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ELASTIC PROPERTIES COMPUTATIONAL METHODS OF MULTIPHASE PRE-IMPREGNATED COMPOSITES

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Abstract. *The aim of the paper is to determine the upper and lower limits of the homogenized elastic coefficients for a 27% fibers volume fraction Low-Shrink Sheet Molding Compound (LS-SMC) based on a homogenization method as well as to compute four averages of the Young's and shear moduli of various LS-SMCs with different fibers volume fractions. Since the fibers volume fraction of common LS-SMCs is 27%, to lighten the approach, a 0.27 ellipsoidal inclusion area situated in a square of side 1 is considered. The plane problem will be considered and the homogenized coefficients will be 1 in matrix and 10 in the ellipsoidal inclusion. The structure's periodicity cell of a 27% fibers volume fraction LS-SMC composite material is presented, where the fibers bundle is seen as an ellipsoidal inclusion. The upper limit of the homogenized coefficients can be estimated computing the quadratic mean of these basic elastic properties taking into account the compounds volume fractions. The lower limit of the homogenized elastic coefficients can be estimated computing the harmonic mean of the basic elasticity properties of the isotropic compounds. A comparison between these moduli and experimental data obtained on a Zwick materials testing machine is also presented.*

Keywords: *Prepreg, Homogenization, Averaging, Elastic coefficients.*

1. INTRODUCTION

The objectives of the paper is to compute the upper and lower limits of the homogenized elastic coefficients for a common 27% fibers volume fraction LS-SMC based on a homogenization method described by Ene and Pasa [5] as well as four averaging methods of the Young's and shear moduli of various LS-SMCs with different fibers volume fractions.

The most obvious mechanical model which features a multiphase composite material is a pre-impregnated material, known as prepreg. In the wide range of prepregs the most common used are Sheet and Bulk Molding Compounds. A Low-Shrink Sheet Molding Compound (LS-SMC) is a pre-impregnated material, chemically thickened, manufactured as a continuous mat of chopped glass fibers, resin (known as matrix), filler and additives, from which blanks can be cut and placed into a press for hot press molding. The result of this combination of chemical compounds is a heterogeneous, anisotropic composite material, reinforced with discontinuous fibers [4], [10].

A typical LS-SMC material is composed of the following chemical compounds: calcium carbonate, chopped glass fibers roving, unsaturated polyester resin, low-shrink additive, styrene, different additives, pigmented paste, release agent, magnesium oxide paste, organic peroxide and inhibitors in various volume fractions. The matrix system plays a significant role within a SMC, acting as compounds binder and being "embedded material" for the reinforcement. To decrease the shrinkage during the cure of a LS-SMC prepreg, filler have to be added in order to improve the flow capabilities and the uniform fibers transport in the mold. For materials that contain many compounds, an authentic, general method of dimensioning is difficult to find.

In a succession of hypotheses, some authors tried to describe the elastic properties of SMCs based on ply models and on material compounds [3], [6]. The glass fibers represent the basic element of LS-SMC prepreg reinforcement. The quantity and roving orientation determine, in a decisive manner, the subsequent profile of the LS-SMC structure's properties. The following information is essential for the development of any model to describe the composite materials behavior: the thermo-elastic properties of every single compound and the volume fraction concentration of each compound. Theoretical researches regarding the behavior of heterogeneous materials lead to the elaboration of some homogenization methods that try to replace a heterogeneous media with a homogeneous one [1], [2], [7], [8], [9]. The aim is to obtain a computing model, which takes into account the microstructure or the local heterogeneity of a material.

