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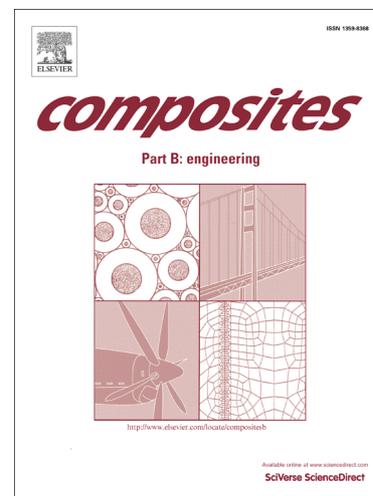
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Enhanced conductivity behavior of polydimethylsiloxane (PDMS) hybrid composites containing exfoliated graphite nanoplatelets and carbon nanotubes

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Abstract

Polydimethylsiloxane (PDMS) hybrid composites consisting of exfoliated graphite nanoplatelets (xGnPs) and multiwalled carbon nanotubes functionalized with hydroxyl groups (MWCNTs-OH) were fabricated, and the effects of the xGnP/MWCNT-OH ratio on the thermal, electrical, and mechanical properties of polydimethylsiloxane (PDMS) hybrid composites were investigated. With the total filler content fixed at 4 wt%, a hybrid composite consisting of 75% x GnP/25% MWCNT-OH showed the highest thermal conductivity (0.392 W/m.K) and electrical conductivity (1.24×10^{-3} S/m), which significantly exceeded the values shown by either of the respective single filler composites. The increased thermal and electrical conductivity found when both fillers are used in combination is attributed to the synergistic effect between the fillers that forms an interconnected hybrid network. In contrast, the various different combinations of the fillers only showed a modest effect on the mechanical behavior, thermal stability, and thermal expansion of the PDMS composite.

Keywords

B. Mechanical properties; B. Thermal properties; B. Electrical properties

1. Introduction

Carbon nanotubes (CNTs) made from graphene layers folded into a cylindrical hexagonal lattice structure exhibit excellent mechanical properties [1,2]. A single carbon nanotube is one hundred times stronger than steel, one-sixth of the density of steel, and exhibits both high electrical and thermal conductivities. The cylindrical shape of the filler results in only a small concentration being required to dramatically change a wide range of the properties of a polymer composite. As a result, carbon nanotubes (CNTs) have attracted a great deal of interest in the polymer composite research community. Compared with the enormous number of studies on the application of CNTs in epoxies, thermoplastics, and fibers, there are relatively few reports dealing with applications of CNTs in rubber [3].

The addition of CNTs to polydimethylsiloxane (PDMS), a type of silicone elastomer, improves the mechanical, electrical, and thermal properties of the resulting composite. Our recent work revealed that the electrical conductivity of a CNT/PDMS composite increased by five orders of magnitude when as little as 1 wt% pristine MWCNTs are added to the PDMS matrix. In contrast, the thermal conductivity of the CNT/PDMS composite showed a 38% improvement with the addition of 4 wt% pristine MWCNTs [4]. The improvement of the thermal conductivity in CNT/PDMS composites has also been reported by Chua et al. [5] and Hong et al. [6]. In a separate work, Wu et al. [7] reported that the elastic modulus of a CNT/PDMS composite increased when 2.0 wt% CNTs were added to the PDMS matrix. Furthermore, the storage modulus and

hardness also increased with the addition of CNTs. The addition of functionalized CNTs has been reported to improve CNT dispersion due to strong interfacial adhesion with polymers. This subsequently results in better mechanical and functional properties of CNT nanocomposites [9].

Another emerging type of carbon-based filler is the exfoliated graphene nanoplatelet (xGnP™). Previous work has demonstrated that xGnPs, which have a graphitic structure, also have many of the superior thermal and electrical properties of CNTs when incorporated into a polymer composite. This might make xGnPs a promising and less expensive alternative filler in polymer composite systems when compared with CNTs [8, 10]. It has also been reported that xGnP-filled high-density polyethylene (HDPE) composites show superior mechanical properties, whereas an xGnP-filled linear low-density polyethylene (LLDPE) displays excellent mechanical properties coupled with good electrical conductivity [11].

In recent years, using combinations of two or more conductive fillers to produce polymer/hybrid fillers composites has gained increased attention. Li et al. [12] made an epoxy-filled composite with hybrid GNP and CNT composites with a total mass fraction of 2 wt% and studied the effect of varying individual CNT/GNP contents on the electrical, flexural, and fracture properties of epoxy composites. Ma et al. [13] studied hybrid fillers of CNT and carbon black (CB). It was found that the addition of CNTs in CB composites enhanced the electrical conductivity of the composites. Yang et al. [14] also combined fibrous 1-D MWCNTs with the planar 2-D multi-graphene platelets to create a 3-D graphene-based architecture that exhibited a synergistic effect on the mechanical and thermal conductivity of epoxy composites. The tensile modulus, tensile strength, and thermal conductivity of 0.9 wt% MGP and 0.1 wt% MWCNT epoxy

