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ENHANCED LUMPED FORMULATIONS FOR ESTIMATING THERMAL PROPERTIES OF POLYMERIC NANOCOMPOSITES USING INFRARED THERMOGRAPHY

Debora Carneiro Moreira
Mariana Cristina de Oliveira Telles
Luiz Carlos da Silva Nunes
Leandro Alcoforado Sphaier
Laboratório de Mecânica Teórica e Aplicada, Programa de Pós-Graduação em Engenharia Mecânica, Universidade Federal Fluminense, Rua Passo da Pátria 156, bloco E, sala 216, Niterói, RJ, 24210-240, Brazil
dcmoreira@id.uff.br, marianatelles@id.uff.br, luizcsn@mec.uff.br, lasphaier@id.uff.br

Abstract. *This paper presents an investigation of a new methodology for estimating thermal properties of polymeric materials using infrared thermography. A simple experimental setup was proposed and the temperature fields in the samples were measured and recorded with an infrared camera. In addition, a heat transfer fin model was considered, together with different lumped formulations, in order to select the approach that best describes the experimental data. Finally, the thermal conductivities of nanocomposite samples were estimated by an adjustment of the one-dimensional solutions to the observed temperature distribution. The temperature fields were well described by the chosen theoretical approach, however, there were significant changes in the estimated thermal conductivity, according to the selected formulation.*

Keywords: *Thermal conductivity, Nanocomposite, Polymer, Fin, Steady State.*

1. NOMENCLATURE

a	bar length
b	bar height
Bi	transversal Biot number
Bi^*	modified Biot number
c	bar thickness
h_∞	convective heat transfer coefficient
k	thermal conductivity
L_*	aspect ratio
m	parameter in the fin model
T	temperature
T_b	basis temperature
T_∞	air temperature

Greek Symbols

θ	dimensionless temperature
ξ	dimensionless length

Subscripts

nc	effective or apparent
r	effective or apparent

2. INTRODUCTION

The development of new materials is often related to the design of novel compounds resulting from the combination of materials with different physical or chemical properties. Over the last two decades, fabrication and properties of polymeric nanocomposites have been continuously investigated, since it has been shown that some of polymer properties may be significantly enhanced by the addition of nanofillers (Njuguna *et al.*, 2008; Park *et al.*, 2012). In particular, the addition of metal oxides nanoparticles to a polymeric matrix may result in a composite with higher thermal conductivity than the polymer while maintaining electrical insulating properties, which is interesting for some applications like electronic packaging (Fu *et al.*, 2010) and as thermal interface materials (Sim *et al.*, 2005).

Although there are many works concerning the investigation on thermal properties of polymeric nanocomposites, changes in macroscopical properties of polymers caused by particle loading are not fully known and cannot be accurately predicted. Besides, there are various experimental techniques that can be employed in order to assess the thermal properties of materials and these different techniques may be responsible for some differences among reported results. The thermal conductivity may be estimated, for example, by the guarded heat flow meter method (Moreira *et al.*, 2011; Nayak *et al.*, 2010), by the transient short hot wire technique (Xie *et al.*, 2006), and by infrared thermography, which is most commonly used for determining the thermal diffusivity of materials (Philippi *et al.*, 1995; Miettinen *et al.*, 2008; Laskar *et al.*, 2008). An infrared camera allows the recording of full temperature fields with non-contact measurements and it is considered a powerful tool in many industrial and scientific applications.

In this sense, this paper describes a methodology for estimating the thermal conductivity of polymeric materials using improved heat-transfer fin solutions, which extend the application of the classical one-dimensional solution to situations with larger values of Biot number. The solution to a partially lumped heat conduction equation was fitted to steady state temperature fields recorded by an infrared camera, in order to estimate the unknown parameters. Estimates of thermal conductivity obtained from classical and improved approaches are also compared with previous experimental results measured by a guarded heat flow conductivity meter.

