3) since for n = 1 the two sets of equations are equivalent and the same assumption (c = const) is made.

The total evaporation rate and the evaporation rate fractions $\varepsilon^{(k)} = \frac{m_{ev}^{(k)}}{m_{ev}^{(T)}}$ when the mass form of the Fick's law based model (9) is used are given by solving the following set of non-linear algebraic equations (see [12]):

$$\sum_{k=1}^{n} \frac{\chi_{\infty}^{(k)} - \chi_{s}^{(k)}}{\left(e^{-\frac{m_{ev}^{(T)}}{4\pi R_{0}\rho D^{(k,m)}}} - 1\right)} = 1 - \sum_{k=1}^{n} \chi_{\infty}^{(k)}; \quad \varepsilon^{(k)} = \frac{\chi_{s}^{(k)} - \chi_{\infty}^{(k)} e^{-\frac{m_{ev}^{(T)}}{4\pi R_{0}\rho D^{(k,m)}}}}{1 - e^{-\frac{m_{ev}^{(T)}}{4\pi R_{0}\rho D^{(k,m)}}}}$$
(10)

Following the same procedure, the solution for the molar form of the Fick's law based equations (8) yields a similar set of equations:

$$\sum_{k=1}^{n} \frac{y_{\infty}^{(k)} - y_{s}^{(k)}}{\left(e^{-\frac{N_{ev}^{(T)}}{4\pi R_{0}cD^{(k,m)}}} - 1\right)} = 1 - \sum_{k=1}^{n} y_{\infty}^{(k)}; \quad \nu^{(k)} = \frac{y_{s}^{(k)} - y_{\infty}^{(k)}e^{-\frac{N_{ev}^{(T)}}{4\pi R_{0}cD^{(k,m)}}}}{1 - e^{-\frac{N_{ev}^{(T)}}{4\pi R_{0}cD^{(k,m)}}}} \tag{11}$$

where

$$\varepsilon^{(k)} = \frac{Mm^{(k)}\nu^{(k)}}{\sum_{p=1}^{n} Mm^{(p)}\nu^{(p)}}; \quad m_{ev}^{(T)} = N_{ev}^{(T)} \sum_{p=1}^{n} Mm^{(p)}\nu^{(p)}$$

The Stefan-Maxwell approach is expected to yield more accurate results than the two Fick's law based models since the former takes into account in a more proper way the inter-diffusion between different species. However the above presented solutions are all affected by approximations among which the constancy of the density is probably the most severe one.

To notice that the differential equation set (8) can be written in a form similar to equation (3):

$$\frac{d}{d\zeta}\Psi = \mathcal{A}^F\Psi + \mathbf{B}^F \tag{12}$$

where

$$\mathcal{A}^{F} = \hat{N}_{ev}^{(T),F} \begin{bmatrix} -\beta^{1} & 0 & \dots & 0 \\ 0 & -\beta^{2} & \dots & 0 \\ \dots & \dots & \dots & \dots \\ 0 & 0 & \dots & -\beta^{n} \end{bmatrix}$$

$$\mathbf{B}^{F} = \hat{N}_{ev}^{(T)} \left[\nu^{(1)}\beta^{1}, \nu^{(2)}\beta^{2}, \dots, \nu^{(n)}\beta^{n} \right]^{T}$$
(13)

and $\beta^k = \frac{D_{ref}}{D^{(k,m)}}$.

Since the two equations ((3) and (12)) have a similar form, their solutions can be written in a unique form as:

$$\Theta\left(\zeta\right) = \mathcal{Z}\left(\zeta\right)\Theta_s$$

where:

$$\mathcal{Z}\left(\zeta\right) = \mathcal{E}\left(\zeta\right) \mathcal{E}\left(1\right)^{-1}$$

being $\Theta(\zeta) = (\Psi(\zeta) - \Psi_{\infty})$ and $\mathcal{E}(\zeta) = (e^{\mathcal{A}^* \zeta} - \mathcal{I})$, and the matrix \mathcal{A}^* is given by equations (4) or (13) for the two models. The differences on the vapour molar fraction distributions predicted by the two models are then ascribed to the differences in the matrices $\mathcal{Z}(\zeta)$, which ultimately depend on the characteristic matrices \mathcal{A} and \mathcal{A}^F .

