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The Laser Flash Method with Repeated Pulses—Optimal Experimental Design Analysis[1]

L. Vozár,[2,3] G. Labudová,[2] and W. Hohenauer[4]

The paper deals with analytical aspects of the laser flash method with repeated pulses, which is a photothermal experimental method for measurement of the thermal diffusivity of solids. It concentrates on the data reduction—an estimation of the thermal diffusivity from the experimental data. Special attention is given to the technique of correction of the width and shape of the heat pulses. Results of sensitivity and optimal experimental design analysis are discussed in detail. It focuses on questions of the influence of setting the experimental parameters, heat pulse period and the number of applied heat pulses, to the sensitivity of the method as well as the optimum time of duration of an experiment.

KEY WORDS: experimental design; flash method; repeated pulses; sensitivity; thermal diffusivity.

1. INTRODUCTION

The laser flash method with repeated pulses is a photothermal experimental method for measurement of the thermal diffusivity of solids. The method is an extension of the standard laser flash method based on an analysis of the thermal response of the test material sample to a heat pulse [1]. The laser flash method with repeated pulses uses a small disk-shaped sample similar to the standard laser flash method. Here the one (front) surface of the

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[2] Department of Physics, Faculty of Sciences, Constantine the Philosopher University, Tr. A. Hlinku 1, SK-94974 Nitra, Slovakia.

[3] To whom correspondence should be addressed. E-mail: vozar@nr.sanet.sk or vozar@tpl.ukf.sk

[4] Department of Materials Research, ARC Seibersdorf research GmbH, A-2444 Seibersdorf, Austria.

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sample is subjected to several repeated heat pulses originated by consecutive laser pulses. The resulting temperature rise on the opposite (rear) face of the sample is measured, and the thermal diffusivity is computed from the temperature rise versus time data [2, 3]. Data reduction—calculation of the thermal diffusivity from the experimental data consists of an estimation of one parameter of interest—the thermal diffusivity and one, or two, other additional (nuisance) parameter(s)—the temperature term (the adiabatic temperature limit), and the Biot number, as it conforms to the ideal adiabatic or, nonideal, analytic theory, respectively.

The aim of an introduction of the flash method with repeated pulses is to overcome particular experimental difficulties connected with an application of highly intensive pulses. Splitting the total energy entering the sample among several consecutive pulses reduces temperature gradients in the sample, which cause problems in case of an investigation of insulators, temperature sensitive materials, large-grain heterogeneous materials, measurements near the phase transition, etc.

The purpose of the present work is to perform experimental design analysis [4] and to discuss the achieved results. Here the concept of sensitivity coefficients S [5] as well as the formalism based on the criterion to maximize the ratio of determinants Δ/Δ_2 of $\mathbf{S}^{\mathrm{T}}\mathbf{S}$, which contains the product of the sensitivities and their transpose [6], is utilized. Results give arguments on how setting the experimental parameters influences the sensitivity of the method as well as it influences the optimum time of duration of an experiment, as presented for the flash methods with extended pulses, the step heating method [7], and other prescribed steady-state [6, 8] or periodic [9] heat flux methods.

2. ANALYTICAL BASIS

The analytical model considers a homogeneous opaque slab of thickness e with uniform and constant thermophysical properties and the density ρ. The sample front face is exposed to instantaneous heat pulses repeated with a period t_{p}, analytically described by the shape $\phi(t) = Q\delta(t - kt_{\mathrm{p}})$; $k = 0, 1, \ldots, p$. Here Q is the heat supplied by a pulse to a unit area of the front face, $\delta(t)$ is the Dirac's function, and $(p+1)$ is the number of pulses. If there is heat transfer between the sample and its environment, governed by Biot numbers H_0 and H_{e} at the front and rear faces, respectively, the transient rear-face temperature $T(t)$ can be expressed in the form of a Fourier series [3],

$$T(t) = T_{\lim} \sum_{n=1}^{\infty} A_n(H_0, H_{\mathrm{e}}) \sum_{i=0}^{k} \exp[v_n(it_{\mathrm{p}} - t)], \tag{1}$$

