

The Effective Thermal Transport in Composite Materials

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1. Introduction

Composite materials are promising materials, which should exhibit an improve on several aspects of the physical properties such as mechanical, thermal, electrical etc.. A composite material is a system of materials composed of two or more components randomly mixed and bonded on a macroscopic scale. For example, metal-alloys Silicon carbides such as Aluminum Silicon Carbide (AlSiC) is made up of Aluminum, Silicon and Carbone on a microscopic scale. It is a metal matrix composite (MMC) packages that have a unique set of material properties. It is ideally suited to thermal management performance, and a functionality that supports high-density interconnection microelectronic packaging applications (Hollecka et al., 1988) & (Zhang et al., 2004). This example is one of the concrete evidences that thermal conduction is an important feature of composites' application for electronic packaging, which are associated with thermal insulation, and heat spreader. General speaking, we should have different composite materials which are suitable for different applications (Xu, & Yagi, 2004), and this can be achieved because of the unique character of the detailed microstructure of the composites.

In particular, a composite material is composed of strengthening items (reinforcement) such as particles, flakes, fibers, etc., embedded in a matrix of metals, ceramics, or polymers. The matrix holds these items to form the desired shape while the items improve the overall physical properties of the matrix. When designed properly, the new combined material exhibits better physical property than would each individual material. When composites are selected over traditional materials such as metal alloys or plastics, it is usually because of one or more desired properties regarding cost, weight, size, surface condition, strength, thermal and electrical conduction etc...

For example, Silicon Carbide SiC/SiC composites, which are relatively cheap, have upper limit for thermal conductivity that exceeds the corresponds obtained in single crystal and high-purity chemically vapor deposited CVD SiC. Effective thermal conductivity (λ_{eff}) of such composites reaches maximum values of $\sim 320 \text{ W/m } ^\circ\text{C}$ at room temperature (Zinkle & Snead, 1999). This value is comparable if not higher than the thermal conductivity of some precious expensive metals such gold and silver.

A new generation of functionally graded fibrous composites called functionally graded materials (FGMs) (Suresh, 1998), (Koizumi, 1993) and (Fung & Hu, 2008). FGMs have dynamic effective thermal properties and the volume fraction of the materials changes

gradiently. It is the non-homogeneous microstructures in these materials that produce continuous graded macroscopic properties, such as the thermal conductivity, specific heat, mass density and elastic modulus. FGMs have been developed as the super-resistant materials for propulsion systems and airframe of the space planes in order to decrease thermal stresses and to increase the effect of protection from heat (Gray et al., 2003) and (Kuo & Chen, 2005). FGMs can reduce the thermal stress in such structures working in high temperature environment. All the effective thermal properties of FGMs can provide great help in predicting the overall behavior under various loading conditions. The least to say is that the theoretical and experimental investigation of the effective thermal properties of all composites including FGMs is an area which has received great interest in recent years.

2. Historical background of theoretical models

Estimation of the effective thermal conductivity of composite material has been subject of many theoretical and experimental investigations. The earliest model was proposed by Maxwell (Maxwell, 1892). He derived an expression for the simplest kind of two-phase dispersion consisting of spherical particles suspended or imbedded into a continuous medium of another material neglecting the interactions between the particles. The derived expression was valid only for very low concentrations of the dispersed phase i.e. for dilute volume fraction.

For non-dilute volume fractions, The interaction between the spherical particles has a significant effect and cannot be neglected, the work of Maxwell was later followed by the work of Rayleigh in 1892 to account for these interactions. By considering a form of a simple cubic lattice of spheres in a homogeneous material and using a series expansion, Rayleigh assumed that the spherical particles as inclusions that form a cubical array, and included the interaction effect of a number of nearby spheres.

Other proposed works for non-spherical inclusions developed by other researchers such as the work of (Polder & Van Santen, 1946), (Reynolds & Hough, 1957), (Hamilton & Crosser 1962), (Rocha & Acrivos, 1974) and others. Extension of these previous works was also carried out by (McPhedran & McKenzie, 1978), and (Sangani & Acrivos, 1983). In the course of models developments Hashin, 1968 proposed a general self-consistent treatment. The treatment provides a physically realistic model of particle to particle interaction for two phase system covering the full range of the volume fraction. The self-consistent field concept is extended to include the contact resistance in the composite reinforced with coated spheres. Benveniste & Miloh (1991) and Felske et. al. (2004) employed effective medium theories to predict the effective thermal conductivity of coated short-fiber composites. Later, Samantray and co-workers (Samantray et al, 2006) proposed the correlations between the inclusions to estimate the effective thermal conductivity of two-phase materials. Recently in 2009, (Fang et. al, 2009) applied the thermal wave method to investigate the unsteady effective thermal conductivity of particular composites with a functionally graded interface. The scattering and refraction of thermal waves by a spherical particle with an inhomogeneous interface layer in the matrix are analyzed, and the results of the single scattering problem are applied to the composite medium.

2.1 Determination of bounds and estimation of λ_{eff}

In general, using a theoretical model to estimate the effective thermal conductivity (λ_{eff}) of a multiphase medium requires the previous knowledge of the thermal conductivity of each

phase, the volume concentrations of the phases, the phase morphology (distribution), the shape of inclusions of the different phases in the solid matrix, and the fractional porosity if the medium is porous.

In this chapter, the problem of determining the bounds and/or estimating the effective thermal conductivity (λ_{eff}) of a composite (multiphase) system has been examined. A comparison between the measured data and the results predicted by theoretical models has been made. Two different categories of composite materials have been investigated, namely wood and ceramics. In particular we investigate the effect of morphology in woods and mineralogy in ceramics.

