OPTIMAL PARAMETERIZATION IN THE MEASUREMENTS OF THE THERMAL DIFFUSIVITY OF THERMAL BARRIER COATINGS

by

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The paper presents an estimation procedure for the measurement of the thermal diffusivity of thermal barrier coatings deposited on thermal conductive substrates using the laser flash method when the thermal contact resistance between the coating and substrate is unknown. The procedure is based on the application of the optimal parameterization technique and Gauss minimization algorithm. It has been applied on the experimental data obtained by using two different samples, one made of PTFE (polytetrafluoroethylene) coating deposited on a stainless steel substrate and the other made of PVC (polyvinylchloride) deposited on a copper substrate.

Key words: *laser flash method, optimal parameterization, parameter estimation, thermal barrier coatings, thermal contact resistance, thermal diffusivity*

Introduction

The laser flash method, originally established by Parker *et al*. [1], is the standard technique for thermal diffusivity measurements of solid materials. In this method, one side of a small disk-shaped sample is exposed to a single pulse laser beam and the temperature change due to the heat diffusion through the sample is detected from the other sample side. Related data reduction implies the consideration of several specific points of a measured temperature signal and the correction of different physical effects such as the heat loss and finite pulse time (see refs. [2] and [3], for example).

Although the laser flash method is not standardized for thermal diffusivity measurements of multi-layer materials or thin films and coatings, there are many papers in literature proposing it for such using. In that sense, Larson and Koyama [4] and Bulmer and Taylor [5] analyzed one-dimensional transient heat conduction through a double-layer sample, taking in consideration finite pulse effects and various pulse shapes, but neglecting the influence of thermal contact resistance between the layers. On the other hand, Chistyakov [6] and Hartmann *et al*. [7] considered the presence of previously measured thermal contact resistance, but ignored heat exchange between sample and environment.

Lee *et al*. [8] contributed to thermal diffusivity measurements of multi-layered structures with the particular analysis of the thermal behavior of layers, while Shoemaker [9] described different limitations of the laser flash method and proposed the using of a two-dimensional model of heat diffusion for measuring thermal diffusivity of such structures. Also, Degiovanni [10] proposed and applied particular transfer functions or quadruples for modeling transient heat conduction through multi-layered systems, while Milošević *et al*. [11] gave the exact analytical solution for the transient temperature of the rear sample side implying both heat loss and pulse duration effects, as well as the influence of a finite thermal contact resistance.

In the case when the thermal contact resistance between the coating and substrate is unknown, an extraction of reliable information on thermal diffusivity of coating can be difficult. A proportional influence of thermal diffusivity and thermal contact resistance on measured data restricts seriously simultaneous estimation of these two properties. Some experimental solutions have been proposed for the measurement of the thermal diffusivity of dielectric films deposited on conducting substrates and related thermal contact resistance (for example, pulsed electrothermal technique by Hobbie and De Reggi [12] or two-dimensional laser flash method by Milošević *et al*. [13]), but, they do not possess some important advantages of the classical laser flash method, such as measurement on high temperatures, small and simple samples, *etc*.

The problem of simultaneous determination of thermal diffusivity of coatings and thermal contact resistance between coating and substrate using the laser flash method can be overcome by the application of an optimal parameterization, which is based on a recently proposed technique by Martinsons [14]. In this approach, instead of estimating a set of original unknown properties, a new group of unknown parameters whose sensitivity coefficients are linearly independent is to be computed in each step of the estimation procedure. New parameters are dimensionless and represent a particular combination of related original properties. In such a way, under certain experimental conditions, like that of a high signal-noise ratio, the simultaneous estimation of thermal diffusivity of coatings and thermal contact resistance can be performed successfully.

Statement of the problem

Mathematical model

In the laser flash method the measuring signal represents the temperature evolution of the rear sample side after the laser pulse absorption at the front sample side. The form of the sample and related physical properties considered in this research are given in fig. 1. Both substrate and coating materials are described by thermal diffusivity, heat capacity, and density, the contact between them by thermal contact resistance, while the heat loss rate by heat transfer coefficients. The laser pulse is described by duration τ, while the energy absorbed at the front surface by parameter Q.

The analytical solution of the transient temperature of the coating surface T can be obtained by using different analytical methods, such as the separation of variables

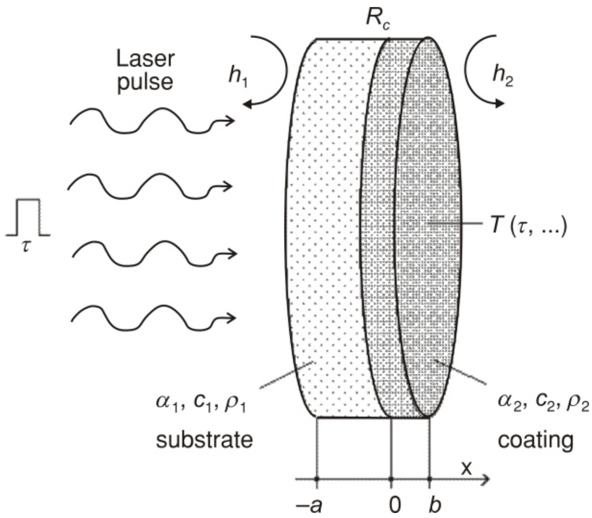

Figure 1. The form of the sample and related parameters

technique, for example. For the particular case given in fig. 1 and assuming the laser pulse of the quadratic form, the explicit solution can be written as:

$$T \quad \frac{Q}{\rho_1 c_1 \tau}\sqrt{\frac{\alpha_1}{\alpha_2}} \; {}^{\infty}_{n \;\; 1} \frac{1}{\beta_n^2 N_n}(\sin\varphi_{1n} \quad s_{1n}\cos\varphi_{1n})(-\sin\varphi_{2n} \quad s_{2n}\cos\varphi_{2n})(\mathrm{e}^{\beta_n^2 \tau} \quad 1)\mathrm{e}^{\;\;\beta_n^2 t} \tag{1}$$

where the functions N_n, s_{1n}, s_{2n}, φ_{1n}, and φ_{2n} are given by:

$$N_n \quad \frac{a}{2\varphi_{1n}} \quad \varphi_{1n}(1 \quad s_{1n}^2) \quad \frac{1}{2}\sin 2\varphi_{1n}(1 \quad s_{1n}^2) \quad s_{1n}(1 \quad \cos 2\varphi_{1n})$$
$$\frac{b}{2\varphi_{2n}}\frac{k_1}{k_2} \quad \varphi_{2n}(1 \quad s_{2n}^2) \quad \frac{1}{2}\sin 2\varphi_{2n}(1 \quad s_{2n}^2) \quad s_{2n}(1 \quad \cos 2\varphi_{2n})$$

$$s_{1n} \quad \frac{k_1\varphi_{1n}\cos\varphi_{1n} \quad ah_1\sin\varphi_{1n}}{k_1\varphi_{1n}\sin\varphi_{1n} \quad ah_1\cos\varphi_{1n}}, \quad s_{2n} \quad \frac{k_2\varphi_{2n}\cos\varphi_{2n} \quad bh_2\sin\varphi_{2n}}{k_2\varphi_{2n}\sin\varphi_{2n} \quad bh_2\cos\varphi_{2n}},$$

$$\varphi_{1n} \quad \frac{\beta_n a}{\sqrt{\alpha_1}}, \quad \varphi_{2n} \quad \frac{\beta_n b}{\sqrt{\alpha_2}}$$

and parameters k_1 and k_2 are related thermal conductivity ($k = \rho c a$). Positive numbers β_n represent the solutions of following transcendental equation:

$$\frac{k_2}{\sqrt{\alpha_2}} s_{1n} \quad \frac{k_1}{\sqrt{\alpha_1}} s_{2n} \quad \frac{k_1 k_2 R_c}{\sqrt{\alpha_1 \alpha_2}} \beta_n \quad 0 \tag{2}$$

However, instead of resolving eq. (2) β_n numbers can be found by the efficient and reliable "sign-count" method proposed by Mikhailov and Vulchanov [15]. According to that procedure, the number of eigenvalues $N(\beta_s, S)$ below an arbitrary value β_s is equal to:

$$N(\beta_s, S) \quad \text{int} \ \frac{\varphi_{1s}}{\pi} \quad \text{int} \ \frac{\varphi_{2s}}{\pi} \quad N[S(\beta_s)] \tag{3}$$

where $N[S(\beta_s)]$ is the number of negative elements of the set $S(\beta_s) = \{d_1, d_2, d_3, d_4\}$, defined by:

$$d_1 \quad \frac{k_1}{a} \varphi_{1s} \cot \varphi_{1s} \quad h_1, \quad d_2 \quad \frac{k_1}{a} \varphi_{1s} \cot \varphi_{1s} \quad \frac{1}{R_c} \quad \frac{1}{d_1} \quad \frac{k_1}{a} \frac{\varphi_{1s}}{\sin \varphi_{1s}} \quad ^2$$

$$d_3 \quad \frac{k_2}{b} \varphi_{2s} \cot \varphi_{2s} \quad \frac{1}{R_c} \quad \frac{1}{d_2 R_c^2}, \quad d_4 \quad \frac{k_2}{b} \varphi_{2s} \cot \varphi_{2s} \quad h_2 \quad \frac{1}{d_3} \quad \frac{k_2}{b} \frac{\varphi_{2s}}{\sin \varphi_{2s}} \quad ^2$$

If one varies the value β_s by using eq. (3) and the principle of dichotomy, the determination of eigenvalues β_n is straightforward. Number of eigenvalues β_n needed for the computation of series in eq. (1) depends on the solution convergence and the required accuracy.

Sensitivity analysis

According to the mathematical model, there are 13 physical parameters: a, b, α_1, α_2, c_1, c_2, ρ_1, ρ_2, h_1, h_2, R_c, τ, and Q. Among them, three parameters have been considered as unknown in this research: thermal diffusivity of coating, α_2, thermal contact resistance, R_c, and absorbed energy per unit of surface, Q. Other 10 parameters have been taken as known with negligible uncertainties.

Let define a column-vector **z** that represents three unknown parameters and a scalar σ_T that corresponds to the standard uncertainty of the measured temperature response which is assumed constant through the whole measurement range. Then, the matrix of sensitivity coefficients, $\mathbf{X_z}$, has dimension $[n \times 3]$, where n is the number of measured data. If one assumes that there is no correlation between measured data, the covariance matrix of measured data **W** is the identity matrix multiplied with the variance σ_T^2 or $\mathbf{W} \quad \sigma_T^2 \mathbf{I}$.

For the best simultaneous estimation of two or more parameters, the determinant of the covariance matrix of related parameters, $\mathbf{W_z}$, should be minimal [16]. If there is no information a priori, the covariance matrix of parameters for estimation is equal to $\sigma_T{}^2(\mathbf{X}_\mathbf{z}^T\mathbf{X}_\mathbf{z})^{-1}$ and its diagonal elements represent the variances of the Cramér-Rao bound of each parameter for estimation, while those extra-diagonal the covariances between the same parameters. In other words, unknown parameters are simultaneously estimable if the extra-diagonal elements of $\mathbf{W}_\mathbf{z}$ tend to zero and those diagonal are kept as small as possible. For satisfying these conditions the sensitivity coefficients from $\mathbf{X}_\mathbf{z}$ must be linearly independent and their reduced forms (which represent the sensitivity coefficients multiplied by related parameters) must be above the level of the standard uncertainty σ_T in the range of consideration.

In the problem of this research, the sensitivity coefficients of the thermal diffusivity of coating and thermal contact resistance are linearly dependent in general. As an example, taking the parametric values from the experimental part of this research, *i. e.*, from tab. 1 (specimen II), tab. 2, and tab. 3 (combination 3-V), the reduced sensitivity coefficients of three unknown parameters and the level of the standard measurement uncertainty is presented in fig. 2. According to that, it can be seen a strong linear dependence between the sensitivity coefficients of thermal diffusivity and thermal contact resistance, $X^*_{\alpha_2}$ and $X^*_{R_c}$.

