

MECHANICAL, RHEOLOGICAL, AND THERMAL PROPERTIES OF
CALCIUM CARBONATE FILLED POLYPROPYLENE/LINEAR LOW
DENSITY POLYETHYLENE COMPOSITES

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'Especially for my beloved parents and wife'

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ABSTRACT

The mechanical properties, melting temperature, glass transition temperature, crystallization temperature and rheological properties of polypropylene (PP) with varying weight percentage of linear low density polyethylene (LLDPE) at 10, 20, 30, and 40 wt % were studied. PP/LLDPE (60/40) was selected and investigated at different fraction of calcium carbonate (0-40%). The influence of CaCO₃ treated with 2% aminopropyltriethoxy coupling agent was also used to facilitate the link between filler and matrix. Tensile test, impact resistance, flexural test, viscosity, shear stress and shear rate of these blends were evaluated. Differential scanning calorimetry (DSC) was used to investigate the miscibility of PP/LLDPE blends and glass transition temperature was determined by dynamic mechanical analysis (DMA). The fractographic analysis of these blends was examined by scanning electron microscopy (SEM). Results indicated that the increase in LLDPE contents lead to decrease the tensile and flexural properties while the impact resistance of PP/LLDPE blends increase. However the increase in CaCO₃ amounts lead to increase both flexural strength and modulus. In the second part of this study, apparent viscosity of PP/LLDPE blends is affected by LLDPE contents due to lack of matrix reinforcement. On the other hand, incorporated of CaCO₃ into PP/LLDPE blends (60:40) has successfully increased the viscosity while CaCO₃ treated by aminopropyltriethoxy (AMPTES) coupling agent enhances the rheological properties. In the third part of this research, thermal properties were studied. Thermogravimetric analysis indicated that the total weight loss of PP/LLDPE/CaCO₃ composites decreases with increasing CaCO₃ loading. Heat deflection temperature of PP/LLDPE blends increases at all CaCO₃ loading.

ABSTRAK

Sifat-sifat mekanik, suhu kecairan, suhu peralihan gelas, suhu pengkristalan dan sifat-sifat reologi bagi polipropilena (PP) dan kepadatan rendah polietelina (LDPE) dengan peratus berat yang berbeza pada 10, 20, 30 dan 40 wt% adalah dikaji. PP/LLDPE (60/40) telah dipilih dan dikaji pada pecahan kalsium karbonat yang berbeza (0-40%). Kesan rawatan CaCO_3 dengan 2% pepadatan aminopropiltriethyloksi juga digunakan untuk memudahkan hubungan antara pengisi dan matrik. Ujian tegangan, ketahanan hentaman, ujian lenturan, kelikatan, tegasan ricih dan kadar ricih untuk campuran tersebut telah dikaji. Pembezaan imbasan kalorimetri (DSC) digunakan untuk mengkaji kebolehlarutan campuran PP/LLDPE dan suhu peralihan kaca ditentukan dengan analisis dinamik mekanik (DMA). Analisis fraktografi bagi adunan tersebut di kaji menggunakan mikroskop electron imbasan (SEM). Keputusan menunjukkan pertambahan kandungan adunan LLDPE mempengaruhi penurunan sifat tegangan dan lenturan bahan tetapi sebaliknya ketahanan hentaman adunan PP/LLDPE meningkat. Manakala, penambahan kandungan CaCO_3 dalam adunan meningkatkan kekuatan dan modulus lenturan. Dalam bahagian kedua kajian ini, kelikatan ketara untuk adunan PP/LLDPE dipengaruhi oleh kandungan LLDPE disebabkan oleh kekurangan tetulang matrik. Sebaliknya, penggabungan CaCO_3 ke dalam adunan PP/LLDPE (60:40) berjaya meningkatkan kelikatan manakala CaCO_3 yang telah dirawat dengan agen gandingan aminopropiltriethyloksi (AMPTES) meningkatkan sifat-sifat reologi adunan. Dalam kajian ini juga, sifat-sifat terma telah dikaji. Analisis pemeteran graviti haba menunjukkan kehilangan jumlah berat untuk komposit PP/LLDPE/ CaCO_3 menurun dengan peningkatan kandungan CaCO_3 . Suhu pesongan haba bagi adunan PP/LLDPE juga meningkat dengan kandungan CaCO_3 .

