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Simultaneous Estimation of the Thermal Diffusivity and Thermal Contact Resistance of Thin Solid Films and Coatings Using the Two-Dimensional Flash Method[1]

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The paper presents a description of the two-dimensional flash method for measuring the thermal diffusivity of deposited thin solid films and coatings, including the possibility of determination of the thermal contact resistance and the thermal anisotropy of the film or coating. Giving an analytical solution of the transient temperature as the consequence of transient two-dimensional heat conduction through the sample, the paper describes the estimation possibilities for the thermal diffusivity and thermal contact resistance, as well as for other typically unknown parameters of the model. Particular attention was given to the influence of "known" parameters on the estimation possibilities of unknown parameters. With such a study, essential information for the modeling of the experiment is provided. The experimental setups and demonstration of the proposed data reduction procedure are presented at the end of the paper. Having different estimation possibilities for measuring different locations of the sample, the results suggest using the data of several transient temperatures for more reliable and accurate simultaneous estimation of the thermal diffusivity and thermal contact resistance of deposited thin films and coatings.

KEY WORDS: coatings; laser flash method; layered materials; thermal anisotropy; thermal contact resistance; thermal diffusivity; thin films; two-dimensional heat conduction.

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1. INTRODUCTION

The laser flash method based on analyzing the rear-side transient temperature response, originally proposed for thermal diffusivity measurements of solid homogeneous materials [1], has been, since the mid-1960s of the past century, the most powerful tool in thermal transport property studies over the widest temperature range. The method has also been the subject of extensive analyses. Research was directed toward both improvement of the accuracy and reliability of its results, and also extension of its applicability regarding the types of examined materials. With the evergrowing demand of industrial applications, the laser flash method has been extended to thermal diffusivity measurements of thin films, coatings, anisotropic, and layered materials, metals in the liquid state, etc.

As a solution for anisotropic single materials, Donaldson and Taylor [2] proposed a radial heat flow or two-dimensional flash method variant, which was later used and further developed by Chu et al. [3] and Amazouz et al. [4]. Lachi and Degiovanni [5] applied a parameter estimation analysis in the method, introducing an optimization regarding the sensitivities of measured values. Using the same principle, Shibata et al. [6] proposed a technique for thermal diffusivity measurements of thin films in the directions parallel and perpendicular to the rectangular sample surface. They used a line-shaped laser beam for one-dimensional (1D) heat flow simulation in the sample plane, described by a simplified analytical equation. Recently, Sheikh et al. [7] suggested circular disk specimens and the solution of a corresponding cylindrical two-dimensional heat conduction equation.

Considering layered materials, Larson and Koyama [8] studied theoretically transient mono-dimensional heat conduction in two-layered samples resulting from step-function heating, but neglecting thermal contact resistance between the layers. Gilchrist and Price [9] developed an apparatus for measuring the thermal diffusivity of thin films and double-layer materials. Bulmer and Taylor [10] revised the mathematical procedure of Larson and Koyama for pulse heating appropriate to the laser flash method. Balageas et al. [11] considered the influence of finite contact resistance, giving also the analytical solution for 1D heat flux. Many authors applied later the principle of the flash method with mono-dimensional heat conduction to measuring the thermal diffusivity of layered materials.

On the other hand, several papers dealt with transient two-dimensional heat conduction in layered and composite materials, but not necessarily related to the laser flash method. Salt [12, 13] investigated an analytical solution in a two-dimensional composite slab when the initial flux had a step change. He analyzed in detail the temperature modes, giving their mathematical and physical interpretation. Mikhailov and Vulchanov [14] proposed a general procedure for resolving Sturm–Liouville problems, proper for the multilayered two-dimensional (2D) heat conduction case. Somewhat later, Mikhailov and Özişik [15] confirmed the procedure offered by Salt for a three-dimensional version of the problem. Yan et al. [16] developed different solutions of 2D conduction in layered media for various boundary conditions. Aviles-Ramos et al. [17] offered the exact transient solution of two-dimensional conduction in composite orthotropic media for inverse evaluation purposes. Abdul Azeez and Vakakis [18] proposed a partially analytical solution of the transient heat diffusion equation in multilayered semi-infinite media using a double integral transformation, which was similarly done by Kozlov and Mandrik [19].

As the laser flash method holds a most important place in measuring thermal diffusivity, this paper tries to extend possible applications of the method to the study of 2D heat flow in layered media, including the finite thermal contact resistance and anisotropy of thin films or coating materials. The paper presents an explicit analytical solution of the problem, an investigation of parameter estimation possibilities using an estimation procedure, as well as a description of a measurement technique for simultaneous determination of the thermal diffusivity and thermal contact resistance.

2. THEORETICAL MODEL

Let a two-layer sample be in the cylindrical form as shown in Fig. 1a. The first layer (substrate) is a homogeneous and isotropic material, and it is exposed to a short laser pulse. The other represents a thin film or coating, deposited on the substrate. Each material is characterized by thermophysical properties such as thermal diffusivity α, heat capacity c, and density ρ. The contact between the layers, which is non-ideal, is described by a finite thermal contact resistance, R_c. Assuming the anisotropy of a thin film or coating, one identifies thermal diffusivity $\alpha_{2\parallel}$ for the parallel and $\alpha_{2\perp}$ for the perpendicular direction to the plane of that layer.

The corresponding cylindrical coordinate system is shown in Fig. 1b. The contact point between the layers and at the center of the sample is designated zero in axial and radial directions. The thickness of the substrate is denoted with a, that of the thin film or coating with b, and that for the sample radius with d. Axial heat transfer coefficients h_1 and h_2, describing boundary conditions between sample and environment, are finite, while radial coefficients h_{r1} and h_{r2} are assumed to be zero. The latter condition is essential for resolving the problem by the separation of variables technique [16].

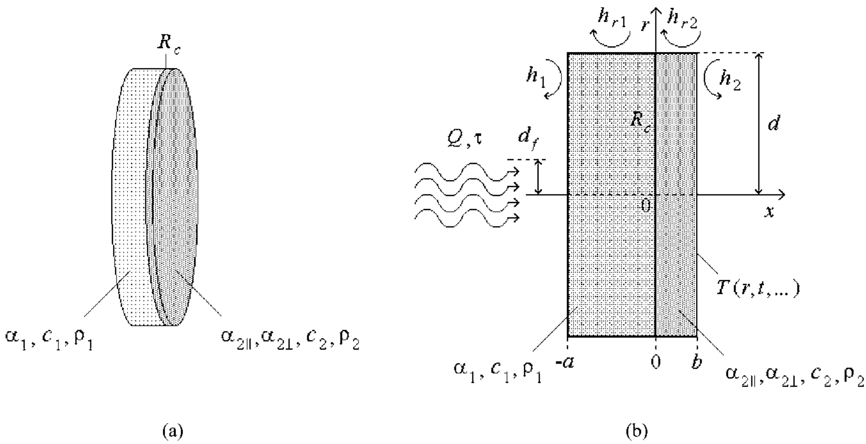

Fig. 1. (a) Two-layer composite sample; (b) Corresponding coordinate system.

