

Effect of testing conditions on the laser flash thermal diffusivity measurements of ceramics

Ana Paula F. Albers^a, Thomaz A.G. Restivo^b,
Luciano Pagano^b, João B. Baldo^{a,*}

^a*Departamento de Engenharia de Materiais, Universidade Federal de São Carlos,
Rodovia Washington Luiz Km 235, São Carlos, CEP 13.565-905, Brazil*

^b*Laboratório de Materiais, Centro Tecnológico da Marinha em São Paulo,
Rodovia Sorocaba-Iperó Km 12.5, Iperó, CEP 18.560-000, Brazil*

Received 28 September 2000; accepted 12 December 2000

Abstract

Several experimental techniques either under steady state or transient heat transfer conditions, have been developed to evaluate thermal conductivity and thermal diffusivity of materials. However, testing difficulties resulting from specimen size, extended testing time and heat losses, have somewhat impaired the applicability of many of them. In this respect, the use of the laser flash technique for thermal diffusivity measurements, is a very convenient alternative, considering its basic modeling equation is independent of the temperature gradient as well as the heat flow, and in addition the heat losses can be analytically treated. Another important advantage of the technique is its rapid experimental execution. In this work, it is presented as an investigation concerning how the testing conditions such as specimen coating, laser power and pulse duration, base line adoption, heat losses correction methods, and specimen thickness, may affect the thermal diffusivity measurements of some ceramic materials using the laser flash technique. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Thermal diffusivity; Ceramics; Laser flash technique

1. Introduction

The laser flash technique is based on a transient heat flux technique for thermal diffusivity measurement. In this sense, the property is measured at selected temperatures.

The theory involved and the general description of the principles of measurement, were thoroughly treated [1–3]. In the present work, it was investigated how testing conditions such as specimen coating or not,

laser power and pulse duration, base line adoption, heat losses correction methods, and specimen thickness, may affect the thermal diffusivity measurements of some ceramic materials using a Holometrix Thermaflash 2200 laser flash equipment.

Basically in the mentioned apparatus, a short laser heat pulse of high intensity hits the front face of a circular thin specimen, previously equilibrated at the desired temperature. The absorbed heat travels through the specimen thickness causing an increase in its rear face temperature, which is properly measured by a detector along the time.

Following Carslaw and Jaeger [4], the temperature distribution in a thermally insulated specimen of

* Corresponding author. Tel.: +55-16-260-8250;

fax: +55-16-261-5404.

E-mail address: baldo@power.ufscar.br (J.B. Baldo).

thickness l is given by

$$T(x, t) = \frac{1}{l} \int_0^l T(x, 0) dx + \frac{2}{l} \sum_{n=1}^{\infty} \exp\left(\frac{-n^2 \pi^2 a t}{l^2}\right) \times \cos \frac{n \pi x}{l} \int_0^l T(x, 0) \cos \frac{n \pi x}{l} dx \quad (1)$$

where a stands for thermal diffusivity in cm^2/s .

When the heat pulse Q (cal/s cm^2) is instantaneously and homogeneously absorbed at a small depth g , from the front specimen surface ($x = 0$), the temperature distribution in any specific time is given by [1,2]

$$T(x, 0) = \frac{Q}{\rho c_p g} \quad 0 < x < g \quad (2)$$

$$T(x, 0) = 0 \quad g < x < l$$

Considering the above boundary conditions, Eq. (1) may be rewritten as [1,2]

$$T(x, t) = \frac{Q}{\rho c_p l} \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n \cos \frac{n \pi x}{l} \frac{\sin \frac{n \pi g}{l}}{n \pi g / l} \times \exp\left(\frac{-n^2 \pi^2 a t}{l^2}\right) \right] \quad (3)$$

