THERMAL EFFUSIVITY ESTIMATION OF POLYMERS IN TIME DOMAIN

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Abstract: *The knowledge about thermophysical properties of materials is necessary to make their correct application. A property which is increasing its importance in heat conduction problems is the thermal effusivity. This property indicates the quantity of thermal energy that a material is able to absorb. It depends on thermal diffusivity and thermal conductivity. The estimation of this property can be made by simulating a transient heat transfer model. In this case a one-dimensional semi-infinite thermal model is used. A resistance heater in contact with the sample generates a heat pulse. The variations of temperature and heat flux are measured simultaneously only on the frontal surface of the sample. This experimental procedure presented here allows in situ measurements, where destructive methods can not be used. In this work the thermal effusivity is estimated in time domain through the minimization of the objective function, defined as the square difference between experimental and theoretical temperatures. The golden section technique is used for minimizing this objective function. A sensitivity analysis and a comparison between the semi-infinite and the finite models were also done to define the number of points to be used in the estimation. Measurements were carried out with three different polymers: polymethyl methacrylate (PMMA), polyvinyl chloride (PVC) and polyethylene (PE). In all cases studied the results are in good agreement with literature. In addition an uncertainty analysis is also presented.*

Keywords*: heat conduction, experimental methods, optimization, thermal effusivity.*

1. INTRODUCTION

The knowledge about materials thermophysical properties is even more necessary to make its correct application. Thermal conductivity λ, thermal diffusivity α and thermal effusivity b are three important properties in heat conduction problems. A common example of their application occurs in civil engineering. Many buildings use the thermal insulation to reduce the heat exchange with environment. It reduces the consumption with heating or air conditioning.

Due to the importance of these properties, methods have been developed to determine their values with accuracy and reliability. The methods which involve transient heat transfer stand out because they have an easy implementation, lower costs and lower measurement time. In these methods a signal is generated, usually an impulse, a periodic function or a step function, in the surface of the sample. The variations of temperature and heat flux are used to calculate the property. Blackwell (1954) presented the hot wire technique to measure thermal conductivity. A wire is inserted inside the sample. It is used as a heater and temperature sensor. A heat pulse is generated and the variations of temperature and heat flux with time are measured. A disadvantage is that it is a destructive method because a hole has to be made in the sample. Also it can not be used in metals because of the thermal contact resistance and the lower time measurement. This method presents good results for insulation materials. Santos *et al.* (2004) and Carvalho *et al.* (2006) used it to measure thermal conductivity of polymers. To measure thermal diffusivity Parker *et al.* (1961) developed the flash method. Since it has been used in various works and receiving proposes of improvment, as made by Sheindlin *et al.* (1998) and Min *et al.* (2007). It is also the most used method to measure thermal diffusivity of different kind of materials. For instance, Iguchi *et al.* (2007) measured α of polymers and Blumm *et al.* (2007) measured α of water and ethylene glycol. The method consists in generate a energy pulse with high intensity in a short time interval on the frontal surface of a thin sample. The variations of temperature and heat flux are measured in the rear face. Using a temperature versus time curve the thermal diffusivity can be calculated. The photoacoustic techniques have been widely used to measure thermal effusivity. These techniques can be used in many kind of materials, including liquids, as in the work of Dadarlat *et al.* (2008). As shown by Benedetto and Spagnolo (1988), the technique is based on the measurement of sound waves intensity or phase. These waves are generated by any type of radiation absorbed by the material. A microphone is used to detect them. Generally, the radiaton source is a light beam. To avoid the reflection the surface of the sample must be opaque. Usually a black paint with known thermal properties is used to avoid this. Despite everything these techniques are restricted to laboratorial experiments. Few in situ measurements are made with this techinique. In situ measurements are made with a contact method, like the one presented by Balageas and Jamet (1974). This kind of method consists in a heater in contact with the surface of the sample. The heat flux is measured with a transducer and the temperature with a thermocouple. The data of temperature and heat flux over time are used to estimate thermal effusivity. The solution of this problem can be made in the frequency or time domain. Krapez (2000), Defer *et al.* (2001) and Antczak *et al.* (2007) used the thermal impedance method to estimate thermal effusivity in frequency domain. Coment *et al.* (2002) and Yesilata e Turgut (2007) estimated the property in time domain.