where $T_{\lim}$ is the adiabatic limit temperature ($T_{\lim} = Q/(\rho ce)$ with c being the heat capacity), t is the time, a is the thermal diffusivity

$$A_n(H_0, H_e) = \frac{2u_n^2(u_n^2 + H_e^2)\left(\cos u_n + \frac{H_0}{u_n}\sin u_n\right)}{(u_n^2 + H_0^2)(u_n^2 + H_e^2) + (H_0 + H_e)(u_n^2 + H_0 H_e)},$$

$$H_0 > 0; \quad H_e > 0 \tag{2}$$

$$v_n = \frac{u_n^2 a}{e^2}, \tag{3}$$

$$k = \left\langle \begin{array}{ll} 0, 1, \ldots, p-1; & kt_p \leqslant t < (k+1)\, t_p \\ p & t \geqslant pt_p \end{array} \right., \tag{4}$$

and u_n are the positive roots of the equation,

$$(u^2 - H_0 H_e)\tan(u) = (H_0 + H_e)\, u. \tag{5}$$

3. DATA REDUCTION

The data reduction, an estimation of the thermal diffusivity, consists of a least-squares fit of the theoretical curves to the measured temperature rise versus time evolution. It is easy to show that the problem of finding the thermal diffusivity can be transformed to solving the algebraic equations [10],

$$\sum_{j=1}^{N} T_j \theta_j(a, H) \sum_{j=1}^{N} \theta_j(a, H) \frac{\partial \theta_j(a, H)}{\partial a} - \sum_{j=1}^{N} T_j \frac{\partial \theta_j(a, H)}{\partial a} - \sum_{j=1}^{N} \theta_j^2(a, H) = 0, \tag{6}$$

$$\sum_{j=1}^{N} T_j \theta_j(a, H) \sum_{j=1}^{N} \theta_j(a, H) \frac{\partial \theta_j(a, H)}{\partial H} - \sum_{j=1}^{N} T_j \frac{\partial \theta_j(a, H)}{\partial H} - \sum_{j=1}^{N} \theta_j^2(a, H) = 0, \tag{7}$$

where T_j is the experimental temperatures measured in the time t_j, $\theta_j(a, H) = T(t_j)/T_{\lim}$ is the analytical dimensionless temperature rise (Eq. (1)) (assuming $H = H_0 = H_e$) and N is the number of points taken into account. The approach also allows an estimation of the adiabatic limit temperature $T_{\lim}$ [11].

4. EXTENDED PULSES

The simple theory considers that the applied heat pulses are instantaneous. This assumption is valid when the heat pulse duration is negligibly small, i.e., in the case of measuring a poor thermal conductive material and/or for sufficiently thick samples. Otherwise, the duration and the shape of the heat pulses should be taken into account. The expression for the temperature rise evolution in the case of square-wave shaped pulses has been given [3]; the other formulas suitable for the usual heat-pulse-shape approximations are summarized elsewhere [12].

The other solution originally proposed for the standard "one pulse" laser flash method consists of adjustment of the effective irradiation time t_g using the center of gravity of the heat pulse defined as [13]

$$t_g = \int_0^{\infty} t'\varphi(t')\, dt'. \tag{8}$$

The correction of the influence of the heat pulse duration consists of shifting the time axis taking the effective irradiation time t_g as the time origin. Then the heat pulse is considered to be instantaneous and the appropriated analytical solution for an ideal heat pulse is utilized in the data reduction [14].

This correction can be effectively used in the case of repeated pulses, as is shown in the Appendix. It consists practically of shifting the time axis to the effective irradiation time t_g of the first heat pulse. Then the analytical solution (Eq. (1)) is considered in the data reduction.