Since the great majority of ceramic solids is opaque to the laser, then g turns out to be a small number, such that the following approximation can be done [1,2]

$$\sin \frac{n \pi g}{l} = \frac{n \pi g}{l}$$

In this manner, the specimen rear face temperature distribution (at $x=l$) can be given by [1,2]

$$T(l, t) = \frac{Q}{\rho c_p l} \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n \exp\left(\frac{-n^2 \pi^2 a t}{l^2}\right) \right] \quad (4)$$

As a consequence the maximum temperature is

$$T_{\max} = \frac{Q}{\rho c_p l} \quad (5)$$

Upon the introduction of two non-dimensional parameters it is possible to write

$$T'(l, t) = \frac{T(l, t)}{T_{\max}} \quad (6)$$

$$t' = \frac{\pi^2 a t}{l^2}$$

Which provides subsidies to rewrite Eq. (4) in the form [1,2]

$$T' = \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n \exp(-n^2 t') \right] \quad (7)$$

The graphical representation of Eq. (7) is shown in Fig. 1. It can be noticed that the abscissa is the thermal diffusivity a . If we assume $T' = 0.5$ in Eq. (7), it results that $t' = 1.37$ and then the thermal diffusivity a can be given by

$$a = \frac{1.37}{\pi^2 t_{1/2}} l^2 \quad (8)$$

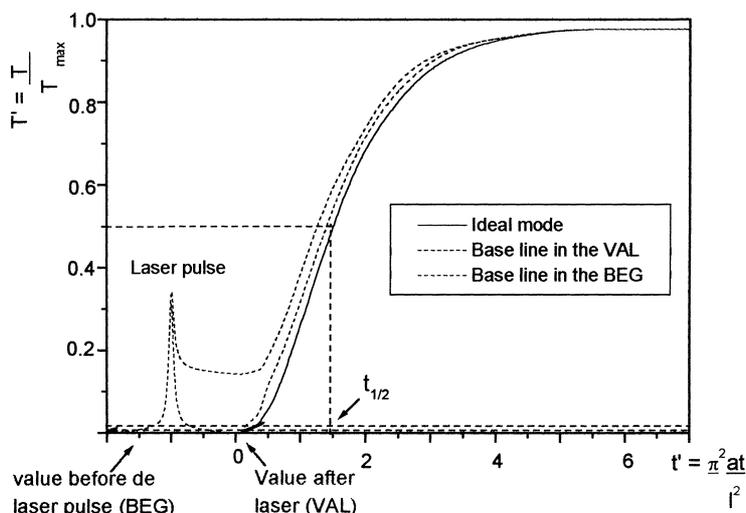


Fig. 1. Temperature distribution at the specimen rear face as a function of time.

where $t_{1/2}$ is the time it takes for the temperature in the specimen rear face to reach half of the maximum value.

Fig. 1 displays the testing measurement “ideal mode” (black line) of typical experimental results [5]. The plot on red is equivalent to the ideal mode, however, using the base line in the value after laser (VAL) mode, meaning that the time counting is based on the minimum value obtained after the laser pulse is shot. This implies an equilibrium temperature at the specie surface before the laser pulse is triggered. When the temperature increase is not obtained in the “ideal mode”, it is proposed a correction method called value before the laser pulse (BEG) mode, implying that the specimen rear face temperature increase starts before the laser pulse ends. In this case, the time counting is also initiated before the laser pulse end. The errors involved in non-ideal mode associated to the ones originated on specimen thickness measurement are considered measurable [5,6].

However, when there are errors associated to deviations of non-ideal mode as a result of: heat gain or losses [7–9], the effect of finite pulse time [10–13], non-uniform heating up of the irradiated surface [14], and the surface energy absorption occurring at a deeper depth than expected, all these are considered non-evaluable errors.

In the present work, aiming at better understanding of the factors that affect thermal diffusivity measurements using the laser flash technique, a series of control parameters related either to the specimen or to the operational conditions, were elected for investigation. Namely they were:

- which type of base line to choose;
- the influence of specimen thickness;
- the type of heat loss correction used (Clark, Taylor, Cowan, Degiovanni);
- laser pulse power and its duration;
- the presence or not (one face or both) of colloidal graphite coating [15–17].