In this work a contact method is used with impulse signals to estimate thermal effusivity of three different polymers: polymethyl methacrylate (PMMA), polyvinyl chloride (PVC) and polyethylene (PE). A semi-infinite and one-dimensional thermal model is used. In this case the medium depends only on its thermal effusivity. In this thermal model the Green's functions are used to solve the heat equation, as made by Fudym (2005) in a tridimensional case. This solution allows calculate the theoretical temperature through numerical methods. In this work the trapezoids rule was used. The solution of the problem is made in time domain. This is made with the minimization of the objective function. This function is defined as the square difference between theroretical and experimental temperatures. The golden section techinique was used to minimizate the function (Vanderplaats, 2005). A sensitivity analysis and a comparison between the semi-infinite and finite models were made to choose the number of points to be used. The results are in good agreement with literature. The higher difference was in PE case, where the difference were 5.13%. This is due to the larger time step and the lower number of experiments.

2. THEORY

2.1. Semi-infinite Thermal Model

The experimental model used is presented in Fig. 1. A semi-infinite sample is subjected to a heat flux on the frontal surface. The heat transfer is one-dimensional with no influence of convection. The temperature measurement is on the same surface.

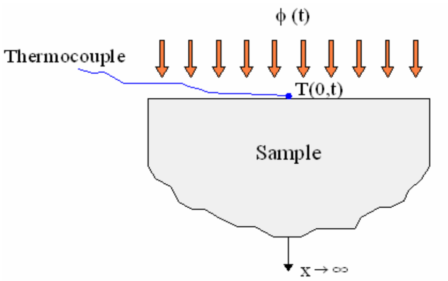


Figure 1. Semi-infinite sample subjected to a heat flux.

In this case the heat diffusion problem can be describe as:

$$\frac{\partial^2 T}{\partial x^2} = \frac{1}{\alpha}\frac{\partial T}{\partial t} \tag{1}$$

The boundary and initial conditions are:

$$\phi(0,t) = -\lambda \frac{\partial T}{\partial x} = \phi_1(t) \tag{2}$$

$$T(x,t)_{x\to\infty} = T_0 \tag{3}$$

$$T(x,0) = T_0 \tag{4}$$

where $\phi_1(t)$ is the heat flux measured on sample surface and T_0 is the initial temperature. To solve this poblem the Green's functions can be used (Beck *et al.*, 1992). The temperature solution on the frontal surface $T(0,t)$ can be written as:

$$T(0,t) = T_0 + \frac{1}{b\sqrt{\pi}}.\int_0^t (t-\tau)^{-1/2}.\phi_1(\tau)d\tau \tag{5}$$

where b is the thermal effusivity, defined as the relation between thermal conductivity λ and thermal diffusivity α square root.

$$b = \frac{\lambda}{\sqrt{\alpha}} \tag{6}$$

Equation (5) is solved numerically by the trapezoid rule method (Ruggiero and Lopes, 1996).

2.2. Thermal Effusivity Estimation

To estimate the thermal effusivity a minimization of an objective function is done. This function is defined as the square difference between experimental, T_e, and theoretical, T, temperatures, defined as:

$$obj = \sum_{i=1}^{n} [T_e(i) - T(i)]^2 \tag{7}$$

where n is the number of points used. To minimize the objective function (Eq. 8) the Golden Section method (Vanderplaats, 2005) is used to determine the thermal effusivity.

2.3. Definition of the Number of Points Used

To choose the number of points n to be used in the estimation procedure, a sensitivity analysis and a comparison between the semi-infinite and the finite models were done. Values of thermal conductivity and diffusivity from literature were used to make these analyses and to calculate the thermal effusivity. The values for PE were obtained from Guimarães *et al.* (1995) and for PVC and PMMA from Lima e Silva *et al.* (2003) (Tab. 1).

Table 1. Literature values used in sensitivity analysis and in comparison between semi-infinite and finite models.

Sample	**λ (W/m.K)**	**α x 10^7 (m²/s)**	**b (W.s$^{1/2}$/m.K)**
PVC	0.156	1.318	429.702
PMMA	0.179	1.14	530.152
PE	0.389	2.33	805.882

2.3.1. Sensitivity Analysis

The sensitivity coefficient, S_b, is defined as the first derivative of the temperature (Eq. 5) with respects to the parameter b.

$$S_b = \frac{dT}{db} = -\frac{1}{b^2\sqrt{\pi}} . \int_0^t (t-\tau)^{-1/2} . \phi_1(\tau) d\tau \tag{8}$$

Figure 2 shows the behavior of S_b for each sample. It can be seen that when S_b becomes constant a little contribution is given to the estimation procedure. For PVC it happens approximately with 1100 points (967.23 s), for PMMA with 900 (900 s) and for PE 160 (1000.496 s).

