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Effectiveness of Several Theoretical Models in Predicting the Mechanical Properties of Coconut Shell Powder/Epoxy Resin Composites

AM Andezai[1], LM Masu[1] and M Maringa[2]

[1]*Vaal University of Technology, Faculty of Engineering and Technology, Department of Mechanical Engineering, Private Bag X021, Vanderbijlpark, Andries Potgieter Blvd, 1911, South Africa.*

[2]*Central University of Technology, Faculty of Engineering and Technology, Department of Mechanical and Mechatronics Engineering, Private Bag X20539, Bloemfontein, 9300, South Africa.*

Abstract

The static elastic properties of coconut shell powder/epoxy resin composites with two different filler sizes and different volume fractions were determined experimentally, using the ultrasonic through thickness test, and the results compared with predictions from existing theoretical models. Coconut shell filler with average sizes of 150 µm and 212 µm were used to manufacture composites with filler weight fractions of 0, 5, 10, 15, 20, 25 and 30%. The theories developed by Einstein, Guth's and separately Smallwood, the Voigt and Reuss rule, and Kerner, were each used to predict theoretical values of elastic and shear modulus for coconut shell powder/epoxy resin composites for these six different filler weight fractions. The Einstein and Voigt Equations predicted values of elastic modulus that were higher than the experimental values with the former being higher, while the Reuss equation predicted values that were lower than the experimental. The Reuss equation gave the best prediction for the values of ultrasonic through thickness elastic and shear modulus, though slightly lower than the experimental results.

Keywords: Weight percentage, coconut shell powder, particle, matrix, reinforcement, modulus

1.0 INTRODUCTION

Composite properties depend on the properties of the constituent materials; fibres, particulates, and matrices used. The strength and stiffness of the composites are directly a function of the properties of reinforcing phases, which carry most of the load, and their volume content [1]. The matrices help to maintain the relative position of the reinforcing phases within composites and, more importantly, transfer load to the reinforcing phases. As a result, the interfacial properties of the reinforcing phase/resin are also important and have a significant effect on properties of composites, for instance toughness and transverse fracture stress. To fabricate high strength composites, all three factors namely properties of the reinforcing phase and matrix, as well as the characteristics of the reinforcing phase/resin interface are critical [1].

Natural fillers are used in the form of fibres or particle. Coconut shell powder (CSP) is a particle filler used to reinforce polymers. The shape and size of filler particles are prominent parameters which influence the mechanical properties of resulting composites. The strength of composites strongly depends on the stress transfer between the filler particles and the matrix. For well bonded particles, the applied stress can be effectively transferred to the particles from the matrix [2, 3]. A reduction of strength occurs in the case of poorly bonded particles. Adhesion at the filler/matrix interfaces and the strength of composites become higher through the use of the right coupling agents. Consequently, the mechanical properties of filler/polymer composites are affected by the size and type of particles, as well as the effectiveness of interfacial adhesion of fillers and matrices. Theoretical models are used to predict the elastic modulus, strength, and fracture toughness of particulate polymer composites and composites in general [4-7]. Although the predictions may not be precise, once a composite has been processed, curve fitting parameters can be introduced into the models to provide a better fit to the empirical data [7].

The particulate filler phase has been studied with reference to amongst other factors weight fraction and particle size [8, 9]. It has been shown in research that the strength of composites generally increase with increasing volume fraction of the reinforcing filler and for a given particulate volume fraction, increases with decreasing size of filler particles [9, 10]. A variety of models are available to describe the modulus, tensile strength, and elongation at rapture of particle-filled composites as a function of filler volume fraction and particle size, including Einstein's equations, Guth's and Smallwood's separate models, Voigt rule, Reuss model, and Kerner's equation, presented here as Equations 1, 2, 3, 4, 5 and 6, in this respectively order [8,11].

$$E_C = E_m(1 + V_p) \quad (1)$$

$$E_c = E_m(1 + 2.5V_p) \quad (2)$$

$$E_c = E_m(1 + 2.5V_p + 14.1V_p^2) \quad (3)$$

$$E_c = E_m v_m + E_p V_p \quad (4)$$

$$E_c = \frac{E_m E_P}{E_m V_p + E_p \nu_m} \quad (5)$$

$$E_c = E_m\left(1 + \frac{\phi_p}{\phi_m}\frac{15(1-\nu_m)}{(8-10\nu_m)}\right) \quad (6)$$

In the foregoing equations, the symbol, *E* stands for Young's modulus, subscript *m* matrix, *p* filler, and *c* composite, while the symbol ϕ_p stands for the volume fraction of filler particles, and ϕ_m volume fraction of matrix.

