Mechanical properties of hybrid SiC/CNT filled toughened epoxy nanocomposite

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Abstract. Mechanical properties of epoxy nanocomposites filled single filler have been extensively studied by various researchers. However, there are not much discovery on the behavior of hybrid nanocomposite. In this study, single and hybrid nanocomposites of toughened epoxy filled CNT/SiC nanoparticles were investigated. The hybrid nanocomposites samples were prepared by combining CNT and SiC nanoparticles in toughened epoxy matrix via mechanical stirring method assisted with ultrasonic cavitations. Epoxy resin and liquid epoxidized natural rubber (LENR) mixture were first blend prior to the addition of nanofillers. Then, the curing process of the nanocomposite samples were conducted by compression molding technique at 130°C for 2 hours. The purpose of this study is to investigate the hybridization effect of CNT and SiC nanoparticles on mechanical properties toughened epoxy matrix. The total loading of single and hybrid nanofillers were fixed to 4% volume are 0, 4C, 4S, 3S1C, 2S2C, and 1S3C. Mechanical properties of hybrid composites show that the highest value of tensile strength achieved by 3S1C sample at about 7% increment and falls between their single composite values. Meanwhile, the stiffness of the same sample is significantly increased at about 31% of the matrix. On the other hand, a highest flexural property is obtained by 1S3C sample at about 20% increment dominated by CNT content. However, the impact strength shows reduction trend with the addition of SiC and CNT into the matrix. The hybridization of SiC and CNT show highest value in sample 1S3C at about 3.37 kJ/m² of impact energy absorbed. FESEM micrograph have confirmed that better distributions and interaction observed between SiC nanoparticles and matrix compared to CNT, which contributed to higher tensile strength and modulus.

1. Introduction
Nanocomposite materials have gained a great attention to the various fields recently. The used of this material in high performances application such as electronics, telecommunications, defense, aerospace possessed high surface area, therefore polymer nanocomposite offer enhanced mechanical, thermal, barrier and flame retardant properties compared to conventionally filled materials [2]. The discovery of CNT by [3] brought the explosion of nanotechnology in various field. Due to this reason, CNT
based product received high demand for bulk production and currently are available in open market [4].

However, there are some problems facing by composite industry in nanocomposite production. According to [5], the major problem in nanocomposite materials production is the dispersion and agglomeration of nanofiller in matrix system. These phenomena will reduce the mechanical properties of nanocomposite and limits their usage in specific applications. The other problem is the use of single nanofiller in a polymer matrix cannot fulfill certain requirement for specific application. Thus, hybrid composite has been proposed in order to solve these problems.

Hybrid nanocomposite is a composite system produced by combining two types or more nanofillers in a matrix system. Hybridization of different nanofiller in one matrix system is a new approached used by several researchers to solve a problem that cannot be attained by single filler [6]. Other researcher [7] claimed that the hybridization of different filler in a matrix system are able to optimized the composite properties and complement the other filler weaknesses. Two or three types of filler present in single matrix not only improved the mechanical properties, but electric, magnetic and microwave absorption as well.

Tensile strength of hybrid composite have similar curve as single nanocomposite but changed from linear to nonlinear due to inconsistent hybrid filler content [8]. Study by [9] on hybrid composite of multiwalled carbon nanotube (MWCNT) and organoclay montmorillonite (OMMT) in epoxy matrix found that impact strength of nanocomposite has been increased and the value is falls between their single filler nanocomposite.

In this study, hybrid composite of CNT and SiC nanoparticle were combined in order to study the effect of their hybridization on mechanical properties of toughened epoxy nanocomposite.

2. Experimental procedures

Toughened epoxy matrix were first prepared by blending epoxy resin with liquid epoxidized natural rubber (LENR) by using mechanical stirrer assisted with ultrasonic cavitations for 1 hour at 1300 rpm. During stirring, the SiC nanoparticles were added and mixed for 15 minutes prior to the addition of CNT addition. Then, the mixture was further mixing for 45 minutes followed by the addition of crosslinking agent at the ratio of 1:3 to the resin amount. After that, the mixture was degassed in vacuum oven for 5 minutes at room temperature for entrapped air removal. The mixture were then poured onto mould and followed by pre-curing at room temperature for 12 hour before proceed to the post cure at 130°C for 2 hours. The mechanical tests were carried out using Testometric M350-10CT Universal Testing Machine. Type I dumbbell shape of tensile sample were prepared according to the ASTM D638. Flexural properties of the hybrid nanocomposite were measures according to ASTM 790 three point bending test while Izod impact tests were performed according to ASTM D256.

3. Results and Discussion

The mechanical properties of single and hybrid nanocomposite were determined by comparing the tensile strength and modulus at different loading condition of CNT and SiC nanoparticles. Measuring the materials behavior under tension load with the present of two different nanofiller were done to study the effects of CNT/SiC nanoparticle loading on mechanical properties of single and hybrid nanocomposites. The tensile properties of toughened matrix, single filled composite and hybrid composite are summarized in figure 1 (a) and (b).

