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Thermochimica Acta 468 (2008) 6–9

thermochimica acta

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Review

Determination of apparent thickness of graphite coating in flash method

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Received 27 July 2007; received in revised form 12 November 2007; accepted 12 November 2007
Available online 24 November 2007

Abstract

When a sample for the measurement of thermal diffusivity is coated with graphite to enhance the absorbance of flash energy in the flash method, thermal resistance of the sample is increased due to the graphite layer itself and the contact resistance between the graphite layer and the sample surface. Such increased thermal resistance is considered as that of the graphite layer whose thickness is the apparent thickness ℓ_{apgr} of the graphite coating whose actual thickness is ℓ_{gr}. Using an equation for the thermal diffusivity proposed by Parker et al. [W.J. Parker, R.J. Jenkins, C.P. Butler, G.L. Abbott, J. Appl. Phys. 32 (1961) 1679], the present study found that the resistance factor defined as $Gr_{\mathrm{eff}} = \ell_{\mathrm{apgr}}/\ell_{\mathrm{gr}}$ is a unique function of the half time, irrespective of the materials, which is given by a correlation equation, $Gr_{\mathrm{eff}} = 4.2454(t_{\mathrm{s+gr}})_{1/2}^{-0.465}$. Therefore, an accurate measurement of the half time enables us to find the thermal diffusivity of the graphite-coated sample material with an uncertainty level of about 0.6. It was found that the present method produces the thermal diffusivity of standard materials within 0.6% difference with respect to the standard data except very high thermal diffusivity materials such as copper and alumina. The advantage of this method lies in avoiding the lengthy mathematical equations (e.g. a three-layer analysis) needed to correct the errors caused by the graphite coating.

Keywords: Apparent graphite thickness; Graphite coating; Thermal diffusivity; Half time; Flash method

Contents

1. Introduction

The thermal diffusivity is one of the most fundamental properties which are extensively used by design engineers and scientists. One of the most popular methods for measuring the thermal diffusivity is the flash method. This method consists of heating the front surface of a sample with a high-intensity short-duration flash pulse and measuring the temperature evolution on the rear surface by using an infrared detector.

Graphite coating on the sample surface is a process that is fundamental in thermal diffusivity measurement by the flash method. It increases both the absorbance of flash energy on the front surface and the intensity of the infrared light which is emitted from the rear surface. Moreover, the graphite coating plays an important role in decreasing the surface roughness [1,2]. However, the graphite coating causes an increase in the thermal resistance of the graphite–sample system and results in an increase of the half time and a decrease of the thermal diffusivity which is the source of error in the thermal diffusivity measurement. This error is more pronounced in the cases of thin samples or highly conductive materials. Hence for reliable measurements the sample thickness of at least 3 mm has been

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doi:10.1016/j.tca.2007.11.012

recommended for high thermal diffusivity material such as aluminum nitride as reported by Hasselman and Merkel [3]. Similar values have been recommended by various equipment makers to minimize the error due to the graphite coating. However, in the practical point of view it is almost impossible to adhere to the recommended thickness for different kinds of material.

Hence a number of studies have been made to resolve the error caused by the graphite coating on sample surface. Various theories have been presented to predict the thermal resistance [4–7] of the graphite coating, yet none is in good agreement with the experimental data. As a result, the thermal resistance of the graphite coating is one of the most complicated problems in thermal diffusivity measurement. Therefore, the principal difficulty of the flash method lies in the estimation of the thermal resistance of thin free-standing coatings. The objective of the present study is to propose a simple method to take into account of the increased thermal resistance due to the graphite coating in the accurate measurement of the thermal diffusivity by the flash method.

2. Measurement principle

In flash method as shown in Fig. 1, assuming one-dimensional heat transfer with adiabatic boundary condition, the temperature rise at the rear surface of the sample at time t can be written as [8]

$$\frac{\Delta T}{\Delta T_{\max}} = 1 + 2\left[\sum_{n=1}^{\infty}(-1)^n \exp\left(\frac{-n^2 2\pi^2 \alpha t}{\ell^2}\right)\right] \tag{1}$$

where α and ℓ are the thermal diffusivity and sample thickness, respectively. ΔT is the temperature rise at the rear surface, $\Delta T_{\max}$ is the maximum temperature rise at the rear surface, and t is time after pulse heating.

