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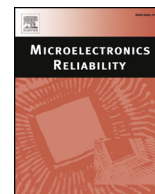
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Effect of temperature and humidity on moisture diffusion in an epoxy moulding compound material

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ABSTRACT

In this paper we propose a new multistep characterisation method to be able to map out the dependency of moisture diffusion parameters of a polymeric material over a range of temperature and humidity conditions in a limited amount of time. We do that by (1) using a moisture sorption analyser which can continuously monitor weight changes with microgram accuracy, (2) using thin samples which speeds up the diffusion process and (3) already switch to the next humidity level at 90 or 95% completion of a diffusion step. A multistep diffusion model was developed to account for the overlapping diffusion steps. This model showed to be extremely accurate for fitting experiments consisting of five absorption and one desorption steps. We show that for temperatures up to 85 °C and humidity level between 0 and 85% RH the diffusion of our material was essentially Fickian with a diffusion coefficient ranging from 3.8×10^{-7} mm²/s at 20 °C to 3.6×10^{-6} mm²/s at 85 °C. The moisture saturation concentrations showed a slightly non-linear variation with the applied humidity level.

1. Introduction

Electronic devices are protected by encapsulating them in highly filled epoxy moulding compound materials (EMCs). However, because of their polymeric nature, these materials are hygroscopic and will absorb environmental moisture which can be detrimental to the product performance and reliability. Absorbed moisture can for example lead to swelling and local stresses with magnitudes which are comparable to thermal stresses and which may eventually lead to cracking [1]. In addition, absorbed moisture may lead to electrochemical migration and corrosion as well as popcorn failure [2]. The understanding of moisture absorption and its related failure mechanisms is important for example for applications in the automotive industry in which under-the-hood electronic components are alternatively exposed to high humidity environments in which they absorb moisture and high temperatures where the moisture suddenly is driven out.

Good quantitative data of moisture diffusion for moulding compounds is crucial for moisture related failure predictions but is often lacking due to the wide variety of available materials and the extensive experimental work required for the testing. Simply assuming a value for the diffusion parameters based on published data for similar materials may lead to large over or underestimations of the actual diffusion process since the diffusion rate can vary as much as a factor 10 to 30

depending on the material, temperature and relative humidity. In current practice the moisture absorption of a moulding compound is determined by periodically weighing the mass increase of a sample which is stored in a fixed temperature and humidity environment. Such a measurement takes about 1 week for a single temperature and humidity combination, meaning that establishing the diffusion characteristics for a range of temperature and humidity levels would take months. In the current paper we present a new and relatively quick procedure for testing the diffusion parameters of a moulding compound over a range of temperatures and relative humidities.

Typical values for the diffusion coefficient of water in unfilled epoxies vary from about 1×10^{-7} mm²/s at 20 °C to 1×10^{-6} mm²/s at 75 °C [3–5]. The diffusion coefficients for moulding compounds at 85 °C range from 2 to 6×10^{-6} mm²/s [6–8] and are thus of the same order of magnitude as those for unfilled epoxies. Moisture diffusivity above 100 °C can be determined with a desorption experiment using a Thermal Gravimetric Analyser (TGA). However, outgassing of solvents and moisture loss during heating up may severely complicate this technique [7]. Fan [8] reported diffusion coefficients during desorption of saturated moulding compounds ranging from 1.6×10^{-6} mm³/s at 100 °C to 2.03×10^{-4} mm³/s at 260 °C.

In literature about moisture diffusion in polymers anomalous effects are frequently reported [9]. Many authors report that the initial Fickian

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absorption is followed by a second, slower stage [1,10–12]. Bao [10] explains this by the plasticizing effect of absorbed water molecules. Desorption and re-absorption experiments show that this effect is irreversible, unless the polymer is rejuvenated by heating above its glass transition temperature. Others [13] attribute these irreversible effects to micro damage induced by the plasticisation process.