composites were improved by 23%, 15%, and 47%, respectively, compared with a pure epoxy system. Kumar et al. [15] used a simple and innovative solution-mixing approach to fabricate polyethylenimine (PEI) filled with MWCNT and xGnP hybrid fillers with a fixed loading of 0.5 wt%. The hybrid composite exhibited a 45% increase in thermal conductivity when compared with pure PEI, whereas the PEI filled with 0.5 wt% single filler xGnPs and or PEI filled with 0.5 wt% single filler MWCNTs only exhibited a 22% or 9% increase, respectively. The hybrid xGnP/MWCNT composite containing equal amounts of two geometrically different fillers (0.25 wt% xGnPs and 0.25 wt% MWCNTs), showed a surface resistivity 8 orders of magnitude lower than that of a composite with 0.5 wt% single filler xGnPs alone. All these results demonstrate that incorporating hybrid fillers of both xGnPs and MWCNTs in combination into polymers can result in a significant change to the behavior of the polymer, especially the electrical and thermal conductivity behavior.

Even though combining CNTs and xGnPs as hybrid fillers has shown synergistic effects in enhancing the electrical and thermal conductivity of composites, especially in epoxy and polyethyleneimine (PEI) [13-15], no comprehensive studies on the use of CNT/xGnP hybrid fillers in an elastomer such as PDMS have been reported. It was therefore decided to investigate the electrical and thermal conductivity and the mechanical properties of PDMS composites. These composites might also display significant synergy with the filler with the potential for significant cost savings because xGnPs are significantly cheaper than CNTs [16].

2. Experimental

2.1 Materials

PDMS (Sylgard 182 Silicone Elastomer) consisting of a base elastomer (Part A) and a curing agent (Part B) was purchased from Dow Corning, USA. The ratio of Part A to Part B is 10:1. MWCNTs-OH, measuring 30–50 nm in diameter and 0.5–2 μm in length, of 95% purity, were purchased from Nanostructured and Amorphous Materials, Inc., USA. The exfoliated graphite nanoplatelets used were purchased from XG Sciences, Inc., USA. Each particle consists of several sheets of graphite with an overall average thickness of 6–8 nm, average particle diameter of 5 μm , and a typical surface area of 120–150 m^2/g . The nanoplatelets have naturally occurring functional groups such as ethers, hydroxyls, or carboxyls that can react with atmospheric humidity to form acids and other compounds. The solvent used to disperse the fillers was toluene (99.9%) purchased from J.T. Baker.

2.2 Fabrication of a PDMS/xGnP/MWCNT-OH Hybrid Composite

MWCNTs-OH and xGnPs were separately dispersed in toluene according to a weight ratio of 1:20 and sonicated in an ultrasonic bath for 1 hour. After that, both the toluene-xGnP and toluene-MWCNT-OH suspension were combined and dispersed for an additional 30 minutes. Part A of the PDMS was then added to the suspension. The mixture was mechanically stirred at 400 rpm and 70 $^{\circ}\text{C}$ for 1 hour to evaporate the toluene. After the mixture was cooled to room temperature, Part B was added, and the mixture was mechanically stirred at 800 rpm and room temperature for 10 minutes. The mixture was cast into a 1.5 mm-thick mold, followed by degassing under vacuum for 1

hour before being cold compressed for 1 minute. Lastly, the compressed mixture was cured in an oven at 150 °C for 2 hours. Based on our recent findings, the PDMS/MWCNT-OH and PDMS/xGnP single filler composites show the highest conductivity value at 4 wt% filler loading [4,17]. Hence, the total weight fraction of the hybrid filler was maintained at 4 wt%. Hybrid composites with an xGnP content of 0%, 25%, 50%, 75%, and 100% were produced according to the above steps.

2.3 Characterization

The surface morphology of the MWCNT-OH and xGnP was observed using the JEOL-2010 high-resolution transmission electron microscope. The thermal conductivity of the composites was obtained using a hot disk thermal constant analyzer (TPS 2500S). The electrical conductivity of the composites was measured with an Advantest R8340 ultra-high resistance meter. This test was carried out at room temperature with a constant voltage of 500 V. Tensile tests were conducted with an Instron 3366 universal testing machine. The fracture surface of the composites was observed using an FEI Quanta 3D environmental scanning electron microscope. Thermogravimetric analysis (TGA) was carried out to study the thermal stability of the composites. The test was conducted using a Perkin Elmer Pyris™ TGA, heated from room temperature to 800 °C (heating rate: 10 °C/min) in a nitrogen atmosphere. A thermomechanical analysis (TMA) was conducted to determine the coefficient of thermal expansion (CTE). Test specimens 1 mm thick and 5 mm in diameter were tested using a Perkin Elmer TMA7 Thermomechanical Analyzer. The expansion probe was rested on the test specimen with a static force of 50 mN, heated from -150 °C to 250 °C at a heating rate of 10 °C/min, and purged with nitrogen gas at the rate of 20 ml/min.

3. Results and Discussion

3.1 Surface morphology of fillers

The TEM images of pristine MWCNTs, MWCNTs-OH, and xGnPs are shown in Figures 1(a-c), respectively. Figure 1(a) reveals that the walls of MWCNTs-OH are heterogeneous, rough, and stained with extra materials compared with pristine MWCNTs (Figure 1(a)). This is believed to be due to acidic etching of the nanotube's surface during chemical (acid) treatment to introduce the functional groups [18]. The TEM image of xGnP particles is shown in Figure 1(c). The xGnP particles were observed to have a platelet shape with a much larger lateral dimension.

