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Elastic moduli of randomly oriented, chopped-fibre composites with filled resin

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An analytical method is developed to determine the effective elastic moduli of a filled resin reinforced by randomly oriented chopped fibres, a three-phase composite such as a filled sheet moulding compound (SMC). The analytical results compare well with experimental results for both filled (glass/calcium carbonate/polyester) and unfilled (glass/polyester) SMC composites. The numerical results also illustrate the important influence of the filler phase on the composite stiffness properties.

1. Introduction

One family of lightweight materials of interest in the automotive industry for vehicle weight reduction is fibre-reinforced plastics (FRP). Among various forms of FRP composites, sheet moulding compound (SMC) appears to be one of the most promising candidates for many potential applications because of its relatively low cost and suitability for high-volume production. There are two kinds of sheet moulding compound: filled SMC (a three-phase composite, i.e. fibre/filler/resin), and unfilled SMC (a two-phase composite, i.e. fibre/resin). Since there are numerous possible compositions of an SMC system (e.g. different types and volume fractions of constituents), it is desirable that the effective material properties of selected composites can be analytically determined so that the material can be tailored for optimal cost, performance, and processing.

In this paper we introduce such an analytical method to determine the effective stiffness properties of three-phase composites by extending theories of two-phase composites. The elastic constants of the filled-matrix phase are calculated according to Hashin's theory [1] on elastic moduli of heterogeneous materials. The elastic constants for the final matrix/fibre composite phase are calculated by Christensen and Waals' [2] expressions for the effective stiffness of randomly oriented fibre composites, in conjunction with the theories of Hill [3, 4] and Hashin [5, 6].

A computer program is also developed to carry out the sequence of computations to determine the effective stiffness properties and density of a composite having specified constituent properties.

Analytical and available experimental results show good agreement. Numerical results illustrate the influence on the effective stiffness properties due to changes of constituent content. The effect of fillers was examined by comparing filled and unfilled SMC.

2. SMC: material compositions

The basic constituents of an SMC system are resin, filler, fibre, and small amounts of additives. The commonly used fibres of automotive-type SMC are coated S-glass filaments combined as rovings and chopped to 25 mm long. The chopped-glass fibres are randomly dispersed in a filled or an unfilled thermosetting resin paste (polyester) to reinforce strength, stiffness, and dimensional stability of the composite. For a filled SMC, a large fraction of low-cost filler (~ 40% by weight) is used to reduce the overall cost of the composite. The most commonly used filler for automotive-type SMC is finely ground calcium carbonate ($CaCO_3$). Other miscellaneous additives, comprising only a very small weight-fraction of the composite, help

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control the chemical reaction, improve the face smoothness, and enhance other desirable characteristics such as fire-resistance. In computing the effective stiffness of a typical SMC-composite, the direct contributions from the additives are ignored and their weights assigned to the resin weight. By doing so, a filled SMC can be identified simply as a three-phase composite (fibre/filler/resin), and an unfilled SMC, a two-phase composite (fibre/resin).

3. Method of analysis

The effective stiffness of a filled composite reinforced by randomly oriented chopped fibres (fibre/filler/resin – a three-phase composite) has not been analysed directly. In this section, we introduce an analysis by extending theories of two-phase composites to determine the effective stiffness properties of such three-phase composites. The basic approach of the proposed method is: the resin and filler are combined to form an "effective" matrix; the resulting matrix is then combined with the chopped fibres to form the final composite.

3.1. Assumptions

We use these basic assumptions in analysing a three-phase composite:

(1) each phase of the constituents is homogeneous and isotropic;

(2) both fibres and filler particles are uniformly dispersed, and the fibres are randomly oriented in the resin;

(3) the fibres lie in the plane of the sheet;

(4) void effects are neglected;

(5) the interfacial bond between the constituents is perfect.

Thus the resin/filler two-phase composite can be regarded as quasi-homogeneous and quasi-isotropic, and the resin/filler/fibre three-phase composite as quasi-homogeneous and, in the plane of the sheet, quasi-isotropic. In other words, we have assumed the existence of "effective" or macroscopically averaged mechanical properties of both phases of the composite. This, of course, is a reasonable approximation only when dealing with effective behaviour of a body containing many fibres or inclusions which are small in comparison to the gross dimensions of the body or region of interest, and when the size of filler particles is small in comparison with the least dimension of the fibres.