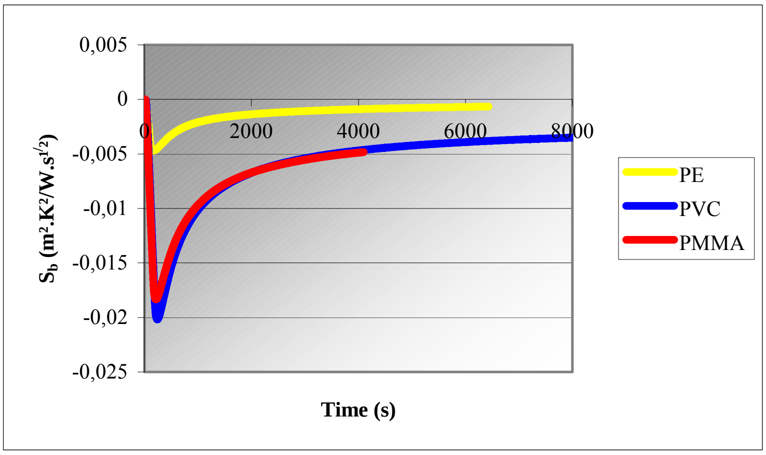


Figure 2. Sensitivity coefficients related to b for PVC.

2.3.2. Comparison Between Semi-infinite and Finite Models

According to Beck et al. (1992), for certain intervals of time the thermal behavior of a finite medium can be considered identical to the semi-infinite. This hypothesis happens as larger the thickness of the medium and smaller the time of heat diffusion. In this work the finite one-dimensional model with heat flux imposed on the frontal surface and insulation on the other surface with thickness L = 50 mm was used (Figure 3).

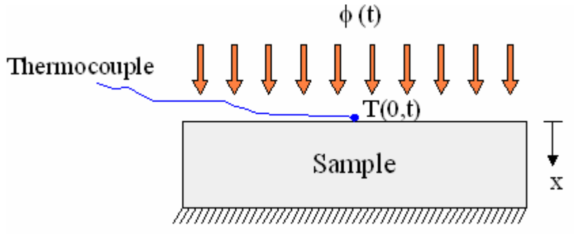
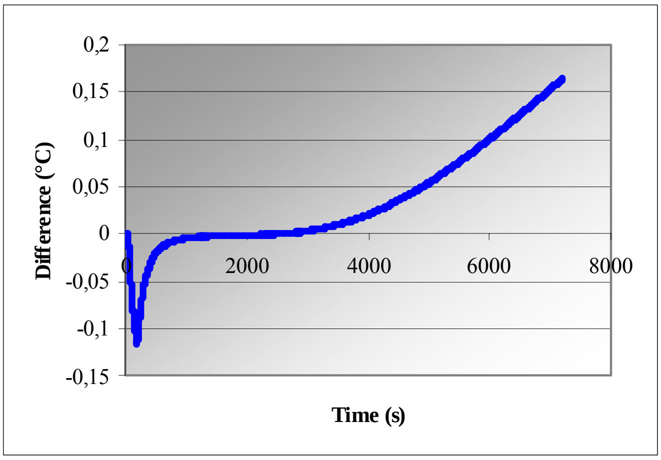
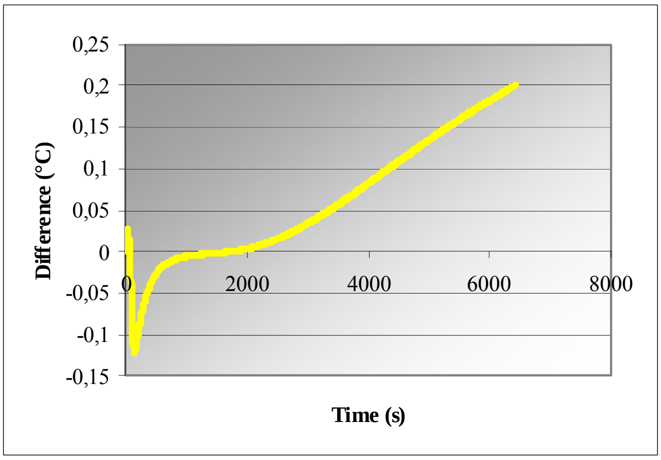


Figure 3. Finite thermal model.