3. MATHEMATICAL FORMULATION

This work deals with a heat conduction problem in a bar with rectangular cross-section and dimensions a, b, and c, in x, y, and z directions, respectively. Figure 1 illustrates the bar with the boundary conditions that were considered.

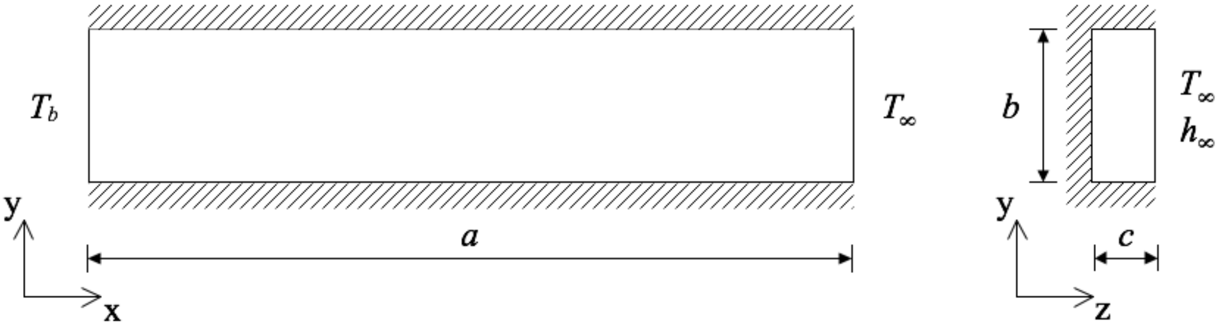

Figure 1. Schematic diagram of a sample in the experimental arrangement.

The multidimensional steady–state heat equation and the boundary conditions in the Cartesian coordinate system are presented below:

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} = 0, \qquad \text{for} \qquad 0 < x < a, \quad 0 < y < b, \quad 0 < z < c, \tag{1}$$

$$T = T_b, \qquad \text{for} \qquad x = 0, \quad 0 < y < b, \quad 0 < z < c, \tag{2}$$

$$T = T_\infty, \qquad \text{for} \qquad x = a, \quad 0 < y < b, \quad 0 < z < c, \tag{3}$$

$$\frac{\partial T}{\partial y} = 0, \qquad \text{for} \qquad y = 0, \quad 0 < x < a, \quad 0 < z < c, \tag{4}$$

$$\frac{\partial T}{\partial y} = 0, \qquad \text{for} \qquad y = b, \quad 0 < x < a, \quad 0 < z < c, \tag{5}$$

$$\frac{\partial T}{\partial z} = 0, \qquad \text{for} \qquad z = 0, \quad 0 < x < a, \quad 0 < y < b, \tag{6}$$

$$k\,\frac{\partial T}{\partial z} + h_\infty T = h_\infty T_\infty, \qquad \text{for} \qquad z = c, \quad 0 < x < a, \quad 0 < y < b. \tag{7}$$

This system of differential equations was partially lumped in y and z directions, resulting in a fin approach (Ozisik, 1993), as follows:

$$\frac{\partial^2 \bar{T}}{\partial x^2} + \frac{1}{c}\left[\frac{h_\infty}{k}(T_\infty - \bar{T})\right] = 0, \qquad \text{for} \qquad 0 < x < a, \tag{8}$$

$$\bar{T} = \bar{T}_b, \qquad x = 0, \tag{9}$$

$$\bar{T} = T_\infty, \qquad \text{for} \qquad x = a. \tag{10}$$

where $\bar{T}$ is the average temperature at the fin cross section:

$$\bar{T}(x) = \frac{1}{b\,c}\int_{z=0}^{c}\int_{y=0}^{b} T(x,y,z)\,\mathrm{d}y\,\mathrm{d}z. \tag{11}$$

Dimensionless formulations are frequently used in order to homogenize systems of differential equations, allow the comparison of different data sets and reduce the number of variables in a given problem. In this context, the dimensionless

