Pergamon

Int. J. Heat Mass Transfer. Vol. 38, No. 15, pp. 2885–2891, 1995

0017-9310/95 $9.50+0.00

0017-9310(95)00032-1

Using non-linear χ^2 fit in flash method

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(*Received* 29 *September* 1994)

Abstract—The paper presents a data reduction method for determination of thermal diffusivity in flash method. It is based on non-linear χ^2 fit to a model which takes into consideration heat losses. The advantage of the method is that it does not need a knowledge of base line and is suitable for data disturbed by noise and mains hum. A comparison with some other data reduction methods is given.

1. INTRODUCTION

The flash method introduced by Parker *et al.* [1] in 1961 has become a widely accepted method for measuring thermal diffusivity of solids. In this method, a small disk-shaped sample is subjected to a very short burst of radiant energy from a laser or a flash lamp and the resulting temperature rise of the rear surface of the specimen is measured by a thermocouple or IR detector. Thermal diffusivity is then computed from the resulting temperature versus time data.

Data reduction methods play a significant role in determination of thermal diffusivity. In the original paper [1] only one or a few data points from the measured temperature versus time curve were used to calculate the thermal diffusivity. The progress in computer technology during the past few decades have resulted in a number of data reduction techniques which are based on analysis of any part of the temperature vs time curve [2–5]. The precision of a method depends upon adequately meeting the boundary conditions of the ideal theoretical model [1]. This assumes uniform and instantaneous heat pulse absorbed in a very thin layer of a homogeneous, isotropic and opaque samples, no heat losses from the sample to the surroundings, temperature independence of the thermal properties within the temperature rise caused by irradiation.

It is often not possible to fulfill these conditions, mainly heat losses and finite pulse time duration are usually unavoidable. In earlier works these effects were accounted for by multiplying the thermal diffusivity determined from the ideal model with the appropriate numerical factor [6, 7]. More recent methods are based on the general mathematical model obtained as a solution of a two-dimensional (2D) heat conduction equation with the heat losses from the sample surface. In those methods the thermal diffusivity is determined either by means of several particular points or by using the temporal moments of the defined temperature interval of the thermogram [8, 9]. Maillet *et al.* [10] studied the influence of measurement noise on the calculation of thermal diffusivity utilizing Degiovanni *et al.* methods. Another approach is based on the knowledge that the response is less perturbed by heat losses at the start of the thermogram [11, 12]. Other papers take the advantage of the Laplace transform to convert the working equation into a simpler one [13, 14].

The presented method is based on numerical fitting to the model which takes into account heat losses and mains hum.

2. THEORY AND DATA REDUCTION

2.1. *Theory*

When performing measurements with the flash method as described by Parker *et al.* [1] the temperature rise at the rear surface of the test specimen is given by :

$$T(l,t) = T_0 + T_1(l,t) \tag{1}$$

$$T_1(l,t) = \frac{Q}{\rho C_p l \pi r^2}\left[1 + 2\sum_{n=1}^{\infty}(-1)^n \exp\left(-\frac{n^2\pi^2\alpha t}{l^2}\right)\right]$$

where T_0 is the initial temperature of the specimen [K], Q is the energy absorbed at the front surface (J), ρ is the density [kg m^{-3}], C_p is the specific heat [J kg^{-1}K^{-1}], α is the thermal diffusivity [m^{2}s^{-1}], l is the thickness [m], r is the radius [m] of the specimen and t is the time [s] elapsed since the flash energy was absorbed at the front surface of the test specimen. After infinite time the rear face temperature will reach its maximum value

NOMENCLATURE

a	thermal diffusivity divided by square thickness	T_{Imax}	maximum temperature rise on back surface
D	Hessian matrix	U	voltage response
B	amplifier gain multiplied by the thermoelectric power of the thermocouple	U_0	baseline voltage
		U_{hum}	additional voltage caused by mains hum
C_p	specific heat	U_{max}	maximum voltage of the response.
L	Biot number		
l	sample thickness		
p	model parameter vector	Greek symbols	
Q	absorbed energy	α	thermal diffusivity
r	sample radius	α_s	thermal diffusivity value used in simulations
T	temperature		
T_0	initial temperature	β	possitive roots of equation (5)
T_I	temperature rise at ideal conditions	λ	LM method weighting factor
T_L	temperature rise when heat losses occur	ω_i	multiples of the basic mains frequency
		ρ	density
t	time	σ	standard deviation.

