ESTIMATION OF THE THERMAL DIFFUSIVITY IN A LARGE ELECTROCERAMIC BODY BY AN INVERE METHOD

Húlan T.,[1] Ondruška J.,[1,2] Kováč J.,[1] Medved' I.,*[1,2] Trník A.,[1,2] and Vozár L.[1]
*Author for correspondence
[1] Department of Physics, Constantine the Philosopher University, 949 74 Nitra, Slovakia
[2] Department of Materials Engineering and Chemistry, Czech Technical University, 166 29 Prague, Czech Republic
E-mail: imedved@ukf.sk

ABSTRACT

We investigate the temperature dependence of the thermal diffusivity for a large ceramic body of a cylindrical shape during firing up to 900 °C. The body was made of a ceramic material used in the production of electroporcelain insulators. We describe the corresponding heat transfer by the standard heat equation and solve the inverse problem by the Levenberg-Marquardt method. The results show that the method allows one to detect the physical-chemical processes occurring in the material during firing, namely, the liberation of physically bound water in the range up to 250 °C, the phase transformation of kaolinite into metakaolinite (dehydroxylation) in the range ~ 450 °C – 650 °C, and solid-state sintering starting at ~ 700 °C.

INTRODUCTION

Physical properties of composite materials like ceramics are necessarily affected by various processes (phase transformations, chemical reactions, etc.) that may take place in the materials during their thermal treatment. In particular, this is true for the temperature dependence of the thermal diffusivity. Isothermal measurement methods – for example, the standard flash method [1,2] – are not capable of registering the influence of such processes. A convenient approach to this problem is the use of inverse methods [3] that enable one to estimate, quite generally, various kinetic parameters from experimental data. In the case of the thermal diffusivity the data correspond to the time and position dependence of the temperature in the material.

In kaolin-based ceramics three main processes occur in the firing range 20 – 900 °C [4-6]:
  (A) the liberation of physically bound water in the range up to 250 °C;
  (B) the phase transformation of kaolinite into metakaolinite (i.e., dehydroxylation) in the range ~ 450 °C – 650 °C;
  (C) solid-state sintering starting at ~ 700 °C.

As a rule, these processes may significantly affect the temperature dependence of the diffusivity in their respective temperature ranges, resulting in dramatic changes in the diffusivity. It is the aim of this paper to determine the thermal diffusivity as depending on the temperature for an alumina ceramic material used in the production of electric insulators, using the inverse approach and compare the results with those obtained by the flash method in which the processes A – C cannot be recorded.

NOMENCLATURE

a	[m^2/s]	Thermal diffusivity
L	[m]	Sample thickness
r	[m]	Radial position
T	[K]	Temperature
t	[s]	Time

| Special characters		
μ	[-]	Damping parameter
ρ	[kg/m^3]	Density

| Subscripts	
bd	Boundary temperature
exp	Temperature data from experiment
est	Estimated temperature values from an inverse analysis
fin	Final time
in	Initial time or temperature
Tr	Transposed matrix

EXPERIMENTAL

Samples were prepared from a wet plastic ceramic mass that was provided by the electro-porcelain plant PPC Čab, Slovakia, a member of the international PPC Insulators group. The initial composition of the ceramic material was: 48 wt. % of kaolin and clay, 30 wt. % of alumina, and 22 wt. % of feldspar. The samples were prepared to be of a cylindrical shape with the diameter 80 mm and length 120 mm (see Figure 1). Four holes of diameter 2 mm were bored in the axial direction in the

