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PET식품 용기에서 발효 모사 식품으로 전이되는 아세트알데히드와 부틸알데히드 예측 모델

Theoretical Migration Estimation of Acetaldehyde and Butyraldehyde from Polyethylene Terephthalate (PET) into Fermented Food Simulants

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ABSTRACT

Objectives: Materials coming into contact with food may result in the migration of chemical substances into the food. To protect consumers from exposure, Regulation (EU) No. 10/2011 specifies the use of standard migration tests. Polyethylene terephthalate (PET), widely used for food packaging materials, has drawn the attention of researchers because unwanted migration of PET into food might occur when consumers reuse packaging material. The aim of this study was to predict and develop a migration model for two components, acetaldehyde and butyraldehyde in PET, into food simulants under conditions of changing pH and solvents, such as those observed in fermented foods like kimchi or sauerkraut.

Methods: Using a migration model based on Fick's second law of diffusion in one dimension, the migration of acetaldehyde and butyraldehyde from PET into a simulant of fermented food at 20 °C over 10 days was evaluated. The simulant for fermented food was modelled as 10% ethanol for three days, followed by 3% acetic acid for seven days.

Results: The migration of acetaldehyde into the 10% ethanol was 0.36 times that of a simulated fermented food system, while that of butyraldehyde was 1.34 times greater. These results may have been influenced by the chemical interactions among the migrants, polymers and simulants, as well as by the solubilities of the migrants in polymers and simulants.

Conclusion: Because food simulants have a limited capacity to mimic real food systems under the current migration model, an appropriate simulant and migration test should be considered in the case of increasing acidity. Furthermore, since the accuracy of the worst-case estimation of migration predicted by the current model is severely limited under changing food conditions, food simulants and their interactions should be further investigated with respect to conservative migration modelling.

Keywords: Food simulants, migration, partition coefficients, polyethylene terephthalate (PET), simulant for fermented food (SfF)

I. Introduction

In the packaging industry, plastics have been widely used because of its lightness, stability, strength and inertness. These features make plastics ideal packaging materials for mass production in a

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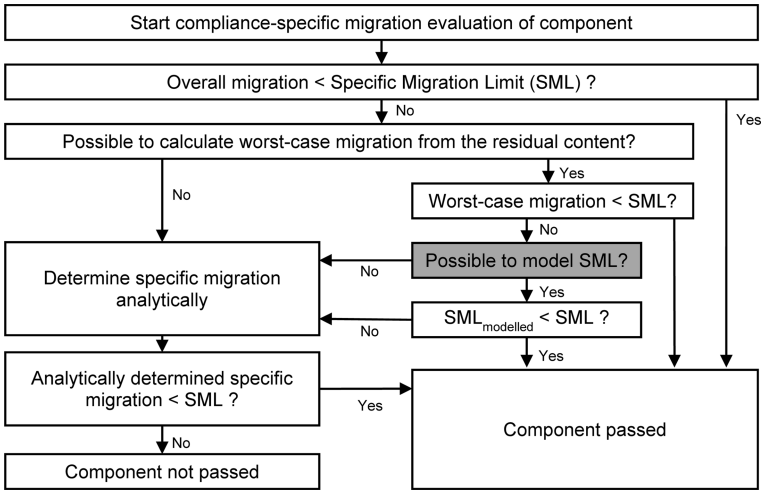

Fig. 1. Generic scheme for the evaluation of specific migration[6)]

wide range of commercial applications. In 2014, plastic production in Europe reached 59 million tons, of which 39.6% was used for packaging materials.[1)] The most important function of a packaging material is to preserve the packaged goods. Because of the principal chemical instability of foods, food packaging must be optimized to ensure food safety. Plastic materials, however, can release low-molecular weight compounds, such as monomers or additives, into foodstuffs via diffusion. When these compounds enter the food system, they may affect the safety and quality of the food product.

In the European Union (EU), materials intended to come into contact with foodstuffs are regulated by the Framework Regulation (EC) No. 1935/2004 (2004).[2)] Plastics are controlled by a specific directive (EU Commission Directive 2002/72/EC, 2002), and specific and overall migration tests should be conducted using food simulants under certain temperature and time conditions.[3)] Because of the complexity of the matrix and the analytical difficulties of quantifying migrating substances in foodstuffs, EU legislators have agreed upon six food simulants to be used instead. These simulants, which were chosen based on research results to resemble the principal food types, are as follows: 3% acetic acid; 10%, 20% and 50% ethanol; vegetable oil; and poly(2,6-diphenyl-p-phenylene oxide) (PPPO). However, the physicochemical properties of the food under consideration should be taken into account when selecting the simulants to be studied. In particular, the simulant should be selected in accordance with the complexity and diversity of the foodstuff under consideration.

