NEW BOUNDS FOR THERMAL CONDUCTIVITY OF RECYCLED POLYMERIC MATRIX COMPOSITES

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Abstract. *Polymeric matrix composites (PMC) are used in engineering applications as they present low density and high strength. However, they are not used in large–scale applications due to their high cost. The large variety of thermoplastic matrices allow us to experiment different types of resin combinations creating the so–called melt-blended matrices. In this model, a binary combination of thermoplastics is used to form a fully recycled melt-blended matrix. The HDPE/PET matrix is assumed to be composed by PET spheroidal inclusions diluted in a HDPE substrate, and the resulting matrix is used to form a unidirectional laminated polymeric matrix composite. Due to chemical reactions involved during the matrices combination process and between the resulting matrix and the E-glass fibers, the weak interface fiber/matrix condition must be considered. To model the thermal-elastic behavior of HDPE/PET-E glass fiber composites a double step homogenization procedure is proposed. The concentric spheres model is applied to obtain the melt-blended matrix effective properties, and to describe the overall composite behavior the composite assemblage model under weak interface condition is considered. The new expression proposed for the effective transverse thermal conductivity is based on the analogy between shear loading and conductivity. The numerical simulations are compared against analytical models, Hashin and Rosen, and the representative volume element approach with good agreement.*

Key words:Bound for Thermal Conductivity, Laminated Polymeric Matrix Composites, Recycled composites

1. NOMENCLATURE

a, b fiber and matrix radii
D_n, D_s, D_t material spring constants
G_A axial shear modulus
$\bar{q}$ heat flux
t interface thickness
ΔT temperature variation
$[u_n]$, $[u_s]$, $[u_t]$ interface displacement jumps
v_f, v_i, v_m fiber, interface and melt-blended matrix volume fractions
γ number of recyclings
μ thermal conductivity
ψ mutation ratio
σ stress
Subscripts:
A, T axial and transverse directions
f, i fiber and interface
$-i$, $+i$ interface's borders to fiber and melt-blended matrix
M, $m1$, $m2$ melt-blended, inclusion and suspension matrices
n, s, t mutual orthogonal system of axis
Superscript:
* effective property

2. INTRODUCTION

Polymeric matrix composites are attractive to engineering applications due to their light weight and high strength. However, their cost is still prohibitive for most of the emergent countries. The use of melt-blended matrices, formed by two immiscible polymers, is an alternative solution for cost reduction of polymeric matrix composites. Such idea is growing in research centers and industries thanks to the large variety of polymeric matrices – in special thermoplastics – and their low cost, e.g. polyethylene terephtalate (PET) has an approximate cost of 12 cents/kg while high density polyethylene (HDPE) costs around 8 cents/kilo. Such low cost is due to the large amount of post-consumer plastic waste generated daily in large cities worldwide.

Researchers like Sanches et al. (1997), Avila and de Miranda (1999) and Avila (1999a) have been studying recycled polymeric composites trying to model its thermal-mechanical behavior in order to use them in new engineering applications. In their work, Sanches et al. (1997) studied the thermal degradation of polycarbonate (PC), polybutylene terephtalate (PBT), and its blend (PC/PBT). They are more interested in the thermal-chemical reactions during elevated temperature situations, no attention is given to the analysis of the mechanical properties. Avila (1999a), however, tries to model the mechanical properties of recycled polymeric matrix laminated composites (PP/PET-E glass). His work is focused on the fiber/matrix interface condition. When recycled polymeric matrix composites are considered , it is important to study the interface fiber/matrix adhesion due to chemical reactions between the melt-blended matrix and the fibers. To be able to model the combined effects of thermal and mechanical loadings, the thermal-elastic properties must be studied and analyzed.

This paper is concerned about the thermal-elastic analysis of melt-blended polymeric

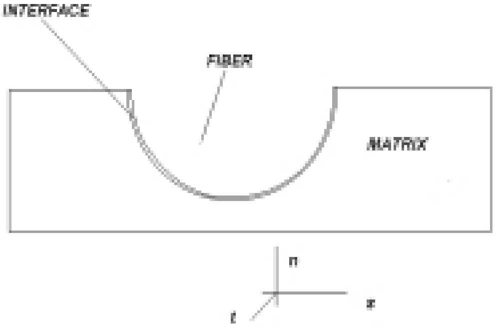

Figure 1: Interface region

matrix composites where the weak interface fiber/matrix condition is applied. Special emphasis is given to the effective thermal conductivity of such class of materials. To do this type of analysis, a double step homogenization procedure is proposed.