2. A HOMOGENIZATION METHOD

We consider a domain Ω from R^3 space, in x_i coordinates, domain considered a LS-SMC composite material, in which a so-called replacement matrix (resin and filler) represents the field Y_1 and the reinforcement occupies the field Y_2 seen as a bundle of glass fibers. Let us consider the following equation [5]:

$$f(x) = -\frac{\partial}{\partial x_i}\left[a_{ij}(x)\cdot\frac{\partial u}{\partial x_j}\right]; \quad a_{ij} = a_{ji}, \tag{1}$$

alternatively, written under the equivalent form:

$$f = -\frac{\partial p_i}{\partial x_i}; \quad p_i = a_{ij}\cdot\frac{\partial u}{\partial x_j}. \tag{2}$$

In the case of SMC materials that present a periodic structure containing inclusions, $a_{ij}(x)$ is a function of x. If the period's dimensions are small in comparison with the dimensions of the whole domain then the solution u of the equation (1) can be equal with the solution suitable for a homogenized material, where the coefficients a_{ij} are constants. In the R^3 space of y_i coordinates, a parallelepiped with y_i^0 sides is considered, as well as parallelepipeds obtained by translation $n_i y_i^0$ (n_i integer) in axes directions.

The functions [5]:

$$a_{ij}^{\eta}(x) = a_{ij}\left(\frac{x}{\eta}\right), \tag{3}$$

can be defined, where η is a real, positive parameter. Notice that the functions $a_{ij}(x)$ are ηY-periodical in variable x (ηY being the parallelepiped with ηy_i^0 sides). If the function $f(x)$ is in Ω defined, the problem at limit is [5]:

$$f(x) = -\frac{\partial}{\partial x_i}\left[a_{ij}^{\eta}(x)\cdot\frac{\partial u^{\eta}}{\partial x_j}\right], \quad u^{\eta}\big|_{\partial\Omega} = 0. \tag{4}$$

Similar with equation (2), the vector $\vec{p}^{\eta}$ defines the following elements [5]:

$$p_i^{\eta}(x) = a_{ij}^{\eta}(x)\cdot\frac{\partial u^{\eta}}{\partial x_j}. \tag{5}$$

For the function $u^{\eta}(x)$, an asymptotic development will be looking for, under the following form [5]:

$$u^{\eta}(x) = u^0(x,y) + \eta^1 u^1(x,y) + \eta^2 u^2(x,y) + \ldots; \quad y = \frac{x}{\eta}, \tag{6}$$

where $u^i(x,y)$ are elements, Y-periodical in y variable. The derivatives of the functions $u^i(x,y)$ behave in the following manner [5]:

$$\frac{d}{dx_i} \to \frac{\partial}{\partial x_i} + \frac{1}{\eta}\cdot\frac{\partial}{\partial y_i}. \tag{7}$$

If the values:

$$u^i\left(x, \frac{x}{\eta}\right) \tag{8}$$

are compared in two homologous points P_1 and P_2, homologous through periodicity in neighbor periods, it can be notice that the dependence in x/η is the same and the dependence in x is almost the same since the distance P_1P_2 is small.

Let us consider P_3 a point homologous to P_1 through periodicity, situated far from P_1. The dependence of u^i in y is the same but the dependence in x is very different since P_1 and P_3 are far away. For instance, in the case of two points P_1 and P_4 situated in the same period, the dependence in x is almost the same since P_1 and P_4 are very close, but the dependence in y is very different since P_1 and P_4 are not homologous through periodicity. The function u^{η} depends on the periodic coefficients a_{ij}, on the function $f(x)$ and on the boundary $\partial\Omega$. The development (6) is valid at the inner of the boundary $\partial\Omega$, where the periodic phenomena are prevalent. Using the development (6), the expressions:

$$\frac{\partial u^\eta}{\partial x_i} \tag{9}$$

and p^η are [5]:

$$\frac{\partial u^\eta}{\partial x_i} = \left(\frac{\partial}{\partial x_i} + \frac{1}{\eta}\cdot\frac{\partial}{\partial y_i}\right)\cdot\left(u^0 + \eta\cdot u^1 + \ldots\right) = \frac{\partial u^0}{\partial x_i} + \frac{\partial u^1}{\partial y_i} + \eta\cdot\left(\frac{\partial u^1}{\partial x_i} + \frac{\partial u^2}{\partial y_i}\right) + \ldots, \tag{10}$$