To assess the safety of polymeric food contact materials according to EU regulations, specific and overall migration should be tested to evaluate whether the mass transfer of compounds from plastic into foodstuff takes place under specified test conditions. For overall migration tests, the total quantity of constituents released may not exceed 60 mg/kg or 10 mg/dm^2 per material or article. With specific migration testing, the amount of an isolated substance must not be present in quantities greater than a fixed value determined according to its toxicological characteristics.[4)] However, the determination of specific migration into food or food simulants is error-prone, costly and time-consuming. Numerous studies published during the last two decades have shown that migration from food contact materials into the food system is a

predictable physical process. It has been shown that migration occurs as a result of the process of diffusion of the migrant in plastic, which can be described by Fick's law of diffusion. Theoretical migration estimation can therefore be used as an alternative tool for regulatory purposes.[5] The generic approach to demonstrating compliance with specific migration limits (SML) is illustrated in Fig 1.[6]

To predict specific migration from plastic materials, the core parameters are the diffusion coefficient of the migrant in the plastic, D_P, and the partition coefficient of the migrant between the plastic and the food simulant $K_{i,F/P}$. One of the recognized models predicts upper-bound migration values validated within the EU project SMT-CT98-7513.[7,8] The mathematical equations used in the present study are listed under "Materials and Methods" as equations 1-4.

In the present study, the foodstuff is assumed to be a fermented food such as kimchi or sauerkraut. In lactic acid fermentation, the production of acid results in a lower pH and an increase in sourness.[9] However, none of the six food simulants has this property, and the migration model does not seem appropriate for a change in pH or other physical properties of the food system.

The main objective of this study was to determine, based on a mathematical model, whether fermented food influences the migration kinetics of selected migrants from polyethylene terephthalate (PET). The results will provide new insight into migration modelling, which should facilitate the inclusion of changeable food properties in the model. To date, this is the first study to investigate migration modelling for fermented food.

II. Materials and Methods

1. Migration modelling

Mass transport between plastic materials and food simulants under non-steady-state conditions is described by Fick's second law of diffusion (Eq. 1):

$$\frac{\partial c}{\partial t} = D_P \frac{\partial^2 c}{\partial x^2} \quad (1)$$

where c is the concentration of the migrant in polymer P at time t and distance x from the origin of the x-axis, and D_P is the constant diffusion coefficient for P.

Cranck (2007) presented the analytic solution in the form of Eq. 2, which can be expressed by the amount of migrant released per unit area A from the polymer into the food simulant at time t:

$$\frac{m_{F,t}}{A} = 0.1 C_{P,0} \rho_P d_P \left(\frac{\alpha}{1+\alpha}\right) \left[1 - \sum_{n=1}^{\infty} \frac{2\alpha(1+\alpha)}{1+\alpha+\alpha^2 q_n^2} exp\left(-D_P t \frac{q_n^2}{d_p^2}\right)\right] \quad (2)$$

with

$$\alpha = \frac{1}{K_{P,F}} \frac{V_F}{V_P} = \frac{c_{F,\infty}}{c_{P,\infty}} \frac{\rho_F}{\rho_P} \frac{V_F}{V_P}$$

where $m_{F,t}$ is the mass (in mg) of the migrant transferred from polymer P into food simulant F after time t (seconds); A is the contact area of P with F (in cm^2); $C_{P,0}$ is the initial concentration (in mg/kg) of the migrant in P; ρ_P is the density of P (in g/cm^3); d_P is the thickness of P (in cm); V_P is the volume of P (in cm^3); V_F is the volume of F (in cm^3); q_n is the positive root of the equation $\tan q_n = -\alpha q_n$; D_P is the diffusion coefficient of the migrant in the polymer (in cm^2/s), and $K_{P/F}$ is the partition coefficient of the migrant between P and F.

