

A Simplified Procedure for the Solution of the Dusty Gas Model Equations for Steady-state Transport in Non-reacting Systems

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ABSTRACT

The Dusty Gas Model equations for diffusion of n -component mixtures in porous media have been conveniently cast into n -dimensional matrix notation. A simplified linearized procedure, involving the assumption of a constant matrix of transfer coefficients, is developed for the calculation of the steady-state fluxes in non-reacting systems. Published experimental data are used to test the accuracy of the linearized procedure and it is concluded that the linearized procedure is of good accuracy.

The procedure can be extended to the analysis of diffusion of non-ideal liquid mixtures in porous media.

1. INTRODUCTION

In recent years the Dusty Gas Model has gained wide acceptance for application to the description of multicomponent ideal gas mixtures in porous media. The theory has been well discussed in two excellent texts by Jackson [1] and Mason and Malinauskas [2]. Even for the simple case of steady-state diffusion in non-reacting systems, the Dusty Gas Model equations must be solved numerically, though exact analytic solutions are available for some very special cases [1 - 3].

In this paper we cast the Dusty Gas Model equations into convenient n -dimensional matrix notation and develop a simplified, linearized procedure for the calculation of the steady-state diffusion fluxes. The developed procedure is tested against published experimental data and also against an available exact analytical solution in a special case.

The results of this study will be of use in design calculations for adsorptive and

membrane separations, catalytic reactors and other situations involving porous transport.

The analysis is extendible to the description of mass transport of non-ideal liquid mixtures in porous media.

2. THEORETICAL DEVELOPMENT

For diffusion in "open space", the correct description of the diffusion process is given by the generalized Maxwell-Stefan diffusion equations [4, 5]

$$d_i \equiv \frac{x_i}{RT} \nabla_{T,p} \mu_i + \frac{(\phi_i - \omega_i)}{c_t RT} \nabla p - \frac{1}{c_t RT} \left(c_i F_i - \omega_i \sum_j^n c_j F_j \right) = \sum_{\substack{j=1 \\ j \neq i}}^n \frac{x_i N_j - x_j N_i}{c_t D_{ij}} \quad i = 1, 2, \dots, n \quad (1)$$

For ideal gas mixtures the following simplifications can be made:

$$\frac{x_i}{RT} \nabla_{T,p} \mu_i = \nabla x_i \quad (\text{thermodynamic ideality}) \quad (2)$$

$$\phi_i = x_i \quad (\text{volume fraction} = \text{mole fraction}) \quad (3)$$

$$c_t = \frac{p}{RT} \quad (\text{ideal gas law}) \quad (4)$$

and so the generalized Maxwell-Stefan diffusion eqns. (1) reduce to

$$d_i \equiv \nabla x_i + \frac{(x_i - \omega_i)}{p} \nabla p - \frac{1}{p} \left(c_i F_i - \omega_i \sum_j^n c_j F_j \right) = \sum_{\substack{j=1 \\ j \neq i}}^n \frac{x_i N_j - x_j N_i}{c_t D_{ij}} \quad i = 1, 2, \dots, n \quad (5)$$

In the Dusty Gas Model the "walls" of the porous material are modelled as the $n + 1$ th component in the mixture, the so-called "dust" species. These dust species are giant molecules ($M_{n+1} \rightarrow \infty$), uniformly distributed in space ($\nabla c_{n+1} = 0$), and are held motionless ($N_{n+1} = 0$) by unspecified external forces acting on them ($c_{n+1} F_{n+1} = \nabla p$). With these model assumptions the set of $n + 1$ Dusty Gas eqns. (5) can be simplified to give (see Jackson [1] and Mason and Malinauskas [2] for detailed derivation)

$$\begin{aligned} \frac{p}{RT} d_i &\equiv \frac{p}{RT} \nabla x_i + \frac{x_i}{RT} \nabla p - \frac{c_i F_i}{RT} \\ &\equiv \frac{1}{RT} \nabla p_i - \frac{c_i F_i}{RT} \\ &= \sum_{\substack{j=1 \\ j \neq i}}^n \frac{x_i N_j^D - x_j N_i^D}{D_{ij}^e} - \frac{N_i^D}{D_{iK}^e} \\ & \quad i = 1, 2, \dots, n \quad (6) \end{aligned}$$

