

Gas and liquid diffusion representation in the source term $G_{diff,e,n}^P$

Summary:

This article explains the algorithmic calculation procedure for gas and liquid diffusion as it is applied to landfill waste diffusion in the LDAT landfill waste degradation and transport model. In the LDAT constitutive equation, landfill gas and liquid diffusion is treated as a component of the source term in the LDAT unsaturated gas and liquid flow sub-model. This component is evaluated using a 'dusty gas' approach which combines a Knudsen/Fickian diffusion model with a Binary/Dusty Gas diffusion model in the case of gas diffusion. In the case of liquid diffusion just the Fickian diffusion model is used. The Binary/Dusty gas diffusion model is derived from the Stefan-Maxwell equation. An explanation is given of the method of calculating the LDAT constitutive equation diffusion source term. The choice of values for the Fickian diffusion coefficients and the Binary diffusion coefficients for the different waste chemical compounds that are involved is discussed.

Gas and liquid diffusion

LDAT solves the landfill process constitutive equations using a finite difference algorithm within a framework of rectangular representative elementary volumes. The space occupied by the waste material in the landfill is represented by a three dimensional array of these elements. Each element e is surrounded by six neighbouring elements some of which may be boundary elements, Figure 1. The suffix em attached to a variable denotes that the variable is evaluated at the interface between the element and its neighbour m . In the case of vectors it also denotes direction, which is positive in the outward direction normal to the interface. A single suffix e attached to a variable denotes that the variable is evaluated at the centroid of element e .

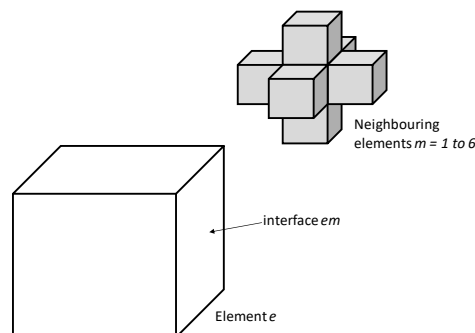


Figure 1 Finite difference three dimensional rectangular element.

Note: Each element has a number, in this case e . There are six interfaces with neighbouring elements. The interfaces are designated by the integer $m = 1$ to 6 . The number of a neighbouring element i is obtained from an array mapping function $i = array_map(e, m)$.

The representation of the impact of the diffusion of gas and liquid chemical compounds in landfill waste is achieved by locating the effect of these in the source term of the LDAT constitutive equation. This equation is derived by assembling the individual equations that represent the

conservation of the mass, $m_{e,n}^P$, of the n^{th} component chemical compound of the waste in phase P (solid, liquid or gas) in the context of a representative elementary volume, V_e as expressed by the following equation,

$$\frac{\partial m_{e,n}^P}{\partial t} = \rho_{e,n}^P G_{e,n}^P V_e - \sum_m \rho_{em,n}^P A_{em,n}^P v_{em,n}^P \quad (1)$$

$m_{e,n}^P = \rho_{e,n}^P z_{e,n}^P V_e$ where $z_{e,n}^P$ is the component compound volumetric concentration or volume fraction (m^3/m^3) and $\rho_{em,n}^P$ is the component density (kg/m^3). For the gas and liquid phases $v_{em,n}^P$ is the interstitial flow velocity (m/day) of component n . $A_{em,n}^P$ is the area associated with the flow or displacement velocity $v_{em,n}^P$. $G_{e,n}^P$ is the source term ($m^3/day/m^3$) associated with component n in phase P , and is composed of four parts, an abstraction/recharge term, a waste degradation term, a gas solubility/phase change term, and a diffusion/dispersion term, as shown by the equation,

$$G_{e,n}^P = G_{abs,e,n}^P + G_{deg,e,n}^P + G_{sol,e,n}^P + G_{diff,e,n}^P \quad (2)$$