The problem of Fig. 3 is solved numerically approximating the heat diffusion equation in finite differences by explicit method. Literature values were used for α and λ (Tab. 1). The difference between the two models is analised and the moment when the difference begins to increase is determined. At this point the hypothesis of semi-infinite medium is no more valid. Figure 4 shows the difference between finite and semi-infinite models for PVC, PMMA and PE samples. For PVC the increasing in difference begins with approximately 2800 points (2462.04 s), for PE with 270 points (1688.337 s) and for PMMA with 3000 points (3000 s). These time intervals are well above than those determined by the sensitivity analysis. In addition it shows that in the time interval used for the thermal effusivity estimation there is no significant difference between the two models, so the thermal model can be considered semi-infinite.



(a)



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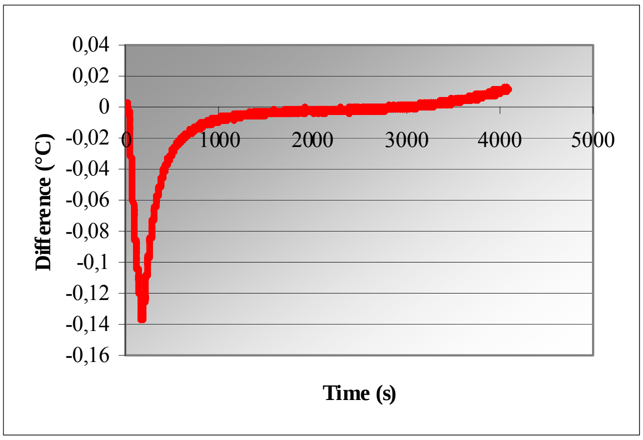


(c)

Figure 4. Difference between semi-infinite and finite models; a) PVC; b) PE; c) PMMA.

2.4. Thermal Contact Resistance Influence

In heat conduction problems involving composite systems, in which the conduction occurs in a material to another, the thermal contact between them has great importance. Usually it is assumed a perfect thermal contact, but this does not occur in practice due to lack of plains, the roughness of the samples and the insertion of sensors such as thermocouples. These spaces are occupied by air, which causes a drop in temperature at the interface of the samples. So the heat transfer occurs through the real contact area and the failures area. To determine the influence of thermal contact resistance in the experiments, a air layer with 0.01 mm of thickness between the transducer and the sample, as shown in Fig. 5, was simulated. Lima e Silva (2000) showed that in center of the transducer and the surround area, the heat transfer can be considered one-dimensional.

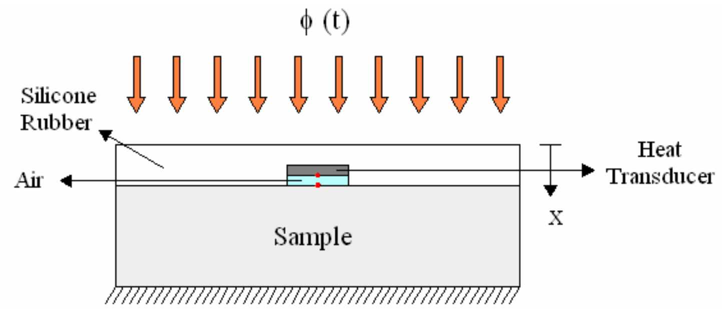
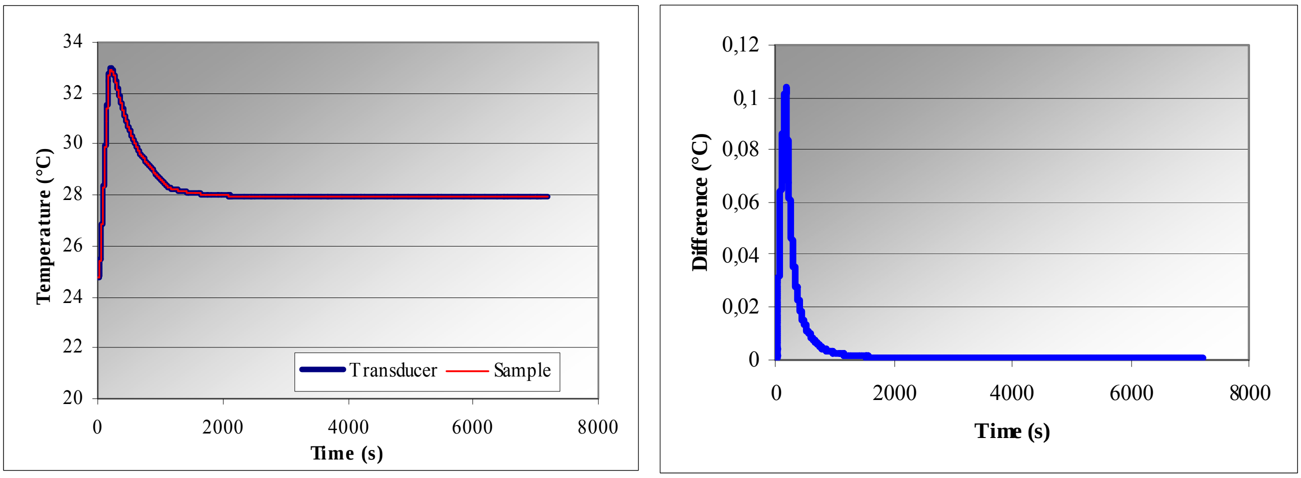


Figure 5. Model adopted to analyze thermal contact resistance influence.

