A new method for the prediction of diffusion coefficients in poly(ethylene terephthalate)—Validation data

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Prediction of the migration is a useful tool in compliance evaluation of food contact materials. In our previous work, such a prediction model had been established for polyethylene terephthalate (PET), which is widely used as packaging material for beverages as well as for meat and cheese. Within the actual study, 263 diffusion coefficients in PET for 66 substances at temperatures between 40°C and 120°C were determined from permeation kinetic experiments. The diffusion coefficients \( D_p \) were compared with the predicted values by use of a log/log plot as well as in direct comparison of the diffusion coefficients. When applying the migration prediction model for compliance evaluation of food packaging materials, it is mandatory that the migration prediction is over-estimative in any case. As a result, the predicted values are in good agreement with the experimental results. The prediction model slightly overestimates the real migration by a factor of 1.3 in average. Reduction of the molecular volume of 20% results in an average over-estimation of 3 of the migration (worst-case). Another finding of this study is that the diffusion behaviour at the glass transition temperature not significantly change. The prediction model is applicable below and above the glass transition temperature.

KEYWORDS
diffusion coefficients, diffusion modelling, permeation kinetics, polyethylene terephthalate

1 | INTRODUCTION

The prediction of the migration is a useful tool in compliance evaluation of food contact materials, especially for packaging polymers with a low diffusion. For low diffusive polymers, the migration process is very time consuming and result at the end in low concentrations in food that challenges the analytical approaches. Increasing the temperature is one of the approaches to increase the speed of the mass transfer. However, higher temperatures like 60°C are far away from realistic storage conditions, which is in most cases room temperature. One of these low-diffusive polymers for which migration prediction is important in food law compliance evaluation is polyethylene terephthalate PET. PET is mostly used for beverage packaging, but also for trays for meat and cheese.

The most important factors influencing the mass transfer from the packaging material into food is the concentration of the migrant in the polymer, the storage time, the storage temperature, the diffusion coefficients \( D_p \), and the partition coefficient \( K_{P/F} \). Other factors are the surface volume ratio and the film thickness of the packaging material. Most of the abovementioned factors are known or analytically available. Only the diffusion coefficients \( D_p \) and the partition coefficients \( K_{P/F} \) are rarely available from the scientific literature. However, for low-diffusive polymers like PET, the partition coefficients are negligible because the equilibrium between the polymer and the food will...
not be reached under normal storage conditions of packed foods. Therefore, the diffusion coefficient $D_P$ is the most important factor in the prediction of the migration for low-diffusive polymers like PET.

Predictive models for the migration from food packaging mater-
ials have been developed with the last 25 years.\textsuperscript{1–5} A comprehensive review on the different approaches is given in the scientific literature.\textsuperscript{6} These prediction models should be over-estimative to show the food regulatory compliance with a safety factor.\textsuperscript{7} Regulation 10/2011 stated that “To screen for specific migration the migration potential can be calculated on the residual content of the substance in the material or article applying generally recognized diffusion models based on scientific evidence that are constructed in a way that must never underestimate real migration levels.”\textsuperscript{8} However, the prediction should be not too over-estimative; otherwise, compliance with current food law cannot be shown in some cases. Therefore, realistic but still over-estimative diffusion coefficients $D_P$ should be available or should be predicted.

In our previous work, a prediction model for the diffusion coefficients in PET was developed.\textsuperscript{9} This prediction model is based on the molecular volume $V$ of the substances and the temperature in Kelvin (Equation 1). The prediction model was derived from two correlations: (i) a correlation between the activation energy $E_A$ and the molecular volume $V$ of the substance and (ii) a correlation between the activation energy $E_A$ and the pre-exponential factor $D_0$ of the Arrhenius equation.\textsuperscript{9} The activation energy of diffusion $E_A$ and the pre-exponential factor $D_0$ were experimentally determined mainly from desorption kinetics of spiked PET sheets into the gas phase.\textsuperscript{10} The parameters $a$ and $b$ are the slope and the intercept of the correlation between the activation energy $E_A$ and the pre-exponential factor $D_0$. The parameters $c$ and $d$ are the intercept and the slope of the correlation between the activation energy $E_A$ and the molecular volume $V$. The parameters $a$ to $d$ for the prediction of diffusion coefficients $D_P$ in PET are given Table 1.

\[
D_P = b \left( \frac{V}{c} \right)^{\frac{a}{d}}
\]  
\textbf{(1)}

Due to the fact that PET is a low-diffusive polymer, high tempera-
tures have to be applied for the desorption kinetics in our previous study. Therefore, most of the activation energies of diffusion $E_A$ were derived from diffusion coefficients $D_P$ above the glass transition temperature $T_g$.\textsuperscript{10} Also the activation energies of an alternative prediction model are determined mainly above $T_g$.\textsuperscript{3} On the other hand, the low diffusion of PET is also responsible that at temperatures below the glass transition temperature only the diffusion coefficients for small molecules are available.\textsuperscript{11–13} As a consequence, only a limited amount of diffusion coefficients are available in the scientific literature which can be used for validation purposes of Equation 1.