$$T_{\text{Imax}} = \frac{Q}{\rho C_p l \pi r^2}. \tag{2}$$

Equation (1) is valid only under the ideal conditions. In the case of heat transfer between the sample and its environment, more complicated expression is used. Watt [15] proposed a solution for 1D heat flow and heat losses from the two parallel surfaces as

$$T(l,t) = T_0 + T_{L_{1,2}}(l,t) \tag{3}$$

$$T_{L_{1,2}}(l,t) = T_{\text{Imax}} \sum_{n=1}^{\infty} Y_n(0)\, Y_n(l) \exp\left(-\frac{\beta_n^2 \alpha t}{l^2}\right)$$

where

$$Y_n(x) = \frac{\sqrt{2(\beta_n^2+L_2^2)}[\beta_n \cos(\beta_n x/l) + L_1 \sin(\beta_n x/l)]}{\sqrt{(\beta_n^2+L_1^2)(\beta_n^2+L_2^2+L_2)+L_1(\beta_n^2+L_2^2)}} \tag{4}$$

and β_n ($n = 1,2,3,\ldots$) are the positive roots of

$$(\beta^2 - L_1 L_2)\tan\beta = \beta(L_1 + L_2). \tag{5}$$

Since the thermocouple and the amplifier have linear voltage vs temperature profiles, at least over a range of a few degrees, the voltage vs time curve can be expressed by :

$$U(t) = U_0 + BT^*(l,t) \tag{6}$$

where U_0 is the baseline of the amplified thermocouple voltage before the flash pulse, B is the amplifier gain multiplied by the voltage response per degree for the thermocouple used, and $T^*(l,t)$ represents either $T_I(l,t)$ or $T_{L_{1,2}}(l,t)$.

The amplified thermocouple voltage readings may be disturbed by superimposed mains hum, adequately expressed by :

$$U_{\text{hum}} = \sum_{i=1}^{N} h_i \cos(\omega_i t + \varphi_i) \tag{7}$$

where h_i, ω_i and φ_i are amplitude, frequency and phase of the first $N(3 \div 5)$ multiples of the basic mains frequency ω_1. This influence can be minimized by numerical smoothing, which requires enough measured points and is not applicable for responses of the order of $1/\omega_1$.

As heat losses usually do not occur during short time measurements, the response model for this case can be then expressed by

$$U(t) = U_0 + BT_I(l,t) + U_{\text{hum}}. \tag{8}$$

In the case of longer responses the influence of mains hum can be minimized by smoothing or during the measurement by using sampling rate synchronized by mains frequency. Heat losses should be taken into account. In this case our model is

$$U(t) = U_0 + BT_{L_{1,2}}(l,t). \tag{9}$$

2.2. *Data reduction method*

Due to the complexity of our response models (8) and (9) we have chosen the Levenberg–Marquardt (LM) χ^2 based fitting method, which enables to process nonlinear models with arbitrary number of parameters. Detailed description of the method, together with source code is given in [16], therefore we give only a brief outline.

The LM method is an extension of inverse-Hessian and steepest descend function minimization methods [16] for the case of χ^2 merit function

$$\chi^2(\vec{p}) = \sum_{i=1}^{N} \left[\frac{y_i - y(x,\vec{p})}{\sigma_i}\right]^2 \tag{10}$$

where y_i are measured data samples, σ_i corresponding standard deviations, $\vec{p} = (p_1, p_2, \ldots p_M)$ the model parameters and $y(x, \vec{p})$ the model being fitted.