The idealized geometric models upon which the theoretical results are based are that of the sphere model [1] for particulate fillers and the cylinder model [2] for chopped fibres. Further, it is assumed that the fibre aspect ratio is sufficiently large so that the end effect of the chopped fibres can be ignored.

3.2. Matrix: resin/filler mixture

Much work has been done to determine the effective stiffness properties of a heterogeneous material with inclusions. The earliest one is perhaps due to Einstein [7]. A rigorous solution based on the theory of elasticity appears to be difficult to obtain. Owing to this reason, bound theorems have been applied to predict the bounds of the elastic moduli of the filled system without considering the exact geometrical distribution of the fillers. Apparently, the first bounds on the Young's modulus of a filled system are due to Paul [8], but their range is too wide to be practically useful.

A closer pair of upper and lower bounds on the bulk and shear moduli of a composite with spherical inclusions was obtained by Hashin [1]. By separately applying constant displacement and constant stress on the boundary of the composite element, the elastic stress fields for both boundary conditions can be obtained from [9]. The strain energy of the composite element is then calculated from the stress fields under both boundary conditions. On the other hand, the strain energy of the composite element can also be expressed in terms of the applied uniform strain or stress and its effective elastic moduli, such as bulk modulus, K, and shear modulus, G. Equating these two expressions of strain energy and using the fact that an admissible strain field is associated with the upper bound, whereas an admissible stress field is associated with the lower bound of the strain energy, one can obtain the values of upper and lower bounds of bulk and shear moduli. In [1], Hashin further proved that these upper and lower bounds of bulk modulus coincide. He also found that there always exists an expression which lies between the upper and lower bounds of the shear modulus. This expression can be regarded as a good approximation of the shear modulus.

When the resin and filler are mixed to form the matrix phase for the final composite, the effective bulk and shear modulus of the (filled) matrix can be obtained based on Hashin's expressions as follows:

$$K_m = K_r + (K_p - K_r) \times \frac{(4G_r + 3K_r)v_p^*}{4G_r + 3K_p + 3(K_r - K_p)v_p^*} \quad (1)$$

$$G_m = G_r\left\{1 + \frac{15(1-\nu_r)(G_p - G_r)G_r v_p^*}{(7-5\nu_r)G_r + 2(4-5\nu_r)[G_p - (G_p - G_r)v_p^*]}\right\} \quad (2)$$

where subscripts m, r, and p refer to matrix, resin, and particulate filler, respectively, ν_r is the Poisson's ratio of the resin and

$$v_p^* = \frac{v_p}{v_p + v_r} \quad (3)$$

with v_p^* being the effective filler volume content of the matrix phase and v_p and v_r being the corresponding volume fractions of the final composite.

We stated earlier that the filled matrix is assumed to be quasi-homogeneous and quasi-isotropic. The elastic modulus and Poisson's ratio, which are the effective stiffness properties needed to represent the filled matrix, thus can be derived as

$$E_m = \frac{9K_m G_m}{3K_m + G_m} \quad (4)$$

$$\nu_m = \frac{3K_m - 2G_m}{2(3K_m + G_m)}. \quad (5)$$

3.3. Composite: fibre/matrix mixture

Once the effective stiffness properties of the filled-matrix phase are known, the three-phase composite can be represented as a fibre/matrix mixture. Although much work has been devoted to analysing the effective moduli of unidirectional and angle-ply reinforced composites, relatively little work has been done to examine these properties of randomly oriented fibre-reinforced composites. The earliest work which treated this type of problem is that by Cox [10], who studied the stiffness and strength properties of paper. In his study, he assumed that the entire load is carried by the fibres alone, and that the elastic moduli of paper is given by the integrated average of the modulus of individual fibres when oriented uniformly from 0 to π. Because the matrix moduli are neglected, Cox's estimates of the elastic moduli are low. Following Cox's idea of integration, others have considered the matrix properties in their derivations. The applicability of most of these results, however, is rather limited. For instance, Nielson and Chen's work [11] is given in numerical form, whereas Halpin and Pagano's [12] predictions are not very good when the fibre volume fraction lies between 0.38 and 0.50. The authors consider the theory of Christensen and Waals [2] to be the most comprehensive among all those published. The two-dimensional results of this theory are summarized below.