We have introduced a superscript D on the fluxes N_i to emphasize the fact that only the diffusional contribution to the transport is being considered here; the parallel viscous contribution arising from the flow of the "mixture as a whole" through the porous medium must be added to N_i^D to obtain the total fluxes

$$N_i = N_i^D + N_i^V \quad (7)$$

The viscous contribution, which is non-separative, is

$$N_i^V = -x_i \frac{B_0 p}{\mu RT} \left(\nabla p - \sum_j^n c_j F_j \right) \quad (8)$$

For a cylindrical pore of radius r_0 , the permeability B_0 is

$$B_0 = \frac{r_0^2}{8} \quad (9)$$

In the Dusty Gas eqns. (6) we have introduced a superscript e on the diffusion coefficients because the diffusion coefficients have to be modified to take account of the porosity ϵ and the tortuosity τ of the medium

$$D_{ij}^e = \frac{\epsilon}{\tau} D_{ij} \quad D_{iK}^e = \frac{\epsilon}{\tau} D_{iK} \quad (10)$$

The bulk gas phase Maxwell-Stefan diffusion coefficients D_{ij} can be estimated by semi-

empirical procedures based on the kinetic gas theory [6]. The Knudsen diffusion coefficient for a cylindrical pore is also obtained from the kinetic gas theory

$$D_{iK} = \frac{2}{3} r_0 \left(\frac{8RT}{\pi M_i} \right)^{1/2} \quad (11)$$

Both the porosity and the tortuosity reduce to unity for a cylindrical pore

$$\epsilon = 1 \quad \tau = 1 \quad (\text{cylindrical pore}) \quad (12)$$

We may combine eqns. (6) - (8) to obtain the working form of the Dusty Gas Model equations [1, 2]

$$\begin{aligned} \frac{p}{RT} d_i &\equiv \frac{p}{RT} \nabla x_i + \frac{x_i}{RT} \nabla p - \frac{c_i F_i}{RT} \\ &\quad + \frac{x_i B_0 p}{\mu RT D_{iK}^e} \left(\nabla p - \sum_j^n c_j F_j \right) \\ &= \sum_{\substack{j=1 \\ j \neq i}}^n \frac{x_i N_j - x_j N_i}{D_{ij}^e} - \frac{N_i}{D_{iK}^e} \\ & \quad i = 1, 2, \dots, n \quad (13) \end{aligned}$$

For the usual cases of importance in chemical engineering the external forces acting on the species $i = 1, 2, \dots, n$, F_i , are absent and eqns. (13) reduce to [1, 2]

$$\begin{aligned} \frac{p}{RT} d_i &\equiv \frac{p}{RT} \nabla x_i + \frac{x_i}{RT} \left(1 + \frac{B_0 p}{\mu D_{iK}^e} \right) \nabla p \\ &= \sum_{\substack{j=1 \\ j \neq i}}^n \frac{x_i N_j - x_j N_i}{D_{ij}^e} - \frac{N_i}{D_{iK}^e} \\ & \quad i = 1, 2, \dots, n \quad (14) \end{aligned}$$

There are n independent eqns. (14), n independent fluxes N_i , n independent gradients (driving forces): $\nabla x_1, \nabla x_2, \dots, \nabla x_{n-1}$, and ∇p .

Summing over the n species, eqn. (14) yields

$$RT \sum_{i=1}^n \frac{N_i}{D_{iK}^e} = - \left(1 + \frac{B_0 p}{\mu} \sum_{i=1}^n \frac{x_i}{D_{iK}^e} \right) \nabla p \quad (15)$$

If the pressure gradients are absent or negligibly small, then eqns. (15) reduce in view of eqn. (11) to

$$\sum_{i=1}^n N_i M_i^{1/2} = 0 \quad (\nabla p = 0) \quad (16)$$

and so equimolar diffusion is only possible if the transferring species all have the same molecular weight.

