A semi-analytical model for landfill gas migration through finite fractured unsaturated landfill cover soil

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ABSTRACT: Landfill gas random diffusion causes serious greenhouse effect on atmospheric environment. The clay cover in landfill is the main barrier to control the methane emission to the atmosphere. Cracks develop when the clayey soil is affected by the wet and dry cycle and the uneven settlement of waste. This paper presents a semi-analytical solution to one-dimensional advection-diffusion of landfill gas transport in a finite unsaturated fractured landfill cover. Influences of fracture widths, methane diffusion coefficient, oxidation rate and degree of saturation on transport of methane in the unsaturated clay cover were analyzed based on the proposed solution. When the fracture width increased from 1mm to 10mm, the surface relative concentration and flux increased about two orders of magnitude. When the first-order oxidation coefficient of methane increased from 10^{-6} s^{-1} to 10^{-5} s^{-1}, the concentration was reduced by a factor of 2, while the surface flux is reduced by 90%. When the degree of saturation decreased from 0.9 to 0.1, the concentration and surface flux of fracture with 10 mm width is reduced by more than 50%.

1 INTRODUCTION

Municipal solid waste landfill (MSW) can be considered as a large-scale biochemical reactor in the long-term landfill process. Due to compaction, rainfall and microbial decomposition, and other biochemical reactions, MSW will produce plenty of secondary pollutants such as landfill gas and leachate (Arigala et al. 1995). Therefore, in order to mitigate landfill gas and other harmful substances affecting the surrounding environment, all kinds of landfill closure system should include waste body, bottom lining system, leachate collection and discharge system, gas control and cover system (Didier et al. 2000).

Methane is the main component of landfill gas, and its disorderly diffusion has a serious greenhouse effect on the atmospheric environment (Li et al. 2016). Though the clay liner in landfill is a major barrier to the diffusion of methane into the atmosphere, due to the seasonal dry and wet cycle, freeze-thaw cycle, landfill and the upper part of the soil layer's temperature difference and uneven settlement, plant root transpiration and other reasons, the compacted clay cover is prone to cracks. Miller and Mishra (1989) have found cracks in the landfill cover with a width of more than 10 mm and a depth of more than 30 mm. With the growth of fractures under the influence of wet and dry cycles, the permeability coefficient of the compacted clay layer also increases linearly (Lu et al. 2016). The dry and wet circulation can lead to the dry cracking of the clay layer (Li et al. 2011) and the deformation of the soil skeleton (Xue et al. 2014a). Xue et al. (2014b) found that the incorporation of aspartic fibers in compacted clay can effectively reduce the emergence of cracks in the cover soil. Czepiel et al. (1996) found that about half of the methane would overflow through these cracks if there were cracks in the landfill cover.

Meanwhile, the parameters in the landfill cover soil such as the diffusion coefficient of methane in the compacted clay liner (CCL) and the first-order oxidation coefficient in the cover soil have a significant effect on the concentration and flux of the cover surface. Wickramarachchi et al. (2011) found that the diffusion ratio of soil and gas of methane in different water content was most affected by soil dry density and particle size.

There have been many previous studies on the release of methane from buildings to the atmosphere in the CCL (Yao et al. 2013). Christopher (2008) assessed the gas-phase transfer processes in landfill with respect to their influence on waste biodegradation. Poulsen et al. (2001) set up the model to study the lateral gas transport in soil adjacent to old landfill. He et al. (2011) worked on the factors on methane oxidation in the landfill cover. Zhan et al. (2017) explored the law of water-gas coupling and migration in the overburden through soil column test and numerical simulation. However, up to now, the migration of methane has not been studied in the presence of fractures in the CCL. In this paper, a dual medium transport model of methane in fractured clay covers has been developed and the semi-analytical solution of the model has been obtained by using Laplace inversion of Stehfest method. On the basis of the proposed solution, the effects of the fracture opening width, the methane permeability coefficient in the CCL, the methane oxidation rate and other parameters on the concentration and flux of methane on the CCL surface were investigated. The solution is relatively simple, but can provide an effective method for the fitting of test data and risk assessment.