At time $t=0$, the laser pulse of energy Q and duration τ, begins to impact the front sample side (substrate) on the central circular surface of radius d_f, as shown in Fig. 1b. Assuming that the pulse energy is absorbed in a very thin volume of thickness ε ($\varepsilon \ll a$) and radius d_f, the heat diffuses axially and radially by conduction from the front to the rear sample side, where the transient temperature is measured.

If one assumes that the thermophysical properties are constant within a small variation of sample temperature, the initial energy is absorbed uniformly in the sample, and, for example, the initial pulse is infinitely short, i.e., in the form of a Dirac pulse ($\tau=0$), the corresponding partial differential equations are

$$\frac{\partial^2 T_1}{\partial x^2}+\frac{1}{r}\frac{\partial T_1}{\partial r}+\frac{\partial^2 T_1}{\partial r^2}=\frac{1}{\alpha_1}\frac{\partial T_1}{\partial t}, \qquad -a \leqslant x < 0, \quad 0 \leqslant r \leqslant d, \quad t>0 \tag{1}$$

$$\frac{\partial^2 T_2}{\partial x^2}+\frac{\lambda_{2\parallel}}{\lambda_{2\perp}}\left(\frac{1}{r}\frac{\partial T_2}{\partial r}+\frac{\partial^2 T_2}{\partial r^2}\right)=\frac{1}{\alpha_{2\perp}}\frac{\partial T_2}{\partial t}, \qquad 0 < x \leqslant b, \quad 0 \leqslant r \leqslant d, \quad t>0 \tag{2}$$

with the following boundary and initial conditions

$$\frac{\partial T_1}{\partial x}=h_1 T_1, \quad x=-a, \qquad \frac{\partial T_2}{\partial x}=-h_2 T_2, \quad x=b, \qquad t>0 \tag{3}$$

$$\lambda_1\frac{\partial T_1}{\partial x}=\lambda_{2\perp}\frac{\partial T_2}{\partial x}, \qquad T_2-T_1=\lambda_1 R_c\frac{\partial T_1}{\partial x}, \qquad x=0, \qquad t>0 \tag{4}$$

$$\lambda_1 \frac{\partial T_1}{\partial r} = 0, \quad \lambda_{2\|} \frac{\partial T_2}{\partial r} = 0, \quad r = d, \quad t > 0 \tag{5}$$

$$T_1 = \begin{cases} \dfrac{Q}{\rho_1 c_1 \varepsilon}, & -a \leqslant x \leqslant -a+\varepsilon, \quad 0 \leqslant r \leqslant d_f, \quad \varepsilon \ll a \\ 0, & -a+\varepsilon \leqslant x \leqslant 0, \quad 0 \leqslant r \leqslant d_f \\ 0, & -a \leqslant x \leqslant 0, \quad d_f < r \leqslant d \end{cases}, \qquad T_2 = 0, \quad t = 0 \tag{6}$$

where λ is the corresponding thermal conductivity ($\lambda = \rho c \alpha$).

This problem can be solved analytically by separating the space and time variables. The solution of temperature at the surface of a thin film, assuming the instantaneous laser pulse ($\tau = 0$), can be expressed in the following form [20]:

$$\begin{aligned} T &\equiv T_2(r,t)|_{x=b} \\ &= Q \frac{\alpha_1}{\alpha_{2\perp}} \frac{d_f^2}{d^2} \left\{ \sum_{n=1}^{+\infty} \frac{1}{N_{n,i=1}} \frac{\gamma_{n,i=1}}{\eta_{n,i=1}} \left[\sin(\gamma_{n,i=1} a) + \frac{s_{1n,i=1}}{s_{2n,i=1}} \cos(\gamma_{n,i=1} a) \right] \right. \\ &\quad \times \left[-\sin(\eta_{n,i=1} b) + \frac{s_{3n,i=1}}{s_{4n,i=1}} \cos(\eta_{n,i=1} b) \right] e^{-\beta_{n,i=1}^2 t} \\ &\quad + \frac{2}{d_f} \sum_{i=2}^{+\infty} \frac{J_1(v_i d_f)}{v_i J_0^2(v_i d)} J_0(v_i r) \sum_{n=1}^{+\infty} \frac{1}{N_{n,i}} \frac{\gamma_{n,i}}{\eta_{n,i}} \left[\sin(\gamma_{n,i} a) + \frac{s_{1n,i}}{s_{2n,i}} \cos(\gamma_{n,i} a) \right] \\ &\quad \left. \times \left[-\sin(\eta_{n,i} b) + \frac{s_{3n,i}}{s_{4n,i}} \cos(\eta_{n,i} b) \right] e^{-\beta_{n,i}^2 t} \right\} \end{aligned} \tag{7}$$

where

$$\begin{aligned} N_{n,i} &= \frac{\rho_1 c_1}{4\gamma_{n,i}} \left\{ 2 \frac{s_{1n,i}}{s_{2n,i}} [1 - \cos(2\gamma_{n,i} a)] \right. \\ &\quad \left. + \sin(2\gamma_{n,i} a) \left(\frac{s_{1n,i}^2}{s_{2n,i}^2} - 1 \right) + 2\gamma_{n,i} a \left(\frac{s_{1n,i}^2}{s_{2n,i}^2} + 1 \right) \right\} \\ &\quad + \rho_2 c_2 \frac{\alpha_1^2}{\alpha_{2\perp}^2} \frac{\gamma_{n,i}^2}{4\eta_{n,i}^3} \left\{ 2 \frac{s_{3n,i}}{s_{4n,i}} [\cos(2\eta_{n,i} b) - 1] \right. \\ &\quad \left. + \sin(2\eta_{n,i} b) \left(\frac{s_{3n,i}^2}{s_{4n,i}^2} - 1 \right) + 2\eta_{n,i} b \left(\frac{s_{3n,i}^2}{s_{4n,i}^2} + 1 \right) \right\} \end{aligned} \tag{8}$$

$$\gamma_{n,i} = \alpha_1^{-1/2} \sqrt{\beta_{n,i}^2 - \alpha_1 v_i^2}, \qquad \eta_{n,i} = \alpha_{2\perp}^{-1/2} \sqrt{\beta_{n,i}^2 - \alpha_{2\|} v_i^2} \tag{9}$$

$$s_{1n,i} = k_1\gamma_{n,i}\cos(\gamma_{n,i}a) + h_1\sin(\gamma_{n,i}a),$$
$$s_{2n,i} = k_1\gamma_{n,i}\sin(\gamma_{n,i}a) - h_1\cos(\gamma_{n,i}a) \tag{10}$$

$$s_{3n,i} = k_{2\perp}\eta_{n,i}\cos(\eta_{n,i}b) + h_2\sin(\eta_{n,i}b),$$
$$s_{4n,i} = -k_{2\perp}\eta_{n,i}\sin(\eta_{n,i}b) + h_2\cos(\eta_{n,i}b) \tag{11}$$

