

**EFFECT OF CONSOLIDATION PRESSURE ON
PHYSICAL AND MECHANICAL PROPERTIES
OF UNSATURATED POLYESTER RESIN-
COCONUT SHELL-TITANIUM DIOXIDE
COMPOSITE**

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UNIVERSITI

**FACULTY OF EARTH SCIENCE
UNIVERSITI MALAYSIA KELANTAN**

2017

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by

SITI NOOR ASMA BINTI MOHAMAD ZANI

A thesis submitted in fulfillment of the requirements for the degree of
Bachelor of Applied Science (Materials Technology) with Honours

**FACULTY OF EARTH SCIENCE
UNIVERSITI MALAYSIA KELANTAN**

2017

DECLARATION

I declare that thesis entitled “Effect of Consolidation Pressure on Physical and Mechanical Properties of Unsaturated Polyester Resin-Coconut Shell-Titanium Dioxide Composite” is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature : _____

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Date : _____

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Effect of Consolidation Pressure on Physical and Mechanical Properties of Unsaturated Polyester Resin-Coconut Shell-Titanium Dioxide Composite

ABSTRACT

This study investigates the effect of different content polyester (UPR), coconut shell (CS) and titanium dioxide (TiO_2) and compaction pressure in UPR-CS- TiO_2 composite. The composite mixture with different composition was mixed and then cold compressed with 200, 300, 400 and 500 MPa. The result showed that the composite with 40 wt% of CS fiber with no addition of TiO_2 has good flexural properties due to the ability of reinforcement (CS) support the loading on composite and the lighter compared to the other composites. UPR-CS- TiO_2 composite density increased with TiO_2 content due to higher agglomeration and void content between CS fiber and TiO_2 particles. The composite with 20 wt% CS and TiO_2 exhibit thickness swelling, water uptake and moisture content reduction. In the most composite composition modulus of rupture (MOR) and modulus of elasticity (MOE) increased with compaction pressure while thickness swelling, water absorption and moisture content decreased.

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Kesan Tekanan Konsolidasi keatas Sifat-Sifat Fizikal dan Mekanikal Resin Poliester Tak Tepu-Tempurung Kelapa-Titanium Dioksida Komposit

ABSTRAK

Kajian ini adalah untuk mengkaji kesan perbezaan kandungan poliester (UPR), tempurung kelapa (CS) dan titanium dioksida (TiO_2) dan tekanan pemadatan dalam UPR-CS- TiO_2 komposit. Campuran komposit dengan komposisi yang berbeza telah dicampur dan telah dimampat sejuk pada 200, 300, 400 dan 500 MPa. Hasil menunjukkan komposit dengan komposisi 40 wt% CS tanpa penambahan TiO_2 mempunyai sifat kelenturan yang baik berdasarkan keupayaan pengukuhan menampung bebanan ke atas komposit dan menunjukkan ketumpatan yang paling ringan berbanding dengan komposit lain. Ketumpatan komposit UPR-CS- TiO_2 meningkat dengan kandungan TiO_2 disebabkan pergumpalan dan rongga udara yang tinggi antara gentian CS dan zarah TiO_2 . Komposit dengan 20 wt% CS dan TiO_2 mempamerkan penurunan ketebalan pembengkakan, penyerapan air dan kandungan. Dalam kebanyakan komposisi komposit, modulus patah (MOR) dan modulus kekenyalan (MOE) meningkat dengan tekanan pemadatan manakala ketebalan pembengkakan, penyerapan air dan kandungan lembapan menurun.

