

## Migration and Diffusion of Diphenylbutadiene from Packages into Foods

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Monitoring of exposure to chemicals from food contact materials is a subject of increasing importance. The concentration of the chemicals and their migration levels, as well as food consumption and packaging usage data, are required to enable calculation of the degree of such exposure. The present study investigated the migration kinetics of diphenylbutadiene (DPBD) from packages into flour, rice, honey, milk powder and toast. Migration was not always negligible, except in honey and skimmed milk powder. Experiments carried out with starch alone enabled us to conclude that diffusion of migrants occurred through starch and fat. Key diffusion parameters were determined (diffusion coefficient and partition coefficient) based on Fick's second equation. The following diffusion coefficients were obtained at 25 °C:  $2.7 \times 10^{-10}$ ,  $3.4 \times 10^{-11}$ ,  $3.2 \times 10^{-10}$ ,  $8.4 \times 10^{-11}$ ,  $8.1 \times 10^{-11}$  cm<sup>2</sup> s<sup>-1</sup>, for wheat flour, rice, milk powder and toast, with 4 and 21% fat, respectively. A very good fit between experimental and predicted data was achieved. The data obtained in the present study confirm the validity of the mathematical model for predicting migration from Food Contact Materials (FCM) into foods.

**KEYWORDS:** Dry foodstuffs; migration; mathematical modeling

### INTRODUCTION

Plastic food packages may contain additives used to minimize degradation during processing, to facilitate processing and to increase stability during storage. The additives, such as antioxidants, dyes, pigments, antifogging agents, stabilizers and plasticizers, are generally present at low levels but may migrate into food packaged with these materials and then be ingested by the consumer (1, 2). The most widely used food packaging material is low-density polyethylene (LDPE).

In order to protect human health and to ensure food safety it is important to monitor the exposure of chemicals that migrate from FCM (food contact materials) into foodstuffs (3). This can be defined as the amount of a substance that may be consumed as a result of its migration from the packaging to the foodstuff, and to calculate it, the concentration of the substance in food, or migration data, and food consumption and packaging usage data are required.

Migration depends on many factors like the chemical and physical nature of the migrant and of the food in contact with the packaging, the surface area of the packaging material in contact with the foodstuff, the time and temperature of the contact, and the type of packaging material (LDPE has intrinsically very high diffusion coefficients and can therefore cause higher migration levels). Several studies have been carried out to calculate food

consumption or packaging usage data, although information is still scarce (4–6). A database has recently been constructed to provide information about the types of food packaging materials used (4). Further work along these lines is underway within the European “FACET” project (<http://www.ucd.ie/facet/>).

According to European Directive 2002/72/EC (7), verification of compliance with the specific migration limits (SMLs) may be ensured by determination of the quantity of a substance in the finished FCM, provided that a relationship between that quantity and the value of the specific migration of the substance has been established either by adequate experimentation or by the application of generally recognized diffusion models based on scientific evidence. However, in order to demonstrate the noncompliance of a food contact material, the estimated migration would need to be confirmed experimentally. In line with this, the Practical Guide (8) of the EU recommends the use of mathematical modeling of migration by the enforcement authorities as a tool to avoid long and expensive analysis.

FCM producers must be conscious that they are responsible for the safety of the product that they sell, and they must also be aware that for correct prediction of migration it is fundamental to know the composition of the FCM. In light of the advantages of mathematical modeling, several studies have developed different models and/or applied models to experimental results (6, 9–12). Some of these have evaluated the effectiveness of such models as tools for compliance testing (13–16). In one interesting study, the authors discuss how food inspectors in Denmark were in favor of

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**Table 1.** Migration Test Conditions

food item	storage temperature (°C)	storage time
wheat flour	25	4, 10, 30, 60, 180 d
	40	1, 2, 4, 7, 10 d
	70	8 h, 16 h, 1, 2, 4, 7, 10 d
rice	25	4, 10, 30, 60, 180 d
milk powder	40	1, 2, 4, 7, 10d
toast (4 and 23%)	25	1, 2, 4, 10, 20 d
	40	1, 2, 4, 7, 10 d
honey	25	1, 2, 4, 10, 20 d
wheat and rice starch	70	8 h, 16 h, 1, 2, 4, 7, 10, 20 d