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LIST OF ABBREVIATIONS AND SYMBOLS

A	Constant related to the Einstein coefficient
ABS	Acrylonitrile butadiene styrene
AMPTES	Aminopropyl triethoxysilane
ASTM	American Standard of testing materials
b	Mean width of the specimens (m)
B	Related to the relative modulus of filler and polymer
CaCO ₃	Calcium carbonate
CO ₂	Carbon dioxide
d	Mean thickness of the specimens (m)
DSC	Differential scanning calorimetry
DTG	Differential thermogravimetric
FTIR	Fourier transform Infra red spectroscopy
HDPE	High density polyethylene
HDT	Heat Deflection Temperature
H ₂ O	Water
LLDPE	Linear low density polyethylene
L	Span between the centers of support (m)
MBS	Methacrylate butadiene styrene
NPDE	Non predefined elastomers
OPE	Oxidized polyethylene
PP	Polypropylene
PE	Polyethylene
phr	Part per hundred resin
R	Organofunctional group

Rpm	Rotational per minute
SEM	Scanning Electron Microscopy
TG	Thermogravimetric
TGA	Thermogravimetry analysis
T_g	Glass transition temperature
T_c	Crystallization Temperature
T_m	Melting temperature
UV	Ultraviolet
X_c	Degree of Crystallinity
W	Ultimate failure load (N)
W	Increment in load (N)
μm	Micrometer
γ	Shear rate
η	Apparent viscosity
τ	Shear stress
ΔH_f	Measured enthalpy of melting
ΔH_c	Measured enthalpy of crystallinity

CHAPTER 1

INTRODUCTION AND BACKGROUND

1.1 Current perspectives and future prospects: An overview

In recent years significant progress had been made in many areas of polymer blend and polymer matrix composite science and technology. Compounding of polymers with inorganic fillers was developed as versatile route leading to novel polymers with improved mechanical, rheological and thermal properties combined with cost and performance, the filled of materials science has lately focus on the research for composite materials that demonstrated positives characteristics of their compounds, the future work tendency to polymer blend and composite research; including established, as well as innovative, applications and new directions for these novel materials.

1.1.1 Polypropylene - polyethylene thermoplastic

Blends of polypropylene (PP) with polyethylene (PE) had attracted much commercial interest. One of the reasons for adding PE to PP is to improve the low temperature impact behaviour. Marked differences in properties between PP blends containing different polyethylene had been reported by (Utracki, 1990 and Dumoulin, 1995). Also it was found that PP and PE have many similarities in their properties, particularly in their swelling, electrical properties and solution behaviour. Despite of the many similarities the presence of a methyl group attached to alternate

carbon atoms on the chain backbone can alter the properties of the polymer in number of ways (Brydon, 1989)

In a 1960 patent application, 50-95 wt% PP was blended with PE for high impact strength and low brittle temperature (Sun Oil Co., 1964). In a Hoechst patent document, also deposited in 1960, PP was blended with 5-70 wt% LLDPE. The blends have good mechanical properties at low temperature (Holzer and Mehnert, 1963). Blends of PP with linear low density polyethylene 20-40% (LLDPE) were patented by Phillips petroleum to give material with high impact resistance and low brittleness temperature. It is worth stressing that there is a great variety of LLDPE with different structures, chemical compositions and molecular weight. Some example of such behaviour has been reported in the patent and open literature. In general, reducing the molecular weight of PP improves the fracture behaviour of PP/LLDPE blends (Utracki, 1990).

PP, due to its high technical and economic significance, has generated enormous scientific interest. However, its low mechanical properties compared with engineering thermoplastics create problems in its successful end-use application in various fields. To overcome these shortcomings, three procedure may be adopted, first is developing the copolymer of PP with other olefin, second is the addition of a nucleating agent that will help in lowering the dimensions of the spherulite, and the third solution is the preparation of a physical blend by incorporating a rubbery material in the desired concentration into a PP matrix. Various elastomers which were used for this purpose are well documented in the literature (Gupta *et al.*, 2004).