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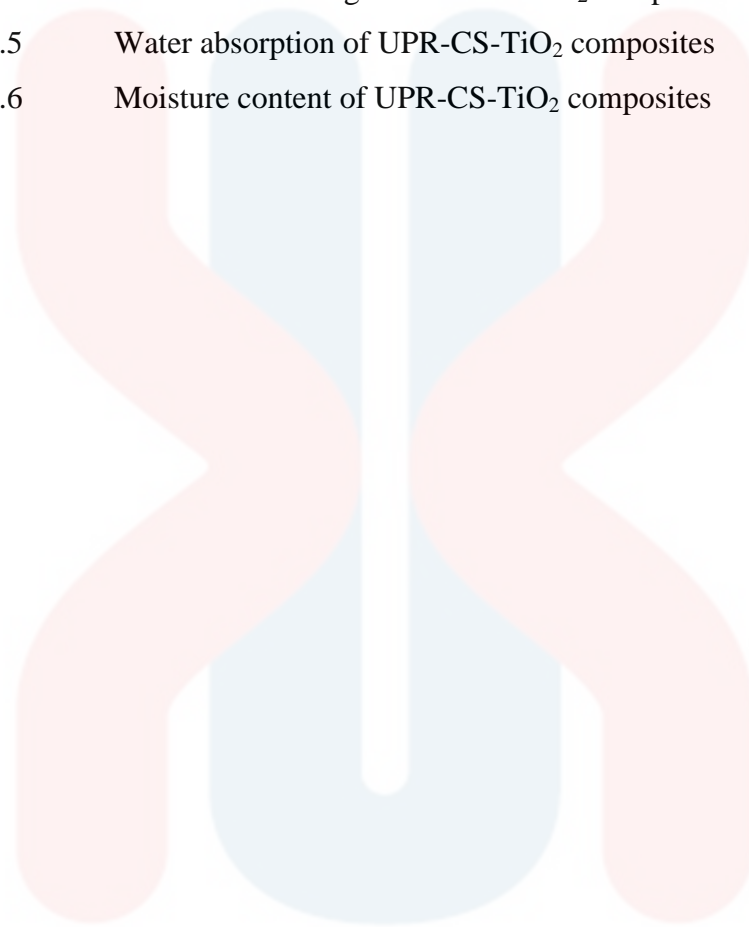
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LISTS OF ABBREVIATIONS

ABBREVIATION	DESCRIPTION
ASTM	American Society for Testing and Materials
ATH	Aluminium Trihydroxide
BMC	Bulk Molding Compound
CMCs	Ceramic Matrix Composites
CS	Coconut Shell
DBTDL	Dibutyltin Dilaurate
FESEM	Field Emission Scanning Electron Microscopy
IEM	Isocyanatoethyl Methacrylate
LMC	Liquid Molding Compound
MEKP	Methyl Ethyl Ketone Peroxide
MMCs	Metal Matrix Composites
MOE	Modulus of Elasticity
MOR	Modulus of Rupture
MWCNT	Multiwalled Carbon Nanotubes
NFPCs	Natural Fiber Reinforced Polymer Composites
OPS	Oil Palm Shell
PMCs	Polymer Matrix Composites
PE	Polyethylene
PEEK	Polyetheretherketone
PES	Polyethersulfone
PI	Polyimide
PP	Polypropylene
PVC	Polyvinyl chloride
SEM	Scanning Electron Microscopy
SMC	Sheet Molding Compound
THF	Tetrahydrofuran
TiO ₂	Titanium Dioxide
UPR	Unsaturated Polyester Resin
UV	Ultraviolet

LISTS OF SYMBOLS

SYMBOL	DESCRIPTION
%	Percentage
>	Greater than
°C	Degree Celsius
g	Gram
MPa	Mega pascal
M_0	Initial weight of sample before heated in an oven
M_1	Final weight of sample after heated in an oven.
m_0	Mass of composite in the air
m_1	Mass of composite in the water
mm	Millimeter
nm	Nanometer
ρ	Density
ρ_w	Density of water
T_0	Initial thickness of sample before immersion in distilled water
T_1	Final thickness of sample after immersion in distilled water
μm	Micrometer
W_0	Initial weight of sample before immersion in distilled water
W_1	Final weight of sample after immersion in distilled water
wt%	Weight percentage