Designating the time when the temperature rise reaches $\Delta T/\Delta T_{\max} = 1/2$ as $t_{1/2}$, the thermal diffusivity can be obtained from Eq. (1) as follows [8]

$$\alpha = \frac{0.138785\ell^2}{t_{1/2}} \tag{2}$$

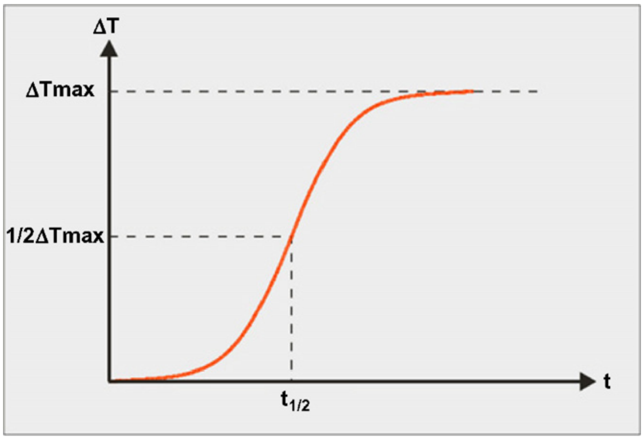

Fig. 1. Theoretical curve of temperature rise at the rear surface of the sample.

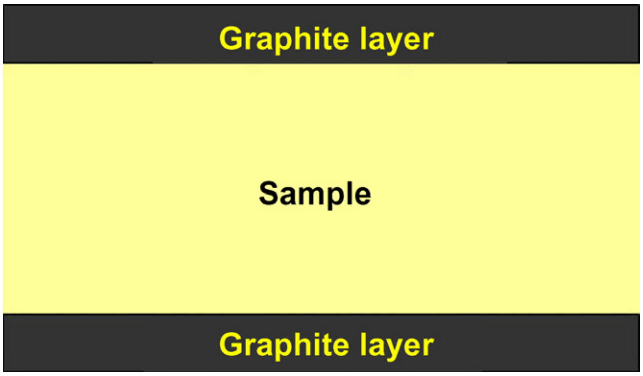

Fig. 2. Sketch of the sample coated by two graphite layers.

When the sample is coated with graphite on both sides as sketched in Fig. 2, it increases the half time as shown in Fig. 3, and thus, the measured thermal diffusivity is decreased as dictated by Eq. (2).

Fig. 3 shows the time lag and change in $t_{1/2}$ due to the thermal resistance for a black sample with and without graphite coating on both surfaces.

To nullify the effect of thermal resistance of the graphite coating, the thickness of graphite coating must be taken into account in the thickness parameter ℓ.

Consequently, α must be written as

$$\alpha = \frac{0.138785(\ell_{\mathrm{s}} + \ell_{\mathrm{apgr}})^2}{(t_{\mathrm{s+gr}})_{1/2}} \tag{3}$$

Here ℓ_{s} is the sample thickness and ℓ_{apgr} is an apparent graphite thickness of the graphite coatings on both surfaces. The half time of coated graphite sample $(t_{\mathrm{s+gr}})_{1/2}$ can be measured by the flash method.

The graphite thickness (ℓ_{gr}) can be calculated as follows

$$\ell_{\mathrm{gr}} = 0.5093\frac{(m_{\mathrm{s+gr}} - m_{\mathrm{s}})}{\pi/4(1.27)^2} \tag{4}$$

Here m_{s} is the sample mass and $m_{\mathrm{s+gr}}$ is the mass of graphite coated sample. Sample diameter is 1.27 cm and the density of the graphite is 0.5093 g/cm^3. Now, a resistance factor Gr_{eff} is

