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AN APPARATUS FOR THE THERMAL DIFFUSIVITY MEASUREMENT USING THE LASER FLASH METHOD WITH REPEATED PULSES

L. Vozár [1], W. Hohenauer [2]

Department of Physics, Faculty of Natural Sciences,
Constantine the Philosopher University (1);
Department of Materials Research, Austrian Research Centers Seibersdorf (2)

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Abstract: The paper deals with the measurement of the thermal diffusivity using the laser flash method with repeated pulses. The flash method with repeated pulses was proposed as an extension of the classical 'one pulse' laser flash method aimed to overcome particular experimental difficulties associated with the measurement of some types of insulators, temperature sensitive materials and large-grain heterogeneous materials. Unlike the other approaches, based on reducing the energy intensity and simultaneously increasing the exposure time (the flash method with extended pulse), or substituting step (continuous) heating for pulse irradiation (the step heating method), respectively, in the flash method with repeated pulses the pulse energy consists of several consecutive pulses periodically applied to the sample front face. The thermal diffusivity is calculated from the resulting temperature rise of the rear face, in a similar way to the standard laser flash method.

The paper presents the mathematical basis of the flash method with repeated instantaneous pulses. The simple adiabatic mathematical model as well as the more realistic non-ideal theory that considers heat loss from the sample is reviewed here. The built experimental apparatus that is regularly used for serial measurement of the thermal diffusivity in the Austrian Research Centers (ARC) is described here in details. Results of the thermal diffusivity measurements of CrNi austenitic steel are presented and compared.

Nomenclature

a - thermal diffusivity [$m^2 \cdot s^{-1}$];
A_n, u_n - parameter;
H_0, H_e - Biot number;
c - specific heat [$J \cdot kg^{-1} \cdot K^{-1}$];
e - sample thickness [m];
k, n, i - index;
N - number of points;
$p+1$ - number of pulses;
Q - pulse heat energy a unit area [$J \cdot m^{-2}$];
t - time [s];
t_p - period of pulses [s];
$T(t)$ - temperature [°C];
$T_{\lim}$ - maximal temperature rise [°C];
δ - Dirac's function;
ρ - density [$kg \cdot m^{-3}$].

Introduction

The laser flash method, proposed by Parker et al [1], has become the most popular method of measuring the thermal diffusivity of solids. Here the front face of a plane sample receives a short pulse of radiant energy provided by a laser. The resulting temperature rise on the opposite (rear) face of the sample is measured, and the thermal diffusivity is computed from the temperature rise vs. time data. Several review articles have summarized the current achievement in the theory and praxis of the method [2-4].

The flash method with extended pulse [5] and the step heating method [6] were proposed in order to decrease unacceptable large temperature gradients in the sample. They are based on simultaneously reducing of the intensity of the energy source and increasing the exposure time, or substituting step (continuous) heating for pulse irradiation. Finally the flash method with repeated pulses was proposed, where the pulse energy is split into several consecutive (laser) pulses and applied periodically to the sample front face. The thermal diffusivity is here estimated from the resulting temperature rise of the rear surface as in the standard laser flash technique [7,8].

Analytical basis

The ideal model considers a homogeneous opaque thermally insulated slab of thickness e with uniform and constant thermophysical properties and the density r. The sample front face is exposed to instantaneous heat pulses repeated with the period t_p, analytically described by the shape $\phi(t) = Q\delta(t - kt_p)$; $k = 0,1,..., p$. Here Q is the heat supplied by a pulse to the unit area of the front face, $\delta(t)$ the Dirac's function and $(p+1)$ is the number of pulses. The temperature $T(t)$ at the rear face conforms the equation [7]

$$T(t) = T_{\mathrm{lim}} \left\{ k + 1 + 2\sum_{n=1}^{\infty} (-1)^n \sum_{i=0}^{k} \exp\left[n^2\pi^2 \left(it_p - t \right) \frac{a}{e^2} \right] \right\} . \tag{1}$$

Here T_{lim} is the maximal temperature rise ($T_{\mathrm{lim}} = Q/\rho ce$ with c being the heat capacity), t is the time, a the thermal diffusivity and

$$k = \left\langle \begin{array}{ll} 0, 1, ..., p - 1\ ; & kt_p \le t < (k+1)t_p \\ p & t \ge pt_p \end{array} \right. . \tag{2}$$