2 MATHEMATICAL MODEL

The movement of methane in the compacted clay liner can be divided into two processes: movement of methane in the fracture and the migration of methane in CCL matrix. The two processes are coupled, and the two equations are established by using the relationship between the gas flux and the gas concentration ratio at the boundary of the fracture. The model was developed under the following assumptions:

(1)The diffusion of gas in the cracks along the width of the crack and the mechanical dispersion are filled with cracks.

(2)The permeation of gas in the compacted clay liner skeleton is not significant, taking into account only the molecular diffusion of the gas in the soil.

(3)The transport rate of the gas in the fracture is much greater than its rate of transport in the compacted clay.

Therefore, the movement of methane takes into account the following effects: advection in fractures, molecular diffusion along fractures, molecular diffusion from fractures to porous CCL, and methane oxidation in the cover soil, as shown in Figure 1.

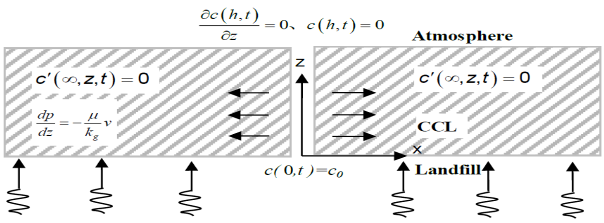

Figure 1. Diagram of gas transport in fractured clay cover

Due to the existence of pores or fractures in the cover soil of a different degree, making the oxygen in the atmosphere enter into the soil and react with the methane (Ng et al. 2015). The governing equation in fracture is (Tang et al., 1981)

$$\frac{\partial c}{\partial t}+v\frac{\partial c}{\partial z}-D\frac{\partial^2 c}{\partial z^2}+\lambda c+\frac{q}{b}=0 \tag{1}$$

The governing equation in matrix is

$$\frac{\partial c'}{\partial t}-D'\frac{\partial^2 c'}{\partial x^2}+\lambda c'=0 \tag{2}$$

where z is the vertical distance from the bottom of CCL, t is time, c is the concentration of methane in the fracture, c' is the concentration of methane in the CCL, $2b$ is the crack width, v is the Darcy flow velocity of methane moving along the fracture direction, q is the methane diffusion flux perpendicular to the fracture, D is the diffusion coefficient of methane in the air, D' is the diffusion coefficient of methane in the matrix, and λ is the first order oxidation reaction rate of methane.

Since the gas flux at the interface between the fracture and the clay cover is equal, the above two equilibrium equations are related as follows:

$$q=-\theta D'\left.\frac{\partial c'}{\partial x}\right|_{x=b} \tag{3}$$

where θ_g is the volume gas content of the CCL, and q is the gas flux at the interface of the fracture and clay cover. In fact, the compacted clay cover in this paper is a porosity-fracture dual media in which pores and cracks are interconnected and have different porosities. In general, the porosity of the fracture media is less than the porosity of the porous media (Garg & Achari 2010), and the dual-media model usually considers the whole system as a continuous or equivalent continuous medium for the mathematical modeling (Hantush & Govindaraju 2003).

The boundary conditions for the fractures are

$$c(0,t)=c_0 \tag{4a}$$

$$\frac{\partial c(h,t)}{\partial z}=0,\ c(h,t)=0 \qquad \text{(4b)、(4c)}$$

where c_0 is the initial concentration in landfill.

The initial conditions of the governing equation (1) are

$$c(z,0)=0 \tag{4d}$$

The boundary conditions of the governing equation (2) are

$$c'(b,z,t)=c(z,t) \tag{5a}$$

$$c'(\infty,z,t)=0 \tag{5b}$$

The initial conditions of the governing equation (2) are

$$c'(x,z,0)=0 \tag{5c}$$

The flow rate of methane in the fractures can be determined by the following equation (Gary & Achari 2010):

$$\frac{dp}{dz}=-\frac{\mu}{k_g}v \tag{6}$$

where dp/dx is the pressure gradient in fracture, μ is the dynamic viscosity coefficient, and k_g is the permeability coefficient of methane.