4 Results and discussions

This section reports the results of a comparison among the multi-component vaporisation models developed in the previous section. A parametrical analysis has been performed, assuming a single spherical drop vaporising in stagnant air at steady-state conditions, under atmospheric pressure. Different drop compositions and temperatures have been considered. The binary diffusion coefficients and the mass and molar gas densities are calculated at reference temperature and concentrations, according to the formulas [23]:

$$T_{ref} = \frac{2T_s + T_{\infty}}{3}; \quad y_{ref}^{(k)} = \frac{2y_s^{(k)} + y_{\infty}^{(k)}}{3}$$
(14)

The diffusion coefficient of the k-component in the gas/vapour mixtures $D^{(k,m)}$ is approximated by the Wilke formula [9]:

$$D^{(k,m)} = \frac{1 - y_{ref}^{(k)}}{\sum_{\substack{j=0\\j \neq k}}^{n} \frac{y_{ref}^{(j)}}{D_{jk}}}$$

while the binary diffusion coefficients D_{jk} are calculated according to the Fuller et al. formula [24] at reference temperature conditions.

The vapour concentrations at drop surface, needed to calculate $y_s^{(k)}$, were evaluated by Raoult's law. In the following analysis the vapour concentration of all components in the bulk gas were taken equal to zero.

Figure 1(a) shows the effect of drop temperature on the total drop evaporation rate of a two component drop made by 50% ethanol and 50% acetone, under steady-state and isothermal (i.e. $T_s = T_{\infty}$) conditions, as predicted by the S-M model and by the two Fick's law based models (equations (10) and (11)). The results show that both Fick's models generally underpredict the evaporation rate for temperatures larger than ambient conditions and the differences increase with temperature. The maximum difference is achieved with both the forms of the Fick's model at the highest drop temperature and it is equal to about 10%, as shown in Figure 1(b), which reports, as function of temperature, the relative difference of the total mass evaporation rate between the results from the Fick's models and the S-M model, $\Delta m_{ev}^{(T)}\% = \frac{m_{ev}^{(T),Fick} - m_{ev}^{(T), S-M}}{m_{ev}^{(T), S-M}} \times 100.$

The effect of drop temperature on the mass evaporation rate fractions, $\varepsilon^{(ethanol)}$, for ethanol component is shown in Figure 2, which enlightens that the evaporation rate fractions predicted by the Fick's models, for the selected range of operating conditions, differs from the S-M model predictions by *less than* 5%. This general behaviour is confirmed also for other liquid mixtures, as reported in the four graphs of Figure 3, which plots the effect of drop temperature on the total mass evaporation rate, under steady-state isothermal evaporating conditions, for drops made of two, three and four components, for some alkanes, hydrocarbons and/or alcohols and water. As above mentioned, the Fick's law based models *generally* underestimate the evaporation rate, particularly at higher temperatures.

The effect of drop composition was studied by varying the mass fraction $\chi_l^{(k)}$ of one component in a two-component drop. Figure 4(a) reports the total mass evaporation rate for a drop made by ethanol and acetone, under steady-state isothermal conditions with $T_s = 325$ K. The evaporation rate is calculated by the S-M model and the two Fick's models, varying the mass fraction of ethanol from 0 up to 1. As above mentioned, since constancy of molar density is assumed when the S-M equations are solved, the molar form of the Fick's law based model and S-M model yield identical results when a single-component drop is considered (i.e. when $\chi_l^{(k)}$ is equal to 0 or 1). The largest deviation on the absolute value of the evaporation rate is found when none of the species mass fraction prevails on the other. The corresponding values of the ethanol evaporation rate fraction ($e^{ethanol}$) as function of ethanol mass fraction are shown in Figure 4(b). The differences among the evaporation rate fractions predicted by the three models are relatively small.