With respect to heat losses, it has to be pointed out that Clark and Taylor [7] use the period before the attainment of the maximum temperature, while Cowan [8], the period of time after the attainment of the maximum temperature. On the other hand, Degiovanni uses the integral of the temperature distribution.

2. Materials and methods

Three different ceramics were investigated. The first was pyroceram 9606, a standard provided by the laser flash equipment producer. The other two were uranium dioxide (UO₂) and aluminum oxide (Al₂O₃) both processed in the authors laboratory.

The aluminum oxide specimens were produced from ALCOA's A-16 calcined alumina, after firing for 2 h at 1500°C, dry pressed cylindrical bodies 0.001–0.002 m thick and 0.012 m diameter under 75 MPa pressure. The degree of densification related to the theoretical density was 91.5%. The uranium dioxide specimens were produced by sintering at 1700°C under hydrogen similar pellets, dry pressed at 270 MPa. The degree of densification obtained in this case was 95.3%.

Thermal diffusivity measurements were done on a Neodymium glass laser flash Termaflash 2200 from Holometrix, whose wavelength was 1.06 μm and maximum power of 15 J. The equipment producer recommends a power of only 10% of the maximum for test temperatures under 400°C, 50% of full power for test temperatures under 600°C and full power for test temperatures above 600°C. The suggestion for the laser pulse duration is $\tau = 0.0004$ s. These last two settings may be altered as wanted.

3. Results and discussion

3.1. Effect of colloidal graphite coating

In Figs. 2–4, it is shown the thermal diffusivity behavior of the three investigated ceramics as a function of the presence or not of colloidal graphite coating, depending also on the manner the coating was applied. That is, in only one face (frontal or rear) or both specimen faces. In this part of the study, it was used the laser power and pulse time recommended by the equipment instructions manual and the Cowan's heat losses correction procedure. The choice of base line (BEG or VAL) was made considering the rear face temperature increase response.

It is well known that the colloidal graphite coating is used to increase the specimen heat absorption keeping this absorption constant, even when the specimen color or its shade is changed. Secondly, the coating

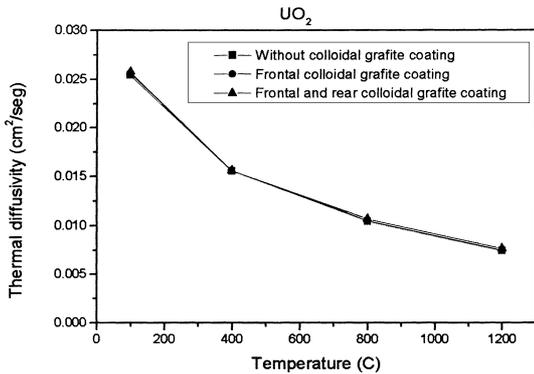


Fig. 2. Thermal diffusivity vs. temperature of the UO₂ ceramic, for different conditions of colloidal graphite coating. Base line choice was the VAL mode.

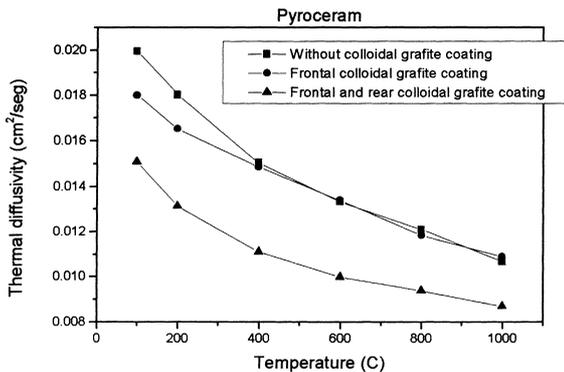


Fig. 3. Thermal diffusivity vs. temperature of the pyroceram 9606 ceramic, for different conditions of colloidal graphite coating. Base line choice was the VAL mode.