Composite materials are usually classified by type of matrix such as polymer composites, cement, and metal matrix composites, which are typically divided into the three main categories of fibre-reinforced, particle-reinforced, and structural composites. The particle sizes are nearly the same in all directions or elongated for a distributed phase of particle-reinforced composites. Fibre-reinforced composites have strong fibres, which are continuous or discontinuous and are enclosed by a weaker matrix material. The work of this matrix is to protect the fibres and transmit load to the fibres [12, 13].

The mechanical properties of fibre reinforced composites (FRCs) are dependent on the type, length and shape of fibres used. For example, randomly oriented, evenly distributed short fibres of lengths between 1-30 μm form composites that are not fully isotropic. This type of fibre reinforcement is used mostly on low performance parts that are produced in great numbers. When FRCs use fibres that run all the length of the composite in only one direction, the mechanical properties of the composites vary with the orientation of the applied load with reference to the orientation of the reinforcing fibres. Such composites have highest mechanical properties in the longitudinal direction of the reinforcing fibres [14].

Filler particles are incorporated into materials such as plastics to lessen the use of additional expensive binder material and to enhance some mechanical and physical properties of the materials [15]. Thus, for example, the addition of industrial waste (fly ash) filler to kenaf fibre nano composites was observed to provide considerable improvement of tensile strength of kenaf fibre nanocomposites [16]. In sago starch-filled linear low-density polyethylene (LLDPE) composites, in which sago starch acts as a filler, tensile strength and elongation at breakpoint were observed to decrease with increasing sago starch content while the modulus was found to increase [11].

Ultrasonic through-transmission or pulse-echo techniques depend on the use of high-frequency mechanical oscillations for the detection of damage mechanisms, by measuring the signal amplitude and/or the time-of-flight of the ultrasonic signal to locate and size defects in a material [17]. Ultrasonic techniques are known to have the capability to characterize elastic properties of composites by processing the phase velocities of elastic waves that propagate in various directions in materials. They are more precise than static methods, especially for measuring shear properties [18]. Many authors have studied the ultrasonic velocity and mechanical properties of materials by use of the pulse echo method [19, 20, 22].

The research work presented here focuses on the use of theoretical models in predicting the mechanical properties of coconut shell powder/epoxy resin composites for different filler sizes and volume fractions and comparison of the predicted results with those obtained experimentally.

2. EXPERIMENTAL PROCEDURE

2.1 Materials

The materials shown in Table 1 were used to prepare CSP/epoxy resin composites.

Table 1: Details of the resin system used in the present work

Details of matrix, reinforcement, and release system		
Matrix Details		
Manufacturer	Gurit	
Resin type	Epoxy	
Resin Identification	Prime 27	
Matrix Hardener		
Product Name	Prime 27 slow hardener	
Filler Details		
Filler material	150 μm CSP	212 μm CSP
Release System		
Wax	Ram Wax	Ram Wax