2. Moisture diffusion theory

The modelling of moisture diffusion in polymers has been the subject of a large number of studies (e.g. [7,14,15]). Here we first discuss the basics of Fickian diffusion and then briefly discuss the most important observed deviations from this theory. In the last part of this section we discuss how the weight gain due to multiple Fickian diffusion steps can be modelled.

2.1. Standard diffusion theory

The moisture uptake in a flat sample can be described with the one dimensional diffusion equation with diffusivity, D (in mm^2/s) and concentration C (in mg/mm^3).

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (1)$$

If a sample is exposed to an environment with a relative humidity level RH , the moisture is first saturating the surface layers, resulting in a surface concentration C^{sat} . During a diffusion experiment with constant humidity setting the moisture slowly penetrates in the sample but the surface layer concentration remains stable at C^{sat} . With this as the boundary conditions and a uniform concentration C_0 as the initial condition, the general solution for the diffusion equation is given as [15].

$$C(x, t) = C_0 + (C^{\text{sat}} - C_0) 4 \sum_{n=0}^{\infty} \frac{(-1)^n}{B_n} \cos\left(\frac{B_n x}{h}\right) \exp\left(\frac{-B_n^2 D t}{h^2}\right) \quad (2)$$

where $B_n = (2n + 1)\pi$ and h is the sample thickness. This expression shows how the moisture ingresses in a sample until it is fully saturated and has a uniform concentration C^{sat} . Experimentally concentration profiles are difficult to measure but the overall concentration change is easy to obtain by monitoring the changes in sample weight. The relation between sample weight and overall concentration follows by integrating Eq. (2) over the sample volume:

$$m(t) = WL \int_0^h C(x, t) dx \quad (3)$$

The mass change is usually reported relative to the dry sample mass, m_{dry} :

$$M(t) = \frac{m(t)}{m_{\text{dry}}} = \frac{\text{mass of absorbed water}}{\text{mass of dry sample}} \quad (\text{in wt\%}) \quad (4)$$

With this, Eq. (2), can be rewritten as.

$$M(t) = M_0 + (M^{\text{sat}} - M_0)[1 - f(t)] \quad (5)$$

where

$$f(\tau) = \sum_{n=0}^{\infty} \frac{8}{B_n^2} \exp[-B_n^2 \tau], \quad \tau = \frac{Dt}{h^2}$$

Here we introduced τ as the dimensionless diffusion time and M^{sat} as the moisture saturation content, i.e. the weight change after full saturation of the sample. M^{sat} increases with the applied humidity level but the effect of temperature is not well established in literature [19]. Some studies report a clear increase of the saturation values at higher temperatures [12,21] whereas in other papers the temperature effect is neglected [22] or attributed to a second diffusion step [1,8]. This lack of conclusiveness may result from the long experimental times and the

scarcity of data at different temperature levels. Based on Henry's law, M^{sat} is supposed to change linearly with the applied relative humidity, RH , but in order to allow for a possible non-linear variations and temperature effects we propose to use a second order approximation:

$$M^{\text{sat}} = a_1 * RH + a_2 * RH^2 \quad (6)$$

in which the coefficients a_1 and a_2 are allowed to vary with temperature. The saturation concentrations typically range from 3 to 7 wt% for unfilled epoxies [3] and 0.25–0.6 wt% for moulding compounds [6,8].

Note that a simple evaluation of the $f(\tau)$ term in Eq. (5) shows that for $\tau > 0.02$ the error of neglecting the $n = 1$ and subsequent terms is $< 2\%$ such that $f(\tau > 0.02) \cong \frac{8}{\pi^2} e^{-\pi^2 \tau}$ can be considered as a good and useful approximation for most of the diffusion process. This approximation can also be used to estimate the time a sample needs to reach saturation. The time to reach 95% of the ultimate saturation level for a Fickian diffusion process is given by

$$t_{0.95} = \frac{-h^2}{\pi^2 D} \ln\left[\frac{8}{\pi^2}(1 - 0.95)\right] \quad (7)$$

For a 1 mm thick moulding compound sample with $D = 1.0 \times 10^{-6} \text{ mm}^2/\text{s}$ it takes about 90 h to reach 95% saturation, for a 2 mm sample this will take 4 times as long, since diffusion scales quadratically with the thickness. For samples with a non-negligible side surface area, the extra water uptake through the side surfaces can be approximately accounted for by using the correction given by Shen [16]:

$$D^{\text{corr}} = \frac{D^{\text{meas}}}{\left(1 + \frac{h}{L} + \frac{h}{W}\right)^2} \quad (8)$$

where L and W denote the sample length and width.

