Modelling of tensile properties glass powder/epoxy composites post-cured in an oven and in microwaves

H Ku, J Epaarachchi M Trada and P Wong

Faculty of Engineering and Surveying
& Centre of Excellence in Engineered Fibre Composites
University of Southern Queensland

Journal of Reinforced Plastics and Composites, **Modelling of tensile properties glass powder/epoxy composites post-cured in an oven and in microwaves** will appear in print in Volume 32 Issue 10 May 2013 pp. 15 - 25.

Abstract: This paper discusses a simplified approach to analyse the mechanical properties of randomly distributed particulate composites. Mechanical properties of glass powder filled vinyl ester resins were experimentally investigated. The analytical results were compared with experimental results and a very good correlation was found. Further, the experimental results and the predictions showed that the strength of the composites is less than the strength of the matrix material, for all three composites tested.

Keywords: Modelling, epoxy resin, mechanical properties and glass powder

Introduction

Powder fillers and particulate materials are now very popular in the manufacturing of composite materials because the weights of composite structures can be substantially reduced, and at the same time keeping or even improving the properties of the materials. The particulate sizes dispersed in the matrix of the particulate composite are in the range of µm to nm. The ease of fabricating complex parts, low cost and isotropic nature are some of the significant characteristics of particulate composites. Fillers were frequently added to polymers to increase stiffness and reduce the cost of the composite by lowering the amount of matrix materials. Nowadays, there are many inorganic and metallic fillers that have been used to modify the physical, mechanical, electrical and thermal properties of the particulate-polymer composites. Recently, the advances in nano-technology have pushed the particulate- polymer composite to a new level. Now, nanoclay and carbon nano-tubes are some of the latest fillers being used with conventional fillers such as hollow glass spheres, clay, silicon oxides and calcium carbonate. Research work done on the particulate-polymeric composites has shown significant improvements in physical and mechanical properties by the addition of a small quantity of nano-scale particles [1-4].

Extensive work has been done on short fibre composites and particulate-polymer composites. It has been well known that the addition of small amount of short-fibre fillers will increase of the properties of composites significantly [4]. On the hand, the significant governing factor that controls the properties of particulate-polymer composites is the aspect ratios of the particles, which is usually one for spherical particles. Due to this reason the properties of particulate-polymer composite are significantly different to the short fibre composites. Some notable work by Tavman, Martin et al. and Fu et al. [1, 2, 5] suggested that the addition of metallic or organic materials fine particles of the size of micrometers will improve the modulus of the composites significantly. However, they explained that the strength of the composite varies significantly according to the stress transfer between the particles and the matrix. If the bonding between matrix and the particles are effective, then the strength will increase. The work detailed in this paper investigated the characteristics and the properties of glass powder filled epoxy particulate-polymeric composites.

Experiments

The reinforcement was glass powder (glass hollow sphere) particulates and they were made 0 % to 35% by weight in the cured epoxy composites. As the raw materials of the composites are liquid and glass hollow spheres, the tensile test specimens were cast to shape. The resin is an opaque liquid and is first mixed with the catalyst. After that the glass powder is added to the mixture, they are then mixed to give the uncured composite. The mixture of glass powder, resin and accelerator was blended with mechanical blender to ensure a more homogenous mixture. The mixture was poured into the moulds and the moulds with composites were located under the fume cabinet. Before pouring the mixture, plate surface was covered by a glossy paper to facilitate removal of composite after solidification.

After initial 24-hour curing when the test pieces were removed from the mould, they were cut into pieces. Half of the test pieces were then post-cured in an oven. Oven temperatures and times were:

- 16 hours at 40°C
- 16 hours at 50°C
- 8 hours at 60°C

The samples were then tensile tested.