2.2 Fabrication of the Composites

The coconut shell was dried in the open air and ground into powder thereafter using a pulverizing machine, and the powder subsequently sieved in accordance with BS 1377:1990 standard. The composite was fabricated using the method of hand lay-up. Firstly, the moulds for casting the test specimens were fabricated from glass with planer dimensions of 350 x 550 x 35 mm and a thickness of 7.5 mm. The mould was then coated with a transparent film of "ram-wax" on the inside to facilitate removal of the cast material from the mould upon cure. Coconut shell powder (CSP) was then added into acetone in a glass jar and the mixture stirred at a low frequency of 10 Hz using a scientific ultra-sonic bath at a room temperature of 23^{o}C for an hour in order to ensure an even distribution of the CSP in the acetone. The suspension was then added to epoxy resin in a jar, stirred and heated to a temperature of 140^{o}C in order to evaporate the acetone thus leaving behind coconut shell powder dispersed in epoxy resin. The mixture was then placed in a scientific ultra-sonic bath and stirred till the mixture temperature cooled down to 21^{o}C. After this, hardener was added into the mixture of resin and coconut shell powder in the ratio of 3.57:1(100:28) resin: hardener as recommended by the supplier and stirred thoroughly for about 10 minutes to obtain a uniform mixture. The thoroughly stirred mixture was then placed in a vacuum chamber for degassing under a -90 kPa vacuum for 20 minutes to remove any entrapped air bubbles. The mixture was thereafter poured into the glass mould, sealed, and allowed to cure for 8 hours. The cured composite was later post-cured in an oven for 8 hours at 65^{o}C. This whole process

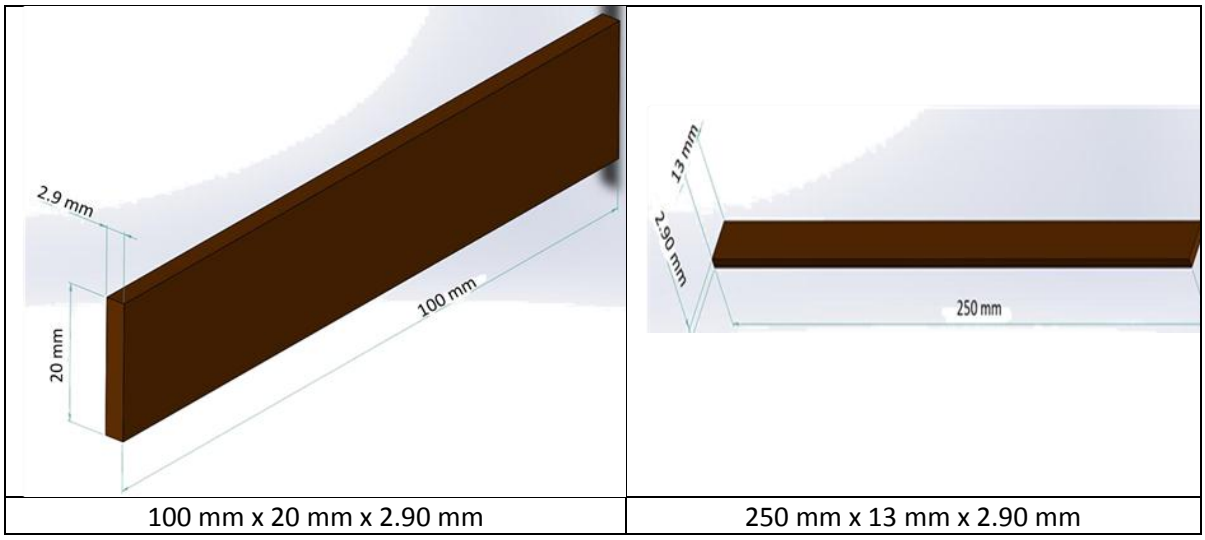


Figure 1: Schematic drawings of ultrasonic test specimens according to AST ASTM E2580 - 17

was repeated for the different weight percentages of (5, 10, 15, 20, 25 and 30) of the coconut shell particles. Thereafter, the specimens were cut to standard dimensions for each test to be carried out, using a CNC machine. The specimens were then used to determine the mechanical properties of elastic and shear moduli using ultrasonic testing equipment in accordance with ASTM standards E2580-17 [21]. A schematic drawing of the test specimens used with their dimensions is shown in Figure 1.

Figure 2a and 2b show the pouring of thoroughly mixed CSP and epoxy resin composite into a mould and cured composite inside an enclosed glass mould, respectively.