$$p_i^\eta(x) = p_i^0(x,y) + \eta\cdot p_i^1(x,y) + \eta\cdot p_i^2(x,y) + \ldots, \tag{11}$$

where:

$$p_i^0(x,y) = a_{ij}(y)\cdot\left(\frac{\partial u^0}{\partial x_j} + \frac{\partial u^1}{\partial y_j}\right),\quad p_i^1(x,y) = a_{ij}(y)\cdot\left(\frac{\partial u^1}{\partial x_j} + \frac{\partial u^2}{\partial y_j}\right), \ldots \tag{12}$$

represent the homogenized coefficients.

3. APPLICATION FOR A 27% FIBERS VOLUME FRACTION LS-SMC

In the case of a LS-SMC composite material which behaves macroscopically as a homogeneous elastic environment, is important the knowledge of the elastic coefficients. Unfortunately, a precise calculus of the homogenized coefficients can be achieved only in two cases: the one-dimensional case and the case in which the matrix- and inclusion coefficients are functions of only one variable. For a LS-SMC material is preferable to estimate these homogenized coefficients between an upper and a lower limit. Since the fibers volume fraction of common LS-SMCs is 27%, to lighten the calculus, an ellipsoidal inclusion of area 0.27 situated in a square of side 1 is considered. The plane problem will be considered and the homogenized coefficients will be 1 in matrix and 10 in the ellipsoidal inclusion. In fig. 1 the structure's periodicity cell of a LS-SMC composite material is presented, where the fibers bundle is seen as an ellipsoidal inclusion.

Let us consider the function $f(x_1, x_2) = 10$ in inclusion and 1 in matrix. To determine the upper and the lower limit of the homogenized coefficients, first the arithmetic mean as a function of x_2-axis followed by the harmonic mean as a function of x_1-axis must be computed. The lower limit is obtained computing first the harmonic mean as a function of x_1-axis and then the arithmetic mean as a function of x_2-axis. If we denote with $\varphi(x_1)$ the arithmetic mean against x_2-axis of the function $f(x_1, x_2)$, it follows:

$$\varphi(x_1) = \int_{-0.5}^{0.5} f(x_1,x_2)dx_2 = 1,\quad for\quad x_1 \in (-0.5;\quad -0.45)\cup(0.45;\quad 0.5), \tag{13}$$

$$\varphi(x_1) = \int_{-0.5}^{0.5} f(x_1,x_2)dx_2 = 1 + 9.45\sqrt{0.2025 - x_1^2},\quad for\quad x_1 \in (-0.45;\quad 0.45). \tag{14}$$

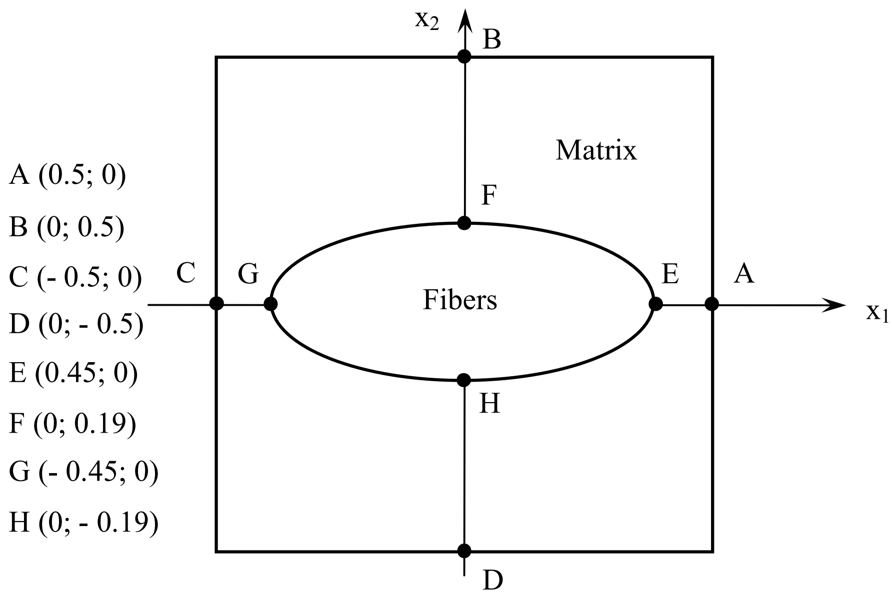

Figure 1. Periodicity cell of a LS-SMC material with 27% fibers volume fraction [10].

