

Dielectric Properties of MWCNT-Muscovite/Epoxy Hybrid Composites

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Abstract—In the present work, the dielectric properties of Epoxy/MWCNT-muscovite HYBRID and MIXED composites based on a ratio 30:70 were studied. The multi-wall carbon nanotubes (MWCNT) were prepared using two methods: (a) MWCNT-muscovite hybrids were synthesised by chemical vapour deposition (CVD) and (b) physically mixing muscovite with MWCNT. The effects of different preparation of the composites and filler loading were evaluated. It was revealed that the dielectric constants of HYBRID epoxy composites are slightly higher than MIXED epoxy composites. It was also indicated that the dielectric constant increased by increasing the MWCNT filler loading.

Keywords—MWCNT-Muscovite, Epoxy, Dielectric Properties, Hybrid Composite.

I. INTRODUCTION

THE extraordinary and remarkable structural, mechanical [1], thermal and electrical properties, which have low density, excellent strength, toughness, chemical stability and high surface area have attracted much attention from the researcher [2] since the first discovery of carbon nanotubes.

Recently, CNT/polymer composites have generated much interesting research due to their potential applications in aerospace and automotive materials [3], capacitors [4], sensor [5], electromagnetic shielding [6], and in optical devices [7]. Combining CNTs with various polymer matrices, such as polyurethane [8], polyamides, polypropylene [9] and epoxy [10] to improve the electrical and mechanical properties, has been intensively investigated.

Epoxy resin is the most common thermoset material used as a matrix for advanced composites due to its superior thermal, mechanical and electrical properties and chemical characteristics as well as its low shrinkage upon curing, volatility during curing reactions, curing over a wide temperature range and the control of degree of cross linking. Epoxy resins can be cross-linked through a polymerisation reaction with a hardener at room temperature or at an elevated temperature. Curing agents used at room temperature are usually from aliphatic amines, whereas aromatic amines and acid anhydrides are used for high temperature.

In this study, MWCNTs are used as filler due to their exceptional electronic properties with high aspect ratio. Polymeric matrix composites containing dispersed electric conductive fillers can be a good candidate for dielectric materials. To reduce the dielectric loss in the composites, the

electrical connection between the dispersed MWCNT must be blocked. Epoxy was used as the blockage material to utilise good insulating properties as the MWCNT composites have high electrical conductivity. MWCNT/epoxy composites were prepared and morphological and electrical properties of the composites were investigated. Characteristics such as morphological properties of HYBRID and MIXED composites and dielectric constant of MWCNT-muscovite/epoxy were discussed.

II. EXPERIMENTAL

A. Materials

The crystal clear epoxy resin and hardener were purchased from Euro Chemo-Pharma Sdn. Bhd. Epoxy resin crystal clear is diglycidyl ether of bisphenol A (DGEBA) based and epoxy hardener crystal clear is trimethylhexamethylene diamine (TMD) based. MWCNT (95%, 10-30nm) was purchased from Skyspring Nanomaterials, Inc. Muscovite was obtained from Bidor Mineral Sdn. Bhd. with an average particle size of 5µm. Nickel (II) nitrate hexahydrate (98%, $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) was purchased from Merck.

B. Synthesis of MWCNT-Muscovite by CVD Process

Ni/Muscovite catalyst was prepared by mixing nickel nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) with muscovite in distilled water with the presence of sodium hydroxide (NaOH). After 24 hours at room temperature, the precipitate catalyst was washed, filtered and dried in an oven at 110°C for 2 hours. Then, the catalyst was calcined at 900°C for 10 hours before performed in hydrogen gas at 400°C for reduction process. In the presence of methane and nitrogen atmosphere, the catalyst was reacted at 800°C for 30 minutes. The synthesis of MWCNT was performed in a custom-made horizontal furnace using nickel as a metal catalyst, similar to earlier reports [11]. The final product was named HYBRID composite.

C. Fabrication of MWCNT-Muscovite/Epoxy Composites

An appropriate amount of MWCNT to prepare 1%, 3%, and 5% MWCNT was dispersed into muscovite powder with a ratio MWCNT to muscovite of 30:70. The mixture powders were milled with ceramic ball (50-100 mm diameter) at 30 rpm for 24 hours.