In addition to the linear dependence, for high values of thermal diffusivity of coating their reduced sensitivity coefficients may be below the level of standard deviation and the estimation of this parameter becomes impossible. However, while the level of the standard measurement uncertainty can be minimized by reducing the measurement noise, the linear dependence between the sensitivity coefficients can be changed only by using the optimal parameterization.

Applied estimation procedure

If there is no information *a priori* of parameters for estimation, one should apply the maximum likelihood (ML) estimator. According to this estimator, assuming the Gauss distribution of errors, the iterative equation for minimizing the difference between measured and computed values is:

$$\mathbf{z}^{(k+1)} = \mathbf{z}^{(k)} + [\mathbf{J}_\mathbf{z}^{(k)}]^{-1}\{\mathbf{X}_\mathbf{z}^{\mathrm{t}(k)}\mathbf{W}^{-1}[\mathbf{Y} - \mathbf{T}^{(k)}\mathbf{z}^{(k)}]\} \tag{4}$$

where $\mathbf{Y}$ and $\mathbf{T}$ are the column-vectors of n measured and computed values, respectively, and $\mathbf{J}_\mathbf{z}$ is the Fisher information matrix equal to:

$$\mathbf{J}_\mathbf{z}^{(k)} = \mathbf{X}_\mathbf{z}^{\mathrm{t}(k)}\mathbf{W}^{-1}\mathbf{X}_\mathbf{z}^{(k)} \tag{5}$$

The diagonal elements of the inverse Fisher information matrix are the variances of parameters for estimation, while the extra-diagonal correspond to the covariances between them. While the first ones are responsible for the precision of estimated parameters, the extra-diagonal elements describe the possibilities of simultaneous estimation of two related parameters. In that sense, the parameters are optimal for the estimation when all extra-diagonal elements of the matrix $\mathbf{J}_{\mathbf{z}}^{\;1}$ are asymptotically equal to zero. In other words, in order to find a set of optimal parameters **p**, a diagonal of the inverse Fisher information matrix needs to be extracted.

Diagonalization of any square matrix means to find its eigenvalues and eigenvectors, but it has no sense if parameters for estimation have any physical dimension [14]. From that reason, one must substitute all related parameters with physical dimension with their dimensionless counterparts. In this case, dimensionless parameters that correspond to thermal diffusivity of coating α_2, thermal contact resistance R_c, and absorbed energy Q can be defined as[*]:

$$q_1 \quad \frac{\tau}{a^2}\alpha_2 \tag{6}$$

$$q_2 \quad \frac{\rho_1 c_1 \alpha_1}{a} R_c \tag{7}$$

$$q_3 \quad \frac{1}{\rho_1 c_1 a} Q \tag{8}$$

Each of them is a product of only one unknown and several known parameters with physical dimension. In above definitions, only known properties which relate to the substrate are used because they are better known in practice than those which correspond to the coating.

Having the column-vector of dimensionless parameters **q**, a following non-linear combination can be created:

$$p_j \quad {}_{m\;1}^{\;3}\; q_m^{r_{jm}} \tag{9}$$

where j = 1, 2, and 3 and the numbers r_{jm} are to be found from the process of diagonalization. Applying the logarithmic transformation of both sides of eq. (9), one obtains:

$$\log p_j \quad {}_{m\;1}^{\;3}\; r_{jm} \log \mathbf{q}_m \tag{10}$$

or log **p** = **R** log **q** in the matrix notation where **R** is the matrix of r_{jm} elements. Having the matrix of sensitivity coefficients of logarithmic parameters with a typical element:

[*] This set of dimensionless parameters is a result of the, so called, π-theorem, described in [14]. However, only unknown parameters have been substitute with the hew parametric set.

$$(\mathbf{X}_{\log \mathbf{q}})_{ij} = \frac{\partial T_i}{\partial (\log \mathbf{q}_j)} = \mathbf{q}_j \frac{\partial T_i}{\partial \mathbf{q}_j} = (\mathbf{X}^*_{\mathbf{q}})_{ij} \tag{11}$$

the inverse Fisher information matrix of logarithmic parameters log **q** becomes:

$$\mathbf{J}^{-1}_{\log \mathbf{q}} = \left[\left(\mathbf{X}^{*\mathrm{t}}_{\mathbf{q}} \mathbf{W}^{-1} \mathbf{X}^*_{\mathbf{q}}\right)^{-1}\right]^{-1} \tag{12}$$

Once the matrix $\mathbf{J}_{\log \mathbf{q}}$ is created, the diagonalization of its inverse can be applied as:

$$\mathbf{J}^{-1}_{\log \mathbf{q}} = \mathbf{U}^{\mathrm{t}} \mathbf{V} \mathbf{U} \tag{13}$$

where the eigenvalues are diagonal elements of the diagonal matrix **V**, while the eigenvectors are the columns of the transformation matrix **U**. The matrix **U** is orthogonal, *i. e.*, $\mathbf{U}^{\mathrm{t}} = \mathbf{U}^{-1}$. Because the eigenvectors are linearly independent, substituting the matrix **R** from eq. (10) by the matrix $\mathbf{U}^{\mathrm{t}}$, the vector **p** becomes the optimal set of parameters with the typical element:

$$p_j = \prod_{m=1}^{3} q_m^{(\mathbf{U}^{\mathrm{t}})_{jm}} \tag{14}$$

The eigenvalues of the diagonal matrix **V** represent the variances of logarithmic parameters or the square of the relative standard deviation of parameters **p**, *i. e.*:

$$\mathbf{V}_{jj} = \sigma^2_{\log p_j} = \left(\frac{\sigma_{p_j}}{p_j}\right)^2 \tag{15}$$

These values indicate which optimal parameter can be estimated precisely. Namely, only parameters with small logarithmic variances are used for the correction in the iterative procedure.