The inverse-Hessian method takes advantage of the fact that a function f can be at a point $\vec{p}$ which is sufficiently close to the searched minimum approximated by a quadratic form. In that case one can directly compute the minimizing parameters as

$$\vec{p}_{\min} = \vec{p} - D^{-1}\nabla\chi^2(\vec{p}) \tag{11}$$

where

$$D = \left[\frac{\partial^2\chi^2}{\partial p_k\, \partial p_l}\right]$$

is the Hessian matrix of χ^2 at $\vec{p}$ and $\nabla\chi^2(\vec{p})$ is the gradient of χ^2 with respect to p_k,

$$\nabla\chi^2(\vec{p}) = \left(\frac{\partial\chi^2}{\partial p_1}, \ldots, \frac{\partial\chi^2}{\partial p_M}\right).$$

From equation (10) we obtain

$$\frac{\partial^2\chi^2}{\partial p_k\, \partial p_l} = 2\sum_{i=1}^{N}\frac{1}{\sigma_i^2}\left[\frac{\partial y(x_i,\vec{p})}{\partial p_k}\frac{\partial y(x_i,\vec{p})}{\partial p_l} - [y_i - y(x_i,\vec{p})]\frac{\partial^2 y(x_i,\vec{p})}{\partial p_k\, \partial p_l}\right]. \tag{12}$$

Since the term $[y_i - y(x_i, \vec{p})]$ in equation (12) should be a random measurement error of each point which is uncorrelated with the model, the second derivatives cancel out. Thus, for the case of χ^2 function minimization is the Hessian dependent only on first derivatives of the model function :

$$\frac{\partial^2\chi^2}{\partial p_k\, \partial p_l} = 2\sum_{i=1}^{N}\frac{1}{\sigma_i^2}\left[\frac{\partial y(x_i,\vec{p})}{\partial p_k}\frac{\partial y(x_i,\vec{p})}{\partial p_l}\right]. \tag{13}$$

If we introduce

$$c_{kl} = \frac{1}{2}\frac{\partial^2\chi^2}{\partial p_k\, \partial p_l}, \quad b_k = -\frac{1}{2}\frac{\partial\chi^2}{\partial p_k}$$

$$\text{and} \quad \delta p_l = p_{l\min} - p_l,$$

equation (11) can be rewritten as a set of linear equations

$$\sum_{i=1}^{M} c_{kl}\delta p_l = b_k. \tag{14}$$

In the iterative procedure, the system (14) is solved for δp_l, which define the next approximation of the searched parameters.

The steepest descend function minimization method works well also for the points, which are far from the searched minimizing point, where approximation by the quadratic form is impossible. The next approximation can be in this case computed by

$$\delta p_k = \text{const.}\, b_k. \tag{15}$$

The main idea of the LM method is to vary smoothly between the inverse-Hessian and steepest descend methods : the latter is used far from minimum, while the first in its vicinity, where approximation by the quadratic form is possible.

The authors proposed, first, to approximate the constant in equation (15) by $1/\lambda c_{kk}$. From (15) we then get

$$\lambda c_{kk}\delta p_k = b_k \tag{16}$$

and, second, to replace the system (14) by

$$\sum_{i=1}^{M} c'_{kl}\delta p_l = b_k \tag{17}$$

where

$$c'_{kl} = \begin{cases} c_{kk}(1+\lambda) & \text{if } k = 1 \\ c_{kl} & \text{otherwise} \end{cases}. \tag{18}$$

During the iterative process, the contribution of both minimization methods can be controlled by the factor λ : when far from the minimum, λ should be large and (17) goes to (16), otherwise λ should decrease and (17) goes to (14).

2.3. *LM technique in the case of flash method*

In this section we mention some details concerning the implementation of the LM method for the models (8) and (9). Generally, it is necessary to provide a function which evaluates the model and its partial derivatives with respect to the fitted parameters.

The case of the model (8) without heat losses and with superimposed mains hum was straightforward. The fitted parameters were U_0, B, $a = \alpha/l^2$ and h_i, ω_i and φ_i. Parameter a was used instead of α due to instability of the fitting process caused by its small value.

Although there are no special requirements on the fitted model function for the LM technique, we simplified the second, heat loss model (9) by a presumption that heat losses from both rear and front faces of the specimen were equal ($L1 = L2 = L$). In that case we obtain :

$$T_L(l,t) = T_{l\max}\sum_{n=1}^{\infty} Y_n \exp(-\beta_n^2 at) \tag{19}$$

where

$$Y_n = \frac{\beta_n^2\cos\beta_n + L\beta_n\sin\beta_n}{0.5(\beta_n^2 + L^2) + L}. \tag{20}$$

From (5) we then get

$$(\beta^2 - L^2)\tan\beta = 2\beta L. \tag{21}$$

The following parameters should then be searched for : $T_{l\max}$, a, U_0 and L.