Coefficients v_i and $\beta_{n,i}$ ($n, i = 1, 2,..., +\infty$) represent positive roots of two transcendental equations:

$$v_i J_1(v_i r) = 0 \tag{12}$$

and

$$k_{2\perp}\eta_{n,i}s_{1n,i}s_{4n,i} - k_1\gamma_{n,i}s_{2n,i}s_{3n,i} - k_1k_{2\perp}R_c\gamma_{n,i}\eta_{n,i}s_{2n,i}s_{4n,i} = 0 \tag{13}$$

For each value of v_i one has to solve Eq. (13) and find the roots $\beta_{n,i}$.

If the initial pulse is of finite duration τ, the solution can be obtained using the theorem of superposition proposed by Watt [21], according to which the sample temperature is a function of the pulse evolution. If the pulse variation is described by $\Theta(t, \tau)$, the transient temperature of the rear sample side becomes for $t > \tau$

$$T(r, t, \tau > 0) = \left[\int_0^\tau \Theta(t', \tau)\, dt'\right]^{-1} \int_0^\tau \Theta(t', \tau)\, T(r, t, \tau = 0)\, dt' \tag{14}$$

where $T(r, t, \tau = 0)$ is the temperature from Eq. (7). For $t \leqslant \tau$, the same expression is valid, but with parameter t instead of τ for the upper limits of both integrals. In practice, when the characteristic duration of the temperature response is much longer than τ, the temperature is well described only by Eq. (7).

The transient temperature is mathematically expressed by Eq. (7) or Eq. (14) for every moment t and coordinate r. Nevertheless, in reality, when an infrared detector is used for temperature measurement, the temperature response represents an average temperature of a region "seen" by the IR detector. One should, therefore, extend the expression for the transient temperature given by Eq. (7) or Eq. (14) to that which would comprise a supposed radius of the detected area, r_s.

The influence of the finite detected region, where the region is concentric with the sample surface, has been studied by Yamane et al. [22]. In this case, however, the detected region is removed from the sample center and an auxiliary reference system must be considered. It is presented on Fig. 2a, where r_0 is the distance between the center of the sample surface and the center of the detected region, and r'' is the coordinate of a single

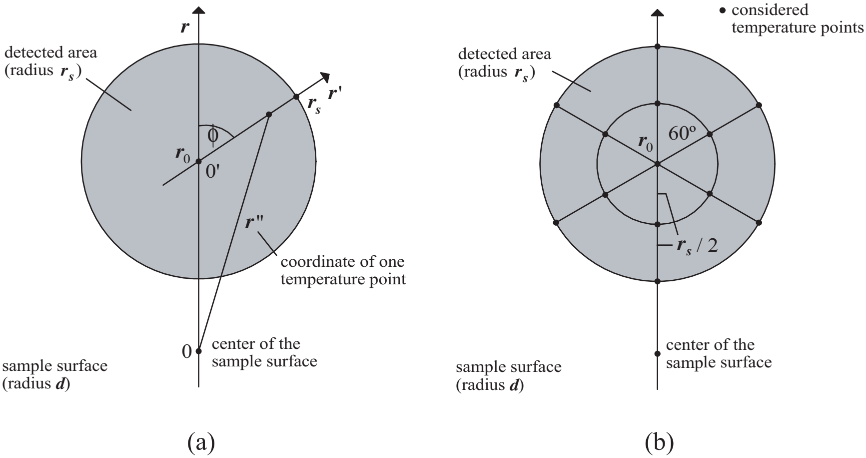

Fig. 2. (a) Detected surface and corresponding coordinate system; (b) Considered temperature points for parameter estimation.

temperature point within the detected region with respect to the main reference system. Since r'' is equal to $r'' = \sqrt{r_0^2 + 2r_0 r' \cos\phi + r'^2}$, the integral form of the transient temperature that corresponds to the detected area becomes

$$T(t) = \int_{\phi=0}^{\phi=2\pi} \frac{1}{2\pi} \int_{r'=0}^{r'=r_s} \frac{2r'}{r_s^2} T(r'', t)\, dr'\, d\phi \tag{15}$$

where $T(r = r'', t)$ is the temperature from Eq. (7) or Eq. (14). Since there is no analytical solution of Eq. (15), one must perform one of the methods of numerical integration. In this paper, the solution of temperature was approximated by using the Newton–Cotes formulas over a limited number of the temperature points within the detected region (12 distributed symmetrically, plus one at the center, as shown in Fig. 2b). Taking into account that only 4 of the 8 temperatures of the symmetrical points from the left and right sides of axis r are different, the following approximate formula of Eq. (15) was applied:

$$\begin{aligned} T(t) &= \int_{\phi=0}^{\phi=2\pi} \frac{1}{2\pi} \int_{r'=0}^{r'=r_s} \frac{2r'}{r_s^2} T(r'')\, dr'\, d\phi \\ &\approx \frac{5}{5184} \frac{1}{r_s} [76 r'_1 T(r''_{0,1}) + 19 r'_2 T(r''_{0,2}) + 376 r'_1 T(r''_{1,1}) + 94 r'_2 T(r''_{1,2}) \\ &\quad + 500 r'_1 T(r''_{2,1}) + 125 r'_2 T(r''_{2,2}) + 200 r'_1 T(r''_{3,1}) + 50 r'_2 T(r''_{3,2})] \end{aligned} \tag{16}$$

where $r'_i = i r_s/2$, $r''_{j,i} = \sqrt{r_0^2 + 2r_0 r'_i \cos\phi_j + r_i'^2}$, and $\phi_j = j\pi/3$.

Having thus the final form of the transient temperature (Eq. (16)), one can perform a parameter estimation procedure, which includes the study of the parameter estimation possibilities.

3. PARAMETER ESTIMATION

3.1. Estimation Procedure

There are several parameter estimation procedures, but the most frequently used are of the gradient type. Generally, all of them are based on minimization of differences between experimental data and calculated theoretical values. Among methods of the gradient type, the Gauss iterative procedure is usually used in nonlinear estimation, i.e., when sensitivity coefficients depend on proper parameters. A detailed description of this procedure is given by Beck and Arnold [23]. Milošević et al. [24] proposed it for estimation of the thermal contact resistance of a double-layer sample, in the case of 1D heat conduction, including the influence of "known" parameter uncertainties. In this paper, the same estimation procedure is applied, as well as the principle of estimation possibility analysis.