To determine the influence of the air layer, the temperatures on the surface of the transducer and the sample were compared. Figure 6 shows that the difference between these temperatures increased with the rise of heat flux. However for a air layer of only 0.01 mm the largest difference found was only 0.103 °C. It represents an error of only 0.32%. So the effect of thermal contact resistance can be despised on thermal effusivity estimation. In addition, to reduce the contact resistance the heat transducer is placed under pressure with a thermal paste.



(a) **(b)**

Figure 6. a) Temperature evolution on the heat transducer and on the PVC sample; b) Difference between temperatures on the heat transducer and on the sample.

3. EXPERIMENTAL PROCEDURE

Figure 7 shows a diagram of the assembly made to perform the thermal effusivity estimation of three polymers: PMMA, PVC and PE. The samples have dimensions of 305 x 305 x 50 mm. The lateral dimensions are larger than the thickness to ensure a uniform and one-dimensional heat flux on their surfaces. The heater used has the same lateral dimensions and a thickness of 1.4 mm. Its electrical resistance is 22 Ω. To measure the heat flux a transducer with dimensions of 50 x 50 x 0.1 mm was used. The temperature measurement on the contact surface is made through a K type cable thermocouple (30 AWG). Signs of heat flux and temperature are acquired by an acquisition system HP Series 75000 controlled by a computer. To avoid the influence of convection on the samples sides, the experiments were conducted inside an oven.

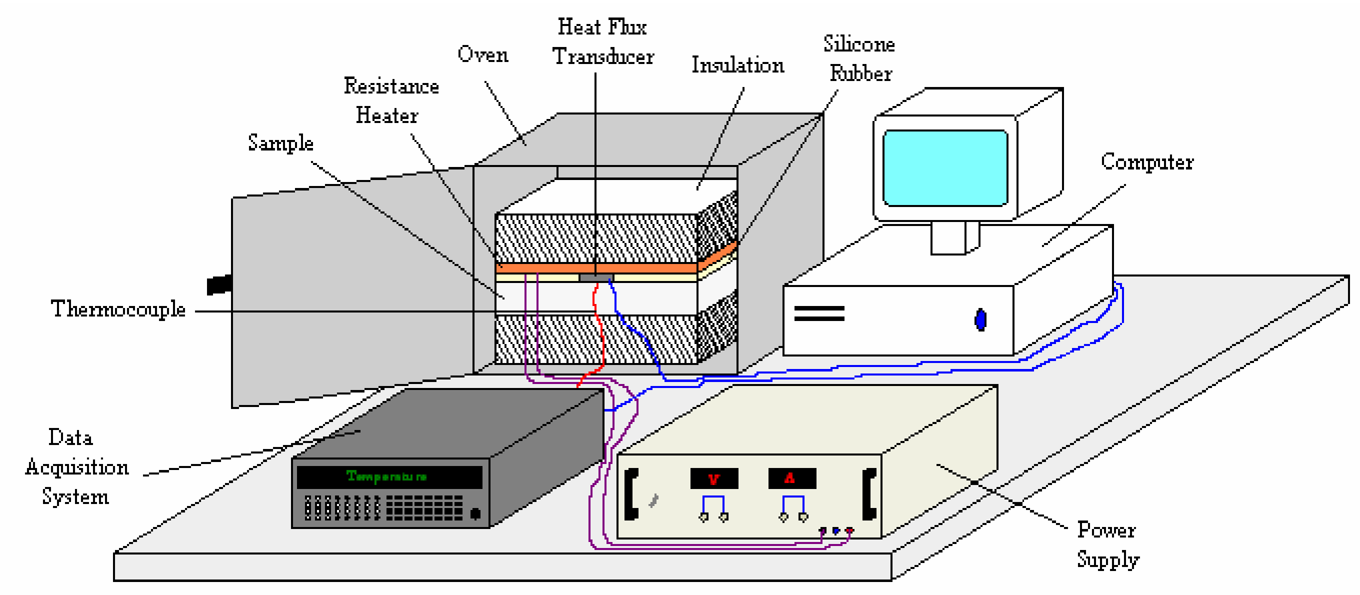


Figure 7. Scheme of the experimental apparatus.