2.2. Non-linear and anomalous diffusion effects

There is a relatively large amount of studies which report clear non-linear diffusion effects. This can be either during a single absorption run in which an initially Fickian diffusion is followed by a second Fickian diffusion with a different diffusion coefficient (2-stage diffusion) [1,10,17,18]. vanLandigem [18] argues that this is probably caused by incomplete cure, resulting in a heterogeneous structure. Placette [1] on the other hand argues that the second stage reflects the water molecules which are chemically bonded to the polymer and which will release more difficult during desorption, eventually resulting in a residual moisture content which can only be released by drying at an elevated temperature.

Anomalous effects are also observed if a previously wet specimen is first dried and then again subjected to a wet environment. The initial diffusion rate then appears to be faster than the rate during the first time diffusion process [10]. They show that the changes during the 2nd absorption stage are irreversible and that the second and third absorption tests are simply continuations of the previous tests when plotted on the overall time scale. The authors were able to model their observations using a two-stage diffusion process. The moisture induced 'irreversible' structural network changes can be removed by annealing above T_g .

2.3. Multiple humidity steps diffusion

Eq. (5) above describes the water uptake during a single applied step in relative humidity. Diffusion however is a slow process and it may happen that the sample is already being exposed to a new humidity condition before equilibrium is reached for the first exposure step. This may for example occur if during a measurement the sample is taken out of the humidity chamber to determine its weight or, in practice, when an electronic device is exposed to fluctuating humidity conditions during transport. We can model the overall weight change caused by a

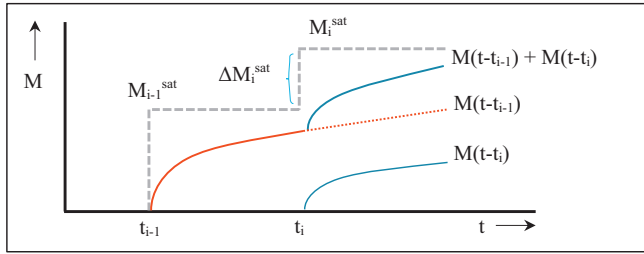


Fig. 1. Schematic representation of weight gain due to humidity steps at t_{i-1} and t_i .

series of humidity steps analogously to creep strain due to a series of applied stress steps. Essential is that in the summation a stress or humidity step which is applied at time t_i creates a contribution proportional to $f(t-t_i)$ which continues to the end, even if the applied humidity level (or stress) has already changed or been removed (see also Fig. 1).

Mathematically this can be written as

$$M_{tot}(t) = M_0 + \sum_{i=1}^{steps} \Delta M_i^{sat} [1 - f(t - t_i)] \quad (9)$$

where M_i^{sat} is the moisture saturation content due to humidity level RH_i applied at time t_i and $\Delta M_i^{sat} = M_i^{sat} - M_{i-1}^{sat}$. Eq. (9) above is valid for both absorption and desorption steps. Desorption towards a non-zero residual moisture content, M^{res} , can be modelled by defining $\Delta M_i^{sat} = M^{res} - M_{i-1}^{sat}$. The equation is originally derived for a series of humidity steps at constant temperature. However, if M_i^{sat} and $f(t)$ are considered as temperature dependent, Eq. (9) can still be used to predict the moisture content of a sample subjected to a series of arbitrary humidity and temperature steps. In Section 4 we will use Eq. (9) to interpret the moisture diffusion behaviour for a series of incomplete diffusion steps at a constant temperature.