**Table 2.** Nutritional Composition of the Selected Dry Foodstuffs

foodstuff	water (g/100 g)	protein (g/100 g)	carbohydrates (g/100 g)	fat (g/100 g)	mineral (g/100 g)
whole milk powder	2.5	25	35	26	7.0
skimmed milk powder	3.2	36	53	1.0	6.8
wheat flour	14	11	69	1.5	0.54
rice	13	6.8	78	0.62	0.53
honey	19	0.38	75	0.0	0.22

using migration modeling for compliance purposes, despite a lack of basic information such as the detailed composition of the materials (17).

Migration into dry foods was reported for migrants such as phthalates, diisopropyl-naphthalenes (DIPN) and certain volatile compounds (18). The aim of the present study was to explore the migration kinetics of an optical brightener, diphenylbutadiene (DPBD), from a FCM (LDPE) into different dry foodstuffs. DPBD was selected as model migrant. Foodstuffs were selected in order to represent a wide range of dry foods. Assays were performed at different times and temperatures in order to study actual and more extreme storage conditions. Key parameters of migration (diffusion and partition coefficients) were calculated according to a model based on Fick's second equation.

## MATERIALS AND METHODS

**Plastic.** The film used was a well-defined reference material for specific migration testing. It is a LDPE film (thickness 444  $\mu\text{m}$ ) spiked with fluorescent dye 1,4-diphenyl-1,3-butadiene (DPBD) and was produced by Fraunhofer IVV (Freising, Germany). The concentration of the migrant in the polymer ( $C_{P,0}$ ) was  $121.4 \text{ mg kg}^{-1} \pm 3.1\%$ . This value was derived from a trial certification exercise (19).

**Sampling.** Several dry foodstuffs were chosen for the study: honey, milk powder (whole and skimmed), wheat flour, rice and toast (containing 4 and 23% of fat). Crystalline casein, amorphous soy protein and wheat and rice starch were also included. Foodstuffs were bought in a local supermarket (honey, skimmed milk powder, toast containing 23% fat). The time and temperature chosen to perform the kinetic curves are summarized in **Table 1**. Three temperatures were selected for the assays: 25 °C, which corresponds to room temperature; 40 °C, which is the highest temperature to which foodstuffs are predicted to be exposed; and 70 °C, which is the temperature for accelerated assays. Samples were analyzed throughout several days, always in duplicate. The composition of samples is shown in **Table 2**, in accordance with the corresponding information provided on the label.

**Migration Tests.** Two methods were used to carry out migration tests, the glass washer method and the cell method (20, 21). Honey was subjected to the cell method because of its consistency. The amount of honey (approximately 10 g) that filled a cell was weighed accurately, and the cell was turned over and placed in contact with the plastic containing the DPBD (contact surface: 0.08  $\text{dm}^2$ ). Samples were stored under different conditions (see **Table 1**).

The amounts of all other foodstuffs (flour, rice, toast and milk powder) as well as casein, soy protein, wheat and rice starch required to fill a glass washer of area 0.1  $\text{dm}^2$  and height 0.8 mm were weighed accurately, and

the washers were then placed in contact with the plastic containing the DPBD (one side only). The samples were then wrapped in aluminum foil and placed inside a transparent plastic bag. The samples were packed under vacuum atmosphere to enable better contact between the sample and the plastic contaminated with DPBD, and then stored under different conditions (see **Table 1**). All analyses were conducted in duplicate.

**Chemicals and Standard Solutions.** All reagents were analytical grade. Ethanol, acetonitrile (ACN) and hexane were from Merck (Darmstadt, Germany). Ultrapure water was prepared with a Milli-Q filter system (Millipore, Bedford, MA). Diphenylbutadiene (DPBD) (CAS 538-81-8) was supplied from Aldrich (purity, 98%).

