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Preparation and Characterization of Tensile Properties of PMMA/SiC Nanowhiskers Nanocomposite Films: Effect of Filler Loading and Silane Treatment

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ABSTRACT

The preparation of nanocomposites through melt mixing was challenging as the nanofillers tend to form agglomeration. The silicon carbide nanowhiskers (SiCNWs) filled poly (methyl methacrylate) (PMMA) thin film in this study was prepared by means of solution casting. Acetone with low toxicity was used as solvent to dissolve the PMMA pellets. A coupling agent, silane was used to enhance the properties of composite films. Besides, the untreated and treated SiCNWs were filled into PMMA matrix, respectively with the filler loading varied from 0.2 to 0.8 wt%. The universal testing machine was used to investigate the tensile properties of composites. It was found out that the tensile strength of the PMMA was reduced in the presence of SiCNWs. However, the tensile strength had increased with the rise of filler loading. At 0.8 wt% of SiCNWs, the composites' tensile strength was comparable to virgin PMMA. Meanwhile, the SiCNWs had reduced the elongation at break but increased the elastic modulus of PMMA/SiCNWs nanocomposite films. In addition, silane surface treatment on SiCNWs had improved the tensile strength and ductility but lowered the elastic modulus of the nanocomposites. The improvement was due to the enhancement of interfacial adhesion between SiCNWs and PMMA.

Keywords. PMMA, solution casting, silicon carbide nanowhiskers, nanocomposites, silane

1. INTRODUCTION

Poly(methyl methacrylate) (PMMA) is one of the common petroleum based thermoplastics. It is known to have good thermal stability, surface hardness, biocompatibility and environmental inert. Recently, PMMA nanocomposite films incorporated with zinc oxide, tin oxide and titanium dioxide, respectively were reported as promising materials in the development of optoelectronic, organoelectronic, and microelectronic devices [1]. Besides, PMMA also can be used as porous biomedical implant [2] while composites of PMMA filled with alumina are utilized in biomedical applications [3].

The properties of PMMA can be customized by incorporation of different types of functional fillers. In this study, silicon carbide in nano-sized is selected as filler in the fabrication of PMMA composites thin films. Silicon carbide has high thermal conductivity and mechanical strength, wide energy band gap and excellent oxidation resistance [4]. Its unique properties of SiC had

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drawn the interest of many researchers. Kamil et. al. for example reported the physical properties of SiC nanoparticles filled PMMA [5]. They found that SiC improved thermal conductivity, surface hardness and surface roughness of PMMA for the application of denture fabrication.

The processing of polymer composites can be conducted by using melt mixing method such as extrusion, injection, or compression moulding. These processing methods require relatively high cost of equipment. In addition, high temperatures where temperatures above the melting point of the polymer is required for the processing. Thus, it further increases the cost of production. Solution casting offers a simple and economic way to prepare composites thin films. Previously, authors had reported the preparation PMMA thin films filled with palm kernel shell [6]. Moreover, it is not easily to obtain homogeneous nanocomposites through melt mixing. The nano-sized fillers with high surface energy tend to form agglomeration and resulting in poor dispersion of fillers. Therefore, the PMMA/SiCNWs nanocomposites were prepared by means of solution casting. The effect of SiCNWs loading and silane on the tensile properties of PMMA/SiCNWs nanocomposites were investigated.

2. METHODOLOGY

2.1 MATERIALS

The polymethyl methacrylate (PMMA) was purchased from LG Chemical, Korea with the MFI value of 2.50 g/10 min at 230 °C and specific gravity of 1.18 g/cc. The silicon carbide nanowhiskers (SiCNWs) that served as filler was purchased from Nano Amor, USA. The range of diameter and length were 0.1 – 2.5 μm and 2.0 – 5.0 μm, respectively. The coupling agent used was 3-(Trimethoxysilyl)propyl methacrylate (TMSPMA). It was obtained from Sigma-Aldrich Chemie GmbH. The acetic acid and acetone were purchased from HmbG Chemicals, Syarikat Saintifik Jaya, Shah Alam, Selangor.