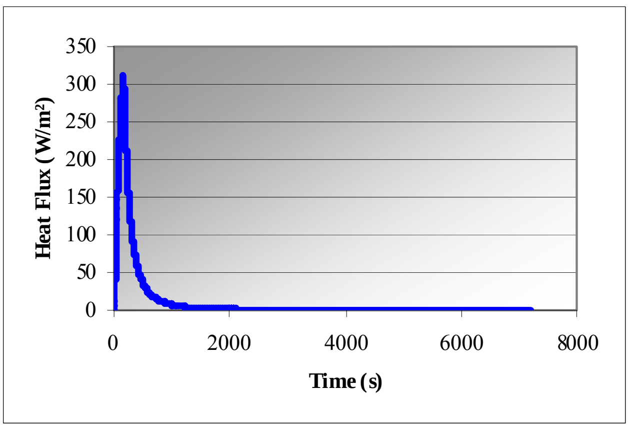
4. RESULTS

Table 2 shows the number of experiments realized, time interval of acquisition and the number of points measured and used on thermal effusivity estimation for PVC, PMMA and PE samples. The difference among the procedures is because the data were obtained from three different works. For PE (Guimarães *et al.*, 1995), PMMA (Lima e Silva *et al.*, 1998) and PVC (Lima e Silva *et al.*, 2003).

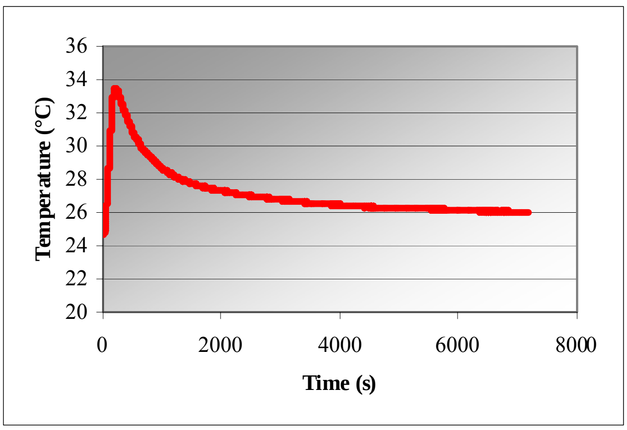
Table 2. Number of experiments, time interval, points measured and used on thermal effusivity estimation.

Sample	Experiments	Time Interval (s)	Total points	Points Used
PVC	51	0.8793	8192	1100
PMMA	42	1.0000	4097	900
PE	21	6.2531	1030	160

In all experiments the heat flux and the temperature evolution has the same behavior, as shown in Fig. 8. An impulse signal of heat flux imposed to the sample surface results in a temperature increase. After this impulse the temperature begins to decrease.



(a)



(b)

Figure 8. a) Heat flux evolution; b) Temperature evolution.

The results of thermal effusivity estimation are shown on Tab. 3. When compared with literature values (Tab. 1) the biggest difference found was for PE sample, with 5.13%. This is due to the larger time interval used and also the lower number of experiments.

Table 3. Thermal effusivity average, standard deviation and difference with literature values.

Sample	b ($W.s^{1/2}/m^2.K$)	Standard Deviation (%)	Difference (%)
PVC	427.84	1.41	0.43
PMMA	507.06	1.53	4.36
PE	847.18	3.06	5.13

5. UNCERTAINTY ANALYSIS

Uncertainty can be described as a portion of the measurement that can not be considered as a true value. Each time a measurement is taken it depends upon a mechanical, electrical or visual point of reference to assingn an appropriate value. These values, no matter how carefully they are obtained, contain some uncertainty (Taylor, 1997). The uncertainties are used to evaluate the precision of the result. That is why it is important to keep low values for them. In this work the procedure to determine the uncertainty in the estimation of *b* is based on linear propagation of ucertainties of the variables, heat flux and temperature, and the numerical calculus of these signals. The hypothesis of linear propagation is used because the objective function is based on the difference between experimental and theoretical temperatures. The uncertainty for experimental temperature U_{Exp} provides from the uncertainties of the data acquisition system U_{Data}, the thermocouple U_{Therm} and the thermal contact resistance U_R.