Aim of the study was the determination of diffusion coefficients $D_P$ in PET which should be used to validate the prediction of diffusion coefficients from Equation 1 with the parameters from Table 1. The diffusion coefficients should be determined with another method not used in our previous study in order to exclude possible artefacts from the high temperature desorption method. These independent diffusion coefficients should be compared with the predicted values from Equation 1.

\section{RESULTS}

Within this study, a permeation method was applied to determine the diffusion coefficients $D_P$ for several substances in PET. From the experimental permeation data, the diffusion coefficients $D_P$ were derived from the so-called lag time according to Equation 2, where $l$ is the thickness of the PET film. This method was applied in previous studies for the determination diffusion coefficients in PET,\textsuperscript{14} polyamide PA6,\textsuperscript{15} polyethylene naphthalate PEN,\textsuperscript{16} and general purpose polystyrene GPPS.\textsuperscript{17} However, in these studies, the permeants were limited to homologous rows of $n$-alkanes and $1$-alcohols. In the actual study, we determined also other substances than $n$-alkanes and $1$-alcohols towards their permeation through a thin PET film and derived diffusion coefficients for a broad range of substances and functional groups.

\[
\text{lag time} = \frac{L^2}{6D_P}
\]  
\textbf{(2)}

The permeants used within this study are summarized in Table 2. The substances cover various molecular volumes $V$, functional groups, and therefore also polarities. Overall, 66 substances were tested at 14 temperatures between 40°C and 120°C. In addition, the diffusion coefficients were determined also at different gas phase concentrations in the lower space of the permeation cell. The gas phase concentrations used for the permeation tests are far below the saturation vapour pressure at each temperature. The applied low concentrations avoids swelling of the PET material. Swelling of the polymer might lead to higher diffusion coefficients. The gas phase concentrations in the lower space of the permeation cell are given in the supporting information together with the corresponding diffusion coefficients $D_P$.

As a result, overall 263 diffusion coefficients $D_P$ were experimen-
tally determined from these lag time experiments. The diffusion coeffi-
cients were compared with the predicted values from Equation 1 with the parameters given in Table 1. This comparison is visualized in Figure 1 for each of the tested temperatures between 40°C and
**TABLE 2** Substances used in the permeation experiments, their molecular weight volume