cylindrical samples in which thermocouples Pt-PtRh10 were placed to measure the time evolution of the temperature (see Figure 1). The radial distances of the thermocouples were as 0 cm (the samples' center), 1 cm, 2 cm, and 3 cm. The heating rate was chosen as 2 °C/min. The experimentally measured time dependences of the temperatures at the four positions are shown in Figure 2.
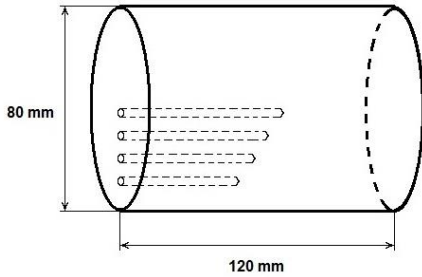
Figure 1 A cylindrical sample with holes for four thermocouples
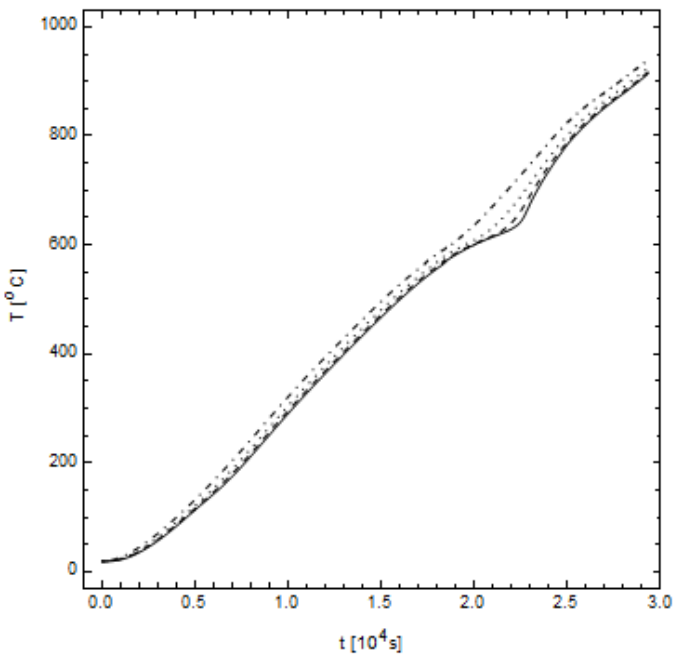
Figure 2 The measured temperature data at the radial positions 0 cm (full line), 1cm (dashed line), 2 cm (dotted line), and 3 cm (dot-dashed line)

For the sake of comparison of results, we also employ the standard flash method [7,8] to measure the thermal diffusivity. The method is based on the measurement of a thermal response of a studied material to a short heat pulse. The response is measured near one face of the material, while the pulse initially impacts and heats the opposite face. Since the time dependence of the thermal response, $T(t)$, can be determined analytically, it is possible to calculate the thermal diffusivity by fitting the theoretical dependence to experimental data. To simplify the analysis, one usually uses just a single experimental value of the thermal response $T(t)$, namely, at the time $t_{1/2}$ at which the response attains 1/2 of its maximal value. Then the thermal diffusivity can be calculated from the simple formula a = 0.1388 $L^2/t_{1/2}$, where L is the sample thickness. In our measurements we used a device whose highest operating temperature is 650 °C (see [7, 8] for details).

THEORETICAL

We assume that the heat transport in the cylindrical samples was solely due to conduction and that it can be described by the one-dimensional sourceless heat conduction equation [9]:

$$\frac{\partial T(r,t)}{\partial t} = a \frac{1}{r} \frac{\partial}{\partial r}\left(r \frac{\partial T(r,t)}{\partial r} \right) \qquad (1)$$

The thermal diffusivity $a = a(T)$ generally varies with the temperature. Using experimentally obtained values of T in dependence on r and t, we shall employ an inverse method to estimate the values of $a(T)$.

Direct Problem

The direct problem consists in solving Eq. (1) for $T(r,t)$, as long as initial and boundary conditions are specified and the diffusivity a is a known function of the temperature. Our experimental situation corresponds to the initial condition:

$$T(r,t_{in}) = T_{in}(r), \qquad 0 \le r \le R \qquad (2)$$

Here $T_{in}(r)$ is the radial distribution of the temperature at an initial time t_{in} and $R = 3$ cm is the maximal radial position for which Eq. (1) is solved. The boundary conditions are as follows:

$$T(R,t) = T_{bd}(t), \quad \frac{\partial T(0,t)}{\partial r} = 0, \qquad t_{in} \le t \le t_{fin} \qquad (3)$$

Here $T_{bd}(t)$ is the time dependence of the temperature at the largest radial position R and t_{fin} is a final time up to which the direct problem is solved. The second boundary condition expresses the radial symmetry of the temperature distribution in a cylindrical body.

Once the function $a(T)$ is known, Eqs. (1) – (3) can be solved, yielding a unique solution $T(r,t)$ in the solution domain $0 \le r \le R$ and $t_{in} \le t \le t_{fin}$. Hence, the direct problem is a well-posed type of mathematical problem: its solution satisfies the requirements of existence, uniqueness, and stability with respect to the input data [10].

Inverse Problem

The inverse problem of estimating the thermal diffusivity $a(T)$ is opposed in formulation to the direct problem: knowing experimental values of $T(r,t)$ for an ensemble of positions r_k and times t_l, the dependence $a(T)$ is estimated from Eqs. (1) – (3). This is not a well-posed type of a mathematical problem in the above sense. Nevertheless, it can be reformulated as such (for example, as a minimization problem [10]). Indeed, it has been conveniently used to estimate heat and mass transfer

parameters in many applications (see, for example, [11-19]).