The non-ideal model that is more realistic in a wider temperature range considers 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 Fourier series

$$T(t) = T_{\mathrm{lim}} \sum_{n=1}^{\infty} A_n(H_0,\ H_e) \sum_{i=0}^{k} \exp\left[u_n^2 \left(it_p - t \right) \frac{a}{e^2} \right], \tag{3}$$

where

$$A_n(H_0, H_e) = \frac{2u_n^2 \left(u_n^2 + H_e^2 \right) \left(\cos u_n + \dfrac{H_0}{u_n} \sin u_n \right)}{\left(u_n^2 + H_0^2 \right)\left(u_n^2 + H_0^2 \right) + \left(H_0 + H_e \right)\left(u_n^2 + H_0 H_e \right)},\ H_0 > 0; H_e > 0 \tag{4}$$

and u_n are the positive roots of equation

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

Data reduction

The data reduction – an estimation of the thermal diffusivity consists of a least squares fitting of the theoretical curves, to the measured temperature rise vs. time evolution. It is based on minimizing of the function (Gembarovič et al 1990)

$$R = \sum_{j=1}^{N} [(T_j - T(t_j)]^2, \tag{6}$$

where T_j are the experimental temperatures measured in times t_j, N is the number of points taken into account and $T(t_j)$ is the analytical expression for the temperature rise vs. time calculated in the time t_j. As described elsewhere [9,10] the problem of finding the thermal diffusivity is transformed to solving the algebraic equations.

The theory considers that the applied heat pulses are instantaneous. The assumption is valid when the heat pulse duration is negligible 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 pulse should be taken into account. The expression for the temperature rise evolution in the case of square-wave shaped pulses has been given [8], the other formulas suitable for the heat pulse approximations were summarized elsewhere [4]. The correction originally proposed for standard 'one pulse' laser flash method based on adjustment the effective irradiation time using the center of gravity of the heat pulse, taking this time as the time origin and considering the appropriated analytical solution with an ideal instantaneous heat pulse [11] can be utilized also in the case of the flash method with repeated pulses as shown in [10].

Experimental apparatus

The laser flash measuring system is the home-made experimental apparatus built and installed at the Materials Research Division of the Austrian Research Centre in Seibersdorf, where it is regularly used for serial measurements of the thermal diffusivity of solids. It consists of Nd:Cr:GGG (galium-gadolinium garnet doped with neodymium) glass laser (BLS400, Baasel Lasertech) working at wave length λ = 1.064 μm with the justified pulse energy ~10 J.cm^{-2}. The transient temperature is measured by the liquid nitrogen cooled HgCdTe infrared detector (HCT-80, Infrared Associated, Inc.) with preamplifier (PPA-15-DC). The detector has a time constant of about 300 ns and is set to detect radiation from the central square area (~4 mm^2) at the sample rear face. The sample is supported in a horizontal position in the vacuum chamber. A short tantalum tube acts as the resistance heater and allows measurements in the temperature range from 20 up to 1850 °C. The furnace is powered by a DC current from the power source (TN 10-5000, Heinzinger Elektronik). The sample temperature sensor consists of the steel encapsulated K-type (NiCr/Ni) thermocouple of 1 mm in diameter, or spot-welded S-type (Pt/PtRh10) thermocouple made from wires of 0.35 mm in diameter (Heraeus). All data acquisition and control is performed using the standard measurement hardware (PCI 20001C-A carrier, PCI 20002M 12bit A/D, 20003M 12bit D/A, PCI 20007M Timer/IO, Burr-Brown).

The apparatus is constructed along two axes. The laser is placed horizontally. The laser beam is reflected by a bending mirror and follows vertically through a glass window (BK7) into a water-cooled stainless-steel vacuum chamber. The vacuum is stabilized using the turbo pump (TPH 110, Pfeiffer Wakuumtechnik) at values of 10^{-5} Pa order. The sample holder consists of three molybdenum rods that fix the sample in a horizontal position in the central zone of the furnace. The construction allows the irradiation of the lower (front) face of the sample and the measurement of temperature and temperature response on the upper (rear) face of the sample. The detachable top of the vacuum chamber fixes the IR temperature sensor that is focused with CaF_2 lens and mechanical iris. The chamber top contains the movable tubes that allow the setting and, through a window, the checking of the thermocouples' position.