3.2. Estimation Possibilities

Some parameters are more suitable for estimation, some less. In that sense there are certain criteria that must be satisfied in order to obtain reliable results for a given estimator. These criteria are based on the analysis of the reduced forms of sensitivity coefficients [23],

$$X_j^* = p_j \frac{\partial T}{\partial p_j}, \tag{17}$$

where p_j is the jth parameter of the model and T is the model response. They are compared in order to find such a set of parameters whose sensitivity coefficients show both an acceptable level of linear independence and also absolute comparability.

There is also one condition for good estimation possibilities. In practice, there is always some finite uncertainty of "known" parameter values, which can considerably influence the final results of estimations. Besides the uncertainty of experimental data, σ, the variance-covariance matrix should, therefore, also be composed of the variances of "known" parameters, giving thus the total variance of the temperature response as [24]

$$\sigma_{\text{tot}}^2 = \sigma^2 + \sum_k \left(\sigma_{p_k} \frac{\partial T}{\partial p_k} \right)^2, \tag{18}$$

Table I. Values of Parameters *a priori* for the Estimation Analysis in Two Examples

Parameter for estimation	Example 1 Al (substrate), Ni (thin film) Value *a priori*	Example 2 Steel (substrate), PTFE (coating) Value *a priori*
$\alpha_{2\perp} \times 10^6\ (m^2 \cdot s^{-1})$	23	0.29
$\alpha_{2\parallel} \times 10^6\ (m^2 \cdot s^{-1})$	23	0.29
$R_c \times 10^6\ (m^2 \cdot K \cdot W^{-1})$	0.5	50
$\rho_2\ (kg \cdot m^{-3})$	8900	2100
$c_2\ (J \cdot kg^{-1} \cdot K^{-1})$	444	1100
$h_1 = h_2 = h\ (W \cdot m^{-2} \cdot K^{-1})$	50	50
$Q\ (J \cdot m^{-2})$	1000	1000

where p_k is the kth "known" parameter. These variances increase the general uncertainty of an estimation procedure and estimates, whose reduced sensitivity coefficients are less than the level of the total standard deviation from Eq. (18), and have a small influence on the temperature response, making their accurate estimation difficult.

Estimation possibilities of the thermal diffusivity, density, and specific heat[5] of a thin film or coating, the thermal contact resistance, as well as those of the absorbed energy Q, and heat transfer coefficient h, will be considered in two examples, one with a thin film, the other with a coating. The values of all parameters as well as maximum uncertainties of "known" parameters used in this study are presented in Tables I and II.

Example 1. Let the sample consist of aluminum as a substrate and of nickel as a thin film. Both of these materials are good heat conductors, especially aluminum. The transient temperatures of the nickel surface, computed using Eqs. (16), with parameter values from Tables I and II, and for different distances from the sample center, are presented in Fig. 3 on a logarithmic time scale. One can distinguish very fast responses in the region that corresponds to laser pulse impact from back sample side ($0 < r \leqslant d_f$), as well as slower responses outside this region ($r > d_f$). This is expected because the heat flux is initially much higher in the axial than in the radial direction. Also, the temperature amplitude decreases as the distance r becomes larger. An interesting result is about the time at which the temperature maximum is reached. Namely, in the laser pulse region, it decreases

[5] Having the same position in the model's equations, parameters ρ_1 and c_1, and ρ_2 and c_2 are treated integrally as two single parameters: $\rho_1 c_1$ and $\rho_2 c_2$.

Table II. Values and Uncertainties of "Known" Parameters for Two Examples

	Example 1 Al (substrate), Ni (thin film)		Example 2 Steel (substrate), PTFE (coating)	
"Known" parameter	Value	Maximum uncertainty (%)	Value	Maximum uncertainty (%)
r (mm)	0,3,5,6,7,8	$0.1 \div 100$[a]	0,3,5,6,7,8	$0.1 \div 100$[a]
r_s (mm)	1	10	1	10
a (mm)	0.2	1	1.55	0.1
b (μm)	5	10	100	0.5
d (mm)	25	0.04	25	0.04
d_f (mm)	5	0.04	5	0.04
τ (ms)	0.1	10	0.1	10
$\alpha_1 \times 10^6$ ($cm^2 \cdot s^{-1}$)	97.1	2	4	2
ρ_1 ($kg \cdot m^{-3}$)	2710	0.1	7820	0.1
c_1 ($J \cdot kg^{-1} \cdot K^{-1}$)	902	2	460	2

[a] From $r = 8$ mm to $r = 0$ mm where the minimal difference in distance is $\Delta r = 5$ μm.

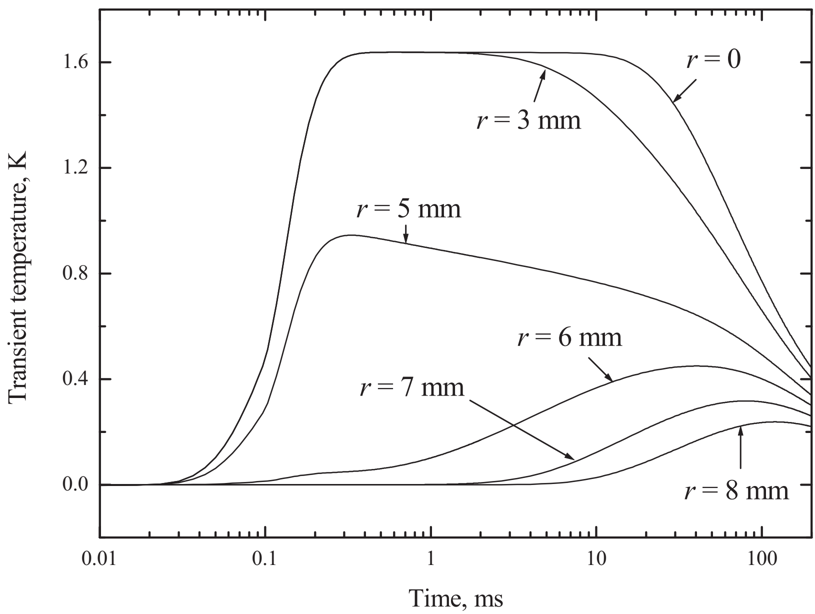

Fig. 3. Temperature responses of the back sample side at different distances r from Example 1.

first, but very soon it begins to increase with the distance r, as it does outside the region. The shift of the time of the temperature maximum is a consequence of radial heat flow that affects responses of the rear sample side gradually.

On the same figure one can note the influence of the finite area of radius r_s from which the temperature was actually measured. Namely, as a consequence of the extended detector view and vicinity of the sample central region, the transient response at $r = 6$ mm begins to rise very soon, although its "real" characteristic response (implying $r_s = 0$) is much larger.

