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AIP Conf. Proc. 2065, 030037 (2019)

<https://doi.org/10.1063/1.5088295>



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Calculation of the Drying Process of Hygroscopic Polymer Pellets in Desiccant Dryers and With Additional Microwave Application

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Abstract. Hygroscopic polymers absorb water and bind it within; therefore an effective drying down to an acceptable moisture level is essential for a successful production. If not dried properly, the residual moisture can cause major problems for converting and also significantly affect the product quality. Usually, data sheets give standard recommendations for drying temperatures and times; however, these do neither take into account the actual current moisture up-take of the plastic pellets, nor the current conditions. In this work, a simulation model is presented, calculating the heat and mass transfer within the polymer pellet in order to predict the drying progression. In addition to approaches from the literature describing the diffusion coefficient of water in plastic with a complex function and a high number of material parameters, a linear function was used to model the dependence of diffusion from moisture and temperature. Moreover, the effect of microwave application for an accelerated drying was included by calculating the resulting temperature rise from dielectric heating. The simulation of the drying process shows a very good conformity with experimental results. The linear approach used in this work leads to good results and allows to reduce the complexity of the whole model compared to approaches from the literature. The calculation of a temperature rise through dielectric heating also shows satisfying agreement with that measured in the real process from a prototype for microwave drying on an industrial scale. Therefore, the model is a reliable tool to predict required drying times for different polymers and under variation of drying conditions.

Keywords: Drying, Polymers, Diffusion, Microwaves, Simulation.

PACS: 81.05.Lg; 82.20.Wt; 84.40.-x; 66.30.-h

INTRODUCTION

The majority of engineering plastics are so called hygroscopic plastics, i. e. they absorb moisture and bind it within. Therefore, they need to be dried before processing and converting, as the presence of moisture would otherwise lead to material damage [1], ranging from simple surface defects like clouding and loss of transparency or gloss to severe changes in the performance characteristics of the final part due to molecular degradation via hydrolysis. [2] Also, the viscosity during processing can be changed, leading to wastage.

Most State-of-the-Art (SoA) dryers for plastic pellets are so called desiccant dryers, using a stream of dehumidified hot air to dry the pellets convectively. The drying performance is therefore physically limited by the speed of heat conduction and diffusion of water particles within the polymer [3, 4]. The efficiency limitations of such dryers can only be reduced but not completely be overcome, as they are a direct consequence of the process design itself. [5]

In spite of being of high relevance for the industry, common drying practices only rely on standard recommendations without deeper understanding of the mechanisms and influencing factors. The storage time of opened or even unopened bags or silos as well as the current humidity of the ambient air are crucial factors often neglected. They all influence the moisture content of the plastic pellets and therefore lead to drying requirements different from the recommendations on the data sheets, which may lead to insufficient drying. A generally applicable method to predict required drying times and parameters is not available.

In preceding works the authors have investigated the application of microwaves to accelerate the drying process and increase its efficiency [6] The idea behind microwave-based drying (MW drying) is to use the microwaves for a) volumetric heating of the pellets, and b) increasing the partial pressure of the bound moisture due to the microwave stimulation of water molecules (Figure 1). Even though the potential of this technology has been discussed since the 80s [7], it is not yet well established in plastics processing, even though it is considered promising [6]. The authors' previous findings showed that drying times can be significantly reduced, especially when drying at lower

temperatures [6]. However, microwave supported drying poses an additional challenge to converters, as the parameters have to be carefully adjusted to not cause thermal damage or melt the pellets and there is only little knowledge on how to select the right settings. Different polymers react differently to microwave application due to their, depending on their dielectric properties. [7, 8]

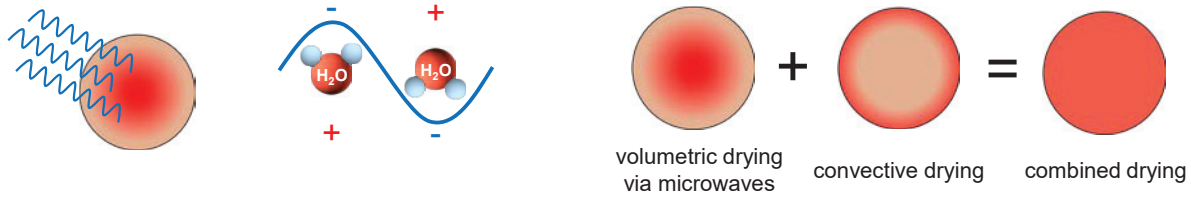


FIGURE 1. Principle of microwave supported drying

This paper investigates the drying kinetics of conventional as well as microwave supported drying. A model is presented, which is able to calculate the drying progress of polymer pellets over time for given conditions, therefore allowing to predict required drying times and adjust the drying process to different circumstances.