3.2 Morphological Study of Hybrid Fillers in PDMS

SEM was used to observe the surface morphology of the fracture surfaces and the distribution of hybrid fillers in the PDMS. Figures 2(a) and 2(b) show the fracture surface of PDMS filled with 75% and 50% xGnPs, respectively, at a high magnification. An even higher magnification is shown in Figure 2(c) to show the fracture surface of PDMS filled with 75% xGnPs. It is observed that varying the xGnP/MWCNT-OH ratio (% xGnP) has some effect on the dispersion of fillers. From Figure 2(a), the xGnPs and MWCNTs-OH are better distributed in comparison to the xGnPs and MWCNTs-OH in Figure 2(b). Small clusters of agglomerated nanotubes can be observed in Figure 2(b), as indicated by the red circles. Decreasing the amount of xGnPs and increasing the amount of MWCNTs-OH increases the probability of the nanotube fillers forming agglomerates.

The highest magnification reveals that there is geometrical synergy between the nanoplatelets and nanotubes in which the nanoplatelets and nanotubes form an interconnected hybrid network structure, which consists of nanotubes forming multiple junctions among themselves and the exfoliated graphite nanoplatelets. Figure 2(c) shows that the MWCNTs-OH formed multiple junctions among themselves and the exfoliated graphite nanoplatelets, as indicated by yellow circles. It is deduced that this combination of the fillers, when incorporated within the PDMS matrix, shows an even higher interconnected network with more junctions. Figure 3 illustrates the suggested formation of an interconnected hybrid network between xGnPs and MWCNTs-OH. The nanotubes form junctions among the nanoplatelets by connecting two or more together. Such a network has also been reported by Yang et al. [14] and Kumar et al. [15] for other hybrid polymer composite systems.

3.3 Thermal Conductivity

The effect of varying the ratio of xGnPs to MWCNTs-OH at a fixed total filler weight fraction on the thermal conductivity of PDMS hybrid composites is shown in Figure 4. By varying the ratio of xGnPs to MWCNTs-OH from 0 to 100%, the thermal conductivity of hybrid composites might be expected to be in the range between the thermal conductivity of PDMS/4 wt% xGnP single filler composite (0.359 W/m.K) and PDMS/4 wt% MWCNT-OH single filler composite (0.244 W/m.K). However, all the values measured were higher than might be expected from a simple Rule of Mixtures. The highest thermal conductivity is obtained with 75% xGnPs. The obtained thermal conductivity of 0.392 W/m.K exceeds the thermal conductivity measured for either single filler system, being 9% higher than the value for the PDMS/4 wt% xGnP.

This phenomenon of a superior high thermal conductivity of hybrid composites that exceeds the thermal conductivity of either single filler composite has also been observed by other researchers [14, 15]. Kumar et al. [14] used a simple and innovative solution mixing approach to fabricate polyethylenimine (PEI) filled with MWCNT-COOH and xGnP hybrid fillers with a fixed loading of 0.5 wt%. The hybrid composite exhibited a 45% increase in thermal conductivity compared with pure PEI, whereas the PEI filled with 0.5 wt% single filler xGnPs or PEI filled with 0.5 wt% single filler MWCNTs-COOH only exhibited 22% and 9% increases, respectively. They proposed that this was related to a synergistic effect between the fillers, which form an interconnected hybrid network. From Figure 2(c), it is observed that the nanotubes are branching out from the nanoplatelets, forming multiple junction pathways and bridging the graphite nanoplatelets. This special type of hybrid morphology created extra pathways for heat flow, bypassing the polymer matrix, which resulted in an increase in thermal conductivity.

3.4 Electrical Conductivity

Figure 5 shows the plot of electrical conductivity of PDMS filled with xGnP/MWCNT-OH composite as a function of filler ratio. The trend obtained is similar to the thermal conductivity result of PDMS hybrid composites. Increasing the xGnP content from 0% to 100% of the filler content in the hybrid system increases the electrical conductivity of the hybrid composites. The highest electrical conductivity is again observed in PDMS filled with 75% xGnP, confirming that this ratio is an optimum ratio for a range of properties. The measured electrical conductivity is 1.24×10^{-3} S/m, approximately two orders of magnitude higher than that of composites filled

with single fillers of xGnPs and MWCNTs-OH. This relatively high electrical conductivity for composites filled with 75% xGnPs is again most likely due to the formation of an interconnected hybrid network structure between nanotubes and nanoplatelets. It is suggested that the formation of this interconnected hybrid network facilitates the electron transport throughout the polymer, thus giving rise to an increased electrical conductivity [15]. Figure 2(c) supports this explanation because it is observed that MWCNTs form conducting channels between the nanoplatelets, thus reducing the gap, facilitating electron hopping, and hence conducting electricity more readily. Such a synergistic effect with a high contact surface area within the hybrid fillers was also observed in previous works [14, 15]. Kumar et al. [15] reported that this synergy for the electrical conductivity of a hybrid composite of PEI filled with 0.25 wt% MWCNT-COOH and 0.25 wt% xGnP was 8 orders higher than that of PEI/0.5 wt% xGnP and a single order higher than that of PEI/0.5 wt% MWCNT-COOH.

