MODELLING LAMINATE RECYCLED POLYMERIC MATRIX COMPOSITES

Antonio F. Ávila

Department of Mechanical Engineering, Universidade Federal de Minas Gerais, 6627 Antonio Carlos Avenue – 31270-901 Belo Horizonte – Brazil

SUMMARY: The purpose of this paper is drawing the potential use of fully recycled melt-blended matrices into polymeric matrix composites, and its application for construction of low cost houses in emergent countries. To achieve such goal a two-step homogenization procedure is proposed. The melt-blended matrix is homogenized by applying the concentric spheres model under weak interface condition and the linear functional of transformation. The overall composite effective properties are estimated from the composite cylinder assemblage model under weak interface condition for each lamina. FE simulations of beam problems are performed to evaluate the possible application of recycled PMCs into construction of low cost houses; the results are compared against conventional materials.

KEYWORDS: Computer modeling, recycling, polymeric matrix composites, weak interface condition.

INTRODUCTION

Polymeric matrix composites (PMC) are used into engineering applications as they present low density and high strength. However, they are not used into large-scale applications due to their high cost. The large variety of thermoplastic matrices allows us to experiment different types of resin combinations creating the so-called melt-blended matrices. The objective is not only cost reduction but also increase specifics properties such as strength. One low cost source of such type of resins is recycled thermoplastics. Post-consumer polyethylene terephthalate (PET), for example, has a cost of five cents/pound while the polystyrene (PS), high and low density polyethylene (HDPE, LDPE), and polypropylene (PP) have an even lower cost. Their cost range from three to one and half cents/pound. Such low cost is due to the large amount of post-consumer plastic waste generated daily on large cities worldwide. A city in an emergent country with a population of three million inhabitants, for example, produces each day around 400 ton of plastic waste. Countries like the U.S. has already begun to search for solutions by funding research on the recycling field [1]. Our purpose is to develop new technologies into the polymeric waste recycling field so that we can apply these new technologies into the construction of low cost houses in emerging world countries privileging a social-environmental approach.

TWO STEP HOMOGENIZATION PROCEDURE

The major problems on the melt-blended PMC are the chemical reactions between the combined matrices, and the resulting matrix and the fibers. To be able to model such phenomena, a two-step homogenization procedure is proposed. This homogenization

procedure is based on phenomenological observations and it is applied only for a group of melt-blended matrices - binary blends made of recycled materials. Such resulting melt-blended matrix is then applied to unidirectional fibers to get laminated PMCs.

Binary blends have been studied by Acierno and Di Maio [2] with regard to the possible recycling uses. They are more interested in the manufacturing/re-manufacturing process in special how the extrusion process is affected by the polymer granulometry and the phases viscosities. Their approach is essentially experimentalist, and they do not consider the existence of a third phase, e.g. a copolymer or a compatibilizer. Effects of different types of compatibilizers for recycling blends as in Park et al.[3] will not be considered into this paper. The reason is the virtual impossibility of modeling such type of chemical reactions by the micromechanical approach.

Many researchers as Vaccaro et al.[4], Teh et al.[5], study blends microstructures and their influence on the mechanical properties. They show that for most of immisible thermoplastics the resulting array can be assumed as spheroid inclusions in a suspension (see Figure 1). The idea of a spherical domain is also applied by Lyngaae-Jørgensen & Utracki [6] for calculation of dual phase continuity in polymer blends. However, their attention is focused on the percolation phenomena and its influence on phase continuity.

Considering such type of microstructure configuration it is feasible, to assume the concentric spheres model [7,8] for computing the effective properties. Kolarik [9] proposes a scheme to predict the Young's modulus and the yield (or tensile) strength of binary blends. However, he considers a perfect interface adhesion. Such assumption can not be applied when recycled materials are involved. According to Nicolais et al.[10], there is an interface formation between the matrices and the effective material properties are highly affected by the interface conditions. To be able do model such interfacial phenomena, it is assumed the weak interface condition proposed by Hashin [11]. Besides the weak interface condition a functional of transformation is also applied due to the material properties changes during the recycling process. According to Mancini et al.[12], the mechanical properties of recycled thermoplastics are highly dependent of the number of recycles suffered by the material. Therefore, for the first step of the homogenization procedure, the linear functional of transformation must be incorporated into the concentric spheres model. This procedure is called pre-homogenization. Once this step is completed the overall composite effective properties are obtained on variation of the Composite Cylinder Assemblage model (CCA) under weak interface condition proposed by Ávila [13]. This procedure is so-called post-homogenization procedure.