During the last few years, investigations of the melting and crystallization behaviour of polypropylene have become a subject of increasing interest. This is due to development of some new technologies, similar studied were conducted by (Silva *et al.*, 2005; Mader *et al.*, 1994; Wulforth and Tetzlaff, 1992) the thermoplastic polymers are used as matrices for fiber-reinforced composite materials. Short glass-fiber-reinforced PP widely uses as a light, stiff, and strong material, having a higher temperature resistance than the PP homopolymer. Thus, a system consisting of

polymer matrix, elastomer, and filler (glass fiber) could have been an attractive material for numerous engineering applications.

PP is a semi crystalline polymer that has a specific physical and mechanical properties leading PP to be a distinctive polymer in many application produced in different tacticities (atactic, isotactic, and syndiotactic polypropylene) and degree of crystallinity. Generally, the isotactic and syndiotactic polypropylene have good rheological, mechanical and physical properties because of a high stereoregularity of chains, which increases the intermolecular forces and entanglements of PP chains as a result, the degree of crystallinity increased.

PP has been widely used for injection molding, extrusion, and film blowing processes due to the remarkable properties. However, polypropylene exhibits a poor impact resistance in specific applications particularly at low temperature because of high degree of crystallinity and relative high glass transition temperature (T_g) which is about $-10\text{ }^{\circ}\text{C}$ compare to the glass transition temperature for PE. For these reasons, PP blended with other polymers to improve the impact strength. Many research studies have been reported the blending of PP with impact modifier such as ethylene-propylene copolymer, ethylene-propylene-diene terpolymer, and rubber particles. It has well known that blending of Linear Low Density Polyethylene (LLDPE) with PP strongly influencing the morphology and mechanical properties of PP matrix, Liu *et al.* (2005) who investigated variety of fillers applied in polymer to achieve different performance properties. Different organic and inorganic materials were used as a filler to enhance properties of the polymer such as biodegradation, low cost production, and reinforced polymer.

1.1.2 Calcium carbonate and talc as fillers

Potschke and co-worker (2000) studied the natural organic and inorganic materials have been widely used in plastic composite such as calcium carbonate (CaCO_3), talc, kaoline, silica, starch and cellulose. One of the most commonly

material mixed with polymers as extenders or for reinforcement is calcium carbonate. CaCO_3 is natural and environmental friendly material. It has used in film blowing to produce plastic bags and other plastic products (Stamm, 1999). It has shown that CaCO_3 improved flexural modulus, and shrinkage of the plastic bags when mixed with polyethylene at weight ratio of 6%. Da Silva *et al.* (2002) explored the torque studies of filled PP with talc at different filler content. They found that when low filler content was added a decrease in torque values in relation to PP was observed. The result can be related to the fact that at low filler content a good dispersion in the PP matrix can be achieved resulting in a better interfacial interaction between PP and filler. At high filler content (15 wt%) an increase in torque value in relation to PP was observed which resulted from filler agglomeration.

Coupling agents are bi-functional molecules containing organic and inorganic ends used to improve compatibility, as a result, the mechanical properties and chemical resistance of composites improved. Surface modification of the filler particles with a coupling agent has been widely used to enhance the polymer-filler interface. Many coupling agents are used for polypropylene blend, Kremer *et al.* (2000). But the most widely used coupling agents are silane and titanate based compounds. The chemical structures of the coupling agents allow to interacting both the surface of the filler and the polymer matrix. It was used to eliminate weak boundary layers, a tough and flexible layer, develop a highly cross linked interphase region such as electric cables composed of cross linked polyethylene, improve the wetting between polymer and substrate, form physical bonds between the polymer and the filler, and acidify the substrate surfaces (Bledzki and Gassan, 1999; Brydson *et al.*, 1990). The inorganic group (X) of silane molecule will hydrolyze to produce silanol, which forms a siloxane bond with inorganic material (CaCO_3). The organic group (Polymer) of the silane molecule will react with the polymer to produce a covalent bond as result both of them are tightly bound together as showed in Figure 1.1.

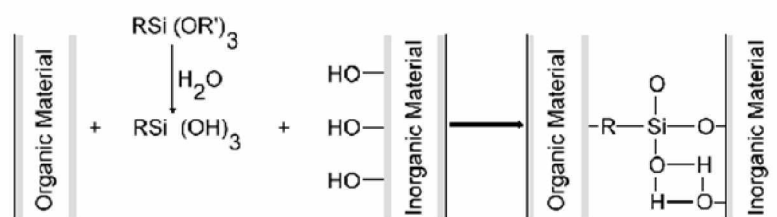


Figure 1.1: Mechanism of silane coupling agent

1.2 Problem statements

One of the most important aspects in the materials development of engineering thermoplastics is to achieve a good combination of properties and processability at a moderate cost. As far as mechanical properties are concerned, the main target is to strike a balance of stiffness, strength and toughness.

Adding filler to polymers will affect the following:

- Increase stiffness (modulus) but reduced toughness (impact strength).
- Increase the viscosity and decrease melt elasticity of the polymer.
- Increase the thermal stability of composites.

Hence, studies the mechanical, rheological, and thermal properties are important to achieve properties according to the requirements applications.

The study was focus on the effect calcium carbonate as a filler to the PP/LLDPE blends, many studies have been carried out to use natural resources to the polymer in order to an improvement for the plastic products such as mechanical, physical and electrical, several factors that influence and affects the properties of the composites need to be considered. Thus the optimum formulation of the composite will be investigated. Therefore, the questions that need to be answered in this area of the research are:

- 1) What is the optimum of treated CaCO_3 should be added to the PP/LLDPE blends polymers to achieve a balance mechanical, thermal, rheological properties and processibility?
- 2) What is the processes condition that effect to both of flexural modulus and impact resistance?

In spite of plenty studies have been reported by using CaCO_3 filled polypropylene. However, there is not much study have been applied calcium carbonate in particularly treated by aminopropyltriethoxy (APMTES) coupling agent. The study was particularly focused on using treated CaCO_3 . Based on theoretical fundamentals of PP/LLDPE blends, the practical side was important to design better processing equipment and determine optimal processing conditions, thus. The effect of the filler loading, type and treatment of the filler were studied with regards to the rheology behaviour and as well as the extruder swell, also the mechanical phenomena of the blends.

1.3 Objectives of research

The objectives of this works are:

- 1) To study the effects of LLDPE contents to PP on mechanical, thermal and rheological properties.
- 2) To investigate the effect of CaCO_3 loading on the mechanical, thermal and rheological properties.
- 3) To determine the effect of silane coupling agent on the mechanical, thermal and rheological properties incorporated of calcium carbonate into PP/LLDPE blends.

1.4 Strategic of the research

Blending process is great interest in polymer processing for manufacturing of many plastic products when low cost and good material performance are needed. Polypropylene has a limit of impact resistance compared with polyethylene and other plastics materials. It has been used as an engineering material for production of many products in several applications. In addition, polypropylene exhibits poor impact strength particularly at relative low temperature. Linear low density polyethylene may be blended with PP when high impact strength products are required. Different inorganic materials have been used as additives to improve both processing parameters and material properties. Calcium carbonate has been used as filler in the polymer processing to improve the mechanical properties of the polymers. The purity of CaCO_3 in some mining areas reached to 99.5% (Doufnoune *et al.*, 2006). It is great interest to investigate the use of calcium carbonate as filler for PP-LLDPE composite and explore new applications. This research was focused on the addition of treated calcium carbonate with polypropylene and linear low density polyethylene blends.

1.5 Scope of research

1. In the first part, the sample preparation includes the following stages:
 - Dry blending,
 - Mechanical mixing of samples,

In the second part, twin screw extruder used to produce pellets of PP/LLDPE blends followed by injection moulding of PP/LLDPE used to study the effect of mechanical, thermal, and rheological properties of PP/LLDPE, optimal results of blending were used. In additional of CaCO_3 particle size is $2.6\mu\text{m}$ treat with 3-

aminopropyltriethoxy silane was used to study the effect of mechanical, thermal and rheological properties. CaCO_3 weight ratios were at 10%, 20%, 30% and 40%.

2. Mechanical properties of PP blending with varying percentages of LLDPE were determined especially,

- Tensile properties
- Impact properties
- Hardness test

3. Melting and crystallization behaviour of a series of blends of PP with varying percentages of LLDPE were investigated.

4. Based upon the optimum formulation of PP/LLDPE blends. (10-40 wt%) weight percentages of CaCO_3 treat by Aminopropyltriethoxy (AMPTES) were conducted to determine the effects of mechanical, thermal, and rheological properties.

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