The inverse approach to the estimation of the diffusivity is a nonlinear problem due to its temperature dependence. Thus, we consider only the data for which the measured temperatures are within a relatively small interval so that $a(T)$ can be approximated by a constant. To this end, we split the studied temperature range 20 °C – 800 °C into n small intervals of width 4 °C and represent $a(T)$ by constant values $a_1, \ldots, a_n$ in these intervals. We use $\boldsymbol{a}$ to denote the vector $(a_1, \ldots, a_n)$ of estimated values of the thermal diffusivity in the n temperature intervals.

The inverse problem is reformulated as a minimization of the weighted least-square norm:

$$S(\boldsymbol{a}) = [\boldsymbol{T}^{\text{exp}} - \boldsymbol{T}^{\text{est}}(\boldsymbol{a})]^{\text{Tr}} \boldsymbol{W} [\boldsymbol{T}^{\text{exp}} - \boldsymbol{T}^{\text{est}}(\boldsymbol{a})] \qquad (4)$$

Here $\boldsymbol{T}^{\text{exp}} = (T^{\text{exp}}(r_k, t_l))$ is the vector of the $4m$ measured values of the temperature at the four thermocouple positions r_k and m times t_l, while $\boldsymbol{T}^{\text{est}}(\boldsymbol{a}) = (T^{\text{est}}(r_k, t_l))$ is the vector of the $4m$ estimated values of the temperature at positions r_k and times t_l as obtained from the solution of the direct problem with $a(T)$ represented by $\boldsymbol{a}$. The matrix $\boldsymbol{W}$ is a diagonal matrix with diagonal elements equal to the inverse of the variances of the measurements.

To solve the inverse problem, we employ the Levenberg-Marquardt method [20,21]. It is based on an iterative procedure in which the diffusivity vector $\boldsymbol{a}^{(i+1)}$ in the $(i+1)$-th step is related to the diffusivity vector $\boldsymbol{a}^{(i)}$ in the preceding step as $\boldsymbol{a}^{(i+1)} = \boldsymbol{a}^{(i)} + \Delta\boldsymbol{a}^{(i)}$, where the corresponding change is:

$$\Delta\boldsymbol{a}^{(i)} = \frac{\boldsymbol{J}_i^{\text{Tr}} \boldsymbol{W} [\boldsymbol{T}^{\text{exp}} - \boldsymbol{T}^{\text{est}}(\boldsymbol{a}^{(i)})]}{\boldsymbol{J}_i^{\text{Tr}} \boldsymbol{W} \boldsymbol{J}_i + \mu_i} \qquad (5)$$

The sensitivity matrix $\boldsymbol{J}_i = \partial\boldsymbol{T}^{\text{est}}(\boldsymbol{a}^{(i)})/\partial\boldsymbol{a}^{(i)}$ is the $4m \times n$ matrix whose n columns contain the derivatives of the $4m$ estimated temperature values in the i-th step with respect to the i-th step value a_j (with $j = 1, \ldots, n$) and μ_i is a damping parameter. At the beginning of the iteration procedure μ_i is usually taken large and then it is gradually decreased. However, if instabilities are generated during the iteration procedure, μ_i should be increased, making the Levenberg-Marquardt method rather stable. Therefore, if in the $(i+1)$-th step the norm S_{i+1} is less (more) than the value S_i in the i-th step, we set $\mu_{i+1} = \mu_i/10$ ($\mu_{i+1} = 10\mu_i$).

The solution algorithm for the Levenberg-Marquardt method may slightly vary, depending on the specific problem. We employed the version described in [11,13,22] with the stopping criterion given as:

$$\frac{|a_j^{(i+1)} - a_j^{(i)}|}{|a_j^{(i+1)}|} \le \varepsilon \qquad (6)$$

The prefixed positive tolerance was chosen as $\varepsilon = 0.01$. The evaluation procedure in the domain $[0, R] \times [t_{\text{in}}, t_{\text{fin}}]$ was repeated until the stopping criterion (6) was satisfied. Afterwards, the same procedure was initiated for the next domain, with the starting value of the diffusivity taken as its resulting value in the previous interval. To carry out the Levenberg-Marquardt inverse analysis, we employed the numerical computing environment MATLAB.