Pre-homogenization procedure

The pre-homogenization procedure proposed is based on phenomenological observations. The objective here is to obtain the melt-blended matrix effective properties of a binary system. As we are dealing with fully recycled thermoplastics, it is needed to estimate how the material properties change after each recycling process. To achieve such goal, it is proposed a transformation functional that can artificially change the mechanical properties of a virgin material. This functional is highly dependent on the material properties and on the manufacturing/re-manufacturing process. Such linear functional will be called mutation ratio due to the material changes during the recycling process. Figure 2 shows an example of the mutation ratio for a specific thermoplastic, PET. A 5th order polynomial approximation is used to represent this linear functional.

$$\psi = -0.009\gamma^5 + 0.1146\gamma^4 - 0.5494\gamma^3 + 1.2211\gamma^2 - 1.1672\gamma + 1 \qquad (1)$$

where ψ is the mutation ratio, γ the number of recycling, and the R^2 parameter for this curve fitting is equal to unity.

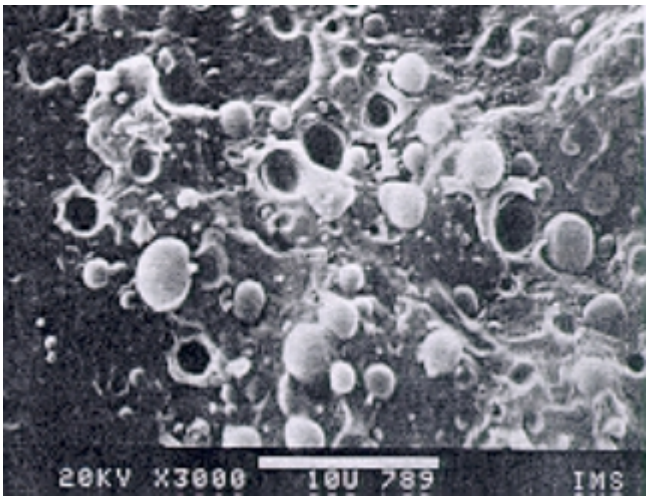

Figure1: Microstructure of PP/PET blend [4]

The mutation ratio is a linear functional and follows the same properties as cited in Szabó and Bab□ska [14]. The physical interpretation of the mutation ratio is the rate of changing of the mechanical properties, e.g. strength, during the recycling process. The number of recycling processes are material dependent, and it is a usual practice among to manufactures/re-manufactures consider that the material is not able of recycling anymore when a significant decrease on strength from one recycling process to another is observed (see reference 15 for more details). In our case, we will postulate that any material with mutation ratio below 0.4 will be considered useless for engineering applications.

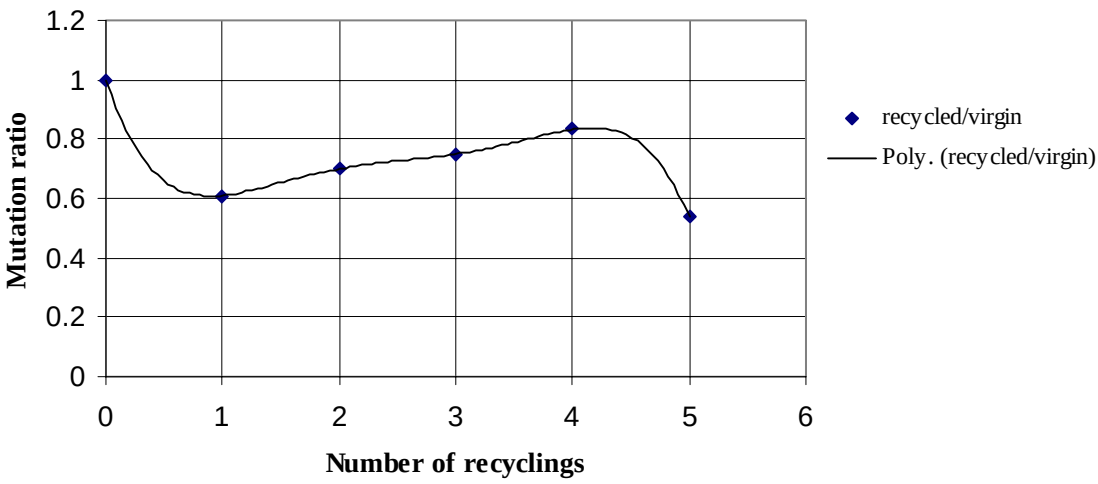

Figure 2: PET mutation ratio

It is considered that each phase of the melt-blended matrix is isotropic, and the inclusions have the spherical shape. The overall composite behavior is assumed to be isotropic. By