2.2 SAMPLES PREPARATION AND CHARACTERIZATION

SiCNWs was silanized with 6 ml of TMSPMA dissolved in a 100 ml mixture of ethanol/water (80/20). Then, the pH of the solution was adjusted to 4 by adding some acetic acid. The mixture was magnetic stirred homogeneously for 2 hours at room temperature. Then SiCNWs was filtered and washed with ethanol to remove excess silane. After that the silanized SiCNWs was dried in an oven for 48 hours at 80 °C.

By using solution casting, the untreated and treated SiCNWs filled PMMA composites were prepared. PMMA pellets were dried in oven at 70 °C for 24 hours to remove excessive moisture. Then they were dissolved in acetone by weight ratio 1:6 and stirred 6 hours using magnetic stirrer. The SiCNWs was added into the PMMA solution and subjected to vigorous stirring process until homogenous. After that, the mixture was ultrasonicated for 1 hour at 35 °C. Lastly, the mixture was casted into petri dish and dried for 24 hours at room temperature. A similar procedure was repeated with TMSPMA treated SiCNWs to produce treated PMMA/SiC composites. Table 1 shows the formulation of the untreated and treated SiCNWs filled PMMA composites.

Table 1. Formulation of untreated and treated PMMA/ SiCNWs nanocomposites.

Materials	Untreated PMMA/ SiCNWs	Treated PMMA/ SiCNWs
PMMA (wt %)	100, 99.8, 99.6, 99.4, 99.2	100, 99.8, 99.6, 99.4, 99.2
SiC (wt %)	0, 0.2, 0.4, 0.6, 0.8	0, 0.2, 0.4, 0.6, 0.8
TMSPMA (vol %)	-	6

The PMMA/ SiCNWs nanocomposites in the form of thin films were subjected to tensile test by using Universal Testing Machine (UTM) Instron 5569 and with reference to ASTM D882. Rectangular specimens were prepared in the dimension of 80 mm × 15 mm with the crosshead speed of 1 mm/min. For each formulation, at least five samples were prepared and tested. The values of tensile strength, Young's modulus and elongation at break of the composite films were taken and the average values were calculated.

3. RESULTS AND DISCUSSION

The tensile strength of both untreated and treated PMMA/SiCNWs nanocomposite films at various SiCNWs loading is shown in Figure 1. Initially, presence of 0.2 wt% SiCNWs had reduced 25% tensile strength of PMMA/SiCNWs nanocomposites. The nano-sized SiCNWs has high surface energy and tends to form agglomeration. This agglomeration then acts as stress concentrator that weakens the tensile strength of PMMA/SiCNWs composites. This finding was in agreement with the literature that showed nanoparticles like carbon nanotubes had negative impact on mechanical properties of nanocomposites due to the agglomeration [7]. However, the rise of SiCNWs had resulted in increased tensile strength of the composites. The tensile strength of the composite at 0.8 wt% SiCNWs loading was found to be comparable to that of the virgin PMMA. The low interfacial interaction between PMMA matrix and SiCNWs had caused mechanical rupture occurred at interface region. The hydrophilic SiCNWs was not compatible with the hydrophobic PMMA. Hence, the issue of incompatibility nature had weakened the interfacial interaction by causing voids and gaps between filler and matrix. Lule *et. al.* reported that the SiC particles had reduced the tensile strength of polybutylene adipate terephthalate/polycarbonate blends as a results of the poor interfacial adhesion and voids formation [8]. Therefore, addition of coupling agent was aimed to enhance the interfacial bonding between SiCNWs and PMMA.