For the LM technique, it is necessary to compute all partial derivatives of the model function with respect to the parameters being fitted :

$$\frac{\partial T_L}{\partial T_{l\max}} = \sum_{n=1}^{\infty} Y_n \exp(-\beta_n^2 at) \tag{22}$$

$$\frac{\partial T_L}{\partial a} = -T_{\mathrm{lmax}} t \sum_{n=1}^{\infty} Y_n \beta_n^2 \exp(-\beta_n^2 at) \tag{23}$$

$$\frac{\partial T_L}{\partial L} = T_{\mathrm{lmax}} \sum_{n=1}^{\infty} \left(\frac{\partial Y_n}{\partial \beta_n} - 2 Y_n \beta_n at \right) \frac{\partial \beta_n}{\partial L} \exp(-\beta_n^2 at) \tag{24}$$

$$\frac{\partial T_L}{\partial U_0} = 1. \tag{25}$$

We see that in order to compute $\partial T_L / \partial L$ it is necessary to know $\partial \beta_n / \partial L$, which we can get from equation (21):

$$\frac{\partial \beta_n}{\partial L} = \frac{2(\beta + L \tan \beta)}{2\beta \tan \beta + \dfrac{\beta^2 - L^2}{\cos^2 \beta} - 2L}. \tag{26}$$

In order to evaluate the derivatives (22)–(24) as well as the model (19) the positive roots of the equation (21) have to be found. Since this equation cannot be solved analytically, we solved it numerically using the Newton–Raphson method [16]. A more detailed description of solving a similar equation can be found in [17].

3. EXPERIMENTAL SET-UP

The proposed data reduction has been tested on a set of measurements made by the apparatus which is briefly described below and its principal scheme is depicted in Fig. 1. The cylindrical test specimen was irradiated by the light from a flash lamp. A quartz rod (length 40 cm) was used as the light pipe to irradiate the front surface of the test specimen placed in the center of the 40 cm long heating furnace. The whole assembly is placed in a vacuum chamber.

The temperature response was measured by an intrinsic chromel–alumel thermocouple (diameter 125 μm) where each thermocouple wire was individually attached to the test specimen. The thermocouple signal was led to an amplifier based on 725 and 741 μA in the standard configuration of gain approximately 10^5. In order to have a possibility to measure shorter responses, capacitance feedback filters, usually used to remove mains hum, were avoided and software solution of problem was preferred. A 12-bit successive approximation A/D converter controlled by a personal computer was used to digitize the amplified thermocouple response. The start time of the transient temperature recording, i.e. the time when the flash was fired, was detected by a phototransistor.

Dependent on the total measurement time, two ways of setting reading pulse frequency for sampling the thermocouple output were used :

(1) for test specimens with long response time $((l^2/\alpha) > 3)$ a signal derived from the mains frequency was used to minimize mains hum and

(2) for test specimens with short response time $((l^2/\alpha) < 3)$ a software adjustable timer built into the A/D converter was used.

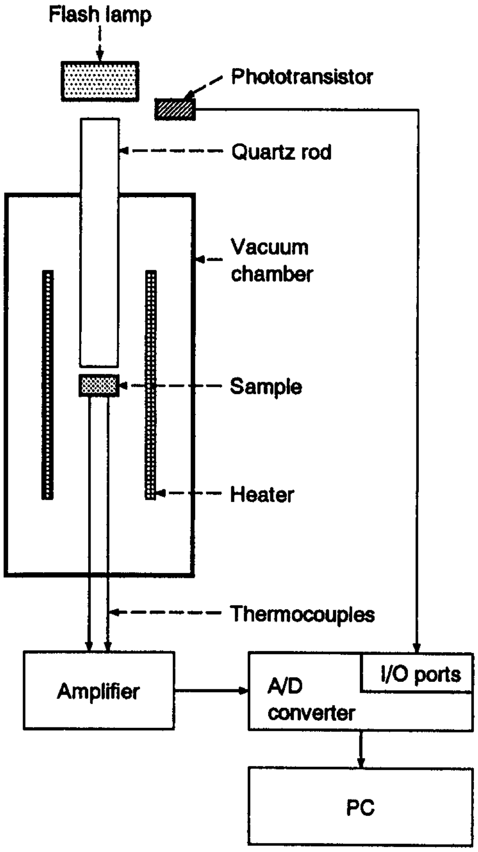

Fig. 1. Experimental set-up.