Wood is a heterogeneous porous material with known anisotropy due to its intrinsic distribution of phase morphology (inter layers arrangement). It should be mentioned that measurements of the thermal conductivity as a function of phase morphology (grain size and orientation) indicate preferential heat conduction along the conducting chain of grains. Its low for random distribution of grains and high for layered (parallel) grains.

The phenomena of heat transfer in wood depend on the geometry of the wood, as well as porosity. Such heterogeneous medium containing the three phases, its thermal conductivity is only an apparent conductivity because it results from complex exchanges concerning simultaneous conduction in solids, and fluids (gases and/or liquids). Regarding the theoretical models to estimate wood conductivity, there are various models of heat conductivity are given in the literature, for example, see references given by Gronli (1996). In this study, we will use one of these models which given by (Kollmann & Cote, 1968). The influences of density, porosity and anisotropy on thermal transport in wood are investigated. To estimate the effective thermal conductivity (λ_{eff}) in a fibrous wood like structure, a model (Kollmann, & Cote, 1968) based on a weighting bridge-factor (ξ) between two limiting parallel and serial conduction cases have been used. The model indicates an increase in the effective conductivity as the density/porosity increases/decreases which was in agreement with our investigation regarding the influence of microstructure on the heat conduction in wood. Moreover, indicates significant difference between the longitudinal and the transverse directions.

At this point, it is worth mentioning that a worldwide research and development efforts are underway to examine the potential use of a wide range of non-destructive testing (NDT) and non-destructive evaluation (NDE) technologies for evaluating wood and wood-based materials as fiber-based materials or multiphase composites—from the assessment of standing trees to in-place structures (Brashaw et al., 2009).

Ceramics on the other hand are multiphase mechanically strong, relatively non-porous material with unknown anisotropy but with known volume fractions and conductivities of phases. The thermal transport in such material depends mainly on its mineralogy (microstructure details) of its constituents. Seven heterogeneous samples of Ceramics marbles and glasses have been selected. The tested models include those of the effective medium theory (EMT) (Noh et. al 19), Hashin and Shtrikman (HS) bounds (Hashin & Shtrikman, 1962) and Wiener bounds, (Wiener, 1912). These models can be used to characterize macroscopic homogeneous and isotropic multiphase composite materials either by determining the bounds for the effective thermal conductivity and/or by estimating the overall conductivity of the random mixture. It turns out that the most suitable one of these models to estimate λ_{eff} is the EMT model. This model can be used to characterize macroscopic homogeneous and isotropic multiphase composite materials after determining the parallel and serial bounds (Wigner bounds) of the overall conductivity of the random

mixture. We used Wiener bounds, as a preliminary indicator to validate the homogeneity condition of the investigated samples, i.e. the measured values for the conductivity should be within the limits of the Wigner bounds. After a comparison between the experimental data of the effective thermal conductivity with the corresponding theoretical estimation, it turns out that the EMT model is a suitable one to estimate λ_{eff} . This model is a mathematical model based on the homogeneity condition, which satisfies the existence of a statistically homogeneous medium that encloses inclusions of different phases. Numerical values of thermal conductivity for the samples that satisfy the homogeneity condition imposed by the effective medium theory are in best agreement with the experimentally measured ones.

3. Experimental techniques

The reliability of a specific technique to measure thermal properties is determined by several factors, such as the speed of operation, the required accuracy and performance under various environmental conditions, the physical nature of material, and the geometry of the available sample. However, in most techniques the main concern is to obtain a controlled heat flow in a prescribed direction, such that the actual boundary and initial conditions in the experiment agree with those assumed in the theory. There are several methods (techniques) used to measure the effective thermal conductivity of composites. Indeed, the choice of technique depends on the type of application, size of the sample and the available sample geometry; however they are divided into two groups, the steady state and the non-steady state (transient) methods.

3.1 Steady state techniques

In the 1st group, the sample is subjected to a constant heat flow. In the past, much attention has been focused on the problems of this group. Examples of steady state are the well-known guarded hot plate and heat flow meter techniques for thermal conductivity (ISO 8302, 1991), (Gawin et al., 2004) and (Sombatsompop, 1997) which are typically suited for bulk applications that require large and thick samples. For these methods, testing times can be long due to the need for thermal equilibrium, which can be of the order of 24 hours, and temperature gradients across the sample can be large. The principle of the heat flow meter method is based on mounting the test-sample between two plates, a 'hot' and a 'cold' plate, then a heat flux transducer is used to measure the heat flow through the sample. Using measurements of the heat flux, temperature difference across the sample and sample thickness the thermal conductivity can be found. The method is a quasi-steady state method and is a variant of the guarded hot plate technique thus the instrument and sample must be allowed to reach isothermal condition (thermal equilibrium) before measurements are made. It should be mentioned that this method is a comparative method and thus the instrument must be calibrated using a specimen of known thermal conductivity. More details and analysis can be found in ASTM E1530.

On other hand, these steady state methods may not be suited to testing molten composites which include soft materials such as plastics and polymers. In this type of composites, in addition to other practical factors related to the molten state of the soft materials, they are vulnerable to degradation during performing the measurements and before the required thermal equilibrium is attained. The thermal properties of such composites are influenced by the level of crystalline and molecular structure of the molten which are directly related to its thermal history (Sombatsompop, 1997).