The crux of Christensen and Waals' theory is that the stiffness of a two-dimensional, randomly oriented fibre composite can be generated from that of transversely isotropic, unidirectionally aligned fibre composites by considering the fibres as evenly oriented from 0 to π. Adopting Cox's concept of integration, the elastic moduli of a two-dimensional composite are derived from those of a transversely isotropic, unidirectionally aligned fibre composite as

$$P_c = \frac{1}{\pi}\int_0^{\pi} P(\theta)\mathrm{d}\theta \quad (6)$$

where P_c represents the composite moduli, and $P(\theta)$ represents the elastic moduli of an unidirectional composite oriented at an angle θ with respect to the material axis.

A transversely isotropic material has five independent elastic constants. These five elastic constants, in terms of engineering constants, are designated as E_{11}, ν_{12}, G_{23}, G_{12} and K_{23}, where the 2–3 plane is assumed to be the isotropic plane and the 1-direction the fibre direction. As derived by Hill [3, 4], the modulus and contraction ratio for the uniaxial condition are given by:

$$E_{11} = v_f E_f + v_m E_m + \left[4 v_f v_m (\nu_f - \nu_m)^2 \Bigg/ \left(\frac{v_m}{K_f + G_f/3} + \frac{v_f}{K_m + G_m/3} + \frac{1}{G_m}\right)\right] \quad (7)$$

$$\nu_{12} = v_f \nu_f + v_m \nu_m + \left[v_f v_m (\nu_f - \nu_m) \times \left(\frac{1}{K_m + G_m/3} - \frac{1}{K_f + G_f/3}\right) \Bigg/ \left(\frac{v_m}{K_f + G_f/3} + \frac{v_f}{K_m + G_m/3} + \frac{1}{G_m}\right)\right] \quad (8)$$

where the subscript f refers to fibre, and the matrix volume fraction v_m is

$$v_m = v_r + v_p = 1 - v_f. \quad (9)$$

Based on an elaborate variational principle developed by Hashin and Shtrikman [13], Hashin [5, 6] obtained a pair of upper and lower bounds on G_{23}, G_{12} and K_{23}. The lower bound results, recommended for application, are

$$G_{23} = G_m\left[1+\left\{v_f\bigg/\left[\frac{G_m}{G_f-G_m}+\frac{(K_m+\frac{7}{3}G_m)v_m}{2(K_m+\frac{4}{3}G_m)}\right]\right\}\right] \quad (10)$$

$$G_{12} = G_m\frac{G_f(1+v_f)+G_m v_m}{G_f v_m+G_m(1+v_f)} \quad (11)$$

$$K_{23} = K_m+\frac{G_m}{3}+\left\{v_f\bigg/\left[\frac{1}{K_f-K_m+\frac{1}{3}(G_f-G_m)}+\frac{v_m}{K_m+\frac{4}{3}G_m}\right]\right\} \quad (12)$$

Using Hill and Hashin's results given by Equations 7 and 8 and Equations 10 to 12, Christensen and Waals applied the concept of Equation 6 to obtain the effective stiffness properties of a two-dimensional randomly oriented fibre composite as

$$E_c = \frac{1}{u_1}(u_1^2-u_2^2) \quad (13)$$

$$\nu_c = \frac{u_2}{u_1} \quad (14)$$

where the subscript c refers to the final composite and

$$u_1 = \tfrac{3}{8}E_{11}+\frac{G_{12}}{2}+\frac{(3+2\nu_{12}+3\nu_{12}^2)G_{23}K_{23}}{2(G_{23}+K_{23})} \quad (15)$$

$$u_2 = \tfrac{1}{8}E_{11}-\frac{G_{12}}{2}+\frac{(1+6\nu_{12}+\nu_{12}^2)G_{23}K_{23}}{2(G_{23}+K_{23})} \quad (16)$$

with $E_{11}, \nu_{12}, G_{12}, G_{23}$, and K_{23} given by Equations 7, 8 and 10 to 12.