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variables are defined:

$$\xi = \frac{x}{a} \qquad \text{and} \qquad \theta = \frac{\bar{T} - T_\infty}{T_b - T_\infty}, \tag{12}$$

and the dimensionless formulation of the one-dimensional heat conduction problem is given as:

$$\frac{\mathrm{d}^2\theta}{\mathrm{d}\xi^2} - m^2\,\theta = 0, \qquad \text{for} \qquad 0 < \xi < 1, \tag{13}$$

$$\theta = 1, \qquad \text{at} \qquad \xi = 0, \tag{14}$$

$$\theta = 0, \qquad \text{at} \qquad \xi = 1. \tag{15}$$

where

$$m = \sqrt{\frac{\mathrm{Bi}^*}{L_*^2}}, \qquad \text{and} \qquad L_* = \frac{c}{a}. \tag{16}$$

It is important to mention that Bi^* is a modified Biot number, which is equal to the transversal Biot number for the classical lumped approach. The modified Biot number resultant from classical and improved lumped approaches for $H_{0,0}/H_{0,0}$ and $H_{1,1}/H_{0,0}$ approximations are defined as follows (Corrêa and Cotta, 1998):

$$\mathrm{Bi}^*_{CLSA} = \mathrm{Bi} = \frac{h_\infty\, c}{k}, \qquad \mathrm{Bi}^*_{IL-00/00} = \frac{\mathrm{Bi}}{1 + \frac{\mathrm{Bi}}{4}}, \qquad \mathrm{Bi}^*_{IL-11/00} = \frac{\mathrm{Bi}}{1 + \frac{\mathrm{Bi}}{3}} \tag{17}$$

In order to assess the thermal conductivity augmentation (k_{nc}/k_r) of the nanocomposites, the parameter m was fitted to the experimental results and the relation m_{nc}/m_r was calculated. The recorded temperature distribution was previously averaged in the y direction. The convective heat transfer coefficient (h_∞) was assumed to be the same for all cases.

4. EXPERIMENTAL METHODOLOGY

4.1 Materials and manufacturing

The samples were composed of an epoxy matrix filled with different amounts of alumina (Al_2O_3) nanoparticles, varying from 0% to 10% in volume fraction (ϕ). The epoxy resin (ER) RR515, from SILAEX, is based on a diglycidyl ether bisphenol A and was polymerized by the addition of an aliphatic amine hardener in a proportion of 1:4 by weight. The dispersed phase was composed of 200 nm α-Al_2O_3 spherical nanoparticles, provided by NanoAmor. Table 1 shows the thermophysical properties of the nanoparticles. More details about the nanocomposites manufacturing may be seen in (Moreira *et al.*, 2012).

Table 1. Thermophysical properties of α-Al_2O_3 nanoparticles.

Property	Al_2O_3
thermal conductivity (W m^{-1} K^{-1})	30
true density (kg/m^3)	3700
morphology	spherical
particle size (nm)	200
purity	≥99.99%

The employed specimens were bars with uniform rectangular cross section and dimensions 68 mm × 12 mm × 5 mm. According to previous results, the thermal conductivity of the samples ranged from 0.20 W m^{-1} K^{-1} (for neat epoxy samples) to 0.38 W m^{-1} K^{-1} (for 10% alumina loaded nanocomposites).