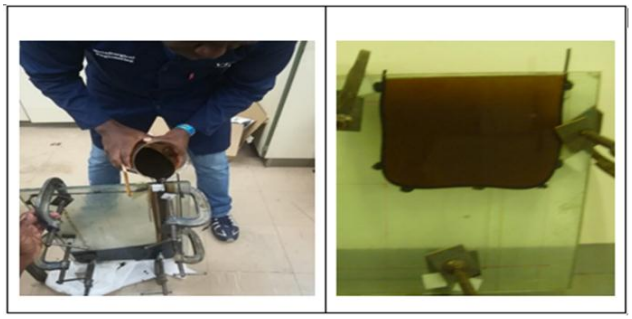


Figure 2: Pouring of the mixed reinforced coconut shell powder composite into a mould and a cast CSP epoxy resin/coconut particle composites.

2.3 Ultrasonic Through Thickness Testing of Coconut Shell Reinforced Composites

The non-destructive testing (NDT) technique used in this work is referred to as the pulse-echo technique. The composite specimens meant for ultrasonic characterisation were cut out of the moulded composite block and machined down to the two sizes of 250 mm × 13 mm × 2.9 mm and 100 mm × 20 mm × 2.9 mm, for both the 150 µm and 212 µm coconut shell filler particle size composite specimens, respectively. The velocity of sound was determined from the pulse-echo technique by the measurement of the time of flight between two successive back-wall echoes. Ultrasonic measurements of the velocities of compression and the shear waves in the specimens were performed at ten different locations along the length of each sample and the corresponding average used in this investigation. Three specimens were used for each weight percentage of coconut shell powder reinforced composite in these tests. The ultrasonic waves were generated using the ultrasonic flaw detector-Epoch LT panametric flaw detector, with a longitudinal transducer of 8 MHz at 0^o (10mm ø) and a shear transducer of 4 MHz at 70^o (10 mm ø).

The velocities of the propagated ultrasonic waves in this setup was obtained using Equation 7.

$$V = \frac{2t}{\Delta T} \quad (7)$$

where V is the velocity of a respective ultrasonic wave through the thickness of the composite, ΔT time interval for return of a respective ultrasonic wave, measured using an oscilloscope, and t thickness of the test specimens.

Figure 3 shows the ultrasonic NDT instrument used in this setup with the longitudinal waves at different velocities on its screen magnified in the two images on the right.

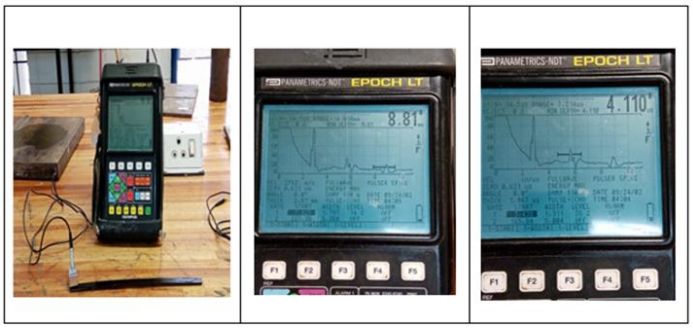


Figure 3: Ultrasonic testing equipment showing the transmitted and reflected ultrasonic waves

The longitudinal ultrasonic wave velocity (V_L) and the transverse or shear ultrasonic wave velocity (V_T) were evaluated using Equation 7. These two ultrasonic velocities were then used to calculate the shear moduli, and elastic modulus of the material according to Equations 8, and 9, respectively [22].

$$G = \rho V_T^2 \quad (8)$$

$$E = \frac{\rho V_L^2 (1+\nu)(1-2\nu)}{1-\nu} \quad (9)$$

where, E is elastic modulus, and G is the shear modulus of the material tested.