The test specimens measured in the present work were prepared from two standard reference materials from NIST, stainless steel† (diameter 12.7 mm, length 6.14 mm) and graphite‡ (diameter 12.7 mm, length 3.63 mm). The front surface of the stainless steel specimen was covered by a thin graphite layer to enhance absorption of the radiant flash energy. The presented data were measured at room temperature.

4. RESULTS AND DISCUSSION

The data reduction method as well as the data acquisition software were implemented in the C language. $\alpha_{T_{1/2}}$ value was used as an initial estimate of the diffusivity for the iterative process. Although this value was usually far from the optimum for responses with large noise and/or large heat losses, only a few iterations were necessary to find the solution. A typical processing time of a response containing 500 measured points on a 33 MHz 486 personal computer was of the order of seconds.

The proposed procedure has been tested on Monte

†Type : SRM 1462, NIST (Gaithersburg, MD).

‡Type : SRM 8425 (Bar 64, pos. 20-25), NIST (Gaithersburg, MD).

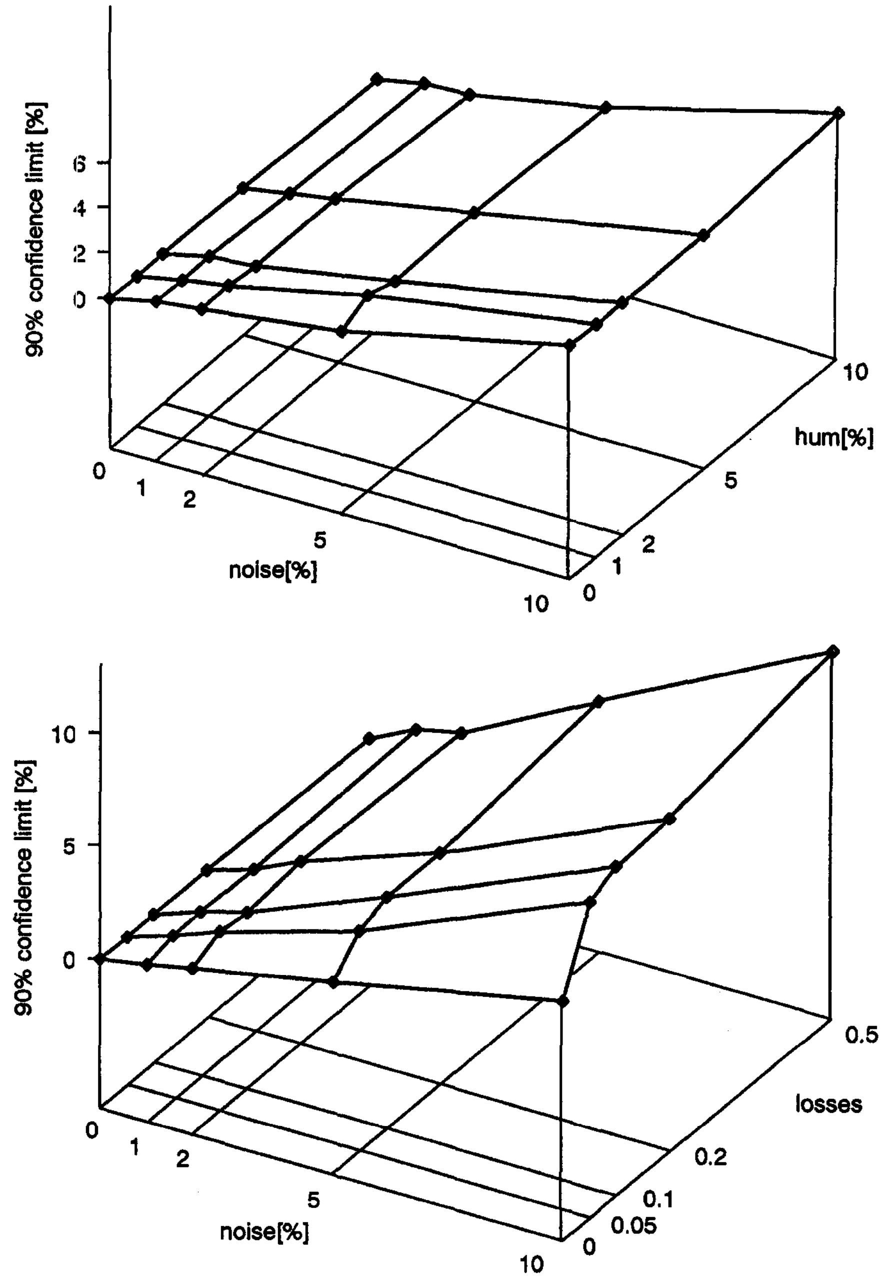

Fig. 2. 90% confidence limits for the thermal diffusivity α obtained by Monte Carlo simulation and LM fitting procedure.

Carlo simulated thermograms with $\alpha_s = 3.77 \times 10^{-6}$ m^2s^{-1}. Figures 2(a) and (b) show 90% confidence interval width of the fitted thermal diffusivity α as a function of noise and hum (model (9)), and noise and heat losses (model (8)), respectively. The 90% confidence interval is a region around α_s that contains 90% of results obtained by fitting the model parameters to a simulated noisy response. 100 noisy thermograms were computed for each combination of noise level, mains hum amplitude and Biot number L.