3.2 Non-steady state(transient) techniques

In the 2nd group, a periodic or transient heat flow is established in the sample. In comparison with the first group, the transient or non-steady state techniques for thermal conductivity are appealing in that the test time is comparatively short, small specimens can be measured, and formed products can be tested (e.g. films, sheets and mouldings). Two Examples of this group are the transient plane source (TPS)(Gustafsson , 1991) to determine the thermal conductivity, thermal diffusivity and their anisotropy (Suleiman et al. 1999) & (Fan et al.,2006), and the transient line-source probe method (Dawson et al., 2006).

The transient line-source probe technique, also known as the needle probe method, is a development of the hot wire method but is suited for testing molten composites in both their molten and solid states (Dawson et al., 2006). A thermocouple and linear heating element are enclosed and casted in a form of a needle, which is typically 50-100 mm in length and 1.5 - 2 mm in diameter. The heater is positioned along the length of the needle and the thermocouple is anchored at the middle position. The needle is immersed and held in the sample until temperature equilibrium is achieved at the desired temperature. Then, by applying a voltage across the resistance heater during a specified time period a heat wave propagates through the sample in the radial direction. The thermal conductivity is determined from the temperature-time profile of the probe. It should be noted that the temperature-time profile (the rate of temperature rise) is sensitive to and depended on the magnitude of the thermal conductivity of the test-sample. It is low profile for higher conductivity and visa versa. Due to this sensitivity and other factors such as the finite probe dimensions and end effects, the technique requires the probe to be calibrated using a reference material with known thermal conductivity (ASTM D5930-01).

More attention will be given to the TPS technique due the fact that most of the experimental data and results discussed in this work are obtained using the TPS technique.

The principle of this technique is simple. The sample is initially kept at thermal equilibrium, and then a small disturbance is applied to the sample in a form of a short heating pulse through the heater/sensor. The temperature profile is characterized by a transient temperature rise of a plane heat source/sensor, known as Hot Disk (Gustafsson, 1991). The hot disk is a thin resistive element, in the form of a bifilar spiral, which acts as both a heat source and a temperature sensor, a schematic drawing is depicted in fig 1. Measurements are simply performed by recording the voltage (resistance/temperature) variations across the sensor during the passage of a heating current in a form of a constant electrical pulse. Figure 1 shows actual recordings of low conductivity ceramic and relatively high conductivity steel samples. These recordings correspond to relatively low temperature rises (around one degree) across the samples. Using this type of sensor, it is possible to measure both the heat input and the temperature increase, from which both the thermal conductivity and thermal diffusivity are simultaneously determined. The theory of the TPS method is based on a three-dimensional heat flow inside the sample, which can be regarded as an infinite medium, if the time of the transient recording is ended before the thermal wave reaches the boundaries of the sample. To validate the theory of the method, the diameter of the sample should be larger than the diameter of the disk as shown schematically in figure 1. In other words; the sample is large enough so that it can be regarded as infinite medium provided that the time of the transient recording is ended before the thermal wave reaches the boundaries of the sample and produces edge effects. Edge effects can be prevented by satisfying a criterion concerning the probing (penetration) depth. The criterion is associated

with the total time of the transient event which is proportional to the square of the overall dimension of the conduction pattern or the distance from the conducting pattern to the nearest free surface of the sample. This is evident from the expression for the probing depth (Gustafsson, 1991) $\Delta_p = \beta (t_{\max} \kappa)^{1/2}$, where t_{\max} is the total time of the recording, κ is the thermal diffusivity of the specimen and β is an adjustable parameter that is related to the relative sizes of the sensor and the specimen, and indirectly correlated with the experimental accuracy. The probing depth and the specimen size are intimately connected in the sense that the shortest distance from any point on the sensor element to the nearest point on any of the free surface of the specimen must always exceed Δ_p to avoid edge effects. Edge effects due to the reflected thermal wave at the boundaries disturb the measurements and affect the accuracy. To achieve high accuracy in our measurements, a β -value of 1.42 seems to define reasonably well the probing depth. The assessment of the temperature increase $\Delta T(t)$ in the heater depends on several factors such as the power output in the sensor element, the design parameters of the sensor, and the thermal transport properties of the surrounding specimen. For the disk-shaped sensor, the thermal conductivity and diffusivity can be obtained from $\Delta T(t)$ that is given by the following equation:

$$\Delta T(t) = P_o (\pi^{3/2} a \lambda)^{-1} D(t) \quad (1)$$

Here P_o is the total output power, λ is the thermal conductivity of the test-sample, and a is the radius of the sensor. $D(\tau)$ is the theoretical expression (Suleiman, 1994) of the time dependent temperature increase, which describes the conducting pattern of a disk-shaped sensor, assuming that the disk consists of a number m of concentric ring sources.