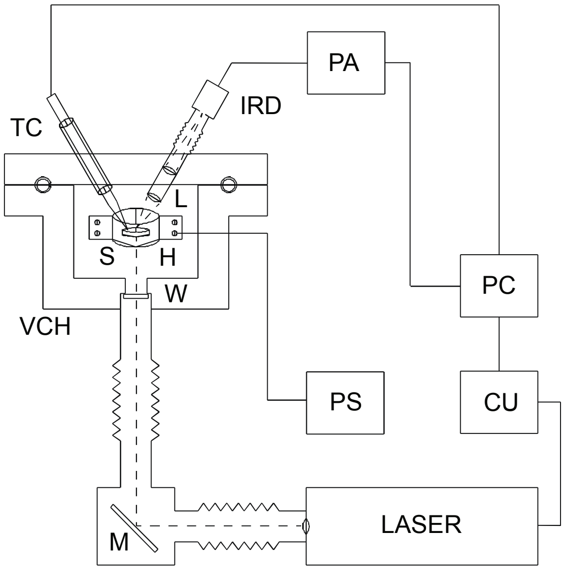

Fig. 1 Schematic view of the experimental apparatus
TC - thermocouple, IRD - infrared detector, PA - preamplifier, L - lens, S - sample, H - heater, W - window, VCH - vacuum chamber, M - mirror, PS - power source, PC - personal computer, CU - controller unit)

The software package is built in Borland's C++ and runs on a PC under MS DOS. The program performs data acquisition and device control, data storage and retrieval, processing of the temperature vs. time recordings, printing and saving of the results. The software consists of several independent functional sections that include data storage in the ASCII form (SAVE) and data retrieval (LOAD), smoothing and filtering of the temperature vs. time curve using the fast Fourier transformation (SMOOTH, FFT FILTER), data reduction - calculation of the thermal diffusivity (DATA REDUCTION), presentation (PRINT, PRINT-SCREEN, PLOT RESULTS) and saving (SAVE RESULTS) of results, and utilities for setting and changing program parameters of built in procedures for data acquisition and data reduction, and sample information (SETTINGS). The correction section (CORRECTION) allows various user-friendly interactions with the data such as zoom, manual setting of the base-line (temperature level before the laser pulse application) and the maximal temperature rise T_{max} and others. The device control section (RUN) allows half automated data acquisition. When selected, it starts to observe the sample temperature behavior. Once the thermal equilibrium is achieved the measurement sequence starts with the recording of the rear face temperature rise vs. time data. After 10% of the chosen duration of the experiment has elapsed the defined sequence of the laser pulses is applied (the exact time is monitored using a fast photodiode). Then the data are ready for processing. The software calculates the thermal diffusivity by various data reduction methods based on the ideal analytical model or models that take account of heat losses and finite pulse time duration. The software gives the possibility of studying two- and three-layered structures (estimation of the thermal diffusivity or the thermal contact resistance). The software allows work in a cycle of a previously defined sequence of functional operations (CYCLE).

Experimental results

The accuracy of the experimental apparatus has been checked measuring several standard or reference materials. Fig. 2 presents results of the thermal diffusivity estimation of austenitic steel X10NiCrMoTiB1515 (Nr.1.4970) - a material that had been intensively investigated by German Thermopysical Society [12]. Comparison shows that deviation between the experimental and recommended data are within +/- 1.3 %. The standard accuracy of the thermal diffusivity measurement is in general better than 5% (3% between 20 and 1500 °C).

Fig. 3 presents some of the typical experimental curves measured on the steel in a vacuum at 300 °C under different measuring conditions. Results of the thermal diffusivity estimation are summarized in Tab. 1. A relatively good agreement of the thermal diffusivity values obtained utilizing the standard 'one-pulse' flash method (No. 1) and the flash method with repeated pulses can be seen. Any significant difference between the dispersion of the thermal diffusivity values estimated from 'several pulses' recordings and the usual dispersion of the standard 'one pulse approach' of the used experimental apparatus haven't been observed.