MODELING OF DRYING

Depending on the kind of pelletizing process, plastic pellets are either spherical or cylindrical. The calculation of heat and mass transfer within a pellet is therefore done in spherical or cylindrical coordinates. Both mechanisms can be regarded as one-dimensional in a spherical pellet. For cylindrical pellets, however, this assumption might not hold when the diameter of the cylinder is not substantially smaller than its height. Within this paper only spherical coordinates are taken into account. The following general assumptions can be made and are illustrated in Figure 2: Before the beginning of the drying process the initial temperature T_0 and the initial moisture content X_0 are evenly distributed within the pellet. As soon as the drying process starts and a convective medium with a temperature $T_\infty > T_0$ and a humidity $Y < Y_{eq}$ (which is the humidity in thermodynamic equilibrium with X_0) is applied, temporal and spatial gradients of temperature and humidity occur within the pellet.

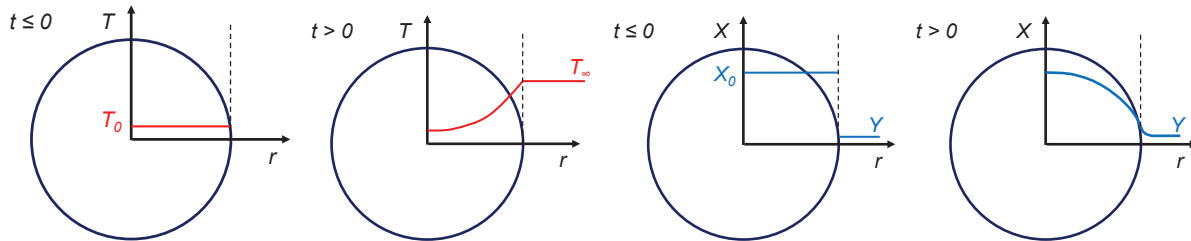


FIGURE 2. Temperature and moisture gradients within the pellet before and during drying

Transient Heat Transfer

The general 2nd grade partial differential equation for a spherical coordinate system is [9]:

$$\frac{\partial T}{\partial t} = \frac{a_K}{r^2} \cdot \frac{\partial}{\partial r} \left(r^2 \cdot \frac{\partial T}{\partial r} \right), \quad (1)$$

with a as the thermal diffusivity, t the temporal coordinate, r the spatial coordinate. The following initial and boundary conditions apply according to the assumptions given above:

$$T(r, t = 0) = T_0, \quad (2)$$

$$\left. \frac{\partial T}{\partial r} \right|_{r=0, t>0} = 0, \quad (3)$$

$$\left(\frac{\lambda}{\alpha} \cdot \frac{\partial T}{\partial r} + T \right)_{r=R, t>0} = T_\infty - \frac{\beta}{\alpha} \cdot \Delta h_W^V \cdot (Y - Y_{eq}). \quad (4)$$

Eq. (2) describes the initial temperature, Eq. (3) binds the problem at the symmetry axis and Eq. (4) describes the convective heat transfer at the pellet surface with the thermal conductivity λ , the heat transmission coefficient α and

the mass transmission coefficient β . However, as the thermal conductivity of the polymer and the mass transmission coefficient to the air are both much smaller than the heat transmission coefficient to the air, the boundary condition can be simplified to:

$$T_{r=R,t>0} = T_{\infty}. \quad (5)$$

Thus, the heat transfer is fully described and can be solved by means of finite difference method.

Transient Mass Transfer

Principally, the transient mass transfer follows the same laws as the heat transfer. It is expressed by Fick's 2nd law of diffusion, which, according to literature, is usually applicable to describe the diffusion of water within polymers [10]. In spherical coordinates it is:

$$\frac{\partial X}{\partial t} = \frac{D}{r^2} \cdot \frac{\partial}{\partial r} \left(r^2 \cdot \frac{\partial X}{\partial r} \right), \quad (6)$$

with D as the diffusion coefficient of the water in the polymer. As Eq. (1) and (6) are mathematically equivalent, the initial and boundary conditions can be described analogously.:

$$X(r, t = 0) = X_0, \quad (7)$$

$$\left. \frac{\partial X}{\partial r} \right|_{r=0,t>0} = 0 \quad (8)$$

$$-\rho_p \cdot D \cdot \left. \frac{\partial X}{\partial r} \right|_{r=R,t>0} = \rho_a \cdot \beta \cdot (Y_{eq} - Y) \quad (9)$$

Eq. (9) describes the equilibrium between the mass flow leaving the pellet at its surface and the mass flow carried away by the convective medium. ρ_p and ρ_a are the respective densities of the polymer and the air. The mass transmission coefficient β can be determined according to [11] and the equilibrium humidity Y_{eq} can be determined according to [10]. Thus, the mass transfer is fully described and can be solved by means of finite difference method.

