



**EFFECT OF TYRE DUST LOADING AND
COMPATIBLIZER ON PROPERTIES OF RECYCLED HIGH
DENSITY POLYETHYLENE/ ETHYLENE VINYL
ACETATE/ TYRE DUST COMPOSITES**

by

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**School of Material
UNIVERSITI MALAYSIA PERLIS**

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LIST OF ABBREVIATIONS

| | |
|-------------------|--|
| ASTM | American Society for Testing and Materials |
| BF | Bamboo fiber |
| CL | Caprolactam |
| CMC | Ceramic matrix composites |
| CaCO ₃ | Calcium carbonate |
| Ca | Calcium |
| CB | Carbon black |
| CFF | Chicken feather fiber |
| DSC | Differential Scanning Calorimetry |
| DMA | Environmental stress cracking resistance |
| EVA | Ethylene Vinyl Acetate |
| ESCR | Environmental stress cracking resistance |
| ESP | Egg shell powder |
| FTIR | Fourier transform infrared analysis |
| FDT | Final decomposition temperature |
| Fe | Iron |
| GTR | Ground tyre rubber |
| HDPE | High Density Polyethylene |
| ISO | International Organization Standardization |
| LDPE | Low density Polyethylene |

| | |
|---------------------|--|
| LDI | Lysine-based diisocyanate |
| LNR | Liquid natural rubber |
| LLDPE | Linear bonded low density polyethylene |
| MAH | Maleic anhydride |
| Mg(OH) ₂ | Magnesium hydroxide |
| MAPE | Maleic anhydride polyethylene |
| NR | Natural rubber |
| PTE | Passenger tyre equivalent |
| PE | Polyethylene |
| PET | Polyethylene terephthalate |
| PEgMAH | Polyethylene grafted maleic anhydride |
| PMCs | Polymer matrix composites |
| PMMA | Polymethyl methacrylate |
| PS | Polystyrene |
| PC | Polycarbonate |
| PP | Polypropylene |
| PPE | Polyethylene ether |
| PLA | Poly (lactic acid) |
| PVA | Polyvinyl alcohol |
| Phr | Per hundred resin |
| PVC | Polyvinyl chloride |
| r-HDPE | Recycled High Density Polyethylene |
| SBR | Styrene butadiene rubber |

| | |
|-------|--|
| SEBS | Polystyrene-b-(ethylene-co-butylene)-b-styrene |
| SEM | Scanning Electron Microscopy |
| TD | Tyre Dust |
| TGA | Thermogravimetric analysis |
| T_g | Glass transition temperature |
| T_m | Melting temperature |
| TPSS | Thermoplastic sago starch |
| V_f | Volume fraction |
| WTD | Waste tyre dust |
| WGRT | Waste ground tyre dust |
| XPS | X-ray photoelectron spectroscopy |
| Zn | Zinc |

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LIST OF SYMBOLS

| | |
|--------------------|--------------------------------------|
| $^{\circ}\text{C}$ | Degree Celsius |
| % MS | Mass swell percentage |
| H_f | Enthalpy of fusion of the composites |
| H°_f | Enthalpy of fusion of HDPE |

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Kesan Muatan Habuk Tayar dan Pengserasi terhadap Sifat-sifat Komposit Polietilena Ketumpatan Tinggi Kitar Semula/ Etilena Vinil Asetat/ Habuk Tayar

ABSTRAK

Komposit polietilena berkumpatan tinggi kitar semula/ Etilena vinil asetat/ Habuk tayar telah dikaji. Komposit ini telah disediakan dengan menggunakan Brabender Plasticorder pada suhu 160 °C dengan kelajuan pemutar 50 rpm. Kesan pengisian TD dan pelbagai jenis pengserasi terhadap sifat-sifat tegangan, peratusan jisim pengembangan, sifat morfologi, spektroskopi inframerah dan pencirian haba bagi r-HDPE/EVA/TD telah dikaji. Pengserasi yang digunakan dalam kajian ini adalah malida anhidrida (MAH), polietilena dicantumkan dengan malida anhidrida (PEgMAH), dan kaprolaktam (CL). Peningkatan jumlah muatan pengisi cenderung kepada pengurangan kekuatan tegangan dengan jumlah muatan pengisi sebanyak TD5 (6.7%), TD10 (21.31%), TD15 (24.95%), TD20 (37.92%), TD25 dan pemanjangan pada takat putus dengan jumlah muatan pengisi sebanyak TD5 (13.9%), TD10 (25.65%), TD15 (36.97%), TD20 (43.67%), TD25 (50.93%) bagi komposit r-HDPE/EVA/TD. Selain itu, modulus keanjalan dengan jumlah muatan pengisi sebanyak TD5 (12.87%), TD10 (18.08%), TD15 (26.136%), TD20 (44.41%) and TD25 (72.781%) dan kestabilan haba telah meningkat. Pada jumlah muatan pengisi yang sama (TD15) serta kemasukan pengserasi (MAH, PEgMAH, dan CL) dalam komposit r-HDPE/EVA/TD mempamerkan peningkatan dalam sifat-sifat tegangan (2.08%, 2.68%, and 6.41%), modulus keanjalan (12.67%, 16.52%, and 25.2%), dan kestabilan haba tetapi penurunan ditunjukkan pada pemanjangan pada takat putus (7.42%, 10.54%, 13.65%), dan peratusan jisim pengembangan (4.09%, 6.53%, 9.61%) jika dibandingkan dengan komposit r-HDPE/EVA/TD. Mikroskop penskanan electron (SEM) juga menunjukkan penambahan pengserasi dapat menambahbaik lekatan antara muka antara TD dan r-HDPE/EVA sebagaimana ditunjukkan di dalam mikroskop SEM. FTIR membuktikan kehadiran MAH, PEgMAH, dan CL di dalam komposit polietilena berkumpatan tinggi kitar semula/ Etilena vinil asetat/ Habuk tayar. Kehadiran kaprolaktam (CL) sebagai pengserasi dalam komposit r-HDPE/EVA/TD menunjukkan ianya adalah pengserasi terbaik berbanding yang lain.