4.2 Infrared thermography

The samples are allocated in a heat insulating holder and subjected to a uniform base temperature. Convection heat transfer occurs at one of the sample's surfaces and its temperature fields are recorded by an infrared camera Flir A325G. The experimental setup may be seen in Fig. 2. All specimens were coated with the same black paint, in order to magnify and standardize the emissivity of the surface facing the infrared camera. A Peltier element was used as the heat source

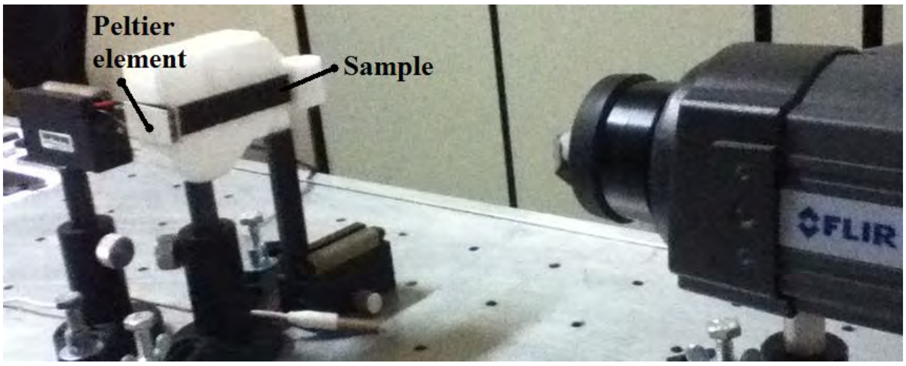

Figure 2. Experimental setup.

and some images were recorded when the steady state regime was reached, which could be verified by the temperature profile remaining constant.

As was mentioned before, the heat transfer coefficient (h_∞) was considered to remain constant in tests with distinct samples, in order to evidence the thermal conductivity augmentation of epoxy/alumina nanocomposites; however, in natural convection, it is known that temperature may affect significantly the convective heat transfer coefficient. On the other hand, higher heat transfer coefficients are achieved in forced convection, and even using enhanced lumped formulations, high values of Biot number should be avoided, to maintain the validity of fin model. In this sense, all samples were tested under three conditions: natural convection on the free surface, and forced convection with two different fans; with the objective of enlightening the best experimental arrangement for our purposes.

4.3 Guarded heat flow meter

Cylindrical specimens were also fabricated to be tested by a commercial equipment. A thermal conductivity meter, *LaserComp* Fox-50, was used for measuring the thermal conductivity of epoxy/alumina nanocomposites by the guarded heat flow meter method. This equipment operation is based in thermal resistances principle: the specimen is confined between two plates with distinct but known temperatures (Figure 3) and the heat flow is measured by one sensor in each plate, such that the average heat flow and the temperature difference may be used to calculate the thermal resistance of the sample. Sample thickness is also measured by the equipment and it is possible to set a value for the thermal contact resistance. The thermal conductivity of the sample is calculated based on values of total thermal resistance, contact thermal resistance and sample thickness. Further details about the tests performed with this equipment may be found in (Moreira, 2011; Moreira *et al.*, 2011).

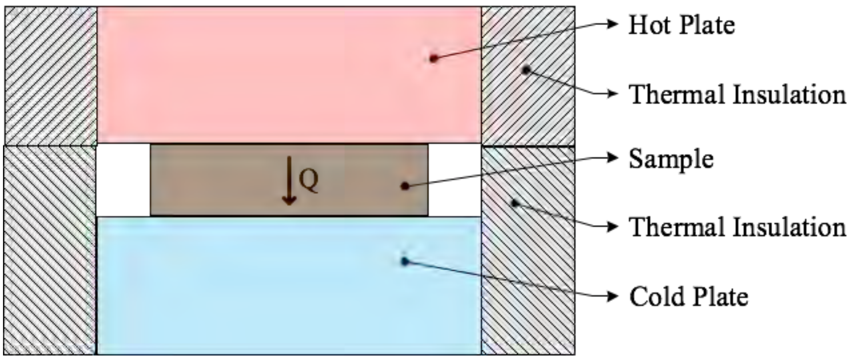

Figure 3. Representative diagram of the guarded heat flow meter method

Tests were performed at four temperature setpoints (0°C, 25°C, 50°C, 75°C), which are the mean temperatures between upper and lower plates. A temperature difference of 20°C between plates was maintained and the thermal contact resistance was set as 0 K/W. Thermal paste was used to decrease thermal contact resistance between samples' surfaces and the plates.