Thus in order to calculate the diffusion/dispersion component of the source term, $G_{diff,e,n}^P$, during a solution of the LDAT constitutive equation, it is necessary to be able to estimate the liquid and gas diffusion velocities, $v_{diff,em,n}^P$, across the interfaces of elements. This is because the source term

$G_{diff,e,n}^P$ is given by,

$$\rho_{e,n}^P G_{diff,e,n}^P V_e = - \sum_m \rho_{em,n}^P z_{em,n}^P A_{em}^P v_{diff,em,n}^P \quad (3)$$

The diffusion equation applied in the LDAT code is one derived from the 'dusty gas' equation used by (Moody et al. 1992). For the gas phase this is given below as equation (4).

$$\frac{1}{\rho} \nabla \cdot (\rho z_{em,n}^G / z_{em}^G) = - \frac{z_{em,n}^G v_{diff,em,n}^G}{D_n^k} + \sum_{j \neq n} \frac{z_{em,j}^G z_{em,n}^G / z_{em}^G (v_{diff,em,j}^G - v_{diff,em,n}^G)}{D_{nj}} \quad (4)$$

On the right hand side of equation 4), there are two terms contributing to the concentration gradients of the gas component n , a Knudsen/Fickian term and a Binary/ Dusty Gas diffusion term, the latter being based on the Stefan-Maxwell equation. For the purpose of calculating diffusion velocities in the liquid phase the Stefan-Maxwell term is ignored.

Equation (4) provides n equations that can be solved for the n velocities $v_{diff,em,n}^G$ across interface em . It is convenient to solve for the product

$$u_n = z_{em,n}^G v_{diff,em,n}^G \quad (5)$$

which may then be used directly to obtain the diffusion mass flow in equation (3).

Note that $\rho_{em,n}^G$ and $z_{em,n}^G$ are calculated according to the direction of $v_{diff,em,n}^G$. If +ve outwards then $\rho_{em,n}^G$ is the density of the component in element e . If -ve then $\rho_{em,n}^G$ is the density of the component in the interface element m . $z_{em,n}^G$ is treated similarly.

Writing the diffusion coefficient matrix as $a_n = \frac{1}{\rho} \nabla (\rho z_{em,n}^G / z_{em}^G)$ equation (4) becomes,

$$a_n = -\frac{1}{D_n^k} u_n + \sum_{j \neq n} \frac{z_{em,j}^G z_{em,n}^G / z_{em}^G (v_{diff,em,j}^G - v_{diff,em,n}^G)}{D_{nj}} \quad (6)$$

And further, using equation (5),

$$a_n = -\frac{1}{D_n^k} u_n + \sum_{j \neq n} \frac{z_{em,n}^G}{z_{em}^G D_{nj}} u_j - u_n \sum_{j \neq n} \frac{z_{em,j}^G}{z_{em}^G D_{nj}} \quad (7)$$

Equation (7) is in fact a set of equations, one for each compound, each of which may be designated by 'equation (7n)' etc. It may be noted that if the Knudsen/Fickian diffusion coefficients are the same

this set of equations may be added together to demonstrate that $\sum_{n=0}^N u_n = \sum_{n=0}^N z_{em,n}^G v_{diff,em,n}^G = 0$, which

is the condition that the net volume diffusion flux is zero across an interface. This is because

$$\sum_{n=0}^N a_n = 0 \quad (\text{the sum of the concentration gradients is zero because } \sum_{n=0}^N z_{em,n}^G / z_{em}^G = 1), \text{ and the } j^{\text{th}}$$

term in the sum $\sum_{j \neq n} \frac{z_{em,n}^G}{z_{em}^G D_{nj}} u_j$ in equation (7n) cancelled out by the n th term in the sum

$$-u_j \sum_{i \neq n} \frac{z_{em,i}^G}{z_{em}^G D_{ji}}$$

in equation (7j). Note that all components of both gas and liquid phases should be involved in the diffusion process in order to provide the condition that the net volume diffusion flux is zero across an interface. This means that a Knudsen/Fickian diffusion coefficient should be specified for all fluid components, and a binary diffusion coefficient should be specified for all gas components.