3.5 Mechanical Properties

The mechanical properties of PDMS hybrid composites (tensile strength, modulus at 50% elongation (M50), and elongation at break) were obtained from a tensile test and are presented in Figure 6. As shown in the figure, the hybrid fillers have a more modest effect on the tensile properties of the composite. It is worth noting that the tensile properties of all the hybrid composites exhibit an approximately linear relationship in between the values for PDMS filled with 4 wt% xGnP and PDMS filled with 4 wt% MWCNT-OH. This observation is rather different from the conductivity results because both the thermal and electrical conductivity of PDMS filled with 4 wt % xGnP/MWCNT-OH at a ratio of 75% xGnP/25% MWCNT-OH exceeded the thermal

conductivity of the PDMS filled with a single filler. This indicates that the interconnected hybrid network does not affect the tensile properties of the hybrid composites as significantly and there is no synergistic effect on the tensile strength of hybrid composites. In fact, based on the results of the tensile test, the mechanical properties of hybrid composites appear to be governed by the Rule of Mixtures. The Rule of Mixtures for hybrid composites states that as the proportion of filler A decreases, the contribution of filler A toward the strength of hybrid composites also decreases. The same is true for filler B as well [19]. Thus, by varying the content of xGnPs from 0% to 100%, the tensile strengths and M50s of hybrid composites should be linearly proportional to their composition.

The tensile strengths of PDMS/4 wt% MWCNT-OH and PDMS/4 wt% xGnP single filler composites are 0.86 MPa and 0.72 MPa, respectively. The tensile strengths of PDMS hybrid composites with 25%, 50%, and 75% xGnP are 0.84 MPa, 0.84 MPa, and 0.75 MPa, respectively. The same trend is also observed for M50, as increasing the xGnP ratio from 0 to 100% decreases the M50 somewhat. Both nanotubes and nanoplatelets have similar moduli of approximately 1 TPa, and thus the PDMS hybrid composites should, if they both follow the Rule of Mixtures, have a similar modulus if the total filler content remains the same [12]. It is also observed that the rule of mixtures works reasonably well for the elongation at break of the hybrid composites. All the mixtures lie in the range between the elongation at break of PDMS/4 wt% MWCNT-OH and PDMS/4 wt% xGnP.

3.6 Thermal Stability

Table 1 summarizes the thermal stability data obtained for the PDMS hybrid composites. In general, adding hybrid fillers into PDMS and varying the ratio of xGnP/MWCNT-OH does not significantly affect the thermal stability of composites. This may result from the relatively good thermal stability of the PDMS and also may be a result of the relatively small amount of filler that is encountered in this work. The T_5 of PDMS filled with hybrid fillers shows only a few degree Celsius improvement as the xGnP content in hybrid fillers is increased from 0 to 100% of the filler content. The residue at 800 °C is also consistently in the range from 52% – 54%. This shows that this small a fraction of a hybrid filler has little effect on the thermal stability of PDMS composites. This is expected because the thermal stability is commonly governed by the filler content [20, 21]. The composites with 4 wt% total hybrid fillers should produce more or less the same thermal stability as the composites filled with 4 wt% single filler.

It is however noted that the thermal stability for PDMS hybrid is the best when filled with 75% xGnPs. The T_5 obtained for this composite is 424 °C, which is approximately 6 °C higher than the single filler system of MWCNTs-OH and 4 °C higher than that of the single filler system of xGnPs. In addition, the T_{10} obtained for the same composite is 508 °C, which is approximately 23 °C higher than the single filler system of MWCNTs-OH and 2 °C higher than that of the single filler system of xGnPs. The slightly better thermal stability of PDMS hybrid composite might again be attributed to the synergistic effect of the hybrid structure formed between the xGnPs and MWCNTs-OH [12]. Another possible reason suggested by Huxtable et al. [22] is that the high thermal stability of a composite is due to the high thermal conductivity of the

composite, which helps facilitate a greater heat flow and dissipation throughout the composite. This is in agreement with the current result because the thermal conductivity obtained for PDMS filled with 75% xGnP is the highest among all the composites investigated.

3.7 Coefficient of Thermal Expansion

The T_g and coefficient of thermal expansion data for PDMS hybrid composites are summarized in Table 2. The T_g for all three hybrid composites lies in the range between the T_g of composites filled with MWCNTs-OH and xGnPs single filler, which is approximately $-76.5\text{ }^\circ\text{C}$ and $-77.2\text{ }^\circ\text{C}$, respectively. Increasing the xGnP content from 0% to 100% of the filler content in the hybrid system decreases the T_g for PDMS hybrid. It is however noted that the difference is very small and is almost at the limit of the sensitivity of the test equipment at less than $1\text{ }^\circ\text{C}$.

