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Development of a structure-based model for moisture diffusion in multiphase lipid networks

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ABSTRACT

Moisture diffusion in lipids has been traditionally described by generic models that do not offer complete information about its structural dependency. Due to the lack of a predictive structure-based model in literature, we do not know the crystallization conditions needed to favorably alter structure and decrease moisture transport in lipids. The objective of this study was to develop a mathematical model to predict effective moisture diffusivity based on fractal structure and void fraction in lipids. Predicted water uptakes using the proposed model were in better agreement (RMSE 0.01–0.03) with experimental data when compared to models from literature (RMSE 0.01–0.16). Clearly, accounting for the fractality of fat crystal networks and vapor phase diffusion in the void fraction were critical to the success of the proposed model over all other models. Interestingly, moisture diffusion occurred predominantly in the void fraction compared to the liquid oil fraction of all the lipid samples.

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1. Introduction

In multiphase foods, the moisture diffusivity in the medium is known to be influenced by the diffusion rates in the different phases, the relative volume fractions, and the geometric distribution of the phases (Ghosh et al., 2002). An example of multiphase systems is a food lipid, which consists of solid fat crystals dispersed in liquid oil with their relative volumes depending on the physicochemical nature of the lipid at that temperature (Ghosh et al., 2002; Marangoni, 2002; Tang and Marangoni, 2006a). Interestingly, Loisel et al. (1997) have observed the presence of void spaces in lipids which make up the third phase in the multiphase lipid network. They visualized lipids to be a network of cavities (voids) enclosed by fat crystals embedded in the liquid oil matrix. When exposed to moisture, a permeating water molecule traveling through the lipid network encounters three phases in its path-fat crystals, liquid oil, and voids. The mass transfer of water can be controlled by modifying the chemical composition and the network structure (Martini et al., 2006; Roca et al., 2007). Moisture diffusion occurring due to the chemical nature of triacylglycerol (TAG) molecules in the liquid phase is described by a molecular diffusivity

value, D (Cussler, 2009). It has been found that presence of impenetrable particles, such as fat crystals, at meso or nanoscale always hinders the diffusion of foreign molecules in a material (Guillard et al., 2003). One of the reasons behind a decrease in diffusion is the increase in tortuosity of the diffusion path due to the presence and distribution of impermeable solid fat crystals (Aguilera et al., 2004; Ghosh et al., 2002; Yuan et al., 2009). As a result, the effective moisture diffusivity value (D_{eff}) for the network is lesser than the molecular diffusivity in the liquid oil (Martini et al., 2006; Roca et al., 2007).

To understand the impact of the lipid network structure on moisture diffusion, it is necessary to quantify the effect of solid-liquid ratio, porosity, and spatial distribution of fat crystals on the moisture diffusivity values. Moreover, a structure-based diffusion model must be adopted or developed to establish clear correlations between structure and diffusion. For this purpose, the objective of this study was to adopt mathematical and empirical models from literature and examine their applicability for prediction of moisture diffusion in lipids. Then, a diffusion model based on structural attributes of lipid networks was developed. The past and proposed models were validated using measured structure and diffusion data and the most accurate predictive model was selected. Finally, simulations of the chosen diffusion model were conducted to enhance the understanding of how various structural inputs influence moisture diffusion in lipids.

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Nomenclature			
A	Cross-section area (m ²)	l	Thickness of lipid layer (m)
a	Crystal size (m)	m	Exponent equivalent to $D_{\text{box}}/2 - D_{\text{box}}$
B	Permeability coefficient (m ²)	M_{∞}	Equilibrium moisture uptake at long time (arbitrary units)
C_0	Initial moisture concentration in lipid (kg m ⁻³)	M_t	Moisture uptake at time, t (arbitrary units)
C_1	Uniform moisture concentration in agar gel (kg m ⁻³)	nT	Number of terms
D_a	Molecular diffusivity of water vapor in air (m ² s ⁻¹)	p	Empirical parameter in the tortuosity-porosity correlation
D_{box}	Box-counting fractal dimension	q	Empirical parameter in the tortuosity-porosity correlation
D_c	Diffusivity in the continuous liquid oil phase (m ² s ⁻¹)	SEDeffi	Standard error of the diffusivity coefficient for sample i
D_d	Diffusivity in the dispersed fat crystals (m ² s ⁻¹)	WUR	Water uptake ratio (M_t/M_{∞})
D_{eff}	Effective diffusivity of moisture (m ² s ⁻¹)	Δx	Thickness of element (m)
$D_{\text{eff}i}$	Mean value of the diffusivity coefficient for sample i	ε	Effective porosity of the system
d_i	Distance covered by sample i in a certain time	ζ	Volume fraction of liquid oil
$D_{i,\text{eff}}$	Effective moisture diffusivity in the liquid oil fraction (m ² s ⁻¹)	θ	Empirical parameter accounting for void structural attributes
$D_{v,\text{eff}}$	Effective water vapor diffusivity in the void fraction (m ² s ⁻¹)	v	Volume fraction of void spaces
j	Mass flux of moisture (kg m ⁻² s ⁻¹)	τ	Tortuosity of the diffusional path
K	Parameter equivalent to tortuosity factor in the Kozény- Carman equation	Φ	Solid fat content or SFC