The solution of eqns. (14) to obtain the n fluxes N_i requires to be carried out numerically in the general case though analytic solutions have been given for some very special cases [1-3, 7]. A convenient simplified procedure for the solution can be developed by first casting eqns. (14) into n -dimensional matrix notation

$$(N) = -\frac{p}{RT} [B]^{-1}(d) \quad (17)$$

where (d) represents an n -dimensional column matrix of driving forces

$$d_i \equiv \nabla x_i + x_i \left(1 + \frac{B_0 p}{\mu D_{iK}^e} \right) \frac{\nabla p}{p} \quad i = 1, 2, \dots, n \quad (18)$$

The $n \times n$ dimensional matrix of inverted diffusion coefficients $[B]$ has the elements

$$B_{ii} = \frac{1}{D_{iK}^e} + \sum_{\substack{j=1 \\ j \neq i}}^n \frac{x_j}{D_{ij}^e} \quad i = 1, 2, \dots, n \quad (19)$$

$$B_{ij} = -\frac{x_i}{D_{ij}^e} \quad i, j = 1, 2, \dots, n \quad (i \neq j) \quad (20)$$

The fluxes N_i can be calculated explicitly if we make the following simplifications:

(i) assume that the elements of $[B]$ are constant along the diffusion path; an averaged value for the mole fractions \bar{x}_i must be used in eqns. (19) and (20).

(ii) the gradients in eqns. (18) are estimated by assuming *linear* composition and pressure profiles along the length of the diffusion path δ :

$$-d_i \cong \frac{\Delta x_i}{\delta} + \bar{x}_i \left(1 + \frac{B_0 \bar{p}}{\mu D_{iK}^e} \right) \frac{\Delta p}{\delta} \frac{1}{\bar{p}} \quad (21)$$

With the above simplifications, eqn. (17) yields the values of the n fluxes explicitly. It remains now to check the accuracy of the linearized procedure suggested above.

3. TESTS OF THE LINEARIZED MODEL

Mason and Malinauskas have carried out extensive experimental investigations to test

the validity of the Dusty Gas Model; their work is conveniently summarized in ref. 2. We use their experimental data to test the accuracy of the linearized model developed above.

In one series of experiments [2], the fluxes for diffusion of a mixture of helium and argon, across a porous septum of thickness 4.47 mm, were measured. The pressure drop across the system was maintained at zero level, while the total system pressure was varied as shown in Fig. 1. The mole fraction driving force Δx_1 was maintained at 0.9628, with a mean composition $\bar{x}_1 = 0.5$. Figure 1 compares the linearized model predictions with the experimental data. Except for the flux of helium at high system pressure \bar{p} , the agreement between linearized theory and experimental data is extremely good. At high system pressures bulk diffusion is controlling and the discrepancy between experiment and theory could be due to an error in the estimation of the binary gas phase diffusivity D_{12} . The diffusivity values used in the model calculations correspond to those used by Mason and Malinauskas [2].

In a further set of experimental results with the system helium-argon, the total system pressure \bar{p} was maintained constant at

