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RESEARCH ARTICLE

Elastic modulus prediction for hybrid polymer composites

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Abstract: To improve on the mechanical properties of polymers in general, the concept of hybrid composites was developed by using two or more different reinforcements in the same matrix, or by using two or more different sizes of the same reinforcement (auto-hybrid composites). In this case, most of the literature results showed that the resulting elastic modulus can be well approximated by the simple rule of mixture (linear additive law) from the tensile modulus of each reinforcement used alone. But is some cases, a positive deviation from this linear approximation was reported up to a point where an optimum composition can give a modulus above the value of both reinforcements used separately. In this work, a simple model is presented to show that positive deviations are possible and the optimum reinforcement ratio is around 25/75 in terms of the lowest/highest reinforcing particle. The model is also compared with literature data where good qualitative agreements are obtained as a first approximation.

Keywords: hybrid polymer composites, elastic modulus, optimum composition

1 Introduction

Composite materials have been around for many years, but new developments are still being made. Although metals, glasses and ceramics have high rigidity and strength, lower production/processing costs and lightweight structures made polymer matrices more interesting. In this case, several types of reinforcement can be used to improve on the mechanical, physical and thermal properties of the matrix. Today, synthetic (man-made) and natural (bio-sourced) reinforcements are available under different sizes (nano-, micro- and milli-meter) and geometries (fibers, spherical, ellipsoidal, platelets, *etc.*). It is known that the two most important parameters controlling the composites properties are particle content and interfacial adhesion, although dispersion/distribution and orientation are also important to optimize the final properties.

Over the years, several techniques have been introduced to improve on the composites overall behavior. One of the most recent one was the concept of hybrid composites where two or more different reinforcements are introduced into the same matrix, each one having its own specific advantages. Even more recently, the concept of auto-hybridization was proposed where a single type of reinforcement is introduced into a matrix, but using different sizes (particle size distribution)[1] . This also led to the development of multi-scale composites by blending reinforcements having sizes varying over a wide range (orders of magnitude)[2] . More information on hybrid composites[3] and composite foams[4] , mainly based on natural reinforcement, can be found in the literature.

One important aspect of hybrid composites is that synergistic effects can be produced between the different reinforcement used leading to properties much higher than expected, especially when using the simple linear rule of hybrid mixtures (RoHM).[5,6] For example, Ramezani Kakroodi et al.[7] combined different concentration of hemp fibers in polypropylene and observed that for a total hemp content of 20% wt., an optimum tensile modulus of 518 MPa was obtained for a 20/80 (weight ratio) of short/long fibers compared to a composite having 100% short fibers (425 MPa) or 100% long fibers (472 MPa). The effect was also seen at 30% wt. total fiber content, but to a lower level. Finally, the same trends were observed for tensile strength and flexural modulus values. Figure 1 presents typical results obtained for polypropylene/hemp auto-hybrid composites. More results on auto-hybrid composites based on different sizes of agave and pine fibers are also available[8] . So the objective of this work is to present a simple model to predict the properties of these complex materials and to determine the optimum composition for a binary mixture

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of particles.

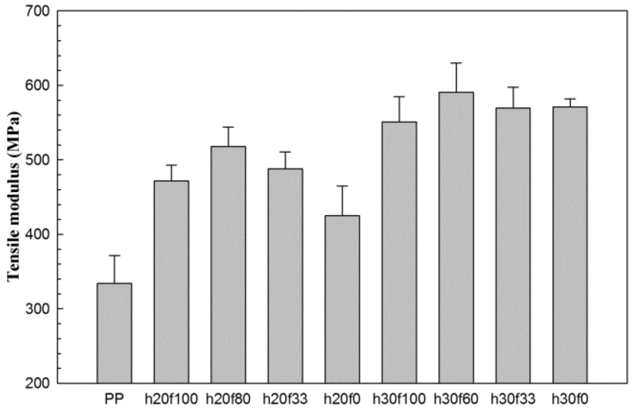

Figure 1. Tensile modulus as a function of composition for hemp fibers (h) in a polypropylene (PP) matrix. The first and second numbers report the total fiber content (wt.%) and the amount of long fibers (%) compared to short ones[7]

2 Modeling

2.1 Literature

Several models have been proposed to predict the mechanical properties of composite materials and a review on the most important ones can be found in Kalaprasad *et al.*[9] . Their main differences (complexity level) is associated to the assumptions (simplifications) made to get the final results. The simplest model is the linear rule of mixture (or Voigt model, E_V) given by:

$$E_V = V_f E_f + (1 - V_f)\, E_m \tag{1}$$

where the parameters E and V are the elastic modulus and volume fraction respectively, while indices f and m refer to the fibers (reinforcement) and matrix, respectively. Similarly, the inverse rule of mixture (or Reuss model, E_R) can be written as:

$$E_R = \frac{E_f E_m}{V_f E_m + (1 - V_f)\, E_f} \tag{2}$$

The Voigt model assumes a parallel reinforcement orientation in the matrix with respect to the applied stress direction, while the Reuss model assumes that the reinforcements are normal to the applied stress direction. Based on Equation (1-2), the model of Hirsch (E_H) was proposed as a combination of the series and parallel orientation models to give:

$$E_H = xE_V + (1 - x)E_R \tag{3}$$

where x is a lumped parameter taking into account the effect of interfacial stress transfer, as well as fiber orientation, length and diameter[9] . It can also be seen as a weighing parameter (ranging between 0 and 1) to account for the relative contribution of each model. Nevertheless, other factors such as fiber ends and stress amplification effects should be accounted for, especially in complex cases where synergistic or antagonistic effects are produced. For example, the Tsai-Pagano (E_{TP}) model is written as[10] :

$$E_{TP} = \frac{3}{8}E_f + \frac{5}{8}E_m \tag{4}$$

for a random short fiber orientation distribution.

For hybrid composites (E_{HC}), a similar approach as Hirsch was proposed leading to the linear rule of hybrid mixtures (RoHM):

$$E_{HC} = V_1 E_1 + (1 - V_1)\, E_2 \tag{5}$$

where indices *1* and *2* refer to both types of reinforcement used inside the hybrid composites. But this model only predicts a linear variation between the properties of both composites, made from a single reinforcement having modulus E_1 or E_2, based on their relative volume fraction.

2.2 Proposed model

Based on the simple RoHM model presented in Equation (5), a relative dimensionless modulus can be defined as:

$$\frac{(E_{HC} - E_2)}{(E_1 - E_2)} = V_1 \tag{6}$$

But Equation (6) can be modified to account for different factors as described above. In this case, a nonlinear model can be proposed as:

$$\frac{(E_{HC} - E_2)}{(E_1 - E_2)} = f\,(V_1) \tag{7}$$

As the simplest case, a polynomial function can be used as a first approximation:

$$f\,(V_1) = a + b\,(V_1) + c\,(V_1)^2 + d\,(V_1)^3 \tag{8}$$

where the parameters a, b, c, and d are function of the different factors influencing the composites response such as interfacial stress transfer, reinforcement dispersion/distribution and orientation, as well as particle orientation, length and diameter. But for the model to be valid and physically sound, the following constraints must be satisfied to recover the values of the single reinforcement composites (limiting cases):

$$a = 0 \quad \text{and} \quad b + c + d = 1 \tag{9}$$

So Equation (8) can be rewritten as:

$$f(V_1) = b(V_1) + c(V_1)^2 + (1 - b - c)(V_1)^3 \quad (10)$$

The main advantage of Equation (10) is that positive (synergistic) or negative (antagonistic) deviations from the linear rule of Equation (6) (the case where b = 1 and c = 0) can be obtained depending on the signs and values of both parameters b and c. Furthermore, a maximum modulus and its condition (optimized V_1 value) can be determined for a specific system by solving:

$$f'(V_1) = b + 2c(V_1)^2 + 3(1 - b - c)(V_1)^2 = 0 \quad (11)$$

to give:

$$(V_1)_{MAX} = \frac{c + (3b^2 + 3bc + c^2 - 3b)^{1/2}}{3(b + c - 1)} \quad (12)$$

Typical examples are presented in Figure 2 for the linear case, as well as for positive and negative deviations with different values of the parameters b and c.

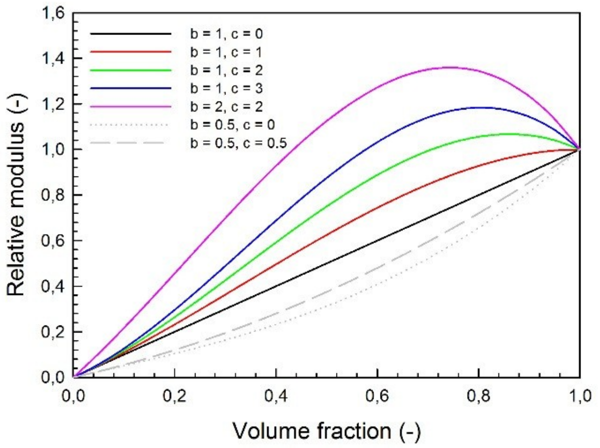

Figure 2. Typical curves for the relative hybrid composite modulus (defined by Equation (6)) as a function of the volume fraction of the first component for different values of the model parameters (b and c)

The curves presented in Figure 2 show that the maximum value of the relative modulus can be substantially higher (up to 40%) than the modulus on the most reinforcing component (V_1= 100%) used alone. Furthermore, it can be seen that the optimum hybrid composition (V_{1MAX}) is in the range V_1 = 60-100% which is in agreement with the data reported in the literature (see Figure 1 for two examples).[7, 8]

3 Results

3.1 Data taken form the literature

Figure 3 presents some data taken on a blend of long hemp fibers (index *1*) and short hemp fibers (index *2*) in a polypropylene matrix for a 20% wt. total reinforcement content[7] . It is clear that for the tensile modulus, a maximum in the curve is present. Based on these data, a non-linear regression method was applied (Sigmaplot v.11) to get the parameters of Equation (10) and the values are reported in Table 1. In this case a good fit is obtained. Figure 3 also shows that a similar trend can be seen for the tensile strength of these samples. In this case, Equation (7) can be modified by changing the modulus (E) by any other property making the proposed model very general. A good fit is also obtained, but the model parameters are different for both properties. This is a clear indication that each model parameter is influenced not only by the materials' properties, but also by the type of property (deformation) measured.