$$U_{Exp}^2 = U_{Data}^2 + U_{Therm}^2 + U_R^2 \tag{9}$$

The uncertainty of theoretical temperature provides from the uncertainties of the heat flux $U_{H.F.}$ and the error of the numerical calculus with the trapezoid rule U_{Num}.

$$U_{Theo}^2 = U_{H.F.}^2 + U_{Num}^2 \tag{10}$$

The uncertainty of the heat flux provides from the uncertainties of the heat transducer $U_{H.T.}$ and the data acquisition system U_{Data}.

$$U_{H.F.}^2 = U_{Data}^2 + U_{H.T.}^2 \tag{11}$$

Combining Eqs. (10-12), the total uncertainty of objective function can be calculated by:

$$U_{Obj}^2 = U_{Exp}^2 + U_{Theo}^2 = 2.U_{Data}^2 + U_{Therm}^2 + U_{H.T.}^2 + U_R^2 + U_{Num}^2 \tag{12}$$

The value for the uncertainty of the data acquisition system is based in a operation range between 20 and 35 °C, auto-zero in position on with NPLC = 1, range of 125 mV with one hour of warmup.

$$U_{Data} = 1.00\% \tag{13}$$

The uncertainty for the thermocouple is based on the maximum fluctuation of the controller bath, 0.1 °C, and on the average temperature measured, 30.5 °C.

$$U_{Therm} = 0.66\% \tag{14}$$

The uncertainty resultant of the thermal contact resistance is estimated with the maximum difference between the temperatures on the transducer and on the sample surface with an air layer with 0.01 mm thickness. This difference is 0.103 °C, which results in an uncertainty of:

$$U_R = 0.32\% \tag{15}$$

The uncertainty of the heat transducer is estimated from its previous calibration and considers the uncertainties of tension, current and area of measurement. Tension and current were measured with a digital multimeter Goldstar with resolution of 0.01 V for tension and 0.001 A for current. The area was measured with a precision escalimeter Trident with an uncertainty of 0.001 m.

$$U_{H.T.} = \sqrt{U_I^2 + U_V^2 + U_A^2} = \sqrt{1.4^2 + 1.33^2 + 0.01^2} = 1.93\% \quad (16)$$

The numerical uncertainty is estimated with the maximum difference between the temperature calculated by the trapezoid rule and the temperature calculated analytically (Lima e Silva *et al.*, 1998). This difference is 0.152 °C, so the maximum uncertainty is estimated in:

$$U_{Num} = 0.47\% \quad (17)$$

Substituting Eqs. (14-18) in Eq. (13), the uncertainty for the objective function, which by the hypothesis of linear propagation is the uncertaity in the estimation of thermal effusivity, is:

$$U_{Obj} = 2.55\% \quad (18)$$

6. CONCLUSIONS

This work showed a non-destructive method for a thermal characterization of a conductive system. It allows an in-situ measurement, which is important for some operations. The implementation of the system needs only sensors to measure the heat flux and temperature on the surface of material. The thermal contact resistance can be more important in in-situ measurements because of the irregularities of the surface. To reduce this effect a thermal paste with high thermal conductivity can be used. Also the estimation of the thermal effusivity presented good agreement when compared with literature values, but it could be observed that a shorter time step presents better results

7. ACKNOWLEDGEMENTS

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8. REFERENCES

Antczak E., Defer D., Elaoami M., Chauchois A. and Duthoit B., 2007, “Monitoring and Thermal Characterization of Cement Matrix Materials Using Non-Destructive Testing”, NDT&E International, Vol. 40, pp. 428-438.

Balageas D. and Jamet J., 1975, “Effusivity Measurement with an Apparatus of ‘Touchau’ Type”, International Journal of Heat and Mass Transfer, Vol. 18, pp. 933-940.

Beck, J. V., Cole, K. D., Haji-Sheikh, A. and LItkouhi, B., 1992, “Heat Conduction Using Green’s Functions”, Washington D. C., Hemisphere Publishing Corporation, 552 p.

Benedetto G. and Spagnolo R., 1988, “Photoacoustic Measurement of the Thermal Effusivity of Solids”, Applied Physics A: Solids and Surface, Vol. 46, pp. 169-172.

Blackwell, J. H., 1954, “Transient-Flow Method for Determination of Thermal Constants for Insulating Materials in Bulk”, Journal of Applied Physics, Vol. 25, pp. 137-144.

Blumm, J., Lindemann, A. and Min, S., 2007, “Thermal Characterization of Liquids and Pastes Using the Flash Technique”, Thermochimica Acta, Vol. 455, pp. 26-29.