From Table 2, it is observed that both the CTE below the T_g and the CTE above the T_g of PDMS hybrid composites decrease with increasing xGnP content. The CTE below the T_g and the CTE above the T_g for PDMS/4 wt% MWCNT-COOH is $65.6 \times 10^{-6}/^\circ\text{C}$ and $362.2 \times 10^{-6}/^\circ\text{C}$, respectively. Incorporating hybrid fillers into PDMS according to an xGnP content of 25%, 50%, and 50% decreases the CTE below the T_g by 9%, 16%, and 19%, respectively. As for the CTE above the T_g , adding hybrid fillers into PDMS according to the xGnP content of 25%, 50%, and 75% decreases the CTE above the T_g by 10%, 13%, and 17%, respectively. The reduction of the CTE with an increasing xGnP content can be attributed to the lower CTE of PDMS/xGnP composites.

4. Conclusion

Combining xGnPs and MWCNTs-OH into a hybrid filler system increases the electrical and thermal conductivity of the composite compared with the single filler composite. For a total filler content of 4 wt%, the optimum ratio of xGnP/MWCNT-OH is 75% xGnP/25% MWCNT-OH. At this ratio, the thermal conductivity and electrical conductivity obtained is the highest among all composites produced, with a value of 0.392 W/m.K and 1.24×10^{-3} S/m, respectively. In addition, the reduction in the CTE is also the highest for the hybrid composite with this ratio. The mechanical properties and thermal stability of hybrid composites fall in the range between the properties of a PDMS/xGnP single filler composite and a PDMS/MWCNT-OH single filler composite, approximately following the Rule of Mixtures.

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References

- [1] Yue D, Liu Y, Shen Z, Zhang L. *J. Mater. Sci.* 2006;(41):2541-2544.
- [2] Bokobza L. *Vib. Spectrosc.* 2008;(51):52-59.

- [3] Das A, Stockelhuber K, Jurk R, Saphiannikova M, Fritzsche J, Lorenz H, Klüppel M, Heinrich G. *Polymer* 2008;(49):5276-5283.
- [4] Kong KTS, Mariatti M, Rashid AA, Busfield JJC. *Polym. Bull.* 2012; (69:8):937-953
- [5] Chua TP, Mariatti M, Azizan A, Rashid AA. *Compos. Sci. Technol.* 2010;(70):671-677.
- [6] Hong J, Lee J, Hong CK, Shim SE. *Curr. Appl. Phys.* 2010;(10):359-363.
- [7] Wu C-L, Lin H-C, Hsu J-S, Yip M-C, Fang W. *Thin Solid Films* 2009;(517):4895-4901.
- [8] Kalaitzidou K, Fukushima H, Drzal LT. *Carbon* 2007;(45):1446-1452.
- [9] Ma P-C, Siddiqui NA, Marom G, Kim J-K, *Compos Part A- APPL S*, 2010; (41:10): 1345-1367
- [10] Kim S, Drzal LT, *Adhes J. Sci. Technol.* 2009;(23):1623-1638.
- [11] Kim S, Seo J, Drzal LT. *Composites Part A* 2010;(41):581-587.
- [12] Li J, Wong P-S, Kim J-K. *Mater. Sci. Eng. A* 2008 (483-484) 660-663.
- [13] Ma PC, Liu MY, Zhang H, Wang SQ, Wang R, Wang K, Wong YK, Tang BZ, Hong SH, Paik KW, Kim JK, *Appl Mater Interfaces* 2009; (1:5): 1090-1096.
- [14] Yang S-Y, Lin W-N, Huang Y-L, Tien H-W, Wang J-Y, Ma C-CM, Li S-M, Wang Y-S. *Carbon* 2011;(49):793-803.
- [15] Kumar S, Sun LL, Caceres S, Li B, Wood W, Perugini A, Maguire RG, Zhong WH. *Nanotechnology* 2010;(21):105702.
- [16] Fukushima H, Kalaitzidou K, Drzal LT. *Electrical and barrier properties of exfoliated graphite nanoplatelet (xGnP) reinforced nanocomposites, ICCM-16, Kyoto, 2007.*

- [17] Kong KTS. Studies on the processing and properties of conductive single filler and conductive hybrid filler filled silicone elastomer composites, MSc. Thesis, Universiti Sains Malaysia, Penang, 2012.
- [18] Xing Y, Li L, Chusuei CC, Hull RV. *Langmuir* 2005;(21):4185-4190.
- [19] Summerscales J. Hybrid Composites. Plymouth University, 2010 [online lecture notes] Retrieved from: <
<http://www.tech.plym.ac.uk/sme/MATS324/PowerPoint/MATS324A6%20hybrid.ppt>>
- [20] Chen X, Wang J, Lin M, Zhong W, Feng T, Chen X, Chen J, Xue F. *Mater. Sci. Eng. A* 2008;(492):236-242.
- [21] Chua TP. The effect of surface modification on multi-walled carbon nanotubes on silicone rubber composites, MSc. Thesis, Universiti Sains Malaysia, Penang, 2009.
- [22] Huxtable ST, Cahill DG, Shenogin X, Xue L, Ozisik R, Barone P, Usrey M, Strano MS, Siddons G, Shim M, Koblinski P. *Nat. Mater.* 2003;(2):731-734.

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Fig. 1: TEM micrographs of (a) pristine MWCNT (b) MWCNT-OH and (c) xGnP.

Fig. 2: SEM images on fracture surface of PDMS hybrid composites filled with (a) 75% xGnP and (b) 50% xGnP at a high magnification; (c) 75% xGnP at an even magnification.