3. Experimental

The standard method to characterise moisture absorption in electronic packaging materials is to put a series of relatively large samples (2 to 10 mm thickness) in a preconditioned oven and manually measure the weight gain of these samples using an electronic balance. The problem is that the humidity and temperature control of the oven is likely to be affected after each opening. In addition, the sample may lose some of its moisture during cooling down before each weight

measurement. Both effects will underestimate the moisture uptake. Moreover, for experiments which run over months, care should be taken to carefully calibrate the balance before each measurement. The sensitivity of these laboratory balances is typically limited to 0.01 mg which requires large enough samples.

In this study we aim at a detailed moisture diffusion characterisation which involves an as wide as possible humidity and temperature range. In our case we choose to measure series of five humidity levels at five different temperatures. If each of the humidity steps would require 90 h (see Eq. (7) above) such an analysis would take two to three months of measurement time. In order to both speed up the measurement time and increase the measurement accuracy we propose to change the standard way of measuring in the following three ways

1. We will use a dynamic sorption analyser which continuously measures the weight change of a sample in a chamber with programmable temperature and humidity. In this way samples do not need to be removed from the humidity chamber and measured manually such that part of the error sources related to the traditional diffusion measurements can be avoided. This Dynamic Vapor Sorption technique is commonly used to analyse powders and other materials [22]. Since the measurement sensitivity of the sorption analyser is 0.1 μg (about at least 100 times better than with a conventional balance) and measurements are taken continuously (i.e. each 6 s) the accuracy is expected to increase largely compared to the traditional way.
2. We reduce the sample thickness as much as possible by polishing it down to about 0.2 mm. With respect to a 1 mm thick sample this reduces the diffusion time with a factor 20 to 25. To reach 95% saturation then requires only about 4 h (Eq. (7)). We cannot further reduce the sample thickness since moulding compounds are inhomogeneous and contain silica filler particles with about 20 μm diameter. The sample size thus should always be 50 to 10 times larger than that of the fillers.
3. In contrast to the standard DVS method we shorten the measurement time by switching to the next humidity step at 90 to 95% of the full saturation levels. This can be done without loss of accuracy in determining the diffusion parameters by using the multistep diffusion model, Eq. (9).

3.1. Dynamic sorption analyser

Fig. 2 shows the TA-Instruments Q5000 sorption analyser which is

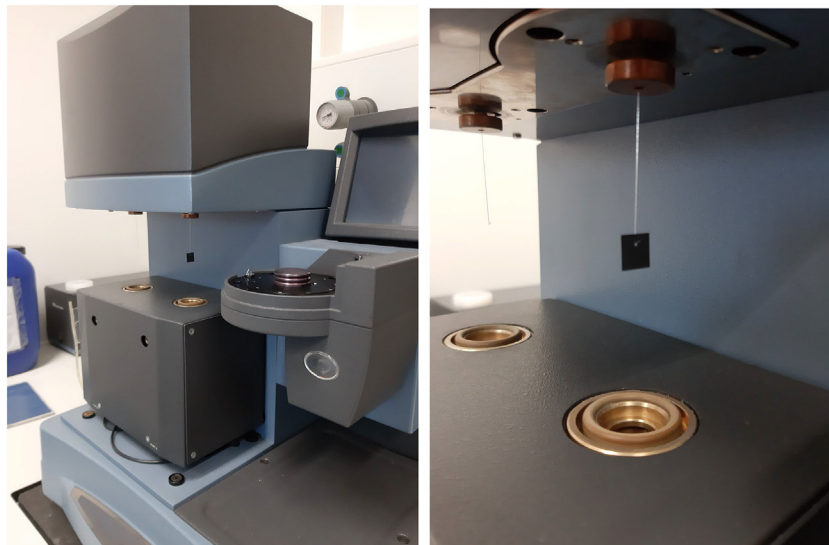


Fig. 2. Q5000 moisture analyser (left), and detail of sample on hook (right).

used during this study. In its standard configuration the instrument balances a reference pan (left) and a pan containing the sample (right). After loading the sample the humidity chamber closes and is conditioned to the desired temperature and humidity program, usually starting with a drying step to ensure starting with a fully dried sample. Temperatures and relative humidity can be programmed between 5 to 95 °C and up to 95% RH, respectively. However at temperatures above 80 °C the maximum attainable humidity level is lower due to instrument limitations. The apparatus stores 5 measurements per minute and the measurement sensitivity is 0.1 µg.