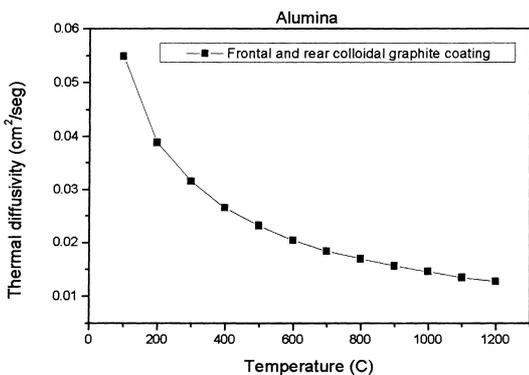


Fig. 4. Thermal diffusivity vs. temperature of the Al₂O₃ ceramic, for different conditions of colloidal graphite coating. Base line choice was the BEG mode.

plays an important role on decreasing surface roughness [15–17]. In certain situations (semi-transparent medium), the front face coating is used to prevent the laser transpassing and the rear face coating to prevent the infrared detector to be affected before the heat pulse travels to the rear face [5].

It was found that for the black ceramic (UO₂) without coating, with coating in either of the two faces or both, there were no changes in the thermal diffusivity values obtained in any testing condition.

On the other hand, the thermal diffusivity versus temperature behavior for specimens of pyroceram 9606 was strongly dependent on the presence or not of colloidal graphite coating. In this sense, it was found that when the coating was only applied to the front face of the specimen, the measured thermal diffusivity values were quite close in magnitude to those obtained for specimens without any coating. On the other hand, the thermal diffusivity values were substantially smaller when the two specimen faces were coated with colloidal graphite. The results indicate that the absence of the coating layer on the rear face of a semi-transparent ceramic permits that radiation hit the sensor when heat is still entirely contained inside the bulk body. This causes a decrease in $t_{1/2}$, and consequently, a virtual increase of the thermal diffusivity. In addition, the magnitude of the thermal diffusivity values obtained in specimens without and with coating on the front face, were much higher than those obtained when the coating was applied to both sides.

In the case of the alumina, it was not possible to make measurements without coating both faces of the specimen. This fact may be due to effects related to the white color of the material and its translucency to the laser radiation.

An analysis of the results indicates when working with non-black materials, it is a must to use the colloidal graphite coating on both faces of the specimen. Moreover, the presence of the coating in only one side may alter the measurements. In this manner, it was decided to use the graphite coating on both faces of the specimens for all the subsequent tests.

3.2. The effect of laser pulse power and duration

Considering that the equipment used permits the change in the laser pulse power and time duration, in Figs. 5 and 6, it is shown that the thermal diffusivity

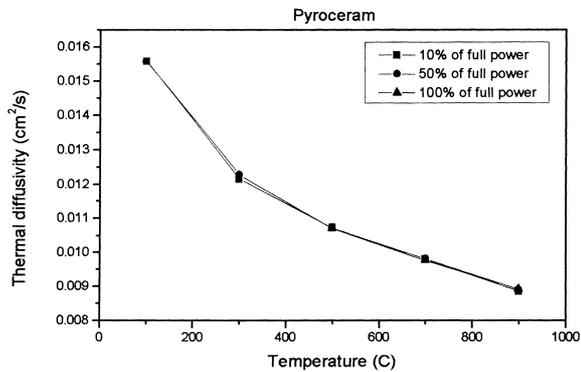


Fig. 5. Thermal diffusivity as a function of temperature for the pyroceram 9606 material under different percentage settings of full laser pulse power. Base line chosen was the VAL mode.