| Substance             | CAS no. | Molecular weight (g/mol) | Molecular volume (Å³) | Temperature range (°C) | Amount of diffusion coefficients measured |
|-----------------------|---------|--------------------------|-----------------------|------------------------|-------------------------------------------|
| Ethane                | 74-84-0 | 30.1                     | 45.76                 | 70                     | 1                                         |
| n-Propane             | 74-98-6 | 44.1                     | 62.56                 | 70–120                 | 7                                         |
| Acrylonitrile         | 107-13-1| 53.1                     | 57.23                 | 40–80                  | 4                                         |
| Acetone               | 67-64-1 | 58.1                     | 64.74                 | 70–100                 | 6                                         |
| n-Butane              | 106-97-8| 58.1                     | 79.36                 | 70–120                 | 7                                         |
| Methyl formate        | 107-31-3| 60.1                     | 57.16                 | 70–100                 | 5                                         |
| 1-Propanol            | 71-23-8 | 60.1                     | 70.82                 | 80–95                  | 4                                         |
| Pyrrole               | 109-97-7| 67.1                     | 69.03                 | 70–100                 | 7                                         |
| Cyclopentane          | 287-92-3| 70.1                     | 85.80                 | 90–110                 | 5                                         |
| 2-Butanone            | 78-93-3 | 72.1                     | 81.54                 | 70–110                 | 10                                        |
| Tetrahydrofuran       | 109-99-9| 72.1                     | 77.98                 | 90–95                  | 2                                         |
| n-Pentane             | 109-66-0| 72.2                     | 96.16                 | 70–100                 | 7                                         |
| 1-Butanol             | 71-36-3 | 74.1                     | 87.62                 | 80–100                 | 6                                         |
| 1,3-Dioxolane         | 646-06-0| 74.1                     | 70.17                 | 90–100                 | 3                                         |
| Ethyl formate         | 109-94-4| 74.1                     | 73.97                 | 60–100                 | 7                                         |
| Benzene               | 71-43-2 | 78.1                     | 84.04                 | 50–100                 | 11                                        |
| Pyridine              | 110-86-1| 79.1                     | 79.89                 | 70–100                 | 7                                         |
| Cyclopentanone        | 120-92-3| 84.1                     | 87.98                 | 90–115                 | 6                                         |
| Cyclohexane           | 110-82-7| 84.2                     | 102.60                | 95–105                 | 3                                         |
| Dichloromethane       | 75-09-2 | 84.9                     | 56.51                 | 40–50                  | 2                                         |
| 2-Pentanone           | 107-87-9| 86.1                     | 98.34                 | 80–100                 | 5                                         |
| n-Hexane              | 110-54-3| 86.2                     | 112.96                | 70–100                 | 14                                        |
| 1,4-Dioxane           | 123-91-1| 88.1                     | 86.97                 | 90–115                 | 5                                         |
| 2-Methyl-1,3-dioxolane| 497-26-7| 88.1                     | 86.75                 | 90–110                 | 4                                         |
| n-Propyl formate      | 110-74-7| 88.1                     | 90.77                 | 60–100                 | 8                                         |
| 1-Pentanol            | 71-41-0 | 88.2                     | 104.42                | 80–100                 | 6                                         |
| Toluene               | 108-88-3| 92.1                     | 100.61                | 75–100                 | 11                                        |
| Aniline               | 62-53-3 | 93.1                     | 95.33                 | 75–100                 | 6                                         |
| 1,1-Dichloroethene    | 75-35-4 | 96.9                     | 67.44                 | 40–80                  | 6                                         |
| Cyclohexanone         | 108-94-1| 98.1                     | 104.79                | 100–115                | 4                                         |
| 1,1-Dichloroethane    | 75-34-3 | 99.0                     | 73.09                 | 60–80                  | 4                                         |
| 1,2-Dichloroethane    | 107-06-2| 99.0                     | 73.31                 | 75–90                  | 5                                         |
| n-Heptane             | 142-82-5| 100.2                    | 129.77                | 80–100                 | 6                                         |
| 2-Hexanone            | 591-78-6| 100.2                    | 115.15                | 80–100                 | 5                                         |
| n-Butyl formate       | 592-84-7| 102.1                    | 107.57                | 70–100                 | 7                                         |
| 1-Hexanol             | 111-27-3| 102.2                    | 121.22                | 80–100                 | 6                                         |
| Ethylbenzene          | 100-41-4| 106.2                    | 117.41                | 80–100                 | 5                                         |
| 2,6-Lutidine          | 108-48-5| 107.2                    | 113.01                | 75–100                 | 6                                         |
| 4-Vinylcyclohexene    | 100-40-3| 108.2                    | 124.17                | 95–110                 | 3                                         |
| 2-Heptanone           | 110-43-0| 114.2                    | 131.95                | 85–100                 | 4                                         |
| n-Octane              | 111-65-9| 114.2                    | 146.57                | 85–110                 | 7                                         |
| 1-Heptanol            | 111-70-6| 116.2                    | 138.03                | 90–100                 | 4                                         |
| n-Pentyl formate      | 638-49-3| 116.2                    | 124.37                | 90–100                 | 4                                         |
| 2-Aminobenzonitrile   | 1885-29-6| 118.1                   | 112.19                | 75–100                 | 6                                         |
| Chloroform            | 67-66-3 | 119.4                    | 70.07                 | 60–90                  | 5                                         |
| n-Propylbenzene       | 103-65-1| 120.2                    | 134.21                | 85–100                 | 5                                         |
120 °C. As a result, the predicted diffusion coefficients (solid line in Figure 1) is in good agreement with the experimentally determined diffusion coefficients.