Effect of Tyre Dust Loading and Compatibilizer on Properties of Recycled High Density Polyethylene/ Ethylene Vinyl Acetate/ Tyre Dust Composites

ABSTRACT

The composites of recycled high density polyethylene (r-HDPE)/ ethylene vinyl acetate (EVA)/ tyre dust (TD) were studied. The composites were prepared using Brabender Plasticorder at 160 °C with rotor speed of 50 rpm. The effect of TD content and several of compatibilizers on tensile properties, mass swell percentage, morphological properties, spectroscopy infrared and thermal characterization of r-HDPE/EVA/TD composites were investigated. The Compatibilizer used in this study were maleic anhydride (MAH), polyethylene grafted maleic anhydride (PEgMAH), and caprolactam (CL). The increasing of filler loading tends to decrease the tensile strength with the filler loading of TD5 (6.7%), TD10 (21.31%), TD15 (24.95%), TD20 (37.92%), TD25 (44.06%) and elongation at break with the filler loading TD5 (13.9%), TD10 (25.65%), TD15 (36.97%), TD20 (43.67%), TD25 (50.93%) of r-HDPE/EVA/TD composites. Besides, the modulus of elasticity with the filler loading of TD5 (12.87%), TD10 (18.08%), TD15 (26.136%), TD20 (44.41%) and TD25 (72.781%) and thermal stability increased. At similar filler loading (TD 15) with the incorporation of compatibilizer (MAH, PEgMAH, and CL) in r-HDPE/EVA/TD composites presented an improvement in tensile properties (2.08%, 2.68%, and 6.41%), modulus of elasticity (12.67%, 16.52%, and 25.2%) respectively and thermal stability but lower elongation at break (7.42%, 10.54%, 13.65%) and mass swell percentage (4.09%, 6.53%, 9.61%) compared to r-HDPE/EVA/TD composites. Scanning electron microscopes analysis also shows that the incorporation of compatibilizers improved the interfacial adhesion between TD and r-HDPE/EVA phase as shown in SEM micrographs. FTIR analysis proved the presence of MAH, PEgMAH and CL in the r-HDPE/EVA/TD composites. The presence of Caprolactam (CL) as compatibilizer in r-HDPE/EVA/TD composites shows good best compatibilizer compared than others.

CHAPTER 1

INTRODUCTION

1.1 Research Background

Over past few decades, it is well known that polymer have substituted many conventional and waste materials in many applications. This is for the reason that polymer have more advantages than conventional material. By considering the economic and environmental value polymer composite is one of the most significant and popular method of plastic production that filling of polymer with the waste material (Wool & Sun, 2011). Polymeric composites are formed by combining fiber and polymer resins which also known as fiber-reinforced plastic (Strong, 2008). Fiber-reinforced polymer composites got a lot of attention among materials as many researcher and engineers are considering developing an environmental friendly material that can be replacing currently used glass or carbon fiber in fiber-reinforced composites (Supri et al., 2011). The tensile properties of polymer composites can be improved as additives or coupling agent was added in the system (Supri et al., 2010). The additives that make physical interaction with the polymers may involve in the formation of the covalent bond. It is known as reactive compatibilizer (Zhou et al., 2006).

The addition of small additives can greatly enhance the mechanical and thermal properties of polymers and elastomers (Kalia et al., 2009). Besides that, a new combination

of properties can be achieved by the addition of filler to polymer blends that also can expand their range of application.