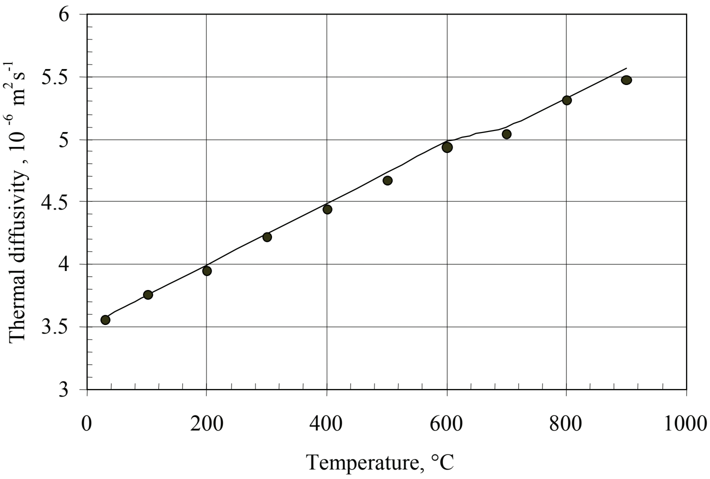

Fig. 2 Thermal diffusivity of austenitic steel X10NiCrMoTiB1515
measured values - circle, literature data -solid line

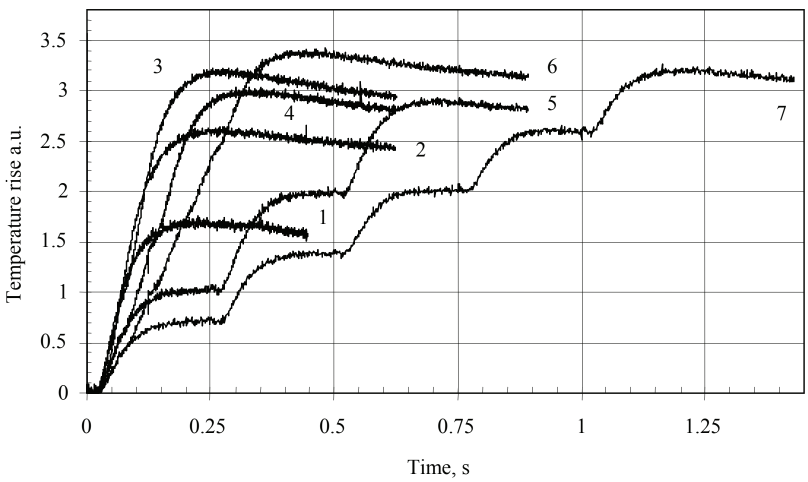

Fig. 3 Experimental temperature rise vs. time curves
material - CrNi steel, curves labels correspond to the notation in the Table 1

Tab. 1

Results of themal diffusivity estimation of austenitic CrNi steel. *f* is the pulse frequency, t_p the period and *p*+1 the number of pulses, *a* and a_R are the measured and reference thermal diffusivities ($a_R = 4.24 \cdot 10^{-6}$ m^{2}s^{-1})

Case	*f*, Hz	t_p, s	*p*+1	*a*, 10^{-6}m^{2}s^{-1}	$(a - a_R)/a_R$, %
1			1	4.25	0.24
2	16	0.062	2	4.33	2.12
3	64	0.016	3	4.34	2.36
4	16	0.062	3	4.26	0.47
5	4	0.250	3	4.24	0
6	16	0.062	5	4.43	4.48
7	4	0.250	5	4.2	-0.94

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Устройство для измерения коэффициента температуропроводности с использованием метода лазерной вспышки с повторными импульсами

Л. Возар[1], В. Гогенауер[2]

*Кафедра физики, факультет естественных наук,
Университет им. Константина Философа (1);
Кафедра исследования материалов,
Австрийский исследовательский центр Сайберсдорфа (2)*

Ключевые слова и фразы: метод лазерной вспышки; повторные импульсы; температуропроводность.

Аннотация: Рассматривается измерение коэффициента температуропроводности при использовании метода лазерной вспышки с повторными импульсами. Метод вспышки с повторными импульсами был предложен как усовершенствование классического метода "одноимпульсной" лазерной вспышки с целью преодоления специфических экспериментальных трудностей, связанных с измерением некоторых типов изоляционных материалов, чувствительных к изменению температуры, и крупнозернистых однородных материалов. В отличие от других подходов, основанных на сокращении интенсивности энергии и одновременном увеличении времени воздействия (метод вспышки с продленным временем воздействия импульса), или замене ступенчатого (непрерывного) нагревания на импульсную иррадиацию (шаговый метод нагревания), соответственно, метод вспышки с повторными импульсами основан на применении энергии импульса, состоящей из нескольких последовательных импульсов, периодически подводимых к фронтальной стороне образца. Коэффициент температуропроводности рассчитывается из итогового повышения температуры у задней стороны образца способом, аналогичным стандартному лазерному методу вспышки.