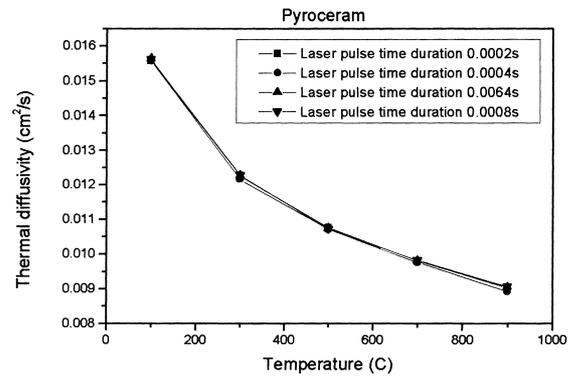


Fig. 7. Thermal diffusivity as a function of temperature for the pyroceram 9606 under laser pulse duration from 2×10^{-4} to 8×10^{-4} s. Base line VAL mode.

behavior as a function of temperature of pyroceram 9606 and alumina specimens, upon changing the laser power. It has to be pointed out in this case that for testing temperatures under 500°C it was not used 100% of the full power. The heat losses correction factor chosen was the one proposed by Cowan, while the pulse duration was the one recommended by the equipment maker.

The power setting suggested by the equipment manual of operations is intended to protect the infrared detector. By changing the laser pulse at levels of 10, 50 and 100% of the total available power, it was not observed any relevant variations in the individual values of thermal diffusivity versus temperature

obtained for each of the three different ceramic materials studied (Figs. 5 and 6).

3.3. The effect of laser pulse time duration

In Figs. 7 and 8, the thermal diffusivity versus temperature behavior for the pyroceram and alumina materials, when the laser pulse time duration was varied from 2×10^{-4} to 8×10^{-4} s is shown. In this case, the laser power used was the one recommended by the equipment maker, and the heat losses correction method was the one proposed by Cowan and the base line chosen was VAL mode for pyroceram and BEG mode for the alumina.

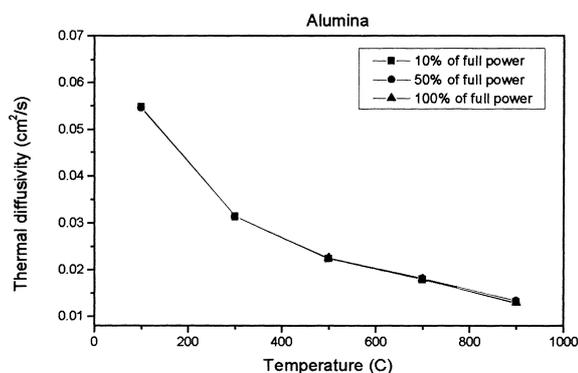


Fig. 6. Thermal diffusivity as a function of temperature for the alumina material under different percentage settings of full laser pulse power. Base line chosen was the BEG mode.

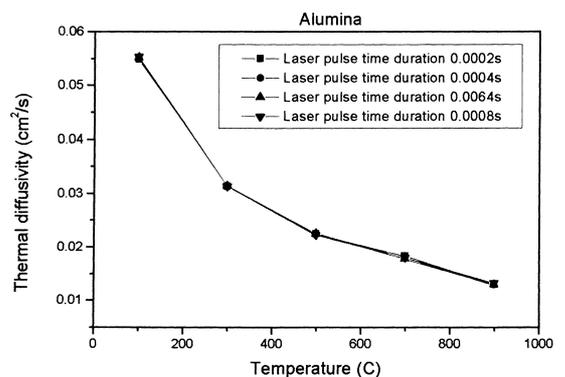


Fig. 8. Thermal diffusivity as a function of temperature for sintered alumina specimen under laser pulse duration from 2×10^{-4} to 8×10^{-4} s. Base line BEG mode.

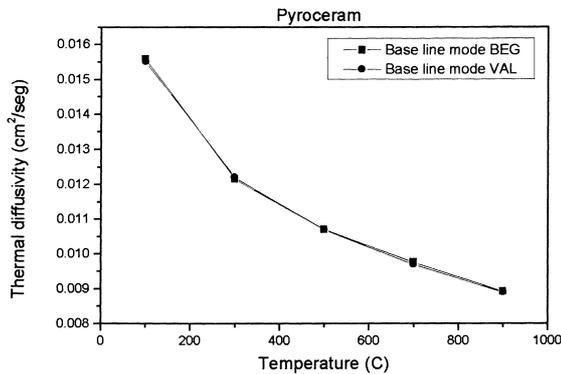


Fig. 9. Thermal diffusivity vs. temperature for the pyroceram 9606 under VAL and BEG base line choices.