assuming such hypotheses, the melt-blended matrix effective bulk modulus – lower and upper bounds - can be described by:

$$K^*_{m(+)} = \psi_1 K_1 v_1 + \frac{\psi_2 K_2 v_2}{1 + 3\psi_2 / aD_n} \tag{2}$$

$$K^*_{m(-)} = \left(\frac{v_1}{\psi_1 K_1} + \frac{v_2}{\psi_2 K_2} + \frac{3v_2}{aD_n} \right)^{-1} \tag{3}$$

where K is the phase bulk modulus, G represents the each phase shear modulus; v_1 and v_2 define the suspension and the inclusion volume fractions, respectively. The lower bound is denoted by the minus sign between parenthesis, and upper bounds by plus sign also between parenthesis. The effective properties are identified by the superscript *.

According to Hashin [11], the variables D_t, D_n, and D_s are the interface parameters on the n, s, t system of coordinates. Such parameters are spring-type constants acting at the interface region. For the case of isotropic interface, the spring-type constants are defined by:

$$D_n = \frac{\psi_3 (K_3 + 1.33 G_3)}{t} \tag{4}$$

and

$$D_s = D_t = \frac{\psi_3 G_3}{t} \tag{5}$$

where *3* indicates interfacial properties and *t* the interface thickness. It is assumed that the interface properties follow the modified rule of mixtures [16]. However, the mutation ratio is presumed to be described by the properties of a linear functional [14].

The effective melt-blended matrix shear modulus upper and lower bounds are described by the following expressions:

$$G^*_{m(+)} = \psi_1 G_1 v_1 + \frac{\psi_2 G_2 v_2}{1 + 5\psi_2 G_2 / (2D_n + 3D_s + 3D_t) a} \tag{6}$$

and

$$G^*_{m(-)} = \left[\frac{v_1}{\psi_1 G_1} + \frac{v_2}{\psi_2 G_2} + \frac{v_2}{a} \left(\frac{0.8(D_s D_t) + 1.2 D_n (D_t + D_s)}{D_n D_s D_t} \right) \right]^{-1} \tag{7}$$

where a is the inclusion radius. An important relation between the interface parameters D_n and D_s is proposed by Hashin[11], and it can be helpful for this case.

$$\frac{D_n}{D_s} = \frac{2(1 - v_3)}{1 - 2v_3} \tag{8}$$

where v_3 is the interface Poisson's ratio. It should be noticed that Eqn.(8) is valid for isotropic interfaces only.

By using the theory of elasticity relations [16] the Young's modulus and Poisson's ratio can be calculated. Once these two bounds are calculated, the effective properties of the melt-blended matrix are completely defined and the pre-homogenization procedure is over.

Post-homogenization procedure

Once the melt-blended matrix effective properties are defined, the resulting matrix is applied to virgin unidirectional fibers to obtain laminated PMC. Virgin fibers are used in our case due to the lack of information concerning the performance of recycled fibers. Eriksson et al. [17] point out that during the recycling process the fibers may suffer severe damage as a result of too intensive processing or excessive use of reground material. The cumulative effect of regrinding and remodeling can cause a significant reduction in most strength parameters. However, a final conclusion about such relationship is not completed formulate yet.

It is important to mention that we are dealing with two sets of interfaces. The first one occurs on the pre-homogenization procedure and it is matrix/matrix interface. The second is a fiber/matrix interface, and it appears on the post-homogenization procedure.

The overall composite effective properties are calculated by applying the mutation ratio to each composite constituents and using the variation of the composite cylinder assemblage model proposed by Avila [13]. Recalling the linear functional properties [14] the melt-blended matrix mutation ratio can be calculated as:

$$\psi_m^* = v_1\psi_1 + v_2\psi_2 \tag{9}$$

where the effective mutation ratio of the melt-blended matrix is designed by ψ^*_m. As we are considering only virgin fibers, the effective mutation ratio for the fibers is equal to the unity.