In order to prepare dielectric specimens, the desired amount of DGEBA epoxy was drawn into a container and sonicated for a few seconds to lower the resin viscosity. The hybrid MWCNT-muscovite was mixed with epoxy resin varying MWCNT contents of 1%, 3%, and 5% and sonicated for 5 minutes at 60°C. Then, the TMD epoxy hardener was added

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into the mixture and stirred manually at room temperature, followed by degassing of the mixture in a vacuum oven at room temperature for 30 minutes to 1 hour. The same method was applied to mixed MWCNT-muscovite/Epoxy composites. Then, the mixture was cast into the flat silicon mould and cured at 120°C for 2 hours.

D. Characterisation by FESEM and EDX Analysis

The morphology of the MWCNT-muscovite hybrid and mixed composite was analysed by Field Emission Scanning Electron Microscope (FESEM, Zeiss Leo Supra-35VP) operating at 5.0kV to 30kV. Analysis of the composition of the carbon (C), muscovite ($\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{OH})_2$) and percentage of carbon in the composites were analysed using the Dispersive X-Ray (EDX) spectroscopy.

E. Dielectric Properties

Dielectric constants of the composite were performed using RF Impedance, Hewlett Packard 4219B at room temperature using ASTM D149. The frequency of the sample was measured at 500MHz to 1GHz. The samples for dielectric property measurement were 2mm thick.

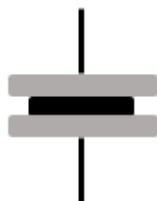


Fig. 1 Schematic of dielectric testing

III. RESULT AND DISCUSSION

A. EDX Analysis

From the EDX analysis, as shown in Fig. 2, 28.97% based on weight percentage was attributed to deposition MWCNT on the muscovite particle. It is proven that carbon nanotube growth, successfully performed by the methane decomposition process that the EDX analysis detected, carbon is the highest element in the composites. Using this percentage, the MIXED composites were prepared. The other visible percentage found in the EDX analysis could be attributed to the presence of the nickel catalyst and muscovite particles.

B. SEM Observation of Hybrid and Mix MWCNT-Muscovite/Epoxy Composites

Fig. 3 represents the muscovite particles before the growth of carbon nanotubes with cleavage flake shape. It is noted that muscovite is platy with a very smooth surface.

Figs. 4 (a)-(d) depict the dispersion of MWCNT via chemical vapour deposition and physical mixing process. From morphological analysis, it shows that the MWCNT was successfully deposited on the muscovite surface for both of the MWCNT/muscovite HYBRID and MIXED composites. It reveals that the carbon nanotube species have a diameter ranging from 10 to 30nm and have entanglement and diverse

worm-like orientation for both HYBRID and MIXED composites. It is clearly shown in Fig. 4 (a) that the MWCNT are grown from the muscovite particles for HYBRID composites. Meanwhile, the MWCNT and muscovite particles are physically attached to each other for MIXED composites in Fig. 4 (b). Furthermore, the MWCNT tend to agglomerate due to the van der Waals interaction.

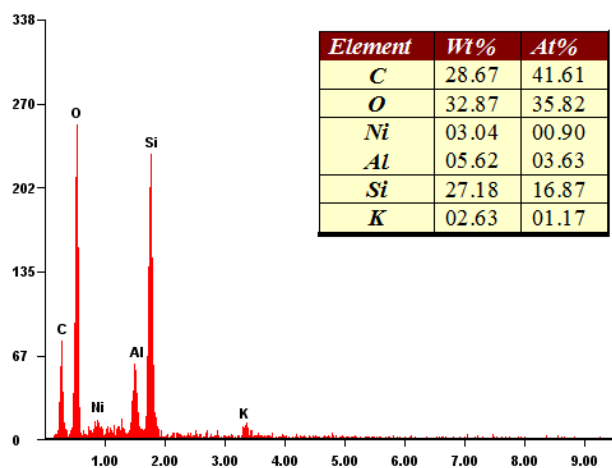


Fig. 2 EDX analysis of CNT via CVD

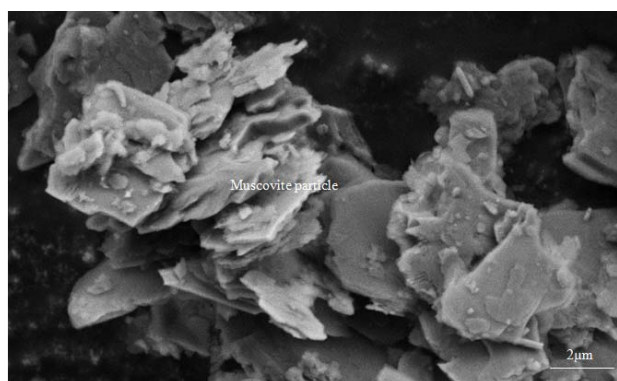
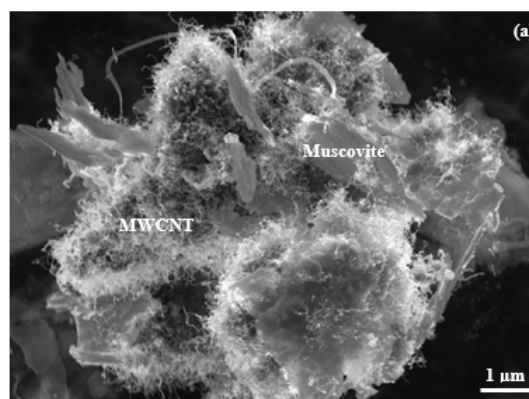


Fig. 3 Muscovite clay particles at magnification 5kV



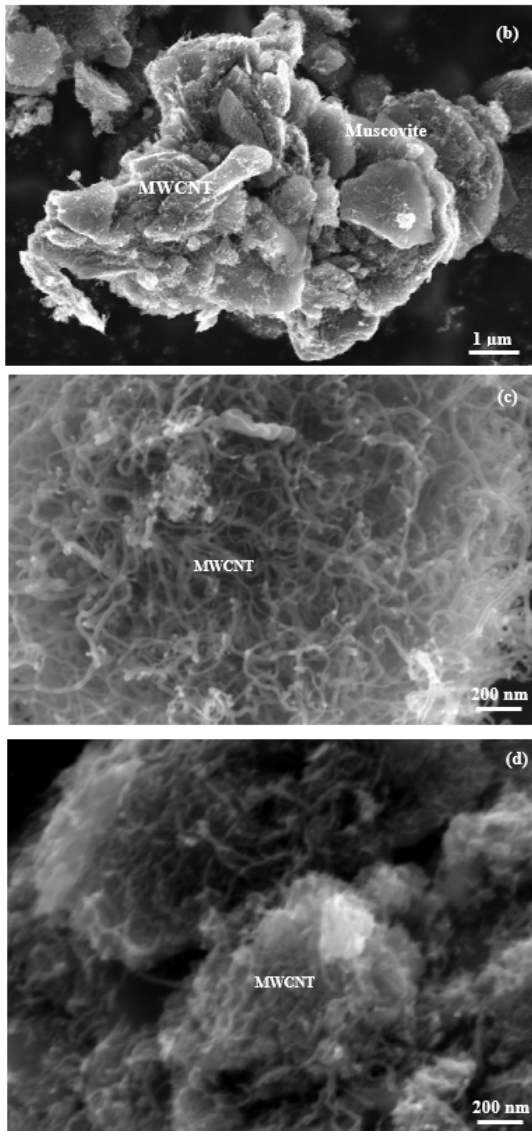


Fig. 4 (a) MWCNT-muscovite Hybrid composite at magnification 5kv, (b) MWCNT-muscovite Mixed composite at magnification 5kv, (c) MWCNT-muscovite Hybrid composite at magnification 30kv, and (d) MWCNT-muscovite Mixed composite at magnification 30kv

Fig. 5 shows the fracture section of 3% MWCNT-muscovite composites. It was discovered that MWCNT-muscovite are well dispersed for HYBRID composites. On the other hand, distribution between the CNT-Muscovite epoxy composite clearly shows that the CNT and muscovite particles are dispersed at different places. Poor dispersion of the MWCNT on the MIXED composites has been attributed to both weak interactions between the MWCNT and the polymer matrix.