3.2. Sample

We used a commercial epoxy based moulding compound with 90.6 wt% silica filler, a density of 2.04 g/cm³ and a glass transition temperature of 131 °C (determined as the 1 Hz tan δ DMA peak value). The sample is rectangular, measures about 10 by 10 mm and has a thickness of 0.216 mm. In order to allow unhindered diffusion from all sides, the sample and reference pans were removed and the sample (in which a 0.5 mm hole was drilled) was hung directly on the hook of the instrument (see Fig. 2, right). The dry sample mass was 40.11 mg.

3.3. Procedure

Runs with stepwise humidity levels of (0%), 20%, 40%, 60%, 85% and 0% RH were programmed at 5 different temperatures (20, 40, 60, 80 and 85 °C). The final step from 85% to 0% RH measures the desorption. The duration of the humidity steps was programmed as 240 min, except for the 20 °C experiment where the double amount of time was taken. During desorption a longer time period was chosen to verify if after desorption the original weight was obtained. Each series took about 1 day and since all measurements ended with a complete desorption the same sample is used for all measurements.

3.4. Data evaluation procedure

The measurements resulted in one humidity step series for each temperature. The fitting of these partially completed absorption step series is done with a MatLab script using the nonlinear least square curve fitting function and Eqs. (6) and (9) for the diffusion model function. This results in single value for the coefficients a_1 , a_2 , the diffusion coefficient D and residual moisture content M^{res} for each temperature series. As an option the absorption and desorption steps can be evaluated separately. This evaluation procedure is repeated for each of the applied temperatures.

4. Results

Fig. 3 shows the typical obtained data of a single humidity step experiment. In order to find out if the absorption and desorption part should be modelled with different diffusion coefficients we first evaluated the absorption and desorption parts separately and then evaluated all data in one go. The black dotted lines show the applied humidity levels and the dashed lines represent the measured relative mass changes of the experiment at 40 °C (the upper data series is shifted upwards for convenience). The full lines represent the model fits over the full data range (purple; upper curve) and for separate absorption and desorption fits (blue; lower curve). The measurement took about 1500 min (25 h). As can be seen in both cases the data is excellently described by the model with only minor differences being present in the desorption parts. The determined diffusivity constants range from 0.98×10^{-6} and 0.93×10^{-6} mm²/s for the absorption and desorption parts and 0.975×10^{-6} mm²/s for the full data series.

Fig. 4 shows the experimental results for the experiments of all 5 selected temperatures. Preliminary experiments showed that at 85 °C the 85% RH humidity condition could not be reached such that the

85 °C experiment was programmed with only a maximum of 60% RH. The results for the experiment at 20 °C were plotted versus time/2 since there the actual humidity steps took 480 min instead of the standard 240 min. In order to determine how this changes for the other temperature series we decided to evaluate the absorption and desorption parts separately for the rest of the data series. It can be observed that at the end of each humidity step the mass uptake is levelling off but that the plateau values are not reached yet. The desorption curves clearly show that the mass loss rate rapidly increases at higher temperature (keep in mind that for the 20 °C curve the time scale is multiplied by a factor 0.5). The desorption curves also show that after 5 to 10 h the equilibrium values are not yet reached and the samples still contain about 0.01% of water. Most of this remaining water was removed during the sample conditioning for the next experiment (at a higher temperature). The dashed lines are predictions of a Fickian diffusion process according to Eq. (4) above. It can be seen that all absorption steps and saturation concentrations are captured well. The largest deviations appear to be for the desorption process at the higher temperatures.