The final composite density can be expressed by

$$\rho_c = (\rho_p-\rho_r)v_p^* v_m+\rho_r v_m+\rho_f v_f \quad (17)$$

with v_p^* and v_m defined in Equations 3 and 9. The basic constituent volume fractions can be related to the corresponding given weight fraction as follows:

$$v_r = \rho_p\rho_f w_r/(\rho_p\rho_f w_r+\rho_f\rho_r w_p+\rho_r\rho_p w_r)$$
$$v_p = \rho_f\rho_r w_p/(\rho_p\rho_f w_r+\rho_f\rho_r w_p+\rho_r\rho_p w_f)$$
$$v_f = 1-v_r-v_p. \quad (18)$$

A computer program has been developed to carry out the sequence of computations. With the aid of the program the effective material properties can be easily obtained for any mixture and for a range of compositions.

The proposed method is validated by comparing analysis and test results in the next section. Those results also show the influence on the effective composite properties resulting from changing the contents of the constituents.

4. Results and discussion

4.1. Data used for numerical results

The general composition of SMC considered in this study is based on a typical SMC system.* The composition of SMC-50† is listed in Table I [14]. In this study we have ignored the specific contributions to the stiffness properties from the additives except fillers. The total weight fractions of the neglected additives (about 3%) are accounted for by adding them to the weight fraction of the resin. By doing so, the SMC-50 is thus represented as a three-phase composite as follows:

resin: polyester $w_r = 0.34$
filler: calcium carbonate $w_p = 0.16$
fibre: glass $w_f = 0.50$.

This composition is used as a baseline to generate other compositions. In particular, the resin weight fraction ($w_r = 0.34$) is kept constant for all filled SMC, and the changes in weight concentration of fibres is offset by equal changes of filler weight concentration. The fibre volume fraction of a filled and an unfilled SMC as functions of fibre weight fraction are shown in Fig. 1. The shaded area shown in Fig. 1 represents the corresponding filler volume fraction of a typical filled SMC. It is

*Note that the composition of a typical SMC can differ slightly from manufacturer to manufacturer.

†Nominal fibre weight fraction of a typical SMC composite is denoted by the number after the letter, e.g. SMC-50 contains 50% chopped fibre by weight.

TABLE I SMC-50 composition

Matrix paste	
resin	100 parts
filler (calcium carbonate)	50 phr
misc. additives:	
inhibitor (benzoquinone):	500 ppm
catalyst (T-butyl perbenzoate);	1 phr
mould release (zinc stearate):	25 phr
thickener (magnesium):	0.5 phr
Compound	
matrix paste	50 parts
glass	50 parts

ppm, parts per million resin.
phr, parts per hundred resin.

important to note that the resin volume fraction of the manufacturer's filled SMC system essentially remains constant (~ 45%) as indicated in Fig. 1. This is done to ensure adequate wetting, and thus good mechanical bonding, of the glass fibres and filler particles. Not more resin is used than is necessary because it costs more than the filler. The material properties of the SMC constituents are listed in Table II.

4.2. Comparison of analytical and experimental results

SMC-type composites usually show appreciable sample-to-sample scatter in their elastic properties. The scatter can be affected in interfacial bonding, fibre distribution and alignment, and voids as discussed earlier, and by fabrication techniques and test methods. Very little complete data for randomly oriented chopped-fibre composites are

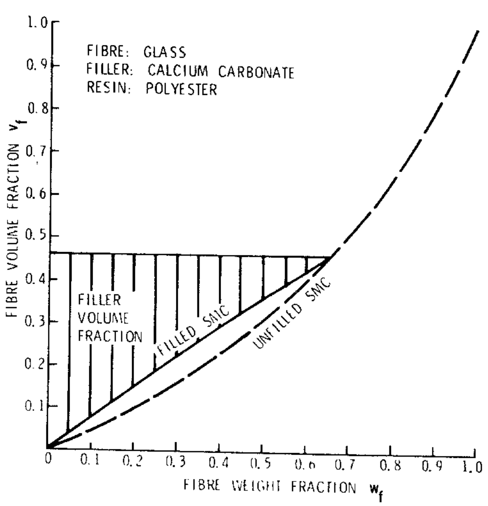

Figure 1 Relationship of fibre weight and volume fractions for a filled and an unfilled SMC.