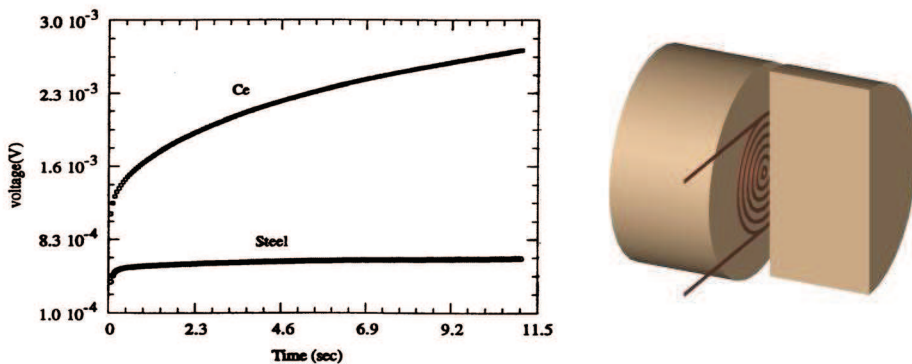


Fig. 1. A typical voltage recordings and a schematic drawing of sample/sensor experimental set-up

This technique has an additional advantage of using a flat thin sensor, which makes it more suitable to substantially, reduce the contact resistance between the specimen and the sensor. This technique has been used several times to report thermal conductivity measurements over a wide range of temperatures (Gustafsson, 1991) & (Suleiman et al, 1996) including the

data presented in next section. A full complete description of the experimental capability regarding precision/accuracy and reproducibility of the measured data of various applications is given elsewhere (Suleiman, 1994).

4. Results and discussion

Two types of multiphase composite materials have been selected, namely; wood and ceramics. As it was mentioned in section 2, in particular we investigate the effect on thermal conduction due to structural morphology in woods and structural mineralogy in ceramics.

4.1 Woods

It is known that wood is a fiber-based ligno-cellulosic material (Avramidis & Lau, 1992). In this type of materials, the crystalline structure of cellulose chains may be altered, due to temperature variations, leading to a permanent loss in strength and considerable changes in physical behaviour including its ability to conduct heat. The purpose of this study is to investigate the relation between the thermal conductivity of wood and the various factors affecting this property. The transient hot-disk method was used at 20 degree Celsius to perform measurements of the thermal conductivity (λ).

The investigated wood samples were hardwood from birch trees used for energy conversion purposes (approximate age of 10-15 years) with a stem diameter of approximately 100 mm. The samples were cut from the stem at a distance of one meter above the ground level. The samples were prepared from different stems, each sample consisting of two disks. Six samples were selected so that sufficient uniformity of density, grain size and orientation, and other physical characteristics could be obtained. The measurements were performed on this six samples on oven-dry basis at room temperature. To insure good thermal contact, in the experimental arrangement the sensor was clamped between the sample halves that consist of two identical cylinder-shaped pieces, each having a diameter of 30 mm and a thickness of 10 mm, see figure 1. The two disks that are required for each test sample were cut from successive portions of the same stem to obtain as much as possible identical and uniform characteristics. To study the influence of fiber direction on thermal conduction the disks were cut parallel (three samples) or perpendicular (three samples) to the fiber direction. Their surfaces were smoothened with sandpaper and equilibrated to the same testing laboratory conditions and then the measurements were performed along the prescribed directions. Although the theory of the TPS method is based on a three-dimensional heat flow inside the sample, however, the samples were cut in a specified direction to maintain a unidirectional-measured property and to minimize any additional contribution of heat flow from the perpendicular directions to the assigned direction of measurement. By cutting the sample in a specified direction, it can be regarded as a unidirectional infinite medium if the time of the transient recording is ended before the thermal wave reaches the boundaries of the sample and produces edge effects. These measurements were done on the longitudinal (along the grain) direction and on the transverse direction (a combination of both radial and tangential directions to the annual rings), the arrows in figure 2 illustrate the measurements directions. Fan and his co-workers (Fan et al, 2006) proposed a fractal model based on SEM images to predict both the tangential and the radial effective thermal conductivities of wood. They used improved TPS method to validate the model, obtaining credible and precise results.

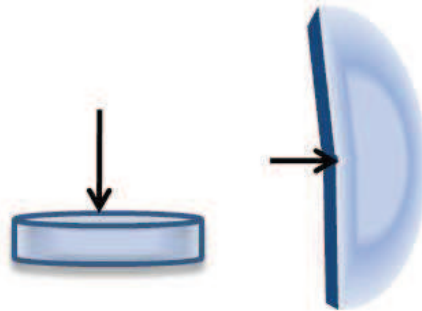


Fig. 2. Schematic drawing; the arrows indicate the longitudinal and transverse measurements directions, respectively

All samples were oven-dried at a temperature of 120 °C until they reached a moisture free condition at constant mass. At a temperature of 120 °C both the free water and the bound water in wood are released, and thus the samples were completely dry. To prevent presence of any moisture, the samples were wrapped by Al-foil during the measurements and kept in plastic containers at all other times. Based on oven-dry masses and volumes, the average values of the apparent densities were calculated at the time of measurements for each sample.

Wood is a porous material and porosity is a parameter, which is likely to affect the magnitude as well as the temperature dependence of the thermal conductivity. This porosity r can be estimated using

$$\rho = 1 - (\rho_{ave}/\rho_{th}) \quad (2)$$

where ρ_{ave} is the average apparent density of the sample, ρ_{th} is the assumed theoretical density of a compact solid wood free from voids. Its value is taken to be 1500 kg/m³ (Kollmann & Cote, 1968). The accuracy of the thermal conductivity data has been estimated to be better than 5%. Care was taken not to increase the temperature in the samples more than one degree Celsius. A larger temperature difference will mask any changes that occur in a narrow interval around a phase transition (Suleiman et. al, 1996). The mean values of five runs of λ at 20 °C, are represented in Fig. 3 for both longitudinal (filled circles) and transverse (open circles) directions. The variations of the thermal conductivity λ as a function of density and porosity are given by the lower and upper axis, respectively. The accuracy was within 3-5% for the conductivity measurements. According to the estimated porosity given in the figure, and considering the fact that the samples have similar thermal conductivity behavior in terms of their thermal profiles, the longitudinal /axial samples would be expected to have somewhat higher λ -values than the transverse samples as shown in the figure. They exhibit the same behavior that of the density but with negative slopes as given by Eq.(2); the conductivity decreases as porosity increases. Although the few data points in the figure show the expected general trend for the conductivity, the figure does not explain why, at fixed temperature (20 °C), there is a difference in the slope as density increases or porosity decreases. Our discussion will be focused on λ and its relation to microstructure (density and/or porosity) and anisotropy. The porosity is typically in the range from 0.5 to 0.8. Voids due to the porosity serve as scattering centers for phonons, and they take up a fraction of the heat conduction volume of the material leading to a lower thermal conductivity. From the