5. SENSITIVITY ANALYSIS

The simulated curves were calculated using the formula (Eq. (1)). The temperature T_{lim} was chosen to be 1 K. Axial and radial heat losses from the sample were assumed to be the same $H = H_0 = H_e = 0.05$. Sensitivities are defined as

$$S_\beta = \beta \frac{\partial T}{\partial \beta}, \tag{9}$$

where T is the rear face temperature and β is the appropriate parameter—thermal diffusivity a, the adiabatic limit temperature T_{lim}, or the Biot number H.

Figure 1 presents the simulated temperature rise and sensitivities versus dimensionless time ($\tau = at/e^2$) curves. Five pulses that follow with the time period $\tau_p = at_p e^{-2} = 0.1$ are considered here. Because of the linear

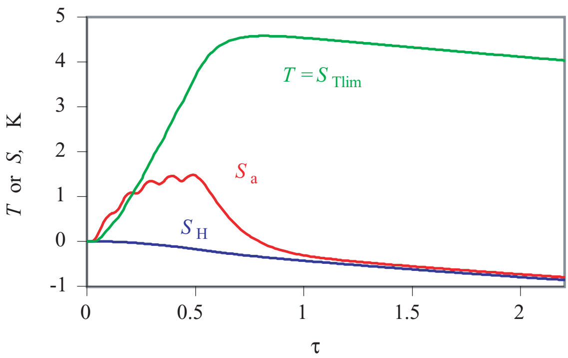

Fig. 1. Simulated temperature rise T, sensitivity to thermal diffusivity S_a, sensitivity to limit temperature $S_{T_{\lim}}$, and sensitivity to the Biot number S_H vs. dimensionless time τ curves.

dependence of the temperature rise on the adiabatic limit temperature $T_{\lim}$, the temperature rise curve corresponds to the sensitivity to the adiabatic limit temperature $T_{\lim}$ versus time curve $S_{T_{\lim}}$. The figure shows, that curves $S_{T_{\lim}}$, S_a (S_a is the sensitivity to the thermal diffusivity) and S_H (S_H, sensitivity to the Biot number), have different shapes and the curves are uncorrelated. This indicates that a least-squares-fitting-based data reduction process can be effectively used for unique estimation of desired parameters a, $T_{\lim}$, and H [6]. Figure 1 shows that the sensitivity to thermal diffusivity results from its magnitude being comparable to the temperature. This confirms that the flash method with repeated pulses is reasonably sensitive to changes in the thermal diffusivity. Maximum values of sensitivity to the thermal diffusivity curves lie periodically after each pulse application around the half time $\tau_{0.5}$ (the half-time $\tau_{0.5} = at_{0.5}e^{-2} \approx 0.139$ is the time corresponding to a rise in the temperature to half of its maximum value in the standard flash method). These points correspond to the optimal points in a simple data reduction experiment. When the temperature rise versus time curve reaches its maximal value, the curve become practically insensitive to changes in the thermal diffusivity. These facts indicate that the thermal diffusivity estimation should be based on analyzing the rising part of the temperature rise versus time curve. The sensitivity to the Biot number is smaller than the sensitivity to thermal diffusivity, especially for the rising part of the temperature rise versus time curve. The Biot number influences the thermal response less than the thermal diffusivity, which is a positive influence since we consider this parameter as a nuisance one.

Figure 2 shows how the sensitivity to the thermal diffusivity depends on the period of the applied heat pulses. This figure shows the sensitivity to

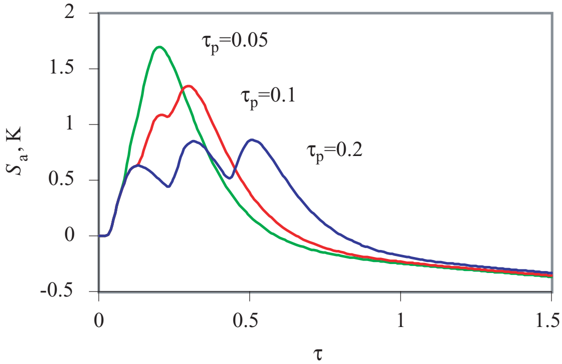

Fig. 2. Sensitivity to thermal diffusivity S_a vs. dimensionless time τ curves calculated for different heat pulse period τ_p. Maximum values correspond to the optimum times for a single measurement.

the thermal diffusivity versus time curves calculated for three applied pulses using three different heat pulse periods $\tau_p = 0.05$, 0.1, and 0.2. We see, that the maximum in sensitivity increases and is shifted to shorter times when the heat pulse period decreases. The entire sensitivity for a longer heat pulse period is spread over longer times. Figure 3 emphasizes the synergetic effect of the sensitivity of the method on a decrease of the heat pulse period τ_p.