The plateau values at each humidity and temperature level vary from 0.085% at 20 °C/85%RH to 0.145% at 80 °C/85%RH. Since in simulation tools like Ansys saturation values expressed as concentrations are required as input, from now on we convert the saturation values to concentrations using $C^{sat} = \rho M^{sat}$. These saturated concentration values as a function of RH and temperature are shown in Fig. 5. In terms of concentration the saturation levels vary between 0.5 and 3 mg/cm³. The deviation from linearity is negligible at the lower temperature and is small but noticeable at 80 and 85 °C. Fig. 5 also clearly shows the saturation concentrations increase with temperature. In an attempt to model this behaviour we assumed a linear temperature dependency in addition to the quadratic humidity dependency of Eq. (6) and fitted the saturation data to.

$$C^{sat} = (a_{10} + a_{11}T) * RH + (a_{20} + a_{21}T) * RH^2 \quad (10)$$

Coefficient a_{20} did not contribute much and was set to zero for the fitting. The results are shown as the dashed lines in Fig. 5 and the values of the coefficients are listed in Table 1.

The next step of the modelling is to consider the temperature dependency of the diffusion coefficient. Fig. 6 shows that the diffusion coefficient for absorption increases from 3.8×10^{-7} mm²/s at 20 °C to a 10 times higher value at 85 °C. It furthermore shows that for temperatures above 60 °C the desorption appears to be a bit faster than the absorption.

$$D(T) = D_0 \exp\left(-\frac{E_a}{kT}\right) \quad (11)$$

The temperature dependency is fitted to the Arrhenius equation, Eq. (11) in which D_0 is the Arrhenius constant, E_a the activation energy and $k = 8.83 \times 10^{-5}$ eV/K the Boltzmann constant. The fitting is done by plotting $\ln D_0$ versus $1/T$ (in K) and evaluating E_a and D_0 from the linear fit. The Arrhenius constant D_0 amounts to 0.057 mm²/s for modelling absorption and 0.547 mm²/s for the desorption phase (Table 1). The corresponding activation energies are given as 0.307 and 0.371 eV, respectively. The diffusion coefficients obtained by forced fitting on the full data sets are very similar to those of the absorption part only. The Arrhenius fits can be seen to describe the temperature dependent diffusion behaviour quite well (see Fig. 6).

5. Discussion

In this study we evaluated the moisture diffusion of a commercial moulding compound as used in the electronics industry for microchip encapsulation. We performed the measurements with an automated high precision sorption analyser and were thus able to generate data with much higher precision than reported in most of the previous studies. This data then allowed us to study in detail how the saturation

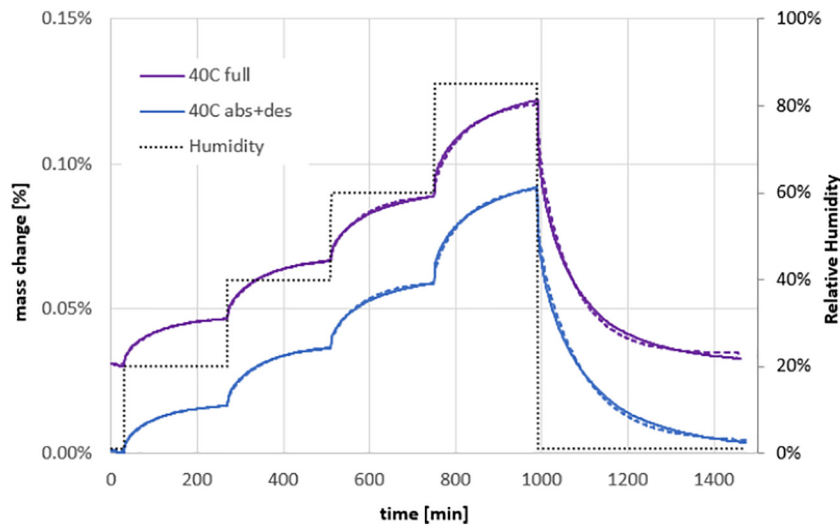


Fig. 3. Dashed lines: measured mass change of the 40 °C series. Full lines: model fits to Eq. (9) either fitted to the full data series (upwards shifted curves) or to absorption and desorption parts fitted separately (lower curve).