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Composite can be defined as the material which consists of two or more distinct materials on macro scale with different properties to form a new material with a different property (Gupta, 2014). The primary phase of a composite material is called a matrix and the secondary phase is called reinforced material (Gupta, 2014). Matrix acts as a binder that holds the reinforcing material together by surface connection while the reinforced material is able to support high tensile loads although the matrix imparts rigidity to the composite (Al-bahadly, 2013). Composite is divided into three types which depend on the matrix used such as metal matrix composites (MMCs), polymer matrix composites (PMCs) and ceramic matrix composites (CMCs). The reinforced material can be either synthetic or natural fibers (Gupta, 2014; Chung, 2010; Callister & Rethwisch, 2010).

Unsaturated polyester resin (UPR) is one of the most important classes of thermosetting polymer. Thermoset is the plastic that cannot be melted once cured because of the presence of crosslink structure (Al-bahadly, 2013) due to the completion of polymerization process (Chung, 2010). UPR is a very famous material that is used as a matrix for fiber reinforced plastic (Smith & Hashemi, 2010) besides epoxy resin. Although UPR is not as strong as epoxy resin, it is very low in cost compared to epoxy resin (Smith & Hashemi, 2010) and it also has the capability to produce high strength composite due to their high strength and high modulus properties. In addition, the other UPR properties are the capability to cure at room

temperature, high resistance to water, transparency (Kargarzadeh *et al.*, 2014) and good corrosion and chemical resistance (Abdullah *et al.*, 2015). Other than can be apply as matrix in composite industries, UPR also has been used in variety of fields such as marine, aerospace (Kargarzadeh *et al.*, 2014; Smith & Hashemi, 2010) automobile, building panel and electronic equipment (Abdullah *et al.*, 2015; Smith & Hashemi, 2010).

There are many natural fibers that are already been found as potential alternatives to steel, asbestos, glass and polymer fibers such as sisal, jute, cotton, flax, hemp and kenaf. This is because of their good reinforcing effect, environmental friendliness and ready in fibrous form. They also can be extracted from plant leaves at low cost (Wei & Meyer, 2016). Natural fiber which consists of large amount of cellulose (Foruzanmehr *et al.*, 2015; Dai & Fan, 2014) content is known for composite properties enhancement. Cellulose based natural fiber such as coconut shell (CS) can be used as reinforced material in polymer composite since it mostly composed cellulose. According to Soltani *et al.* (2014), the higher cellulose content the better mechanical properties.

CS is one of the parts of coconut tree (*Cocos nucifera*) that extracted from coconut fruit. *Cocos nucifera* is a *Arecaceae* family (palm family) that wide spread in the tropics (Latinwo & Agarry, 2015; Adedayo *et al.*, 2012; Okafor *et al.*, 2012). *Cocos nucifera* is commonly grow on tropical beaches and this species has been cultivated for 400 years (Adedayo *et al.*, 2012). According to Adedayo *et al.* (2012), the *Cocos nucifera* comes from the south-east Asian Peninsula (probably Malaysia) and mostly can be found in pacific, Indonesia, Philippines, South Asia, East and West Africa and the Caribbean. Based on the past research, the CS widely use as

activated carbon which is use as absorbent in water purification (Babel & Kurniawan, 2004; Raj & Joy, 2015) due to its high surface area and porosity (Raj & Joy, 2015). Based on the research by Bledzki *et al.* (2010), CS is used as reinforcement in composite resulting in high thermally stable, better at elongation break and better Charpy impact strength compared to soft wood composite.

The way to improve composite properties such as improve tensile strength, provide resistance to abrasion, increased the toughness and thermal stability is by adding the filler (Callister & Rethwisch, 2010). Filler is divides into two categories which are organic and inorganic fillers. Walnut shell and corn cob residue are example for organic filler while for inorganic filler are alumina, kaolin and aluminium hydroxide (Yang & Frazier, 2016).