A primary stock solution of DPBD was prepared in ethanol (1.0  $\text{mg mL}^{-1}$ ). Intermediate standard solutions of DPBD were prepared in ACN and hexane (0.1–10.0  $\mu\text{g mL}^{-1}$ ). The solutions were stored in a refrigerator (4–10 °C) for up to 5 days.

**Sample Preparation.** Extraction was performed as follows (22, 23): 10  $\pm$  0.01 g of sample was extracted with 10 mL of hexane and shaken for 10 min. Organic phases were separated by centrifugation (1036g for 10 min). Extraction was repeated with 10 mL of hexane, and the supernatants were then pooled and evaporated in a rotary evaporator. The fatty liquid residue obtained was dissolved in 10 mL of ACN. Finally, the solution was filtered and a 50  $\mu\text{L}$  aliquot was injected in the HPLC.

**Chromatographic Conditions.** The HPLC system (Hewlett-Packard, Waldbronn, Germany) was fitted with a HP1100 quaternary pump, a degassing device, an autosampler, a column thermostating system and a diode array UV detector. HP ChemStation chromatographic software was used for data acquisition. Chromatographic separation was performed with a Kromasil 100 C18 column (15  $\text{cm} \times 0.4 \text{ cm i.d.}$ , 5  $\mu\text{m}$  particle size) (Teknokroma, Barcelona, Spain) at 30 °C.

A gradient elution method was used. Within the first 2 min the mobile phase was 65% ACN/35% water, after which the ACN was increased to 100% within 15 min. The total run time of each analysis was 30 min to clean the column. The flow rate was 1.0  $\text{mL min}^{-1}$ .

**Mathematical Modeling.** Migration of compounds from plastic packages into foodstuffs depends on many factors, but for a given migrant–polymer system and under controlled/fixed time/temperature conditions, migration greatly depends on the physicochemical characteristics of food, especially the fat content (24).

To assess migration of additives and contaminants from food-packaging films, mathematical modeling based on Fick's second law (eq 1) was used. This differential equation provides a general description of migration of an additive or contaminant from an amorphous polymeric packaging film:

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

where  $C_P$  ( $\text{mg kg}^{-1}$ ) is the concentration of the migrant in the polymer, P, at time  $t$  (s) and position  $x$  in P and  $D_P$  is the diffusion coefficient in P ( $\text{cm}^2 \text{ s}^{-1}$ ). Crank (25) solved the partial differential equation and formulated initial and boundary conditions, from which eq 2 was obtained as a solution. This equation describes the migration rate of a substance from a polymer P into a food F in contact with the polymer (26):

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

where  $m_{F,t}/A$  ( $\text{mg cm}^{-2}$ ) represents the amount of the migrated compound after the contact time  $t$  (s) of F with P. The contact area is  $A$  ( $\text{cm}^2$ ), the initial concentration of the migrant in P is  $c_{P,0}$  ( $\mu\text{g g}^{-1} = \text{mg kg}^{-1} = \text{ppm}$ ), the densities of P and F are  $\rho_P$  ( $\text{g cm}^{-3}$ ) and  $\rho_F$  ( $\text{g cm}^{-3}$ ), respectively, and the thickness of P is  $d_P$  (cm). The volumes  $V_P$  ( $\text{cm}^3$ ) and  $V_F$  ( $\text{cm}^3$ ) of polymer and food are used to calculate  $\alpha$ , as  $\alpha = (V_F/V_P)/K_{P,F}$ , where the partition coefficient  $K_{P,F} = c_{P,\infty} \rho_P / c_{F,\infty} \rho_F$  is the ratio of the migrant concentrations ( $w/v$ ) in P and F at equilibrium. The parameters  $q_n$  are the positive roots of the transcendent equation:  $\tan q_n = -\alpha q_n$ .