From Fig. 2(a) we can see that the confidence limit width is nearly independent on superimposed mains hum amplitude, with maximum width 6% of α_s for 10% noise. On the other hand, Fig. 2(b) shows an increase of the confidence limit also for growing value of Biot number L. However, for the small level of noise and heat losses, which were observed in our measurements, its width does not exceed 5% of α_s.

A similar procedure based on Monte Carlo simulation was chosen also for a comparison of the proposed LM method with some other methods. For the calculation of thermal diffusivity by these methods a software package [18] was used. Two sets of 100 thermograms were simulated with $\alpha = 3.77 \times 10^{-6}$

Table 1. Simulation results for various data reduction methods and two levels of noise ($\alpha = 3.77 \times 10^{-6}$ m^{2}s^{-1}, $L = 0.1$).

	Noise [1%]			Noise [5%]		
	α_{mean} [m^{2}s^{-1}]	D [%]	$C90$ [%]	α_{mean} [m^{2}s^{-1}]	D [%]	$C90$ [%]
Degiovanni [5]	3.908×10^{-6}	3.665	2.849	4.076×10^{-6}	8.108	7.782
Takahashi [15]	3.832×10^{-6}	1.633	5.587	3.880×10^{-6}	2.923	33.730
Gembarovič [16]	3.818×10^{-6}	1.276	3.187	3.915×10^{-6}	3.835	17.570
LM	3.771×10^{-6}	0.046	0.73	3.768×10^{-6}	-0.040	3.580

Table 2. Thermal diffusivity α of stainless steel and graphite (10^{-6} m^{2}s^{-1}).

	α_{meas} [10^{-6} m^{2}s^{-1}]	α_{NIST} [10^{-6} m^{2}s^{-1}]
Stainless steel	3.67 ± 0.08	3.77
Graphite	76.00 ± 0.19	76.60

m^{2}s^{-1} and Biot number $L = 0.1$. Both sets were disturbed by additive noise with standard deviation 1% and 5%, respectively.

Table 1 shows mean value of thermal diffusivity (α_{mean}), its deviation from the ideal value ($\alpha_s - \alpha_{mean}/\alpha_{mean}$) ($D$) and 90% confidence limit (C90) for each method. These results indicate that the presented LM method is the least sensitive to measurement noise.