Fig. 3: Schematic illustrating the formation of interconnected hybrid network between xGnP and MWCNT-OH.

Fig. 4: Thermal conductivity of PDMS hybrid composites as a function of xGnP content.

Fig. 5: Electrical conductivity of PDMS hybrid composites as a function of xGnP content.

Fig. 6: Tensile properties of PDMS hybrid composites as a function of xGnP content.

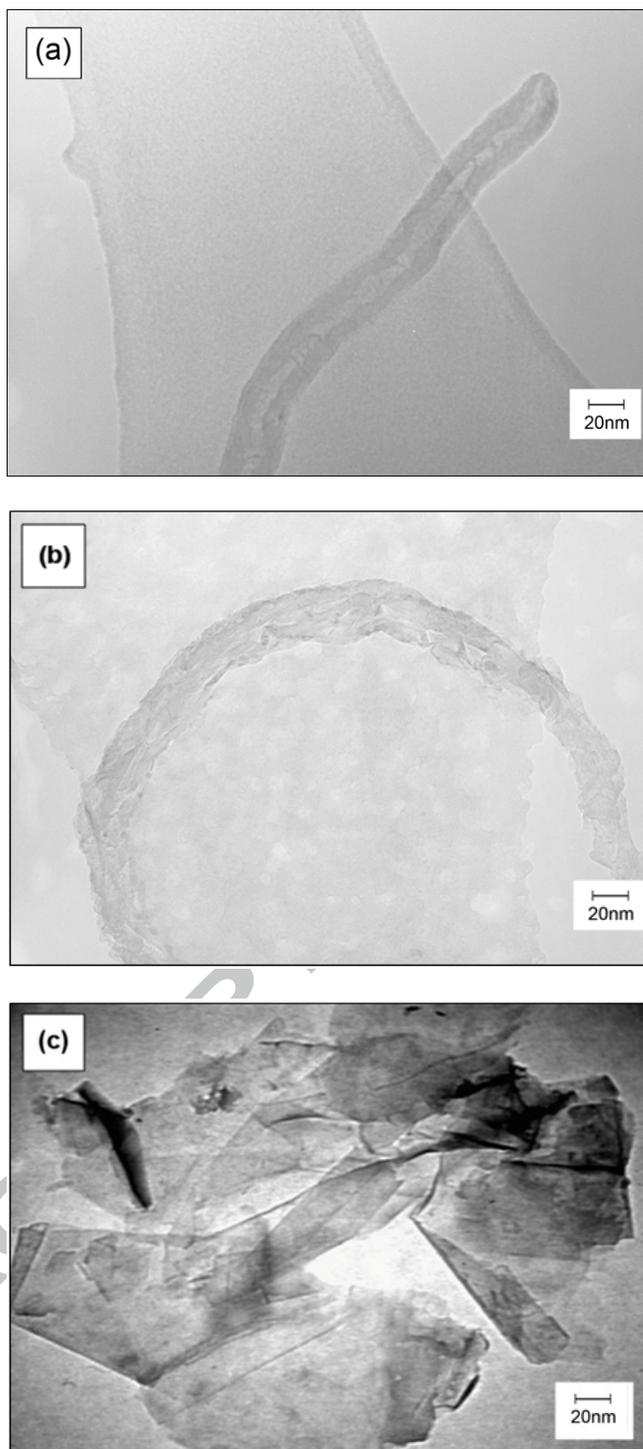


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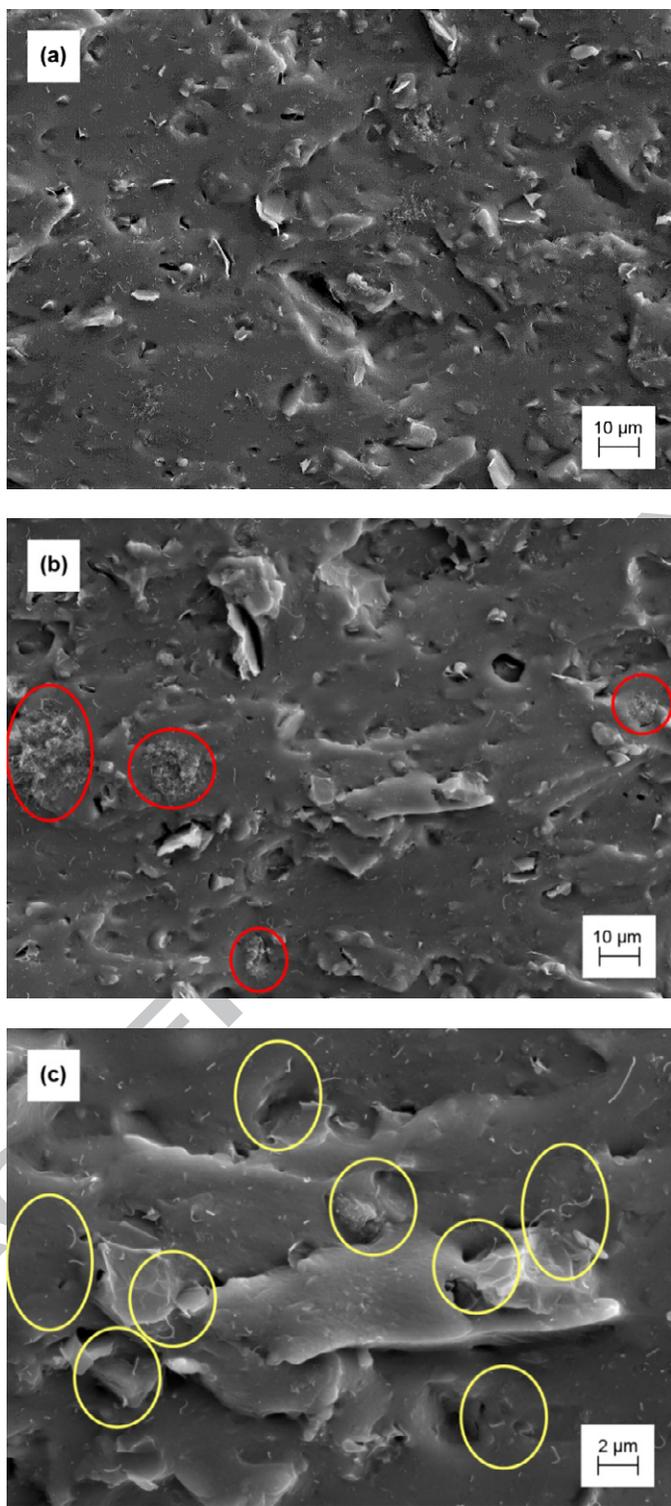