Diffusion Coefficient of Water in Polymers

The right choice of a model for the diffusion coefficient is crucial for a successful simulation of the drying process. In conventional material modelling the Arrhenius equation is often applied. However, this approach does only include temperature dependency, whereas for solvents in polymers it is known from the literature that the diffusion coefficient is also dependent on concentration [4]. An established model including this is the free-volume approach in the formulation of Vrentas and Duda. This approach has already been successfully applied by the authors in preceding works [12]. It can be shown that the dependency of the diffusion coefficient on concentration follows a linear correlation, as seen in Figure 3, which makes it possible to choose a simpler approach. Thus, the diffusion coefficient can be described as a polynomial of degree 1 with the coefficients being dependent from temperature:

$$D(T, X) = c(T) + s(T) \cdot X. \quad (10)$$

This much simpler approach is used and evaluated within this work.

Microwave Heating

The application of microwaves leads to a raise of energy level within the polymers, mainly due to dissipation and resulting temperature increase. The effect of microwaves on a material and the amount of energy absorbed by it is characterized by the dielectric loss factor ε'' . When this factor is known, as well as the electric field strength E_i and the frequency of the field f , the dissipated volume-specific power π_{diss} is:

$$\pi_{diss} = 2 \cdot \Pi \cdot f \cdot \varepsilon_0 \cdot \varepsilon'' \cdot E_i^2. \quad (11)$$

The vacuum permittivity ε_0 is a natural constant. Thus, π_{diss} is directly proportional to ε'' . However, as the electric field strength is unknown in many cases, the power P_{mw} of the magnetron used to generate the microwaves can be used instead: Within a temporal interval Δt the temperature rise ΔT through microwaves is then:

$$\Delta T = \frac{2 \cdot \Pi \cdot f \cdot \varepsilon_0 \cdot \varepsilon'' \cdot E_i^2}{c_p \cdot \rho_p} \cdot \Delta t = \frac{P_{mw} \cdot \varepsilon'' \cdot k}{m_K \cdot c_p} \cdot \Delta t, \quad (12)$$

with the specific heat capacity c_p of the polymer and its density ρ_p . In Eq. (13) k is a proportional factor and m_K is the total bulk mass of the plastic pellets. Assuming a homogeneous distribution of the microwaves within the pellets, the additional temperature rise for a point in time t can be added to the solution of Eq. (1) at any local point r .

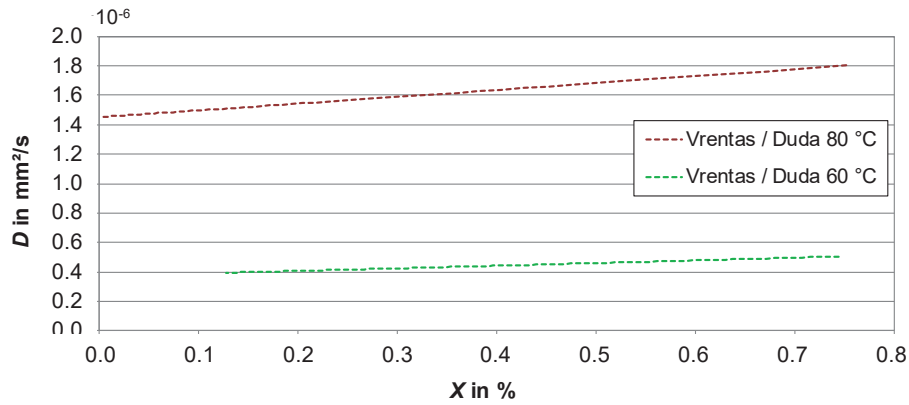


FIGURE 3. Diffusion coefficient determined according to Vrentas and Duda

RESULTS

The described model was verified by comparing calculation results with experimental tests including five different polymers: polyamide 6 (two different grades), polyethylene terephthalate, polylactid acid and polyhydroxybutyrate-valerate. All materials were characterized via differential scanning calorimetry to determine thermal properties and then pre-conditioned for drying tests. Tests were done with a SoA desiccant dryer from Bierther GmbH, Bad Kreuznach, Germany. For microwave application a pilot plant dryer designed and built within the EU-funded project “HiPerDry” was used. Details on the materials as well as the test set-up and the experimental results can be found in preceding publications from the authors [6].

The partial differential equations and the respective initial and boundary conditions were solved using the numerical mathematics platform Matlab® from MathWorks, Natick. For this, a pre-existing solver *pdepe* was modified. Results for the simulation in comparison with the experimental data are presented in Figure 4. The graphs show the residual moisture content X in the plastic pellets over drying time t . It is obvious that calculated drying progress and experimental results are in good compliance. Therefore, the presented model is a suitable tool to predict drying times for different polymers and under different drying conditions. Only the left part of the curves for 80 °C seem to implicate a higher simulated drying rate than can be found in reality. The reason for this is that heat losses of the air, while the pellets are brought from their initial temperature to the setting point, are neglected in the model.