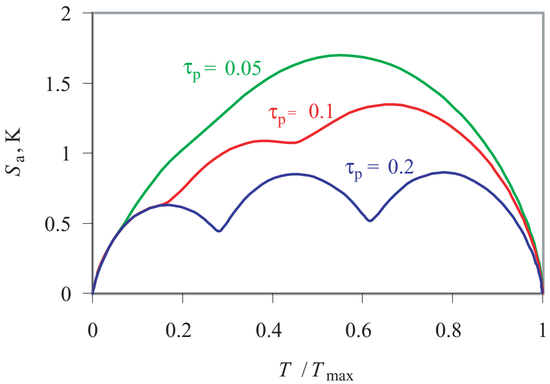

Fig. 3. Sensitivity to thermal diffusivity S_a vs. dimensionless temperature rise T/T_{max} curves, where T_{max} is the maximum temperature rise, calculated for different heat pulse period τ_p.

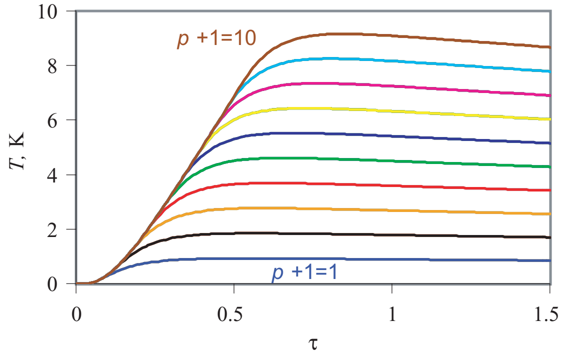

Fig. 4. Simulated temperature-rise curves calculated considering various numbers of pulses ($p+1=1, 2, \ldots, 10$) assuming the same energy for a pulse ($T_{\rm lim}=1$ K; Case No. 1).

Here the sensitivities are shown as a function of the dimensionless temperature rise $T/T_{\max}$, where $T_{\max}$ is the maximum temperature increase. We see that if the period $\tau_{\rm p}$ decreases to zero, the maximum sensitivity occurs at approximately the temperature that corresponds to half of the maximum temperature increase. Enlargement of the period $\tau_{\rm p}$ causes several maxima of the sensitivity curves that occur at half-times $\tau_{0.5}$ for every single heat pulse.

To view the influence of the number of pulses, the following calculations were performed. Two different cases were considered: the first, when all the pulses have the same energy, and the second, when we assume the same total energy enters the sample and the energy of a heat pulse depends

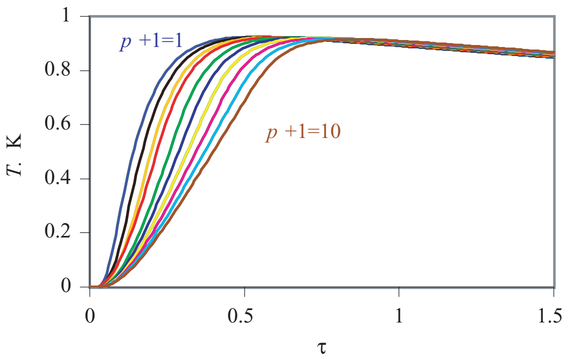

Fig. 5. Simulated temperature-rise curves calculated considering various numbers of pulses ($p+1=1, 2, \ldots, 10$) assuming the same total energy enters the sample ($T_{\rm lim}=1/(p+1)$ K; Case No. 2).