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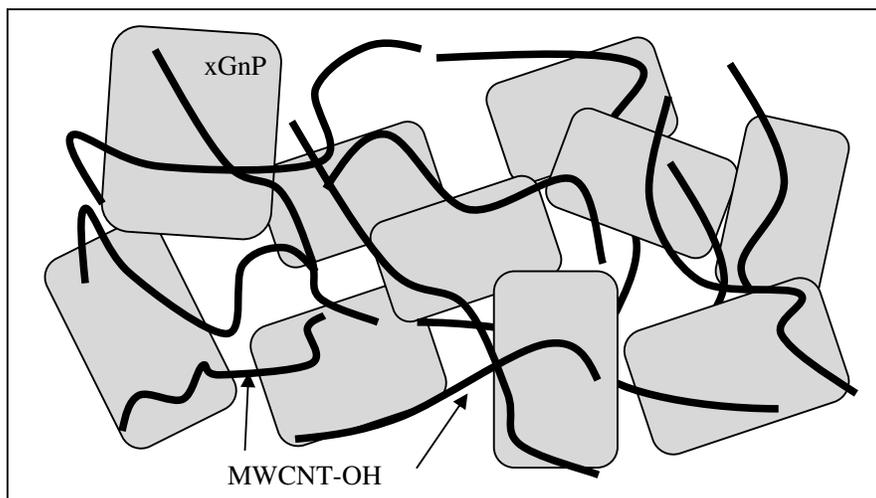


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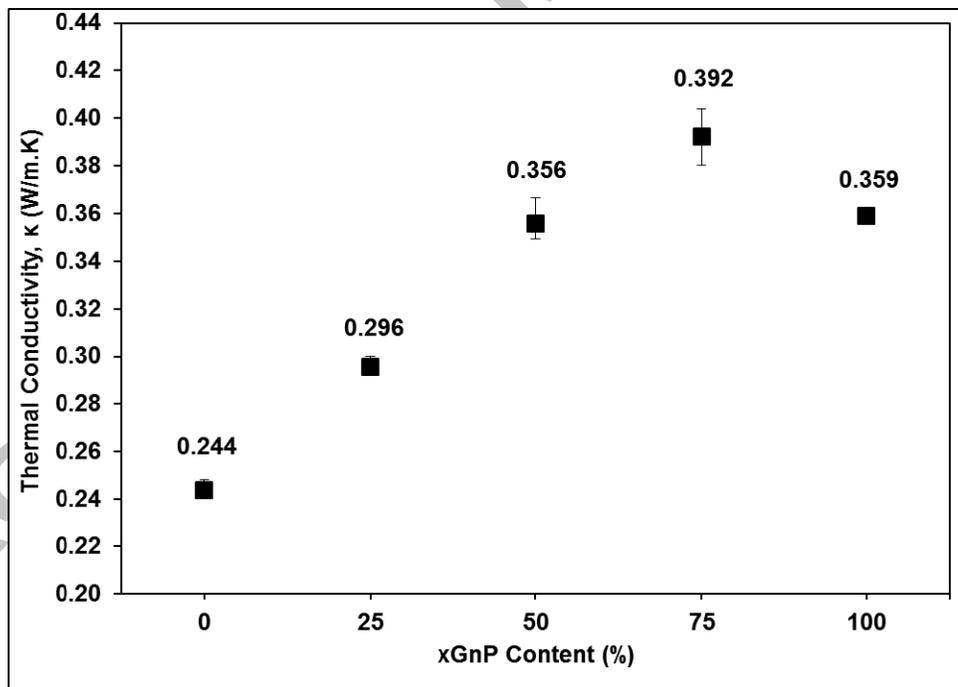


Fig. 4: Thermal conductivity of PDMS hybrid composites as a function of xGnP content.