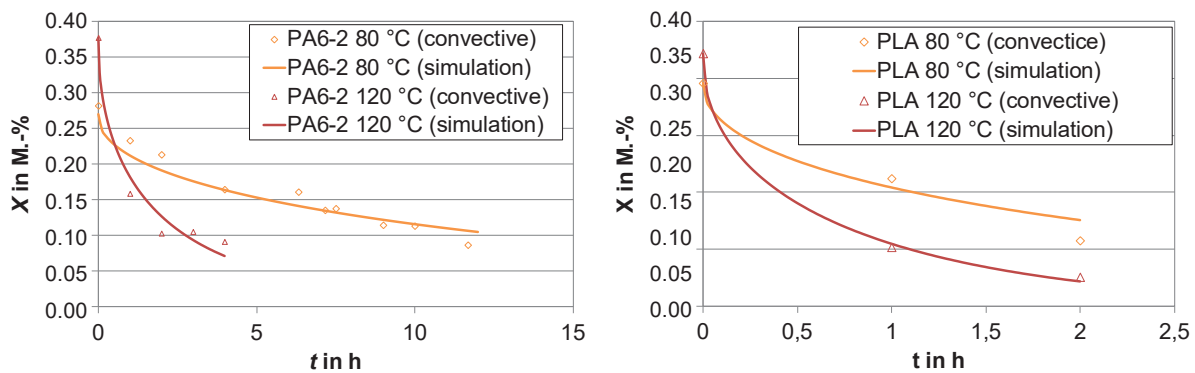


FIGURE 4. Comparison between experimental results (dots) and simulation (lines) for PA6-2 and PLA

To evaluate the approach for the temperature rise calculation, the pellet temperatures measured within the dryer were used. To avoid biases due to heat losses from the air until the pellets are fully heated up, only measurements after

this first phase were used for the comparison. The results are shown in Figure 5 for periods of 2 and 4 hours. They show a good agreement between calculations and measurement data, especially for 80 °C. For 50 °C the calculations predict a lower temperature than measured. It has to be noted that for 50 °C the dielectric loss factor of the materials could not be measured and was extrapolated from the data from higher temperatures, which is likely to be the reason for the differences. However, all deviations are comparatively small, which indicates that the model is suitable to predict the temperature rise from microwave application with a high accuracy.

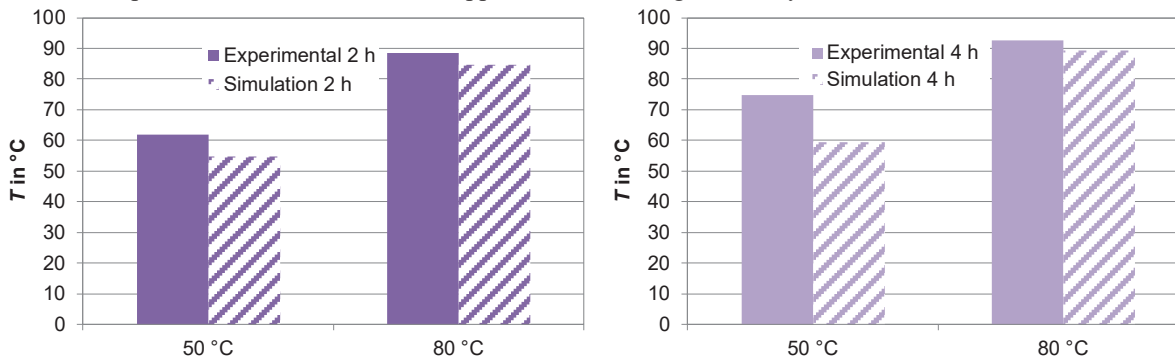


FIGURE 5. Comparison of measured and calculated temperatures

CONCLUSION

The simulation of the drying process shows a very good conformity with experimental results. The linear approach do describe the diffusion coefficient of water in polymer used in this work leads to good results and allows to reduce the complexity of the whole model compared to approaches from the literature. The calculation of a temperature rise through dielectric heating also shows satisfying agreement with that measured in the real process from a prototype for microwave drying on an industrial scale. Therefore, the model is a reliable tool to predict required drying times for different polymers and under variation of drying conditions.

ACKNOWLEDGMENTS

The findings presented in this paper were funded by European Union's Seventh Framework Program under Grant Agreement No 606425. The Authors would like to thank the whole HiPerDry project consortium for the good and trustful cooperation.

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