C.Dielectric Properties of MWCNT-Muscovite/Epoxy Composites

Fig. 6 shows the frequency-dielectric constant dependence of the MWCNT/epoxy for HYBRID and MIXED composites as a function of CNT content at room temperature for the filler

loading 1%, 3% and 5% MWCNT. It is evident that the dielectric constants of the samples decreased with increasing frequency. It is also observed that under the same filler loading the dielectric constant of HYBRID epoxy composites is higher than MIXED epoxy composites. This is because of the increase of the interfacial adhesion between the HYBRID composite and the epoxy matrix due to the good dispersion of the MCNT on the muscovite particles. The large length-to-diameter ratio of MWCNT causes them to be easily tangled, which ultimately affects the properties. Poor dispersion in the direct contact between the conductive fillers and the matrix leads to high dielectric losses. Increasing of filler loading will increase the dielectric constant. This is because the dielectric constant and electric conductive depends on the configuration of the conductive fillers. It is proven, as per Fig. 6, the dielectric constant of the composites with the MWCNT loading < 5% shows a weak frequency dependence.

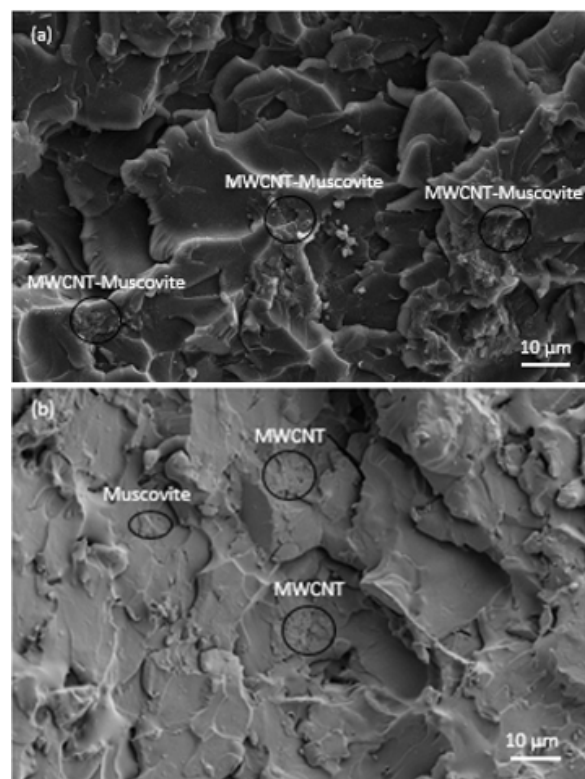


Fig. 5 (a) Fracture of Hybrid Composites and (b) Fracture of Mixed Composites at magnification 500x

IV. CONCLUSION

This work has proven that chemically synthesised MWCNT improves the dispersion of reinforcement and matrix composites, which give more homogeneous distribution compared to physical mixing. The dielectric properties of the composites are fully enhanced using the CVD preparation process. It also found that the frequency dependence of the dielectric constant is also dependent on the filler loading level.

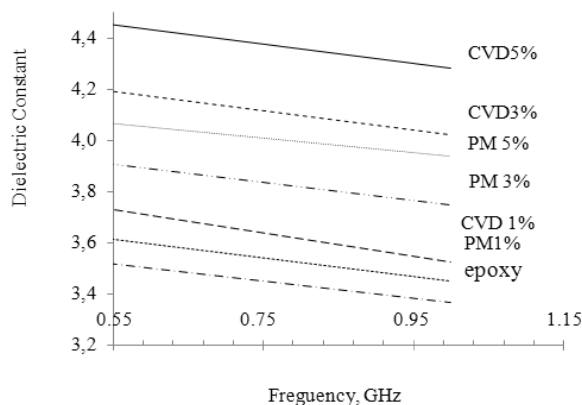


Fig. 6 Dielectric constant of epoxy, 1%, 3%, and 5% of hybrid and Mixed MWCNT-muscovite/Epoxy composites

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