available. One set of experimental data for which it appears that particular care had been exercised in obtaining the results for a range of glass concentrations is that Manera [15]. These data are shown in Figs. 2 and 3 for the elastic modulus E_c and the Poisson's ratio ν_c of an unfilled SMC, respectively. Shown also in these figures are the analytical results obtained in this study along with Manera's approximate solution and an analysis based on the simple rule of mixtures. Manera used the classical laminate analogy in conjunction with invariants defined by Tsai and Pagano [16] and with Puck's micromechanics formulation [17]. He further introduced some numerical approximations to simplify the formulation. In order to obtain adequate accuracy, it is suggested that equations be used only over the range of 0.1 to 0.4 fibre volume content, and 2 to 4 GPa of the matrix Young's modulus. Manera's tests were carried out on randomly oriented chopped glass fibres (5 cm long) and polyester composites ($E_f = 73$ GPa, $\nu_f = 0.25$, $E_m = 2.25$ GPa, and $\nu_m = 0.4$). Each test data point shown in Figs. 2 and 3 represents the average of ten individual values for flexure tests, five for tensile tests, and five for the Poisson's ratio tests. He also showed that there is no significant difference in the values of elastic modulus obtained by flexure and tensile tests.

As shown in those figures, very good agreement is observed between Manera's experimental results and the analytical results obtained from the method proposed in this study (by letting $v_p = 0$) for both the elastic modulus and the Poisson's ratio. The proposed method also results in better estimates than does Manera's solution for the complete range of fibre concentrations tested. The results also show that solutions based on the rule of mixtures greatly over-estimate the elastic moduli of the composite.

The analytical results of the effective modulus of a filled SMC (glass/calcium carbonate/polyester) versus the fibre weight fraction are compared with

TABLE II Selected material properties of SMC constituents

Constituents	Elastic modulus E (GPa)	Poisson's ratio ν	Specific density ρ
Fibre: glass	72.4	0.22	2.54
Resin: polyester	3.5	0.45	1.12
Filler: calcium carbonate	41.4	0.21	2.70

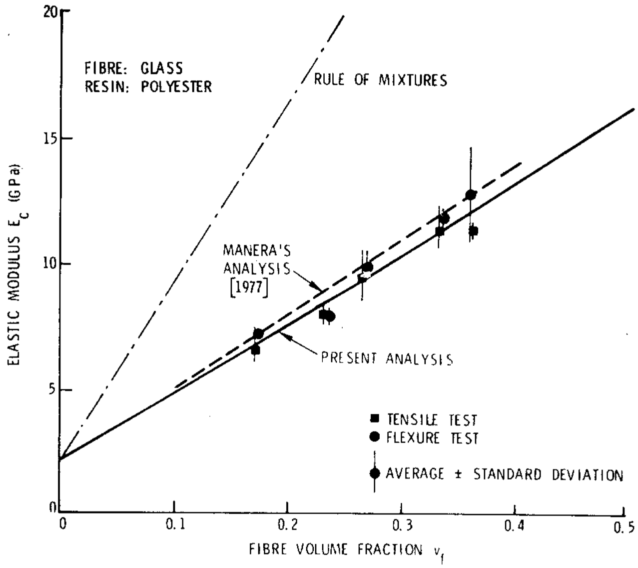

Figure 2 Comparison of analytical and experimental results for elastic modulus of an unfilled SMC.

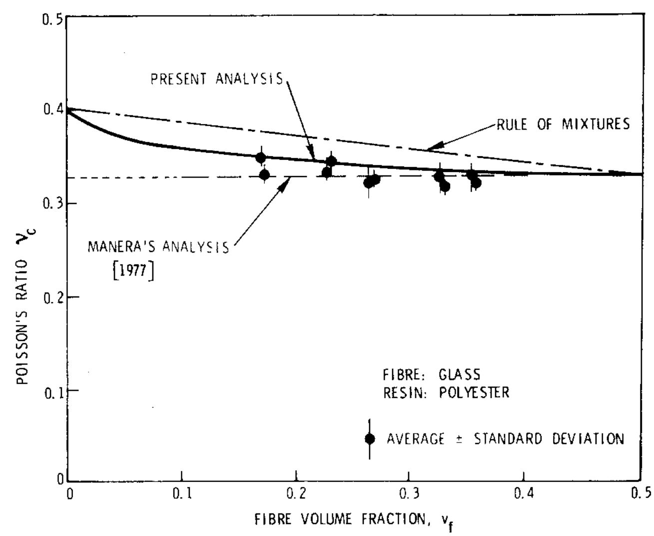

Figure 3 Comparison of analytical and experimental results for Poisson's ratio of an unfilled SMC.