2. Diffusion coefficient

A simple approach to estimating D_P was reported in the 1990s, using the relative molecular mass of the migrant, M_r, a material-specific coefficient A_P and the absolute temperature T, based on empirical relationships.[10] Instead of D_P, a polymer-specific upper-bound diffusion coefficient, D_P*, can be

Table 1. "Upper-bound" A_P' values for selected polymers. A_P': material-specific coefficient, τ: major simplification of the Pringer model (Eq. 4)

Polymer	A_P'	τ	T(°C)
LDPE/LLDPE[a]	11.5	0	< 80/< 100
HDPE[b]	14.5	1577	< 90
PP (homo and random)[c]	13.1	1577	< 120
PP (block copolymer)	11.5	0	< 100
PS[d]	−1.0	0	< 70
HIPS[e]	1.0	0	< 70
SBS[f]	10.5	0	< 70
PET[g]	3.1	1577	< 70
PEN[h]	5.0	1577	< 175
PA[i] (6,6)	2.0	0	< 100

a. LDPE: low-density polyethylene; LLDPE: linear low-density polyethylene
b. HDPE: high-density polyethylene
c. PP: polypropylene
d. PS: polystyrene
e. HIPS: high-impact polystyrene
f. SBS: poly(styrene-butadiene-styrene)
g. PET: polyethylene terephthalate
h. PEN: poly(ethylene 2,6-naphthalate)
i. PA: polyamide

estimated ($D_P \leq D_P$*). Hence, migration estimations using D_P* values lead to "worst-case" or "overestimated" values.[11] D_P* can be estimated using Eq. 3:[12]

$$D_P = D_0 e\left(A_P - 0.1351 \cdot M_r^{\frac{2}{3}} + 0.003 \cdot M_r - \frac{R \cdot 10454}{R \cdot T}\right) \quad (3)$$

$$A_P = A_P' - \frac{\tau}{T} \quad (4)$$

The polymer-specific parameter, A_P, is a function of temperature, as shown in Eq. 4. It describes the diffusion behaviour of the polymer matrix in relation to the migrants. The parameter τ and the constant 10454 in Eq. 3 both contribute to the diffusion activation energy, E_A = (10454+τ) R, where R = 8.3145 (J/mol K) is the gas constant. By the empirical relationship, one group of plastics, such as low-density polyethylene (LDPE), is represented by τ = 0, and another group of plastics, such as high-density polyethylene (HDPE) and polypropylene (PP), is represented by τ = 1577.[12] A list of A_P' and τ values, as currently applied in the Practical Guidances on the Application of Migration Modelling for the Estimation of Specific Migration, is shown in Table 1.[13]

3. Partition coefficient

The transfer of migrant i from food to polymer takes place until thermodynamic equilibrium is reached. The partition coefficient, $K_{i,F/P}$, is defined as the ratio of the migrant concentration in the polymeric material, C_P, to its equilibrium concentration in the foodstuff, C_F:[14]

$$K_{i,\,F/P} = \frac{C_P}{C_F}$$

When K = 1, the system is at equilibrium and the migrant concentration in the polymeric material equals that in the food. If K > 1, more of the migrant is present in the polymer than in the food. Current migration modelling recommends establishing these values in a generalized and conservative way with respect to a "worst-case" scenario, i.e. K = 1, which means that the substance is very soluble in the foodstuff.[5]

Tehrany and Desobry (2004) observed that the migration process is influenced by compounds that are volatile between food and packaging. Various factors such as the chemical and physical structure of the food, the packaging, the migrant, the pH, the fat and water content of the food affect the partitioning of the migrant between the packaging and the food.

For the prediction of partition coefficients, Elmira and colleagues (2006) studied a mathematical model in a food/packaging system by using quantitative structure-property relationships (QSPR), based on molecular descriptors.[15] The experimental values of the partition coefficients were determined at 20°C. The identified molecular descriptors are the

Table 2. Physicochemical properties of food simulants, polymers and migrants. MW: molecular weight; LUMO: lowest unoccupied molecular orbital; HLB: hydrophilic-lipophilic balance. Polymer abbreviated as follows: PET: polyethylene terephthalate

Compound	Name	Polarity	MW	LUMO migrant	HLB migrant
Polymer	PET	9.65	-	-	-
Simulant	Water	16	18.01	-	-
	10% ethanol	15.28	20.82	-	-
	3% acetic acid	15.75	19.27	-	-
	95% ethanol	9.16	44.66	-	-
Migrant	Acetaldehyde	-	44.05	0.1511	8.23
	Butyraldehyde	-	72.1	0.1521	4.59