present data related to density and porosity, the following can be deduced: Firstly, a density variation of 20% could alter the conduction by 15% as it is the case for the longitudinal direction at 20 °C. Secondly, the presence of other heat conduction obstacles in addition to voids, such as rays and cell boundaries could also affect the conduction process.

However, it seems that conduction through voids is the dominant factor influencing heat conduction in this type of wood samples. Peculiarities in bulk structure or inherent chemical composition of the different samples appear to have no influence upon thermal conduction, other than that due to the unique microstructure of each sample.

Regarding the anisotropic nature of wood there is no dispute about direction dependency of thermal conductivity in both hard and soft woods, see for example the review by Steinhagen (1977). Thermal conductivity in the longitudinal direction (parallel to the grain) is greater than conductivity in the transverse direction. This may be attributed to orientation of the molecular chains within the cell wall. The long-chain linear polymers (cellulose) that comprise the cell wall, are arranged in bundles called microfibrils. These microfibrils are most closely aligned with the longitudinal axis of the cell. Obviously, the thermal conduction is higher along the length of a microfibril (parallel direction) than across a series of microfibrils (series direction), (Parrott & Stuckes, 1975).

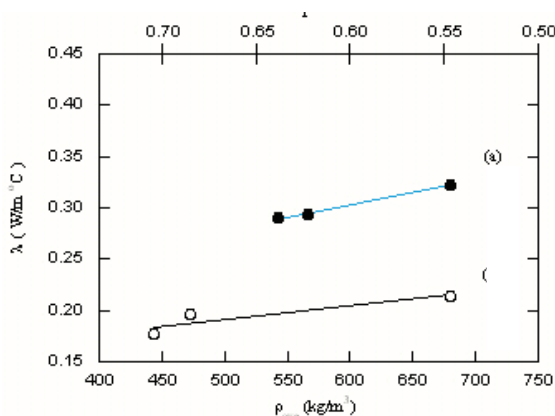


Fig. 3. Thermal conductivity as a function of density (lower axis) and porosity (upper axis) in the longitudinal (solid circles) and the transverse (open circles) directions, respectively

There might be some debate about the direction dependence of the transverse conductivity (the difference in conductivity between the tangential and radial directions with respect to the annual rings). Radial conductivity may be higher than the tangential conductivity and this may be attributed to the influence of the radially oriented wood rays. According to Steinhagen (Steinhagen, 1977), it appears that the ratio of the tangential versus radial conductivity is primarily determined by the volume of the ray cell in hardwoods and by latewood volume in softwoods. However, in our case, no appreciable difference would be expected between the radial and the tangential conductivities, since our samples have a uniform structure throughout the annual rings. Furthermore, our samples are cut from young trees, i.e. the samples are mostly free from latewood.

On the other hand, our data show a significant difference between the longitudinal and the transverse directions. This difference is expected to be larger than what we have measured. The reason for this is that, in order to satisfy the theoretical principle of the Hot-Disk

method and to maintain unidirectional measured properties, we have cut the samples in a specified direction, and this will not eliminate any contribution of heat flow from the perpendicular directions to the assigned direction of measurement. Thus, the resultant measured thermal conductivity in our case is an averaged value, which is lower than the actual value in the direction of measurement.

Kollmann & Cote 1969 model gives the effective thermal conductivity of a wood-like fibrous structure by using a weighting bridge-factor (ξ) between the two limiting cases of Wiener bounds. Namely: conduction in parallel layers (λ_{\parallel}) of gas, liquid and solid phases and serial layers (λ_{\perp}) in relation to the heat transfer direction. Furthermore, the model includes the influence of the radiation in the gas phase. According to this model, the effective thermal conductivity (λ_{eff}) for pre-dried wood can be written as:

$$\begin{aligned}\lambda_{\text{eff}} &= \xi \lambda_{\parallel} + (1 - \xi) \lambda_{\perp} \\ \lambda_{\parallel} &= (1 - r) \cdot \lambda_{\text{fiber}} + r(\lambda_g + \lambda_{\text{rad}}) \\ \lambda_{\perp} &= \frac{1}{\frac{(1-r)}{\lambda_{\text{fiber}\perp}} + \frac{r}{(\lambda_g + \lambda_{\text{rad}})}}\end{aligned}\quad (3)$$

where r is the porosity, $\lambda_{\text{fiber}\parallel}$ is the thermal conductivity of the cell-wall substance/fibers along the grain, $\lambda_{\text{fiber}\perp}$ is the corresponding conductivity perpendicular to the grain, and λ_g is the gas(air) conductivity. The irradiative thermal conductivity λ_{rad} , at 20 °C, is very small and can be neglected.

A plot of the effective thermal conductivity (λ_{eff}) for various values of the bridge-factor is shown in Fig. 4. This plot was generated by applying Eq. (2) and using $\lambda_g = 0.0258 \text{ W/m } ^\circ\text{C}$ for dry air, $\lambda_{\text{fiber}\parallel} = 0.766 \text{ W/m } ^\circ\text{C}$ and $\lambda_{\text{fiber}\perp} = 0.430 \text{ W/m } ^\circ\text{C}$ for the wood fibers at a density of 1500 kg/m^3 . The values of $\lambda_{\text{fiber}\parallel}$ and $\lambda_{\text{fiber}\perp}$ have been used by Gronli, (Gronli, 1996) and were taken from mean values at room temperature reported by Maku, (Maku, 1954) and by Siau, (Siau, 1984).

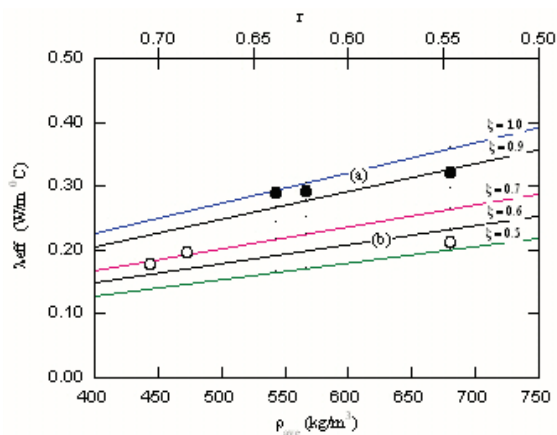


Fig. 4. The effective thermal conductivity according to eq (3) for different ξ values as a function of density(lower axis) and/or porosity (upper axis). The symbols of the data points are the same as the corresponding ones in Fig. 3

Our data in the longitudinal direction at 20 °C lie in the range $\xi = 0.8-1.0$. The data in the transverse direction are in the range $\xi = 0.5-0.7$. At 20 °C, our values for both directions are in agreement with literature data. Gronli (1996) obtained ξ in the range from 0.8 to 1.0 for the longitudinal direction and in the range from 0.35 to 0.6 for the transverse direction. Kollmann et al. (1968) found $\xi = 1$ for the longitudinal direction and $\xi = 0.58$ for the transverse direction.

Furthermore, the model indicates an increase in the effective conductivity as the density increases or porosity decreases, which agrees with our interpretation regarding the influence of microstructure on the heat conduction in wood. Therefore, such measurements can be used to probe the effect of anisotropy (morphology) in wood samples.

It should be noted that there are some constraints imposed upon using this model, such as a limited number of samples, using mean values for $\lambda_{\text{fiber}\parallel}$ and $\lambda_{\text{fiber}\perp}$, and limiting the analysis to room temperature data. Such constraints may introduce a degree of uncertainty in the conclusions. However, there is a tendency of a good agreement between the measured and calculated thermal conductivities and the results are supported by prediction of the model proposed by Kollmann and Cote (Kollmann & Cote, 1968) in spite of the various assumptions involved. Furthermore, in this attempt the purpose of using this model for these six samples, just to examine the possibilities of determining the bounds for estimating the effective thermal conductivity.

4.2 Ceramics

Seven samples of ceramic materials; glasses and marbles with different mineralogy and effective conductivities were selected. These samples were labelled from P1 to P7. These materials are classified into two types of samples, rock marbles and ceramic glasses. The rocks were two grey-red gneissic granite samples labelled P1 & P2 and the other five were either provided through a private communication with Dr. Vlastimil Boháč from the Institute of Physics, Slovak Academy of Sciences (SAS) or selected from the A-Z materials database within the www.amazon.com website. The effective thermal conductivity (λ_{eff}) of such composite multiphase samples with unknown isotropy requires the previous knowledge of the thermal conductivity of each phase, the volume concentrations of the phases, and the shape of inclusions of the different phases in the solid matrix.

Table 1 shows the volumetric fractions of constituents of the seven samples which were determined either by the point-counting method for the marbles or using X-ray (XRF) analysis for the ceramic glasses. The effective thermal conductivity of these samples depends on its mineralogy and the microstructure details of its constituents. Moreover, it appears that for these samples, the major influence on the effective conductivity is due to the magnitude of different conductivities of the constituents and their volumetric composition. For these homogeneous and isotropic (with unknown morphology) multiphase composite materials, we have used the EMT Model mentioned in sec 2.1 to estimate the overall conductivity of the random mixture after determining the serial and parallel conduction (Wiener) bounds. Wiener was the first one to introduce these two bounds.

According to the definitions of Wiener bounds, a dry rock sample consisting of a mixed orientation of mineral layers (parallel and serial distributions of phases), is expected to have an effective conductivity value within these two bounds (homogeneity condition). Hashin & Shtrikman, 1962, introduced a model narrowing the range between Wiener bounds by imposing requirements on the isotropy of the medium. They derived new

bounds for an effective magnetic permeability of a composite material by means of variational theorems. The HS model can be directly applied to different transport properties including thermal conductivity (Helsing & Grimvall, 1991). The corresponding equations of HS bounds for three dimension multiphase media can be expressed in terms of Upper (λ_U) and Lower (λ_L) bounds which are functions of the highest and lowest values of λ_i 's of the different phases and their volume fractions P_i 's. (Porfiri et. al 2008) & (Suleiman, 2010).

