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Impact of the number and sampling time of water content measurements on the identifiability of a concentration-dependent water diffusivity

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ABSTRACT

The aim of this work was to determine the impact of the experimental conditions (number and sampling time) of measurements of the water content on the identifiability of a concentration-dependent water diffusivity. Water contents simulating a drying process were generated from a diffusion model with the addition of Gaussian noise. An infinite slab with Dirichlet boundary conditions was considered. The coefficients describing a concentration-dependent water diffusivity were estimated from the water content by least-squares minimization. The identifiability of the coefficients was investigated from the model sensitivity functions and using the asymptotic method, the profile likelihood method, and Monte Carlo simulation. The inner product sensitivity matrix was full rank, indicating that the coefficients were locally structurally identifiable. The coefficients were estimated more accurately from water content obtained over the whole drying process, because sampling times concentrated in a short period increased the correlation between the coefficients and increased their uncertainty. The coefficients were practically identifiable from the water content for noise intensity below 0.4%. Above that threshold, the number of measurements of the water content required for the practical identifiability of the coefficients increased linearly with the noise intensity of the water content.

1. Introduction

Efficient drying operations and accurate estimation of the drying rate can be achieved by modeling the movement of water inside a product. For drying operations controlled by a diffusion mass transfer mechanism, estimation of the water diffusivity is required for model application. Because it cannot be measured directly, the water diffusivity of a product (the input variable of the drying model) is generally estimated from measurements of the product's global water content (the output variable of the drying model) by least-squares minimization. A proper experimental design, in terms of the number of global water content measurements and their sampling time, is critical for an accurate estimation of the water diffusivity. Experiments should be designed considering the structural and practical identifiability of the water diffusivity. Structural identifiability refers to the unicity of the least-squares objective function for the theoretical situation of perfect (noise-free) measurements of the output variable [1–4]. The input variables are structurally identifiable if the model mapping from the input variable space to the output variable space is injective [5–7]. Practical identifiability refers to the accuracy of the input variable estimate from experimental (noisy) measurements of the output variable [8]. Practical identifiability reflects the sensitivity of the input variable to measurement noise and can be assessed from the confidence interval of the estimate [8].

The identifiability of a constant diffusivity (independent of the position, time, or concentration) and a convection mass transfer coefficient has been investigated. Analysis of the least-squares objective function by Mercier et al. [9] showed the structural identifiability of the water diffusivity and the convection mass transfer coefficient from local or global measurements of the water content. However, a high number of measurements is required for the practical identifiability of the coefficients if the measurement noise is above 2% for global water content and above 5% for local water content. Mercier et al. [9] also showed that the confidence intervals of the water diffusivity and the convection mass transfer coefficient need to be calculated using a systematic method, such

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Nomenclature

A	coefficient of the concentration-dependent water diffusivity
$\hat{A}$	least-squares estimate of A
CI	95% confidence interval
$cor_{D_0,A}$	correlation between D_0 and A
cov	covariance matrix
D	water diffusivity ($m^2\,s^{-1}$)
D_0	coefficient of the concentration-dependent water diffusivity ($m^2\,s^{-1}$)
$D_{0.3}$	water diffusivity for a water content $M = 0.3$ ($m^2\,s^{-1}$)
$\hat{D}_0$	least-squares estimate of D_0 ($m^2\,s^{-1}$)
FIM	Fisher information matrix
Fo	Fourier number ($Fo = \frac{D_{0.3}t}{w^2}$)
M	water content (kg H_2O kg dry matter^{-1})
$\bar{M}_i$	global water content without noise (kg H_2O kg dry matter^{-1})
$\bar{M}_i^*$	global water content with noise (kg H_2O kg dry matter^{-1})
n	number of water content values
$n_{\min}$	minimum number of water content values for the practical identifiability of the coefficients D_0 and A
S	state variable sensitivity
$\bar{S}$	global (according to x) state variable sensitivity
SSE	error sum of squares
t	time (s)
w	product half-thickness (m)
x	thickness coordinate (m)

Greek symbols

ε	noise
λ	sensitivity matrix
ρ	square root of the determinant for the inner product sensitivity matrix
σ	standard deviation of the noise
σ^2	variance of the noise

Subscripts

0	initial
E	equilibrium
i	time i

as the profile likelihood method, because the local asymptotic method assumes symmetric confidence intervals and underestimates the uncertainty of the coefficients. In accordance with Mercier et al. [9], Martinez-Lopez et al. [10] showed that local measurements of the output variable provide more accurate estimates of the coefficients than global measurements do and that local measurements hence promote the coefficients' practical identifiability. Wan et al. [11] studied the impact of the number of measurements of the output variable on the practical identifiability of the coefficients for different geometries and showed that the coefficients are practically identifiable if the ratio of noise to the number of measurements is low and if the Biot number (dimensionless ratio between the internal and external resistances) is close to unity. Wan et al. [11] also showed the importance of higher-order nonlinearities in the asymptotic estimation of the coefficient confidence intervals.

The identifiability of a position-dependent coefficient has been investigated by DuChateau [12] and Teergele and Danai [13]. DuChateau [12] demonstrated the identifiability of a position-dependent diffusivity in a parabolic partial differential equation from the controllability of the adjoint problem. Teergele and Danai [13] used the sensitivity of the output variable for the coefficients to determine the impact of the sensor location on the identifiability and applied their method to the 2D dispersion of pollutants in air described by an advection–diffusion equation.

To our knowledge, the identifiability of a concentration-dependent diffusivity for a transient process has not been investigated. For hygroscopic products, including many polymers and food products, the water diffusivity decreases during drying as the ratio of bound to free water increases [14–17]. The dependence of the water diffusivity for the water content is generally expressed using an exponential function:

$$D = D_0 \exp(AM) \tag{1}$$

where D is the water diffusivity, M is the local water content, D_0 is a coefficient determining the water diffusivity at the limit $M \to 0$, and A is a coefficient describing the sensitivity of the water diffusivity for the water content. The dependence for the water content modifies the identifiability, given that the diffusivity becomes both position- and time-dependent and that two coefficients, D_0 and A, need to be estimated from the water content.

The aim of this work was to assess the structural identifiability of a concentration-dependent water diffusivity from global water content and to determine the experimental conditions (number and sampling time of measurements of the global water content) required for its practical identifiability. To that end, global water content values were generated by adding Gaussian noise to drying model predictions. The coefficients describing a concentration-dependent water diffusivity, D_0 and A, were estimated from these global water content values by least-squares minimization. The structural identifiability of the coefficients was investigated from the least-squares objective function and the drying model sensitivity functions. The practical identifiability was assessed from the confidence intervals calculated using the asymptotic method and the profile likelihood method. The impact of the number and sampling time of measurements of the global water content on the accuracy of least-squares estimates was investigated according to the noise intensity using Monte Carlo simulation.

2. Methods

2.1. Drying model

Drying of an infinite slab was described using a diffusion model with a concentration-dependent water diffusivity:

$$\frac{\partial M}{\partial t} = \frac{\partial}{\partial x}\left[D_0 \exp(AM)\frac{\partial M}{\partial x}\right] \tag{2}$$

where t is the drying time, and x is the coordinate in the mass transfer direction. Eq. (2) was solved considering uniform initial water content M_0 (Eq. (3)) and Dirichlet boundary conditions (Eq. (4)):

$$M(x, t = 0) = M_0 \tag{3}$$

$$M(x = -w, t) = M(x = w, t) = M_E \tag{4}$$

where M_E is the equilibrium water content, and w is the product half-thickness, where the origin of the drying model was established.