5. RESULTS AND DISCUSSION

The dimensionless temperature distribution in the x direction of epoxy/alumina nanocomposites are displayed in Fig. 4. It is possible to observe that the temperature decaying is smoother for higher loaded samples, which may be explained by the fact that the addition of alumina nanoparticles is responsible for an increase in the thermal conductivity

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of the epoxy resin.

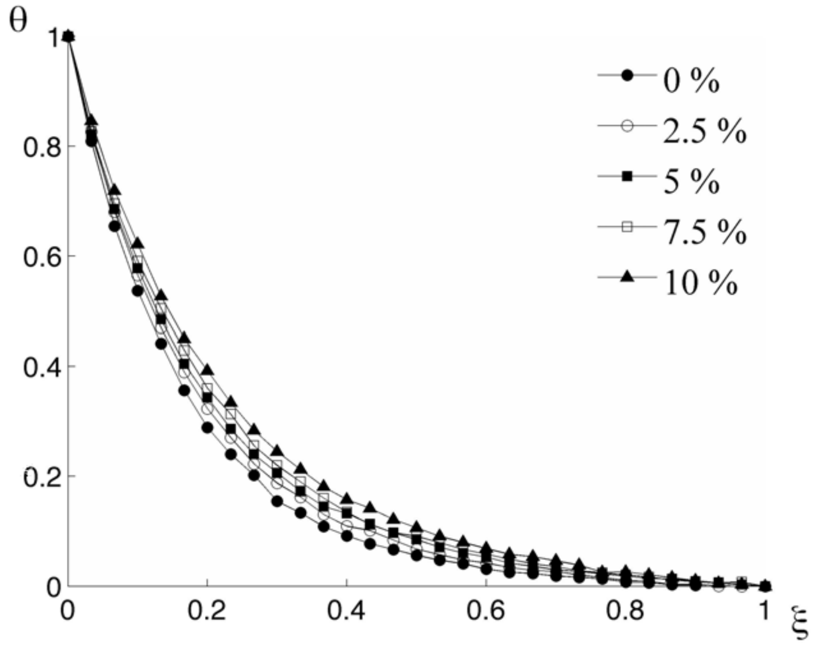

Figure 4. Dimensionless temperature distribution along the x axis of nanocomposite samples.

Figure 5 shows the estimated thermal intensification of the nanocomposites (k_{nc}/k_r) using the three different lumped approaches for each experimental arrangement: with no fan, where it occurred natural convection only, and with fans 1 and 2, respectively. It is important to remember that fan 1 is slower than fan 2. The results are compared with previous results of thermal intensification, obtained from direct measurements of the thermal conductivity of the same nanocomposites by the guarded heat flow meter (GHFM) method (Moreira, 2011).