The upper limit is obtained computing the harmonic mean of the function $\varphi(x_1)$:

$$a^{+} = \frac{1}{\int_{-0.5}^{0.5} \frac{1}{\varphi(x_1)} dx_1} = \frac{1}{\int_{-0.5}^{-0.45} dx_1 + \int_{-0.45}^{0.45} \frac{dx_1}{1+9.45\sqrt{0.2025 - x_1^2}} + \int_{0.45}^{0.5} dx_1}. \tag{15}$$

To compute the lower limit, we consider $\psi(x_2)$ the harmonic mean of the function $f(x_1, x_2)$ against x_1:

$$\psi(x_2) = \frac{1}{\int_{-0.5}^{0.5} \frac{1}{f(x_1, x_2)} dx_1} = 1, \quad for \quad x_2 \in (-0.5; \ -0.19) \cup (0.19; \ 0.5), \tag{16}$$

$$\psi(x_2) = \frac{1}{\int_{-0.5}^{0.5} \frac{1}{f(x_1, x_2)} dx_1} = \frac{1}{1 - 3.42\sqrt{0.0361 - x_2^2}}, \quad for \quad x_2 \in (-0.19; \ 0.19). \tag{17}$$

The lower limit will be given by the arithmetic mean of the function $\psi(x_2)$:

$$a_{-} = \int_{-0.5}^{0.5} \psi(x_2) dx_2 = \int_{-0.5}^{-0.19} dx_2 + \int_{-0.19}^{0.19} \frac{dx_2}{1 - 3.42\sqrt{0.0361 - x_2^2}} + \int_{0.19}^{0.5} dx_2. \tag{18}$$

Since the ellipsoidal inclusion of the LS-SMC structure may vary angular up to ± 30° against the axes' centre, the upper and lower limits of the homogenized coefficients will vary as a function of the intersection points coordinates of the ellipses, with the axes x_1 and x_2 of the periodicity cell. The LS-SMCs micrographs make obvious this angular variation of the fibers' bundles, the extreme heterogeneity and the layered structure of these materials as well as the glass fibers and fillers distribution. The micrographs show that there are areas between 100 – 200 μm in which the glass fibers are missing and areas where the fibers distribution is very high.

We can consider both resin and filler system as a distinct phase compound which can be called replacement matrix, so that a LS-SMC can be regarded as a two phase compound material. If we denote E_r the basic elastic property of the resin, E_F the basic elastic property of the fibers, E_f the basic elastic property of the filler, φ_r the resin volume fraction, φ_F the fibers volume fraction and φ_f the filler volume fraction, the Young's modulus of the replacement matrix (E_{RM}) can be estimated computing the harmonic mean of the basic elastic properties of the isotropic compounds, as follows:

$$E_{RM} = \frac{2}{\frac{1}{E_r \cdot \varphi_r} + \frac{1}{E_f \cdot \varphi_f}}, \tag{19}$$

so, for the entire composite, the Young's modulus has been computed using the rule of mixtures:

$$E_C = E_F \cdot \varphi_F + E_{RM} \cdot (1 - \varphi_F). \tag{20}$$

If we denote P_M, the basic elastic property of the matrix, P_F and P_f the basic elastic property of the fibers respective the filler, then the upper limit of the homogenized coefficients can be estimated computing the quadratic mean of these basic elastic properties taking into account the compounds volume fractions:

$$A_q = \sqrt{\frac{(P_M \cdot \varphi_M)^2 + (P_F \cdot \varphi_F)^2 + (P_f \cdot \varphi_f)^2}{3}}. \tag{21}$$

The lower limit of the homogenized elastic coefficients can be estimated computing the harmonic mean of the basic elasticity properties of the isotropic compounds:

$$A_h = \frac{3}{\frac{1}{P_M \cdot \varphi_M} + \frac{1}{P_F \cdot \varphi_F} + \frac{1}{P_f \cdot \varphi_f}}. \tag{22}$$

Intermediate limits between the quadratic and harmonic means are given by the arithmetic and geometric means written below:

$$A_a = \frac{P_M \cdot \varphi_M + P_F \cdot \varphi_F + P_f \cdot \varphi_f}{3}, \tag{23}$$

$$A_g = \sqrt[3]{P_M \cdot \varphi_M \cdot P_F \cdot \varphi_F \cdot P_f \cdot \varphi_f}, \tag{24}$$

where P and A can be the Young's modulus or the shear modulus.

4. RESULTS AND DISCUSSION

Typical elasticity properties of the LS-SMC isotropic compounds and the composite structural features for a 27% fibers volume fraction LS-SMC are given as input data in the theoretical approach. For the unsaturated polyester resin, following data have been used: Young's modulus: < 5 GPa; shear modulus: < 4 GPa; volume fraction: 30%. For glass fibers: Young's modulus: < 80 GPa; shear modulus: < 35 GPa; volume fraction: 27%. For filler: Young's modulus: < 55 GPa; shear modulus: < 25 GPa; volume fraction: 43%.

According to equations (19) and (20), the longitudinal elasticity moduli E_{RM} (for the replacement matrix) and E_C (for the entire composite) can be computed. A comparison between these moduli and experimental data obtained on a Zwick materials testing machine is presented in fig. 2.

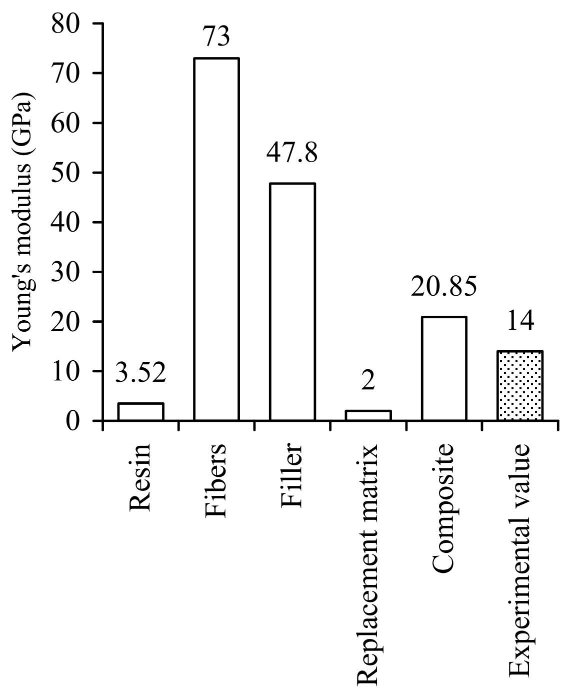

Figure 2. Young's moduli E_{RM} and E_C for a 27% fibers volume fraction SMC material [10].

From fig. 2, it can be noticed that the Young's modulus for the entire composite is closer to the experimental value unlike the Young's modulus for the replacement matrix. This means that the rule of mixtures used in equation (20) give better results than the inverse rule of mixtures presented in equation (19), in which the basic elastic property of the filler and the filler volume fraction can be replaced with fibers Young's modulus and fibers volume fraction, appropriate for a good comparison.

According to equations (15) and (18), the upper and lower limits of the homogenized coefficients for a 27% fibers volume fraction LS-SMC material have been computed and presented in table 1. The results show that the upper limit of the homogenized coefficients decreases with the increase of angular variation of the ellipsoidal inclusion unlike the lower limit which increases with the increase of this angular variation.

Table 1. Upper and lower limits of the homogenized coefficients for a 27% fibers volume fraction LS-SMC material [10]

Angular variation of the ellipsoidal inclusion	Upper limit a^+	Lower limit $a\_$
0°	2.52	0.83
± 15°	2.37	0.851
± 30°	2.17	0.886

Fig. 3 shows the Young's moduli of the isotropic LS-SMC compounds, the upper and lower limits of the homogenized elastic coefficients as well as a comparison with the experimental value.

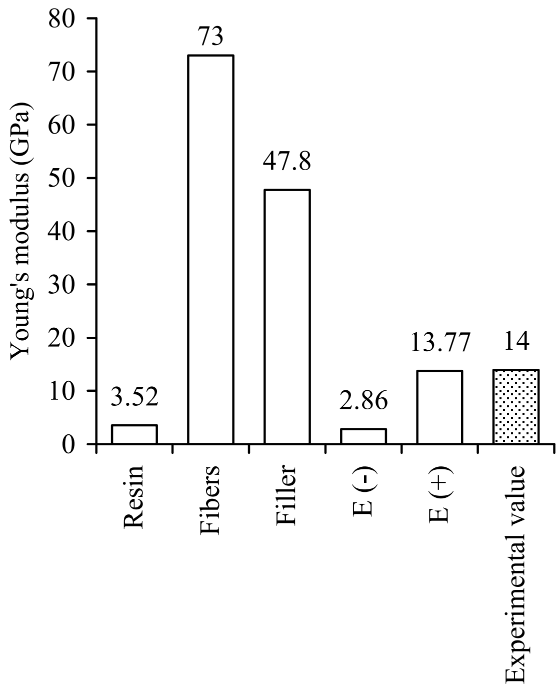

Figure 3. The Young's moduli of the isotropic LS-SMC compounds, the upper (E+) and lower limits (E-) of the homogenized elastic coefficients [10].

Fig. 4 presents the shear moduli of the isotropic LS-SMC compounds, the upper and lower limits of the homogenized elastic coefficients and a comparison with the experimental value obtained on a Zwick materials testing machine.

According to equations (21) – (24), the averaging methods of Young's and shear moduli of various LS-SMCs with different fibers volume fractions present following distributions in figs. 5 and 6. By increasing the fibers volume fraction, the difference has been equally divided between matrix and filler volume fraction. The averaging methods to compute the Young's and shear moduli of various LS-SMCs with different fibers volume fractions show that for a 27% fibers volume fraction LS-SMC, the quadratic and arithmetic means between matrix, fibers and filler Young's moduli respective shear moduli give close values to those determined experimentally.

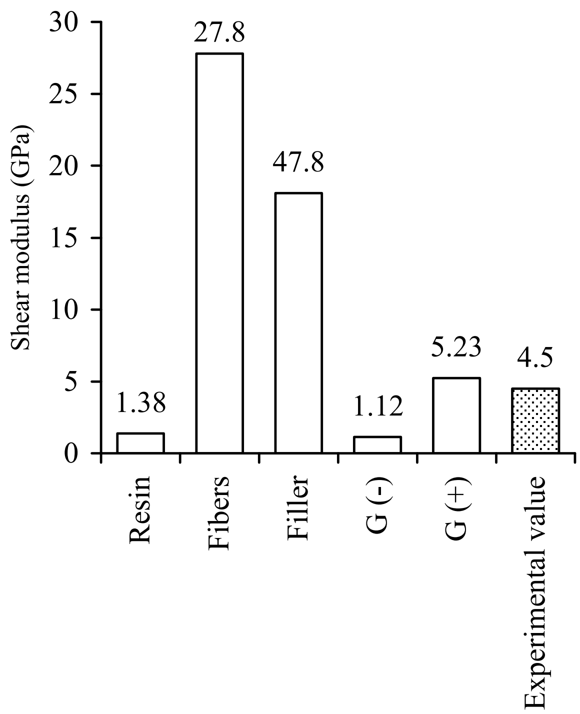

Figure 4. The shear moduli of the isotropic LS-SMC compounds, the upper (G+) and lower limits (G-) of the homogenized elastic coefficients [10].

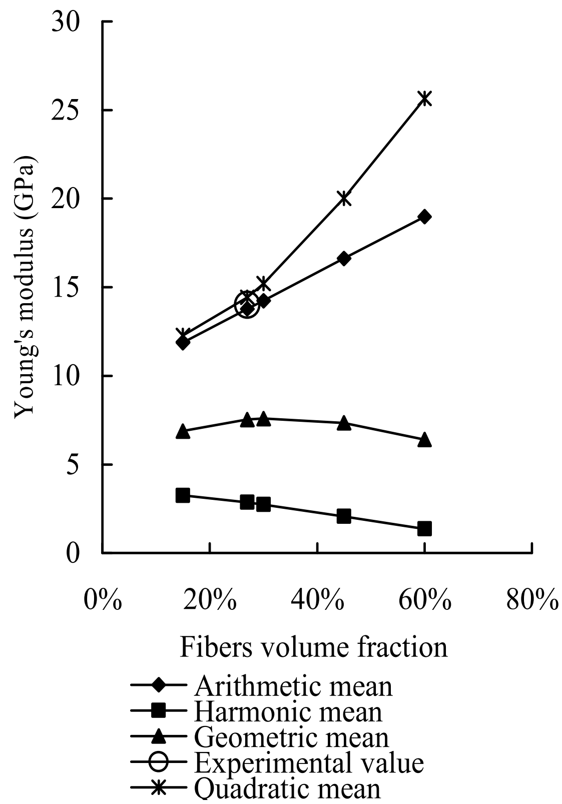

Figure 5. Four averaging methods to compute the Young's moduli of various LS-SMCs with different fibers volume fractions.

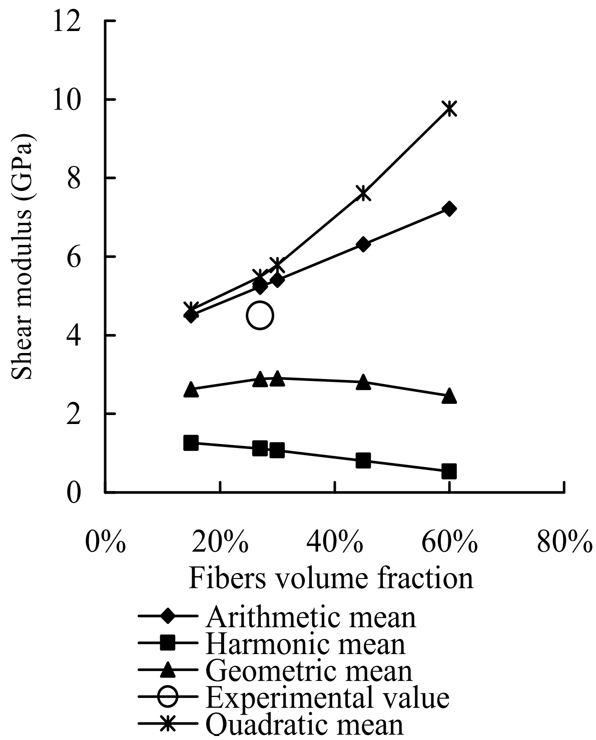

Figure 6. Four averaging methods to compute the shear moduli of various LS-SMCs with different fibers volume fractions.

5. CONCLUSIONS

The presented results suggest that the environmental geometry given through the angular variation of the ellipsoidal domains can lead to different results for same fibers volume fraction. This fact is due to the extreme heterogeneity and anisotropy of these materials. The upper limits of the homogenized elastic coefficients are very close to experimental data, showing that this homogenization method give better results than the computed composite's Young's modulus determined by help of the rule of mixtures. The proposed estimation of the homogenized elastic coefficients of pre-impregnated composite materials like Low-Shrink Sheet Molding Compounds (LS-SMCs) can be extended to determine the elastic properties of any multiphase, heterogeneous and anisotropic composite material.

6. REFERENCES

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