According to Wang *et al.* (2016), TiO₂ is known as transparent metal oxide semiconductor which is the one type of ceramic group. TiO₂ is widely used as photocatalysts because of its wide band gap (Salim *et al.*, 2015; Wang *et al.*, 2016) and having a great lifetime of electrons and photogenerated holes (Salim *et al.*, 2015). Other than that, TiO₂ also has been used in make-up, biomedical devices (Wang *et al.*, 2016), solar cell (Maria *et al.*, 2015; Nguyen *et al.*, 2013), energy storage device (Maria *et al.*, 2015), self-cleaning coating (Nguyen *et al.*, 2013; Maria *et al.*, 2015) and odor inhibition (Nguyen *et al.*, 2013). These widely applications of TiO₂ are because of their superior properties such as high chemical stability (Maria *et al.*, 2015; Salim *et al.*, 2015), high thermal stability (Salim *et al.*, 2015), good delivery ability (Deivanayaki *et al.*, 2013), low density, high surface area (Deivanayaki *et al.*, 2013), non-toxicity (Wang *et al.*, 2016; Salim *et al.*, 2015; Luo

et al., 2014; Zhao *et al.*, 2016) and low cost (Salim *et al.*, 2015; Deivanayaki *et al.*, 2013; Luo *et al.*, 2014).

In this study, the composite will be produced from unsaturated polyester resin (UPR)-coconut shell (CS) and TiO₂ as a filler using compression molding method. The physical and mechanical properties of the composites also will be determined.

1.2 Problem Statement

CS consist of carbohydrate components such as hemicellulose, cellulose and lignin whereas the composition (%) of hemicellulose, cellulose and lignin are 21%, 34% and 27%, respectively (Sarki *et al.*, 2011; Bledzki *et al.*, 2010). The high cellulose content in CS makes it possible as reinforced material for improvement of properties of unsaturated polyester-based composite. Moreover, CS is widely available, cheap and environmental friendly. However, there are not much study has been discussed on physical and mechanical properties of CS in unsaturated polyester resin-based composite. Besides that, the addition of TiO₂ as filler in unsaturated polyester resin-CS composite should be evaluated in order to determine the compatibility of reinforced materials in composite.

1.2 Objectives

The objectives of this research are:-

- i. To determine the effect of consolidation pressure on physical and mechanical properties of unsaturated polyester resin (UPR)-CS-TiO₂ composite.
- ii. To investigate the different compositions of CS particles and TiO₂ in unsaturated polyester resin (UPR)-based composite prepared by compression molding.

1.3 Expected Outcome

This study deals with preparation and characterization of physical and mechanical properties of unsaturated polyester resin (UPR)-CS-TiO₂ composite using compression molding. The variation on consolidation pressure will affect the physical and mechanical properties of composite. The potential of CS and TiO₂ also can be measured by evaluating its different compositions in composite.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter was explained in detail about composite materials includes the phases that involved in composite and classification of composite. Besides that, this chapter also discussed the properties and the application of unsaturated polyester resin (UPR)-based composite and natural fiber. The types of filler and fabrication of composites also will be discussed in this chapter.

2.2 Composite Materials

Composite materials was produced by combination of the best properties of the constituent phases which are matrix phases and reinforcement phases (Callister & Rethwisch, 2010). It is can be comprised with two or more components and form naturally by casting process (Chung, 2010). Moreover, composite materials can be classified according to types of material that are used as matrix phase such as metal, polymer and ceramic. Other than that, it also can be classify based on the shape of filler which are particulate, fiber or layer formed (Chung, 2010; Thomas *et al.*, 2012). This is due to their properties that can be tailored for vary of applications such as aerospace, automobile, electronic, construction, energy and biomedical (Chung, 2010). The properties of tailored composite depend on the choosing on their distribution, component, proportion, morphologies, degree of crystallinity and the composition of the interface between components as well as the structure (Chung, 2010).