After the diagonalization of the matrix $\mathbf{J}_{\log \mathbf{q}}$ the matrix of reduced sensitivity coefficients of the optimal parameters **p** can be computed using the simple formula:

$$\mathbf{X}^*_{\mathbf{p}} = \mathbf{X}^*_{\mathbf{q}} \mathbf{U}^{\mathrm{t}} \tag{16}$$

The usual matrix of sensitivity coefficients of the optimal parameters $\mathbf{X}_{\mathbf{p}}$ is obtained by dividing each column of the matrix $\mathbf{X}^*_{\mathbf{p}}$ with the related parameter **p**.

Changing the vector and matrix **z** and $\mathbf{X}_{\mathbf{z}}$ in eq. (4) by **p** and $\mathbf{X}_{\mathbf{p}}$, respectively, the Gauss iterative equation becomes:

$$\mathbf{p}^{(k+1)} = \mathbf{p}^{(k)} + [\mathbf{X}^{\mathrm{t}(k)}_{\mathbf{p}} \mathbf{W}^{-1} \mathbf{X}^{(k)}_{\mathbf{p}}]^{-1} \{\mathbf{X}^{\mathrm{t}(k)}_{\mathbf{p}} \mathbf{W}^{-1} [\mathbf{Y} - \mathbf{T}^{(k)}(\mathbf{p}^{(k)})]\} \tag{17}$$

As stated above, parameters with a large related value from the matrix **V** cannot be estimated precisely, so they should be fixed in the current iteration. Obviously, a new optimal set of parameters needs to be created for each iteration step.

There are three conditions which must be satisfied for reaching the convergence as described in [14]: the cost function should be minimal, the correction of **p** from eq. (17) must tend to zero, and the orthogonal matrix **U** from eq. (13) must tend to identity matrix. If the criterion of convergence is satisfied, the final values of the initial parameters **q** are extracted from the set of optimal parameters **p** from the last iteration. Using eq. (14) and the fact that the matrix **U** is orthogonal one obtains:

$$q_j = \sum_{m=1}^{3} p_m (\mathbf{U}^t)^{-1}_{jm} = \sum_{m=1}^{3} p_m (\mathbf{U})_{jm} \tag{18}$$

On the other hand, the final uncertainties of the parameters **q** can be derived from the inverse of the Fisher information matrix from the final iteration

$$[\mathbf{J}_\mathbf{p}^{\text{final}}]^{-1} = [\mathbf{X}_\mathbf{p}^{t\,(\text{final})}\mathbf{W}^{-1}\mathbf{X}_\mathbf{p}^{(\text{final})}] \tag{19}$$

which are equivalent to the diagonal elements of the matrix $\mathbf{V}^{\text{final}}$.

Experiments

In order to demonstrate the application of proposed estimation procedure, two samples have been taken for the experiment. The first, made of a coating of the polytetrafluoroethylene (PTFE) doped with carbon and deposited on a stainless steel substrate, and the second, made of with a polyvinylchloride (PVC) coating on a copper substrate.

Sample 1: PTFE+C – coating, stainless steel – substrate

The coating deposition is a common process in the industry of kitchen tools, especially pans. The coating of polymer is useful for the protection of metallic body and its thermal isolation. For the purpose of this research, a commercially available pan of the producer SITRAM® has been selected for the specimen preparation.

Specimens and measurements

The bottom of the pan was made of a thick stainless steel base coated by a thin layer of PTFE doped with 30% of carbon fibers. The central part of the pan's bottom was about 200 mm in diameter and about 3 mm in thickness. Two specimens in the form of disk were manufactured from that part of the pan: one of the steel basement only, and other with the original polymer coating. The first specimen was used for classical thermal diffusivity measurements of the steel basement, while the second for the measurements of thermal diffusivity of coating. Data obtained from the first specimen were necessary for the measurements performed on the second sample.

Temperature responses were recorded using the laser flash apparatus of following properties: the ruby pulse laser beam of about 15 mm in diameter, 30 J of maximum output energy, and 1 ms of pulse duration. The specimens were held in a specially designed vacuum chamber which has two windows, allowing the pulse heating of front and the temperature detection of the rear specimen side. Transient temperature response was measured by a sensitive InSb IR detector and recorded by a high-precision data-acquisition system. All the measurements were performed at room temperature.

All the values of parameters related to specimen dimensions, pulse duration, and average noise of temperature responses are presented in tab. 1. Data on specimen diameters D are only informative as far as they are not involved in the physical model and estimation procedure from above. The relative noise of measured transient temperatures was different for two specimens due to different emissivities of two related surfaces.

Table 1. Specimen dimensions, pulse duration, and average relative standard measurement uncertainty for sample 1

Specimen	a [mm]	b [μm]	D [mm]	τ [ms]	σ_T [%]
I	2.01	–	10	1	1.1
II	2.07	80			0.6

Thermophysical properties of substrate and coating

In order to determine the heat capacity of the substrate stainless steel, a chemical analysis with the mass spectroscopy method was carried out. According to that, the used stainless steel had the following composition in weight percent: 84.1% Fe, 14.4% Cr, 0.8% Mo, 0.3% Ni, 0.17% Si, 0.06 Mg and Mn, 0.05 V, 0.04 Cu, and 0.01 Zn. By applying the Kopp-Neumann rule and reference data taken from Touloukian and Buyco [17], the value of 450 J/kgK for the heat capacity of the substrate material was obtained. However, in order to verify this value, the heat capacity was also determined experimentally by the ballistic calorimetry method*. Results of those measurements suggested a value of 460 J/kgK, which was only about 2.5% higher than the computed one.

The density of the steel was measured directly by using the dimensions and mass of specimen I, and the value was 7449 kg/m^3.

* This formula is valid when the temperature difference between the surface and environment is small ans the heat exchange by convection is negligible, which is the case when the experiments are performed at the room temperature and under the vacuum conditions.

The thermal diffusivity of steel was determined by the standard laser flash method and, in total, 8 temperature responses were recorded from specimen I. The final value of $12.5 \cdot 10^{-6}$ m^2/s was obtained for this property.