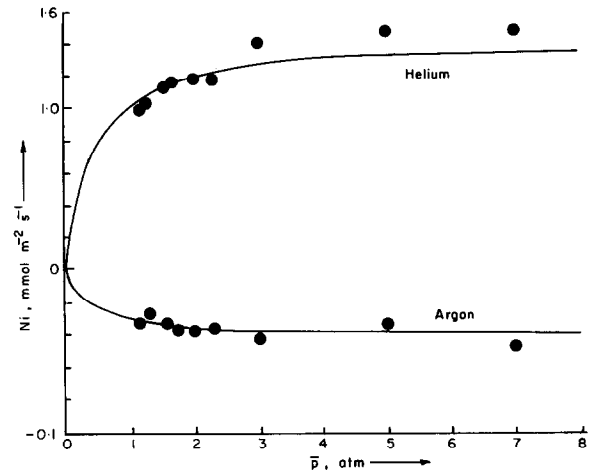


Fig. 1. Comparison of experimental data for the fluxes N_i with predictions of the linearized model. Experimental data given in Fig. 16 of ref. 2 for the system helium-argon. The experimental conditions are $\Delta x_1 = 0.9628$; $\bar{x}_1 = 0.5$; $T = 298.15$ K; $\Delta p = 0$; length of diffusion path $\delta = 4.47$ mm. The model parameters used in the calculations are: $B_0 = 2.13 \times 10^{-18}$ m²; $p D_{12}^e = 1.06 \times 10^{-8}$ atm m² s⁻¹; $D_{1K}^e = 3.93 \times 10^{-8}$ m² s⁻¹; $D_{2K}^e = 1.24 \times 10^{-8}$ m² s⁻¹; gas mixture viscosity $\mu = 22.8$ Pa s.

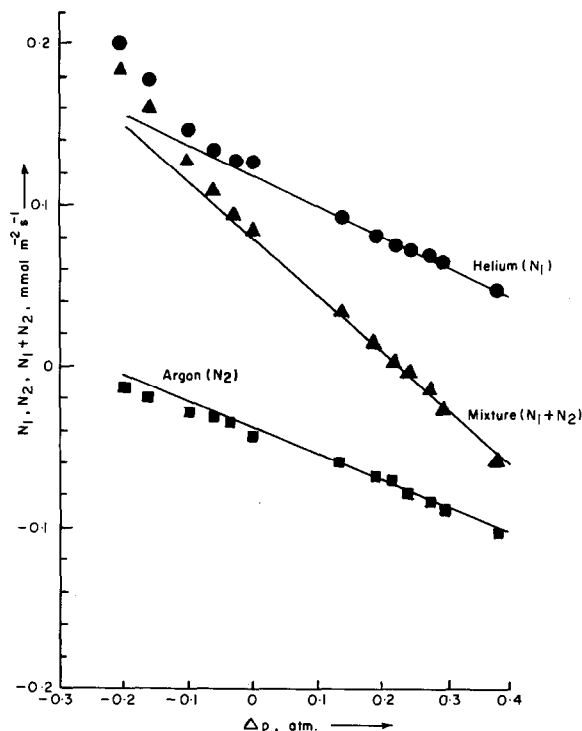


Fig. 2. Comparison of experimental fluxes N_1 , N_2 and $N_1 + N_2$ for the system helium (1)-argon (2) with predictions of the linearized model. The data are taken from Fig. 19 of ref. 2. The experimental conditions are: $\Delta x_1 = 0.9628$; $x_1 = 0.5$; $T = 298.15$ K; $\bar{p} = 1.96$ atm; length of diffusion path $\delta = 4.47$ mm. The model parameters used in the calculations are the same as those in Fig. 1.

1.96 atm, while the Δp across the porous septum was varied over a wide range as shown in Fig. 2. The composition driving force was maintained at $\Delta x_1 = 0.9628$. Figure 2 shows a comparison of the linearized model predictions with the experimental data. A positive Δp hinders the transport of helium and aids the transport of argon. The agreement between linearized theory and experiment is good for positive values of Δp . For negative Δp , the

pressure gradient acts in a manner such as to aid the transfer of helium and counter the transfer of argon. Since the flux of helium is significantly greater than that of argon, the countering effect of Δp on the argon transport serves to introduce larger absolute deviations between model predictions and experiment in comparison with the situation for positive Δp . Put another way, the accuracy of the linearized model can be expected to be reduced when the Δp term serves to counter the smaller flux and aid the larger flux. The relative deviations between experimental data and linearized model predictions may still be of acceptable accuracy for chemical engineering design calculations.