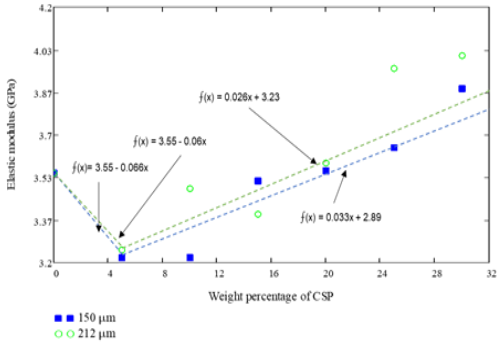
3 RESULTS AND DISCUSSION

3.1 Ultrasonic Through Thickness Testing of Coconut Shell Powder Reinforced Epoxy Resin Composites

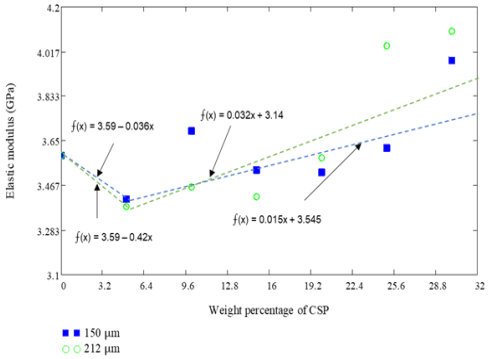
The results of elastic modulus, and shear modulus of CSP/epoxy resin composites with different weight percentages of the CSP filler are presented in Figures 4-8, together with their accompanying discussions.

3.1.1 Elastic and Shear Modulus

Figure 4 (a and b) shows the elastic modulus for both 150 µm and 212µm particle sizes of CSP/epoxy resin composites for test specimens of size 100 mm x 20 mm x 2.9 mm, and 250mm x 13mm x 2.9 mm.



a) (100 mm x 20 mm x 2.9 mm)

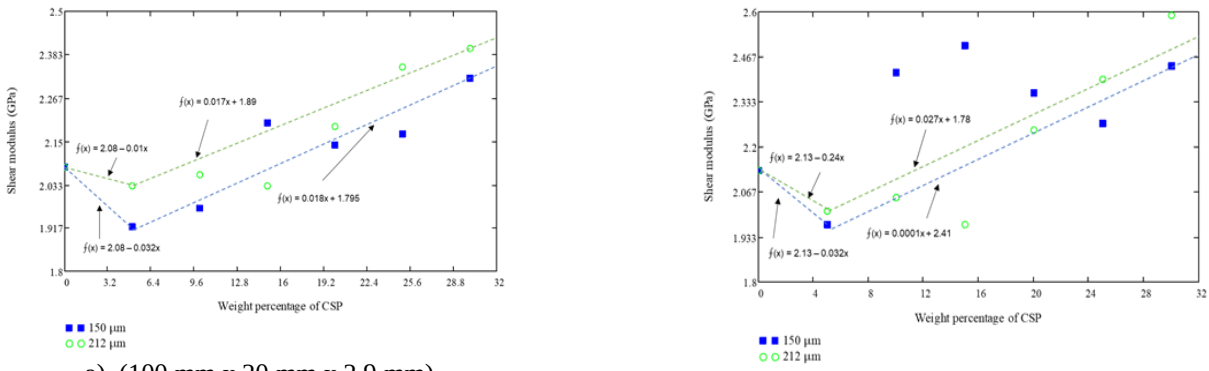


b) (250 mm x 13 mm x 2.9 mm)

Figure 4: Elastic modulus of CSP/epoxy resin composites at different weight percentages, for specimens with dimensions of 100 mm x 20 mm x 2.9 mm and 250 mm x 13 mm x2.9 mm