test data [14] as shown in Fig. 4. The test data shown in Fig. 4 represent the mean values of the tensile modulus of coupon specimens cut from 30.5 cm x 45.7 cm (12 in. x 18 in.) plaques 2.5 ± 0.25 mm thick. Half the specimens were loaded normal and half parallel to the major axis of the moulded plaque. The results show that the analytical solution based on proposed methods can be used satisfactorily to represent the effective average elastic modulus for the filled SMC systems.

4.3. Effect of fibre

A typical computer output of the analysis of the (filled) SMC* is shown in Fig. 5. The values E_c, ν_c, G_c, ρ_c, v_f of a filled SMC composite are given for fibre weight fractions ranging from 0 to 0.5. As expected, the higher the glass content, the stiffer the composite. The degree of increase, however, is fairly low. For example, an increase of glass weight content by 50% (from SMC-30 to SMC-45) results in only a 10% increase in the elastic modulus. The effect on G_c and ν_c is even less. It is also interesting to note that the density of the composite, ρ_c, remains nearly constant over the range of glass content studied.

4.4. Effect of filler

The effect of filler on the effective elastic modulus is illustrated in Fig. 6, by comparing filled SMC composites (fibre/resin/filler) and unfilled composites (fibre/resin) of various fibre weight fractions. The effective elastic moduli of the

*As discussed earlier, the resin weight fraction is kept constant for a filled SMC ($w_r = 0.34$).

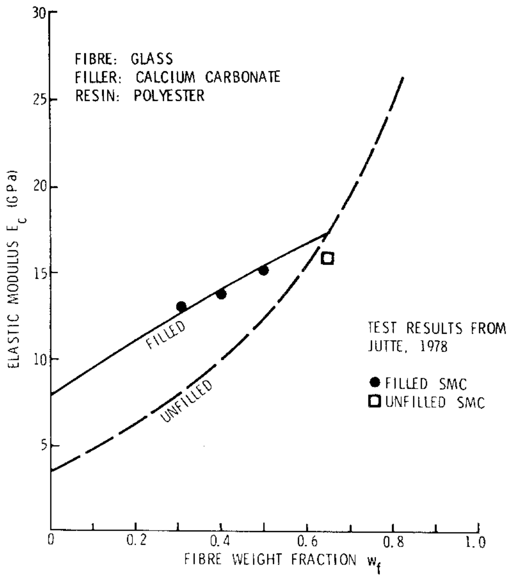

Figure 4 Comparison of analytical and experimental results for filled and unfilled SMC.

corresponding matrix phases alone (resin/filler for the filled SMC and resin for the unfilled SMC) are also shown in the figure. The vertical separation between each corresponding pair of curves (composite and matrix) represents the stiffening contributed by the fibres. The separation between the curves for the unfilled and the filled SMC matrix represents the contribution of fillers to the stiffness of the final composite.

Based on the assumed composition with resin weight fraction being constant ($w_r = 0.34$), the matrix fibre content possible in SMC without degrading the bonding is about 66% by weight, in which case the amount of filler reduces to zero. The results in Fig. 6 (glass/calcium carbonate/polyester) show that, when reinforced by the same amount of glass, filled SMC is always stiffer than unfilled SMC. Hence, the filler phase not only is an important constituent in promoting fibre flow and reducing the overall cost, but also is an important contributor to the final stiffness properties of the composite. For example, in the case of SMC-30, the effective elastic modulus of the filled matrix is more than double that of the unfilled matrix. On the other hand, the added glass fibres (30% by weight) only double the effective elastic modulus from that of the filled matrix.