A more stringent test of the linearized model would be to test this against experimental data for a true multicomponent system with species made up of widely different sizes. Remick and Geankoplis [8] have provided such data for the diffusion of helium-neon-argon in a capillary diffusion cell. The experiments were performed in such a way that $\Delta p = 0$ across the diffusion path. The system pressure \bar{p} was varied and so were the composition driving forces Δx_i . The experimental data are summarized in Table 1. The predictions of the linearized model for the fluxes N_i are given in Table 2. Also given in Table 2, for comparison purposes, are the predictions of the fluxes N_i using an *exact* analytical procedure which we had presented earlier [3]. Comparison between the exact and linearized model predictions shows that the linearized model is an excellent approximation over a wide range of system pressure values. The small percentage deviations between model (exact or linearized) and experimental values of N_i point to the applicability of the Dusty Gas

TABLE 1

Experimental ternary diffusion data (Remick and Geankoplis [6]) for the system helium (1)-neon (2)-argon (3)

Run No.	\bar{p} (mm Hg abs)	t (°C)	Mole fractions				Fluxes (10^{-5} kmol m^{-2} s^{-1})		
			x_{10}	x_{16}	x_{20}	x_{26}	$-N_1$	N_2	N_3
1	0.450	27.6	0.0472	0.9471	0.5241	0.0343	3.7333	1.100	0.5966
2	1.100	28.3	0.0652	0.9610	0.5099	0.0251	6.810	1.923	1.065
3	4.09	27.2	0.0572	0.9619	0.5134	0.0244	19.98	5.064	2.843
4	22.66	27.8	0.0622	0.9625	0.5102	0.0237	44.14	9.437	5.495
5	303.19	27.5	0.0539	0.9601	0.5051	0.0252	49.25	12.81	7.472

TABLE 2

Model predictions (linearized and exact) and % deviations from measured data for N_i . The model parameters are: $\delta = 9.6$ mm; $2r_0 = 0.391$ μm ; $M_1 = 4$; $M_2 = 20.18$; $M_3 = 39.95$; $\mu = 22$ $\mu\text{Pa s}$; $\mathfrak{D}_{12} = 106.8$ $\text{mm}^2 \text{s}^{-1}$; $\mathfrak{D}_{13} = 73.4$ $\text{mm}^2 \text{s}^{-1}$; $\mathfrak{D}_{23} = 31.6$ $\text{mm}^2 \text{s}^{-1}$ (values at 1 atm); $\epsilon = 1$; $\tau = 1$

Run No.	Model	Predicted fluxes (10^{-5} kmol $\text{m}^{-2} \text{s}^{-1}$)			% Deviations from expt. (%)		
		$-N_1$	N_2	N_3	e_1	e_2	e_3
1	Exact	3.439	0.834	0.496	-7.88	-24.19	-16.94
	Linearized	3.438	0.834	0.495	-7.90	-24.21	-16.97
2	Exact	7.611	1.837	1.103	11.76	-4.5	3.55
	Linearized	7.600	1.834	1.101	11.60	-4.63	3.41
3	Exact	20.318	4.909	2.940	1.69	-3.06	3.41
	Linearized	20.124	4.862	2.912	0.72	-3.99	2.42
4	Exact	40.594	9.844	5.848	-8.03	4.32	6.42
	Linearized	39.093	9.479	5.633	-11.43	0.44	2.51
5	Exact	51.536	12.276	7.582	4.64	-4.17	1.47
	Linearized	48.518	11.552	7.141	-1.49	-9.82	-4.43

Model to the system considered. It must be pointed out here that our model predictions given in Table 2 appear to be somewhat superior to the predictions of the "exact" model of Remick and Geankoplis [8], who used the analytical solution of Cunningham and Geankoplis [9]. This latter procedure does not involve matrix algebra and it is possible that their model calculations had not fully converged; calculations of multi-component mass transfer using matrix methods are known to provide rapid convergence [7].