It is assumed that at the beginning of the mass transfer the migrant is homogeneously distributed in P and that there is no boundary resistance

**Table 3.** Diffusion Coefficients,  $\alpha$ ,  $K_{P/F}$  and RMSE Values for Different Foodstuffs

foodstuff	temp (°C)	$D$ (cm <sup>2</sup> /s)	$\alpha$	RMSE (%)	$K_{P/F}$	$K_{F/P}$
flour	25	$2.7 \times 10^{-10}$	0.52	2.10	4.00	0.250
	40	$4.5 \times 10^{-10}$	0.62	3.27	3.35	0.298
	70	$1.7 \times 10^{-9}$	0.67	4.15	3.10	0.323
rice	25	$3.4 \times 10^{-11}$	0.84	0.67	2.47	0.404
	40	$8.8 \times 10^{-11}$	1.40	1.13	1.48	0.674
powder milk	25	$3.2 \times 10^{-10}$	1.40	2.11	1.48	0.674
	40	$1.8 \times 10^{-9}$	0.80	0.44	2.60	0.385
toast 4% fat	25	$8.4 \times 10^{-11}$	0.40	1.22	5.19	0.193
	40	$1.6 \times 10^{-10}$	1.00	2.02	2.08	0.481
toast 23% fat	25	$8.1 \times 10^{-11}$	50.00	6.24	0.04	24.068
	40	$1.3 \times 10^{-9}$	3.00	13.86	0.69	1.444
wheat starch	70	$1.3 \times 10^{-10}$	0.05	0.44	41.55	0.024
rice starch	5	$2.7 \times 10^{-10}$	0.11	1.25	18.06	0.055

for the transfer between P and F. The migrant is homogeneously distributed in F, and the total amount of the migrant in P and F remains constant during the migration process.

Experimental data for different dry foodstuffs were fitted to eq 2, by nonlinear regression, with commercial software (Solver tool in Microsoft Excel 2003). From the series of experimental data on migration level ( $\mu\text{g dm}^{-2}$ ) plotted against time, the model parameters  $\alpha$  and  $D$  were calculated for each sample at different temperatures (see Table 3).

To measure the fit between experimental and estimated data, the % root of the mean-square error (% RMSE) was calculated as (27)

$$\text{RMSE (\%)} = \frac{1}{M_{P,0}} \sqrt{\frac{1}{N} \sum_{i=1}^N ((M_{F,t})_{\text{experimental},i} - (M_{F,t})_{\text{predicted},i})^2} \times 100 \quad (3)$$

where  $N$  is the number of experimental points per migration curve;  $i$  is the number of observations;  $M_{P,0}$  is the initial amount of migrant in the polymer ( $\mu\text{g}$ ).

The key parameters for determining the migration process are diffusion coefficient and  $K_{P/F}$ , where  $K_{P/F}$  corresponds to the relative solubility of the migrant at equilibrium between the plastic and the foodstuff (16, 28).

## RESULTS AND DISCUSSION

**Selection of the Foodstuffs.** Flour was chosen because it has high carbohydrate content and low water and fat contents, it is rich in protein, and it has high specific surface area. Milk powder is widely used for baby-food and has a high specific surface area and high fat content; lactose is the main carbohydrate in powdered milk, and it is presented in the amorphous form. Rice has a high carbohydrate content, it is rich in proteins and it has a low water content. Toast is representative of bakery products (bread) and has a porous structure. Honey is a semifluid natural product with very high carbohydrate content, low water content, no fat, no proteins, high viscosity and low pH (3.9). Crystalline casein, soy protein powder, wheat and rice starch were also analyzed, and represented protein and carbohydrate nutrients, in order to provide a better understanding of the food parameters/nutrients that have the greatest effects on migration.