3. DOUBLE STEP HOMOGENIZATION PROCEDURE – S^2HP

The double step homogenization procedure is composed of two individual homogenization procedures. They are applied in a such way that there is a one-way influence from the pre-homogenization procedure (also called primary homogenization) into the post-homogenization (sometimes called secondary homogenization). As we are dealing with recycled composites where the weak/imperfect interface is considered, it should be worth to establish the differences between perfect and imperfect interface conditions. Hashin (1990) states that for a perfect interface condition it is assumed that tractions and displacements are continuous across the interface. To be able to model the imperfect/weak interface situation it is assumed that normal and tangential interface displacement jumps are each proportional to their associate traction components as in Hashin (1992). Thus, with reference to Fig. 1,

$$\sigma_{nn}^m = \sigma_{nn}^f = D_n\,[u_n] \tag{1}$$

$$\sigma_{ns}^m = \sigma_{ns}^f = D_s\,[u_s] \tag{2}$$

$$\sigma_{nt}^m = \sigma_{nt}^f = D_t\,[u_t] \tag{3}$$

The spring constants are material parameters which have the dimension of stress divided by their length. Hashin (1990) calls these constants the interface parameters. Infinite values of these parameters are equivalent to perfect bonding, where the displacement jumps go to zero.

3.1 Pre-Homogenization - Concentric Spheres Model

On this phase of the S^2HP, the melt-blended matrix is assumed to be formed by spheroidal inclusions in a suspension solution. It is also assumed that the two matrices are perfected bonded. Therefore, there is no interface between the two blended matrices. The mechanical properties of such blend can be estimated using the models developed by Hashin and Shtrikman (1963) and Christensen and Lo (1979), in which the idea of con-

Figure 2: Melt-blended matrix microstructure

centric spheres is presented, are applied. By analyzing Figure 2, it is possible to conclude that the actual microstructure can be represented by the concentric spheres model. The choice of the concentric spheres array is due to the possibility of applying association in series of thermal conductivity for the two blended matrices (Incropera & Dewitt, 1996) .

To be able to use the thermal conductivity association in series proposed by Incropera & Dewitt (1996), some preliminary assumptions must be made. It is assumed that the overall melt-blended matrix is isotropic and its components are also isotropic. The melt-blended matrix effective thermal conductivity is given by

$$\mu_M^* = \psi_1 \mu_{m1} v_{m1} + \psi_2 \mu_{m2} v_{m2} \tag{4}$$

The mutation ratio is a linear functional defined by Avila (1999b) that artificially change the material properties due to the recycling process. For the PET, for example, Avila (1999c) proposed the following 5th order polynomial expression:

$$\psi = -0.009\gamma^5 + 0.1146\gamma^4 - 0.5494\gamma^3 + 1.221\gamma^2 - 1.1672\gamma + 1 \tag{5}$$

It should be mentioned that the following expression must be applied to the melt-blended components.

$$\frac{v_{m1}}{v_m} + \frac{v_{m2}}{v_m} = 1 \tag{6}$$

3.2 Post-Homogenization - Composite Cylinder Assemblage Model

The post-homogenization procedure is based on a modified version of the composite cylinder assemblage model where a weak interface is considered (Hashin, 1990;Avila, 1999a). In the present model the interface properties are calculated using either the rule of mixtures or the modified rule of mixtures (Tsai & Hahn, 1980). The overall composite behavior is considered transversely isotropic due to the composite configuration (unidirectional). In this case, it is observed that the inclusion of the weak interface fiber/matrix causes no significant influence into the axial thermal conductivity. Considering a random array of fibers, a representative volume element can be isolated, and the effective axial thermal conductivity can be written as,

$$\mu_A^* = \mu_M^* v_m + \psi_f \mu_f v_f + \mu_i v_i \tag{7}$$

where

$$v_f = \left(\frac{a}{b}\right)^2 \tag{8}$$

$$v_m = \frac{b^2 - (a+t)^2}{b^2} \tag{9}$$

$$v_i = \frac{(a+t)^2 - a^2}{b^2} \tag{10}$$

We will assume that fibers are always virgin materials due to the problems involved in their recycling process. Therefore, for us ψ_f is equal to one.