Besides, the tensile strength of the treated and untreated PMMA/SiCNWs nanocomposites were compared. In a similar study, Ike-Eze and coworkers reported that silane treatment was the most effective surface treatment reagent, compare to others treatment such as acetic acid and sodium sulphite, to improve the tensile properties of banana fiber reinforced polyester composites [9]. At similar filler loading, the silane treated PMMA/SiCNWs nanocomposites films had higher tensile strength compared to the untreated ones. The silane treatment on SiCNWs enhanced the tensile strength as much as 10.3% at 0.8 wt% filler loading while the least increment of tensile strength of treated PMMA/SiCNWs nanocomposites was 4.2% at 0.6 wt% SiCNWs loading, relative to the untreated PMMA/SiCNWs nanocomposites.

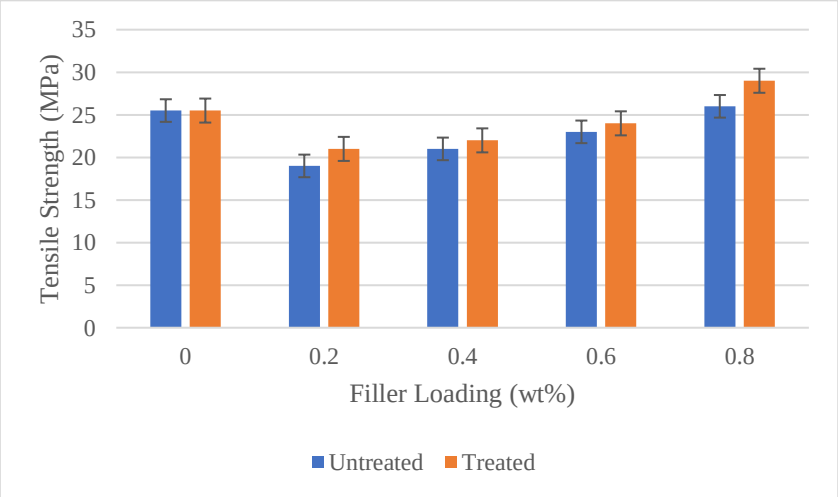

Figure 1. Tensile strength for treated and untreated PMMA/SiCNWs nanocomposites at different filler loading.

Figure 2 illustrates the effect of SiCNWs loading on the elongation at break of untreated and treated PMMA/SiCNWs nanocomposites. It was found that presence of SiCNWs in PMMA reduced the ductility of PMMA composites. The elongation at break of the composites decreased with the increase of SiCNWs loading. SiCNWs restricted the movements of PMMA polymer chains under tensile stress. As the SiCNWs loading increases, the nanocomposites became more compact, and it further prevent deformation to take place. On the other hand, silane treatment could improve the ductility of PMMA/SiCNWs composites. The treated nanocomposites had higher elongation at break compared to untreated nanocomposites at similar SiCNWs loading. The elongation at break of the composites was increased from 2.8 % to 4.0 % when 0.2 wt% of treated SiCNWs was added into PMMA. The increment continued to rise with the SiCNWs loading and reach 56.5% at 0.8 wt% SiCNWs loading. It was believed that treated SiCNWs can minimize agglomeration and distribute better in the PMMA matrix. Similar findings were reported by Zhang et. al. [10] in the study of poly(butylene adipate-co-terephthalate) (PBAT)/calcium carbonate composite films. In their study, silane was capable in preventing agglomeration of particles and minimizing driving force for crack initiation.