The entire composite is assumed to be transversely isotropic, and due to this assumption, only five constants are needed to describe the composite behavior. The new expression for the composite effective bulk modulus is given by,

$$K^* = \psi_m^* k_m^* + \frac{v_f}{\frac{1}{\psi_m^*}\left(\frac{1}{k_e/\psi_m^* - k_m^*} + \frac{v_m}{\left(k_m^* + G_m^*\right)}\right)} \tag{10}$$

where

$$k_e = \frac{k_f}{1 + \frac{2k_f h}{b\psi_i\left(K_i + G_i\right)}} \tag{11}$$

where the subscript *i* represent the fiber/matrix interface, *h* the interface thickness, *b* the fiber diameter, and the subscripts *f* and *m* the fiber and the melt-blended matrix properties, respectively.

The effective axial Young's modulus and the axial Poisson's ratio expressions have the same shape as in Hashin [18] or Avila [13], and for conciseness will not be shown. However, the effective axial shear modulus is defined by the following new expression:

$$G_A^* = \psi_m^* G_m^* + \frac{v_f}{\frac{b\psi_i G_i + G_f h}{\psi_i G_f G_i - \psi_m^*\left(bG_i + hG_f\right)} + \frac{v_m}{2\psi_m^* G_m^*}} \tag{12}$$

Following Avila [13], the effective transverse shear modulus must be calculated by applying the generalized self consistent scheme (GSCS). By applying the GSCS we obtain a 8 by 8 determinant equation as,

$$
\begin{bmatrix}
1 & -1 & 1 & 1 & 1 & 1 & 0 & 0 \\
1 & 1 & \frac{2+\eta_m}{1-\eta_m} & 1 & \frac{\eta_m}{1+\eta_m} & -1 & 0 & 0 \\
g^* & 3g^* & 0 & 1 & -\frac{2}{1+\eta_m} & -3 & 0 & 0 \\
g^* & -3g^* & \frac{3}{1-\eta_m} & 1 & \frac{1}{1+\eta_m} & 3 & 0 & 0 \\
0 & 0 & 0 & 1 & -\frac{2}{(1+\eta_m)v_f} & -\frac{3}{v_f^{\,2}} & 0 & -g \\
0 & 0 & \frac{3v_f}{1-\eta_m} & 1 & \frac{1}{(1+\eta_m)v_f} & \frac{3}{v_f^{\,2}} & -\frac{3gv_f}{1-\eta_f} & -g \\
0 & 0 & v_f & 1 & \frac{1}{v_f} & \frac{1}{v_f^{\,2}} & -v_f & -(p+1) \\
0 & 0 & \frac{(2+\eta_m)v_f}{1-\eta_m} & 1 & \frac{\eta_m}{(1+\eta_m)v_f} & -\frac{1}{v_f^{\,2}} & -\frac{(3q+2+\eta_f)}{1-\eta_f} & -(q+1)
\end{bmatrix} = 0 \qquad (13)
$$

where η represents ratio between the phase transverse shear modulus and the bulk modulus, g is ratio between the fiber and the matrix transverse shear moduli. The other constants are defined as,

$$
g^* = \frac{G_T^*}{\psi_m^* G_m^*} \qquad p = \frac{G_f h(1-2v_i)}{\psi_i b(1-v_i)} \qquad q = \frac{2hG_f}{\psi_i b G_i} \qquad (14)
$$

It is important to notice that Eqn. (14) is for the particular case of isotropic interface. Hashin [18] points out that the determinant of Eqn. (13) is a quadratic expression in g^*.

The effective transverse Young's modulus and transverse Poisson's ratio can be obtained by the transverse isotropy relations as cited in Jones [16]. For conciseness, these expressions will not be shown in this paper. Once all effective properties are completely described by a set of algebraic equations the post-homogenization procedure is over.

