A RELEASE MODEL CONSIDERING CHEMICAL LOSS FROM A DOUBLE-LAYER MATERIAL INTO FOOD

by

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The migration of chemicals from packaging materials into food is predictable by various mathematical models. However, the loss of chemicals makes the predictions more complicated. In this article, a mathematical model considering chemical instability is developed to quantify the release of chemicals through double-layer packaging films based on Fick's diffusion and first order reaction. At the same time, two different loading modes are considered in the loss function. The release model is solved numerically to elucidate the effects of diffusivity value, distribution of chemical and mass transfer at the interface of material and food on the migration process, and the loss of the chemicals in food is also elucidated.

Key words: *model, migration, numerical simulation, chemical instability*

Introduction

Food safety refers not only to the safety of food itself, but also to the safety of food packaging materials. The migration of chemicals in food packaging materials has attracted extensive attention and in-depth research from food packaging enterprises and scholars [1-9]. The ability to predict the migration is critical to assess the hazards. However, the current predictive models have some limitations, because the instability of migrants in food are not considered [10-13]. Experiments show that the migrants in packaging materials are unstable in some foods, such as hydrolysis and thermal degradation [14-17]. Instability affects the reliability and accuracy of migration model prediction.

The objective of this paper is to develop a migration model for two-layer film consisting of regenerative layer and original layer. In this model, one direction migration, the same diffusion coefficient, partition coefficient between material and food, mass transfer coefficient between material and food, most importantly, chemical instability and different loading ways of loss function, are considered. A series of equations are developed based on Fick’s diffusion and first order reaction, and solved numerically by compact difference scheme from the finite difference method.

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Diffusion modeling and assumptions

Assumptions

Packaging materials and food volume are limited in finite packaging-finite food migration modeling, and the concentration of chemicals in food varies with time, and the mass transfer resistance and the distribution at the packaging-food interface are also considered.

The assumptions of this model are: two layers are in perfect contact, migrant is from the regenerative layer to food, fig. 1. At the initial stage, the initial concentration of migrant is uniform in the regenerative layer, the migration is in one direction, *i. e.* from the regenerative layer into food, there is no migration from the packaging into air, there is no migration of food into the package, and there is a finite coefficient of mass transfer, h_m, from original layer to food, the migration in each layer follows Fick's diffusion, and diffusivities of migrant in each layer are constant, the partition coefficient k_{PF} (ratio of chemical concentration in packaging material to chemical concentration in food at balance) of a migrant is constant at the interface of original layer and food.

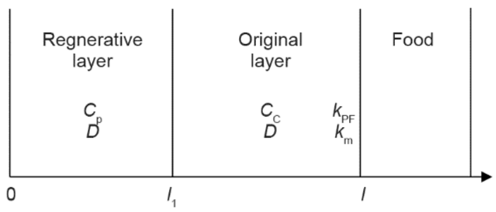

Figure 1. Scheme of model system

Model and difference scheme

A migration prediction model is established for the migration of contaminants in double layer packaging materials through the original layer as functional barrier layer into the limited volume food. The thickness of the barrier layer is $l_2 = l - l_1$.

The migration of chemicals from material into food can be described by the following diffusion equation.

Let $C_P = u$, $C_C = v$, we have the following governing equations:

$$\frac{\partial u}{\partial t} = D\frac{\partial^2 u}{\partial x^2}, \quad 0 < x < l_1 \tag{1}$$

$$\frac{\partial v}{\partial t} = D\frac{\partial^2 v}{\partial x^2}, \quad l_1 < x < l \tag{2}$$

Initial conditions ($t = 0$):

$$u(x,0) = C_{\text{in}}, \quad 0 < x < l_1, \tag{3}$$

$$v(x,0) = 0, \quad l_1 < x < l, \tag{4}$$

Boundary conditions ($t > 0$):

$$\frac{\partial u}{\partial x} = 0, \quad x = 0 \tag{5}$$

$$D\frac{\partial u}{\partial t} = D\frac{\partial v}{\partial t}, \quad x = l_1 \tag{6}$$

$$v(l_1,t)=u(l_1,t), \quad x=l_1 \tag{7}$$

$$-D\frac{\partial v}{\partial x}|_{x=l}=h_{\mathrm{m}}[v(l,t)-k_{\mathrm{PF}}C_{\mathrm{F}}], \quad x=l \tag{8}$$

The packaging film is divided into a uniform mesh in the direction along film thickness, fig. 2, in which 1 is divided into M parts (space step is h), time step is τ.