В статье представлено математическое обоснование метода вспышки с повторными мгновенными импульсами. Рассматривается простая адиабатическая математическая модель и более реалистическая неидеальная теория, которая учитывает утечки тепла из образца. Детально описана созданная экспериментальная установка, которая регулярно используется для измерения коэффициента температуропроводности в австрийских исследовательских центрах. Представлены результаты измерений коэффициента температуропроводности CrNi аустенитной стали.

Errichtung fьr die Messung des Koeffizientes der Temperaturleitfдhigkeit bei der Benutzung der Methode des Laserblitzes mit den wiederholenden Impulsen

Zusammenfassung: Es wird die Messung des Koeffizientes der Temperaturleitfähigkeit bei der Benutzung der Methode des Laserblitzes mit den wiederholenden Impulsen betrachtet. Die Methode des Laserblitzes wurde als Vervollkommnung der Klassikmethode des «einimpulsischen» Laserblitzes mit Zweck der Überwindung der spezifischen experimentellen mit der Messung der einigen Typen verbundenen Isolationsstoffe vorgeschlagen, die zur Temperaturveränderung und zu den grobkörnigen gleichartigen Stoffen empfindlich sind. Zum Unterschied von den anderen Standpunkten, die sich auf der Reduktion der Energieintensivität und gleichzeitiger Steigerung der Zeiteinwirkung (die Methode des Blitzes mit der verlängerten Zeit der Impulseinwirkung) oder auf dem Ersetzen der ununterbrochenen Heizung mit der

Impulsirradiation (schrittliche Heizungsmethode) gründen, ist die Methode des Blitzes mit den wiedercholenden Impulsen auf der Benutzung der Impulsenergie gegründet. Diese Energie besteht aus einigen aufeinanderfolgenden zur Frontalseite des Musters periodisch zuführenden Impulsen. Der Koeffizient der Temperaturleitfähigkeit wird aus der gesamten Temperaturerhöhung der Rückseite des Musters berechnet. Dieses Verfahren ist der standarten Lasermethode des Blitzes ähnlich.

In dem Artikel ist die mathematische Begründung der Blitzmethode mit den wiederholendenden augenblicklichen Impulsen dargestellt. Es wird das einfache adiabatische Modell und eine realistischere unideale den Wärmeverlust aus dem Muster berücksichtigte Theorie betrachtet. Es ist die geschaffene für die Messung des Koeffizientes der Temperaturleitfähigkeit in Österreichischer Forschungszetren regelmäßig benutzende Experimentellanlage in Daitals beschrieben. Es sind die Ergebnisse der Messungen des Koeffizientes der Temperaturleitfähigkeit CrNi des austenitischen Stahls dargestellt.

Dispositif pour la mesure du coefficient du transfert de température par l'utilisation de la méthode de l'éclat du laser avec les impulsions répétées.

Résumé: On examine le changement du coefficient du transfert de température par l'utilisation de la méthode de l'éclat du laser avec les impulsions répétées. La méthode de l'éclat du laser a été proposée comme le perfectionnement de la méthode classique de l'éclat du laser «à une impulsion» afin de surmonter des difficultés spécifiques expérimentales qui sont liées aux mesures de certains types de matériaux isolés sensibles aux changement de la température et des matériaux homogènes granulés. A la différence d'autres approches qui sont fondées sur la réduction de l'intensité de l'énergie et du temps de l'interaction (méthode de l'éclat avec le temps de l'action d'impulsion prolongé) ou bien sur le changement du chauffage avec un palier (continu) sur l'irradiation d'impulsion (méthode du chauffage pas à pas), la méthode de l'éclat avec les impulsions répétées est fondée sur l'application de l'energie de l'impulsion qui se compose de quelques impulsions sucsessives qui sont périodiquement emmenées au côté frontal de l'échantillon. Le coefficient du transfert de température est calculé à partir de l'augmentation finale du côté d'arrière de l'échantillon par la méthode analogue à la méthode de l'éclat du laser standartisée.

Dans l'article on a présenté le fondement mathématique de la méthode de l'éclat du laser avec les impulsions répétées. On examine le simple modèle mathématique adiabatique et la théorie non-idéale plus réaliste qui tient compte des pertes de la chaleur à partir de l'échantillon. On a décrit en détails l'installation expérimentale qui est utilisée réguliérement pour la mesure du coefficient du transfert de température dans les centres d'études de l'Australie. On a cité les résultats des mesures du coefficient du transfert de température de l'acier austénite CrNi.