Marble Samples	Density (g cm ⁻³)	Quartz	Alkali-felds	Plagioclase	Biotite	Chlorite	Muscovite	Epidote	Zircon	Sphene
		%								
P1	2.79	27.20	24.00	35.20	9.00	0.80	0.80	2.60	0.00	0.40
P2	2.53	22.20	64.10	0.00	1.70	0.00	10.20	1.40	0.40	0.00
Ceramic Samples	Density (g cm ⁻³)	Al ₂ O ₃	SiO ₂	Fe ₂ O ₃	TiO ₂	CaO	MgO	K ₂ O	Na ₂ O	Impurity
		%								
P3	2.76	57.83	36.14	0.68	0.27	0.12	0.25	3.45	0.38	0.41
P4	2.51	47.50	46.30	0.90	0.29	0.23	0.25	3.36	0.56	0.61
P5	2.75	59.98	34.48	0.65	0.25	0.16	0.25	3.32	0.50	0.41
P6	2.50	41.90	55.20	0.80	0.50	0.30	0.20	0.50	0.40	0.00
P7	2.52	46.00	16.00	0.00	0.00	0.00	17.00	10.00	0.00	7 (B ₂ O ₃)

Table 1. The volume fractions of the samples components

Using the more confined limits of Hashin and Shtrikman, it is reasonable to estimate the effective thermal conductivity of the multiphase isotropic medium by taking the mean value of the upper and lower limits (Horai et al., 1972), i.e.,

$$\lambda_{\text{eff(HS)}} = \frac{\lambda_U - \lambda_L}{2} \quad (3)$$

It should be mentioned that according to Chen (Chen et al, 1977) at the outset because of boundary resistance, the Hashin & Shtrikman (1962) bounds, derived under the assumption of temperature continuity between the two phases, are no longer valid in case of relatively large temperature variations at the boundaries.

Tables 2 shows the thermal conductivities λ 's of the marbles and glasses samples components, respectively. These values of λ_i 's for the rocks were taken from Landolt-Börnstein, Numerical Data and Functional Relationships in Science and Technology (1994). The corresponding λ 's values of the individual components for the classes samples were collected from the 88th edition of CRC hand book of chemistry and physics (Lide ed, 2008). The conductivity range for the impurities were estimated from the weighted average value of the other components conductivities using the limits of the upper and lower bounds defined by the above mentioned bounds.

Component	Quartz	Alkali-felds	Plagioclase	Biotite	Chlorite	Muscovite	Epidote	Zircon	Sphene
λ [W/m °C]	7.69	2.15*	1.84*	1.17	5.14	2.32	2.82	4.45	2.33

Component	Al ₂ O ₃	SiO ₂	Fe ₂ O ₃	TiO ₂	CaO	MgO	K ₂ O	Na ₂ O	Impurity
λ [W/m °C]	3	1.45	12.5	12	14	42	2*	0.5*	1.6-2.0

*The thermal conductivity of these components slightly varies with the chemical composition - a mean value was therefore used. The range of values for impurity was obtained from the weighted average value, for more details see the text.

Table 2. The thermal conductivity λ [W/m °C] of the components of the marbles and glasses samples, respectively

In general, the utility of bounds gives a narrower range to predict a good match to the real value of the effective conductivity only if the conductivities of the components are not too different in magnitude. It is possible to evaluate λ_{eff} using the EMT model which is dependent on the existence of statistically homogeneous medium surrounding inclusions of different phases. As it was stated in sec 2.1, in this model, it is assumed that the phases with λ_i 's are distributed in such a way that the material can be considered isotropic and homogeneous, then according to EMT, the effective conductivity $\lambda_{\text{eff(EMT)}}$ is determined self consistently from the formula:

$$\sum_i P_i \left(\frac{\lambda_{\text{eff(EMT)}} - \lambda_i}{\lambda_{\text{eff(EMT)}} + \lambda_i} \right) = 0 \quad (4)$$

This formula has been derived from the solution to the problem of dilute spherical inclusions of one phase embedded in a matrix of a second phase (Noh et al. 1991). However, equation (4) designates a symmetric representation to all phases without singling out a certain phase as dilute. The solution of this equation will have a physical meaning under certain imposed conditions such as considering $\lambda_{\text{eff(EMT)}}$ as continues function of the volume fraction P.

Table 3 shows the values of the calculated thermal conductivities at room temperature using effective medium theory (EMT), Hashin & Shtrikman, and Wiener bounds. The calculated values indicates that there are four out of the seven samples namely; P3, P5, P6, and P7 do not satisfy the theoretical necessary rule imposed by the parallel and serial bounds. According to this rule and by definition, the measured values should be within the limits of the parallel and serial bounds. This can be seen from the obtained values of the parallel and serial bounds.. In other words, these samples they will not satisfy the validity condition (homogeneity condition) and therefore, have been exempted from our discussion regarding the models validities. The average deviations of $\lambda_{\text{eff(EMT)}}$ from the measured values of these four samples are within 28-29%. Therefore, for the samples P3, P5, P6, and P7 the mismatch between the theoretical models and the measured values should not be attributed to models validities. The most proper reason is that the distribution of phases within these samples is not uniform within the composite matrix.