As it can be seen, the change in laser pulse time duration did not affect the thermal diffusivity versus temperature behavior on both materials.

3.4. The effect of base line choice

In this work, the effect of the base line adoption for the calculations was also investigated. The laser pulse power and pulse time duration used were the ones suggested by the equipment maker. The heat losses correction method was the one proposed by Cowan. The thermal diffusivity versus temperature behavior for the different base line options are shown in Figs. 9 and 10.

As it was mentioned before, in order to calculate the thermal diffusivity values, it is necessary to use data

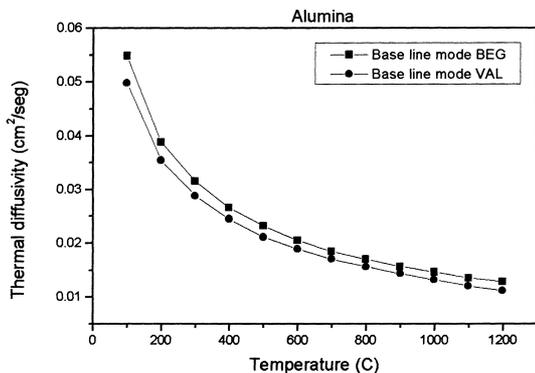


Fig. 10. Thermal diffusivity vs. temperature for the sintered alumina under VAL and BEG base line choices.

relative to the temperature distribution with time on the specimen rear face (Fig. 1). It was found that for the pyroceram 9606 material, the temperature increase starts after the end of laser pulse duration, which is indicative of VAL mode. On the other hand, it was found that in the case of the sintered alumina, the rear face temperature rise occurred before the end of the laser pulse, which is indicative of BEG mode. The thermal diffusivity versus temperature behavior is displayed in Figs. 9 and 10, indicate that in testing situations indicative of BEG mode for the choice of base line, if the measurement is done under VAL mode base line choice, there will be significant differences in the measured values resulting in inaccurate values. However, for materials for which the VAL mode is indicated, if the measurement is done under BEG base line choice apparently there will be no significant differences in the measured values obtained in either base line mode choice. Considering that the temperature increase before the laser pulse ends is a non-ideal measuring condition, the use of the BEG mode base line choice, may be considered a correction method as well. In addition, the results shown in Figs. 8 and 9 lead us to conclude that the alterations on the base line (BEG or VAL mode) is an efficient way of correction for obtained results.

3.5. The effect of heat losses correction methods on the thermal diffusivity measurements

The heat losses involved on the laser flash technique measurements, may be taken into consideration by means of correction methods proposed by Clark and Taylor [7], Cowan [8] and Degiovanni [9]. The effect of correction method on the thermal diffusivity measured values for each of the three ceramics is shown in Figs. 11–13. The laser pulse power and duration were the ones recommended by the equipment maker. It has to be pointed out that the correction method proposed by Degiovanni is not valid when the base line choice is BEG mode-based.

It was found that the Degiovanni proposed correction method maximizes heat losses comparatively to the other correction methods. Similarly, the correction method proposed by Clark and Taylor maximize the heat losses comparatively to Cowan's method. However, considering that the Degiovanni's method cannot be applied under BEG mode, and in the Clark and

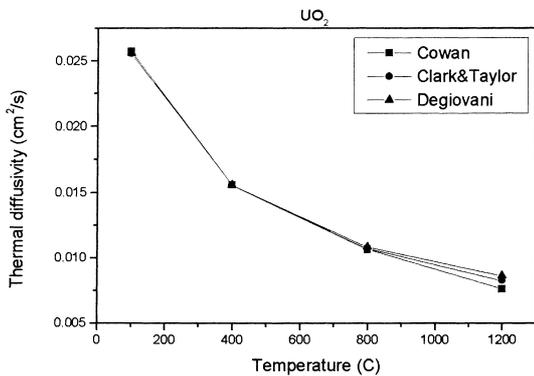


Fig. 11. Thermal diffusivity vs. temperature for the UO₂ sample, using different heat losses correction methods, base line choice was VAL mode.