Regarding the sample coating, the values of its heat capacity and density were taken from the database www.matweb.com as 1100 J/kgK and 2100, respectively, for the PTFE with 30 to 50% of carbon filling. On the other hand, being one of three parameters for estimation, the value of the coating thermal diffusivity was used only as the initial value in the first step of the iterative eq. (4). In that sense, eight *a priori* values in the range from $0.06 \cdot 10^{-6}$ to $40 \cdot 10^{-6}$ m^2/s were taken for the estimation procedure. The range was chosen by considering the reference value for pure PTFE, which was about $1.12 \cdot 10^{-6}$ m^2/s [19].

The same logic was applied for the thermal contact resistance where, in total, nine values from $0.6 \cdot 10^{-6}$ to $800 \cdot 10^{-6}$ m^{2}K/W were used for the first step of the estimation procedure. At the same moment, however, due to very good estimation possibilities for the absorbed energy *Q*, only one *a priori* value of this property was taken and it was 50 J/m^2.

Finally, the values of heat transfer coefficients for surfaces of both materials were computed using the approximate formula*, $h \quad 4\sigma_{sb}\varepsilon T_{\text{ref}}^3$ where σ_{sb} is Stefan-Boltzmann constant (5.67 10^{-8} W/m^{2}K^4), ε emissivity of related surface, and T_{ref} specimen referential temperature. Taking the emissivity of the steel surface as 0.5 [18] and assuming the emissivity of the coating surface as 0.8, the heat transfer coefficients h_1 and h_2 for the referential temperature of 24 °C are equal to 3.0 and 4.8 W/m^{2}K, respectively.

All the value s of known thermophysical properties of substrate and coating are given in tab. 2, while the a priori values of parameters for estimation in tab. 3 Each *a priori* value of the thermal diffusivity of coating and thermal contact resistance has been numerated in order to analyze conveniently estimation possibilities and discuss following results.

Table 2. Known thermophysical properties of the substrate and coating of sample 1

Specimen	ρ [kg/m^3]	ρ_2 [kg/m^3]	c_1 [J/kgK]	c_2 [J/kgK]	α_1 [10^{-6} m^2/s]	h_1 [W/m^{2}K]	h_2 [W/m^{2}K]
II	7449	2100	460	1100	12.5	3.0	4.8

Sensitivity analysis

Having the values of all known and a priori parameters, the computation of related sensitivity coefficients and analysis of the estimation possibilities could be performed. As a result, the reduced sensitivity coefficients of three original parameters for estimation from tab. 3 (3-V combination of *a priori* parameters) and the level of standard measure-

* This formula is valid when the temperature difference between the surface and environment is small ans the heat exchange by convection is negligible, which is the case when the experiments are performed at the room temperature and under the vacuum conditions.

Table 3. *a priori* values of unknown parameters of sample 1

Specimen	$\alpha_{2\,ap}$ [10^{-6} m^2/s]	$R_{c\,ap}$ [10^{-6} m^{2}K/W]	Q_{ap} [J/m^2]
II	0.06 (estim. 1) 0.1 (estim. 2) 0.4 (estim. 3) 0.8 (estim. 4) 2 (estim. 5) 6 (estim. 6) 10 (estim. 7) 40 (estim. 8)	0.6 (estim. I) 1 (estim. II) 4 (estim. III) 8 (estim. IV) 20 (estim. V) 60 (estim. VI) 100 (estim. VII) 400 (estim. VIII) 800 (estim. IX)	50

ment uncertainty are already presented in fig. 2. However, if one substitutes the original parameters from tab. 3 with those from eqs. (6) to (8) and performs the optimization as described in section 3, one obtains a new set of parameters. Thus, for example, for combination 3-V, the transformation matrix **U** and the matrix of eigenvalues **V** are equal to:

$$\mathbf{U} \quad \begin{matrix} 3.603\ 10^{1} & 9.328\ 10^{1} & 1.835\ 10^{5} \\ 9.328\ 10^{1} & 3.603\ 10^{1} & 4.880\ 10^{3} \\ 4.559\ 10^{3} & 1.741\ 10^{3} & 1 \end{matrix} \tag{20}$$

$$\mathbf{V} \quad \begin{matrix} 34.5 \\ 0.915 \\ 0.012 \end{matrix} \tag{21}$$

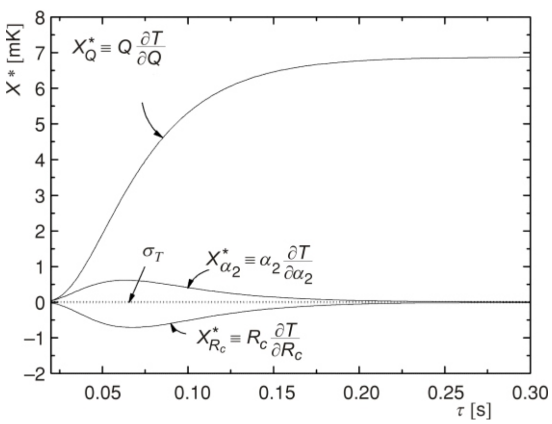

Figure 2. An example of the reduced sensitivity coefficients of three original unknown parameters

The reduced sensitivity coefficients of the parameters created from eqs. (14) and (20) are given in fig. 3. It can be seen that all the sensitivity coefficients are linearly independent, so the simultaneous estimation of related parameters is theoretically possible. However, because the reduced sensitivity coefficient of the optimal parameter p_1 is below the level of standard measurement uncertainty, this parameter cannot be estimated and should be fixed during the actual iteration step.