The other half of the samples were post-cured in microwaves for 4 minutes using a power level of 320 W and the temperature reached was 40 ^{o}C. The temperature was measured using an Oakton TempTestr Infra Red handheld thermometer. Allow the samples to cool in the oven cavity to room temperature. The samples were again heated to 50 ^{o}C by exposing them to a power level of 320 W for 6 minutes. Allow the samples to cool in the oven cavity to room temperature. The samples were again heated 60 ^{o}C by exposing them to a power level of 320 W for 8 minutes. Allow the samples to cool in the oven cavity to room temperature.

The processes were equivalent to heating the samples in a conventional oven. The samples were then tensile tested. The mechanical property testings were carried out on MTS 100T universal material testing machine. The rate of extension, 1 mm per minute, was in accordance with an Australian Standard (Australian Standard 1145.2, 2001) [6].

Modelling

Various models are available to predict the mechanical properties such as yield and tensile strength as well as the elastic modulus and elongation at breakage of composite materials of particulate composites. . In general the properties of mixtures are predicted by developing relationships between the different constituents of the composites, in this case the matrix, epoxy resin, and the filler, glass powder.

Studies have shown that when the aspect ratio, which is determined by dividing the length of the particle by its diameter, is at unity, the particles can be considered spherical. The Young’s modulus of the composite is governed by a number of factors including the particulate loading and particle size. Fu et al. (2008) conducted this study and in doing so found that adhesion at the interface between the matrix and the filler has negligible effect on the modulus of particulate composites. It must be noted that small differences in particle size tend to have little to no effect on the mechanical properties of the composite and only when the particle size is reduced into the nano scale will there be a major difference [2].

In this study, the density of the glass powder used is 0.6 g/cc. The density of epoxy resin used is 2 g/cc. The elastic modulus of the epoxy resin is 2.71 GPa. For composites with 10 W/t % of glass powder, the mass of glass powder in 100 g of the composite is 10 g and that of the epoxy resin is 90g. The volume of the glass powder in 100g of the composite = $\frac{mass}{density}$ =

$\frac{10}{0.6}$ = 16.67 cc. The volume of the epoxy resin in 100g of the composite = $\frac{mass}{density} = \frac{90}{2} =$ 45 cc. The volume fraction of the glass powder = $\frac{16.67}{16.67+45}$ = 0.27. The volume fraction of the resin = 1 - 0.27 = 0.73. The volume fractions of other W/t % of glass powder and epoxy resin considered are shown in Table 1.

Results and Discussion

Yield strengths

According to Nicolais and Narkis' prediction,

$$\sigma_{yc} = \sigma_{ym} [1 - (v_f/v_m)2/3] [1] \qquad (1)$$

where σ_{yc} is the yield strength of the composite

σ_{ym} is the yield strength of the matrix

v_f is the volume fraction of the filler

v_m is the volume fraction of the matrix

Values of yield strength of the composites obtained from Nicolais and Narkis' prediction and experiments are shown in Table 2 [7].

Figure 1 shows the values of yield strength of the composites obtained from Nicolais and Narkis' prediction and experiments [7]. It can be found that the values of yield strength of the samples post-cured in microwaves were higher than their counterparts post-cured in an oven. The values of yield strength obtained from Nicolais and Narkis' prediction were much lower than those from experiments, particularly at higher particulate loading.

Wong's model was therefore developed to suit these types of particulates, e.g. glass powder. In Wong's model, $\sigma_{yc} = \sigma_{ym} [1 - 0.15 (v_f/v_m)]$ (2)

The composites used by Nicolais and Narkis in their study were SAN (Styrene-acrylonitrile)/glass beads and those used in this study was glass powder filled epoxy resins. It can be argued that the matrix-filler interaction at interface of the constituent materials used in this study was better than that used by Nicolais and Narkis; they therefore arrived at an equation giving lower yield strengths at higher filler contents, using the data of this study.

Figure 2 illustrates the yield strengths of glass powder filled epoxy composites post-cured in an oven and from Wong's model. The yield strength of neat resin, σ_{ym} used is that of neat resin post-cured in an oven. It can be found that the model predicted the values of yield strength of the composites very accurately and was within the 5 % markers.