Among the waste material in industrialized region, the waste automotive material represents one of the problematic areas to be addressed such as tyre. Scrap vehicle tyre make a significant contribution to the generation of waste. For instance, the rate of scrap tyre generation in industrialized countries approximately one passenger car tyre equivalent to 9kg per capita per year (Snyder, 1998). In addition, it is estimated that an additional 2-3 billion scrap tyre are stockpiled on abandoned piles around the world that the cumulative scrap tyre generation approximately 10 years (Brown, June 2001). Tyres give an extensive defiance to sustainability of environment (Aisien et al., 2003; Adhikari et al., 2000). Other than that tyres have become a largest volume of waste rubber and do not degrade efficiently. This has attracted a thoughtful concern over the danger obtain to public health as well as environment. A number of possible applications of various form waste rubber in broad disciplines have been studied and reported. Tyre dust is one of the inorganic filler that can be used in polymer composites. Noriman et al. (2010) reported that the addition of waste tyre dust seems effective in improving the overall tensile properties, swelling behavior and morphology of the polymer blends.

High density polyethylene (HDPE) is suitably used as a packaging and manufacturing products due to its properties such as large strength to density ratio and has little branching. HDPE is stronger than the standard PE that can act as an effective barrier against moisture and remains solid at room temperature (Salih et al., 2013).

It is well known that recycling contributes to reduction in resources consumption and pollution. Recycled high density polyethylene (r-HDPE) is used to manufacture lawn, garden products, buckets, crates, office products, and automobile parts. Application of r-HDPE is often limited due to its low impact strength and Young's modulus properties, particularly at low temperature and high temperature loading conditions (Dikobe & Luyt, 2006). Blending r-HDPE with different polymer is an economic and effective way to improve these shortcomings (Duquesne et al., 2003). r-HDPE/EVA blends are widely used in many applications such as multilayer packaging, shrinkable film, and wire and cable coating (Nyambo et al., 2009). Blending EVA at different ratios with r-HDPE may improve the toughness, transparency, environmental stress cracking resistance (ESCR), and the capacity of the filler carrying (Faker et al., 2008).

Recently numerous engineers and researchers have been conducting some studies on the mechanical properties, especially interfacial performances of the composites based on the inorganic filler due to the poor interfacial bonding between the hydrophilic inorganic filler and the hydrophobic polymer matrices (Zhang et al., 2002). Incorporation of suitable compatibilizer onto the filler surface is an obvious solution in order to modify interaction or improve the bonding of filler to polymer by either altering the strength or changing the size of the interaction (Supri et al., 2010).

The compatibilizer has been used to improve dispersion, adhesion and compatibility for systems containing filler and the matrix in the composites (Liu et al., 2008). The agent modified the interface by interacting with the polymer, thus forming a link between the components (John et al., 2011). The main objectives of this study are therefore to examine the effect of tyre dust loading on the mechanical properties and thermal behavior of

recycled high density polyethylene/ ethylene vinyl acetate blends. Besides that, the compatibility of the recycled high density polyethylene/ ethylene vinyl acetate/ tyre dust (r-HDPE/EVA/TD) composites has been investigated by adding various type of compatibilizer.

Data obtained from this study will be useful in areas of potential applications such as construction material, electrical and electronic, appliance housing and automobile.

1.2 Problem Statement

Polymer blending is an alternative approach to obtain new materials with desirable properties based on commercially available polymer rather than to design and synthesize completely new polymer. Polymer blend providing materials with full desired properties at the lowest price meanwhile offer the means for industrial waste plastic. Others there can improve process ability, quick formulation changes and high productivity (Yu et al., 2006). Unfortunately, there are some of the major issues involved with polymer blending have been introduced. Blending two different kind of polymer are most likely become immiscible blends. These immiscible polymer blends lead to a phase separated and not thermodynamically stable (Lipatov, 2006).

The addition of tyre dust filler to polymer blends is meant of achieving new combination of properties and expands their range of application. Awang et al. (2008) reported that the addition of waste tyre dust seems effective in improving the overall tensile properties, swelling resistance, and morphology of polypropylene-based blends. However,

with different polarities of the polymer blends will produce the immiscible blends that lead to incompatibility polymer blends.

The incorporation of compatibilizer onto the filler surface is an accessible solution in order to modify interaction or increase the bonding of filler onto polymer by either altering the strength or changing the size of the interaction. Recycle waste rubber in one form to another is as old as the industrial use of rubber itself. In 1910, natural rubber cost nearly as much as silver, and it thus made perfect sense to reuse much as possible of this valuable commodity. Until now, the average recycled content of all rubber products was over 50 %. By 1960, the recycling content in rubber products dropped to around 20 %. As of 1990, the established tyre and rubber industry used only 2 % recycled material. Therefore, one of the various problems as enters 21st century the problem is waste disposal management. Abundant of waste rubber have created serious environmental problem because all waste rubber is non degradable material. Recycling contributes to reduce in resource consumption and pollution.

To solve this environmental issues, tyre dust obtained from waste rubber has been used in recycled high density polyethylene/ ethylene vinyl acetate/ tyre dust composite as a filler to achieved good combination of mechanical properties and process ability.