Prediction of Mechanical properties of Epoxy composites containing mono and hybrids particulate fillers.

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Abstract: This paper reports the mechanical properties such as tensile strength and tensile modulus of epoxy composite systems containing micro particulates of Cu, Al and hybrid (Al-Cu) which are randomly distributed. The experimental results are compared with strength prediction equations of Biggs, Ross Murphy & Todd, Langley & Green and Wong. Experimentally determined tensile modulus was compared with Reuss and Counto equations. The values obtained from above equations was not in line with experiment. Hence in view of the above, a new strength predictive equation is developed which is empirical in nature and exhibits good correlation with experimental data.

1. Introduction
Epoxy is one of the structural polymers which are widely used. Epoxy resins have excellent resistance to heat, moisture, chemicals and good compatibility with many materials [1]. However, cross link structure of epoxies makes them brittle and poor crack resistance [2]. A combination of epoxy and particulate fillers can improve physical, mechanical, wear resistance, thermal and electrical conductivities [3]. The epoxy filled particulate polymer composites have wide acceptance in industrial applications due to its ability to meet specific requirement and ease of manufacturing [4]. Generally, all fillers improve modulus as they are stiffer than matrix whereas similar improvement is not seen with respect to strength [5]. The strength of micron size filler reinforced polymer composites depends on the strength of interface between matrix and reinforcement. The effective load transfer from matrix to reinforcement can happen with good interface. Further filler volume fractions, intrinsic strength of filler and processing method adapted have significant impact on polymer composite strength. The size, shape and distribution of particle in a matrix and interface have a role in strength of a polymer [6, 7]. Prediction of strength and modulus of a polymer composite to an extent reduce experimentation. Prediction models which predict composite strength accurately with one type of filler may not be accurate with other filler. Hence prediction of strength of a particulate filled polymer composites is not at all simple. There are various analytical equations developed by researchers from time to time to predict tensile strength and tensile modulus of particulate filled epoxy composites. Further, the exactness of a predictive model is measured with experimental data to validate its usefulness. The present study compares the effect of reinforcing micron sized particulate metallic fillers of Al and Cu as mono and hybrid (Al-Cu) filler. Further the strength of the epoxy composites found by experiment was compared with strength predictive models. The variation was found to be more. In view of this a new empirical equation is developed which exhibits good correlation with experiment values.

2. Materials and Methods
Epoxy resin of type LY556 and an amine hardener HY951 supplied by Huntsman India Ltd was the host matrix and the fillers aluminium and copper were procured from Loba chemie India ltd. The size of Al fillers is 10-80µm and Cu fillers are of 5-15µm shown in figure 1.

2.1 Composite preparation
The predetermined amounts of reinforcements were preheated to remove any moisture present in it and were dispersed in epoxy. The mixtures were stirred continuously until the reinforcements were properly...
distributed. The resin and hardener were mixed in the ratio 10:1 by weight. Hardener was added to the mixture with continuous stirring, followed by pouring the mixture into a prepared mould. The cast specimens were allowed to cure in the mould for 24 hours at room temperature followed by post curing at $50^\circ C$ for 30 minutes, $70^\circ C$ for 60 minutes and $85^\circ C$ for 120 minutes. The particle volume fraction is calculated using the equation.

$$V_f = \frac{W_f}{W_f + (1-W_f)\frac{\rho_{par}}{\rho_{mat}}}$$

(a) Al  
(b) Cu

Figure 1: SEM of particulates of aluminium and copper

3 Mechanical Tests

The tensile tests were conducted on a flat dog bone type specimen having dimensions 165 X 20 X 3mm on LYOD’S universal testing machine. ASTM D 638 test procedure was followed to evaluate the tensile properties [8]. The specimens were pulled at a constant loading rate of 1mm/min till the specimen breaks. For each type, 5 specimens were tested. The strength of the composites was determined using the equation

$$\sigma_c = \frac{P}{b d}$$

Where, $\sigma_c$ = ultimate tensile strength (MPa), $P$ = load (N), $b$, $d$ = width and thickness (mm)

The modulus of the composites was determined using the equation

$$E = \frac{(AP)}{(AL)} \frac{L}{bd}$$

P = load (N), $b$, $d$ = width and thickness (mm), $E$ = young’s modulus of elasticity (GPa).

The Scanning electron microscopy of particulate fillers and fractured surfaces were captured by Joel make 5600 LV scanning electron microscope. The predictive equations used are shown in table 1.