4. EXTENSION TO NON-IDEAL LIQUID MIXTURES

Using the generalized Maxwell-Stefan eqns. (1) as the starting point it is possible to parallel the derivation of the Dusty Gas Model valid for the general case of non-ideal fluid mixtures. The final equations obtained in this manner have the same form as eqn. (13), with the following alterations:

- (a) ∇x_i is to be replaced by $\frac{x_i}{RT} \nabla_{T,p} \mu_i$
- (b) $\nabla x_i + x_i \frac{\nabla p}{p}$ is to be replaced by $\frac{x_i}{RT} \nabla_T \mu_i$
- (c) the pressure is to be replaced by $c_i RT$

except in the ∇p term

With the above alterations, the final Dusty Fluid Model equations take the form

$$\begin{aligned}
 c_i d_i &\equiv \frac{c_i}{RT} (\nabla_T \mu_i - F_i) \\
 &+ \frac{c_i B_0}{\mu D_{iK}^e} \left(\nabla p - \sum_{j=1}^n c_j F_j \right) \\
 &= \sum_{\substack{j=1 \\ j \neq i}}^n \frac{x_i N_j - x_j N_i}{\mathfrak{D}_{ij}^e} - \frac{N_i}{D_{iK}^e} \\
 & \quad i = 1, 2, \dots, n \quad (22)
 \end{aligned}$$

Equation (22) represents the most convenient starting point for the discussions on transport of non-ideal fluid mixtures in porous media. Though eqns. (22) are formally equivalent to the corresponding Dusty Gas Model equations (13), there are some important limitations to eqns. (22). The diffusion coefficients D_{iK}^e representing transport of component i through the porous media have to be determined empirically. The same comments hold for the pair generalized Maxwell-Stefan diffusion coefficients \mathfrak{D}_{iK}^e . Equations (22) must therefore be viewed as an important aid to the interpretation of diffusion data in porous media. By distinguishing between the two types of coefficients \mathfrak{D}_{ij}^e and D_{iK}^e these coefficients can be better interpreted in terms of the molecular and thermodynamic properties of the fluid mixture and the structure of the porous medium.

In practical cases the most important external body force to be considered is the electrostatic potential, arising in the transport processes involving charged species

$$F_i = -z_i \mathcal{F} \nabla \phi \quad (23)$$

Due to the condition of electroneutrality in the bulk fluid mixture we must have

$$\sum_{i=1}^n c_i z_i = 0 \quad \sum_{i=1}^n c_i F_i = 0 \quad (24)$$

and so eqns. (22) simplify to

$$\begin{aligned} c_i d_i &\equiv \frac{c_i}{RT} \nabla_T \mu_i + c_i z_i \frac{\mathcal{F}}{RT} \nabla \phi + \frac{c_i B_0}{\mu D_{iK}^e} \nabla p \\ &= \sum_{\substack{j=1 \\ j \neq i}}^n \frac{x_j N_j - x_i N_i}{D_{ij}^e} - \frac{N_i}{D_{iK}^e} \\ & \quad i = 1, 2, \dots, n \end{aligned} \quad (25)$$

The set of eqns. (25) can be cast into n -dimensional matrix notation

$$(\mathbf{N}) = -c_i [\mathbf{B}]^{-1} (\mathbf{d}) \quad (26)$$

where the elements of $[\mathbf{B}]$ are given as before by eqns. (19) and (20). The linearization procedure can be extended to the non-ideal fluid mixture case. We calculate the elements of $[\mathbf{B}]$ at some averaged composition and assume it to be composition independent. The gradients are approximated by assuming linear profiles

$$-d_i \cong \frac{x_i}{RT} \frac{\Delta_T \mu_i}{\delta} + x_i z_i \frac{\mathcal{F}}{RT} \frac{\Delta \phi}{\delta} + \frac{x_i B_0}{\mu D_{iK}^e} \frac{\Delta p}{\delta} \quad (27)$$

With the above simplifications the fluxes N_i can be determined explicitly from eqn. (26). We firmly believe that the above procedure will be an invaluable aid to the interpretation of experimental data on transport processes in membranes.