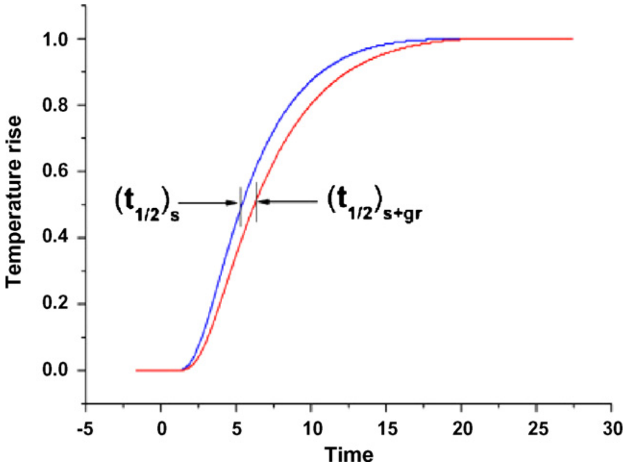

Fig. 3. Time lag due to thermal resistance of graphite coating.

introduced as the ratio between the apparent graphite thickness (ℓ_{apgr}) and the graphite thickness (ℓ_{gr}), i.e.,

$$Gr_{\mathrm{eff}} = \frac{\ell_{\mathrm{apgr}}}{\ell_{\mathrm{gr}}} \tag{5}$$

The information about Gr_{eff} enables us to find the apparent graphite thickness. A method to find Gr_{eff} is explained as follows.

3. Experiments and results

Thermal diffusivity experiments were performed for known samples. Both sides of the samples were coated by spraying a thin layer of graphite. Graphite was purchased from GRAPHIT 33(Kontakt Chemie, Germany). The standard samples used were polycrystalline alumina, pyroceram 9606, pyrex 7790, copper, pure iron, inconel 600, stainless steel 310 supplied by NETZSCH and unknown samples (A and B) of black color and smooth (low roughness). The circular disc-shaped samples of 12.7 mm in diameter were prepared with commercially spray colloidal graphite paint. Mass of the sample was weighed by a microbalance before and after graphite coating and it was used to estimate the thickness of the graphite coating. The experiments have been performed with (LFA 447) at room temperature. A Xenon flash lamp was used to produce the heat pulse on the front surface of the sample. The length of the heat pulse can be varied from 0.1 to 0.5 ms. The NETZSCH Nanoflash adopts an improved Cape-Lehman model [9] to eliminate possible errors caused by the transient heat loss and the finite laser pulse. Details of the experimental method are described by Kim and Kim [10]. The disc sample may be heated non-uniformly because the beam intensity is not uniform across the beam. However, since the heat is transferred almost one-dimensionally through the rather thin disc in less than a half second, the measurement error due to the non-uniform heating seems to be minimal.

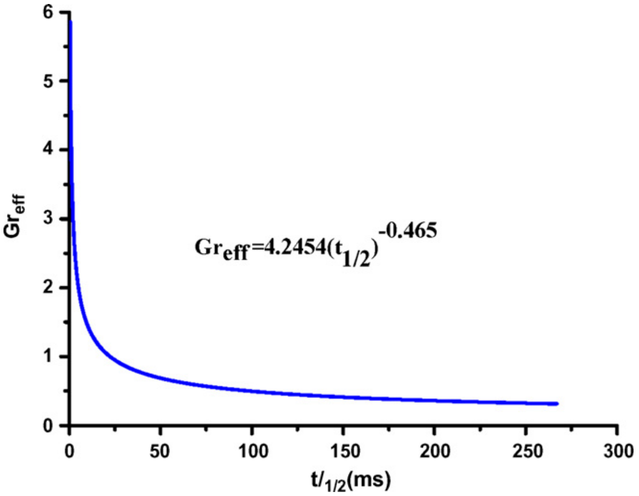

Fig. 4. Correlation of graphite effect (Gr_{eff}) with half time ($t_{1/2}$).