**Migration into Dry Foodstuffs.** The EU conventional value as regards the relation for the plastic area in contact with food/food weight is  $6 \text{ dm}^2 \text{ kg}^{-1}$ . This value is obtained by assuming that a person weighing 60 kg consumes 1 kg of packaged food per day over a lifetime. Nevertheless when the additive is readily soluble in the food/simulant, it is considered acceptable to increase this ratio to improve the sensitivity of the analysis. In the present study, when the migration cell was used, the ratio was  $8.04 (0.0804 \text{ dm}^2 10 \text{ g}^{-1})$ , and when the glass washer method was applied, it was  $9.89 (0.0989 \text{ dm}^2 10 \text{ g}^{-1})$ . The ratio of volumes of liquid

(food) and package plays an important role when it is lower than 10 (29).

The EU legislation on FCM is based on the assumption that migration estimation should be conservative, and thus actual migration values should be overestimated to ensure consumer health. According to the current EU legislation the migration test at 40 °C for 10 days is the strictest for any foodstuff stored at room temperature and should thus yield the highest migration levels. However, the kinetics of migration for milk powder revealed higher migration levels in the samples incubated at 25 °C for 180 days than in those incubated at 40 °C for 10 days. This important result suggests the need for a critical review of the currently prescribed time/temperature conditions for migration tests. This key question about the suitability of food simulants for simulating foods has recently been addressed (30).

Results show that migration ( $M$ ) is negligible in honey and skimmed milk powder (always below the quantification limit) but not in the other foods. For similar time/temperature conditions, the migration obtained was in the following order:

$$M_{\text{whole milk powder}} > M_{\text{toast with fat}} > M_{\text{flour}} > M_{\text{rice}}$$

Whole milk powder and toast (with 23% fat) have the highest fat contents of the foodstuffs selected. It is therefore possible to conclude that fat content greatly affects the migration of DPBD in dry foods. However this is not the only factor that explains the migration process. In order to study the influence of other constituents and/or conditions on the migration level, accelerated tests (carried out at 70 °C) were performed with protein and starch.

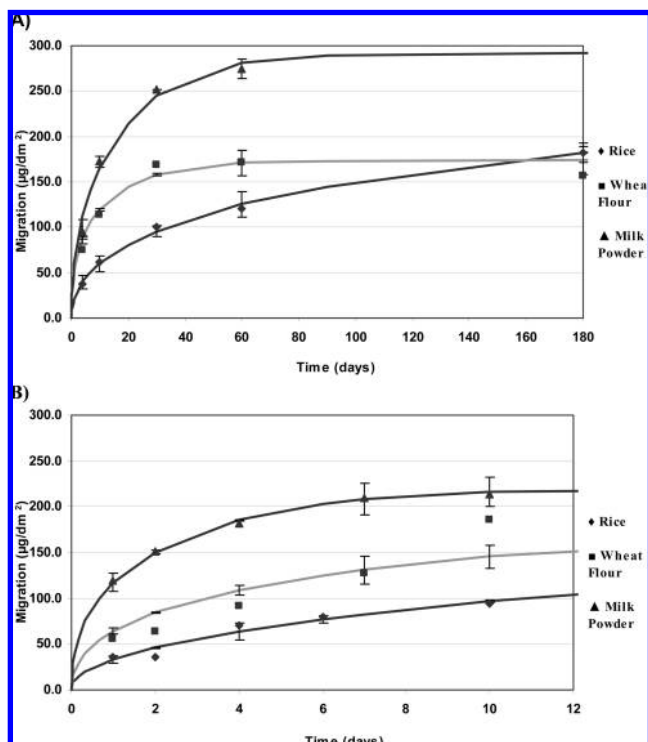
Two proteins were chosen, one in crystalline form (casein) and another in amorphous form (soy protein). The proposed idea was to test, assuming that the protein contributed to migration, whether the form (crystal or amorphous) may also influence the  $M$  levels. However, it was found that migration into both proteins was negligible (lower than  $10 \mu\text{g dm}^{-2}$  at 70 °C).