The elastic moduli of the control samples of pure epoxy resin are seen to be 3.54 GPa in Figure 4a and 3.59 GPa in Figure 4b. This is a small difference of about 1.4% and can safely be ignored. The figures show that with the introduction of coconut shell powder into epoxy resin matrix, there was an initial decrease of stiffness with increasing weight fraction till about 5wt% for the 150 µm and 212 µm filler particles CSP/epoxy resin composites in Figure 4(a and b). A small difference between this value for the 150 µm and 212 µm filler particles CSP/epoxy resin composites is barely visible in Figure 4(b). This weight fraction identifies the minimum reinforcing filler volume fractions, below which increasing the content of the reinforcing phase leads to a continuous decrease in the magnitude of the elastic modulus of the resulting composites below that of the pure matrix. This initial decrease is attributed to there being too few of CSP filler to transfer the load to, which is consistent with the work of Andezai et al. [23]. The critical filler weight fraction, the weight fraction below which the filler has no positive reinforcing effect, is seen in the two graphs in Figure 4(a and b), to be 19% and 21%, and 18% and 22.4% for the 212 µm and 150 µm filler particles CSP/epoxy resin composites, respectively. The linearised curves show highest values of elastic moduli of 3.79 GPa and 3.87 GPa, and 3.77 GPa and 3.91 GPa for the 212 µm and 150 µm filler particles CSP/epoxy resin composites, at a filler volume fraction of 32wt% of filler in Figure 4(a and b), respectively. The results plotted in Figure 4(a) show that the larger particles enhanced the elastic modulus of the epoxy resin more for all weight fractions and led to smaller reductions below the critical weight fraction in contradiction with the established effect of particle-size. The case for Figure 4(b) is mixed with lower values for the larger particles till a weight fraction of 11.2% where a crossover occurs. It is possible that the failure to adhere to the established fact of a higher reinforcement for smaller size particles is a result deviation of the shape of fillers from spherical to elongated, with a greater aspect ratio greater for the 212 µm than the 150 µm filler particles. This forms the subject of further research. The differing values of the curves shown in Figure 4(a and b) is likely to be the result of uneven distribution of the reinforcing fillers in the parent moulding from which the specimens were cut and is unlikely to be an effect of the different dimensions of the specimens.

Figure 5 (a and b) shows the shear modulus for both 150 µm and 212 µm particle size CSP/epoxy resin composites for test specimens of dimensions 100 mm x 20 mm x 2.9 mm and 250mm x 13mm x 2.9 mm.



a) (100 mm x 20 mm x 2.9 mm)

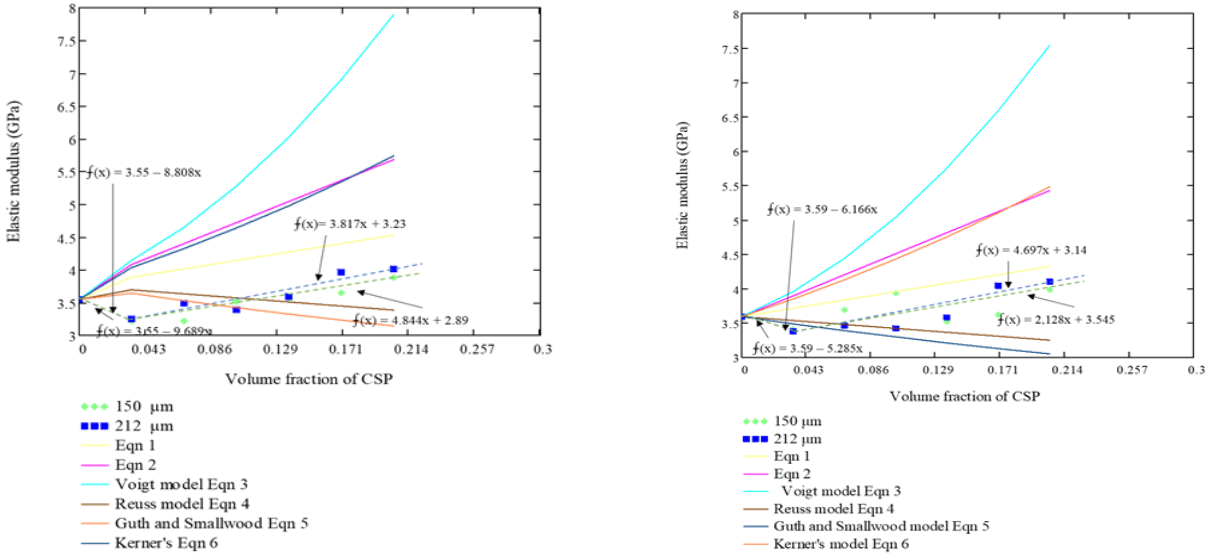
b) (250 mm x 13 mm x 2.9 mm)