Figure 5 and 6 show similar results for two-component drops made by n-tetradecane and n-hexadecane at 500 K (close to the boiling point of pure n-tetradecane) and for drops made by different composition of n-decane and 3-pentanone at 370 K (close to the boiling point of pure 3-pentanone), respectively. As a general rule, the three models yield comparable predictions of the mass evaporation rate fractions, $\varepsilon^{(k)}$, at any drop composition, whereas the predictions of the total mass evaporation rate may differ considerably.

The effect of non-isothermal drop evaporation are taken into account in these models through the values of the reference temperature at which molar and mass densities and diffusion coefficients are calculated (equation (14)). Figure 7 reports the steady-state total mass evaporation rate as predicted by the S-M and by the two Fick's models, for a two-component drop made by 50% ethanol and 50% acetone at 325 K, vaporising in stagnant air, which temperature, T_{∞} , varies from 325 K up to 1000 K. The results show that the effect of non-isothermal conditions does not significantly change the above described evaporation behaviour, and the Fick's law based models consistently underpredict the total evaporation rate.

5 Conclusions

A new multi-component drop evaporation model was developed on the basis of an analytical solution of the Stefan-Maxwell equations. The model was compared to Fick's law based models, previously developed, by evaluating the steady-state total evaporation rate and evaporation rate fractions for different gas and drop temperature and compositions.

The predictions from the Fick's law based models generally underestimate the total evaporation rate, particularly at higher temperatures, for different drop compositions (alkanes, hydrocarbons, alcohols and/or water). The three models predict comparable values of evaporation rate fractions, also under high evaporation rate conditions. The largest deviation on the absolute value of the evaporation rate is found when none of the species mass fraction prevails on the other.

The effect of non-isothermal conditions does not modify the general results previously reported.

6 Figure captions

Figure 1. Effect of drop/gas temperature on (a) total mass evaporation rate and (b) percentage deviation of total mass evaporation rate, as predicted by S-M and the two Fick's law based models (F^{massic} and F^{molar}), for a 50% ethanol / 50% acetone drop.

Figure 2. Effect of drop/gas temperature on ethanol evaporation rate fraction, as predicted by S-M and the two Fick's law based models (F^{massic} and F^{molar}), for a 50% ethanol / 50% acetone drop.

Figure 3. Effect of drop/gas temperature on total mass evaporation rate, as predicted by S-M and the two Fick's law based models (F^{massic} and F^{molar}), for (a) 50% n-tetradecane / 50% n-hexadecane drop, (b) 90% n-decane / 10% 3-pentanone drop, (c) 33% n-octane / 33% n-decane / 34% n-dodecane drop, (d) 25% water / 25% methanol / 25% ethanol / 25% butanol drop.

Figure 4. Effect of ethanol liquid mass fraction on (a) total mass evaporation rate and (b) ethanol evaporation rate fractions, as predicted by S-M and the two Fick's law based models (F^{massic} and F^{molar}), for ethanol / acetone drops ($T_s = T_{\infty} = 325$ K).

Figure 5. Effect of n-tetradecane liquid mass fraction on (a) total mass evaporation rate and (b) n-tetradecane

evaporation rate fractions, as predicted by S-M and the two Fick's law based models (F^{massic} and F^{molar}), for n-tetradecane / n-hexadecane drops ($T_s = T_{\infty} = 500$ K).

Figure 6. Effect of n-decane liquid mass fraction on (a) total mass evaporation rate and (b) n-decane evaporation rate fractions, as predicted by S-M and the two Fick's law based models (F^{massic} and F^{molar}), for n-decane / 3pentanone drops ($T_s = T_{\infty} = 370$ K).

Figure 7. Effect of gas temperature on total mass evaporation rate, as predicted by S-M and the two Fick's law based models (F^{massic} and F^{molar}), for a 50% ethanol / 50% acetone drop ($T_s = 325$ K).

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