Further work should be done to investigate the uniformity of different phases within these samples in order to estimate the values of λ 's and draw any affirmative results. However,

for the other three samples namely; P1, P2 and P4 the average deviations of $\lambda_{eff(EMT)}$ from λ_{meas} did not exceeding 3%. It is also obvious that, the mean value of HS bounds $\lambda_{eff(HS)}$ for these samples is rather good estimation to the measured (λ_{meas}) value at room temperature; The best approximation in this context, were obtained from the effective medium approach represented by the solution of equation (4). The self-consistent solutions of this equation for the three samples yielded nine roots. Each solution resulted in only one positive root that has a physical meaning. The deviation of $\lambda_{eff(EMT)}$ from the experimental values is less than 3.3%. Thus, of all of the calculated approximations, the EMT method appears to be the best for estimating the effective value of thermal conductivity only for samples that satisfy the homogeneity condition.

sample	The number of components	Parallel & Serial Wiener bounds		H S mean of U & L bounds	The E. M. T Model	Measured Value
	n	λ_P	λ_S	$\lambda_{eff(HS)}$	$\lambda_{eff(EMT)}$	λ_{meas}
P1	9	3.50	2.32	2.94	2.93	2.85
P2	9	3.40	2.56	3.02	2.97	2.97
P3	9	2.59	2.12	2.38	2.36	3.32
P4	9	2.46	1.96	2.21	2.19	2.27
P5	9	2.61	2.14	2.39	2.39	3.32
P6	8	2.56	2.06	2.33	2.31	5.00
P7	6	8.66	2.00	2.56	3.12	1.50

Table 3. The estimated λ 's [W/m °C] calculated using the corresponding models

At this point, it should be noted that for these ceramic composites there are some constrains imposed upon using these models such as limiting the number of samples, limiting the analysis to room temperature data, and neglecting the anisotropy of the thermal conductivity of the samples or the components that constitute the samples. In these circumstances such constrains may introduce a degree of uncertainty in the results. However, regards of all that, it seems that assuming isotropic and homogeneous conditions for ceramic samples, may still lead to a good agreement between the values of the measured and calculated thermal conductivities of such heterogeneous samples. Furthermore, in this attempt, the purpose of applied the models for these samples, just to examine the possibilities of determining the bounds and/or estimating the effective thermal conductivity of a multi-phase composite system given the volume fractions and the conductivities of the components.

In some cases where structure phase morphology (interlayer structure) details are very important an accurate knowledge of the thermal conductivity of composites could only be obtained experimentally. The experimental measurements can then be used as a probe to monitor microstructure changes.

Finally, these theoretical models can only be utilized provided the investigated samples satisfy the necessary condition that the measured values for the conductivity should be within the limits of the Wigner bounds. According to our calculations for the selected samples, the effective medium approach produced calculated values that agreed best with the measured values.

5. Conclusions

Four different models are tested to investigate the problem of determining the bounds and/or estimating the effective thermal conductivity (λ_{eff}) of composite (multiphase) systems given the volume fractions, the conductivities of the components and porosity (for woods). Three of the tested models namely; the effective medium theory (EMT), Hashin and Shtrikman (HS) bounds, Wiener bounds were applied on the ceramic samples and the fourth weighted bridge-factor model based on Wiener bounds was applied on wood as fiber-based materials or multiphase composites samples.

The effective thermal conductivity of wood at 20 °C slightly increases in both longitudinal and transverse directions. The effect of density and porosity on the thermal conductivity may be attributed to the presence of other scattering mechanisms such as voids, and cell boundaries. It seems that the porosity (conduction through voids) is the dominant influencing factor on the heat conduction in wood. Regarding the anisotropic nature of wood our results indicate that the thermal conductivity in the longitudinal direction (parallel to the grain) is greater than conductivity in the transverse direction. This may be attributed to orientation of the molecular chains within the cell wall. The long-chain linear polymers (cellulose) that comprise the cell wall, are arranged in bundles called microfibrils. These microfibrils are most closely aligned with the longitudinal axis of the cell. Our data in the longitudinal direction at 20 °C lie in the range $\xi = 0.8$ -1.0. The data in the transverse direction are in the range $\xi = 0.5$ -0.7. At 20 °C, our values for both directions are in agreement with literature data.

Although we did not take into account the possibility of anisotropy of thermal conductivity in the ceramic samples, it seems that, assuming isotropic and homogeneous conditions, the thermal conductivity of such samples may be calculated from their contents with rather good accuracy. The deviation of $\lambda_{\text{eff(EMT)}}$ from the experimental values is less than 3.3%. However, the possible consequences of a large anisotropy in their thermal conductivities may not be disregarded and further investigations are needed. These theoretical models can only be utilized provided that the investigated samples satisfy the necessary condition that the measured values for the conductivity should be within the limits of the parallel and serial bounds. According to our investigations for the selected samples, the effective conductivity can be used as a probe to monitor microstructure changes in both morphological and mineralogical aspects. An extended version of this work is planned to be published in the University of Sharjah international press.

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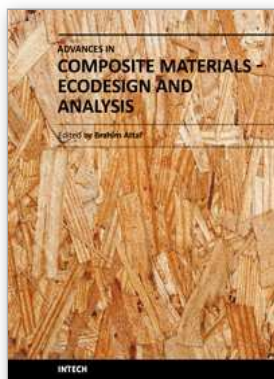
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By adopting the principles of sustainable design and cleaner production, this important book opens a new challenge in the world of composite materials and explores the achieved advancements of specialists in their respective areas of research and innovation. Contributions coming from both spaces of academia and industry were so diversified that the 28 chapters composing the book have been grouped into the following main parts: sustainable materials and ecodesign aspects, composite materials and curing processes, modelling and testing, strength of adhesive joints, characterization and thermal behaviour, all of which provides an invaluable overview of this fascinating subject area. Results achieved from theoretical, numerical and experimental investigations can help designers, manufacturers and suppliers involved with high-tech composite materials to boost competitiveness and innovation productivity.

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