3 DISCUSSION

Within the study, 263 diffusion coefficients of various substances in PET were determined and compared with the predicted values from Equation 1 and the parameters given in Table 1. The experimental diffusion coefficients were good agreement with the predicted values. Only 22 diffusion coefficients were above the predicted diffusion coefficients from Equation 1 (8.4%). The biggest difference was found for acrylonitrile at 40 °C, where the predicted diffusion coefficient was only a factor of 0.44 lower than the experimental diffusion coefficient. The diffusion coefficients of 241 substances were predicted equal or higher as the experimental diffusion coefficients. The average over-estimation factor was 2.6 (median 2.3) compared to the experimental diffusion coefficients. This means the modelling parameters from Table 1 slightly over-estimates the migration in average. However, when using prediction models for compliance evaluation, the migration should be over-estimative in any case.8

From the results of the correlation between the activation energy of diffusion $E_A$ and the molecular volume $V$, a reduction of 20% of the molecular volume of the diffusant is proposed to create this over-estimation. This results in a virtual molecule with a higher predicted diffusion coefficient (worst-case). These worst-case diffusion coefficients derived from the virtual volume reduction of 20% are given in Figure 1 as dotted lines. As a result, all experimental diffusion coefficients are below the worst-case predicted diffusion coefficients when applying the virtual volume V-20%. In average, the over-estimation factor is 9.2 (median 7.7) for the 263 diffusion coefficients. From the results for acrylonitrile at 40 °C, it can be concluded that a volume reduction of only 9.4% is sufficient to predict equal or higher diffusion coefficients as the experimental data. Using 20% reduction in the molecular volume results in an additional safety factor compared to 9.4%.

On the other hand, in some case the predicted diffusion coefficients should be lower than the experimental values. For example in the food law compliance evaluation of repeated-use food contact articles. In the evaluation of repeated-use articles the third migration contact is evaluated in experimental migration tests. As a consequence, in the evaluation of repeated-use articles the migration prediction models need to be under-estimative, otherwise the third contact will have a too low predicted migration value. To predict always lower diffusion coefficients the volume of the molecule should be virtually raised by 60.5%, which was calculated from the experimental diffusion coefficient of cyclopentane at 90 °C.

Figure 2 show the log/log plot of the experimental diffusion coefficients and the predicted diffusion coefficients according to Equation 1 with the modelling parameters given in Table 1. The slope
of the plot is 0.99 which is very close to the ideal slope of 1.0 and the intercept of 0.2 is also very close to zero. This indicates the prediction of the diffusion coefficient is close to the experimental results. The dotted lines show the 95% confidence interval of the correlation, which means the 95% of the values are in between both dotted lines.
It should be noted here that the majority of the diffusion coefficients are determined above the glass transition temperature. A small portion of diffusion coefficients was determined below $T_g$ (33 diffusion coefficients). The results of this study show that PET is a low diffusion polymer. Measurable diffusion coefficients are only available for low molecular weight molecules, by increasing the temperature and/or in case of permeation tests with thin films. For the tests, a thin 12-$\mu$m PET film was chosen. With thinner films also diffusion coefficients of higher molecular weight substances might be available at temperatures below the glass transition temperatures, but such thinner PET films are not available or cannot be handled because they are too thin. The molecular weight range of the permeants was tried to expand the molecular weight range as much as possible. However, the boundaries are very narrow. Therefore, diffusion coefficients below $T_g$ are only available for low molecular weight substances.

It is important to note that all diffusion coefficients $D_p$ are derived from permeation kinetics into the gas phase. Interactions between the PET polymer and food (simulants) like swelling are therefore excluded. The diffusion coefficients derived from this method can be considered as the pure diffusion coefficients in PET. Swelling effects of food simulants on the PET surface are well-known especially at high temperatures of 60°C, which is mostly used for PET beverage bottle testing.\textsuperscript{12,18,19} These swelling effects significantly increases the migration into the simulants and lead in some cases to migration levels that exceed the specific migration limits. On the other hand, real foods did not significantly swell PET and lead to much lower migration limits. Diffusion modelling is therefore more suitable to realistically predict the migration into food at the end of shelf life compared to experimental migration tests using high ethanolic food simulants.\textsuperscript{19}

It is important to note, that also moisture can swell the PET polymer especially at high temperatures. In order to reduce these swelling effects, the diffusion coefficients in this study were determined at virtually zero levels. Therefore, the pure diffusion coefficients in the PET polymer were determined without any swelling effects. Regarding real application, foods in contact with PET include high moisture conditions, but only at low temperatures. At low temperatures swelling is negligible for moisture but are significant for high ethanolic simulants.\textsuperscript{12,18,19}