Carvalho, G.; Pereira, F. R.; Ramos, V. D.; Costa, H. M. and Almeida, F. L. L., 2006, “Otimização do Cálculo de Condutividade Térmica em Polímeros”, in: V Congresso Brasileiro de Análise Térmica e Calorimetria, Poços de Caldas – MG.

Coment E., Batsale J. C., Ladevie B. and Battaglia J. L., 2002, “A Simple Device for Determining Thermal Effusivity of Thin Plates”, High Temperatures – High Pressures, Vol. 34, pp. 627-637.

Dadarlat D., Neamtu C., Houriez N., Delenclos S., Longuemart S. and Sahraoui A. H., 2008, “Photopyroelectric Measurement of Thermal Effusivity of Liquids by Sample’s Thickness Scan”, The European Physical Journal Special Topics, Vol. 153, pp. 115-118.

Defer D., Antczak E. and Duthoit B., 2001, “Measurement of Low Thermal Effusivity of Building Materials Using the Thermal Impedance Method”, Measurement Science and Technology, Vol. 12, pp. 549-556.

Fudym, O., Battaglia, J. L. and Batsale, J. C., 2005, “Measurement of Thermophysical Properties in Semi-Infinite Media by Random Heating and Fractional Model Identification”, Review of Scientific Instruments, Vol. 76, pp. 044902-044902-10.

Guimarães, G., Philippi, P. C. and Thery P., 1995, “Use of Parameters Estimation Method in the Frequency Domain for the Simultaneous Estimation of Thermal Diffusivity and Conductivity”, Review of Scientific Instruments, Vol. 66, pp. 2582-2588.

Iguchi, C. Y., Santos, W. N. and Gregório Jr., R., 2007, "Determination of Thermal Properties of Pyroelectric Polymers, Copolymers and Blends by the Laser Flash Technique", Polymer Testing, Vol. 26, pp. 788-792.

Krapez J. C., 2000, "Thermal Effusivity Profile Characterization from Pulse Photothermal Data", Journal of Applied Physics, Vol. 87, pp. 4514-4524.

Lima e Silva, S. M. M., Guimarães, G. and Duarte, M. A. V., 1998, "A Correlation Function for Thermal Properties Estimation Applied to a Large Thickness Sample with a Single Surface". Review of Scientific Instruments, Vol. 69, pp. 3290-3297.

Lima e Silva, S. M. M., 2000, "Experimental Techniques Development for Determining Thermal Diffusivity and Thermal Conductivity of Non-Metallic Materials Using Only one Active Surface", Doctorate Thesis, (in Portuguese), Federal University of Uberlândia, 130 p.

Lima e Silva, S. M. M., Ong, T. H. and Guimarães, G, 2003, "Thermal Properties Estimation of Polymers Using Only One Active Surface", Journal of the Brazilian Society of Mechanical Sciences and Engineering, Vol. 25, pp. 9-14.

Min, S., Blumm, J. and Lindemann, A., 2007, "A New Laser Flash System for Measurement of the Thermophysical Properties", Thermochimica Acta, Vol. 455, pp. 46-49.

Parker, W. J., Jenkins, R. J., Butler, C. P. and Abbott, G. L., 1961, "Flash Method of Determining Thermal Diffusivity, Heat Capacity and Thermal Conductivity", Journal of Applied Physics, Vol. 32, pp. 1679-1684.

Ruggiero, M. A. and Lopes, V. L. R., 1996, "Cálculo Numérico: Aspectos Teóricos e Computacionais", Makron Books, 2nd ed., 406p.

Santos, W. N., Gregório, R., Mummery, P. and Wallwork, A., 2004, "Método do Fio Quente na Determinação das Propriedades Térmicas de Polímeros", Polímeros: Ciência e Tecnologia, Vol. 14, pp. 354-359.

Sheindlin, M., Halton, D., Musella, M. and Ronchi, C., 1998, "Advances in the Use of Laser-Flash Techniques for Thermal Diffusivity Measurement", Review of Scientific Instruments, Vol. 69, pp. 1426-1436.

Taylor, J. R., 1997, "An Introduction to Error Analysis: The Study of Uncertainties in Physical Measurements", University Science Books, 2nd ed., 327 p.

Vanderplaats, G. N., 2005, "Numerical Optimization Techniques for Engineering Design", Vanderplaats Research and Development Inc, 4th ed., USA, 466 p.

Yesilata B. and Turgut P., 2007, "A Simple Dynamic Measurement Technique for Comparing Thermal Insulation Performances of Anisotropic Building Materials", Energy and Buildings, Vol. 39, pp. 1027-1034.

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