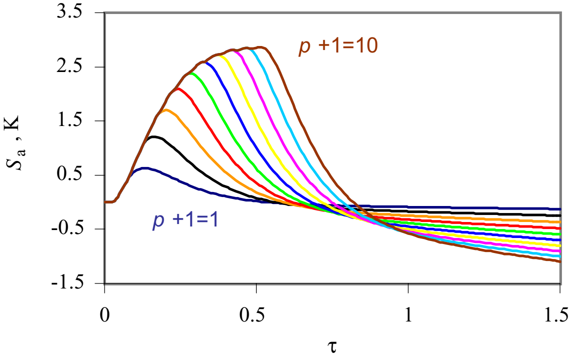

Fig. 6. Sensitivity to thermal diffusivity S_a vs. dimensionless time τ curves (Case No. 1).

on the number of applied pulses. Figures 4 and 5 present simulated temperature rise versus dimensionless time curves calculated for Cases No. 1 and 2—when $T_{\text{lim}} = 1$ and $T_{\text{lim}} = 1/(p+1)$, respectively. Figures 6 and 7, where sensitivities to the thermal diffusivity are shown as a function of dimensionless time and dimensionless temperature rise, respectively, indicate that if the number of pulses increases, the sensitivity of the flash method with repeated pulses increases. The dependence of the thermal diffusivity estimation sensitivity on the applied number of pulses is nonlinear; the increase is larger for a smaller number of pulses. Maximum sensitivity has generally the tendency to occur at higher dimensionless times and higher dimensionless temperature rises, which is a logical consequence of an increase in the overall exposure time.

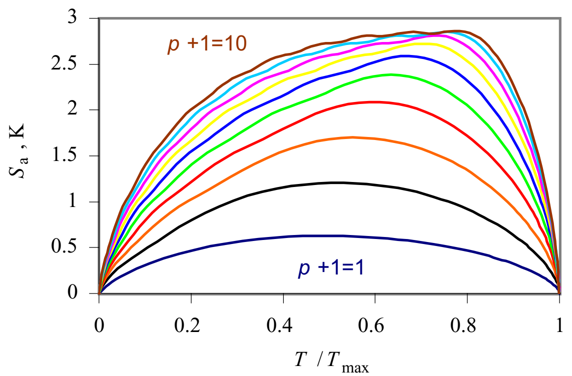

Fig. 7. Sensitivity to thermal diffusivity S_a vs. dimensionless temperature rise T/T_{max} curves (Case No. 1).

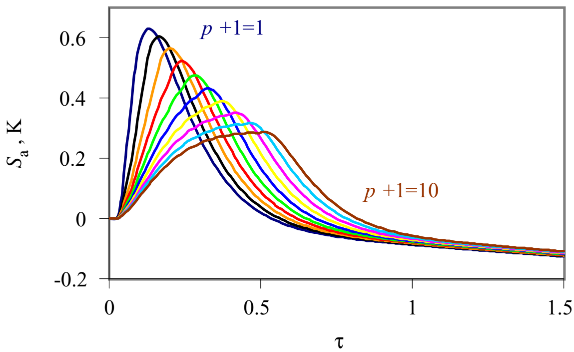

Fig. 8. Sensitivity to thermal diffusivity S_a vs. dimensionless time τ curves (Case No. 2).

On the other hand, Figs. 8 and 9, where sensitivity to the thermal diffusivity, similarly as for Case No. 1, are presented as a function of the dimensionless time and the dimensionless temperature rise, respectively, show the opposite influence. If the number of pulses among which the overall energy is split increases, the sensitivity to the thermal diffusivity decreases. This negative phenomenon is enhanced by the fact that the maximum sensitivity occurs at higher times and higher dimensionless temperature rises when the number of pulses increases as in Case No. 1.