Assuming that the methane oxidation satisfies the first-order kinetic model proposed by Koschorreck and Conrad (1993), the methane first order oxidation constant can also be converted to the methane oxidation rate R_{CH4}, both of which have the following relationship:

$$R_{CH_4}=\int_0^{0.6}\lambda czdz \tag{7}$$

Since the first-order oxidation rate of methane is a function of concentration and depth, here the average value of the rate of methane at the top and the bottom of the CCL is used in the following calculation.

The diffusion coefficient D_m of methane in CCL is given by (Moldrup et al. 2005):

$$\frac{D_m}{D_0}=\frac{\theta_g^{10/3}}{n^2}=n^{\frac{4}{3}}\left(1-S_r\right)^{\frac{10}{3}} \tag{8}$$

where D_0 is the diffusion coefficient of methane in the atmosphere, θ_g is the volume moisture content of the soil, n is the porosity of the soil, and S_r is the degree of saturation of the soil.

3 DERIVATION OF SEMI-ANALYTICAL SOLUTION

Transfer equation (2) into Laplace transform,

$$p\overline{c}'=D'\frac{d^2\overline{c}'}{dx}-\lambda\overline{c}' \tag{9}$$

And the solution of (9) is

$$\overline{c}'=\overline{c}_1\exp\left[-(P/D')^{1/2}\left(x-b\right)\right] \tag{10}$$

where $P=p+\lambda$.

$c_1{}'$ can be obtained by (5a):

$$\overline{c}'=\overline{c}\exp\left[-(P/D')^{1/2}\left(x-b\right)\right] \tag{11}$$

Then

$$\left.\frac{d\overline{c}'}{dx}\right|_{x=b}=-(P/D')^{1/2}\overline{c} \tag{12}$$

Substitute equation (3) into the Laplace transform of equation (1):

$$p\overline{c}+v\frac{d\overline{c}}{dz}+\lambda\overline{c}=\frac{\theta D'}{b}\left.\frac{d\overline{c}'}{dx}\right|_{x=b}+D\frac{d^2\overline{c}}{dz^2} \tag{13}$$

Substitute (12) into (13)

$$\frac{d^2\overline{c}}{dz^2}-\frac{v}{D}\frac{d\overline{c}}{dz}-\frac{1}{D}\left(P+\frac{P^{1/2}}{A}\right)\overline{c}=0 \tag{14}$$

where $A=b\big/\left(\theta\sqrt{D'}\right)$.

The solution form of (14) is

$$\overline{c}=c_2\exp\left(zr_1\right)+c_3\exp\left(zr_2\right) \tag{15}$$

$$r_{1,2}=\frac{v}{2D}\cdot\left\{1\pm\left[1+\frac{4D}{v^2}\left(\frac{P^{1/2}}{A}+P\right)\right]^{1/2}\right\} \tag{16}$$

where $\gamma=v/2D$, $\beta^2=4D/v^2$.

Use (4a) and (4b) to get the undetermined coefficient c_2 and c_3, take the Laplace transform of (4a) and (4b),

$$\overline{c}\left(0,p\right)=\frac{c_0}{P-\lambda} \tag{17}$$

$$\frac{\partial c\left(h,t\right)}{\partial z}=0 \tag{18}$$

From that we can get

$$\frac{c_0}{P-\lambda}=c_2+c_3 \tag{19}$$

$$c_{2,1}\exp(zr_1)\cdot r_1+c_{3,1}\exp(zr_2)\cdot r_2\big|_{z=h}=0 \tag{20}$$

The above formula can be solved as

$$c_2=\frac{c_0}{P\text{-}\lambda}-\frac{c_0}{P\text{-}\lambda}\cdot\frac{1}{1-\exp\left[-\frac{0.6v}{D}\cdot\sqrt{1+\frac{4D}{v^2}(\frac{\sqrt{P}}{A}+P)}\right]\cdot\frac{1-\sqrt{1+\frac{4D}{v^2}(\frac{\sqrt{P}}{A}+P)}}{1+\sqrt{1+\frac{4D}{v^2}(\frac{\sqrt{P}}{A}+P)}}} \tag{21}$$

$$c_3=\frac{c_0}{P\text{-}\lambda}\cdot\frac{1}{1-\exp\left[-\frac{0.6v}{D}\cdot\sqrt{1+\frac{4D}{v^2}(\frac{\sqrt{P}}{A}+P)}\right]\cdot\frac{1-\sqrt{1+\frac{4D}{v^2}(\frac{\sqrt{P}}{A}+P)}}{1+\sqrt{1+\frac{4D}{v^2}(\frac{\sqrt{P}}{A}+P)}}} \tag{22}$$