Equation (7) may be represented as a matrix $[P_{nj}][u_j] = [a_n]$.

$$P_{nj} = \frac{z_{em,n}^G}{z_{em}^G D_{nj}} \quad j \neq n \quad (8a)$$

And

$$P_{nn} = -\frac{1}{D_n^k} - \sum_{j \neq n} \frac{z_{em,j}^G}{z_{em}^G D_{nj}} \quad (8b)$$

The ratios $\frac{z_{em,n}^G}{z_{em}^G}$ should be the average of the ratios on either side of the interface em . In LDAT this average is calculated as the geometric mean of the ratios as this makes the average zero if one or the other is zero.

In the case of the liquid phase the Stephan-Maxwell terms are omitted so that $P_{nj} = 0$ ($j \neq n$) and

$$P_{nn} = -\frac{1}{D_n^k}.$$

(Reinecke 2000) gives a summary of the role of Knudsen diffusivity and explains how a reasonable estimate of the value of D_n^k can be obtained from the equations,

$$D_n^k = \frac{bK^I}{\mu^{L/G}} \text{ and } b = 0.11(K^I)^{-0.39} \quad (9)$$

where K^I is the intrinsic permeability of the porous material in m^2 and b is the Klinkenberg parameter in Pa. For example for values of $K^I = 10^{-14} m^2$ and $\mu^G = 1.8 \times 10^{-5} \text{ kg}/(\text{m}\cdot\text{sec})$ or Pa.sec, $b = 3.17 \times 10^4 \text{ Pa}$, $D_n^k = 1.76 \times 10^{-5} m^2/\text{sec} = 1.52 m^2/\text{day}$.

Furthermore since $K^I = K^L \frac{1}{g} \frac{\mu^L}{\rho^L} = K^G \frac{1}{g} \frac{\mu^G}{\rho^G}$ equation (9) may be expressed as,

$$D_n^k = 0.11 \left(\frac{K^{L/G}}{g\rho^{L/G}} \right)^{0.61} \mu^{L/G-0.39} \quad (9)$$

Some values of binary and Fickian diffusion coefficients are given in Table 2 below. The binary diffusion coefficient values are based on those provided by (Ghabaee and Rodwell 1989).

Compound pair		Binary diffusion coefficient		Dissolved gas	Fickian diffusion coefficient
N ₂	O ₂	1.80		CO ₂	1.66E-04
N ₂	CH ₄	1.85		CH ₄	1.29E-04
N ₂	CH ₄	1.42		H ₂	3.89E-04
O ₂	CH ₄	1.96		H ₂ S	1.22E-04
O ₂	CO ₂	1.41		N ₂	1.62E-04
CH ₄	CO ₂	1.47		NH ₃	1.42E-04
				O ₂	1.81E-04

Table 2 Binary and Fickian diffusion coefficients m²/day

For other compounds default values are used for the binary and Fickian coefficients of 1.5 m²/day and 1.5 x 10⁻⁴ m²/day respectively.

These reference values are adjusted for temperature and viscosity using the equation,

$$D = D_{REF} \frac{T}{T_{REF}} \frac{\mu_{REF}}{\mu}$$

To allow for the variable moisture content in the pore space all diffusion coefficients are multiplied by the relative permeability of the relevant phase.

Ghabaee, K. and Rodwell, W.R. (1989) Landfill gas modelling. A literature survey of landfill gas generation and migration. United Kingdom Atomic Energy Authority (UKAEA), Petroleum Reservoir Technology Division, Winfrith Technology Centre, AEEW - R2567, October 1989.

Moody, R.O., Rodwell, W.R. and Ghabaee, K. (1992) The numerical simulation of the migration of gas produced in landfills. United Kingdom Atomic Energy Authority (UKAEA), AEA Petroleum Services, Winfrith, AEA - APS - 0203, January 1992.

Reinecke, S.A. (2000) The effect of water content on Knudsen diffusion in unconsolidated porous media University of Toronto.