INTERFACE EFFECTS ON THE EFFECTIVE PROPERTIES

A laminated $[\pm 45]_s$ PET/HDPE-E glass transversely isotropic composite is proposed, and the interface effects on the overall composite behavior is studied. In our case, a group of HDPE spherical inclusions, with an average diameter of 5 μm, is diluted into a PET suspension. The volume fraction ratio between the suspension and the inclusion is 80/20. The E-glass fibers volume fraction is 0.30 and its average diameter is 10μm. The fibers and the HDPE inclusions are considered virgin materials, the PET suspension is a recycled material. The material properties listed in Table 1 are from Cheremisinoff and Cheremisinoff [19]. The reader must notice that the material properties shown in Table 1 are for virgin materials. The transformation functional must be applied to artificially change the mechanical properties of the recycled materials.

Figure 3 shows the influence of the interface condition on the effective moduli. As it is expected the weak interface condition causes a decrease on the overall effective moduli. However, there is an increase on the bulk modulus due to the weak interface condition. The

bulk modulus behavior can be explained by the use of the modified rule of mixtures for the interface. Figure 4 shows the Poisson's ratio behavior for the perfect and weak interface conditions. It is also observed an inversion on the transverse Poisson's ratio behavior as a consequence of the bulk modulus inverse behavior. It is important to emphasize that the axial Young's modulus presents a very straightforward behavior and it is not shown in this paper for conciseness. For the perfect interface condition the suspension matrix variation is dominant, i.e. the effective moduli behavior follows the same pattern that one of the suspension matrix. When the weak interface condition is considered there is a change in this pattern due to the interface's influence.

Material	E [GPa]	K [GPa]	G [GPa]	ν
PET	3.10	2.58	1.19	0.30
HDPE	0.84	0.70	0.32	0.30
E-glass	72.40	43.10	29.67	0.22

Table 1: Material properties

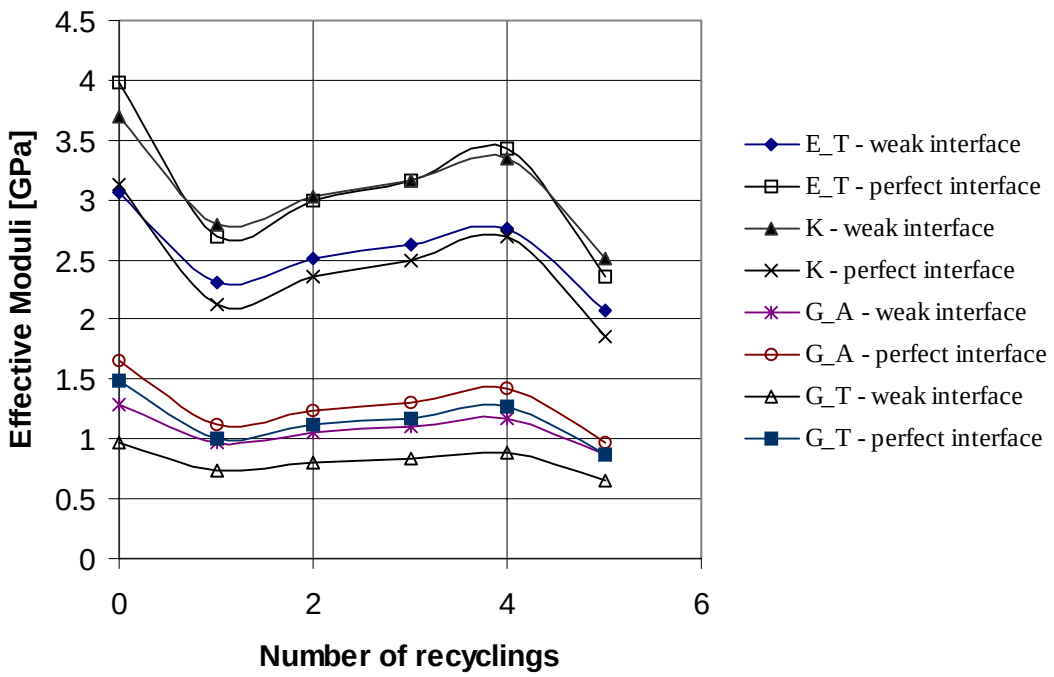

Figure 3:Results: weak x perfect interface condition

The influence of the interface condition on the overall effective moduli is also studied. To be able to study how the two different interfaces, matrix/matrix and fiber/matrix, affect the overall composite effective moduli, it is defined two interface parameters: the first one is the ratio between the inclusion radius and the matrix/matrix interface thickness. This parameter is designated by the Greek letter ϕ. The second interface parameter is the ratio between the fiber radius and the interface fiber/matrix thickness, and it is denoted by ξ.

In this analysis ϕ is changing while ξ is kept fixed. The results are shown on Figures 5 and 6. As ϕ goes to infinity, the results are close to the perfect interface condition, as expected. It seems that there is a direct influence of ϕ on the overall composite effective moduli.

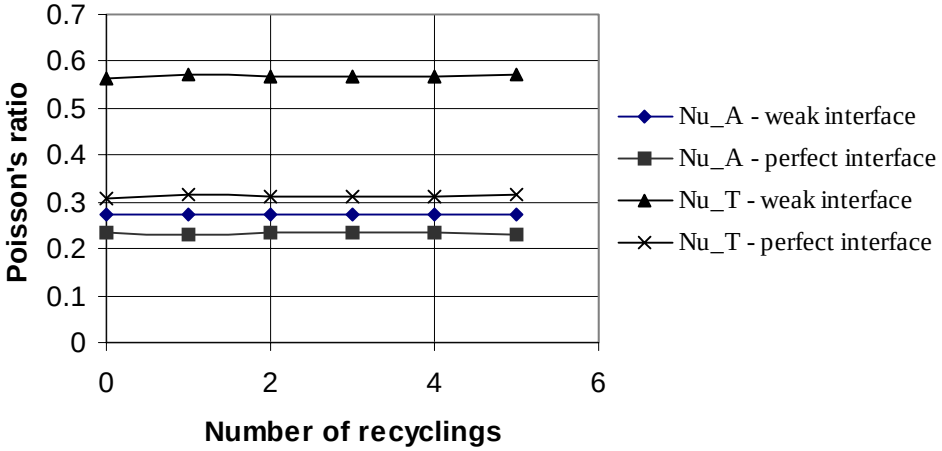

Figure 4: Poisson's ratio – weak x perfect interface condition

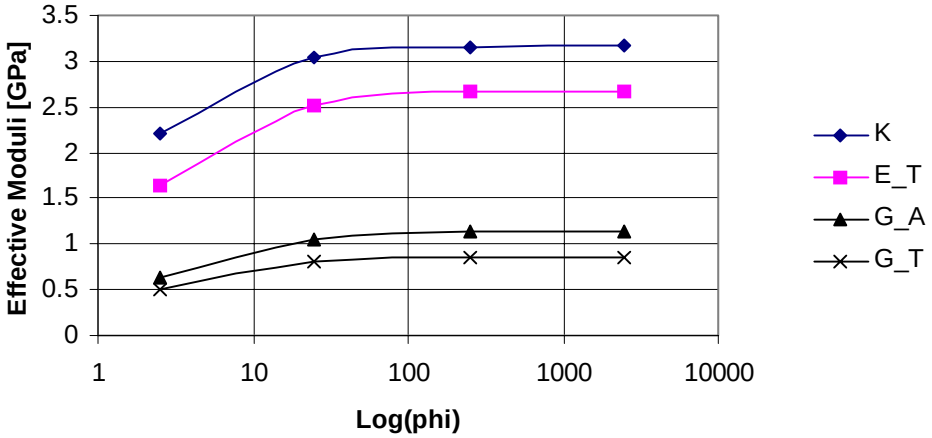

Figure 5: Interface matrix/matrix influence on the effective moduli

The influence of the interface fiber/matrix on the overall composite effective moduli is also studied. For this case the interface parameter ϕ is kept constant while ξ is changing. The results are shown on Figures 7 and 8. The axial shear modulus is practically constant with the interface fiber/matrix changes. This behavior could be caused by the way that the interface properties are calculated. For the axial shear modulus the modified rule of mixtures is close to