The same method can be used to solve the equations (4a) and (4c), and the methane flux J at the fractures at the top of the cover soil can be determined by

$$J=\frac{\overline{c}}{c_0}\cdot v-D\cdot c_0\cdot\frac{\partial\overline{c}}{\partial z} \tag{23}$$

$$\frac{\partial\overline{c}}{\partial z}=\frac{\exp(zr_1)}{p-p\cdot\exp\left[\frac{vh}{D}\cdot\sqrt{1+\frac{4D}{v^2}\left(\frac{\sqrt{P}}{A}+P\right)}\right]}\cdot r_1-\frac{\exp\left[\frac{vh}{D}\cdot\sqrt{1+\frac{4D}{v^2}\left(\frac{\sqrt{P}}{A}+P\right)}\right]}{p-p\cdot\exp\left[\frac{vh}{D}\cdot\sqrt{1+\frac{4D}{v^2}\left(\frac{\sqrt{P}}{A}+P\right)}\right]}\cdot\exp\left(zr_2\right)\cdot r_2 \tag{24}$$

In this paper, the Laplace inversion of the image function of methane concentration at the surface of the CCL was carried out by adopting the Stehfest's numerical inversion method (Stehfest 1970).

4 PARAMETER SENSITIVITY ANALYSIS

In order to study the influence of relevant parameters on the migration of methane in fractured clays, the sensitivity of methane emission flux to the fracture width, diffusion coefficient of methane in the cover soil and the first order oxidation coefficient of methane were analyzed. Yao et al. (2015) found in experiments that the diffusion coefficient of methane in a two-phase gas mixed with oxygen is $2.263\times10^{-5}m^2/s$. At the same time, according to equation (6), it can be calculated that the migration rate of methane in the fracture of clay cover is between 10^{-5} m/s and 10^{-4} m/s. Therefore, the migration rate of methane used below is taken as the average of the two above values, and porosity of the compacted clay cover is 0.4 (Poulsen et al. 2001). In addition, they also obtained the first-order methane oxidation coefficient ranging from $10^{-6}s^{-1}$ to $10^{-5}s^{-1}$ in experiments. The diffusion coefficient of methane in the clay cover lies between $2.13\times10^{-6}m^2/s$ and $6.27\times10^{-6}m^2/s$. The degree of saturation of compacted clay is 0.1 to 0.9. According to equation (9), the range of diffusion coefficient is from $3.83\times10^{-9}m^2/s$ to $5.81\times10^{-6}m^2/s$. In summary, the average of the upper and lower limits $3.14\times10^{-6}m^2/s$ is taken as the diffusion coefficient of methane in the CCL used below.

4.1 *Effect of fracture width*

Camp et al. (2010) obtained a clay fracture width of 2.1 mm to 4.5 mm by a four-point bending experiment. Gourc et al. (2010) investigated the deformation behavior of a clay cap barrier of waste containment system for storing hazardous waste, finding 10mm cracks existing in the landfill cover. Therefore, we selected 1mm, 5mm and 10mm to study the effect of methane migration in fractured clay cover. The diffusion coefficient of methane in the CCL and the first order oxidation coefficient of methane are taken as the average of the data obtained by Yao et al.(2015), respectively, $D'=4.2\times10^{-6}m^2/s$, $\lambda=0.5\times10^{-6}s^{-1}$.

It can be seen from Figure 3 that the methane concentration at the top of the fracture will increase with the development of the fracture opening size. When the fracture width increases from 1 mm to 10 mm, the surface concentration ratio will increase from 0.003 to 0.279. Figure 4 shows the change in methane flux at the fracture of the clay cover surface as time changes when the width of the fracture openings is 1 mm, 5 mm and 10 mm respectively. It can also be seen from Figure 4 that the opening size of the fracture will not have a great impact on the balance of the methane flux, basically reaching the balance state in 30 days. Therefore, the following calculation will continue to the 30^{th} day. However, the fracture width has a greater effect on the final balance value. The total flux of methane produced after 30 days of a 10mm fracture is 10399.18 mol/m^{2}d, which is 44 times of the case with $2b$ = 1mm.

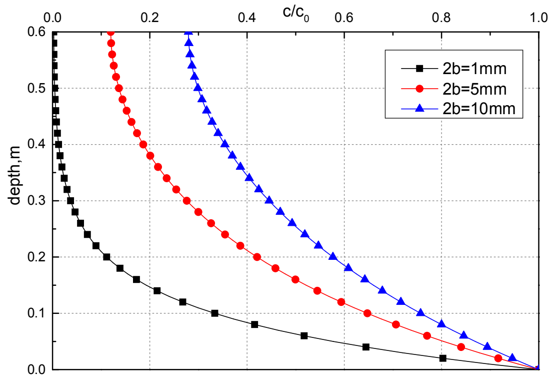

Figure 3. Concentration profiles of methane in fracture for different widths of fractures

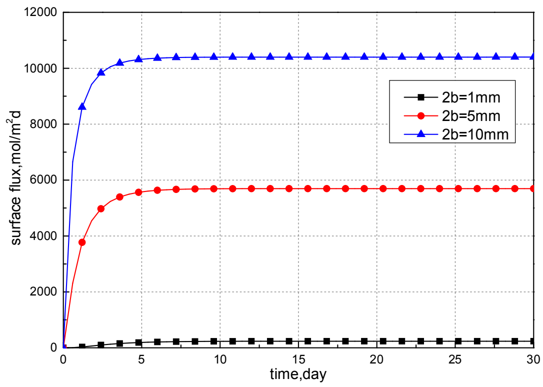

Figure 4. Methane flux at fracture surface vs time for different widths of fractures

4.2 *Effect of first-order oxidation coefficient*

The first order oxidation coefficient of methane is assumed to be $0.5\times10^{-6}s^{-1}$ to $5\times10^{-6}s^{-1}$ (Yao et al. 2015). The remaining parameters are shown in the figure.

Figure 5 shows the relation between the concentration at the fractures of different widths and the first order oxidation coefficient of methane. When λ increased from $10^{-6}s^{-1}$ to $10^{-5}s^{-1}$, the concentration ratio of 1mm crack decreased from 0.046 to 0.003, reducing to 6.5% the original. When the opening size became wider, the concentration ratio reduced from 0.617 to 0.293.

It can be seen from Figure 6 that the rate of flux accumulation is relatively slow in the 1mm fracture. When the first order oxidation coefficient of methane increased from $10^{-6}s^{-1}$ to $10^{-5}s^{-1}$, the surface flux decreased from 2847.87 mol/m^{2}d to 272.11 mol/m^{2}d, which is reduced by about 90.4%. When the crack width increased to 10 mm, the process of flux accumulation became more rapid and the equilibrium flux of $10^{-6}s^{-1}$ methane is 17130.29 mol/m^{2}d, while the faster oxidation one is only 10734.41 mol/m^2. Therefore, the wider the fracture is, the less influence of the change of oxidation coefficient has on the flux.

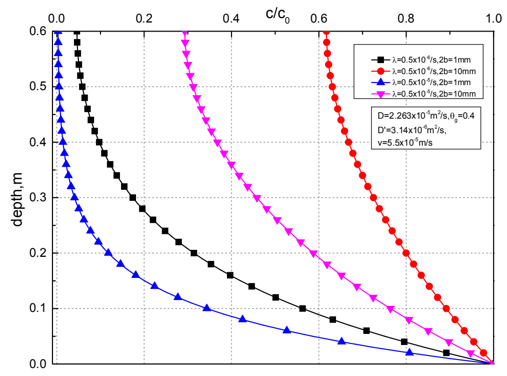

Figure 5. Concentration profiles of methane in fracture with different first order oxidation coefficients（t=30d）