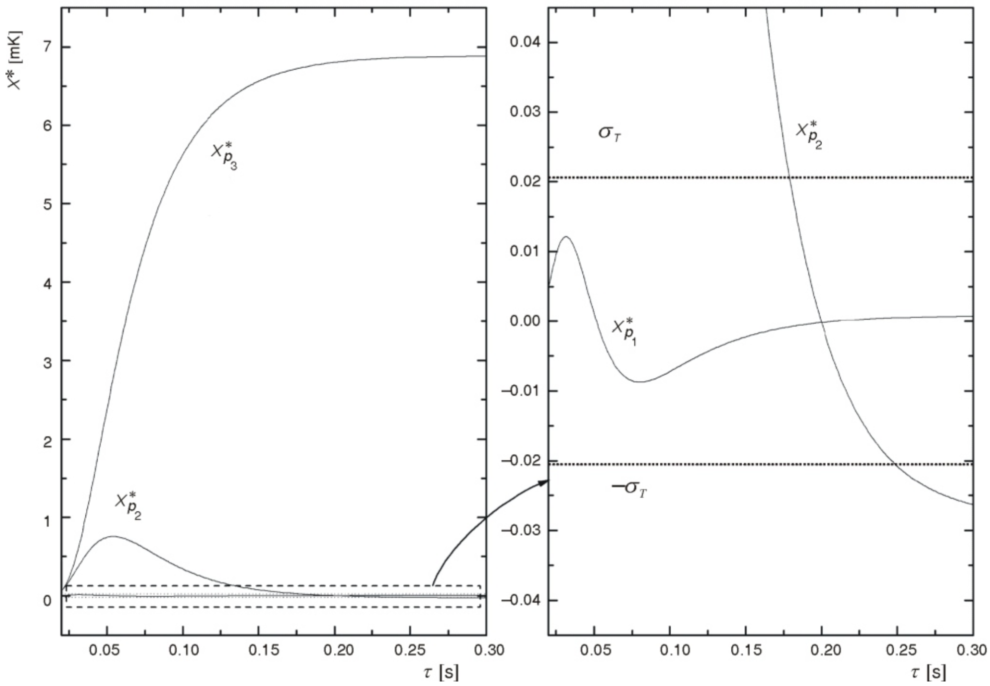

Figure 3. Reduced sensitivity coefficients of three optimal parameters of sample 1

By comparing fig. 2 and fig. 3, the benefit of the optimal parameterization in this case is that the sensitivity coefficients of new parameters are linearly independent making thus possible their simultaneous estimation. However, due to a relatively high level of measurement uncertainty, the identification of certain optimal parameters is impossible. In such a case, the optimal parameterization helps to choose which parameters should be estimated and which fixed in each step of iterative eq. (17).

Experimental results

There were 4 transient temperature responses measured from the back surface of specimen II. According to the *a priori* values from tab. 3 and the values of known param-

Table 4. Final results on thermal diffusivity of the coating and thermal contact resistance of sample 1

Response No.	*a priori* value [m^2K/W]	α_2 [10^{-6} m^2/s]	$\alpha_{2\,av}$ [10^{-6} m^2/s]	$U_{\alpha_2 av}$[%]	R_c [10^{-6} m^2K/W]	$R_{c\,av}$ [10^{-6} m^2K/W]	$U_{R_c av}$ [%]
1	$R_{c\,ap}$ 8 10^{-6}	0.243	0.235	6.2	86.8	83.9	5.7
2		0.242			80.2		
3		0.229			83.8		
4		0.227			84.9		
1	$R_{c\,ap}$ 8 10^{-6}	Diverged	–	–	Same as related *a priori* value	–	
2							
3							
4							

eters from tab. 1 and tab. 2, 72 optimal estimations have been performed for each response, 288 totally. The value of the cost function was used as a criterion of the estimation quality. As a result, two sets of final values have been obtained: first, when the thermal contact resistance is lower than 100 10^{-6} m^2K/W and second, when the same parameter is equal or higher than that value.

All the results of thermal diffusivity and thermal contact resistance, their mean values and expanded uncertainties (coverage factor 2) of the mean values are presented in tab. 4. According to them, the thermal diffusivity has been determined successfully together with the thermal contact resistance only by not assuming a very low thermal contact resistance *a priori* value, *i. e.*, below $8 \cdot 10^{-6}$ m^2K/W.

On the other side, by applying the estimation with the original parameters, in most cases of the *a priori* values the iterative procedure diverges for either thermal diffusivity or thermal contact resistance or both of them, which is the consequence of the mutual linearity of related sensitivity coefficients. The fixation of parameters could help in this case only if one has the results of the optimal estimation.

In comparison with the literature value of $1.12 \cdot 10^{-6}$ m^2/s [19], which corresponds to the pure PTFE, the present results on thermal diffusivity is higher, which is probably due to the carbon filling of the PTFE coating. Also, it should be mentioned that the present results were obtained assuming the perfect knowledge of known properties. If one considered the uncertainties of known parameters, the results would have had larger confidence region.

Sample 2: PVC – coating, copper – substrate

The application of polymer tapes for joining and isolation of electrical conductors is very common. However, polymer tapes represent a thermal barrier for heat transfer between the conductor and environment and in some cases such thermal isolation needs to be quantified, which means a knowledge of thermal transport properties of applied tapes. In this example, therefore, the determination of the thermal diffusivity of a common PVC tape with the commercial name Scotch® has been performed.

Sample and measurements

The adhesive side of a black Scotch® isolation tape was manually pressed to one surface of a thin copper disk. The copper surface was entirely covered by the tape and the surplus of the tape was removed from the edges of the surface by a thin blade. All dimensions of the copper disk as well as the thickness of the tape were measured before the adhesion. Temperature responses were recorded at room temperature using the same apparatus as in the previous example. Sample dimensions, pulse duration, and the standard uncertainty of temperature responses are given in tab. 5.

Table 5. Specimen dimensions, pulse duration, and average relative standard measurement uncertainty for sample 2

a [mm]	b [μm]	D [mm]	τ [ms]	σ_T [%]
1.49	120	10	1	0.6

Thermophysical properties of substrate and coating

In order to measure precisely the tape density, 25 different parts with regular dimensions have been cut from the tape and the average value of about 1360 kg/m^3 was obtained. Considering the heat capacity of the tape material, it was assumed to be 1100 J/kgK using the reference [20].

Regarding the copper substrate, its density was measured directly from the disk, while the values of heat capacity and thermal diffusivity were taken from the Touloukian and Buyco [17] and Maglić and Milošević [21], respectively.

On the same manner as in the previous example, the heat transfer coefficients were computed using the given approximate formula. The emissivity of the free copper surface was taken to be 0.03 [18], while that of the coating was assumed to be 0.8.

Finally, eight *a priori* values of the thermal diffusivity, three of the thermal contact resistance, and one of the absorbed energy were taken for the estimation procedure. All the values of thermophysical properties are presented in tab. 6 (known parameters) and tab. 7 (parameters for estimation).