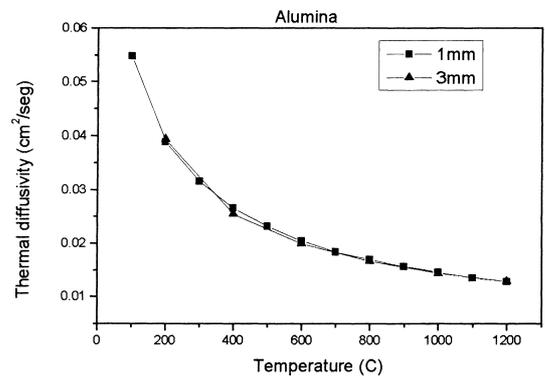


Fig. 14. Thermal diffusivity vs. temperature for sintered alumina specimens having different thicknesses. Correction for heat losses used Cowan's method.

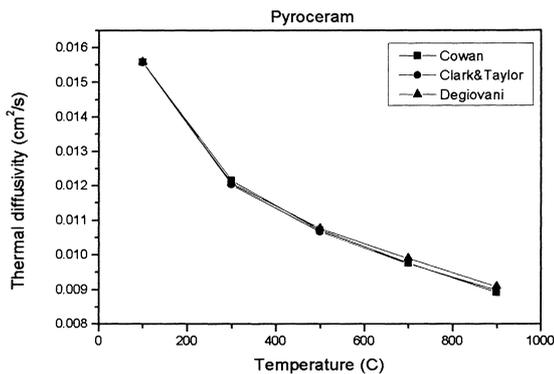


Fig. 12. Thermal diffusivity vs. temperature for the pyroceram 9606 sample, using different heat losses correction methods, base line choice was VAL mode.

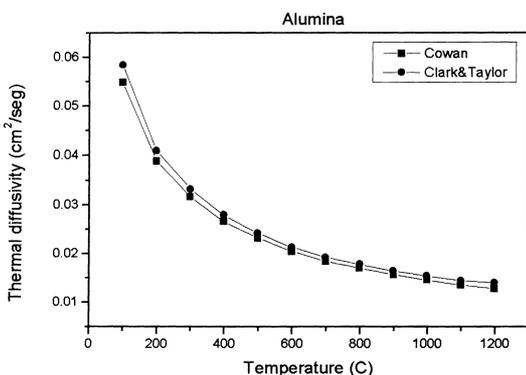


Fig. 13. Thermal diffusivity vs. temperature for the sintered alumina sample using different heat losses correction methods, base line choice was BEG mode.

Taylor method the losses must be limited, it results that Cowan's method is the best choice for most materials and showed to be very efficient for the investigated materials.

3.6. The effect of specimen thickness on thermal diffusivity measurement

In Fig. 14, it is shown the thermal diffusivity versus temperature for the sintered alumina sample, under different specimen thickness condition.

The variation of specimen thickness from 0.001 to 0.003 m on the sintered alumina specimens, did not alter significantly the thermal diffusivity versus temperature results. This fact also indicates that the used heat losses correction (Cowan's) was very efficient.

4. Conclusions

1. It is very important to apply the colloidal graphite coating on both faces of the specimens when thermal diffusivity measurements are done using the laser flash equipment. This procedure guarantees the maximization of heat absorption and topography uniformization of the measuring specimens, at the same time eliminating the sample color effect, resulting in a correct set of conditions for the infrared detector to operate.
2. The change in laser pulse power and duration did not promote alterations on the measured thermal

diffusivity, at least for the tested materials. The possibility of using different base line modes (BEG or VAL), when the testing conditions were not ideal, was very effective on the correction of the results.