The reduced sensitivity coefficients of parameters for estimation at several points r are shown in Fig. 4, also on a logarithmic time scale. One can see different levels of influence and consequently different estimation possibilities. While the parameter Q plays a significant role in the temperature response, the normal thin film thermal diffusivity $\alpha_{2\perp}$ has practically no influence on the transient temperature in this case; influences of the other parameters on the temperature response are different and between these two extremes.

Besides the absolute parameter values, the measuring point r is also very important for estimation possibilities. As r increases, sensitivity coefficients of thermal diffusivities and contact resistance change not only their values, but also their sign (cf. Fig. 4). Having both positive and negative influences on the temperature response, these parameters are principally good for estimation.

Comparing the sensitivity coefficients of parameters for estimation with the total standard deviation σ_{tot} computed from Eq. (18), one considers, however, the real estimation possibilities of parameters for estimation. Such a case is presented in Fig. 5. Several points should be noted: (i) there is no chance to estimate parameters such as $\alpha_{2\perp}$ and R_c, because their influence is much less than the level of σ_{tot}; (ii) in a certain time region, the parallel thermal diffusivity is "visible" for estimation; in another region, it is not; (iii) influences of the other three parameters h, $\rho_2 c_2$, and Q are not affected significantly by σ_{tot}, so their estimation could be accurate and fast; and (iv) the overall uncertainty σ_{tot} varies with the distance r, meaning that the computation of the sensitivity coefficient for every parameter and estimation step is indispensable.

Turning to Fig. 4 again, one can observe that having the sensitivity coefficients almost linearly dependent, simultaneous estimation of $\rho_2 c_2$, and Q may not be accurate, in spite of the high absolute values of their reduced sensitivity coefficients. In such case, one can use an estimation procedure with the optimal set of parameters formed as a logarithmic combination of original ones [25].

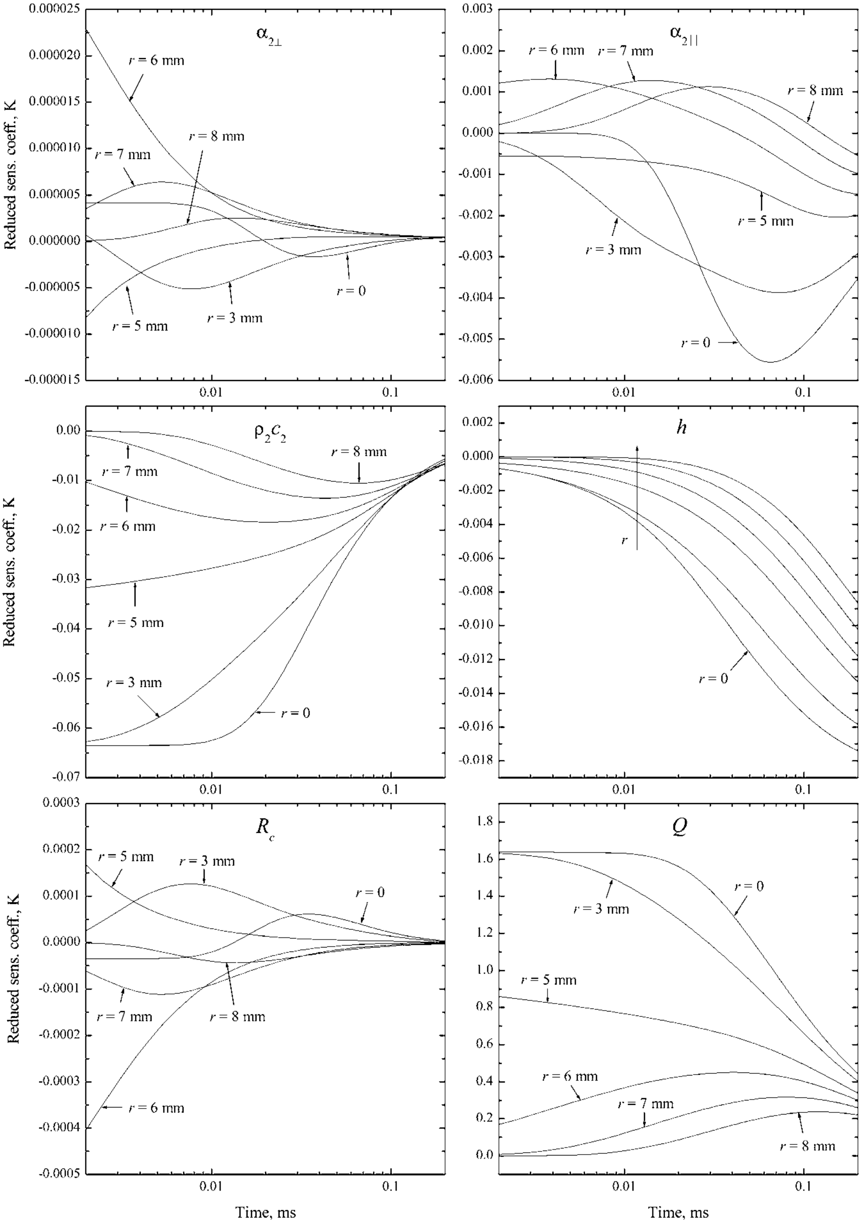

Fig. 4. Reduced sensitivity coefficients of parameters for estimation from Example 1.