2.2.1 Matrix Phase

The matrix phase is a phase that having a continuous character and usually it is more ductile and less hard phase (Thomas *et al.*, 2012). It also can be defined as material that give structure (body) to the composites (Al-bahadly, 2013). The matrix phase is important to hold the reinforcement material together by surface connection. Compared to reinforcement material, matrix is low in strength. However, it is capable to provide resistance to crack propagation (Al-bahadly, 2013). It is also able to transmits applied loads to the reinforcing fiber within the composite and maintain the good orientation and position of reinforcing fiber. As a result, the composite with better mechanical properties will be produced (Al-bahadly, 2013). Among three types of matrix materials that have been used, polymer-based is the most paid attention because its cheapness, flexible and can be reinforced with wide variety of materials. Generally in polymer-based composite, the matrix material is divided into two types which are thermoplastic and thermoset.

Thermoset can be define as a material which initially in a liquid state and will be transform into solid state during the curing process by apply heat or catalyst (Al-bahadly, 2013). Thermoset harden due to polymerization process which is the high crosslink or network structures having covalent bonds with all molecules will be produced (Thomas *et al.*, 2012; Al-bahadly, 2013; Chung, 2010). They cannot be melted and reshape once become a solid state, this prevent the chain from sliding which is resulting in high modulus and increase the creep resistance (Thomas *et al.*, 2012; Al-bahadly, 2013). However, it is more brittle than thermoplastic usually. The most common thermoset resin that has been used as matrix material in composite is polyester, vinyl ester and epoxy. Among of this three, the epoxy is the most popular

due to its high Young's modulus. This can be shown in Figure 2.1 of typical stress-strain curves in which epoxy has the highest strength compared to that of polyester and vinyl ester. However, polyester is the most of choice in industries due to its cost effectiveness and good efficiency compared to other types of resin.

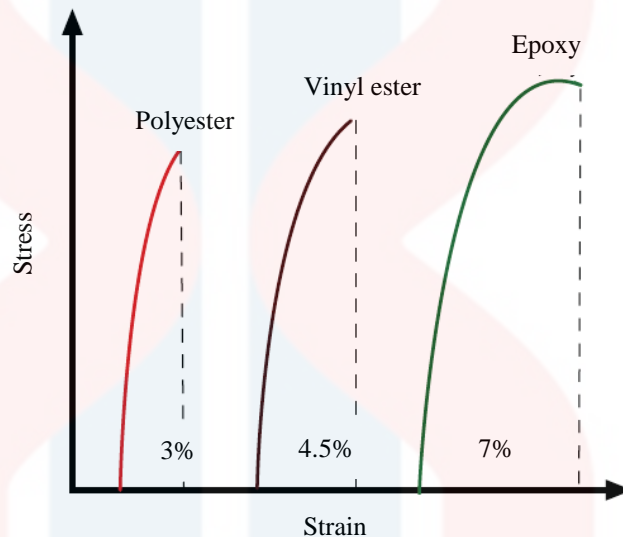


Figure 2.1: Typical resin stress/strain curve (Al-bahadly, 2013).

Meanwhile, thermoplastic is defined as a material that can be melted, reshaped and recycled repeatedly although after cured (Al-bahadly, 2013). Thermoplastic resin is more ductile compared to that of thermoset resin and it is also faster in processing than thermoset resin (Chung, 2010). According to Thomas *et al.* (2012), the structure of thermoplastic can be linear or branched which this structure is bonding with strong intramolecular bonds but weak intermolecular bonds and its structure can be either amorphous or crystalline. The examples of thermoplastic resin are nylon, polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC).

2.2.2 Reinforcement Phase

Reinforcement phase also called dispersed phase (Thomas *et al.*, 2012; Callister & Rethwisch, 2010) is more stronger than matrix phase. This phase is embedded in the matrix in a discontinuous form and it provides good mechanical of composite due to ability support high tensile loads (Al-bahadly, 2013). