STUDIES ON THE PROPERTIES OF HYBRID MINERAL FILLERS FILLED POLYPROPYLENE COMPOSITES

by

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Thesis submitted in fulfillment of the requirements for the degree of

Master of Science

April 2013

DECLARATION

I hereby declare that I have conducted, completed the research work and written the dissertation entitles “Studies on the Properties of Hybrid Mineral Fillers Filled Polypropylene Composites”. I also declare that it has not been previously submitted for the award of any degree or diploma or other similar title of this for any other examining body or University.

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ACKNOWLEDGEMENTS

My deepest gratitude to Allah for the guidance, strength and inspirations that help me to go through this journey and complete my master. I dedicate this thesis especially to my Mama (Noor Azian Zackaria) and Papa (Rizal Vincent Abdullah) for their love, motivation and understanding.

My deepest appreciation goes to my supervisor, Prof. Dr. Ir Mariatti Jaafar who is always there, giving me the guidance, motivation, advice and knowledge while doing this research. The same goes to my co-supervisor, Assoc. Prof. Dr. Samayamutthirian Palaniandy.

Special thanks to all the technicians that help me directly and indirectly along the line especially to Mr. Gnanasegaran N.B Dorai, Mr. Shahril Amir, Mr. Suharuddin, Mr. Mohd Sofi, Mr. Meor Mohammad Noh and Mr. Abd Rashid Selamat.

My special thanks to all of my friends in USM especially Chhor Darong, Fasiha Zaaba, Shazlin Shaari, Komethi Muniandy, Safwan Hamzah, Kouth Phommachanh, Lay Makara and Sarah Ishak for all the greatest moments that we had. To my best friends, Sabrina Mohamad and Norhana Md Drus for always being there through my thick and thin of life. My deepest appreciation to Phoumiphon Nordala for always believes in me and for the support.

Finally, I would like to acknowledge School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia and Japan Advanced of Science and Technology (JAIST) for the research experiences. Last but not least the Ministry of Higher Education for the sponsorship.

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LIST OF ABREVIATIONS

ABS	- Acrylonitrile Butadiene Styrene
ASTM	- American Society for Testing and Material
$CaCO_3$	- Calcium Carbonate
CC	- Calcium Carbonate
CC/M	- Calcium carbonate/Mica
CC/T	- Calcium Carbonate/Talc
CTE	- Coefficient of Thermal Expansion
DMA	- Dynamic Mechanical Analysis
DOC	- Degree of Crystallinity
DSC	- Differential Scanning Calorimetry
EPS	- Expandable polystyrene
GMT	- Glass mat thermoplastic
LDPE	- Low density polyethylene
LLDPE	- Linear-low density polyethylene
M	- Mica
MFI	- Melt Flow Index
PA	- Nylon 6/6
PA12	- Polyamide 12
PC	- Polycarbonate
PE	- Polyethylene
PET	- Polyethylene terephthalate
PMMA	- Polymethyl methacrylate
PP	- Polypropylene

PPO	- Polyphenylene Oxide
PS	- Polystyrene
PUR	- Polyurethane
PVC	- Polyvynil Chloride
SEM	- Scanning Electron Microscopy
SI	- Silica
SI/M	- Silica/Mica
SI/T	- Silica/Talc
T	- Talc
TGA	- Thermogravimetry Analysis
TPO	- Thermoplastic Olefin

LIST OF MAIN SYMBOLS

E’	- Storage modulus
E’’	- Loss modulus
G’	- Dynamic storage modulus
G’’	- Dynamic loss modulus
L	- Original length of sample at room temperature
$T_{5\%}$	- Initial degradation temperature
$T_{50\%}$	- Degradation temperature at 50 wt%
Tan δ	- Tan delta
T_c	- Crystallization temperature
T_g	- Glass transition temperature
T_m	- Melt temperature
T_o	- Onset temperature
V	- Linear burning rate
W_f	- Weight fraction of filler

KAJIAN KE ATAS SIFAT-SIFAT KOMPOSIT POLIPROPILENA TERISI MINERAL HIBRID

ABSTRAK

Dalam kajian ini, sifat-sifat komposit terisi tunggal (mika, silika, talkum dan kalsium karbonat) serta komposit terisi mineral hibrid; silica/talkum (SI/T), $CaCO_3$/talkum (CC/T), silica/mika (SI/M) dan $CaCO_3$/mika (CC/M) di dalam polipropilena dikaji. Oleh kerana Malaysia mempunyai sumber mika yang banyak, maka ada kemungkinan untuk menggantikan talkum yang diimport. Maka dalam kajian ini, sifat-sifat komposit PP terisi tunggal serta hibrid mika dan talkum pada nisbah yang berbeza ke atas sifat-sifat mekanikal, termal, pembakaran dan reologi dikaji. Komposit PP dicampurkan dengan menggunakan Haake serta dimampatkan pada suhu 180 °C dan disediakan mengikut keperluan ujian. Dalam komposit PP terisi tunggal, mika menunjukkan kekuatan moduli dan tensil yang lebih baik berbanding mineral tunggal yang lain. $CaCO_3$ pula menunjukkan kekuatan impak yang lebih tinggi berbanding mineral tunggal yang lain. Dalam sistem hibrid SI/T dan CC/T, sistem CC/T menunjukkan peningkatan sebanyak 11.16% di dalam kekuatan moduli dari nilai PP yang tidak terisi mineral untuk setiap pertambahan T. Tetapi, sistem SI/T menunjukkan penurunan di dalam kekuatan moduli. Di dalam kekuatan tensil, sistem CC/T menunjukkan peningkatan manakala sistem SI/T menunjukkan penurunan untuk setiap kenaikan 10 wt% talkum. CC/T juga menunjukkan sifat termal yang baik dengan menunjukkan nilai CTE yang rendah serta kestabilan termal yang tinggi pada nisbah talkum yang tinggi. Sistem SI/T menunjukkan kekuatan impak dan kadar pembakaran yang lebih bagus berbanding sistem CC/T. Untuk sistem hibrid SI/M dan CC/M, kedua-dua sistem ini menujukkan kenaikan di dalam kekuatan moduli dimana CC/M mempunyai peningkatan yang lebih tinggi berbanding sistem CC/M. Sistem CC/M menunjukkan sifat kehabluran, kestabilan termal dan kadar pembakaran yang lebih baik berbanding sistem SI/M. Berdasarkan kesemua sifat yang dibincangkan, dapat disimpulkan bahawa sistem SI/M mempunyai sifat-sifat yang terbaik berbanding system hibrid yang lain kerana menunjukkan peningkatan yang lebih tinggi di dalam mekanikal, termal dan kadar pembakaran yang lebih baik berbanding dengan sistem hibrid yang lain.

STUDIES ON THE PROPERTIES OF HYBRID MINERAL FILLERS FILLED POLYPROPYLENE COMPOSITES

ABSTRACT

In the present study, properties of single composites (mica, silica, talc and $CaCO_3$) and hybrids of silica/talc (SI/T), $CaCO_3$/talc (CC/T), silica/mica (SI/M) and $CaCO_3$/mica (CC/M) filled polypropylene (PP) composites were investigated. Due to the availability of local resources of mica filler in Malaysia, there is a possibility to replace the imported talc with mica. Hence in this study the properties of single PP composites, talc and mica hybrids filled PP composites at different filler ratio on the mechanical, thermal, flammability and rheological properties of PP composites were investigated. These hybrid composites systems were compounded using Haake internal mixer and hot compressed at 180 °C as per requirements for testing. In single mineral filler filled PP composites, mica showed better Young's modulus compared to the other fillers and high tensile strength at high content of mica filler. $CaCO_3$ filler demonstrated the highest values of impact strength compared to other types of fillers. In hybrid composites of SI/T and CC/T, CC/T system showed 11% increasing in Young's modulus values with respect to the unfilled PP with increasing T. However, SI/T system has a decreasing trend of Young's modulus. The same trend was observed in tensile strength, where CC/T hybrid composites showed increasing trend whilst SI/T hybrid composites exhibited a decreasing trend with every 10wt% increase of talc filler loading. CC/T composites also showed good thermal properties where it exhibits lower CTE values and higher thermal stability with high filler loading of talc. On the other hand, SI/T composites showed better impact strength and flammability if compared to CC/T composites. For hybrid composites of SI/M and CC/M, both composites have improved the stiffness with increasing amount of mica filler where CC/M composites demonstrated higher increment compared to SI/M composites. CC/M composites demonstrate higher crystallization, degradation temperatures and flammability compared to SI/M composites. Thus based on the properties discussed, it can be concluded that SI/M emerged as the best hybrid

composites compared to the other three hybrid composites as it demonstrated higher increment in mechanical, thermal and have good flammability characteristics.

CHAPTER 1
INTRODUCTION

1.1 General Introduction

The growth of modern plastics has expanded for the first 50 years in 20th century where new types of polymers were introduced. The introduction of thermosetting plastics, thermoplastics, natural polymers, modified natural polymers and biodegradable plastic proved to the success of plastic materials industry. There are hundreds of plastic available commercially but only a few are recognized as the commodity thermoplastics due to the high volume consumptions of these thermoplastics as well as relatively low price (Andrady & Neal, 2009).

The world demand of plastic according to the types of resin can be seen in Figure 1.1. Poly (vinyl chloride) (PVC), polypropylene (PP), low density polyethylene (LDPE) and linear-low density polyethylene (LLDPE) represent the highest percent of plastic demand. This is followed by other types of plastics namely expandable polystyrene (EPS), polystyrene (PS), poly (ethylene terephthalate) (PET) and polyurethane (PUR).

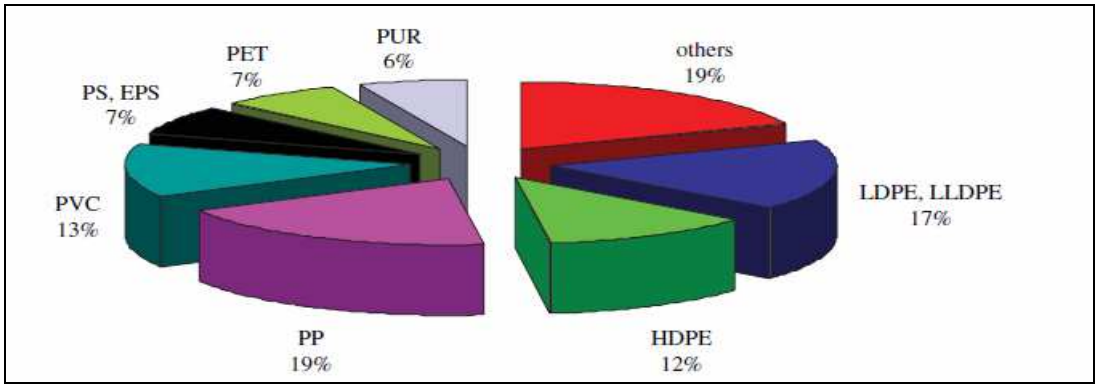

Figure 1.1: World plastic demand by resin types in the year of 2006 (Andrady & Neal, 2009).

Combinations of two or more material bring about the advantage of having the characteristic of each component to complement each other. This holds great potential to pick the most desirable combination of properties from numerous materials to match demands of the various industries. Thermoplastic composites have attracted lots of interest with their unique properties. Combination of polymer and particulate fillers can potentially improve the properties of polymer in terms of mechanical and thermal properties and yet promise cost reduction. Their properties are influenced by several factors namely the filler characteristics, filler content, interfacial adhesion, etc (Deshmukh *et al.*, 2011; Hussein *et al.*, 2011).

One of the commonly used thermoplastic materials is polypropylene (PP). PP offers several advantages including balance of strength together with modulus and chemical resistance at relatively low cost. In today applications, such as in automobiles, particular appliances and some commercial products, it is relatively important to have creep resistance, stiffness, toughness and good performances over a range of temperature and deformations rates (Zhou & Mallick, 2002). In order to

achieve this, the usage of mineral filler filled in PP composites to fulfil the demands had grown exponentially.

Chronologically, inorganic minerals such as glass fiber, talc, calcium carbonate, etc were initially used as fillers in polymer composites. This is closely followed by the natural organic fillers (Morreale *et al.*, 2011). Calcium carbonate is the most commonly type of mineral fillers that are incorporated into plastic, specifically polypropylene (PP) due to its lower price and its filler loading that can be manipulated to high filler loading. Flaky mineral filler such as mica, kaolin and talc on the other hand could bring a good advantage to improve rigidity of composites. It is also important for polymer composites to have exceptionally low coefficient of thermal expansion (CTE) because it can reduce fracture when there is a sudden change in temperature. With the incorporation of mineral fillers mentioned above including silica, this can help in reducing the value of CTE in polymer composites (Nurdina *et al.*, 2009). Apart from that, incorporation of inorganic mineral fillers can help to enhance the performance of polymer composites by improving the stiffness, heat distortion temperature, toughness, dimensional stability and hardness. It is worth to note that the particle shape, loading, size and dispersion of fillers affect the properties of polymer composites (Azizi & Faghihi, 2009; Kaleel *et al.*, 2011).

Studies on hybrid composites is growing in recent years and from the trend observed, hybrid composites proved to be one of the materials with many potentials that can be manipulated into diversified material properties. The importance of hybrid composites can be evaluated into three main aspects. Firstly, the demand of hybrid composites is mainly due to the economic advantage. Through hybridization, reduction of cost is the main focus by combining both expensive and cheap materials

but with advantages from both materials. Specific characteristics can be obtained as combination of hybrid materials could offer wider options to developed materials for specific design and purpose. Finally, through hybrid materials the improvement in mechanical and functional properties could be optimized when hybrid materials interact with each other as it is combined (Babu *et al.*, 2005). With good selection of filler reinforcement and processing techniques, hybrid composites can be produced to suit various practical requirements (Kord, 2011).

Azizi and Faghihi (2009) had studied the hybridization effects of talc and calcium carbonate. It is found that good impact properties can be obtained through the combination of talc and calcium carbonate due to positive characteristics that exist between the two fillers. Hybrid of two types of calcium carbonate also contributes to better mechanical properties compared to those of composites filled with single-filler calcium carbonate.

1.2 Problem Statement

Accelerated growth in the utilization of mineral fillers in plastic composites is provoked by the needs of plastic industry to reduce cost and to improve specific properties in their products. One of the typical thermoplastic namely polypropylene (PP) is the major player in the industry that can be filled with broad range of mineral fillers. It is packaged with good heat stability, low density and chemical resistance but its application is somewhat limited by its low mechanical properties (Yazdani *et al.*, 2006). Thus, properties of the matrix can be altered through the addition of mineral fillers as it is simple and cheap. This is the reason why particulate filled

polymers still become the subject of interest in industrial and research fields. With the incorporation of fillers, properties namely the strength, stiffness, hardness, dimensional stability, etc. could be improved (Guerrica-Echevarriâa *et al.*, 1998).

With its softness, lubricity and together with excellent wetting and dispersion in plastics, talc become one of the important mineral to be used as a filler in polymer. Talc platy structure is a well known reinforcing filler that is applied in various industrial and commercial applications like in packaged components, appliance markets and automotive industry (Saq'an *et al.*, 2008). Other reinforcing filler that has similar platy structure as talc is known as mica. Mica could improve properties of polymer as it has excellent mechanical, electrical and thermal properties (Nurdina *et al.*, 2009).

Calcium carbonate and silica are other type of potential fillers to be used in the polymer. Malaysia's abundant reserves of calcium carbonate located in Perlis, Kedah, Perak, Selangor, Negeri Sembilan, Pahang, Kelantan, Sabah and Sarawak accumulates to a total of 12,000 million tonnes. As for silica, the distribution of this mineral filler in Malaysia could be found in the states of Johor, Perak, Terengganu, Kelantan, Sabah and Sarawak. Mica on the other hand is coming from the state of Perak that is rich in the muscovite type in platy shape. It is then processed into various sizes of ground mica powder and can be used as the reinforcement in plastics (Mineral, 2009).

In the previous work done by several researchers from the year of 2004 to 2010, combination of various mineral fillers such as talc/glass fibers, talc/kaolin and talc/calcium carbonate had been done. It was found out that hybridization of these fillers had significantly improved the mechanical and thermal properties although

properties of fillers such as $CaCO_3$ and kaolin composites are facilitated with the aid of surface treatment and coupling agent. However more attentions are focused towards hybridization of talc/calcium carbonate where at least six researches had carried out the study on talc/calcium carbonate filled PP. Following the improvements by the combination of talc and calcium carbonate, it is motivated to investigate different combination of mineral fillers and to find the partial replacement of talc filler with mica as talc is currently being imported from sources outside Malaysia. Apart from mica, Malaysia is sanctified with vast resources of mineral fillers such as silica and calcium carbonate. With these resources, it is fascinating to study the effect of adding these fillers in PP. Combination or hybridization of these fillers in PP will produce a new material with desired properties. There are several works being done on hybrid fillers composites that investigated the hybrid effects of talc/glass fiber, talc/kaolin, talc/calcium carbonate but they only focus on one type of hybrid composites. Therefore this present study had investigated and compared four types of hybrid composites between talc/calcium carbonate with talc/silica and mica/silica with mica/calcium carbonate hybrid fillers composites.

1.3 Objectives of Study

In this research work, four types of mineral fillers; silica, mica, calcium carbonate and talc in PP composites were investigated. The objectives in this research work are:

1) To investigate different types of mineral fillers and different percentage of filler loadings on the mechanical, thermal, physical, flammability and rheological properties of PP composites.

2) To study the effect of four types of hybrid mineral fillers filled PP composites.
3) To investigate the possibility of replacing talc filler with mica filler in hybrid PP composites.

1.4 Outline of Thesis Structure

Chapter 1: This chapter begins with the general introduction on thermoplastic composites, its advantages and benefits. The problem statement, objectives and the importance of this research works are specifically stated.

Chapter 2: Various literature reviews that related to polymeric materials and mineral fillers are reviewed. In addition, factors that influence the properties of particulate fillers filled in polymer composites, hybridization effects of mineral fillers on polymer composites and the applications of polymer composites in automotive industries are reported in this section.

Chapter 3: In this chapter, detailed information on the materials, methods and testing procedures used to characterize the properties of single and hybrid PP composites are provided. Descriptions on the laboratory equipments used are specifically stated.

Chapter 4: Analysis and discussions on characterization of mineral fillers, single filler composites and hybrid PP composites are reported in this section. Four types of single filler composites and four types of hybrid filler composites are characterized and divided into three sections based on the types of composites. Each section will

have the analysis and discussions that revolves on melt flow index (MFI), density determination, void content, mechanical properties, thermal properties, burning rate, oscillatory shear modulus and morphology of the composites in which the filler loading of mineral filler is fixed at 10 wt%, 20 wt%, 30 wt% and 40 wt% in single composites and 40 wt% for hybrid composites. The effect of filler ratio for hybrid composites were investigated at 10/30, 20/20 and 30/10 ratio in weight percent. Comparison of properties in single and hybrid systems are made to find out the best PP composites systems.

Chapter 5: Conclusions obtained from this research work are presented in this chapter together with the suggestions for future research.

CHAPTER 2
LITERATURE REVIEW

2.1 Thermoplastic: Polypropylene

Polymers can be processed by injection molding, extrusion, compression molding, etc and these polymers can be referred to as “Plastics” (Ibeh, 2011). With basically large molecular weight that consist of recurring monomer units, polymer can be categorized into two; thermoplastics and thermosets. Thermoplastics can be further divided into amorphous and semicrystalline types. These two types of thermoplastics are different in the sense that amorphous thermoplastic have 0% crystallinity but on the other side semicrytalline thermoplastics are determined by various degree of “crystallinity”. Commodity thermoplastics namely polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), and polystyrene (PS) are the four most popular thermoplastic due to the average mechanical properties that they have and cost less with high consumption volume that contributes to the consumption rate of 5%-10% per year (Ibeh, 2011). Table 2.1 shows the consumption volumes of the four major commodity thermoplastics mentioned above.

Table 2.1: Consumption volumes (sales, billions of Pounds) of the four major commodity general purpose thermoplastics (Ibeh, 2011).

Material	**1998 (€)**	**2000 (€)**	**2002 (€)**	**2004 (€)**	**2006 (€)**
Polyethylene (PE)	30.16	33.33	35.71	37.91	38.01
Polypropylene (PP)	13.84	15.58	17.08	18.52	18.37
Polyvinyl Chloride (PVC)	14.68	14.36	15.25	15.88	14.90
Polyestyrene (PS)	6.62	6.68	6.77	6.77	6.19

One type of thermoplastics polymer that is produced from the polymerizing of many propylene monomer is known as PP. The monomers of propylene can be linked through different approach but the common form of PP is fabricated through crystallizable polymer chains that are produced together with a catalyst. Together with this form of PP, semicrystalline solid product would have improvements in physical, mechanical and thermal properties. From semicrystalline PP production, another form of PP is produced as the byproducts which are used in the applications such as adhesions, sealants and caulk. These byproducts are basically soft and tacky and has very poor mechanical and thermal properties (Kissel & Han, 2003).

Azizi & Faghihi (2009) in their report had mentioned that PP is adaptable in many applications as it is packaged with good properties of stiffness, have high service temperature, low in cost, with inertness and clarity. One recognized method to produce PP composites with different properties is through the incorporation of mineral fillers into the polymer with the purpose to further improve both stiffness and its heat deflection temperature as well as to produce PP composites for different types of applications. Thus, mineral filler akin to the name of calcium carbonate, talc,

mica, kaolin, wollastonite and glass fiber are those that are well established fillers to be filled in PP.

2.2 Polymer composites

Polymeric composites are one of the examples of materials that are widely used in majority of commercial applications. The examples of polymeric composites used including poly (vinyl chloride) (PVC), acrylonitrile butadiene styrene (ABS), filled thermosetting resins, glass fiber filled plastics, etc. Polymer composites are preferred over the single polymer due to several advantages such as increased in stiffness, toughness, dimensional stability, improved in mechanical damping and reduction in cost. The end product of composite materials depends on the properties of each component that compose the composites. For instance, shape of filler phase, morphology of the composites system and the interface that exists between the two phases will determine the properties of the composites materials (Nielsen & Landel, 1994). Composites material can be categorized into three general categories as presented in Figure 2.1 (Callister, 2007):

1) Particle-reinforced
2) Fiber-reinforced
3) Structural-reinforced

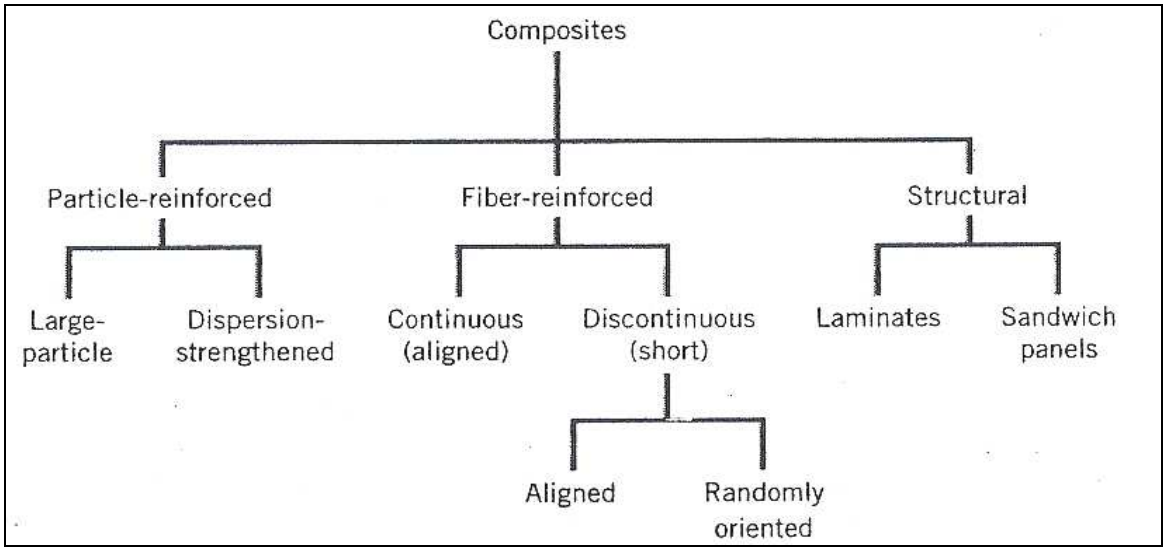

Figure 2.1: Principle composition of composites material (Callister, 2007).

Particle-reinforced composites can be divided into large-particle and dispersion-strengthened compositions. In large-particle compositions, particle-matrix interaction cannot be treated at molecular level; this is opposed to dispersion-strengthened compositions. In particle-reinforced composites, the particulate phase is normally harder and stiffer compared to its matrix. As this particulate filler is incorporated into matrix, it has the ability to restrict the movement of matrix phase. As the consequences of this, matrix will transfer applied stress to the particles. Thus, strong bonding between matrix-particle is important as it influenced the mechanical behavior of composites (Callister, 2007).

Among the particles used in particle-reinforced composites are small mineral particles, aluminium, and carbon black as been reported by Chandramohan & Marimuthu, (2011). Through the reinforcement using particulate fillers, ductility of matrix can be reduced, so as the cost of composites. One of the examples of particle-

reinforced composites is carbon black particle in poly-isobutylene elastomeric polymer (Chandramohan & Marimuthu, 2011). Compared to fiber-reinforced composites, particle-reinforced composites are much easy to process and cost less. Particulate fillers are added during polymer melt in an extruder or it also can be done by injection moulding during polymer processing (NDT n.d.).

Fiber-reinforced composites can range from cloth and paper in phenolic, glass fibers in epoxies, aramid fibers in polyamides, chopped glass fibers in polystyrene and etc. This type of composites could offer unusually high strength and stiffness on a weight basis. It is stated that even by using single fiber-resin the properties of high strength and stiffness could be manipulated over wide ranges. Both specific strength and specific modulus of fiber-reinforced composites can exceed the values obtained by metals (Nielsen & Landel, 1994).

In previous work done by Stefanescu et al., (2011), the studies on fiberglass-reinforced in polymer composites had been reported in the past decades. Due to its high specific stiffness and strength, this polymer composite is commonly applied in the applications that require high fracture toughness and fatigue durability. Example of reinforcement of fiberglass is in poly(methyl methacrylate), (PMMA) based materials. Together with barium titanate ($BaTiO_3$) nanopowder, this fiberglass-reinforced polymer composites have the potential to be applied in multifunctional capacitors as structural dielectrics due the excellent mechanical properties and better energy storage characteristics that they owned.

Structural composites consist of homogeneous and composites material. The outcome properties of this type of composites depend on the respective material properties that compose the structural composites and also depend on the geometrical

design. Two types of structural composites are recognized; laminar composites and sandwich panels. In laminar composites, the fiber-reinforced orientation layers are stacked alternately to each other. Honeycomb core is the example of sandwich panels that consist of face material and core. In this type of structural composites, the core material has low modulus of elasticity while the outer material is stiff and strong (Callister, 2007).

2.3 Typical mineral fillers for polymers

Fillers can be defined as solid materials that are capable of altering the physical and chemical properties of the original materials by means of surface interaction or through its own physical characteristics. By this definition, fillers can be understood as the material that gives significant improvement on large scope of properties for that particular composite. George (1999) had mentioned that fillers work in two ways as it is incorporated into a system. First, filler would contribute to the enhancement of properties by 'donating' its own properties to the compositions. Secondly, through important interactions that takes place between the fillers and the material, it would subsequently lead to the possibility of having strong chemical and physical interactions or it could be no interactions at all between the filler and the material combined.

Cost reduction through filling materials is a well known concept since the early area of polymer industry. Advance developments have been done to further refine the filler such that in fine-particle size technology, particle morphology, surface treatments and the development of coupling agent in order to achieve good

polymer-to-filler bonding so that the mechanical properties could be improved. Better understanding on the effectiveness of individual filler would help in optimizing the filler used. For instance, specific characteristics of different fillers would make them suitable in certain applications that require the properties needed. Basic properties requirements need to be identified before deciding on the optimum filler type and filler loading that will be used. Mostly, two different fillers will be blend together to balance the properties of composites (Lutz & Grossman, 2001).

With various mineral filler available around this world, it leads to the disputed questions whether there is any ideal filler and whether this ideal filler could be developed. The answer is simple as it all comes back to the needs of end applications that require particular set of characteristics in physical, chemical, electrical, etc. Thus, it is essential for researchers and users to identify the pro and cons of the possible mineral that they want to employ to be developed into polymer composites. Among the characteristics of the fillers that should be considered are the cost, the availability, good surface wetting and bonding, good chemical resistance and high strength (Katz & Milewski, 1987).

In Malaysia, over 33 different types of minerals can be found that includes metallic, non-metallic and energy minerals. Table 2.2 shows the statistical information on the amount of productions and value of non metallic mineral in Malaysia.

Table 2.2: Productions (tonnes) and value (RM millions) of non-metallic minerals in Malaysia in the year of 2010 (Mineral, 2009).

Minerals	Productions (tonnes)	value (RM millions)
Clays	25, 000000	175.00
Feldspar	400,000	35.00
Kaolin	500,000	40.00
Calcium Carbonate	22, 000000	220.00
Mica	4515	3.16
Sand And Gravel	23,0000	220.00
Silica	862,626	69.01

2.3.1 Talc

The production of talc can be made from several forms of ore. Talc is known as the reinforcing filler in many plastic applications due to its naturally platy shape. It can act as the extender and together with its high aspect ratio could give significant effect to improve the performance properties of particular compound. Majority of plastic user prefer to use talc that is white in colour and consist of thin platelets. With theoretical formula of $3MgO.4SiO_2.H_20$, the molecular structure of talc can be observed in Figure 2.2. Talc usually comes with the physical properties of white in colour, platy and has high brightness and dry in form from supplier. When platy talc is incorporated into composites, it will usually contribute to the improvement in higher stiffness and creep resistance at ambient or elevated temperature. At 40% of

filler loading, it could triple the stiffness of PP composites up to 600,000 psi compared to the unfilled PP that is only at 200,000 psi. But then, at high filler loading the impact strength of the composites is reduced but this is solved through proper selection of particle size, surface treatments and resin formulation (Radosta & Trivedi, 1987).

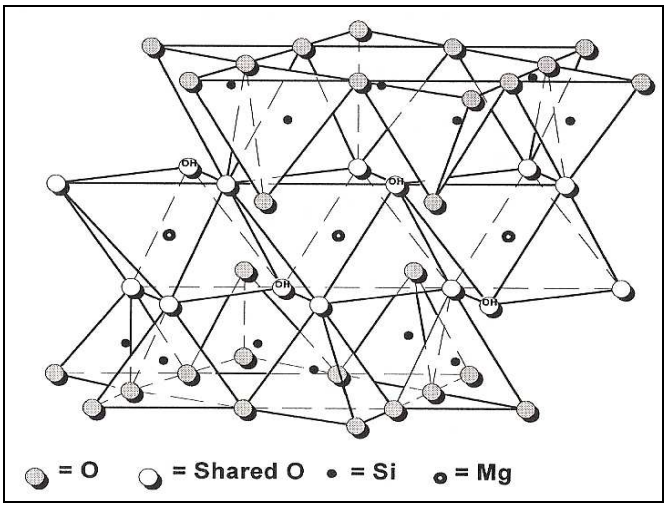

Figure 2.2: Molecular structure of Talc (Ciullo, 1996).

At relatively low shearing force, talc can be delaminated because only some very weak van der Waals forces that holds the planar surfaces of talc platy-like structure. This is the reason why talc feels slippery and thus it is easy to disperse. Important properties owned by talc namely high resistivity, low gas permeability, low abrasiveness and its lubricating effect are due to its plate-like structure (Xanthos, 2005).

The employment of talc in polypropylene (PP) started in 1960s in the application of under the hood automotive parts such as in fan shrouds and blades.

The evolution of talc reinforced in polymer continued with its application to replace stamped metal parts in pump housing, washer tubs and spin basket. This is followed by replacing polyurethane and acrylonitrile-butadiene-polystyrene (ABS) blends when talc is reinforced in thermoplastic olefin (TPO) to meet the demand of fascias and kick plates for lower cost. The usage of talc filler in TPO is extended during 1980s when the Japanese started to apply talc in automotive applications to manufacture automotive parts such as bumper, dashboard and instrument panel (Clark & Steen, 2003). Other applications of talc in industrial and commercial use includes refrigerator jackets, in packaged components and as blocker of infrared in agricultural films (Saq'an *et al.*, 2008).

As filler in polymer, talc is used in various amounts of weight percent from 20% to 40%. The largest plastic used with talc is PP. When talc is filled in PP, several modifications will take place. Talc filled PP would improve the crystallization temperature, mechanical properties and could induce short processing time in injection moulding. The PP nuclei is also increased as talc has the nucleating ability that leads to the present of active sites on composites surface (Ferrage *et al.*, 2002; Sancaktar & Walker, 2004). As talc is incorporated into plastic (especially PP), it would give a good balances between rigidity and impact strength. Finest talc is obtained from advanced milling technology without having to sacrifice on its reinforcing power. With high purity talc, long term thermal stability can be achieved and that make the compounds produced ideal to be use in packaging industry such as in odour-sensitive food contact applications. Talc has addition of benefit in colour processing because less pigment is required due to the whiteness and low yellow index (Murphy, 1996).

2.3.2 Silica

The common availability of silica, its chemical inertness and durability are the main reasons why silica is the major player in industry. Silica could be divided as natural or synthetics minerals. Natural silica consists of crystalline and amorphous. Example of crystalline silica includes sands, ground silica and quartz while amorphous silica is the diatomaceous earth (George, 1999). Fumed silica which is another type of amorphous silica is considered to be non-toxic and non-irritating as it is totally in amorphous state. This is different to crystalline silica that is termed as carcinogenic. Research work on fumed silica in thermoplastic has been investigated and they found that this silica has significant effect as a reinforcing filler when it is incorporated into thermoplastic matrix (Lin *et al.*, 2009). In Malaysia, the deposits of silica could be found in several states such as Johor, Perak, Terengganu, Kelantan, Sabah and Sarawak with estimated amount of 148.40 million tonnes (Mineral, 2009).

Primary particle shapes of silica depends on the natural deposits and it is commonly ranges from round to angular (Rothon, 2003). As it is ground, performance of silica is influenced by the particle size distribution and its acceptable topsize. When silica is applied as filler, it is commonly have the characteristics of low surface area, low-oil absorption and have angular particles that are graded by topsize. High hardness, chemical inertness, heat resistance, low coefficient of thermal expansion and good electrical insulation are the reasons why silica is used in polymer composites (Rothon, 2003).

Silica can function as toughening agent in PP composites. This is confirmed by research work done by Lipponen *et al.,* (2011) that investigated the toughening effect of microsilica compound with PP where microsilica increased the ductile-to-

brittle transition point as high as three orders compared to pure PP. Studies on silica filled in plastic also revealed the ability of silica to reduce coefficient of thermal expansion (CTE) as the result of giant molecular structure owned by silica (Nurdina *et al.*, 2009).

Flammability of silica is studied and its effectiveness as flame retardant is determined in previous work by Kashiwagi *et al.*, (2003). In this case, various types of silica is used and it was figured that low density silica such as fumed silica and silica gel give positive impact on polymer compound as it reduced the heat release and the burning rates of polymer. This is due to the thermal insulation layer that exists when particles from fumed silica and silica gel accumulate at the surface of polymer and prevent polymer concentration near the surface that lead to less flammable material. This result is contradicted when fused silica is used.

2.3.3 Mica

Potassium aluminum silicate hydroxide fluoride or commonly known as muscovite mica as presented in Figure 2.3 have the chemical formula of $KAl_2(AlSi_3O_{10})(F,OH)_2$,. This is the most common mica group. Muscovite mica has the colour of white to colourless and has aluminium silicate layered structures that are bonded by layers of potassium ions. These characteristics make muscovite mica flexible and elastic due to the perfect cleavage of thin sheets of flakes. In particular thermoplastic, mica is used as the reinforcing filler. Another type of mica is known as biotite with the chemical formula of $K(Mg,Fe)_3AlSi_3O_{10}(F,OH)_2$. As oppose to

muscovite mica, this type of mica has a brown colour and thus are less preferred to be used as filler in polymers (Kissel & Han, 2003).

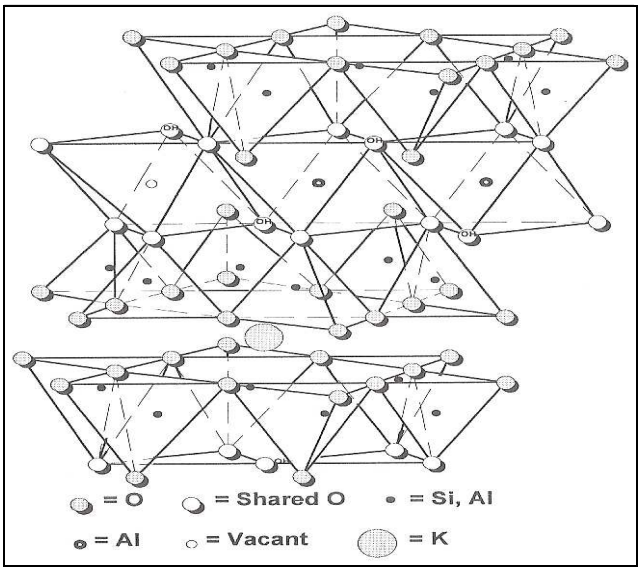

Figure 2.3: Molecular structure of muscovite mica (Ciullo, 1996).

The availability of mica could be found around the world especially in the United States, Canada, France, Korea, Malaysia, Mexico, Russia, Madagascar and India (Hamdani *et al.*, 2009). In Malaysia, the deposits of mica can only be found in the state of Perak (Mineral, 2009). Three common forms of mica are muscovite mica, phlogopite mica and biotite mica. These micas could be found in acidic igneous rocks, ultrabasic igneous rocks and in granites, respectively. As filler for plastic, the characteristics of mica depend on its ability to be processed to thin-plate shaped particles. And thus, higher aspect ratio is expected from mica when compared to other types of minerals fillers (Hamdani *et al.*, 2009).

With its platy shape, mica can bring about the advantage to PP composites. Uniform shrinkage as the results of similar dimension between length and width can eliminate warpage in molded parts. Flow of gases and liquids through PP parts are reduced and this is important when the end applications require good barrier properties (Kissel & Han, 2003).

Kissel & Han (2003) had reported that the increasing demand for material that have larger, thinner and stronger parts as well as lighter weight, higher heat stability and better surface appearance contribute to the use of mica as the reinforcement filler. Automotive, appliances and constructions are the major industries that are affected by the usage of mica-reinforced PP. In automotive applications, mica has the ability to improve scratch resistance and has higher mar resistance compared to talc-filled polyolefins. This is a good advantage of mica, therefore lots of researches are still going on to make good use of mica as filler. Other advantages of mica includes excellent electrical insulator, good thermal and sound dampening effect and it can act as UV radiation blocker at the wavelength below 300nm. In addition, mica has good thermal stability up to 450 °C, soft and non-abrasive. Apart from that, mica has the ability to enhance modulus performance, tensile and flexural strength where these properties improvements are dependent on the orientation, the degree of adhesion and the extent of orientation (Xanthos, 2010).

2.3.4 Calcium carbonate

There is an estimate of 12,000 million tonnes of calcium carbonate ($CaCO_3$) reserves in Malaysia that are located in the states of Perlis, Kedah, Perak, Selangor,

Negeri Sembilan, Pahang, Kelantan, Sabah And Sarawak (Mineral, 2009). Most commercial $CaCO_3$ products are mineral calcite that includes marble, natural limestone, chalk and precipitated calcium carbonate. When $CaCO_3$ is used as the filler, the preferred variant would be white in color with a high degree of mineralogy purity. Other criteria are based on the particle size, shape, surface area and liquid absorptivity. Generally, there are two types of $CaCO_3$ which are natural $CaCO_3$ and synthetic $CaCO_3$ (Ciullo, 1996).

The use of $CaCO_3$ as filler is mainly for its low cost and abundance of sources. The main reasons for its usage are for price reduction and to improve mechanical properties, heat stability, barrier properties and crystallization behavior (D'Haese *et al.*, 2011). Weon and Sue (2006) reported that as $CaCO_3$ is compounded with PP, the strength and fracture toughness are increased. $CaCO_3$ particles can behave as stress concentrators to provoke toughening mechanisms. In hybrid composites of talc/$CaCO_3$, $CaCO_3$ can help in improving the overall impact strength without having to sacrifice much on other mechanical properties.

$CaCO_3$ is one of the important mineral filler that can be associated with automotive applications. These applications includes automotive interior, exterior and under the hood parts. Other applications can be found in household items and furniture. Significant improvement with addition of $CaCO_3$ offered increase in stiffness, heat distortion temperature, retention and impact strength as well as its dimensional stability. It is reported that when 30% of $CaCO_3$ filler is incorporated into PP sheet thermoforming, the stiffness improvement is comparable to other types of polymer such as PS or PVC (Xanthos, 2010).

2.4 Factors that influenced the properties of particulate fillers filled polymer composites

The advantages of wider applications and low cost are the main reasons why particulate filler filled polymer composites are attractive. By adding inorganic material into the polymer matrix, various physical properties like strength, modulus and heat deflection temperature can be enhanced. Generally, factors that contribute to the mechanical properties of particulate filled polymer composites are particle loading, particle shape, particle size and the distribution of particles in polymer composites (Bose & Mahanwar, 2004).

2.4.1 Filler loading

Generally, in most commercial particulate filled polymer, the maximum usage of filler is at 40 wt %. This is due to the decrease of physical properties when the filler loading exceeds 40 wt%. However with the progress in processing and compounding techniques, the future composites might be able to have higher filler loading (Peters, 1998).

Filler loading has significant effect on processing conditions and the final mechanical properties. In previous report by Mortezaei *et al.,* (2011) mentioned that when low filler loading is incorporated into polymer, the filler would behave like dilute particle suspension. The increments of filler loading until it passes the critical loading fraction will affect polymer-filler interactions. During the interactions, absorbed polymer layer is formed on the filler surface while interactions between fillers would influence viscosity of polymer composites. The viscosity continues to