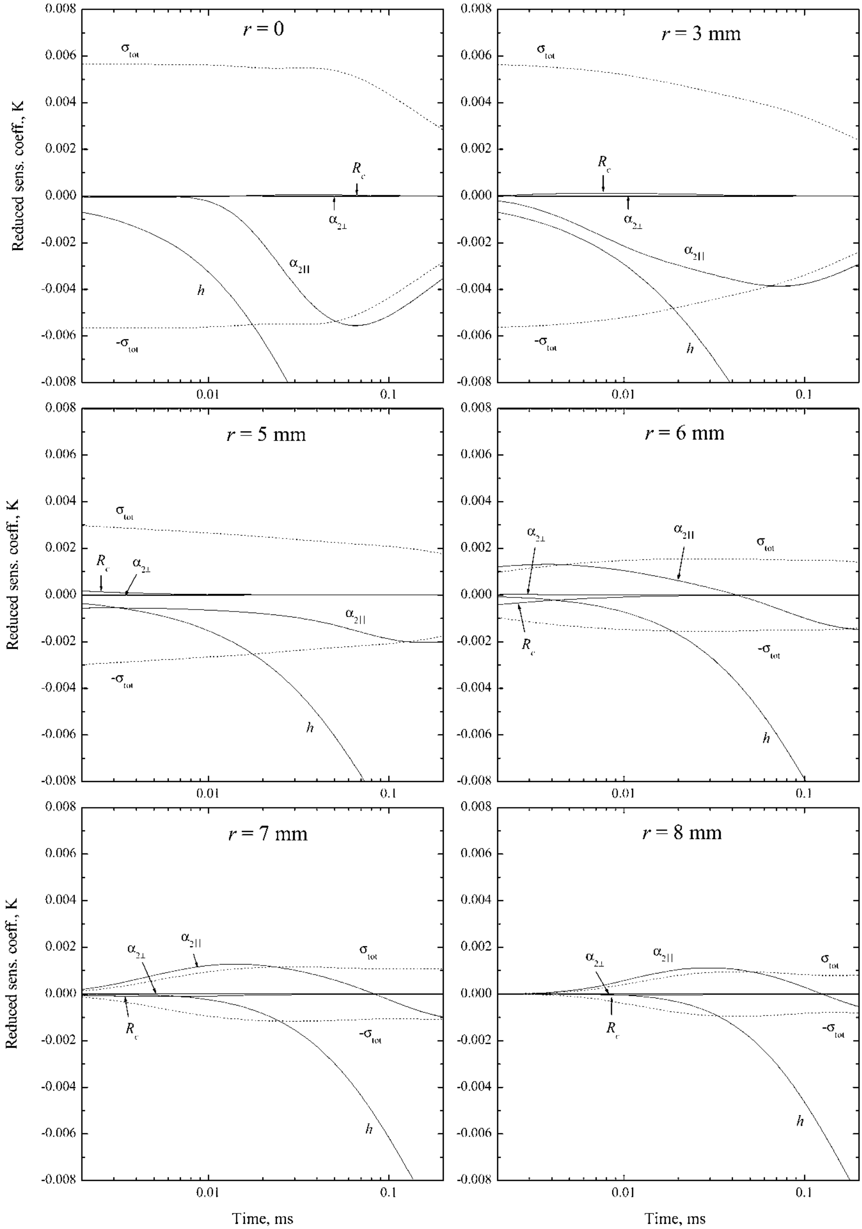

Fig. 5. Reduced sensitivity coefficients of parameters for estimation and total response uncertainty from Example 1.

Example 2. In this example, the first layer (substrate) is made from a steel (Cr-Ni), and the second (coating) from carbon fiber filled PTFE.[6] The first material is an average heat conductor, while the second one is a poor conductor. Such a combination is found in practice in the case of a PTFE-coated frying pan.

Characteristic responses and the influence of parameters for estimation are very different than those from the first example. Reduced sensitivity coefficients are presented in Fig. 6. The normal thermal diffusivity of the coating is more significant than the parallel one, and its influence on the response is always positive. The sensitivity coefficient of the thermal contact resistance is mostly negative and with large values, thus making the estimation of this parameter possible. However, the partial linear dependence between $\alpha_{2\perp}$, $\rho_2 c_2$, and R_c makes, in general, their simultaneous estimation more difficult. As far as the common heat transfer coefficient h is concerned, its influence is generally weaker than in the first case, but the form is the same as before. Similar results are obtained for the parallel thermal diffusivity of the coating.

For a complete analysis of estimation possibilities, the computation of the total response uncertainty is necessary. Having the uncertainties of "known" parameters from Table II, comparisons between reduced sensitivity coefficients and total standard deviation σ_{tot} for different measuring points r are given in Fig. 7. One can observe an attenuation of both the total uncertainty and also reduced sensitivity coefficients as r increases. Nevertheless, the latter always stays higher than the total uncertainty, at least in some time region, except for the sensitivity coefficient of the parallel thermal diffusivity of the coating, whose values are approximately the same as σ_{tot}. The highest estimation possibility for this parameter is in the region of temperature response where the value of its sensitivity coefficient traverses the overall uncertainty range.

As in the previous example, there is a high level of linear dependence between some parameters, in this case, between the normal thermal diffusivity of the coating and the thermal contact resistance. These parameters could be estimated simultaneously by either reducing the overall uncertainty in the region where the sensitivity coefficient of R_c changes sign or using the general optimal estimation procedure applied in Ref. 24.

If transport properties of the material of the coating are isotropic, the problem reduces to the single thermal diffusivity, α_2. Then, with the same parametric values as above, its influence on the temperature response is slightly different than that of the normal thermal diffusivity $\alpha_{2\perp}$ (cf. Fig. 7), and mostly larger than the overall response uncertainty.

[6] Polytetrafluoroethylene.

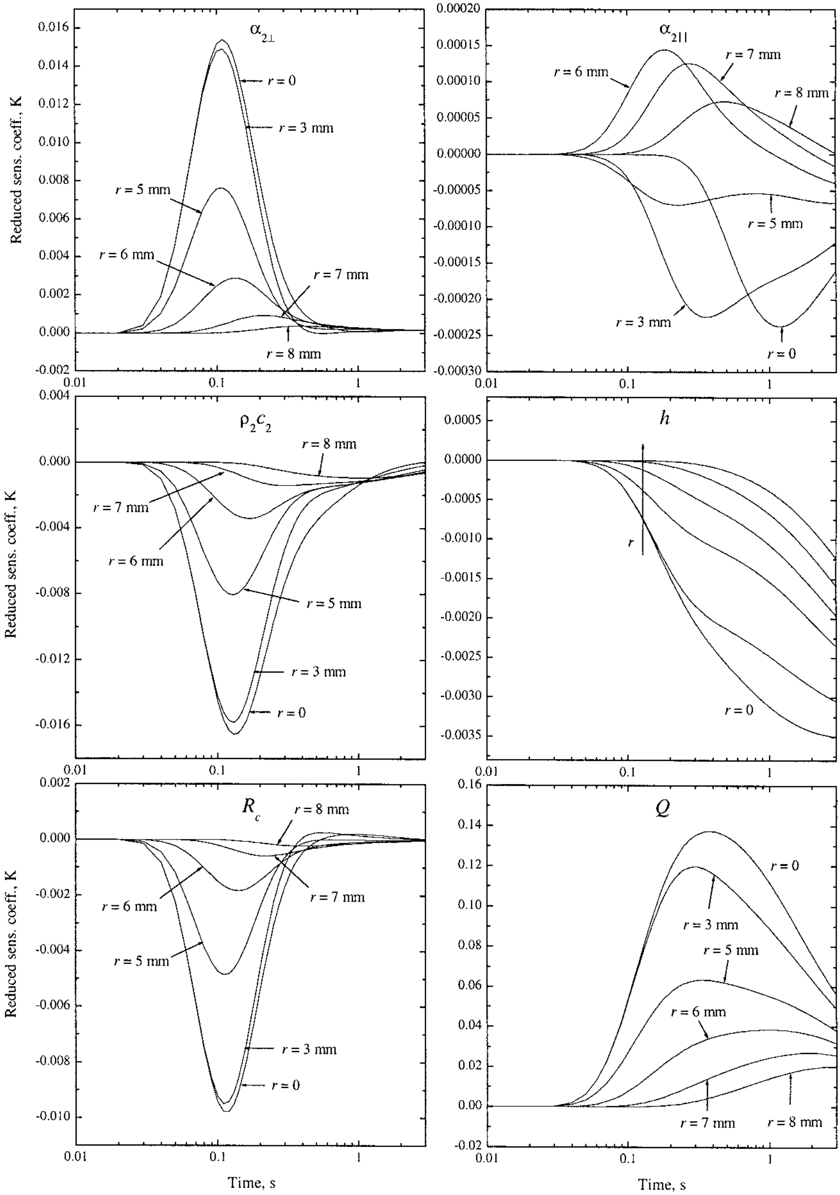

Fig. 6. Reduced sensitivity coefficients of parameters for estimation from Example 2.