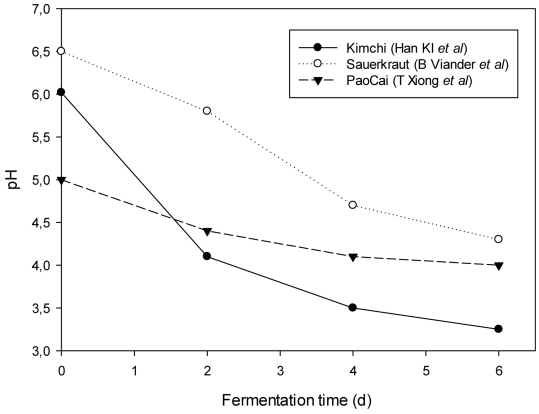

Fig. 2. Changes in pH of kimchi,[18] sauerkraut[19] and pao cai[20] during spontaneous fermentation at 15-25°C

physicochemical properties of each food, its packaging, and the migrant. As listed in Table 2, these descriptors include the polarity of the food, x_1; the polarity of the polymer, x_2; the molecular weight, x_3; the lowest unoccupied molecular orbital (LUMO) of the migrant, x_4; the molecular weight of the food, x_5,; and the hydrophilic-lipophilic balance (HLB) of the migrant, x_6. Based on these molecular descriptors, the model for the calculation of partition coefficient was adapted as follows:

$$K = a_0 + a_1x_1 + a_2x_2 + a_3x_3 + a_4x_4 + a_5(x_1 exp x_3) + a_3(x_1x_4) + a_7x_2x_4 + a_8(x_3x_4) + a_9x_3^2 + a_{10}x_4^2 + a_{11}x_5x_6 \quad (4)$$

Following this principle, S. Parviz *et al.* (2011) developed a QSPR model using the adaptive neuro-fuzzy inference system (ANFIS).[16] This multilayer neural network-based fuzzy system is 41.3 times more precise than the previous model in terms of root-mean-square error (RMSE) values. We therefore collected our data using the ANFIS model.

4. Fermented food

Fermented food generally produces metabolites such as lactic acid bacteria. Lactic acid fermentation spontaneously produces acid, with a resultant decrease in pH and consequent increase in sourness.[9] Several studies have shown that the pH profile of fermented vegetables such as kimchi and sauerkraut decreases sharply during storage (Fig. 2).[17]

During the fermentation of kimchi (fermented Korean side dish), sauerkraut (German fermented cabbage) and pao cai (Chinese pickled cabbage), mean pH decreased from 5.0–6.5 to < 4.5. When selecting the food simulant to use in this migration study, this change in metabolic activity in the food system was taken into account.

The six food simulants that can be used to simulate real foods, together with the types of food they simulate, are as follows[3]:

- Simulant A: 10% ethanol, for foods that have a hydrophilic character
- Simulant B: 3% acetic acid, for foods that have pH < 4.5
- SimulantC: 20% ethanol, for foods with an alcohol content of ≤ 20%
- Simulant D1: 50% ethanol, for lipophilic and alcoholic foods with an alcohol content of > 20% and oil-in-water emulsions
- Simulant D2: vegetable oil, for lipophilic foods that contain free fats on the surface
- Simulant E: PPPO with a particle size of 60–80

mesh and a pore size of 200 nm, for dry foods The selected simulant should be examined under specific temperature and time conditions. However, in the case of fermented foods, a single simulant is not appropriate, since the increasing acidity means that the most appropriate simulant changes from A to B. We therefore simulated fermented foods by using 10% ethanol for three days, followed by 3% acetic acid for seven days. We designated this simulant as "SfF". We modelled the migration of acetaldehyde and butyraldehyde into three simulants (10% ethanol, 3% acetic acid, and SfF) using the following parameter values: initial concentration of the migrant ($C_{P,0}$) = 100 ppm; temperature = 20°C; internal bottle wall surface = 152 cm^2; wall thickness (d) = 0.3 cm; and density of polymer (PET) = 1.38 g/cm^3.