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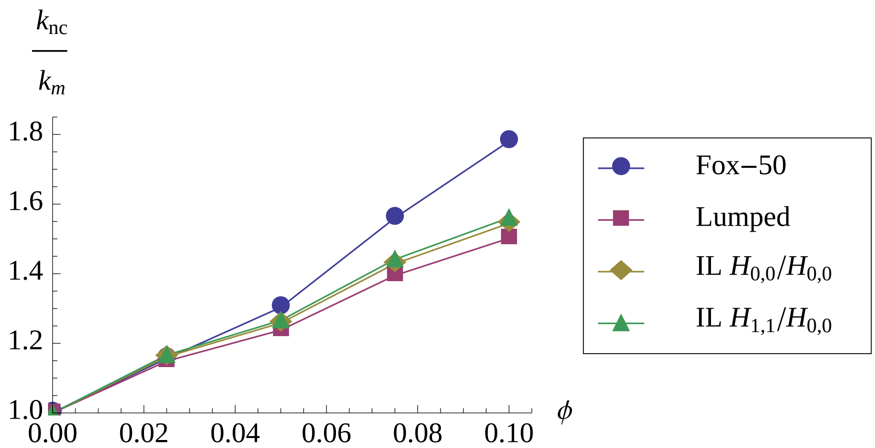
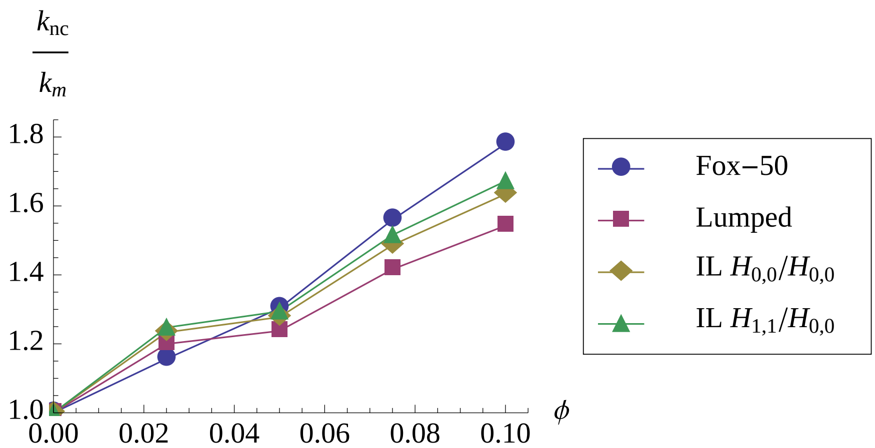
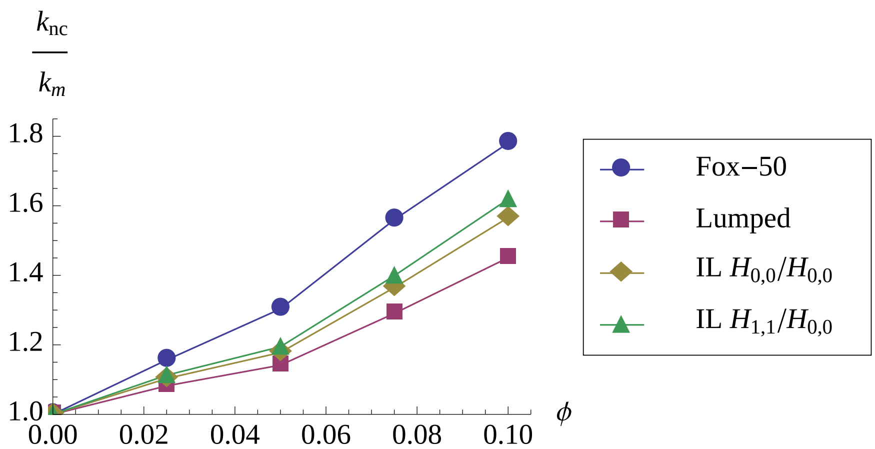


(a) No fan



(b) Fan 1



(c) Fan 2

Figure 5. Comparison of different lumped approaches with Fox-50 results for the thermal intensification of Epoxy/Alumina nanocomposites for three experimental arrangements.

According to Fig. 5(a), results obtained from all three different approaches are closer for the case with no fan, with free convection only. This behavior is expected, since it is possible to consider that the Biot number is smaller in this case, if compared with forced convection cases. It can be noticed that the arrangement with fan 2, which is faster than fan 1, has given thermal intensification results that differ significantly from that attained with Fox-50 device. The observed divergence may be attributed to a high Biot number, which rules out the one-dimensional formulation, even with improved lumped approaches. For that reason, the arrangement with the slower fan (fan 1) was chosen as the best one, since it provides forced convection, damping possible problems in controlling environmental conditions for different tests with free convection, while keeping the Biot number low enough such that the one-dimensional formulation remains valid.