3. The heat losses correction method proposed by Cowan, minimizes the losses and can be used in very abranging testing conditions.
4. It was not found any significant effect on the thermal diffusivity measurements, when the specimen thickness was varied from 1 to 3 mm.

Acknowledgements

The authors would like to thank the São Paulo State Research Founding Agency (FAPESP) for the support under contract no. 96/09830-9. To the Brazilian Navy and the Brazilian Nuclear Commission.

References

- [1] W.J. Parker, R.J. Jenkins, C.P. Butler, G.L. Abbott, Flash method of determining thermal diffusivity, heat capacity and thermal conductivity, *J. Appl. Phys.* 32 (9) (1961) 1679–1684.
- [2] F. Righini, A. Cezairliyan, Pulse method of thermal diffusivity measurements (a review), *High Temperature–High Pressure* 5 (1973) 481–501.
- [3] R. Taylor, Construction of apparatus for heat pulse thermal diffusivity measurements from 300 to 3000 K, *J. Phys. Sci. Instrum.* 13 (1980) 1193–1198.
- [4] H.S. Carslaw, J.C. Jaeger, *Conduction of Heat in Solids*, 2nd Edition, Oxford Science Publications, New York, 1959.
- [5] K.D. Maglic, A. Cezairliyan, V.E. Peletsky (Eds.), *Compendium of Thermophysical Property Measurement Methods*, Volume 2, Plenum Press, New York, 1992.
- [6] D.M. Liu, Effect of specimen thickness on the thermal diffusivity of (Sr, K)Zr₄(PO₄)₆ ceramic via a laser flash technique, *J. Mater. Sci.* 31 (1996) 86–89.
- [7] L.M. Clark III, R.E. Taylor, Radiation loss in the flash method for thermal diffusivity, *J. Appl. Phys.* 46 (2) (1975) 714–719.
- [8] R.D. Cowan, Pulse method of measuring thermal diffusivity at high temperatures, *J. Appl. Phys.* 34 (4) (1963) 926–927.
- [9] Une Nouvelle Technique d'Identification de la Diffusivite Thermique pour la Methode Flash, Degiovani, *Revue Phys. Appl.* 21 (1986) 229–237.
- [10] J.A. Cape, G.W. Lehman, Temperature and finite pulse-time effects in the flash method for measuring thermal diffusivity, *J. Appl. Phys.* 34 (7) (1963) 1909–1913.
- [11] R.C. Heckman, Finite pulse-time and heat-loss effects in pulse thermal diffusivity measurements, *J. Appl. Phys.* 44 (4) (1973) 1455–1460.
- [12] K.B. Larson, K. Koyama, Correction for finite-pulse-time effects in very thin samples using the flash method of measuring thermal diffusivity, *J. Appl. Phys.* 38 (2) (1967) 465–474.
- [13] R.E. Taylor, J.A. Cape, Finite pulse-time effects in the flash diffusivity technique, *Appl. Phys. Lett.* 5 (10) (1964) 212–213.
- [14] J.A. McKay, J.T. Schriempf, Corrections for non-uniform surface-heating errors in flash method thermal diffusivity measurements, *J. Appl. Phys.* 47 (4) 1976 1668–1671.
- [15] D.P.H. Hasselman, Temperature dependence of the thermal diffusivity/conductivity of aluminum nitride, *J. Am. Ceram. Soc.* 73 (8) (1990) 2511–2514.
- [16] D.P.H. Hasselman, Thermal diffusivity and conductivity of dense polycrystalline ZrO₂ ceramics: a survey, *Am. Ceram. Soc., Bull.* 66 (5) (1987) 799–806.
- [17] R.W. Rice, R.P. Ingel, G.E. Youngblood, Thermal diffusivity of partially and fully stabilized (yttria) zirconia single crystals, *J. Am. Ceram. Soc.* 71 (4) (1988) 255–260.