4. Results and Discussion:

4.1 Effect of fillers on tensile strength and tensile modulus

Figure 2(a) presents the tensile strength of unfilled epoxy and epoxy filled with particles of Al, Cu and hybrid (Al-Cu). Neat epoxy exhibits a tensile strength of 39MPa. Al-epoxy composites show a fluctuating behavior in strength and strength decreases with increased fillers. The decreased strength of Al-epoxy composites may be attributed to low dense Al fillers (2.4gm/cc), bigger particles (10 to 80µm) and lower inherent strength (100MPa). Al fillers occupy more volume for a given weight fraction leading to agglomeration, making less coverage of particles by resin, creating weak interface which cannot sustain the applied load and fails at loads lower than neat epoxy [9]. Cu-epoxy composites show an improvement in tensile strength up to 30% Cu reinforcement and strength decreases with 40% reinforcement. The increased strength of Cu-epoxy composites can be attributed to smaller particle size (5 to 15µm), higher density (8gm/cc) and higher inherent strength (200MPa) of Cu fillers. For a given weight fraction, Cu particulates occupy less volume and each particle is well covered by resin to form a good interface. A good interface can effectively transfer load between matrix and reinforcement. The strength of hybrid (Al-Cu) composites also decreases with increased hybrid fillers; however, the strength of these composites is in between Al-epoxy and Cu-epoxy composites. At 40% filler loading,
irrespective of filler type, strength degrades for all filled composites due to agglomeration of particles. Increased fillers reduce the interfacial adhesion due to particle to particle contact, which leads to non-effective transfer of load. Higher particle size makes adhesion poor at the interface leading to poor strength of the composites [10,11 and 12]. Further, dispersed particulates in epoxy have different moduli and poison’s ratio from the resin, creates stress concentration and this magnifies the stress surrounding the particle leading to lowering of strength [13]. Figure 2 (b) presents the tensile modulus of epoxy composites. Cured epoxy exhibits a tensile modulus of 2.51GPa. Tensile modulus of Al-epoxy, Cu-epoxy and hybrid (Al-Cu)-epoxy increases with increased fillers. Modulus of Al-epoxy composites are higher than Cu-epoxy and hybrid (Al-Cu) epoxy due to more particles for a given weight fraction.

Figures 3 to 8 compares tensile strength and modulus predicted by analytical equations with experiment. The values are shown in table 2.

4.2. Evaluation of predictive equations

Bigg’s model is based on the adhesion between filler and matrix [14]. The value of b is taken as 1.1 (good adhesion) in Bigg’s equation. The equation predicts decrease of strength increased volume fraction of fillers. The deviation is more with Al particulate fillers as volume fraction is more. The product b (Ø)\(^{3/2}\) in the equation increases with increased volume fraction. Bigg’s model doesn’t present the exactness of the experimental findings. Prediction of strength by Ross Murphy &Todd [15]is also not in line with experimental results. Increased volume fraction changes the strength marginally. Langley & Green Equation [15] predicts higher strength for Al-epoxy composites while for Cu-epoxy composites the strength is in line with experiment up to 30% Cu and show higher strength for 40% Cu. The predicted strength for hybrid (Al-Cu) composites is in line with experiment up to 20% hybrid fillers over estimates above 20% fillers. The value (1-(Øf/Øm))\(^{3/2}\) increases with increased volume fraction. The prediction by Wong’s equation is reasonably good. The value of b in Wong’s equation is taken as 0.5 for 10 and 20% fillers, 0.7 for 30 and 40% fillers [16].

4.3 New Equation: Most of the predictive models assume that the inclusions of fillers in a matrix, reduce the strength of composites. Furthermore, above models assume that composite strength cannot be higher than the strength of the matrix. But in some cases, increased fillers may increase the strength of the composites signifying that fillers can also contribute to composite strength by creating good interface. Hence the proposed empirical equation (a modified rule of mixture) considers the contribution of constituents i.e. filler and matrix on composite strength.

\[
\sigma_c = \sigma_{m}W_{m} \pm Y\sigma_{f}W_{f}
\]

Where, \(\sigma_{m,f}\) = tensile strength of composite, matrix and filler, \(W_{m,f}\) = weight fraction of matrix and filler, and \(Y = \frac{1}{a + (n-1)d}\)

\[a = 2,\ d = 0.1\ and\ n\ represents\ the\ percentage\ of\ fillers\ by\ weight.\]

The positive sign in the equation is to be considered when the inherent strength of filler is higher.
Figure 3: Tensile strength Al-Epoxy composites

Figure 4: Tensile strength Cu-Epoxy composites

Figure 5: Tensile strength of Hybrid (Al-Cu)- epoxy composites

Figure 6: Tensile modulus of Al-Epoxy composites

Figure 7: Tensile modulus of Cu-Epoxy composites

Figure 8: Tensile modulus of hybrid composites
than epoxy and the negative sign is to be considered for fillers that have lower inherent strength than epoxy. The prediction by new equation is in line with experiment values for Al-epoxy, Cu-epoxy and hybrid (Al-Cu)-epoxy composites. The deviation of new equation is minimum compared to other predictive models. Figures 6 to 8 compares the tensile modulus predicted by analytical equation and experiment. The Reuss equation predicts a marginal improvement in tensile modulus with increased fillers and the values are lower than experimental findings. Count’s prediction is lower than experiment for Al-epoxy composites, higher than experiment for Cu–epoxy and hybrid (Al-Cu)-epoxy composites.