The only unknown variable in Eqs. (7) is the interface's thermal conductivity. By using the analogy between the shear loading and the thermal conductivity proposed by Springer and Tsai (1967), the interface's thermal conductivity can be expressed as

$$\frac{1}{\mu_i} = \frac{1}{v_f + \eta v_m}\left(\frac{v_f}{\psi_f \mu_f} + \eta v_m \frac{1}{\mu_M^*}\right) \tag{11}$$

where

$$\eta = \frac{1}{4\left(1 - v_m\right)}\left(3 - 4v_m + \frac{\mu_M^*}{\psi_f \mu_f}\right) \tag{12}$$

An alternative way of computing the interface's thermal conductivity is given by rule of mixtures (Tsai & Hahn, 1980).

We now have to derive the effective transverse thermal conductivity (μ_T^*) for a two phase composite under weak interface fiber/matrix condition. To do so we use the analogy between the axial shearing and the transverse thermal conductivity. Hashin (1979) observes that the governing differential equations for the axial shearing and the transverse thermal conductivity have the same form, including the boundary conditions. He concludes that there is a mathematical equivalence of the following quantities

$$G_A \longleftrightarrow \mu_T \tag{13}$$

Based on this complete equivalence, the following expression is proposed for modeling the effective transverse thermal conductivity under weak interface fiber/matrix condition. This new expression can be written as

$$\mu_T^* = \mu_M^* + \frac{v_f}{\frac{1}{\mu_e - \mu_M^*} + \frac{v_m}{2\mu_M^*}} \tag{14}$$

where

$$\mu_e = \frac{\psi_f \mu_f}{1 + \frac{\psi_f \mu_f}{\mu_i (a/t)}} \tag{15}$$

Although Hashin (1979) had pointed out the mathematical equivalence between the shear loading and the conductivity he never proposed an expression for modeling thermal

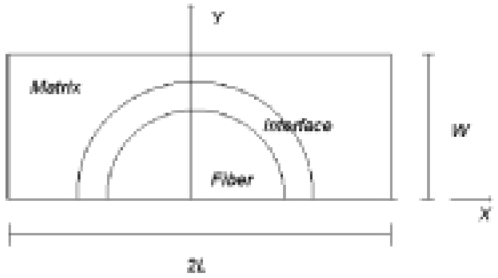

Figure 3: Representative Volume Element

conductivity where the weak interface fiber/matrix was considered. We are going one step further not only by using Hashin's idea and proposing an expression for μ_T, but also by assuming that the interface's thermal conductivity can be modeled by the modified rule of mixtures.

Expressions (7) and (14) represent the axial and the transverse thermal conductivity under weak interface condition for a transversely isotropic composite which complete the double step homogenization procedure. It is important to mention that the thermal conductivity expressions derived by Hashin (1979) for a transversely isotropic unidirectional composites under perfect interface condition are considered in this paper for benchmark purposes.

4. REPRESENTATIVE VOLUME ELEMENT APPROACH - RVE

The representative volume element approach is a numerical technique which isolates a composite's unit cell. Such unit cell represents the composite's microstructure smallest portion which is repeated over the entire composite. Then, after the imposition of specific based boundary conditions and the use of the energy conservation laws it is possible to compute its effective properties. According to Springer and Tsai (1967) and the author's conclusions, it is feasible to assume that the effective axial thermal conductivity can be expressed by Eqs. (7) with accuracy. Therefore, the attention will be focus on the effective transverse thermal conductivity.