5. Characterization of polymers and migrants

PET has been widely used as a food packaging material for many years. The major advantage of PET is its inertness, with good barrier properties against moisture and carbon dioxide, as well as the very low migration tendency of its constituents relative to other packaging materials. Because of the very low diffusion characteristics of PET, there is limited information available concerning migration testing or modelling parameters for this material. However, the migration of additives, catalyst residue and residual monomers from PET plastic is nevertheless considerable.[21] This material's inertness makes PET one of the most recycled types of plastic packaging. As a consequence of the recycling process, post-consumer recycled PET comes into direct contact with foods and beverages. During the first use of the PET, compounds would have migrated from the food it packaged into it. When it is recycled, then, these compounds can remigrate into the food from PET if it is once again used as food packaging.[22] For example, Mancini *et al*.[23] showed that acetaldehyde and butyraldehyde were present in a recycled PET sample subjected only to conventional washing.

Table 3. Partition and diffusion coefficients for each food simulant and migrant in PET at a temperature of 20°C

Simulant	Migrant	$K_{P/S}$	D_P (cm^2/s)
10% ethanol	Acetaldehyde	0.0752	7.0541E-14
3% acetic acid	Acetaldehyde	0.0176	7.0541E-14
10% ethanol	Butyraldehyde	0.0182	3.98579E-14
3% acetic acid	Butyraldehyde	0.0366	3.98579E-14

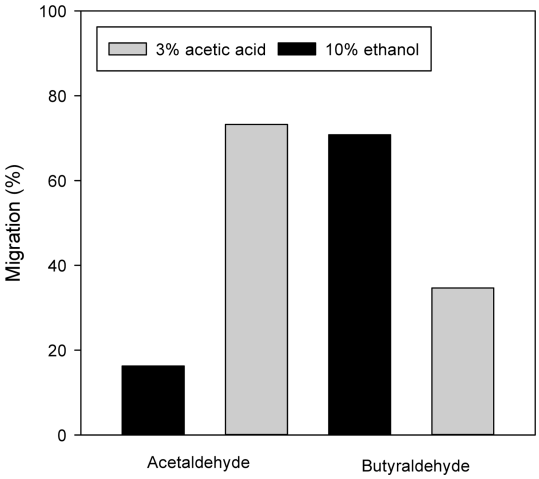

Fig. 3. Percentage of acetaldehyde and butyraldehyde that migrated in 10% ethanol and 3% acetic acid after 10 days, at 20°C.

III. Results

The key parameter to identify for the modelling is the polymer-specific A_P, which is used to calculate the diffusion coefficient, D_P, of a given molecule in a plastic material. With respect to PET, the currently used values are A_P = 6 and τ = 1577, under temperatures of up to 175°C, for worst-case migration modelling.[8] Recently, the European Commission has updated and refined the model for different temperature conditions; below 70°C, A_P = 3.1, and for above 70°C, A_P = 6.4.[5] In this study we aimed to describe the migration model at 20°C, so we used the former value of A_P.

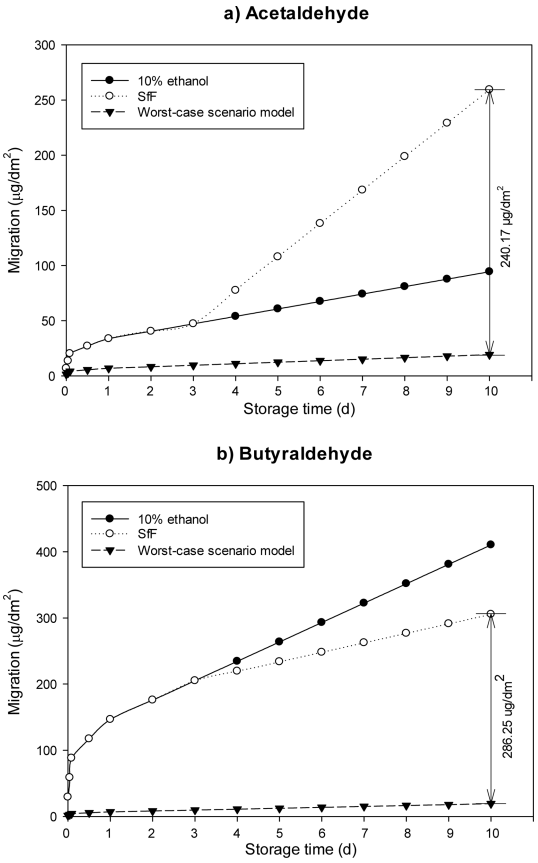

Fig. 4. Migration of acetaldehyde (a) and butyraldehyde (b) from PET into a simulant or foodstuff, as modelled for 10% ethanol, the SfF simulant (10% ethanol for 3 d, followed by 3% acetic acid for 7 d), or according to the worst-case scenario (K=1).