2. Modeling

In this section, a mass transfer model is built from first principles to describe moisture diffusion in a two-layered food with a crystallized lipid layer placed on top of a high-moisture agar gel layer, as illustrated in Fig. 1. The two-layered food system is in a glass enclosure such that the only direction of moisture diffusion is from the gel to the lipid in the positive x direction. The agar gel is assumed to be an infinite source of moisture for this study. Assuming semi-infinite slab geometry for the lipid, no reaction, and no bulk flow; transport equations for diffusion process are developed here.

Consider a differential element of volume $A \Delta x$ in the lipid, where A is the cross-section area (m²), and Δx is the thickness of element (m). The lipid layer is l (m) thick and initially at uniform moisture concentration C_0 (kg m⁻³). At a certain time, t (s), the lipid layer is put in contact with the agar gel of uniform moisture concentration C_1 (kg m⁻³) and the concentration at the lipid-gel interface abruptly increases to C_1 . One-dimensional unsteady state mass transfer of water occurs in the positive x direction and

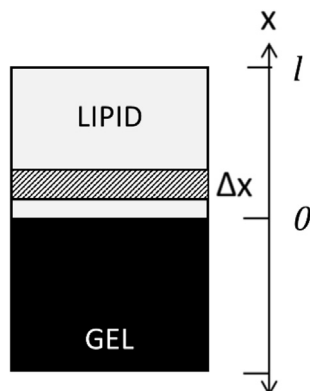


Fig. 1. A two-layered system with a lipid layer placed directly on top of a high-moisture source such as agar gel.

the mass balance for the element is:

solute accumulation in volume $A \Delta x$ = rate of diffusion into the layer at x

– rate of diffusion out of the layer at $x + \Delta x$ which can also be written as:

$$\frac{\partial A \Delta x C}{\partial t} = A (j|_x - j|_{x+\Delta x}) \quad (1)$$

where j is mass flux of moisture (kg m⁻² s⁻¹). Dividing the above equation by $A \Delta x$, we get,

$$\frac{\partial C}{\partial t} = - \frac{((j|_{x+\Delta x} - j|_x))}{(x + \Delta x) - x} \quad (2)$$

As $\Delta x \rightarrow 0$,

$$\frac{\partial C}{\partial t} = - \frac{\partial j}{\partial x} \quad (3)$$

Substituting j with the definition of Fick's I law of diffusion in the above equation, Fick's II law is obtained as,

$$\frac{\partial C}{\partial t} = - \frac{\partial}{\partial x} \left(D_{\text{eff}} \frac{\partial C}{\partial x} \right) \quad (4)$$

where D_{eff} is effective diffusivity of moisture in a lipid containing multiple phases (m² s⁻¹). The above differential equation is solved using the following initial (IC) and boundary conditions (BC):

$$\text{IC: At } t = 0, \quad 0 \leq x \leq L, \quad C = C_0 \quad (5)$$

$$\text{BC - 1: At } t > 0, \quad x = 0, \quad C = C_1 \quad (6)$$

$$\text{BC - 2: At } t > 0 \quad x = L, \quad D_{\text{eff}} \frac{\partial C}{\partial x} = 0 \quad (7)$$

Crank (1975) presents several analytical solutions of Fick's II law of diffusion (equation (4)) under the above boundary conditions

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