INCORPORATION OF MULTI-WALLED CARBON NANOTUBES AND GRAPHENE NANOPLATELETS ON THE MORPHOLOGY AND PROPERTIES OF POLYETHYLENE TEREPHTHALATE NANOCOMPOSITES

By

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Chair : **Md Shuhazlly bin Mamat @ Mat Nazir, PhD**
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In this work, the interaction effect between polyethylene terephthalate (PET) with multiwalled carbon nanotubes (MWCNTs) and graphene nanoplatelets (GNPs) at concentration of 0.1 wt% and 0.5 wt% was examined. The morphology and the properties of PET nanocomposites prepared through melt compounding and injection moulding method were investigated. The presence of MWCNTs and GNPs in the PET matrix was confirmed by the X-ray diffraction (XRD) technique. In contrast, in PET nanocomposites, better dispersion distribution was observed in the 0.1 wt% PET-MWCNTs-GNPs nanocomposites compared to the 0.5 wt% PET-MWCNTs-GNPs nanocomposites resulting in fewer mechanical and physical failures like crack, delamination, and agglomeration. Furthermore, the incorporation of MWCNTs or GNPs into pure PET improved the thermal stability of the nanocomposites as the nanofillers acted as efficient heat sinks, preventing thermal degradation of PET. Additionally, this research demonstrated that PET-MWCNTs-GNPs 0.1 wt% achieved a significant enhancement of up to 270% in Electromagnetic Interference (EMI) shielding compared to neat PET while also increasing electrical conductivity. In terms of mechanical testing, the PET nanocomposites with 0.1 wt% loading were analyzed and PET-MWCNTs-GNPs 0.1 wt% exhibit superior performance in tensile testing, impact testing, flexural testing as well as hardness testing, thus, make this composition emerged as the most promising choice with excellent microwave absorbance and remarkable strength simultaneously.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia (UPM) sebagai memenuhi keperluan ijazah Master Sains

PENGGABUNGAN TIUB NANO KARBON DINDING BERBILANG (MWCNTS) DAN KEPINGAN NANOZARAH GRAFIN (GNPS) PADA MORFOLOGI DAN SIFAT-SIFAT NANOKOMPOSIT POLIETILENA TEREFTALAT (PET)

Oleh

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Dalam kajian ini, kesan interaksi antara polietilena tereftalat (PET) dengan tiub nano karbon dinding berbilang (MWCNTs) dan kepingan nanozarah grafin (GNPs) pada komposisi 0.1 wt% dan 0.5 wt% telah dikaji. Morfologi dan sifat-sifat nanokomposit PET yang disediakan melalui kaedah sebatian cair dan pengacuan suntikan telah dianalisis. Kewujudan GNPs dan MWCNTs dalam PET telah disahkan melalui teknik pembelauan sinar-x (XRD). Walau bagaimanapun, dalam nanokomposit PET, taburan serakan yang lebih baik dapat diperhatikan dalam nanokomposit PET-MWCNTs-GNPs 0.1 wt% berbanding nanokomposit PET-MWCNTs-GNPs 0.5 wt% yang mana kegagalan mekanik dan fizikal seperti retak, delaminasi dan penggumpalan dapat dilihat pada komposisi yang paling tinggi. Selain itu, penggabungan MWCNTs dan GNPs dalam PET dapat meningkatkan kestabilan terma nanokomposit kerana pengisi nano yang bertindak sebab penyerap haba yang cekap dan menghalang degradasi haba di PET. Di samping itu, penyelidikan ini membuktikan PET-MWCNTs-GNPs 0.1 wt% mencapai peningkatan ketara sehingga 270% dalam perisai inteferensi elektromagnetik (EMI) berbanding PET, juga peningkatan dalam penyaluran elektrik. Bagi ujian mekanik, nanokomposit dengan komposisi 0.1 wt% telah dianalisis dan didapati PET-MWCNTs-GNPs 0.1 wt% mempamerkan prestasi unggul dalam ujian tegangan, ujian impak, ujian lentur serta ujian kekerasan seterusnya menjadikan komposisi ini pilihan yang terbaik dengan ciri-cici penyerapan gelombang mikro yang sangat baik dan kekuatan yang luar biasa secara serentak.

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LIST OF ABBREVIATION

CNT	Carbon Nanotubes
DSC	Differential Scanning Calorimetry
EMI	Electromagnetic Interference
FESEM	Field Emission Scanning Electron Microscope
GNPs	Graphene Nanoplatelets
MWCNTs	Multi-walled Carbon Nanotubes
PET	Polyethylene Terephthalate
PNC	Polymer Nanocomposites
TGA	Thermogravimetric Analysis
VNA	Vector Network Analyzers
XRD	X-ray Diffraction

CHAPTER 1

INTRODUCTION

1.1 General Introduction

This chapter will introduce the field involved in this research which is mainly polymer nanocomposite which is categorized under nanomaterials. Nanomaterials, which is one of the characteristics in nanotechnology and nanoscience, is characterized a group of materials in which at least one-dimension measures less than approximately 100 nanometers (Mandal and Thakur, 2017). Polymer nanocomposites or nanofilled polymer that involve nanomaterials is one of the most versatile composites that exhibits a combination of properties for multifunctional applications. In this chapter, the objective background of the study and a structure of the thesis that explain the sequence of each chapter are also stated.

1.2 Nano Materials

Nanomaterial manufactured and developed at a very small scale has exhibited valuable characteristics across various industries. Their specific characteristics that are different forms of bulk materials have attracted widespread attention since the 1990s (Shi et al., 2004). However, it must have at least one dimension less than about 100 nanometres as the size ranges approximately 1-100 nanometres.

The outstanding characteristics of nanomaterials have raised high expectation for a potential new industrial revolution and sparked significant interest in the development of nanocomposites with excellent mechanical properties and multifunctional capabilities. This is evident in the improvement of polymer nanomaterials for food packaging (PNFP) that offer inventive offers to enhance polymer performance while also inducing safety, economic and environmental advantages, hence contributions to the reduction of CO_2 emission (Silvestre et al., 2011). Furthermore, the use of nanomaterials to the design of biosensing is nowadays one of the most active research areas because of large specific surface area and enhancement of sensitivity (Zhu et al., 2015). Other than that, the addition of SiO_2 to cement has been found to result in an improvement in compressive strength and remarkable enhancement of up to 18% in the compression strength of Portland-based concrete (Parveen et al., 2013; Dahlan, 2019).

1.3 Polymer Nanocomposites

In the field of nanotechnology, there is an interest in recent literature concerning nanotechnology based on polymer. This is because polymer nanocomposites represent new alternatives to conventionally filled polymers as this dramatically improved the performance properties of filled distribution nanocomposites when compared to pure polymers due to their filler sizes that typically 10-100 A in at least one dimension.

Polymers have many benefits compared to materials like glass and silicon, including being cost-effective, easy to process and capable of processing results in mass production (Attia et al., 2009). The incorporation of nanoparticles into the polymer matrix is remarkable for its microscopic effects, as it can enhance the optical, mechanical properties, fire-resistant properties and barrier of these new materials. Nevertheless, polymers exhibit lower modulus and strength compared to ceramics and metals. Enhancing the mechanical properties is achieved by using a reinforced polymer with the addition of nanofillers such as fibers, platelets or particles. Nevertheless, it is all subject to the category of filler used, the concentration level, polymer size and the method used to fabricate the polymer (Todorov and Viana, 2007). Besides that, it is highlighted that the technique they prepared, nature of nanofillers and chemistry of polymer matrices significantly impact the properties of nanocomposites (Tjong, 2006). Besides, the combination of two or more polymers are include to improve the polymer's processability especially on high temperature thermoplastics, to improve the physical properties and mechanical of the composite, to make them more desirable than those individual polymers and to meet the market force as the blending technology may be the resources of developing desirable properties from recycled products (Lee, 1993).

A classification scheme of various polymer composite types can be illustrated as in the following Figure 1.1. It consists of three main divisions which are conventional composite, intercalated composites, and exfoliated composites. Conventional composite is where the contact between the clay and the matrix is very weak that can degrade the properties of the resulting composites. An intercalated structure is where the polymer chain is only partially intercalated in between clay layers. Exfoliation composites are where the clay is separated as an individual layer in the polymer matrix which can enhance the contact between the clay and the matrix thus enhancing the mechanical properties of the composites. (Ray et al., 2003; Tjong, 2006)