Figure 5: Shear modulus of CSP/epoxy resin composites at different weight percentages with different particle sizes for specimen dimension of 100 mm x 20 mm x 2.9 mm and 250 mm x13 mm x 2.9 mm

The shear moduli for the control sample of pure epoxy resin are seen to be 2.09 GPa and 2.13 GPa in Figure (5a and 5b), respectively. The percentage difference between these values is small at 1.9% and can safely be ignored. There was an initial decrease in the shear modulus of the composites produced from samples of different weight fractions till 5wt% for both the 150 µm and 212 µm CSP particle sizes in the two figures. As was the case for elastic modulus, this is the minimum weight fraction below which increasing filler content leads to a decrease in the shear moduli of the CSP/epoxy resin composites. The curves in Figure 5(a and b) establish weight fractions of about 9.6% and 16%, and 12% and 16% for the 212 µm and 150 µm filler particles CSP/epoxy resin composites, respectively. Maximum values of shear modulus are derived from the linearised curves in Figure 5(a and b) of 2.43 GPa and 2.35 GPa, and 2.53 GPa and 2.47 GPa for the 212 µm and 150 µm filler particles CSP/epoxy resin composites, at a filler volume fraction of 32wt% of filler in, respectively. The effect of particle-size is seen clearly in these two figures where higher values of shear modulus are registered for the larger particle-size composites at all weight fractions, contrary to expectations from established theory which states that decreasing particle-size leads to an increase of reinforcement effect of the same trend is evident in the curves of Figure 4 (a and b) though to a smaller extent and is likely to be a result of increased agglomeration of particles with decreasing size of the filler particles, that effectively reduces the interfacial area available for bonding.

3.2 Comparison of Theoretical and Experimental Mechanical Properties of CSP/Epoxy Resin Composites

Figures 6 (a and b) shows the curves for the comparison of the elastic modulus of 150 and 212 µm CSP/epoxy resin composite specimens of dimensions of 100 mm x 20 mm x 2.9 mm and 250 mm x 13 mm x 2.9 mm against the volume fraction of coconut shell powder.



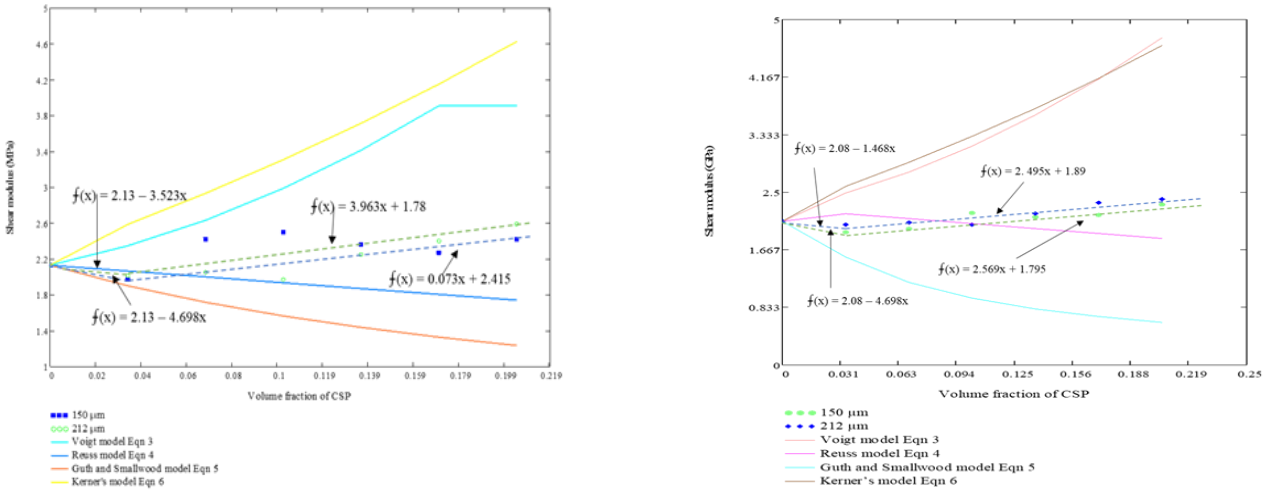
a) (100 mm x 20 mm x 2.9 mm)

b) (250 mm x 13 mm x 2.9 mm)