At the initial time, the chemicals are evenly distributed in the regenerated layer, the compact difference scheme is:

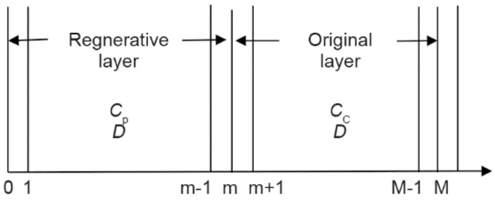

Figure 2. Scheme of model meshing of limited packaging-limited food

$$\frac{1}{12}\left(\frac{u_{i+1}^{k}-u_{i+1}^{k-1}}{\tau}+10\frac{u_{i}^{k}-u_{i}^{k-1}}{\tau}+\frac{u_{i-1}^{k}-u_{i-1}^{k-1}}{\tau}\right)=\frac{D}{h^2}\left(u_{i-1}^{k-\frac{1}{2}}-2u_{i}^{k-\frac{1}{2}}+u_{i+1}^{k-\frac{1}{2}}\right) \tag{9}$$

$$i=1,2,...,m-1; \quad k=1,2,...,nt$$

$$\frac{1}{12}\left(\frac{v_{j+1}^{k}-v_{j+1}^{k-1}}{\tau}+10\frac{v_{j}^{k}-v_{j}^{k-1}}{\tau}+\frac{v_{j-1}^{k}-v_{j-1}^{k-1}}{\tau}\right)=\frac{D}{h^2}\left(v_{j-1}^{k-\frac{1}{2}}-2v_{j}^{k-\frac{1}{2}}+v_{j+1}^{k-\frac{1}{2}}\right) \tag{10}$$

$$j=m+1,m+2,...,M-1; \quad k=1,2,...,nt$$

Initial conditions (t = 0):

$$u_i^0=C_{\mathrm{in}}, \quad 0\le i\le m \tag{11}$$

$$v_j^0=0, \quad m\le j\le M \tag{12}$$

Boundary conditions (t > 0):

x = 0:

$$\left(\frac{5}{12}+\frac{D\tau}{2h^2}\right)u_0^k+\left(\frac{1}{12}-\frac{D\tau}{2h^2}\right)u_1^k=\left(\frac{5}{12}-\frac{D\tau}{2h^2}\right)u_0^{k-1}+\left(\frac{1}{12}+\frac{D\tau}{2h^2}\right)u_1^{k-1}, \quad 1\le k\le nt \tag{13}$$

x = l_1:

$$\frac{1}{12}\frac{u_{m-1}^{k}-u_{m-1}^{k-1}}{\tau}+\frac{5}{12}\frac{u_{m}^{k}-u_{m}^{k-1}}{\tau}+\frac{5}{12}\frac{v_{m}^{k}-v_{m}^{k-1}}{\tau}+\frac{1}{12}\frac{v_{m+1}^{k}-v_{m+1}^{k-1}}{\tau}=$$

$$=\frac{D}{h^2}(v_{m+1}^{k}+v_{m+1}^{k-1}-v_{m}^{k}-v_{m}^{k-1})-\frac{D}{h^2}(u_{m}^{k}+u_{m}^{k-1}-u_{m-1}^{k}-u_{m-1}^{k-1}), \quad 1\le k\le nt \tag{14}$$

$$v_m^{k-\frac{1}{2}}=u_m^{k-\frac{1}{2}}, \quad 1\le k\le nt \tag{15}$$

x = l:

$$\frac{5}{12}\frac{\partial v_M^{k-1/2}}{\partial t}+\frac{1}{12}\frac{\partial v_{M-1}^{k-1/2}}{\partial t}=D\frac{\dfrac{\partial v_M^{k-1/2}}{\partial x}-\dfrac{\partial v_{M-1/2}^{k-1/2}}{\partial x}}{\dfrac{h}{2}}, \quad 1\le k\le nt \tag{16}$$

$$C_{\mathrm{F}}^{k-\frac{1}{2}}V_{\mathrm{F}} + M_{\mathrm{P}}^{k-\frac{1}{2}} + M_{\mathrm{C}}^{k-\frac{1}{2}} = C_{\mathrm{in}}V_{\mathrm{P}}, \quad 1 \le i \le m-1 \tag{17}$$

Then have the following equations:

$$\left(\frac{1}{12}-\frac{D\tau}{2h^2}\right)u_{i+1}^k + \left(\frac{5}{6}+\frac{D\tau}{h^2}\right)u_i^k + \left(\frac{1}{12}-\frac{D\tau}{2h^2}\right)u_{i-1}^k =$$

$$= \left(\frac{1}{12}+\frac{D\tau}{2h^2}\right)u_{i+1}^{k-1} + \left(\frac{5}{6}-\frac{D\tau}{h^2}\right)u_i^{k-1} + \left(\frac{1}{12}+\frac{D\tau}{2h^2}\right)u_{i-1}^{k-1}, \quad 1 \le i \le m-1, \quad 1 \le k \le nt \tag{18}$$

$$\left(\frac{1}{12}-\frac{D\tau}{2h^2}\right)v_{j+1}^k + \left(\frac{5}{6}+\frac{D\tau}{h^2}\right)v_j^k + \left(\frac{1}{12}-\frac{D\tau}{2h^2}\right)v_{j-1}^k =$$

$$= \left(\frac{1}{12}+\frac{D\tau}{2h^2}\right)v_{j+1}^{k-1} + \left(\frac{5}{6}-\frac{D\tau}{h^2}\right)v_j^{k-1} + \left(\frac{1}{12}+\frac{D\tau}{2h^2}\right)v_{j-1}^{k-1}, \quad m+1 \le j < M-1, \quad 1 \le k \le nt \tag{19}$$

The initial conditions ($t = 0$):

$$u_i^0 = C_{\mathrm{in}}, \quad 0 \le x \le m \tag{20}$$

$$v_j^0 = 0, \quad m \le j \le M \tag{21}$$

Boundary conditions ($t > 0$):

$x = 0$:

$$\left(\frac{5}{12}+\frac{D\tau}{2h^2}\right)u_0^k + \left(\frac{1}{12}-\frac{D\tau}{2h^2}\right)u_1^k = \left(\frac{5}{12}-\frac{D\tau}{2h^2}\right)u_0^{k-1} + \left(\frac{1}{12}+\frac{D\tau}{2h^2}\right)u_1^{k-1}, \quad 1 \le k \le nt \tag{22}$$

$x = l$:

$$\left(\frac{1}{12\tau}-\frac{D}{h^2}\right)u_{m-1}^k + \left(\frac{5}{12\tau}+\frac{D}{h^2}\right)u_m^k + \left(\frac{5}{12\tau}+\frac{D}{h^2}\right)v_m^k + \left(\frac{1}{12\tau}-\frac{D}{h^2}\right)v_{m+1}^k =$$

$$= \left(\frac{1}{12\tau}+\frac{D}{h^2}\right)u_{m-1}^{k-1} + \left(\frac{5}{12\tau}-\frac{D}{h^2}\right)u_m^{k-1} + \left(\frac{5}{12\tau}-\frac{D}{h^2}\right)v_m^{k-1} + \left(\frac{1}{12\tau}+\frac{D}{h^2}\right)v_{m+1}^{k-1}, \quad 1 \le k \le nt \tag{23}$$

$$v_m^k = u_m^k, \quad 1 \le k \le nt \tag{24}$$

$x = l$:

$$\left(\frac{1}{12}-\frac{D\tau}{h^2}\right)v_{M-1}^k + \left(\frac{5}{12}+\frac{D\tau}{h^2}+\frac{h_{\mathrm{m}}\tau}{h}\right)v_M^k - \frac{h_{\mathrm{m}}\tau}{h}k_{\mathrm{PF}}C_{\mathrm{F}}^k =$$

$$= \left(\frac{1}{12}+\frac{D\tau}{h^2}\right)v_{M-1}^{k-1} + \left(\frac{5}{12}-\frac{D\tau}{h^2}-\frac{h_{\mathrm{m}}\tau}{h}\right)v_M^{k-1} + \frac{h_{\mathrm{m}}\tau}{h}k_{\mathrm{PF}}C_{\mathrm{F}}^{k-1}, \quad 1 \le k \le nt \tag{25}$$

$$\frac{1}{2}V_{\mathrm{F}}C_{\mathrm{F}}^k + \frac{1}{2}M_{\mathrm{P}}^k + \frac{1}{2}M_{\mathrm{C}}^k = C_{\mathrm{in}}V_{\mathrm{P}} - \frac{1}{2}V_{\mathrm{F}}C_{\mathrm{F}}^{k-1} - \frac{1}{2}M_{\mathrm{P}}^{k-1} - \frac{1}{2}M_{\mathrm{C}}^{k-1}, \quad 1 \le k \le nt \tag{26}$$

Modeling of chemical loss

The loss of migrants in food is simulated using different functions. The chemical residues in packaging materials at time t are:

$$M_{\mathrm{P,t}} = A\int_0^{l_1} u(x,t)\mathrm{d}x \tag{27}$$

$$M_{\mathrm{C,t}} = A\int_0^{l_1} v(x,t)\mathrm{d}x \tag{28}$$

These integrals can be expressed as sums by using the compound trapezoidal method:

$$M_{\mathrm{P,t}} = \frac{1}{2}Ah(u_0^k + u_{\mathrm{m}}^k) + Ah\sum_{i=1}^{m-1} u_i^k \tag{29}$$

$$M_{\mathrm{C,t}} = \frac{1}{2}Ah(v_{\mathrm{m}}^k + v_M^k) + Ah\sum_{j=m+1}^{M-1} u_j^k \tag{30}$$

where $M_{\mathrm{P,t}}$ is the contaminant content in regeneration layer at time t, $M_{\mathrm{C,t}}$ – the contaminant content in original layer at time t, and A – the contact areas between packaging materials and food.

Assuming that a first-order reaction occurs in the food, it is defined by the first-order reaction:

$$-\frac{\mathrm{d}C_{\mathrm{F}}(t)}{\mathrm{d}t} = K_{\mathrm{A}}C_{\mathrm{F}}(t) \tag{31}$$

$$C_{\mathrm{F}}(t) = C_{\mathrm{F0}}\mathrm{e}^{-K_{\mathrm{A}}t} \tag{32}$$

$$C_{\mathrm{F}}(t)V_{\mathrm{F}} = C_{\mathrm{F0}}\mathrm{e}^{-K_{\mathrm{A}}t}V_F \tag{33}$$

$$M_{\mathrm{F}} = M_{\mathrm{F0}}\mathrm{e}^{-K_{\mathrm{A}}t} = M_{\mathrm{F0}}b_1a_1^{c_1t} \tag{34}$$

where K_{A} is the rate of loss reaction, C_{F0} – the migrant concentration in food without considering loss, M_{F0} – the migrant mass in food without considering loss, M_{F} – the migrant mass in food considering loss, V_{F} – the food volume, C_{F} – the migrant concentration in food, b_1 – the front coefficient of loss function in the first order reaction, a_1 – the base number of loss function in the first order reaction, c_1 – the exponential coefficient of loss function in the first order reaction, and a_1, b_1, c_1 together determine the loss ratio of migrant per unit time.