The yield strengths of glass powder filled epoxy composites post-cured in microwaves and from Wong's model are depicted in Figure 3. The yield strength of neat resin, σ_{ym} used is that of neat resin post-cured in microwaves. It can be found that the model predicted the values of yield strength of the composites very accurately and was within the 5 % markers.

Tensile strengths

According to Tavman's model, $\sigma_c = \sigma_m (1-b.v_f^{2/3})$ [1] (3)

where σ_c is the tensile strength of the composite

σ_m is the tensile strength of the matrix

b = 1.1 for densely packed hexagonal packing in the plane of highest density

b = 1.21 for poor adhesion and spherical particles

The values of tensile strength of the composites obtained from Tavman's prediction and experiments are depicted in Figure 4 [1]. The value of b used is 1.21. The tensile strength of the matrix, σ_m used is that of tensile strength of neat resin post-cured in an oven. It was found that the prediction for this model was not satisfactory as the values dropped significantly with particulate loading. Figure 5 shows the values of tensile strengths of glass powder filled epoxy composites post-cured in microwaves and from Tavman's model. The value of b used is 1.21. The tensile strength of the matrix, σ_m used is that of tensile strength of neat resin post-cured in microwaves. It was found that the prediction for this model was not satisfactory as the values dropped significantly with particulate loading; the case was similar to composited post-cured in an oven.

Wong's model was therefore developed to suit these types of particulates, e.g. glass powder, in which adhesion between the filler and the matrix was not too bad, particularly at lower particulate loading. In Wong's model,

$$\sigma_c = \sigma_m (1-b.v_f^{2/3}) \tag{4}$$

but b = 0.5 for samples post-cured in an oven. For samples post-cured in microwaves, the value of b = 0.5 for lower concentration of filler (lower bound) and in this case it was 15 W/t %; after this the value of b has to be increased to 0.7 (upper bound). Figure 6 illustrates tensile strengths of glass powder filled epoxy composites post-cured in an oven and from Wong's model. It was found that the model predicted the results of the experiments quite well.

The tensile strengths of glass powder filled epoxy composites post-cured in microwaves and from Wong's model were depicted in Figure 7. It was found that the model predicted the experimental data accurately.

Young's modulus

Using Neilsen's model, $E_c = E_m (1 + 2.5 V_f)$ (5)

the Young's modulus of the composites were calculated [8, 9]. Figure 8 shows the values of Young's moduli obtained from Neilsen's model and experiments, post-cured in an oven and in microwaves, respectively. It was found that the values of Young's modulus post-cured in microwaves were more reasonable and realistic when compared to those post-cured in an oven. The Young's modulus of resins reinforced with particulate fillers will usually increase slowly to a maximum at a particular filler loading depending on the attributes of the filler, but usually at low concentration of filler. Up to this particular weight content of filler, the adhesion between the particle and the matrix is ideal; after this, the amount of resin can no longer encapsulate the particles completely, leading to the generation of a large number of voids, thus lowering the Young's modulus of the composites [6-8]. *The values from Neilsen's model rose steadily and can be argued to be too high because his model is only suitable for low concentrations of filler*. *The filler in this study is up to 35 wt. %.* However, the model that was proposed for the case when the matrix slips past the particle appears to be accurate for higher concentrations. However, even for lower particulate loading, the model did not work very well [9, 10].

Using the equation developed from Einstein's theory,

$$E_c = E_m (1 + V_f) \quad (6)$$

the Young's modulus of the composites were calculated [10]. Figure 9 shows the values of Young's moduli obtained from Neilsen's model, Einstein's theory and experiments, post-cured in microwaves [5, 8]. It can be found that the Einstein's model predicted the Young's modulus of the composites quite accurately at lower particulate loading (up to 15 W/t % of glass powder) but it was incorrect at higher concentration of fillers. The adhesion between the particle and the matrix was no longer ideal after this W/t % of filler. The amount of resin can no longer encapsulate the particles completely at higher W/t % of filler, leading to the generation of a large number of voids, thus lowering the Young's modulus of the composites [10-11].