5. SEM of Fractured surfaces

The images of shown in figure 9(a-c) has the features of a brittle fracture such as crack, fan blade marks, hackle zones which are the characteristics of brittle fracture [17]. These images correspond to neat epoxy 5A5C-3Ep and 10A10C-3Ep composites. The images of 15A15C-3Ep have the characteristics such as particle removal, void, and formation of cracks which could not sustain the applied load and makes the composites to land up with lower tensile strength.

![Fig 9 SEM of fractured surface of epoxy-mono and hybrid composites](image)

| Table 1 Description of Models |
|-------------------------------|
| **Strength Prediction Models** |
| Bigg’s \[ \sigma_c = \sigma_p (1 - b (\phi^{2/3}) ) \] where \( \sigma_{cp,m} \) = tensile strength of the composite, matrix, \( b = 1.21 \) for poor adhesion between filler and matrix, \( b = 1.1 \) for good adhesion, \( \phi \) = volume fraction of fillers |
| Ross Murphy and Todd \[ \frac{\sigma_c}{\sigma_m} = \left( \frac{1 - \phi^2}{1 - \phi \phi_f} \right)^{3/2} \] where, \( \sigma_{c,m} \) = tensile strength of the composite and polymer matrix, \( \phi \) = volume fraction of fillers |
| Langley and Green \[ \frac{\sigma_c}{\sigma_m} = \left( 1 - \frac{\phi_f}{\phi_m} \right)^{-3/2} \] where, \( \sigma_{c,m} \) = tensile strength of the composite and polymer matrix, \( \phi_{f,m} \) = volume fraction of fillers and matrix. |
| Wong \[ \sigma_c = \sigma_m \left( 1 - b V_f^2 \right) \] \( V_f \) = volume fraction of fillers |

| **Modulus Prediction Models** |
|-------------------------------|
| Reuss \[ E_c = \frac{E_m E_f}{E_f (1 - V_f^2) + E_m V_f^2} \] |
| Counto \[ \frac{1}{E_c} = \frac{1 - V_f^2}{E_m} + \frac{1}{E_f (1 - V_f^2)} \] |

where, \( E_{cmf} \) = Tensile modulus of composite, matrix and filler

\( V_f \) = Volume fraction of reinforcement
Table 2 Comparison of tensile strength & modulus of experiment with prediction models

|                | Tensile strength (MPa) | Tensile Modulus (GPa) |
|----------------|------------------------|-----------------------|
|                | Expt                   | New Equation          | Biggs & Green | Wong | Ross & Todd | Expt | Reuss | Counto |
| 10A-Ep         | 31.55                  | 38.20                 | 33.18         | 42.29 | 36.50       | 28   | 3.03   | 2.63   | 2.79   |
| 20A-Ep         | 36.18                  | 35.82                 | 29.76         | 46.54 | 34.86       | 27.20 | 3.16   | 2.78   | 2.99   |
| 30A-Ep         | 37.22                  | 32.81                 | 26.36         | 53.55 | 31.07       | 27.57 | 4.02   | 2.96   | 3.18   |
| 40A-Ep         | 32.38                  | 29.50                 | 23.37         | 64.11 | 28.93       | 27.77 | 5.17   | 3.21   | 3.41   |
| 10C-Ep         | 41.78                  | 42.00                 | 37.01         | 39.60 | 38.09       | 31.38 | 2.75   | 2.55   | 3.07   |
| 20C-Ep         | 40.81                  | 41.46                 | 34.86         | 40.88 | 38.09       | 31.38 | 2.91   | 2.60   | 3.48   |
| 30C-Ep         | 43.40                  | 39.54                 | 33.98         | 41.57 | 35.81       | 28.42 | 3.05   | 2.65   | 3.93   |
| 40C-Ep         | 37.50                  | 36.96                 | 31.04         | 44.70 | 33.93       | 27.34 | 3.26   | 2.73   | 4.48   |
| 5A5C-Ep        | 40.00                  | 40.10                 | 33.91         | 40.91 | 36.35       | 28.98 | 2.74   | 2.79   | 3.53   |
| 10A10C-Ep      | 41.00                  | 38.90                 | 32.00         | 43.53 | 34.80       | 27.57 | 2.81   | 3.12   | 4.33   |
| 15C15A-Ep      | 39.00                  | 38.44                 | 29.27         | 47.33 | 30.95       | 27.18 | 3.13   | 3.56   | 5.24   |
| 20A20C-Ep      | 34.00                  | 37.89                 | 26.44         | 53.33 | 29.05       | 27.55 | 3.50   | 4.13   | 6.36   |

Summary
Micro particulate characteristics such as higher inherent strength, higher density and smaller size enhance the tensile strength of epoxy composites while filler characteristics such as low density, lower inherent strength and bigger size decrease the tensile strength of epoxy composites. Fillers like Cu which enhances the strength of epoxy can be called as positive fillers while Al fillers reduce the strength of epoxy and can be termed as negative fillers. The deviation of new equation with experimental data is found to be much lower than other predictive models considered in this study.

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