concentration and diffusion coefficient changed with the applied relative humidity level and temperature and find out whether there is a difference between absorption and desorption. We also presented a multistep diffusion model which can be used to better interpret the experimental data. This model effectively takes into account all effects of previously applied humidity steps and thus can correct for the cases in which earlier steps did not reach complete saturation.

We conclude that for the temperature range considered here (i.e. 20 to 85 °C) the absorption at different humidity levels as well as the desorption can be modelled as a Fickian process with a single diffusion coefficient. Thus the diffusion coefficient does not depend on the moisture concentration itself and there is no difference in diffusion speed between absorption and desorption. The largest deviations between the Fickian model and measurements appear at the highest temperature and humidity levels and at the end of the desorption stage. These deviations amount to 0.004 wt% at maximum which corresponds to 1.6 µg. If data would be collected in the traditional way (i.e. intermittent manual measurements and a scatter of typically 0.01 wt%) then these deviations even would not have been noticeable. As can be seen in Fig. 4, the amounts of residual moisture vary between 0.004%

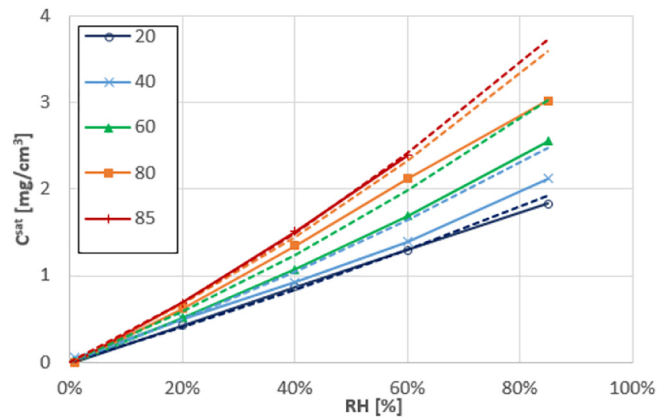


Fig. 5. Moisture saturation concentrations as obtained from the experimental data. Dashed lines are fit to Eq. (10).

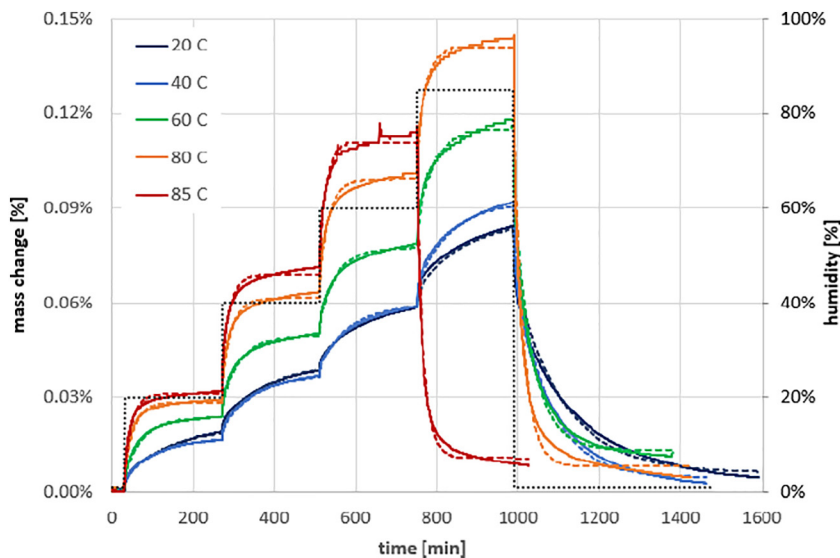


Fig. 4. Humidity steps and observed relative mass change (full lines) as a function of time. Dashed lines are the Fickian model fit for the experiments at different temperatures. Absorption (0–85%RH) and desorption (85–0%RH) are fitted separately. The timescale of the 20 °C experiment is multiplied by a factor 0.5 in the plot.