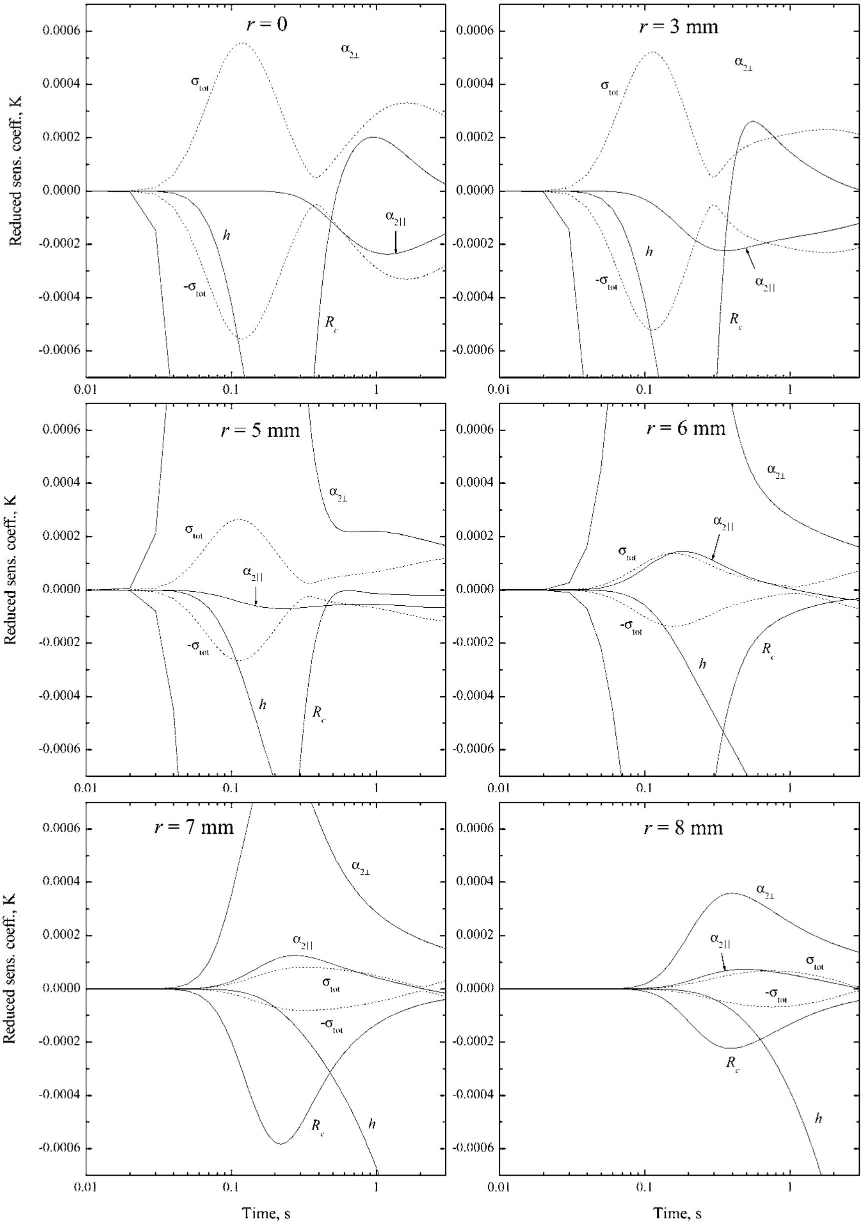

Fig. 7. Reduced sensitivity coefficients of parameters for estimation and total response uncertainty from Example 2.

4. EXPERIMENTAL SETUP AND MEASUREMENT RESULTS

The measurement principle of the present two-dimensional laser flash method is equivalent to that proposed by Shibata et al. [6]. The experimental objective is to measure the transient temperature at an arbitrary point of the rear sample surface away from the center where external heating on the front sample surface takes place. There are two possible ways to accomplish this: to shift the pulse radiation along the front specimen side or to move the detection setup over its rear side. Here, the second solution was more convenient. The experimental setup used in this work is shown schematically in Fig. 8.

The sample is held in specially designed PTFE holders within an aluminum support. In front of the specimen a non-moving mask is fixed, which limits the laser radiation to the central area of the sample surface. At the other side, a moving mask defines the focalizing area for temperature detection. Displacement of the mask is measured with a micrometer mounted on the sample support.

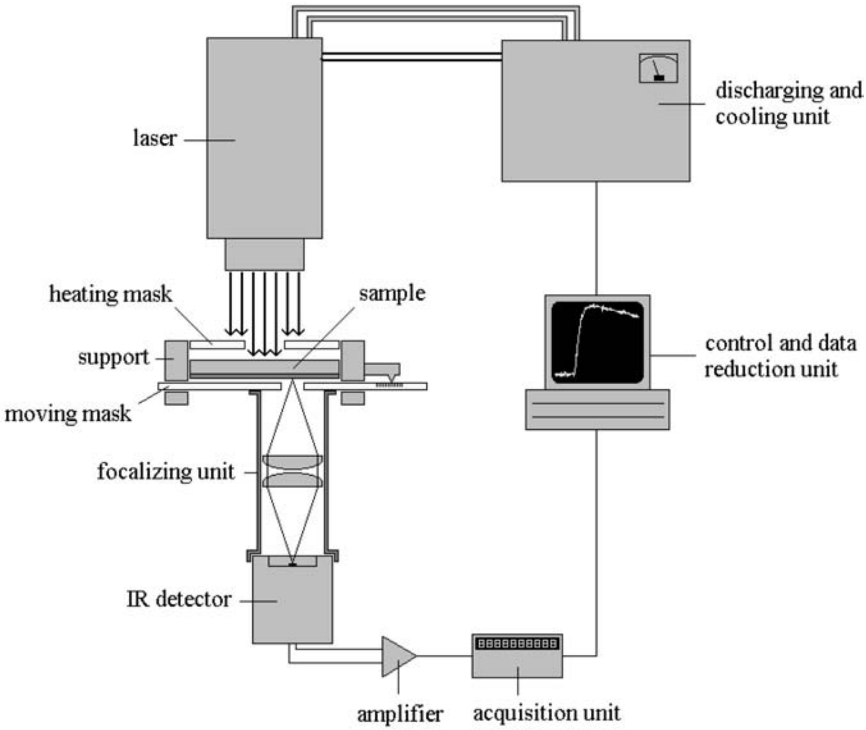

Fig. 8. Experimental setup.