Table 6. Known thermophysical properties of the substrate and coating of sample 2

ρ [kg/m^3]	ρ_2 [kg/m^3]	c_1 [J/kgK]	c_2 [J/kgK]	α_1 [10^{-6} m^2/s]	h_1 [W/m^{2}K]	h_2 [W/m^{2}K]
8930	1361	386.5	1100	121	0.2	4.4

Sensitivity analysis

The reduced sensitivity coefficients of three parameters for estimation from tab. 7 for the combinations of *a priori* values 3-I and 3-III are presented in fig. 4, together with the level of standard measurement uncertainty. According to that, thermal diffusivity can be estimated simultaneously with thermal contact resistance only in cases when the latter parameter has a high value. Otherwise, related sensitivity coefficients are linearly dependent and one needs to apply the optimal parameterization.

Table 7. ***A priori*** **values of unknown parameters of sample 2**

$\alpha_{2\,ap}$ [10^{-6} m^2/s]	$R_{c\,ap}$ [10^{-6} m^{2}K/W]	Q_{ap} [J/m^2]
0.06 (estim. 1)		
0.1 (estim. 2)	1 (estim. I)	
0.16 (estim. 3)	10 (estim. II)	50
0.2 (estim. 4)	100 (estim.III)	
0.6 (estim. 5)	1000 (estim. IV)	
1 (estim. 6)		

Performing described optimization procedure, two optimal sets of parameters for combinations 3-I and 3-III with related transformation matrix **U** have been obtained. Their reduced sensitivity coefficients are given in fig. 5 where one can find that all of

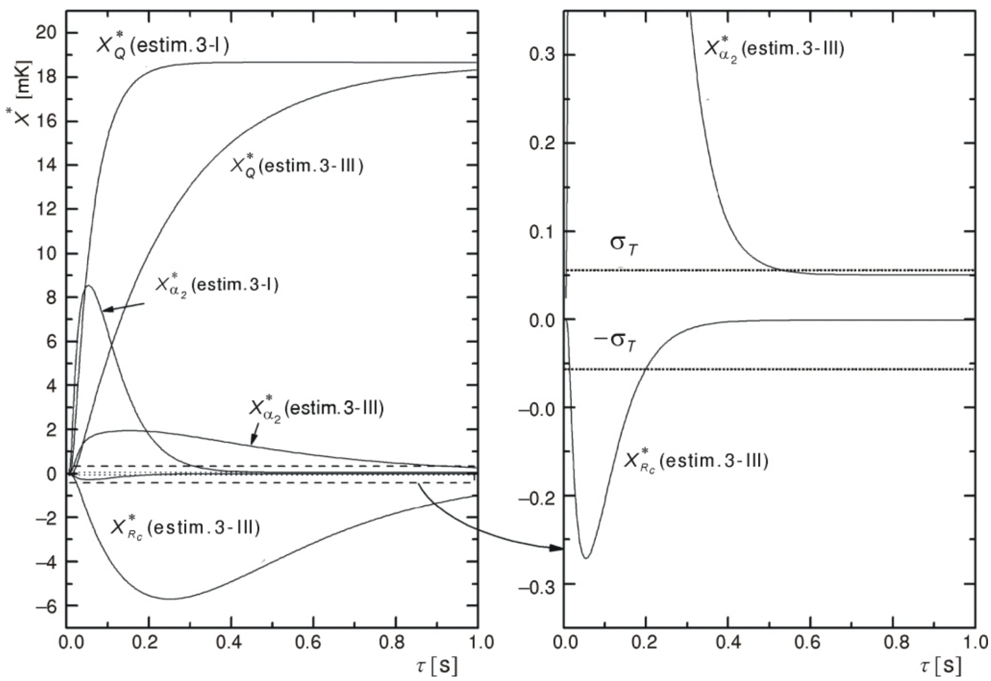

Figure 4. Reduced sensitivity coefficients of three original unknown parameters of sample 2

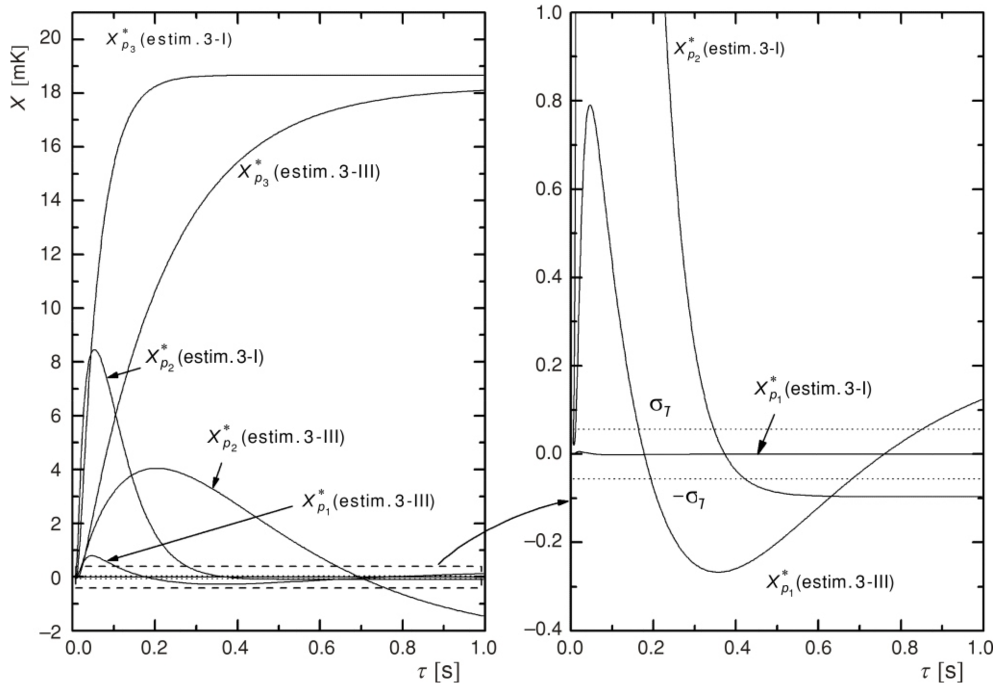

Figure 5. Reduced sensitivity coefficients of three optimal parameters of sample 2

them are linearly independent for corresponding combination of *a priori* values. On the other hand, the reduced sensitivity coefficient of the parameter p_1 from estimation 3-I is completely covered by the standard measurement uncertainty, so it must be fixed in actual step of the iterative procedure. Also, as the parameter p_1 from estimation 3-I is mostly related to dimensionless thermal contact resistance q_2, one can conclude in this case that the parameter R_c cannot be determined if its value is very low as was the case in the previous example.