6. OPTIMUM EXPERIMENTAL DESIGN

The criterion chosen for the optimal design analysis was the ratio of determinants Δ/Δ^2 of $\mathbf{S}^T\mathbf{S}$, which contains the product of the sensitivities

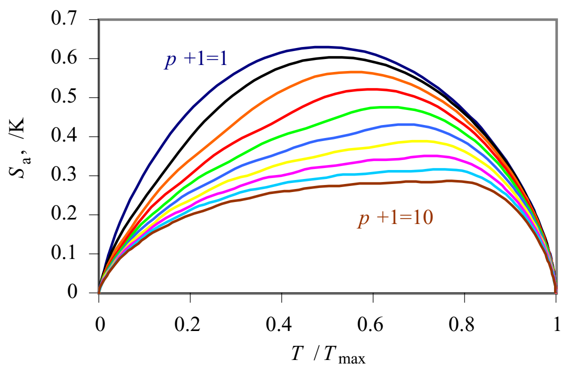

Fig. 9. Sensitivity to thermal diffusivity S_a vs. dimensionless temperature rise T/T_{max} curves (Case No. 2).

and their transpose [6]. As follows from the theory, maximizing the ratio Δ/Δ^2 has the effect to minimize the confidence interval for the resulting estimate of the thermal diffusivity. In the case of three parameters, $\Delta = |\mathbf{S}^T\mathbf{S}|$ is a 3×3 matrix and the determinant is given by

$$\Delta = |\mathbf{S}^T\mathbf{S}| = \begin{vmatrix} d_{11} & d_{12} & d_{13} \\ d_{21} & d_{22} & d_{23} \\ d_{31} & d_{32} & d_{33} \end{vmatrix}. \tag{10}$$

Here

$$d_{ij} = \frac{1}{t_n}\int_0^{t_n} S_i(t)\, S_j(t)\, dt, \qquad i, j = 1, 2, \text{and } 3 \tag{11}$$

where indices 1, 2, and 3 refer to parameters a, T_{lim}, and H, respectively, and

$$\Delta_2 = \begin{vmatrix} d_{22} & d_{23} \\ d_{32} & d_{33} \end{vmatrix}. \tag{12}$$

This criterion corresponds to the case when the thermal diffusivity a is understood as the parameter of interest, and the adiabatic temperature limit T_{lim} and the Biot number H are additional nuisance parameters.

Figure 10 presents the Δ/Δ^2 criterion calculated for various periods τ_p of the applied heat pulses. Results confirm that which comes from sensitivity

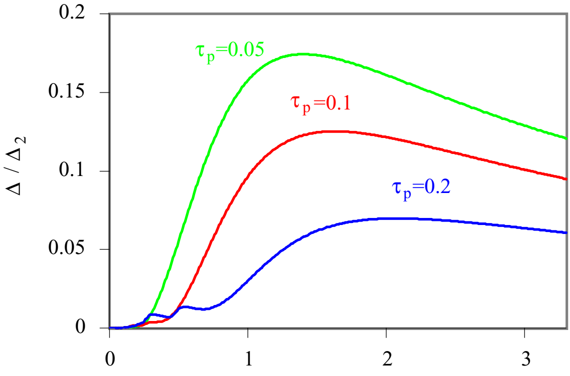

Fig. 10. Results of optimal experimental design analysis (Δ/Δ_2 criterion) calculated for different heat pulse periods τ_p. Shorter pulse period results in increased values of Δ/Δ_2 and in decreases of the optimum time of measurement τ_n.

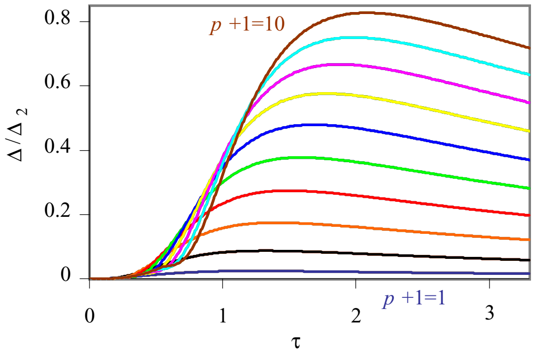

Fig. 11. Dependence of the value of Δ/Δ_2 determinants ratio calculated for different number of pulses $p+1$ assuming the same energy for each pulse (Case No. 1).

analyses. An increase of the period τ_p decreases maximum values of Δ/Δ^2 ratio, which confirms a decrease of the sensitivity to the thermal diffusivity. An increase of the time period τ_p also increases the optimal time of measurement τ_n ($\tau_n = 1.4$, 1.63, and 2.08 for $\tau_p = 0.05$, 0.1, and 0.2, respectively).