As regards starch, comparative assays were carried out with rice and wheat starches. Rice starch caused higher migration than wheat starch but always lower than  $57 \mu\text{g dm}^{-2}$  (value achieved after storage at 70 °C for 20 days, data not shown in Figure 4.) These results are not consistent with those found earlier for rice and flour, in which migration into rice was lower than in wheat flour. However other factors may also be involved, such as surface area. Flour and rice have a similar fat content, but rice has a much lower surface area. Migration may depend on the specific surface area, but with other factors involved.

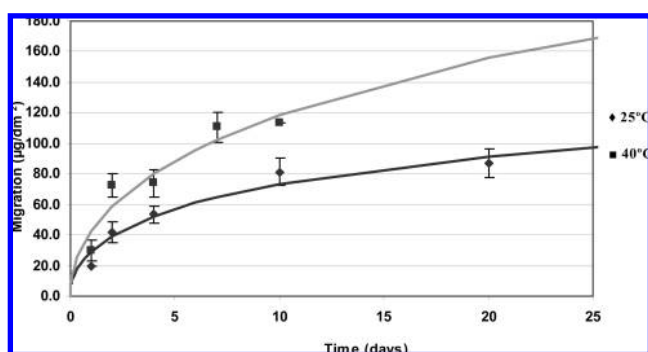
Triantafyllou and co-workers (18) carried out a study of the migration of several substances, namely *o*-xylene, acetophenone, *n*-dodecane, naphthalene, diphenyl ether, 2,3,4-trichloroanisole, benzophenone, diisopropylnaphthalenes, isomeric mixture (DIPN), dibutyl phthalate and methyl stearate, into dry foodstuffs such as semolina (1.9% fat), instant baby cream (13.5% fat), and infant whole milk powder (27.7% fat), and concluded that Tenax (a highly porous and adsorptive polymer considered suitable for use as a food simulant) was the most suitable food simulant for dry foods with low or intermediate fat content, such as semolina and instant baby cream. Moreover, dry food with a high fat content (e.g., whole milk powder) displayed higher migration levels. Powdered milk samples displayed higher benzophenone migration levels than Tenax because benzophenone is a fat soluble compound.

Migration it is not negligible in dry foods and appears to occur through fat and starch, and also depends on the specific surface area. Although specific surface area influences migration  $M$ , fat content appears to be the major contributor, for two reasons.





**Figure 1.** Migration of DPBD into rice, flour and milk powder at 25 °C (A) and 40 °C (B). Data points are mean values of two independent experiments  $\pm$  standard deviation.



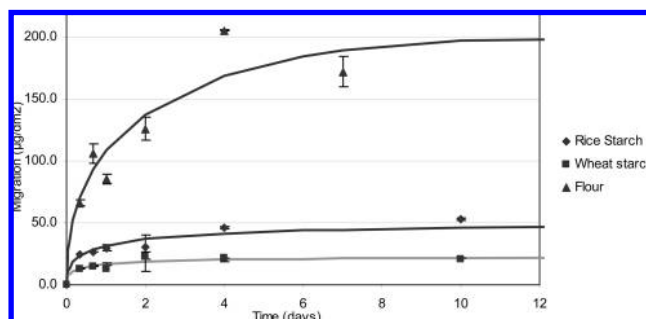
**Figure 2.** Migration of DPBD into toast containing 4% fat, at 25 and 40 °C. Data points are mean values of two independent experiments  $\pm$  standard deviation.

First, the specific surface area of flour is similar to that of milk powder, but because flour has a lower fat content, it causes less  $M$ . Second, skimmed milk powder displays negligible migration, although it has a similar surface area to flour. The influence of carbohydrates on the migration process has already been demonstrated by Sanches-Silva et al. (31). Figure 1 shows the influence of toast fat on the migration of DPBD.