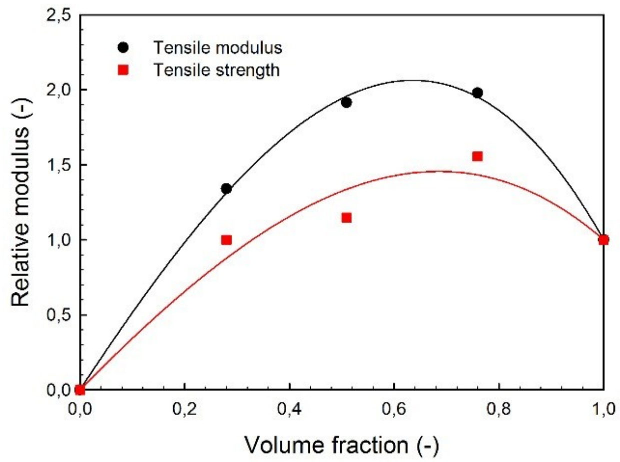

Figure 3. Typical curves for the relative properties of hybrid composites as a function of the volume fraction of the first component for data taken.[7] The lines are regressions with the parameters b and c reported in Table 1

Figure 4 also presents some data for the hemp/polypropylene system[7] , but for a total reinforcement content of 30% wt. In this case, there is also a maximum value from the experimental elongation at break, but the fit is not as good as this parameter is known to have more experimental uncertainty. Nevertheless, a reasonable agreement can be obtained from the simple model proposed. On the other hand, the data on ductility can be represented by a sigmoidal type of curve as obtained from the regression. But again, due to experimental uncertainty, the linear model (dashed line with b = 1 and c = 0) might also be sufficient as reported several time in the literature[6] .

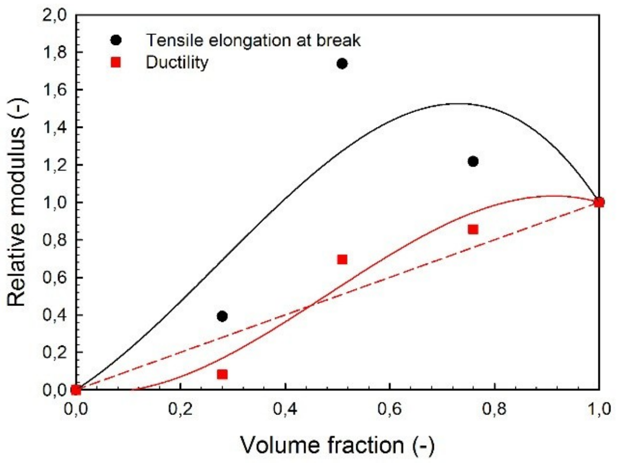

Figure 4. Typical curves for the relative properties of hybrid composites as a function of the volume fraction of the first component for data taken.[7] The lines are regressions with the parameters b and c reported in Table 1

Table 1. Model parameters of Equation (7-8) for hemp/polypropylene auto-hybrid composites[7]

Total hemp content (% wt.)	Property	b	c	$V_{1\,MAX}$
20	Tensile modulus	5.31	-1.39	0.64
	Tensile strength	3.55	-1.05	0.68
30	Tensile elongation at break	1.8	3.63	0.73
	Ductility	-0.47	4.74	0.91

Finally, Table 1 reports on the optimum volume fraction (V_{1MAX}) for each property as calculated by Equation (12). This information would be highly important to optimize the properties of a specific composite based on the model parameters without having to go back in the lab to produce more samples.

4 Conclusion

In this work, a simple model is proposed to represent some synergistic/antagonistic effect in hybrid and auto-hybrid composites defined as positive or negative deviations from the linear rule of mixture. Using only two parameters, several behaviours can be obtained including the linear rule of mixture, as well as positive and negative deviations from the latter. The model can be used to determine the optimum composition for hybrid composites (two types of reinforcement), but could be generalized for more components (three and more). Finally, good fitting with data taken from the literature was possible, but more work is needed to relate the parameters' value to physical factors such as particle dimensions, interfacial stress transfer, orientation, *etc*.

5 Conflict of interest and funding

There is no conflict of interest. Funding for this work was received from the government of the Hunan province (China).

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