The RVE proposed is a variation of the one applied by Springer and Tsai (1967) for computing μ_T^*. In current model, the interface fiber/matrix is introduced (see Fig. 3), and the following boundary conditions are applied,

$$\mu \frac{\partial T}{\partial y} = 0 \quad for \quad y = 0 \quad and \quad y = W \tag{16}$$

$$T = T_{-L} \quad T = T_{+L} \quad at \quad x = \pm L \tag{17}$$

There is no thermal resistance between any phase or the interface. This assumption is

Material	μ [W/m-K]
HDPE	0.5
PET	0.2
E-glass	1.0

Table 1: Material properties

Model Applied	μ [W/m-K]
Rule of Mixtures	0.628
Modified Rule of Mixtures	0.562

Table 2: Interface Thermal conductivity

equivalent to

$$\left|\mu_f \frac{\partial T}{\partial n}\right|_f = \left|\mu_i \frac{\partial T}{\partial n}\right|_{-i} \tag{18}$$

$$\left|\mu_m \frac{\partial T}{\partial n}\right|_m = \left|\mu_i \frac{\partial T}{\partial n}\right|_{+i} \tag{19}$$

Here n represents the normal direction to the considered border. Once the boundary conditions are applied the heat flux ($\bar{q}$) can be computed, and the composite effective transverse thermal conductivity can be defined as

$$\mu_T^* = \frac{2L\bar{q}}{\Delta T} \tag{20}$$

Expression (20) will provide equivalent values of those given by Eqs. (14).

5. NUMERICAL SIMULATIONS

To illustrate the double homogenization procedure, let's consider a fully recycled polymeric matrix laminate composite. It is assumed that PET spheroidal inclusions are diluted in a HDPE matrix. The perfect interface condition is applied to the melt-blended matrix and the weak interface fiber/melt-blended matrix is assumed. Once the HDPE/PET melt-blended matrix is homogenized, the resulting matrix is applied to E-glass unidirectional fibers. The relative fiber/matrix volume fraction is defined as 0.40/0.60, respectively. The ratio HDPE/PET volume fraction is 0.60/0.40. It means that 60% of the melt-blended matrix is composed of HDPE and the remaining is PET. The material properties are from Jones (1994) and they are listed on Table 1. The interface properties evaluated based on the rule of mixtures and the modified rule of mixtures are shown in Table 2.

To be able to analyze how the interface fiber/matrix influences on the overall composite effective properties, two sets of interface conditions are studied. In the first set, the interface properties are calculated by the rule of mixtures. Three different interfaces are considered: perfect interface (interface thickness/fiber diameter equals to zero), and two distinct interface conditions $t/a = 0.1$ and $t/a = 0.02$. In the second set of results, the interface properties are based on the modified rule of mixtures. The same interface

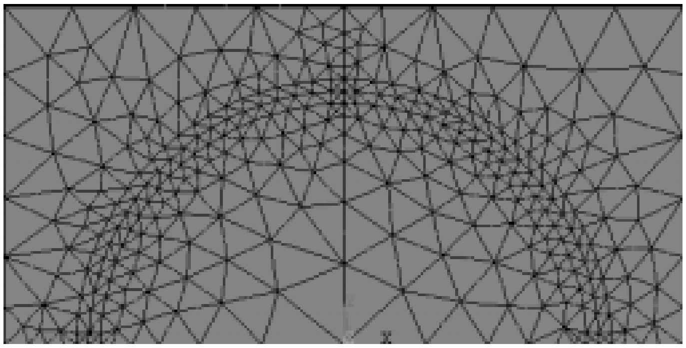

Figure 4: Finite Element Mesh

Interface	S^2HP	RVE	Rosen	Hashin
	μ_A^* [W/m-K]	μ_A^* [W/m-K]	μ_A^* [W/m-K]	μ_A^* [W/m-K]
t/a=0.0	0.7768	0.7768	0.7768	0.7768
t/a=0.1	0.7765	0.7765	–	–
t/a=0.02	0.7768	0.7768	–	–

Table 3: μ_A^* described by the rule of mixtures

conditions are studied. The results from the S^2HP are compared against the RVE approach simulations.

The RVE approach is implemented by using the finite element commercial package NASTRAN. In this case, the six noded triangular elements are used to generate the finite element mesh. The RVE mesh is shown in Fig. 4, and it is composed of 1261 nodes and 608 elements.