Table 3 summarizes the two key parameters of the migration process in PET for each migrant and simulant. The migrant acetaldehyde in 10% acetic acid had the highest $K_{F/P}$ value of the migrants studied, showing that this compound has a higher affinity for PET under acidic conditions. On the other hand, acetaldehyde in 3% acetic acid had the lowest $K_{F/P}$ value.

Acetaldehyde migrated to a greater extent in 3% acetic acid than did butyraldehyde, and vice versa for 10% ethanol (Fig. 3).

For the SfF simulant, there was a marked change in the rate of migration at the time of the change in pH (day 3; Fig. 4). When only 10% ethanol was used, the quantity of acetaldehyde in the simulant after 10 days was 0.36 times that in the SfF. On the other hand, the quantity of butyraldehyde in 10% ethanol was 1.34 times that in SfF after 10 days. Furthermore, when the "worst-case" scenario was modelled (K = 1) using the currently accepted parameter values, the migration estimated was substantially less than that observed for SfF (Fig. 4). The difference between SfF and the "worst-case" scenario was 240.17 μg/dm^2, as 13.47 times of the worst-case predicted value, for acetaldehyde and 286.25 μg/dm^2, as 15.86 times of that, for butyraldehyde.

IV. Discussion

Ideally, the food simulant used in a migration study should simulate the behaviour of specific compounds in a real food system. However, because of the uncertainty concerning whether a food simulant exactly mimics a real food system, a new type of worst-case simulation should be considered. For the migrant acetaldehyde, migration into 10% ethanol was lower than in a simulated fermented food system. In contrast, migration of butyraldehyde in 10% ethanol was greater than in the fermented food simulant. Several studies have demonstrated that migration is greater in low-pH foodstuffs.[24,25] However, our results show that 3% acetic acid alone does not cover the migration that occurs in 10% ethanol, and is thus also unsuitable as a food simulant for fermented food. Leufvén and Hermansson (1993) determined the influence of pH on the sorption of aroma compounds into different polymers. The quantities of aroma compounds extracted varied with pH and polymer type, because of the various interactions between the migrant, the polymer and the solution. These interactions can cause changes in the molecule competition in the polymer, and the simulant may affect the polarity of aromatic compounds. Thus the interactions

between polymers, aroma components and the pH of food systems are complex.[26]

With respect to the migration of the two components into the food simulants 3% acetic acid and 10% ethanol, the migration of acetaldehyde have increased instantly upon the change from ethanol to acetic acid (Fig. 4a). This may be explained by the interaction between acetaldehyde and acetic acid. Because of resonance and inductive effects, acetaldehyde is particularly protonated and becomes more hydrophilic under acidic conditions. As a result, protonated acetaldehyde is highly soluble in 3% acetic acid. On the other hand, the migration of butyraldehyde decreased when the 10% ethanol was changed to 3% acetic acid after three days. The water-octanol partition coefficient, K_{ow}, is expected to increase as the difference between the solubility parameters of water and octanol decreases. For butyraldehyde, log K_{ow} = 0.88, and for acetaldehyde, log K_{ow} = 0.52.[27] Since an increase in K_{ow} reflects greater lipophilicity, we assume that butyraldehyde is much less soluble in 3% acetic acid than it is in 10% ethanol.[28]

V. Conclusions

Despite the inherently low diffusivity of the PET polymer, the partitioning effect enables the estimation of its migration behaviour under different pH conditions. One important objective of this study was to evaluate the effectiveness of the migration model in conditions of decreasing pH. It must be emphasised that the partition coefficient was derived based on different simulants.

From the results of the present study, one important conclusion can be drawn concerning the effect of a change in pH in food systems. Since food simulants have a limited capacity to mimic real food systems under the current migration model, one simulant is inadequate to predict the worst-case scenario. To obtain a more accurate prediction, therefore, food simulants and their interactions should be further investigated with respect to migration modelling.

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