Figures 11 and 12 present results of the study of the influence on the number of pulses. We see the positive influence—an increase of the maximum of the Δ/Δ^2 value only in Case No. 1 (the case of the same heat pulse energy); if the overall energy is only split among the pulses (Case No. 2), the value of the maximum of the Δ/Δ^2 decreases with an increase of the number of pulses. The calculations also give optimum duration of experiment τ_n as summarized in Table I.

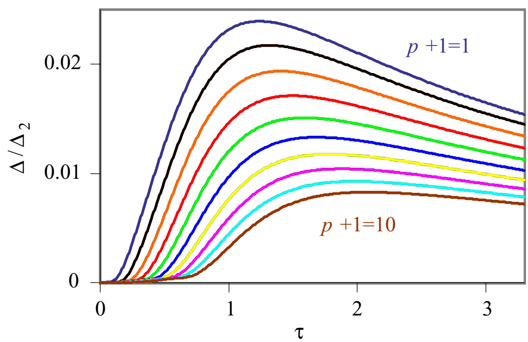

Fig. 12. Dependence of the value of Δ/Δ_2 determinants ratio calculated for different number of pulses $p+1$ assuming the same total energy that enter the sample (Case No. 2).

Table I. Results of Optimal Design Calculation for Various Number of Heat Pulses

	Case No. 1		Case No. 2	
$p+1$	Δ/Δ_2	τ_n	Δ/Δ_2	τ_n
1	0.024	1.24	0.024	1.23
2	0.087	1.31	0.022	1.31
3	0.17	1.40	0.019	1.4
4	0.27	1.50	0.017	1.49
5	0.38	1.59	0.015	1.59
6	0.48	1.69	0.013	1.68
7	0.58	1.78	0.012	1.78
8	0.67	1.88	0.010	1.88
9	0.75	1.98	0.009	1.98
10	0.83	2.08	0.008	2.08

7. CONCLUSIONS

The achieved results of the analyses could be generalized as follows:

1. A decrease of the period τ_p of the applied heat pulses has the effect of an increase of the sensitivity to the thermal diffusivity.
2. Maximum values of the sensitivity to thermal diffusivity curves occur periodically after each pulse application around half-times $\tau_{0.5}$ for each individual heat pulse; if the time period τ_p approaches zero, the maximum of the sensitivity curve occurs near the half-time $\tau_{0.5}$ of the overall temperature rise curve. This is why the thermal diffusivity estimation should be based on analyzing the rising part of the temperature rise versus time curve.
3. An increase in the number of applied heat pulses has a positive influence only when the pulses have the same energy. In the case, when the overall energy is split only among the applied pulses, an increase of the number of pulses has the effect of a decrease of the sensitivity of the method.
4. An increase of the heat pulse period and an increase of the number of pulses have the effect of an increase in the optimal duration of the experiment.

APPENDIX

If the heat pulses are instantaneous, analytically described by the shape $\phi(t) = Q\delta(t - kt_p)$, the expressions for the rear face temperature rise versus time evolution (Eq. (1)) can be written in the simplified form,

$$T_D = \sum_n \sum_i A_n \exp[v_n(it_p - t)]. \tag{13}$$

If the function $\varphi(t)$ describes a heat-pulse shape (assuming that the applied pulses have the same shape), the function $\phi(t) = Q\varphi(t - kt_{\mathrm{p}})$; $k = 0, 1, \ldots, p$ describes shapes of extended repeated heat pulses. Provided that the function $\phi(t)$ is normalized, i.e.,

$$\int_0^{\infty} \phi(t')\, dt' = 1, \tag{14}$$

the exact solution that describes the rear-face temperature rise versus time evolution can be derived in the form [15],

$$T_{\mathrm{E}} = \sum_n \sum_i \left[\int_0^{\mathrm{t}} \varphi(t' - it_p) \exp(v_n t')\, dt' \right] A_n \exp[v_n(it_{\mathrm{p}} - t)]. \tag{15}$$