The results summarized in Tables 3 though 6 are numerical predictions based on S^2HP, the equations prposed by Rosen (1970), the Hashin's (1979) lower and upper bounds and the RVE approach for a temperature variation of 60 ^{o}C. It should be mentioned that the material properties are considered constant for such class of temperature variation. The maximum temperature is such that no thermal-chemical degradation is observed. In a preliminary analysis the temperature variation of 60 ^{o}C may be seen unrealistic, however this value was chosen to avoid numerical problems, e.g. catastrophic cancellation (Carnahan et al., 1969) on the heat flux calculation due to the RVE real size.

It seems that the interface model has direct influence into the overall composite behavior. As the interface parameter (t/a) approaches to zero, which is equivalent to perfect bonding, the results from the S^2HP are closer to the Hashin's 1979 results. For the t/a=0.0 the relative error, considering the Hashin's lower bound as a benchmark, for μ_T^* is around 1.3% for the RVE approach by using the rule of mixture or the modified rule of mixture. The results from the S^2HP approach are practically the same as Hashin's lower bounds and the data from Rosen (1970). When the interface is considered the relative difference between the RVE and the S^2HP approaches are around 3% when the rule of mixutures is applied, and around 0.5% when the modified rule of mixtures is considered. One possible source of uncertainty on the RVE approach is the effective heat flux value.

Interface	S^2HP	RVE	Rosen	Hashin	
	μ_T^* [W/m-K]	μ_T^* [W/m-K]	μ_T^* [W/m-K]	$\mu_{T(+)}^*$ [W/m-K]	$\mu_{T(-)}^*$ [W/m-K]
t/a=0.0	0.7540	0.7442	0.7542	0.8670	0.7540
t/a=0.1	0.7153	0.6938	–	–	–
t/a=0.02	0.7511	0.7286	–	–	–

Table 4: μ_T^* described by the rule of mixtures

Interface	S^2HP	RVE	Rosen	Hashin
	μ_A^* [W/m-K]	μ_A^* [W/m-K]	μ_A^* [W/m-K]	μ_A^* [W/m-K]
t/a=0.0	0.7372	0.7372	0.7370	0.7371
t/a=0.1	0.7713	0.7713	–	–
t/a=0.02	0.7757	0.7757	–	–

Table 5: μ_A^* described by the modified rule of mixtures

Interface	S^2HP	RVE	Rosen	Hashin	
	μ_T^* [W/m-K]	μ_T^* [W/m-K]	μ_T^* [W/m-K]	$\mu_{T(+)}^*$ [W/m-K]	$\mu_{T(-)}^*$ [W/m-K]
t/a=0.0	0.7041	0.7010	0.7042	0.8096	0.7041
t/a=0.1	0.7106	0.7075	–	–	–
t/a=0.02	0.7499	0.7469	–	–	–

Table 6: μ_T^* described by the modified rule of mixtures

In our case, we compute the heat flux on the nodal points at RVE center line, and for each region (fiber, matrix, and interface) the average heat flux is computed. Then, the effective heat flux on the center line can be evaluated by adding each regions average heat flux. As the heat flux was calculated using a piecewise function its value is only an approximation. A possible solution is the use of a more refined mesh.

One interesting observation when analyzing the new expression (Eqs. 14) is that the model used to describe the interface property - the thermal conductivity - has direct influence on the calculation of the effective transverse thermal conductivity.

6. CLOSING REMARKS

The effective thermal conductivity for transversely isotropic recycled polymeric matrix composites are calculated based on the double step homogenization procedure and the representative volume element approach. For the perfect bonding condition, the double step homogenization procedure and the RVE approach give approximately the same results. When the weak interface fiber/matrix condition, is applied there is an average difference between the two methods of calculation on μ_T of 0.5% when the modified rule of mixtures is used to describe the interface's properties, and 3% when the rule of mixtures is used as an interface governing equation. It is the author's conclusion that the modified rule of mixtures describes with more accuracy the interface's properties. The new equation proposed to describe the effective transverse thermal conductivity based on the analogy between the shear loading and the conductivity seems to be adequate. However, more work (numerical and experimental) must be done to get a more accurate interface governing equation and to validate the new expression proposed for the effective transverse thermal conductivity.

7. ACKNOWLEDGMENTS

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