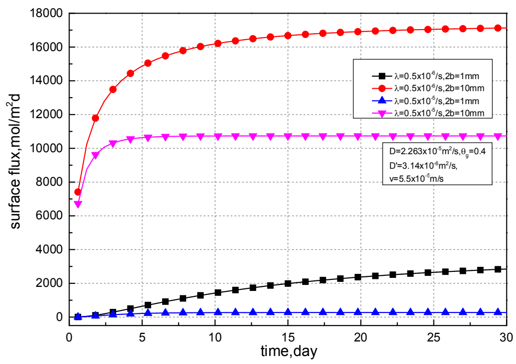

Figure 6. Methane flux at fracture surface vs time with different first order oxidation coefficients

4.3 *Effect of degree of saturation of CCL*

The degree of saturation of the compacted clay cover will affect the diffusion coefficient of methane in the soil through the formula (9). The following parameter values were assumed: S_r=0.1, 0.9 and D_0=2.8×10^{-5}m^2/s (Aubertin et al., 2000). θ_g and D' can be obtained according to the equation (9), and the results were shown in Figure 7.

It is easy to find in Figure 7 that the relationship between the concentration ratio at the top of the fractures in different widths and its saturation. When S_r was reduced from 0.9 to 0.1, the diffusion coefficient of methane in the soil increased from 3.83×10^{-9}m^2/s to 5.81×10^{-6}m^2/s accordingly. The relative concentration of 1mm fracture decreased from 0.579 to 0.001 and the concentration of 10mm decreased from 0.917 to 0.200. This is due to the reduction of lateral migration of gas as it passes through the soil skeleton. Even under the same amplitude increment on the degree of saturation of the CCL, methane concentration ratio will show the different attitude on account of the fracture width. That is, the wider the crack is, the less influence saturation has on the methane concentration.

It can be seen from Figure 8 when the S_r was reduced from 0.9 to 0.1, the surface flux of methane in the fracture with width of 1mm changed from 16470.61mol/m^{2}d to 82.10mol/m^{2}d. When the crack width was up to 10mm, the surface flux decreased from 21173.43mol/m^{2}d to 8271.75mol/m^{2}d. Therefore, the rate of change of surface flux decreases as fracture widen increases even under the same degree of increase of soil saturation.

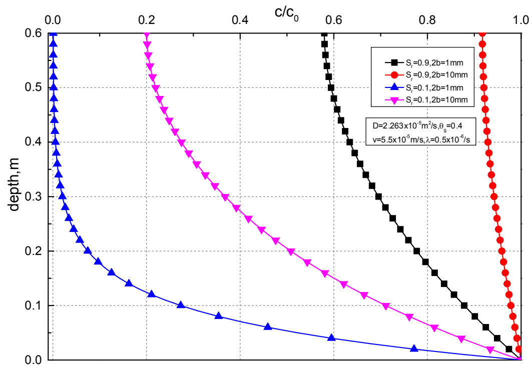

Figure 7. Concentration profiles of methane in fracture with different degree of saturation（t=30d）

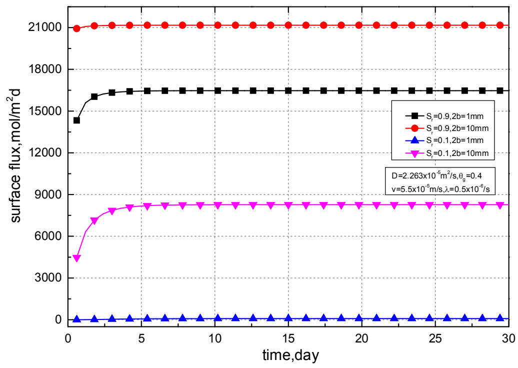

Figure 8. Methane flux at fracture surface vs time with different degree of saturation

5 CONCLUSIONS

In this paper, the semi-analytical solution of methane in the fracture of compacted clay liner is developed on the basis of the method of Laplace transform. The factors affecting methane concentration and flux were discussed, and the following conclusions are drawn:

(1) When the fracture width increases from 1mm to 10mm, the surface concentration can increase by two orders of magnitude under the same situation of oxidation coefficient and diffusion coefficient.

(2) The crack width does not affect the equilibrium time of the methane flux much, but do have a more pronounced effect on the steady-state total flux.

(3) The first-order methane oxidation has a slighter

effect on its migration in the fractured clay cover in the case of the large fractures.

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