The focalizing unit consists of two convex CaF lens and an IR detector holder fixed to the moving mask, directing the detector on the focalizing area. The measurement of the transient temperature is accomplished with a very sensitive InSb photoconductive detector. Its signal is amplified with a built-in amplifier, which reduces efficiently most of the noise level. A ruby pulse laser has an output energy up to 30 J, a pulse diameter of about 15 mm, and a pulse duration of about 1 ms.

As a demonstration, measurements and the estimation procedure were carried out on the sample from the second example with different values for each parameter *a priori*, and with values and uncertainties of "known" parameters r_s, a, b, d, and c_1, taken from Table II. The coating specific heat and density, c_2 and ρ_2, were considered as "known,"[7] and their values were taken from Table I (second example) with maximum uncertainties of 10 and 2%, respectively. The values of the substrate thermal diffusivity and density, α_1 and ρ_1, were obtained separately using a uniform, monolayer sample, made only from the substrate material. The thermal diffusivity of the substrate was measured by the ordinary laser flash method, using the Gauss estimation procedure [26], and the value was $\alpha_1 = 12.5 \times 10^{-6}\ \mathrm{m^2 \cdot s^{-1}}$, while the density is determined statistically as $\rho_1 = 7700\ \mathrm{kg \cdot m^{-3}}$, using different specimens from the same material. The pulse duration was $\tau = 1$ ms, absorbed energy $Q = 100\ \mathrm{J \cdot m^{-2}}$. while the radius of pulse heating was $d_f = 2.5$ mm, instead of 5 mm. The latter parameter influences significantly the characteristic time of the temperature response, considering the distance r, but the shapes of response stay the same as shown in the previous section. The transient temperature was measured at distances 0, 1, 2, 3, and 4 mm from the sample center.

Some experimental data in voltage units proportional to temperature are presented in Fig. 9. Linearity of the IR detector over a small temperature range is implied. A very high signal-to-noise ratio was due to good emissivity of the carbon fiber filled PTFE surface.

Estimation results, obtained by averaging intermediate results derived from the individual set of *a priori* values, are given in Table III. According to these results, there is some level of coating anisotropy where the parallel thermal diffusivity is slightly below the normal value, but this distinction could be covered by their estimation uncertainties. Calculated standard deviations [24] for particular measurements were about 10% for the normal and 33% for the parallel thermal diffusivity, which was expected considering the weaker influence of the latter parameter on the response.

[7] In the case of a very thin film, ρ_2 and c_2 should be considered unknown. However, in order to achieve a better accuracy of estimation results, these parameters were taken as "known," i.e., in practice, previously measured by some other experimental method.

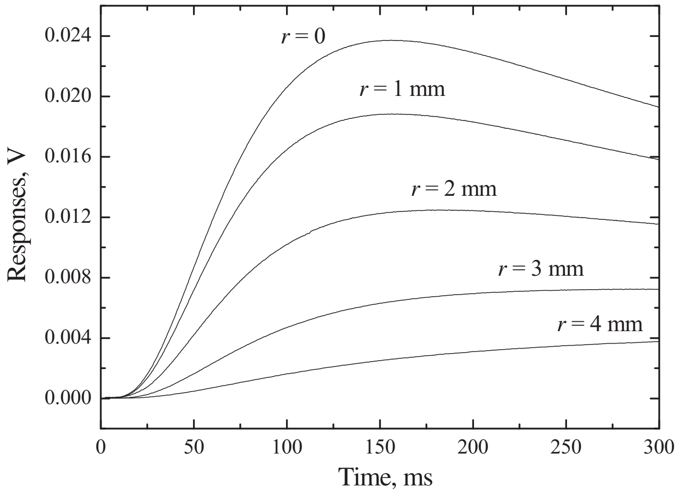

Fig. 9. Experimental data for the steel-PTFE sample.

The standard deviations for the thermal contact resistance, for a given distance r, are quite low. Relatively high general scatter of the results is due to a certain level of linear dependence between the sensitivity coefficients of parameters $\alpha_{2\perp}$ and R_c (cf. Fig. 6).

If the sample is considered homogeneous in the parallel direction, the values for each parameter can be averaged. If not, however, the different uncertainties of "known" parameters across the sample should be taken into account.

5. SUMMARY

An exact two-dimensional analytical solution of the transient temperature of a thin film or coating deposited on a substrate, including anisotropy,

Table III. Values of Estimated Parameters and Their Standard Deviations

r (mm)	$\alpha_{2\perp} \times 10^6$ ($m^2 \cdot s^{-1}$)	$\sigma_{\alpha 2\perp}$ (%)	$\alpha_{2\parallel} \times 10^6$ ($m^2 \cdot s^{-1}$)	$\sigma_{\alpha 2\parallel}$ (%)	$R_c \times 10^6$ ($m^2 \cdot K \cdot W^{-1}$)	σ_{Rc} (%)
0	0.214	9.4	0.180	33.7	342	3.5
1	0.250	8.7	0.182	33.1	346	3.2
2	0.191	8.3	0.183	33.0	397	3.2
3	0.265	13.9	0.186	33.9	539	2.5
4	0.261	10.8	0.189	34.0	402	3.0

the thermal contact resistance, and heat exchange coefficients has been made available [20]. Such a solution, extended for the finite duration of pulse heating and the finite area of the temperature detection, has been used in two examples in the study of estimation possibilities of a thin film or coating thermal diffusivity, of the thermal contact resistance between layers, as well as those of some other typically unknown parameters. In summary:

- The coating thermal diffusivity, perpendicular to the sample plane, is more suitable for estimation than that of a thin film.
- The thin film thermal diffusivity, parallel to the sample plane, could be estimated simultaneously with the thermal contact resistance.
- Going from inside to outside the heating pulse region, sensitivity coefficients of both thermal diffusivities and thermal contact resistance change their forms, thus increasing the estimation possibilities.
- Uncertainties of "known" parameters influence the total variance of the response, thus increasing the final uncertainties of estimated parameters.

Generally, having transient temperature responses at different points r away from the sample center, offers better possibilities for more accurate and reliable estimations than data obtained by the standard flash method, especially when the anisotropy of material is concerned. Therefore, the two-dimensional laser flash method can be, under certain conditions, extended to thermophysical property characterization of deposited thin films or coatings using the parameter estimation procedure.

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