Table 1

Diffusion parameters corresponding to Eqs. (10) and (11). Note that in Eq. (10) RH values are treated as fractional numbers (i.e. 0.20, 0.40) and temperature is in °C.

	D_0 [mm ² /s]	E_a [eV]	a_{10} [mg/cm ³]	a_{11} [mg/cm ³ /°C]	a_{20} [mg/cm ³]	a_{21} [mg/cm ³ /°C]
Absorption part	0.057	0.307				
Full data set	0.239	0.347	1.604	0.018	0	0.017
Desorption only	0.548	0.371				

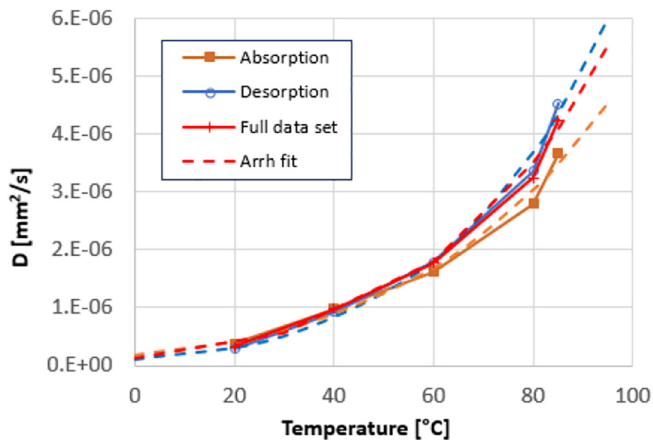


Fig. 6. Diffusion coefficient as a function of temperature and the corresponding Arrhenius fits (dashed lines).

and 0.013% (2 to 5 μg). The largest values are for the 60 to 85 °C experiments which may suggest that this is due to the chemically bound water molecules which are more difficult to release.

The measurements were performed on thin samples for relatively short times. We switched to the next humidity level in the range of 95% to 99% of the final saturation level and did not wait longer to observe a possible second absorption regime. Placette [1], for example, observed the second stage absorption for a 1 mm thick sample to start after 100 h conditioning in a 85 °C/85% humidity setting. This corresponds to a t/h^2 value of $3.6 \times 10^5 \text{ s/mm}^2$ (and a dimensionless diffusion time $\tau = 1.23$). Our humidity steps take 240 min amounting to a t/h^2 of $3.1 \times 10^5 \text{ s/mm}^2$ and a dimensionless diffusion time of 1.1, which may explain why no second stage diffusion was observed in our study.

The moisture saturation levels were seen to increase with the relative humidity in a mildly non-linear way. More interesting to note however is that our results show a clear effect of temperature. At higher temperatures the saturation levels increase and the non-linearity with respect to humidity grows. A possible explanation for this is that at higher temperatures the material expands which allows extra water uptake in the increased free volume and micro cavities. As mentioned before, the literature is still inconclusive about the effect of temperature on the moisture saturation (e.g. [19–22]), so our findings may be a good starting point for more dedicated measurements and a more in depth discussion.

To summarize we can conclude that for mild diffusion conditions (i.e. below 85 °C and RH levels up to 85%) and moderate diffusion duration ($\tau < 1.1$) the diffusion in the moulding compound studied follows a perfect Fickian behaviour and no moisture dependency of the diffusivity nor a difference between absorption and desorption is observed.

CRediT authorship contribution statement

K.M.B. Jansen: Conceptualization, Writing - original draft, Data curation, Formal analysis. **M.F. Zhang:** Writing - review & editing, Investigation, Formal analysis. **L.J. Ernst:** Conceptualization, Supervision. **D.-K. Vu:** Conceptualization, Supervision, Project administration, Funding acquisition. **L. Weiss:** Supervision, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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