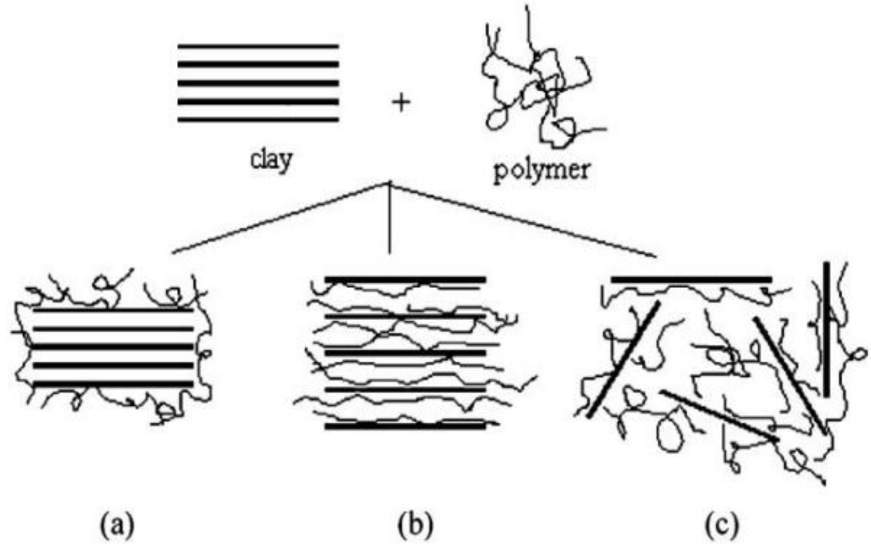

Figure 1.1: Types of polymer nanocomposites structure.

1.3.1 Uses of Polymer Nanocomposites

Polymer nanocomposites (PNC) are a type of nanotechnology that develops the properties of the polymer to make it more versatile to meet the needs of people nowadays. PNC increased significant interest due to their varied applications across multiple industries.

Previous study found that it is predicted that PNC could save 1.5 billion liters of gasoline over the lifespan of one year production of vehicles and lead to decrease carbon dioxide emissions by more than 5 billion kilograms (Zeng et al., 2005). Nanocomposite materials are also potential candidates for bioactive implant materials, catalysts, contact lenses and gas-separation membranes (Tanahashi, 2010). The combination of the properties of the polymer matrix with semiconductor nanoparticles will enable sensor devices selective to certain components in mixtures of gases or vapors (Godovsky, 2000). The MWCNTs have outstanding thermal, electrical and mechanical properties as well as high aspect ratios and large surface areas, making the material idyllic for reinforcement of thermoplastics polymer nanocomposites (Avilés and Toro, 2013).

1.4 Problem Statements

The high consumption of PET bottles in the packing industry leads to a large volume of non-biodegradable waste, which in turn increases serious environmental problems. Alongside its convenience, recycling and reusing plastic waste can add through the involvement of nanotechnology which can lead to various applications.

Despite the numerous industrial applications of PET in the industries, PET also has its own disadvantages, including insufficient mechanical properties, thermal stability and conductivity that in turn, will limit the full utilization of PET (Wang et al., 2015). This sparks the need for fillers or additives that are capable to enhance the properties of the PET matrix and would allow it to be used in more applications. However, the challenge can see in succeeding uniform distribution of MWCNTs and GNPs in the matrix, as well as costly processes involved in preparing carbon nanotubes.

The advancement of electronics has given a rise to new requirement for electromagnetic interference (EMI) shielding materials that cannot be satisfied by conventional metal-based EMI shielding materials. Combining a cost-effective, non-conductive polymer with a very small amount of electrically conductive MWCNTs and GNPs may offer great opportunities for both the electrical and mechanical properties at low manufacturing costs.

However, the nature of MWCNTs is cohesive, chemically inactive, large surface area and surface tension are causing limitation in polymer composites with MWCNTs. This is because of the bundle formation of MWCNTs and fiber to fiber contact which creates many defect sites such as lumps, aggregates and voids in the produced composites. To address this issue, a low amount of MWCNTs should be used as filler, as the lower the MWCNTs loading, the higher the efficiency of the melt blending process to scatter and dispersed the filler in the matrix. An alternative to MWCNTs is Graphene Nanoplatelets (GNPs) which are cost effective and have excellent properties.

Various methods have been employed in fabricating composites such as *in situ* exfoliation, melt-compounding and solution mixing. Melt compounding is a simple and compatible approach for producing GNPs and MWCNTs nanocomposites without the need for solvent that making it suitable for industrial processes.

1.5 Research Objectives

The research aims to study the characterization, thermal properties, mechanical properties and microwave absorbance of PET-GNPs-MWCNTs nanocomposites. PET is as polymer matrix while GNPs and MWCNTs are employed as carbon fillers. This polymer nanocomposite was synthesis by the melt-compounding method and underwent injection molding for characterizations. The objectives of this research are as follows:

- To investigate the microstructure and distribution of PET-GNPs-MWCNTs nanocomposites prepared by injection molding and melt-compounding method and method.
- To study the thermal properties of PET-GNPs-MWCNTs nanocomposites with a different composition ratio of GNPs and MWCNTs.
- To investigate the mechanical properties of PET-GNPs-MWCNTs 0.1 wt% with a different mixture ratio of PET and GNPs/MWCNTs.
- To study the microwave absorbance properties and electrical properties of neat PET and PET nanocomposites with different mixture ratios of nanofillers.

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