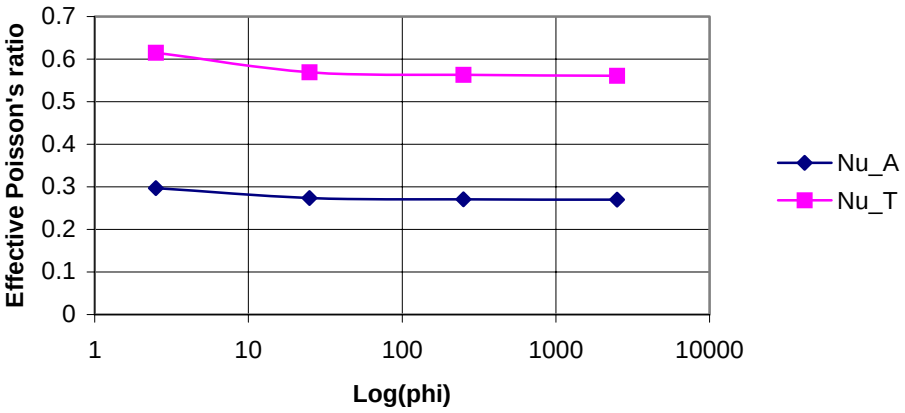

Figure 6: Interface matrix/matrix influence on the Poisson's ratios

the rule of mixtures. The same fact is observed on the axial Poisson's ratio. One possible explanation for this behavior is the cancellation of such effects on the ν_A calculation. It is significant to mention that the elasticity theory is used for the ν_A calculation.

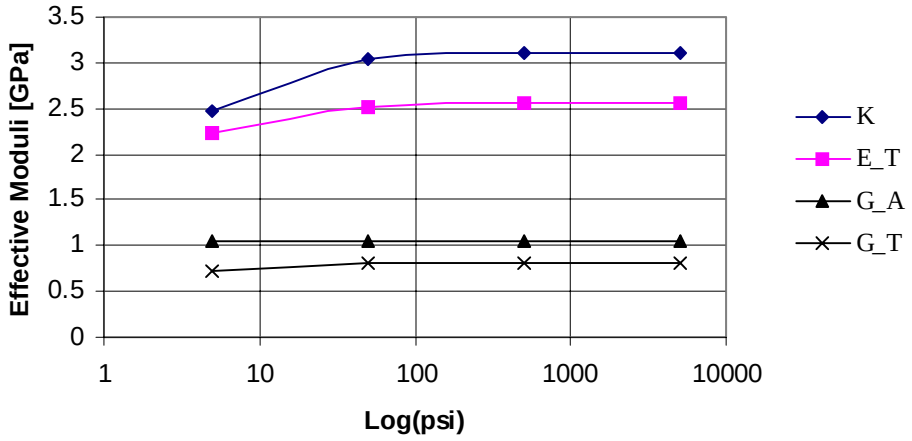

Figure7: Interface fiber/matrix influence on the effective moduli

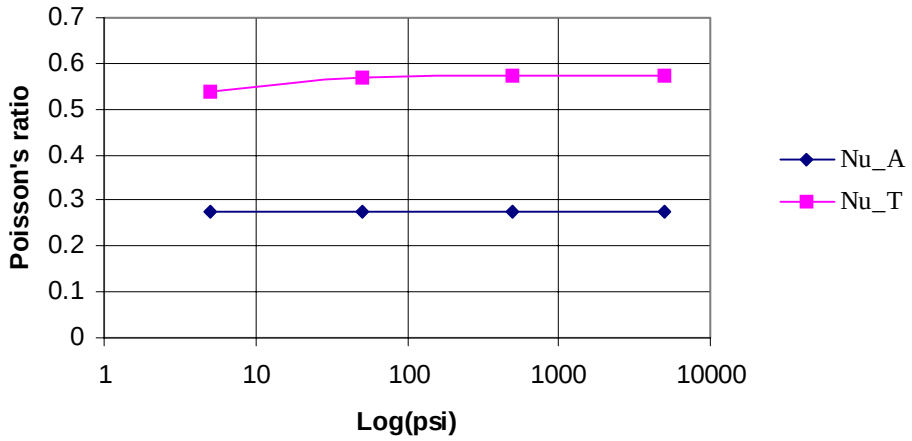

Figure8: Interface fiber/matrix influence on the Poisson's ratios

CONCLUDING REMARKS

The double homogenization procedure proposed is a promising technique to describe the effective moduli of recycled polymeric matrix composites. This procedure is a phenomenological dependent homogenization technique, and it can be applied only for a certain class of PMCs. However, it seems to capture the material properties transformations during the recycling process. The weak interface condition is a crucial feature of the double homogenization procedure. More investigation must be done to describe the interface properties with more accuracy. After all the interface mechanisms of the loading transferring and the interface properties itself is not clear defined yet.

ACKNOWLEDGEMENTS

The author would like to acknowledge the financial support provided by the Universidade Federal de Minas Gerais. Additional support and computer grants were furnished by the Midwest Center for High Performance Computing (CENAPAD- MG/CO).