RESULTS AND DISCUSSION

The experimental data for the time evolution of the temperature at four radial positions in Figure 2 show two notable deviations from a practically linear increase. One occurs in the range 20 – 200 °C and the other one between ~570 °C and 700 °C. In both cases the increase of the temperature is slower than linear. So, they both correspond to endothermic reactions: the reaction at lower temperatures is due to the liberation of physically bounded water, while the reaction at higher temperatures is due to the dehydroxylation (when thermal energy is used for the liberation of chemically bounded water). These two reactions should influence the profile of the temperature dependence of any macroscopic property of the studied ceramic material, including the thermal diffusivity.

The temperature dependence of the thermal diffusivity as obtained by the inverse method applied to the measured data from Figure 2 is shown in Figure 3. A rather fast decrease in a from $\sim 5.0 \times 10^{-7}$ m^2/s by about 45 % in the range 20 – 200 °C is associated with the above-mentioned liberation of physically bounded water. We thus conclude that humidity increases the thermal diffusivity, as should be expected.

In the following range, ~200 °C – 550 °C, no reactions occur in the studied material, and the thermal diffusivity is practically constant, around 2.7×10^{-7} m^2/s.

However, a sudden drop in a to $\sim 0.6 \times 10^{-7}$ m^2/s takes place in the range 550 – 650 °C that corresponds to the interval where the dehydroxylation starts. So, this process affects the thermal diffusivity to a large extent.
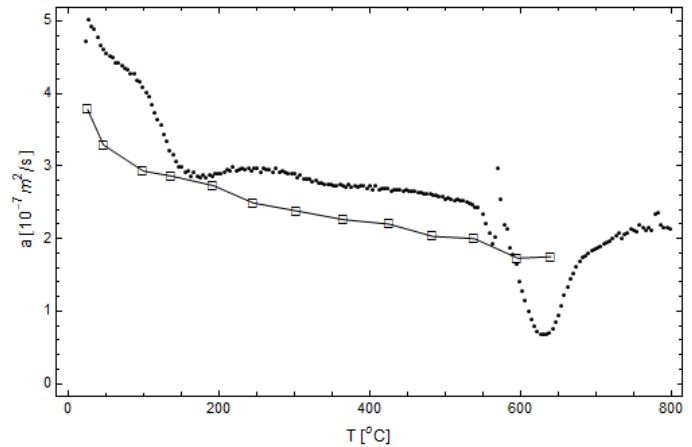
Figure 3 The temperature dependence of the thermal diffusivity calculated by the inverse method (dots) and measured by the flash method (squares)

Finally, above ~ 650 °C where the dehydroxylation is finished, the thermal diffusivity increases fast, to $\sim 2.2 \times 10^{-7}$ m^2/s. After that it is almost constant; at these temperatures solid-state sintering begins.

Accuracy of the dependence $a(T)$ obtained by the inverse analysis and shown in Figure 3 may be verified by using the dependence to solve the direct problem. That is, we may numerically solve the heat equation (1) with the thermal diffusivity from Figure 3 and compare the result with the

experimental data in Figure 2. The two sets of results are in very good agreement in the whole studied temperature range, the root mean square error being only 0.82 K.

Finally, in Figure 3 we also present the results on the temperature dependence of a as measured by the flash method. Clearly, these do not enable one to detect the processes that occur in the studied ceramic material during firing. Indeed, the profile of $a(T)$ exhibits a steady decrease from its initial value ~ 3.8×10^{-7} m^2/s (at 20 °C) to ~ 1.8×10^{-7} m^2/s (at 650 °C). The only exception is between 20 and 150 °C where the decrease is visible and is associated with the liberation of physically bounded water. Note that the thermal diffusivity as obtained by the flash method is at most temperatures slightly lower than the one obtained by the inverse method.

CONLUSIONS

We employed an inverse method to estimate the temperature dependence of the thermal diffusivity for an alumina ceramic material used in the electro-porcelain industry in the temperature range 20 – 800 °C. The results were able to record the physical-chemical processes occurring in the material during firing: the liberation of physically bound water (20 – 200 °C) and the dehydroxylation of kaolinite into metakaolinite (550 – 650 °C). Our numerical results were verified by using the obtained thermal diffusivity to solve the direct problem and compare the resulting temperature field with the experimental data. Very good agreement was achieved. We also employed the standard flash method to measure the thermal diffusivity. However, due to its isothermal nature, the methods yielded results that could not detect the processes taking place in the material during firing.

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