First, the mass (m_{s}), graphite-coated sample mass ($m_{\mathrm{s+gr}}$), and thickness (ℓ_{s}) of the standard samples with known thermal diffusivity (α_{s}) are measured and graphite coating thickness ℓ_{gr} is calculated using Eq. (4). Experiments were repeated for the standard samples to find the apparent graphite thickness ℓ_{apgr}. First the sample thickness ℓ_s is measure, and the half time $(t_{\mathrm{s+gr}})_{1/2}$ is obtained by the experiment and the known standard thermal diffusivity α_{s} is substituted in Eq. (3). This procedure yields the apparent graphite thickness ℓ_{apgr}. Then the resistance factor can be calculated by the defining Eq. (5). The similar experiments were conducted for a set of standard materials and the resistance factors Gr_{eff} were plotted as a function of the half time as shown in Fig. 4. As can be seen Gr_{eff} decreases monotonically as a function of the half time. An empirical correlation function of Gr_{eff} thus found is written as

$$Gr_{\mathrm{eff}} = 4.2454(t_{\mathrm{s+gr}})_{1/2}^{-0.465} \tag{6}$$

Therefore, if the sample thickness ℓ_{s} is measured and the half time is obtained by the flash method, Gr_{eff} can be calculated by Eq. (6). And then the apparent thickness of the graphite coating is obtained by Eq. (5). Finally, the thermal diffusivity is found by Eq. (3).

Table 1 shows the results of thermal diffusivity measurement obtained in this way. It is noteworthy that the resistance factor (Gr_{eff}) is dependant on the thickness as well as the properties of the material. In the case of copper it shows very high value of 3.3 whereas in case of pyrex it is only 0.35. And even for the

Table 1
Comparison of the measured thermal diffusivity (α_{m}) with standard one (α_{s})

Material	m_{gr} (mg)	ℓ_{s} (mm)	ℓ_{gr} (mm)	$t_{1/2}$ (ms)	Gr_{eff}	ℓ_{apgr} (mm)	α_{s} (mm^2/s)	α_{m} (mm^2/s)	Difference (%)
Alumina	0.0010	0.996	0.016	13.76	1.2	0.019	10.23	10.312	−0.80
Pyroceram 9606	0.00146	0.99	0.023	73.12	0.58	0.013	1.926	1.917	0.47
Pyrex	0.00166	0.986	0.026	208.5	0.35	0.009	0.65	0.648	0.31
PuIr1	0.00119	0.984	0.018	6.615	1.76	0.032	21.6	21.185	1.92
Inconel 600	0.00177	1.007	0.027	44.17	0.73	0.02	3.458	3.436	0.64
Copper	0.00172	1.004	0.027	1.681	3.33	0.089	117.2	114.726	2.11
Copper	0.00145	1.998	0.022	5.105	1.99	0.044	117.2	116.597	0.51
Alumina	0.0024	1.985	0.037	52.38	0.67	0.025	10.23	10.198	0.31
Pyroceram 9606	0.00187	1.989	0.029	267.8	0.32	0.009	1.926	1.918	0.42
Stainless Steel 310	0.00157	1.968	0.024	154.7	0.41	0.01	3.352	3.36	0.24
Inconel 600	0.0015	1.995	0.023	159	0.40	0.009	3.458	3.472	0.40
Sample A	0.00163	0.45	0.025	6.282	1.81	0.045	4.986	4.955	0.62
Sample B	0.00166	0.617	0.026	3.856	2.27	0.059	16.838	16.987	0.88

same copper, Gr_{eff} largely depends on the sample thickness. An element uncertainty analysis based on Eq. (3) with uncertainty in ℓ_{apgr} of 0.2% and that in the half time of 0.5% implies that the uncertainty of the measurement of the thermal diffusivity is about 0.6%.

It is seen that the difference is within this range except those of high thermal diffusivity such as alumina, Purl1 and copper. This implies that the standard thermal diffusivities for materials with high thermal diffusivity are less reliable than those of low thermal diffusivity materials.

4. Conclusions

We have developed a simple method to find the apparent graphite thickness of the graphite-coated sample for an accurate measurement of thermal diffusivity. An empirical correlation function for the resistance factor that is the ratio of the apparent graphite thickness and the graphite coating thickness was obtained by measuring the half time of a total of 11 materials with known thermal diffusivity. A couple of black samples were also used to accumulate the data for the resistance factor $Gr_{\text{eff.}}$ It was shown that the empirical resistance factor thus obtained in the present study enables us to find the thermal diffusivity of graphite-coated sample at a very satisfactory level of measurement accuracy with an uncertainty of 0.6%.

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