Experimental results

Totally, four transient temperature responses were measured from the coating surface of sample 2 and 12 estimation procedures, 3 for each response. Final results of thermal diffusivity and thermal contact resistance, their mean values and expanded uncertainties (coverage factor 2) are presented in tab. 8.

The optimal estimation procedure applied in this particular case has given the results as expected: thermal diffusivity and thermal contact resistance have been determined simultaneously with success, but only when the *a priori* value of the thermal contact resistance have been equal or greater than $10 \cdot 10^{-6}$ m^{2}K/W.

Table 8. Final results on thermal diffusivity of the coating and thermal contact resistance of sample 2

Response No.	*a priori* value [m^{2}K/W]	α_2 [10^{-6} m^2/s]	$\alpha_{2\,av}$ [10^{-6} m^2/s]	$U_{\alpha_2 av}$ [%]	R_c [10^{-6} m^{2}K/W]	$R_{c\,av}$ [10^{-6} m^{2}K/W]	$U_{R_c av}$ [%]
1	$R_{c\,ap}$ 10 10^{-6}	0.296	0.273	15.1	496	542	12.8
2		0.268			539		
3		0.280			556		
4		0.247			578		
1	$R_{c\,ap}$ 10 10^{-6}	Diverged	–	–	Same as related *a priori* value	–	–
2							
3							
4							

In comparison to literature data taken from, for example, Touloukian *et al*. [19] ($1.19 \cdot 10^{-7}$ m^2/s at room temperature), the actual thermal diffusivity value is more than twice higher, but reasons for such a difference can be found in material specific preparation and structure. Also, the confidence region of the results would have been larger if the uncertainties of known parameters had been considered. On the other hand, the value of the thermal contact resistance is difficult to compare with other corresponding literature data since it depends on many other physical parameters such as the type of applied adhesive, its density or thickness.

Conclusion

A simultaneous estimation of thermal diffusivity of coatings and thermal contact resistance between the coating and substrate is not possible using the laser flash method due to the linear dependence between related sensitivity coefficients. In order to overcome this problem for the case of thermal barrier coatings, a particular optimal parameterization is proposed. By using the new set of optimal parameters whose sensitivity coefficients are linearly independent and by considering the level of measuring uncertainty, the standard Gauss estimation procedure can be applied with success. The proposed method has been applied in practice on two different substrate-coating structures and obtained results are in agreement with theoretical considerations.

Nomenclature

a – thickness of the substrate, [mm]
b – thickness of the coating, [m]

$c_{1,2}$ – specific heat of the substrate or coating, [$Jkg^{-1}K^{-1}$]
D – diameter of the sample, [mm]
$d_{1,2,3,4}$ – functional elements, [$Wm^{-2}K^{-1}$]
$h_{1,2}$ – radiative heat transfer coefficients, [$Wm^{-2}K^{-1}$]
$\mathbf{I}$ – identity matrix
i – estimation number; estimation index
$\mathbf{J_p}$ – Fisher information matrix of parameters **p**
$\mathbf{J_z}$ – Fisher information matrix of unknown parameters
$\mathbf{J}_{\log \mathbf{q}}$ – Fisher information matrix of logarithmic parameters **q**
j – estimation index
k – iteration index
$k_{1,2}$ – thermal conductivity of the substrate or coating, [$Wm^{-1}K^{-1}$]
m – estimation index
N – number of negative elements of the set S
N_n – function of the solution
n – solution index
$p_{1,2,3}$ – non-linear combinations of dimensionless parameters
p – column-vector of parameters $p_{1,2,3}$
Q – energy per square meter absorbed by the sample, [Jm^{-2}]
$q_{1,2,3}$ – dimensionless parameters, [–]
q – column-vector of dimensionless parameters
R – matrix of r_{jm} elements
R_c – thermal contact resistance, [m^2KW^{-1}]
r_{jm} – auxiliary numbers
S – set of functional elements
$s_{1,2}$ – functions of the solution
T – transient temperature computed from the model, [K]
T – column-vector of the transient temperature computed from the model
t – time, [s]
U – expanded uncertainty of the coverage factor 2, [%]
U – transformation matrix
V – diagonal matrix of eigenvalues
W – variance-covariance matrix of measured data
$\mathbf{W_z}$ – variance-covariance matrix of unknown parameters
$\mathbf{X}_{\log \mathbf{q}}$ – sensitivity coefficients matrix of logarithmic parameters $q_{1,2,3}$
$\mathbf{X_p}$ – sensitivity coefficients matrix of parameters $p_{1,2,3}$
$\mathbf{X_z}$ – sensitivity coefficients matrix of unknown parameters
$\mathbf{X_q}$ – sensitivity coefficients matrix of parameters $q_{1,2,3}$
X^* – normalized sensitivity coefficient, [mK]
Y – column-vector of measured data
z – column-vector of unknown parameters

Greek symbols

$\alpha_{1,2}$ – thermal diffusivity of the substrate or coating, [m^2s^{-1}]
β_n – roots of the transcendental equation
β_s – arbitrary value
$\varphi_{1,2}$ – functions of the solution
$\rho_{1,2}$ – density of the substrate or coating, [kgm^{-3}]
$\sigma_{\log p}$ – standard deviation of the parameter p

$_T$ – standard deviation of measured data, [%]
τ – laser pulse duration, [ms]

Subscripts

ap – *a priori* value
av – average value

Superscripts

final – final iteration
t – transport operator

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