The estimates obtained from the GHFM method were taken at different temperatures and it is possible to observe a slight variation of the thermal conductivity augmentation with temperature. Results of thermal intensification acquired from thermography measurements are plotted together with Fox-50 results. The experiment was arranged with fan 1 and enhanced lumped formulation with $H_{1,1}/H_{0,0}$ approximations were employed.

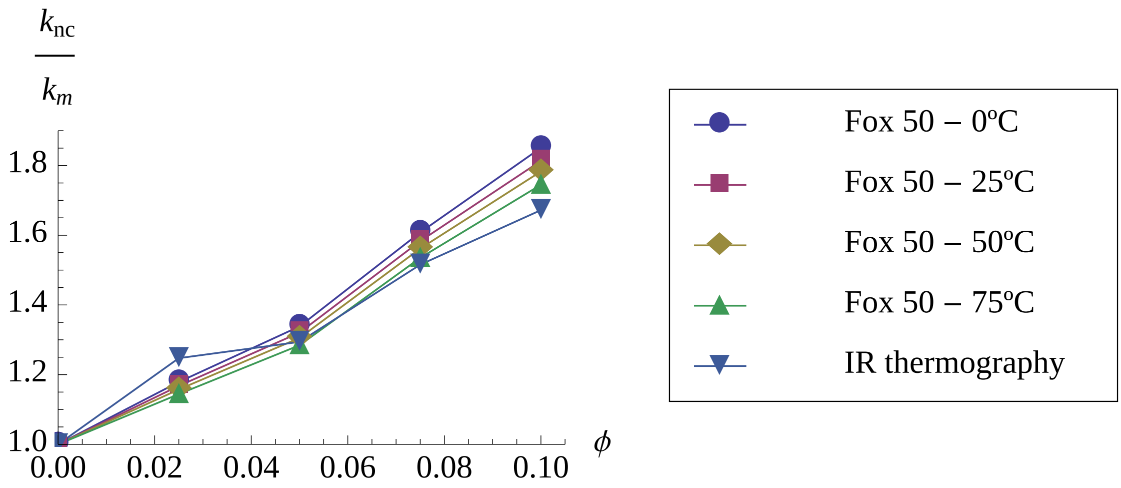


Figure 6. Comparison of the results for the thermal intensification on the epoxy resin estimated by the guarded heat flow meter at different temperatures and the infrared thermography methods.

According to Fig. 6, the thermal intensification estimated by the present method is close to previous data from the GHFM method. However, the observed behavior of thermal intensification is a little distinct, which may be explained by differences in the samples. A further investigation should be carried out and additional data must be collected for a more reliable conclusion.

6. CONCLUSIONS

This paper presented a novel methodology for estimating the thermal conductivity of polymeric materials using temperature fields of a prismatic bar, which were recorded by an infrared camera. A fin model was used and tests were carried out under three conditions: natural convection on the free surface, and forced convection provided by two distinct fans with different velocities. The thermal conductivity augmentation of epoxy/alumina nanocomposites was calculated with the present methodology and compared with measurements obtained by the guarded heat flow meter method using a *LaserComp* Fox-50 conductivity meter.

Besides the classical one-dimensional fin model, an enhanced lumped formulation was used. The improved formulation is based on Hermite approximations, in order to keep the same analytical involvement, while expanding the range of applicability of one-dimensional formulations to larger values of transversal Biot numbers. It was observed that all three approaches presented close results in cases with natural convection, but varied significantly in cases where fans were employed. Also, infrared thermography estimates for thermal conductivity augmentation presented the best agreement with Fox-50 results for the arrangement with the slower fan and enhanced lumped formulation with $H_{1,1}/H_{0,0}$ approximations.

As a final comment, it should be mentioned that the presented results are still preliminary and a more extensive investigation considering other materials and using more measurements will follow this study. In addition, the convective heat transfer coefficient will be calculated, such that, actual values of thermal conductivity may be estimated by the present method.

7. ACKNOWLEDGEMENTS

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