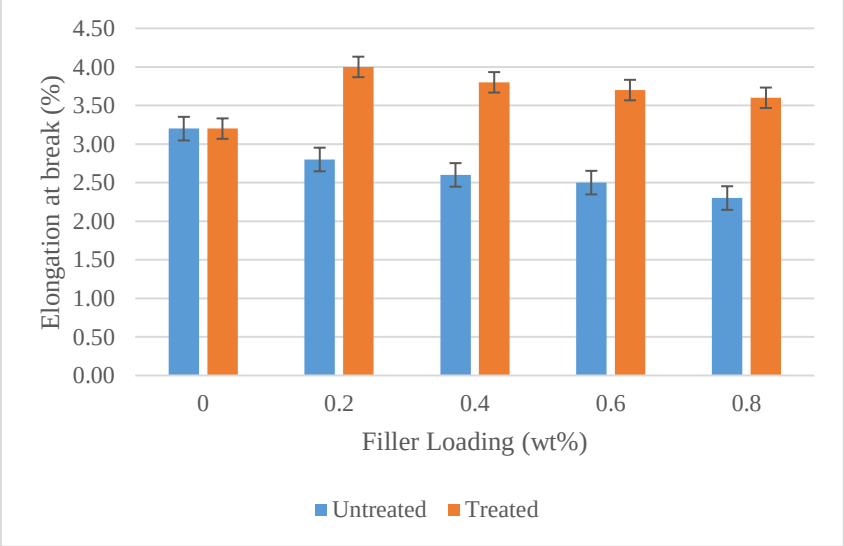

Figure 2. Elongation at breaks for treated and untreated PMMA/SiCNWs nanocomposites at different filler loading.

The elastic modulus of untreated and treated PMMA/SiCNWs nanocomposites films with different SiCNWs loading was shown in Figure 3. It was found that the Young's modulus of the composites increased with increasing SiCNWs loading. In another word, the stiffness of the composites was enhanced with the addition of SiCNWs. At the same time, the Young's modulus of the silane treated PMMA/SiCNWs composites were also increased with the filler loading. However, the treated composites showed lower values of Young's modulus as compared to the untreated counterparts. It is worth noting that the stiffness of the treated composites with SiCNWs loading less than 0.4 wt% was lower than that of the virgin PMMA. As the SiCNWs loading increased to 0.6 and 0.8 wt%, the elastic modulus of composites was increased slightly. Similar findings were reported by Bouza and coworkers in the study of wood flour filled polypropylene composites. In their study, silane was capable of improving filler dispersion and preventing the inclusion of matrix in filler, which reducing the rigidity of composites [11].

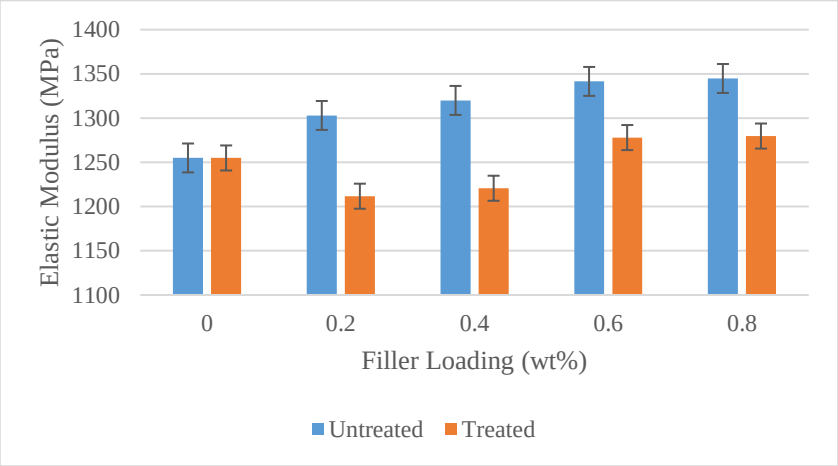

Figure 3. Young's modulus for treated and untreated PMMA/SiCNWs nanocomposites at different filler loading.

4. CONCLUSION

In conclusion, the PMMA/SiCNWs nanocomposites had been successfully prepared by using solution casting. The finding of the study revealed that tensile properties of PMMA/SiCNWs nanocomposites films were dependent on the SiCNWs loading and presence of silane as coupling agent. Though the addition of SiCNWs had initially decreased the tensile strength of composites, the tensile strength of composites at 0.8 wt% was comparable to pure PMMA alone. Moreover, the SiCNWs as filler had reduced the elongation at break of the composites yet increased its Young's modulus. On another hand, presence of silane as coupling agent had enhanced the tensile strength and elongation at break of PMMA/SiCNWs nanocomposites but decreased the Young's modulus of composites. Silane had improved the filler dispersion by minimizing the agglomeration of nano-sized SiCNWs.

5. ACKNOWLEDGEMENT

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