It can be seen that the tensile strength and Young’s modulus increased with the addition of 4% single nanofiller of CNT or SiC nanoparticles in toughened epoxy matrix. However, the addition of 4% SiC nanoparticles shows slightly higher value in tensile strength as compared to 4% CNT and similar trend are observed in Young’s modulus value which, the modulus of SiC nanoparticle composite received greater value than CNT nanocomposite. This is because higher forced is required to break the sample due to plastic deformation increases during crack deflection and pinning [10]. The
matrix chain segment pinning due to the increasing of filler content will increase the stiffness of nanocomposite [11].

![Figure 1](image1.jpg)

**Figure 1.** (a) Tensile strength and (b) Modulus elasticity of toughened epoxy matrix, single and hybrid SiC/CNT nanocomposites.

Figure 2 show the flexural (a) strength and (b) modulus comparison between single and hybrid composite of SiC and CNT filled toughened epoxy nanocomposite. It is obvious that the addition of 4% SiC and CNT in toughened epoxy matrix has increased the flexural strength and modulus. However, CNT nanocomposite of flexural strength and modulus value is higher than SiC nanocomposite.

Hybridization of SiC and CNT in the same toughened epoxy matrix reduces the flexural strength and modulus value but remain higher than matrix. This is due to the domination of CNT content proved that CNT nanotubes provide better flexural properties than SiC nanoparticle. In hybrid nanocomposite sample dominated by SiC content. The presence of large SiC nanoparticle amount restricted the CNT aggregation and increased the tensile strength and modulus. Meanwhile, in hybrid sample dominated by CNT agglomeration acted as a stress concentration sites thus reduced the tensile strength [4].

![Figure 2](image2.jpg)

**Figure 2.** (a) Flexural strength and (b) Flexural modulus of the toughened matrix, single and hybrid SiC/ CNT nanocomposites

The impact strength of single and hybrid nanocomposite of SiC and CNT filled toughened epoxy is shown in Figure 3. There is significant reduction in impact strength with the addition of CNT and SiC nanoparticle in the matrix. However, higher value is observed in CNT compared to SiC.
nanocomposite. The impact strength value of hybrid nanocomposite falls between single nanocomposite of CNT and SiC nanoparticle. Hybrid sample dominated CNT at 1%S+3%C remain higher than sample dominated by SiC of 3%S+1%C.

![Figure 3. Impact strength of matrix, single and hybrid SiC/CNT nanocomposites](image)

Domination of 3%C+1%S hybrid composite is due to high aspect ratio of CNT requires more energy for crack propagation during impact. Therefore, the energy required to break the sample dominated by CNT nanofiller is higher than the sample dominated by SiC. The stiffness behavior of SiC nanoparticle itself contributed to the stiffness of the composites, thus low energy is needed to break SiC nanocomposite sample.

Figure 4 depicts selected FESEM images of the toughened epoxy matrix and single filled of SiC and CNT nanocomposite fractured surfaces. It clearly can be seen in figure 4(a) that a smooth fractured surface of toughened epoxy matrix with the presence of dispersed rubber particles. Rough surface CNT and SiC nanoparticle show a good distribution on figure 4(b) and (c) support the improvement of tensile strength and Young’s Modulus as compared to the matrix. According to [3], rough surface produced by crack propagation changes deviated from original plane. However, there are fiber pullouts of CNT and leave cavity by fiber pullout can be observed samples in figure 4(c) that affected to the reduction of strength in CNT nanocomposite. Meanwhile, the improvement of tensile properties in hybrid nanocomposite sample dominated by SiC nanoparticle proved complete coverage of toughened epoxy matrix onto the SiC nanoparticle surfaces.

Good distribution of SiC nanoparticle will increased the density and mechanical properties of the hybrid composite. Meanwhile, inhomogeneous distribution of CNT nanofiller leads to the nanoparticles interaction by weak Van Der Walls thus reduces reinforcing behavior of filler in matrix [12].
Figure 4. Tensile fracture surface morphology of (a) toughened epoxy matrix (b) 4% S nanocomposite and (c) 4% C (d) 1C3S, (e) 2S2C and (f) 3C1S hybrid nanocomposites.

4. Conclusion
In summary, the presence of CNT and SiC nanoparticles in toughened epoxy matrix has increased 6% of the tensile strength and 12% stiffness respectively. The flexural strength and modulus of SiC and CNT nanocomposite are also increased at about 37% and 25% respectively. However, the impact properties are reduced at about 25% and 47% respectively for both SiC and CNT nanocomposite systems. Meanwhile, the hybridization of SiC with CNT in the toughened epoxy matrix has falls the
mechanical properties between the value of their single nanocomposite. Domination of SiC nanoparticles content are clearly can be seen in higher tensile strength of 47MPa and Young’s modulus of 1442 MPa for 3S1C in hybrid nanocomposite sample. The optimum flexural strength achieved by 1S3C sample at about 85 MPa and the optimum impact strength of hybrid nanocomposite is also observed by 1S3C at about 3.37kJ/m².

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