Figure 6: Comparison of experimental and predicted result of elastic modulus for CSP/epoxy resin composite specimens of dimensions 100 mm x 20 mm x2.9 mm and 250 mm x 13 mm x2.9 mm

The curves in Figure 6 (a and b) show that the values predicted by the Voigt model Equation 3, Kerner's model Equation 6, Einstein Equations 1 and 2, all showed non-linear increases of stiffness with increasing volume content of filler for both 150 µm and 212 µm coconut shell particle reinforced epoxy resin composites. The curves of these models were significantly higher than the experimental results, which implies that the models are of little value in predicting the elastic modulus of CSP/epoxy resin composites. Reuss model Equation 4 and Guth and Smallwood model Equation 5 decreased continuously with the increased volume fraction of the filler particles. Neither the Guth and Smallwood model nor the Reuss model are good in predicting the elastic modulus of the CSP/epoxy resin composites as they have curve profiles that are inverted relative to the experimental curves. With the exception of the Reuss and Guth and Smallwood equations that are close the experimental curves below a volume fraction of 4.3% for Figure 6(b), none of the other equations are useful for predictive purposes and it is recommended that alternative models be developed of sought.

Figure 7(a and b) shows curves of the shear modulus for 150 µm and 212 µm CSP/epoxy resin composite specimens of dimensions of 100 mm x 20 mm x 2.9 mm and 250 mm x 13 mm x 2.9 mm against the volume fraction of coconut shell powder.



a) (100 mm x 20 mm x 2.9 mm) b) (250 mm x 13 mm x 2.9 mm)

Figure 7: Comparison of experimental and predicted result of shear modulus of CSP/epoxy resin composite specimens for specimens of dimensions 100 mm x 20 mm x 2.9 mm and 250 mm x 13 mm x 2.9 mm

It is evident from Figure 7(a and b) that the curves plotted from Kerner's Equation 6, and Voigt Equation 3 give predictions that are way in excess of the experimental values and are hence not useful at all for prediction. The curves or the Guth and Smallwood model, and Reuss model, Equations 3 and 4, respectively, do show values that are proximal to those of the experimental curves for values of volume fraction below about 4% in Figure 7(a). The case is very different for Figure 7(b), where none of the theoretical curves come close to the experimental curves. This disparity for the two figures is expected to arise from different filler volume fractions based on the location of respective specimens on the moulded composite sheet. The closer proximity of the theoretical and experimental curves at lower filler volume fractions, implies better distribution of filler in the specimens related to Figure 7(a) than those for Figure 7(b).

4.0 CONCLUSIONS

The following conclusions can be drawn from this work:

1) The was an initial decrease in the stiffness of the CSP/epoxy resin composites with increasing filler content up until the minimum filler content. This is thought to be a result of filler surface areas that are too small to provide adequate interfacial bonding which them makes them act as voids.
- The effect of particle-size is seen clearly in these two particle sizes of 150 µm and 212 µm where higher values of shear modulus are registered for the larger particle-size composites at all weight fractions. This is likely to be a result of agglomeration of the smaller size filler particles with the attendant reduction in overall available surface area for bonding. Amongst Einstein Equations 1 and 2, Voigt Equation 3, and Kerner's model Equation 6, Reuss model Equation 4 and Guth and Smallwood model Equation 5, only the last two generated curves that were close to the experimental values, and this only at filler volume fractions less than 4%.

5.0 RECOMMENDATIONS

A study of a wider range of CSP particle sizes is recommended in order to determine trends and an optimum particle size. Moreover, the occurrence and nature of agglomeration and the

effectiveness of interfacial bonding particularly at low filler fractions should be studied in more detail.

6.0 CONFLICT OF INTEREST

The researchers have no conflict of interest to disclose with regard to the current research work.

7.0 ACKNOWLEDGEMENT

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