According to the principle of conservation of mass, the amount of chemicals entering food at t time is equal to the difference between the initial amount in packaging material and the residual amount in packaging at time, namely $M_{\mathrm{F,t}} = M_{\mathrm{P0}} – M_{\mathrm{P,t}} – M_{\mathrm{C,t}}$. Because of the instability of migrant in food, assuming that the loss function is expressed in $E(t)$ and loaded into the numerical model in two ways, then the chemical mass $M_{\mathrm{F,t1}}$’ = $M_{\mathrm{F,t}}E(t)$ (loading mode 1) or $M_{\mathrm{F,t1}}$’ = $M_{\mathrm{F,t}} – E(t)$ (loading mode 2) in food at time.

The final amount of chemical in food considering loss are as follows:

$$M_{\mathrm{F,t1}}{}' = M_{\mathrm{F,t}} E(t) = b_1 a_1^{c_1 t} M_{\mathrm{F,t}} \tag{35}$$

or

$$M_{\mathrm{F,t2}}{}' = M_{\mathrm{F,t}} - E(t) = M_{\mathrm{F,t}} - b_2 a_2^{c_2 t} \tag{36}$$

The final expression of chemical mass in food are:

$$M_{\mathrm{P,t}} = \int_0^{l_1} Au(x,t)\mathrm{d}x = \frac{1}{2} Ah(u_0^k + u_{\mathrm{m}}^k) + Ah\sum_{i=1}^{m-1} u_i^k$$

$$M_{\mathrm{C,t}} = \int_{l_1}^{l} Av(x,t)\mathrm{d}x = \frac{1}{2} Ah(v_{\mathrm{m}}^k + v_M^k) + Ah\sum_{i=m+1}^{M-1} v_i^k$$

$$M_{\mathrm{F,t1}}{}' = (C_{\mathrm{in}} Al_1 - M_{\mathrm{P,t}} - M_{\mathrm{C,t}})E(t) = \tag{37}$$

$$= \left\{ C_{\mathrm{in}} Al_1 - \left[\frac{1}{2} Ah(u_0^k + u_{\mathrm{m}}^k) + Ah\sum_{i=1}^{m-1} u_i^k \right] - \right.$$

$$\left. - \left[\frac{1}{2} Ah(v_{\mathrm{m}}^k + v_M^k) + Ah\sum_{i=m+1}^{M-1} v_i^k \right] \right\} (b_1 a_1^{c_1 t}) \quad \text{(loading mode 1)}$$

or

$$M_{\mathrm{P,t}} = \int_0^{l_1} Au(x,t)\mathrm{d}x = \frac{1}{2} Ah(u_0^k + u_{\mathrm{m}}^k) + Ah\sum_{i=1}^{m-1} u_i^k$$

$$M_{\mathrm{C,t}} = \int_{l_1}^{l} Av(x,t)\mathrm{d}x = \frac{1}{2} Ah(v_{\mathrm{m}}^k + v_M^k) + Ah\sum_{i=m+1}^{M-1} v_i^k$$

$$M_{\mathrm{F,t2}}{}' = C_{\mathrm{in}} Al_1 - M_{\mathrm{P,t}} - M_{\mathrm{C,t}} - E(t) = \tag{38}$$

$$= C_{\mathrm{in}} Al_1 - \left[\frac{1}{2} Ah(u_0^k + u_{\mathrm{m}}^k) + Ah\sum_{i=1}^{m-1} u_i^k \right] -$$

$$- \left[\frac{1}{2} Ah(v_{\mathrm{m}}^k + v_M^k) + Ah\sum_{i=m+1}^{M-1} v_i^k \right] - (b_2 a_2^{c_2 t}) \quad \text{(loading mode 2)}$$

Conclusion

The paper reports a mathematical model aiming to prediction of the migration of chemicals from two-layer packaging material into food by considering the loss of chemicals in food. Many important parameters are considered in the model, such as diffusion coefficient, partition coefficient, transmit coefficient at the interface of material and food. Especially, chemical instability and different loading ways of loss function are considered in the model. Numerical solution is given by compact difference scheme from the finite difference method. Recently Liu, *et al.* suggested a fractal release kinetics [18, 19] which can take into account the porous structure on the release process, Liu's fractal model can be extended to the present study, and we will discuss the release process by either a fractal model or a fractional model [20-26] in a future paper.

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