The foregoing linearized treatment has been developed for planar slabs, but can be easily generalized to spherical and cylindrical geometries using generalized distance parameters as has been shown earlier [10].

5. CONCLUDING REMARKS

We have developed a simplified linearized procedure for the calculation of the fluxes N_i

for steady-state diffusion of multicomponent gaseous mixtures in porous media. The linearized procedure is shown to be of acceptable accuracy in a few comparisons with published experimental data. An additional advantage of the use of n -dimensional matrix notation is that this notation is much less awesome than the algebraic form of the Dusty Gas Model equations. Manipulations can be easily carried out.

The linearized procedure can also be extended to the description of the transport of non-ideal fluid mixtures in porous media. This extension will help in the interpretation of experimental data in adsorptive and membrane separations.

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APPENDIX A: NOMENCLATURE

B_0	permeability of the porous medium (m^2)
$[\mathbf{B}]$	matrix of inverted diffusion coefficients defined by eqns. (19) and (20) ($\text{m}^{-2} \text{s}$)
c_i	molar concentration of species i (kmol m^{-3})
c_{n+1}	molar concentration of "dust" species (kmol m^{-3})

c_i	mixture molar concentration (kmol m ⁻³)
d_i	generalized driving force (m ⁻¹)
\mathcal{D}_{ij}	generalized Maxwell–Stefan diffusion coefficients (m ² s ⁻¹)
\mathcal{D}_{ij}^e	effective generalized Maxwell–Stefan diffusion coefficients in porous medium (m ² s ⁻¹)
D_{iK}	Knudsen diffusion coefficient of species i in cylindrical pore (m ² s ⁻¹)
D_{iK}^e	effective Knudsen diffusion coefficient in porous medium (m ² s ⁻¹)
e_i	% deviation between model predictions and experimental values of fluxes
\mathcal{F}	Faraday’s constant (9.65 × 10 ⁷ C (kg equiv) ⁻¹)
F_i	external body force acting on species i (N kmol ⁻¹)
j_i	mass diffusion flux of component i with respect to the mass average mixture velocity (kg m ⁻² s ⁻¹)
M_i	molar mass of species i (kg kmol ⁻¹)
n	number of species in fluid mixture
N_i	molar diffusion flux in fixed coordinate reference frame (kmol m ⁻² s ⁻¹)
p	system pressure (N m ⁻² or atm or mm Hg)
r_0	radius of cylindrical pore (m)
R	gas constant (8314.4 J kmol ⁻¹ K ⁻¹)
t	temperature (°C)
T	absolute temperature (K)
\bar{V}_i	partial molar volume of species i (m ³ kmol ⁻¹)
x_i	mole fraction of species i
z_i	charge on species i

Greek symbols

δ	length of diffusion path (m)
ϵ	porosity of medium

μ_i	molar chemical potential of species i (J kmol ⁻¹)
μ	viscosity of fluid mixture (Pa s)
τ	tortuosity of porous medium
ϕ_i	= $c_i \bar{V}_i$, volume fraction of species i
ϕ	electrostatic potential (V)
ω_i	mass fraction of species i

Subscripts

i, j	referring to species i, j in multi-component mixture
K	Knudsen diffusion coefficient
$n + 1$	“dust” species
0	at position 0 (start of diffusion path)
T	at constant temperature
T, p	at constant temperature and pressure
δ	at position δ (at end of diffusion path)

Superscripts

D	diffusive contribution to the fluxes
e	effective diffusion coefficient in porous medium
V	viscous contribution to the fluxes
$-$	overbar denotes averaged value
\sim	tilde above letter denotes specific quantity

Matrix notation

$[]$	$n \times n$ dimensional matrix
$[]^{-1}$	inverted matrix
$()$	n dimensional column matrix

Vector notation

∇	gradient operator (m ⁻¹)
\cdot	dot product between two vectors

Operators

Δ	difference operator
Σ	summation