REFERENCES

1 Burgiel, J., Butcher, W., Halpern, R., Oliver, D., Tangora, T., Tangora, R. W. and Kirk, D. R., "Cost Evaluation of Automated and Manual Post Consumer Plastic Bottle Sorting Systems, *EPA Report EPA/600/R-94/165*,. 1994.

2. Acierno, D. and Di Maio,L., "The Processability of Waste Thermoplastics. An Experimental Approach with a Simple Two-Component System", *J. Polymer Engineering*, Vol. 14, 1995, pp.127-139.

3. Park, H-D., Park, K-O., Cho, W-J., Ha, C-S. and Kwon, S-K, "The Effect of Various Compatibiliser for the Recycling of PET/HDPE Blends", *Polymer Recycling*, Vol. 2, 1996, pp.283-289.

4. Vacaro, E., Dibenedetto, A.T. and Huang, S. J., "Yield Strength of Low-Density Polyethylene-Polypropylene Blends", *J. Appl. Polym. Sci*., Vol. 63, 1997, pp. 275-281.

5. Teh, J. W., Rudin, A., Yuen, S. Y., Keung, J. C. and Park, C, "LLDPE/PP Blends in Tubular Film Extrusion: Recycling of Mixed Films", *J. Plastic Film and Sheeting*, Vol. 10, 1994, pp.288-301.

6. Lyngaee-Jørgensen, J. and. Utracki, L. A., "Dual Phase Continuity in Polymer Blends", *Makromol. Chem., Macromol. Symp*., Vol. 48/49, 1991, pp. 189-209.

7. Christensen, R. M. and Lo, K. H., "Solutions for Effective Shear Properties in Three Phase Sphere and Cylinder Models, *J. Mech. Phys. Solids*, Vol. 27, 1979, pp.315-330, (1979).

8. Ávila, A. F., "Micromechanical Analysis of Melt-Blended Polymeric Matrices Composites under Weak Fiber/Matrix Interface Condition", *Proceedings of VI PACAM - 6th Pan American Congress of Applied Mechanics*, Vol. 7, 1997, pp. 659-662.

9. Kolarik, J. "Simultaneous Prediction of the Modulus and Yield Strength of Binary Polymer Blends", *Polymer Engineering and Science*, Vol. 36, 1996, pp. 2518-2525.

10. Nicolais, L., Amendola, E., Giannotta, G. and Occhiello, C., "Reactive Blending for Improving Interfacial Behavior", *Composite Interfaces*, Vol. 4, 1997, pp. 269-286.

11. Hashin, Z., "Extremun Principles for Elastic Heterogenous Media with Imperfect Interfaces and Their Application to Bounding of Effective Moduli", *J. Mech. Phys. Solids*, Vol. 40, 1992, pp. 767-781.

12. Mancini, S. D., Zanin, M. and. Bezerra, M. N., "Direct injection molding of recycled PET: A study of viability, *Plástico Moderno*, Vol. 1, 1998, pp. 64-70 (in portuguese).

13. Ávila, A. F., "An Integrated Micro/Macro-Mechanical Model for Elastic-Viscoplastic Analysis of Laminated Metal Matrix Composites Including Debonding Effects, *Proceedings of the IV World Congress on Computational Mechanics*, 1998. in CDROM.

14. Szabó, B. and Babuska, I., *Finite Element Analysis*, John Wiley & Sons, New York, 1991.

15. Pearson, W., Plastics, *in The McGraw-Hill Recycling Handbook*, McGraw-Hill, New York, Chapter 14, 1993.

16. Jones, R. M., *Mechanics of Composite Materials*, Taylor & Francis, 2nd edition 1999.

17. Eriksson, P. A., Albertsson, A. C., Boydell, P., Prautzsch, G. and Månson, J.-A. E. "Prediction of Mechanical Properties of Recycled Fiberglass Reinforced Polyamide 66, *Polymer Composites*, Vol. 17, 1996, pp. 830-839.

18. Hashin, Z., "Thermoelastic Properties of Fiber Composites with Imperfect Interface, *Mechanics of Materials*, Vol. 8, 1991, pp. 333-348.

19. Cheremisinoff, N. P. and Cheremisinoff, P. N., *Handbook of Applied Polymer Processing Technology*, Marcell Dekker, New York, 1996.