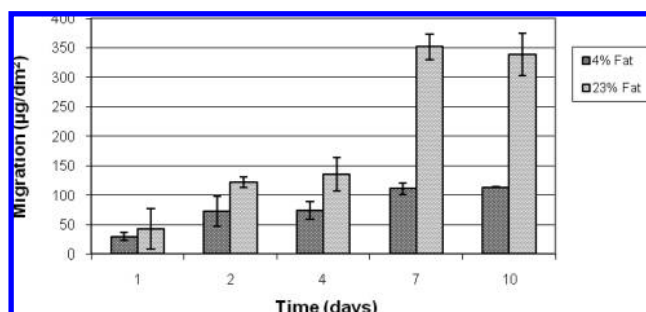
As regards the influence of temperature on the migration process, as expected, higher migration levels were also found at higher temperatures.

**Key Migration Parameters.** The diffusion coefficients and partition coefficients were calculated according to eq 2. The results are summarized in Table 3. The fit of experimental and predicted kinetic migration curves was measured with eq 3, and the % RMSE was always lower than 4%, except for toast with 23% fat.

The measured values and the estimated migration curve for DPBD in rice, flour and milk powder, as a function of time, are



**Figure 3.** Migration of DPBD into flour, rice and wheat starch at 70 °C. Data points are mean values of two independent experiments  $\pm$  standard deviation.



**Figure 4.** Influence of toast fat content on the migration of DPBD. Data points are mean values of two independent experiments  $\pm$  standard deviation.

shown in Figure 2. The corresponding values for toast containing 4% fat are shown in Figure 3. The measured and estimated values obtained for flour, rice and wheat starch at 70 °C are shown in Figure 4.

At 180 days very high migration was observed in milk powder, close to the maximum migration level for this film ( $491 \mu\text{g dm}^{-2}$ ). This value was not taken into consideration to calculate the diffusion coefficient. This may be an outlier value, or the two different types of migration kinetics may occur in milk powder.

At 25 °C, the diffusion coefficient  $D$  presented the following order:  $D_{\text{milk powder}} > D_{\text{flour}} > D_{\text{toast}} > D_{\text{rice}}$ , although  $D_{\text{milk powder}}$  and  $D_{\text{flour}}$  are values of similar magnitude. At 40 °C,  $D_{\text{milk powder}} > D_{\text{toast, 23% fat}} > D_{\text{flour}} > D_{\text{toast, 4% fat}} > D_{\text{rice}}$ .

This order is consistent with the migration levels. The  $D$  values for DPBD have previously been calculated at 25 °C for chocolate ( $2.9 \times 10^{-10} \text{ cm}^2 \text{ s}^{-1}$ ) (32), for margarine containing 61% fat ( $5.1 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$ ) (32), margarine containing 80% fat ( $3.7 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$ ) (32), pork meat ( $1.88 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$ ) (20), and orange juice ( $2.9 \times 10^{-12} \text{ cm}^2 \text{ s}^{-1}$ ) (31). The  $D$  value for chocolate at 25 °C is similar to the  $D$  value for milk powder and flour at the same temperature. At this temperature,  $D$  values for pork and margarines are higher than those achieved for any of the dry foodstuffs considered in the present study and the  $D$  value for orange juice is much lower.  $D$  values of DPBD have also been calculated for cheeses, although only at 5 °C (21): soft cheese ( $3.2 \times 10^{-11} \text{ cm}^2 \text{ s}^{-1}$ ), cottage cheese ( $1.12 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$ ) and Gouda cheese ( $6.34 \times 10^{-11} \text{ cm}^2 \text{ s}^{-1}$ ).

The diffusion coefficient for DPBD in rice and flour was calculated in a previous study (33). At 25 °C this  $D$ , calculated from Moissan's equation was  $7.1 \times 10^{-8} \text{ cm}^2 \text{ s}^{-1}$  and  $4.9 \times 10^{-8} \text{ cm}^2 \text{ s}^{-1}$  for flour and rice respectively. At 40 °C the  $D$  value was  $1.6 \times 10^{-7} \text{ cm}^2 \text{ s}^{-1}$  and  $2.0 \times 10^{-7} \text{ cm}^2 \text{ s}^{-1}$  for flour and rice respectively.