As mentioned above, most of the diffusion coefficients were determined above the glass transition temperature $T_g$ of 81°C. At the glass transition temperature $T_g$, the diffusion behaviour might change. In order to investigate this effect, the diffusion coefficients derived from this study were compared with previous studies. For four permeants, diffusion coefficients were available in the scientific literature from desorption experiments at temperatures between 120°C and 180°C\textsuperscript{20} and from migration kinetics into mineral water and 10% ethanol at temperatures between 23°C and 50°C.\textsuperscript{12,13} The permeation experiments of this study are in between the temperature intervals. The results are visualized in Figure 3 with the correlation of the reciprocal temperature (in Kelvin) and the logarithm of the diffusion coefficient $D_p$ (Arrhenius plot). The results for these four substances show, that at the glass transition temperature $T_g$ no significant change of the diffusion behaviour of PET was detectable. In addition, diffusion coefficients predicted from three independent methods (desorption kinetics,\textsuperscript{20} migration kinetics\textsuperscript{12,13} and permeation kinetics [this study]) for the determination of diffusion coefficients are in good agreement with each other.

4  |  MATERIALS AND METHODS

4.1  |  PET film sample and model chemicals

For the permeation tests, a commercial biaxially oriented PET film was used. The thickness was determined to 11.9 ± 0.1 μm. The glass transition temperature of the investigated PET film was determined by
differential scanning calorimetry (DSC) to 81 °C. The melting range was also determined by DSC to 243–260 °C (peak at 255 °C). The permeation was tested with 66 different substances (Table 2). The substances were purchased with a purity of 99% and used without further purification. The concentration applied in the lower space of the permeation cell are given in the supporting information together with the corresponding diffusion coefficients.

4.2 | Permeation experiments

The 12-μm PET film was placed in a permeation cell with a lower and an upper space separated by the investigated PET film. The permeation cell was made of aluminium and had an area of 191 cm². On both sides, a sealant ring is embedded into the aluminium cell. The PET film is clamped between both sealant rings. The lower space of the permeation cell with a volume of 7437 cm³ was spiked with the permeants. The upper side of the cells was rinsed with pure nitrogen. The constant nitrogen flow moved the permeated substances out of the cell. The nitrogen stream was analysed for the applied permeants by a connected enrichment unit and gas chromatograph with flame ionization detection (GC/FID). Permeation was measured at constant temperatures depending on the permeants in a climate chamber between 40 °C and 120 °C. From these experimental data the permeation rates in μg per cm² and d of the applied permeants are available. Calibration was performed with injections of known amounts of the applied permeants. Several gas chromatographic conditions and columns were applied according to the permeants measured.

4.3 | Molecular volume

The molecular volume $V$ of the molecules was calculated with the free internet program molinspiration.21

5 | CONCLUSIONS

The modelling parameters derived from our previous study9 were validated with 263 diffusion coefficients from 66 different organic substances.
substances with various functional groups, volatility, and polarity. The parameters given in Table 1 in combination with Equation 1 predict the diffusion coefficients $D_P$ of organic substances in PET with a slight average over-estimative factor of 2.6. When using the molecular volume $V$ of the substances minus 20% the modelling parameters are in any case over-estimative which is mandatory for compliance evaluation of food packaging materials according to European Regulation 10/2011. This worst-case prediction results in an average over-estimative factor of 9.2 in the diffusion coefficients. Increasing the diffusion coefficient $D_P$ by a one order of magnitude (factor 10) results in an increase of the migration by a factor of 3.16 (square root of 10). An average over-estimation factor of 9.2 results in a factor of 3.0 higher migration, which seems to be suitable for food law compliance evaluation purposes. The average over-estimation without virtual volume reduction is 1.6, which results in an over-estimation of the migration compared to the predicted migration of only 1.3. The parameters in Table 1 therefore realistically describe the migration from PET. Such a realistic migration prediction is useful for the evaluation of non-intentionally added substances as well as in the evaluation of post-consumer PET recyclates in direct food contact applications.25

Another important finding of this study is, that the diffusion behaviour at the glass transition temperature do not significantly change. Even if the most of the diffusion coefficients were derived above the glass transition temperature the diffusion coefficients $D_P$ can be predicted below the glass transition temperature. On the other hand, assuming that there is a slight change, the diffusion behaviour will be probably higher above the glass transition temperature as below. Therefore, the modelling parameters given in Table 1 can be considered as worst case for temperatures below the glass transition temperature $T_g$ of PET.

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CONFLICT OF INTEREST
The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS
Conceptualization, F.W.; methodology, J.E.; validation, F.W.; writing - original draft preparation, F.W.; review and editing, J.E. Both authors have read and agreed to the published version of the manuscript.

DATA AVAILABILITY STATEMENT
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

Additional supporting information may be found in the online version of the article at the publisher's website.

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