In order to test the proposed fitting method on data from real experiment, a set of measurements on the two standard reference materials was done. For the stainless steel sample with the response time of the order of 10 s, sampling rate derived from mains frequency and the model (9) with heat losses were used. For the graphite sample with response time of the order of 0.5 s the model (8) incorporating mains hum fitting has been applied. The results of both measurements are given in Table 2 together with the value calculated from thermal conductivity, heat capacity and density data given by NIST [19, 20] for the particular material. The deviation between the NIST value and the value measured in this experiment is less than 3%.

Figure 3 depicts a typical response together with the

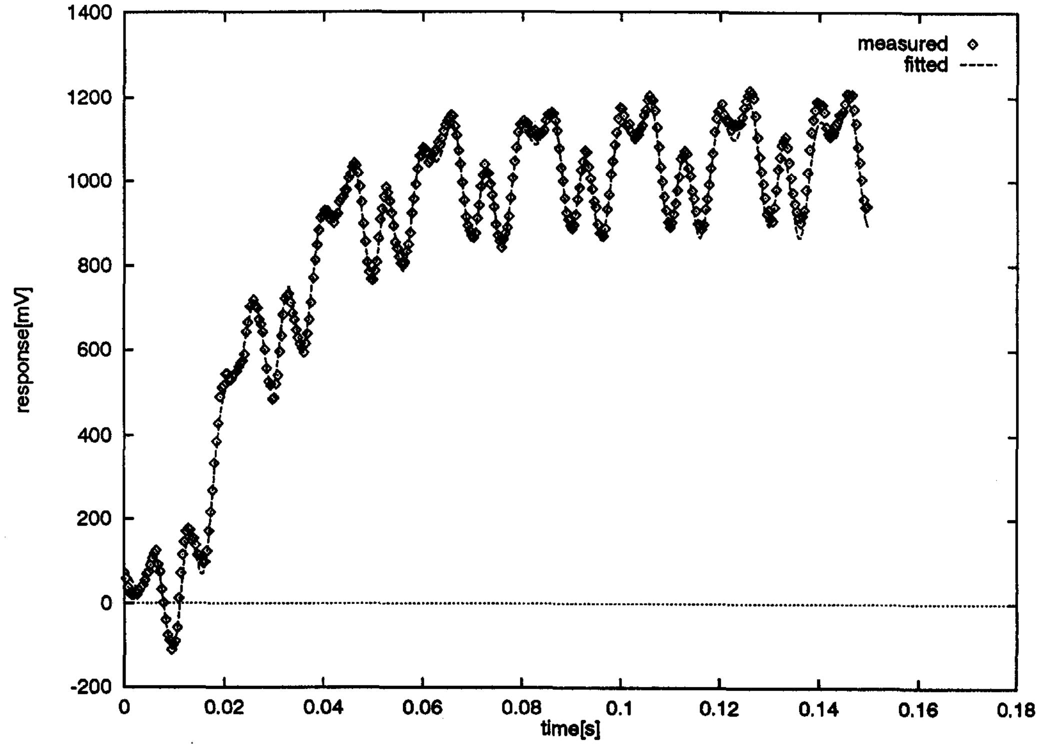

Fig. 3. Typical voltage vs time response of the graphite test specimen.

fitted model (8) for the graphite specimen. It illustrates that the model (8) describes well the influence of mains hum on our short time measurements.

5. CONCLUSIONS

We have outlined use of Levenberg–Marquardt non-linear χ^2 fitting method for data reduction in flash method. Dependent on the duration of a response two models have been proposed. The first procedure allows a set of parameters to be optimized : thermal diffusivity, Biot number, U_0 and U_{max}. Thus the method does not need the exact knowledge of a baseline or maximum amplitude. They are used only for a rough estimation of $\alpha_{T_{1/2}}$ as the starting value for iterative process, so the method is not sensitive to shifts in baseline which may occur after triggering the flash pulse. The next model allows fitting of thermal diffusivity, U_0, U_{max} and U_{hum}. This procedure is suitable for short responses disturbed by mains hum.

The method converges rapidly, consuming time only in the order of seconds when performing calculations on a 33 MHz 486 PC.

Confidence intervals of the LM method for various levels of measurement noise were estimated from Monte Carlo simulated responses. Their comparison with corresponding confidence levels of some other data reduction methods showed that the LM method was the least sensitive to the noise.

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