Because the effective diffusion coefficient of DPBD at the same temperature estimated in the whole system of LDPE + dry foodstuff (rice or wheat flour) is higher than the diffusion coefficient of DPBD at the same temperature with the same dry foodstuff, we can conclude that the mass transport step from the polymer surface to the food is the rate limiting step.

According to Stoffers (34),  $D$  DPBD at 60 °C into ethanol 95% is  $1.2 \times 10^{-8}/3.1 \times 10^{-8} \text{ cm s}^{-1}$ .  $D$  ranged between  $1.1 \times 10^{-8}$  and  $7.1 \times 10^{-8} \text{ cm s}^{-1}$  for the 4 laboratories at 40 °C into 95% ethanol. For accurate values of the partitioning coefficients  $K_{P/F}$ , it is necessary to have data points close to the partitioning equilibrium, which depends on the duration of the migration experiments (34).

The partition coefficient ( $K_{P/F}$ ) was calculated from the  $\alpha$  value and polymer and food volumes. The  $V_P$  for all assays was  $0.439 \text{ cm}^3$ , and the  $V_F$  was  $7.912 \text{ cm}^3$ . The  $K_{P/F}$  values are also shown in Table 3.

The Arrhenius relationship was calculated for wheat flour with the equation

$$D = D_0 \exp\left(-\frac{E_a}{RT}\right) \quad (4)$$

where  $D$  is the diffusion coefficient,  $D_0$  is a constant,  $E_a$  is the activation energy,  $R$  is the universal gas constant ( $8.314 \times 10^{-3} \text{ kJ mol}^{-1} \text{ K}^{-1}$ ), and  $T$  is the temperature (in kelvins).

Higher migration levels were found at higher temperatures and an Arrhenius-type relationship was found for the  $D$  of DPBD in flour.  $D_0$  is  $3.86 \times 10^{-4} \text{ cm}^2 \text{ s}^{-1}$ ,  $E_a$  is  $35264.12 \text{ kJ mol}^{-1}$  and  $r^2 = 0.99$ . The Arrhenius equation can predict  $D$  for any temperature between 25 and 70 °C.

As demonstrated, migration into dry foodstuffs is not always negligible. Consequently, migration testing of FCM in contact with dry foodstuffs should be considered for compliance evaluation.

Results from starch and protein assays also enable us to conclude that migration of DPBD through protein matrices is apparently not feasible. The most surprising result was the migration into rice and flour, both of which have low fat contents. In these cases the results indicate that diffusion of migrants occurs through starch and fat.

The data obtained in the present study are only valid for DPBD, but they confirm the validity of the mathematical model, which will predict the migration of chemicals from packaging into foodstuffs. The mathematical modeling of migration, which has already become a practical tool for both producers of plastic materials and food inspectors, will increase further in importance in this area.

On the basis of mathematical modeling, manufacturers of plastic packaging materials, who generally know the composition of the materials, can adjust the composition and structure of their final materials taking into account the intended food packaging application. They will thus be better able to comply with the regulations without the need for expensive and time-consuming tests in the chemical enforcement laboratories. Finally, food inspectors can also easily confirm the safety of food contact materials.

## ACKNOWLEDGMENT

The authors thank Ms. Patricia Blanco Carro and Mr. Gonzalo Hermelo Vidal for their excellent technical assistance.

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Received for review May 19, 2009. Revised manuscript received October 6, 2009. Accepted October 06, 2009. The study was financially supported by the Xunta de Galicia (Project No. INCITE 08PXIB203096PR) and the European “FOODMIGROSURE” project. The authors are grateful to the “Fundação para a Ciência e Tecnologia”, Portugal, for a postdoctoral contract awarded to Ana Sanches Silva, to the “Science 2007” Programme, and to the “Angeles Alvariño” Programme financed by the “Consellería de Innovación e Industria, Xunta de Galicia” for a postdoctoral contract awarded to R. Sendón.