Wong's model was therefore proposed for these types of fillers. It consisted of two equations, one was for lower particulate loading (lower bound) and the other was for higher filler (upper bound) content.

$$E_c^l = E_m (1 + 0.5V_f) \quad (7)$$

$$E_c^u = E_{m(highest)} (1 - V_f^4) \quad (8)$$

The first Wong's equation, $E_c^l = E_m (1 + 0.5V_f)$, was developed from Einstein's prediction model and it was for lower particulate loading (lower bound), wt % of filler up to 15. The second equation, $E_c^u = E_{m(highest)} (1 - V_f^4)$, had the elastic modulus of the matrix changed so that it was the highest value obtained from the first equation.

Figure 10 illustrates that the values of Young's moduli obtained from Wong's model, Einstein's model prediction and experiments, post-cured in microwaves. It was found that Wong's model predicted the results of the experiments quite closely, hence Wong's model would be suitable for particulate, like glass powder, that would render the adhesion between the particle and the matrix no longer ideal at higher particulate loading, and the amount of resin can no longer encapsulate the particles completely.

Other models

Ishai and Cohen presented a model that depend on an upper and a lower bound [12]. Hsieh et al. performed similar experiments on a particulate filled metal matrix. They used a method of determining upper and lower bounds that used equations established by Voigt and Reuss (Voigt's model, $E_c = E_mV_m + E_fV_f$; Reuss' model, $E_c = \frac{E_mE_f}{E_mV_f + E_fV_m}$) [13]. The Voigt and Reuss bounds were relatively wide apart, and modifications had accordingly been proposed by many researchers. Among these modifications, the Hashin and Shtrikman model (H–S model) had received wide attention. Hashin and Shtrikman treated the system containing one particulate phase and one continuous matrix phase. They employed the "minimum energy" principle and introduced bounds on the bulk modulus, *K* and shear modulus, *G* as

$$K_c^l = K_m + \frac{V_p}{(1/K_p - K_m) + (3V_m/3K_m + 4G_m)} \tag{9}$$

$$K_c^u = K_p + \frac{V_m}{(1/K_m - K_p) + (3V_p/3K_p + 4G_m)} \tag{10}$$

$$G_c^l = G_m + \frac{V_p}{(1/G_p - G_m) + (6(K_m + 2G_m)V_m/5G_m(3K_m + 4G_m))} \tag{11}$$

$$G_c^u = G_p + \frac{V_m}{(1/G_m - G_p) + (6(K_p + 2G_p)V_p/5G_p(3K_p + 4G_p))} \tag{12}$$

where V_p is V_f, volume fraction of the particle or filler. The subscripts m and p denote, matrix and particle, respectively.

The lower and upper bounds on the elastic modulus can be estimated by using the following equations as

$$E_c^l = \frac{9K_c^l G_c^l}{3K_c^l + G_c^l} \tag{13}$$

$$E_c^u = \frac{9K_c^u G_c^u}{3K_c^l + G_c^u} \tag{14}$$

The upper and lower bounds proposed by the H–S model were relatively closer to each other. Therefore, the H–S model provided a more precise expression for the elastic and shear moduli of a two-phase material.

A prediction on the elongation of a composite specimen could be made using a model developed by Nielsen [1]. Tavman mentioned that this model assumed that perfect adhesion was present between the particles and the matrix. Due to the random sizes of the sawdust particles and surface finishes of the filler present this assumption was not entirely accurate, however for the purpose of this study might be used to form a comparison. The equation was $\varepsilon_c = \varepsilon_p(1 - V_f^{1/3})$ (15)

Discussion

ANOVA (analysis of variance between groups) was used to analyse the data statistically. In this study, all the raw data for each tensile property consisted of 8 groups and the number of data in each group was 6. The tensile strength of the composites post-cured in microwaves was first studied. The data entered to a program was illustrated in Figure 11. The results of the analysis were shown in Figure 12 [14]. It was found that the F was 84.43 and that the null hypothesis is incorrect and 'large' F indicates the tensile strengths will be different for different wt % of glass powder. The other tensile properties of the composites were similarly proved by ANOVA with composites post-cured in an oven or in microwaves.