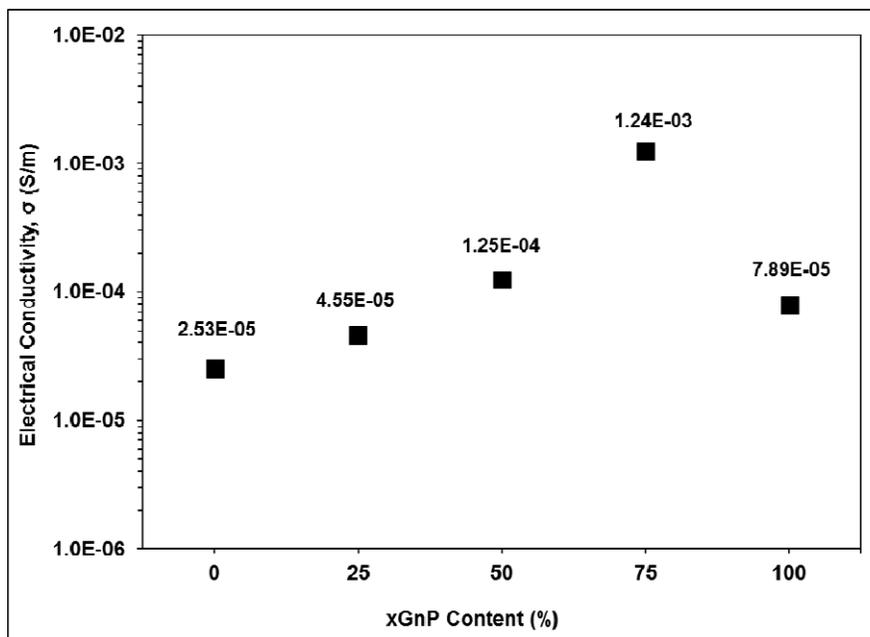


Fig. 5: Electrical conductivity of PDMS hybrid composites as a function of xGnP content.

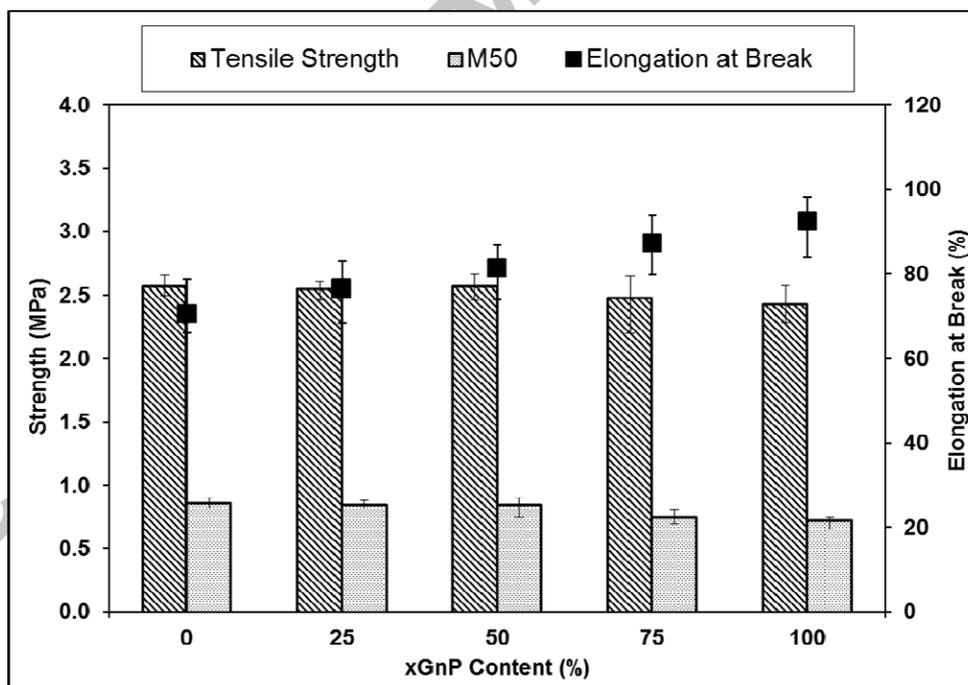


Fig. 6: Tensile properties of PDMS hybrid composites as a function of xGnP content.

Table 1: Thermal stability data obtained from the weight against temperature curves of PDMS hybrid composites.

xGnP content (%)	T ₅ (°C)	T ₁₀ (°C)	Residue at 800 °C (%)
0	418	485	51.5
25	418	495	52.7
50	420	502	53.8
75	424	508	54.3
100	420	506	56.4

*T₅ and T₁₀ represents decomposition temperature at 5% and 10% weight loss, respectively

Table 2: T_g and coefficient of thermal expansion data of PDMS hybrid composites

xGnP content (%)	T _g (°C)	CTE below T _g (x 10 ⁻⁶ /°C)	CTE above T _g (x 10 ⁻⁶ /°C)
0	-76.5	65.6	362.2
25	-77.0	60.0	326.8
50	-77.3	55.3	314.9
75	-77.4	53.6	301.1
100	-77.2	22.4	288.1