5. Summary and conclusions

The analysis presented in this paper provides a practical analytical means to determine the effective stiffness properties of randomly oriented chopped-fibre composites with a filled or an unfilled resin system. Good agreement was obtained with experimental results for both filled and unfilled chopped-glass fibre-reinforced SMC.

The following conclusions are based on the results of this study for a typical automotive-type

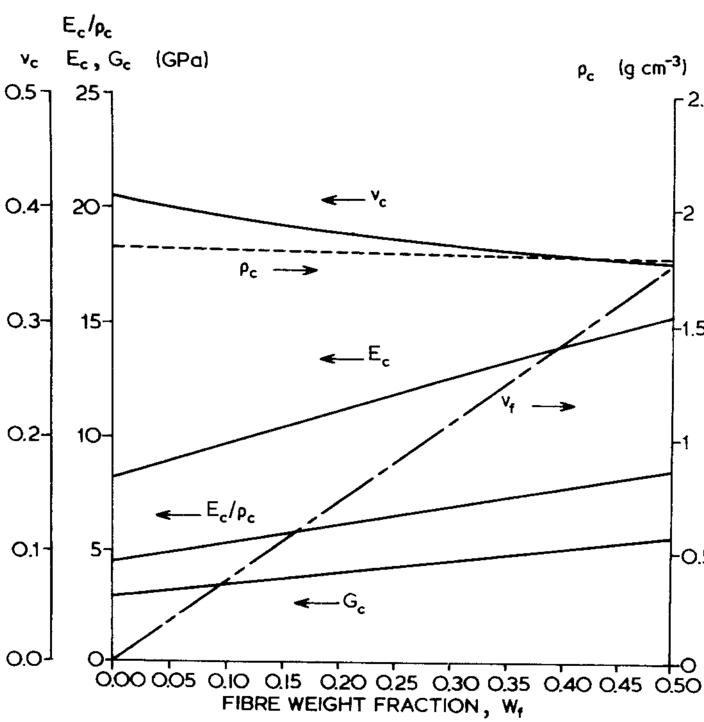

Figure 5 Elastic moduli of a filled (glass/calcium carbonate/polyester) SMC.

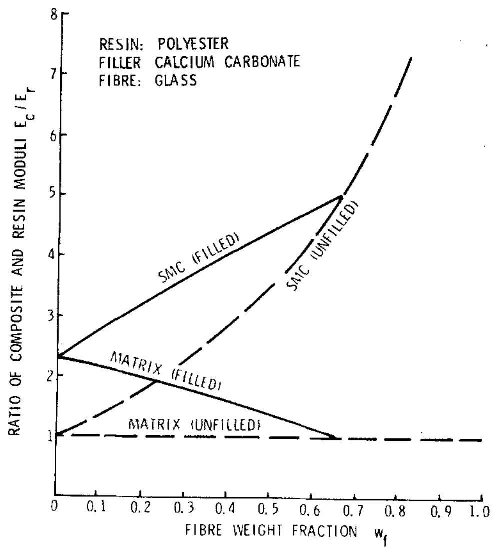

Figure 6 Effect of filler on the elastic modulus of SMC.

SMC composite. The elastic modulus of a filled SMC compared with an unfilled SMC is increased only moderately by increasing fibre content. For instance, a 50% increase in fibre content of a typically filled SMC results in only a 10% gain in elastic modulus of the composite (from SMC-30 to SMC-45); on the other hand, a 30% increase is realized in case of an unfilled SMC. For automotive-type SMC, the filler materials are added for reasons of economy and surface appearance, however, replacing part of the resin with filler also increases the stiffness significantly at the expense of a moderate weight increase.

In applying the proposed method to calculate the effective stiffness of a three-phase composite, one must keep in mind that the imposed assumptions discussed earlier represent idealized conditions for the composite. In reality these assumptions are simply approximations. The facts that the bonding might not be perfect, the fillers might be agglomerated, the fibres might not be uniformly distributed, and there might be voids, all suggest that the estimated results according to the proposed method could be higher than experimentally measured values. The close agreement with the experimental results of the two SMC systems shown in this study, however, suggest that the approximations imposed to simplify the mathematical formulation are propably of secondary importance in influencing the effective stiffness properties of the composites considered.

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