Conclusions

This study has evaluated the mathematical models of many previous workers. In yield strengths, Narkin's prediction was lower than the experimental results of this study. The model developed by Wong matched the results of samples post-cured in microwaves closer. In tensile strengths, Tavman's model gave lower results than those found in this study. Again, Wong's model matched the results of microwave-cured samples closer. As far as Young's modulus is concerned, Neilsen's model gave results higher than those found in this study. Einstein's prediction gave even higher results for Young modulus. On the other hand, Wong's model matched the results of microwave cured samples well.

References

1. Tavman, L H, Thermal and Mechanical Properties of Aluminium Powder-Filled High-Density Polymer Composites, Journal of Applied Polymer Science, 1996, Vol 62, pp. 2161-2167.

2. Fu, S. Feng, X, Lauke B and Mai, Y, Effects of particle size, particle/matrix interface adhesion and particle loading on mechanical propertiesof particulate–polymer composites. Composites B, 2008, Vol. 39, pp. 933-961.

3. Sauer, J A and Richardson, G C, Fatigue of polymers, International Journal of Fracture, 1980, Vol. 16, No. 6, pp. 499-532.

4. Epaarachchi, J, Ku, H and Gohel, K, A Simplified Empirical Model for Prediction of Mechanical Properties of Random Short Fiber/Vinylester Composites, Journal of Composite Materials, 2010, Vol. 44, No. 6, pp.779-788.

5. Martin, M, Hanagud, S and. Thadhani, N N, Mechanical behaviour of nickel + aluminium powder-reinforced epoxy composites, Material Science and Engineering A, 2007, Vol. 443, pp. 209-218.

6. Australian Standard 1145.2 (2001). ‘Determination of tensile properties of plastic materials – Test conditions for moulding and extrusion plastics’.

7. Nicolais, L and Narkis, M, Stress–strain behaviour of styrene–acrylonitrile bead composites in the glassy region, Polymer Engineering Science, 1971, Vol.11, No. 3, pp. 194–203.

8. Al-Hajjaj, A Tahseen and Saki, A, Improving the design stresses of high density polyethylene pipes and vessels used in reverse osmosis desalination plants, Journal of Saudi Chemical Society, 2010, Vol. 14, pp. 251–256.

9. Marrett, C, Moulart, A, Colton, J., Tcharkhtchi, A., Flexible polymer composite electromagnetic crystals, Polymer Engineering and Science, 2003, Vol. 43, No. 4, pp. 822–830.

10. Epaarachchi, Jayantha A and Reushle, Matthew T, Performance of aluminium/vinylester particulate composite, 2010, In: PRICM7: 7th Pacific Rim International Conference on Advanced Materials and Processing, 2-6 Aug 2010, Cairns, Australia.

11. Ray, D, Bhattacharya, D, Mohanty, A K, Drzal, L T and Misra, M, Static and Dynamic Mechanical Properties of Vinylester Resin Matrix Composites Filled with Fly Ash, Journal of Applied Polymer Science. 2006, Vol. 291, pp. 784-792.

12. Ishai, O and Coheno, L J, Effect of Fillers and Voids on Compressive Yield of Epoxy Composites, Journal of Composite Materials July 1968, Vol. 2, pp. 302-315.

13. Hsieh, C L, Tuan, W H and Wu, T T, Elastic behaviour of a model two-phase material, Journal of the European Ceramic Society, 2004, Vol. 24, pp. 3789–3793.

13. ANOVA, http://www.physics.csbsju.edu/stat/anova.html <viewed on 3 Dec 2012>.