Because of the periodicity of the pulses

$$\varphi(t' - it_{\mathrm{p}}) = \varphi(t') \tag{16}$$

and

$$T_{\mathrm{E}} = \sum_n \sum_i I_{1ni} \mathrm{A}_n \exp[v_n(it_{\mathrm{p}} - t)], \tag{17}$$

where

$$I_{1ni} = \int_0^{\mathrm{t}} \varphi(t') \exp(v_n t')\, dt'. \tag{18}$$

If we shift the time axis taking the time t_{g} as the time origin, the temperature T_{D} changes to

$$T_{\mathrm{D}} = \sum_n \sum_i A_n \exp[v_n(it_{\mathrm{p}} - t + t_{\mathrm{g}})] = \sum_n \sum_i I_{2ni} A_n \exp[v_n(it_{\mathrm{p}} - t)], \tag{19}$$

where

$$I_{2ni} = \exp(v_n t_{\mathrm{g}}). \tag{20}$$

Expressing I_{1ni} and I_{2ni} in a Taylor series according to time τ, and ignoring quadratic and higher terms [14], it can be shown, that I_{1ni} is equal to I_{2ni} when

$$t_{\mathrm{g}} = \int_0^{\mathrm{t}} t' \varphi(t')\, dt'. \tag{21}$$

This confirms the above-described concept of the correction of the finite pulses duration.

REFERENCES

1. W. J. Parker, R. J. Jenkins, C. P. Butler, and G. L. Abbott, *J. Appl. Phys.* **32**:1679 (1961).
2. L. Vozár, G. Groboth, W. Hohenauer, and E. Hübner, in *Proc. 10th Int. Conf. Photoacoustic Phototherm. Phenomena* (AIP Woodbury, New York, 1999), pp. 351–353.
3. L. Vozár and W. Hohenauer, *High Temp. High Press.* **33**:9 (2001).
4. A. F. Emerey and A. V. Nenarokomov, *Meas. Sci. Technol.* **9**:864 (1998).
5. K. J. Downing, B. F. Blackwell, and R. J. Cochran, *Num. Heat Transf. B* **36**:33 (1999).
6. J. V. Beck and K. J. Arnold, *Parameter Estimation in Engineering and Science* (Wiley, New York, 1977), pp. 419–480.
7. L. Vozár and G. Groboth, *High Temp. High Press.* **29**:191 (1997).
8. R. Taktak, J. V. Beck, and E. P. Scott, *Int. J. Heat Mass Transf.* **36**:2977 (1993).
9. A. Haji-Sheikh, Y. S. Hong, S. M. You, and J. V. Beck, *J. Heat Transf.* **120**:568 (1998).
10. J. Gembarovič, L. Vozár, and V. Majerník, *Int. J. Heat Mass Transfer* **33**:1563 (1990).
11. L. Vozár and W. Hohenauer, in *Proc. VIth Int. Asian Thermophys. Prop. Conf.*, Guwahati, India (in press).
12. L. Vozár, *Flash Method for the Thermal Diffusivity Measurement. Theory and Praxis* (Univerzita Konštantína Filozofa, Nitra, 2001), pp. 22–25.
13. T. Azumi and Y. Takahashi, *Rev. Sci. Instrum.* **52**:1411 (1981).
14. A. Degiovanni, *Int. J. Heat Mass Transfer* **30**:2199 (1987).
15. D. A. Watt, *Brit. J. Appl. Phys.* **17**:231 (1966).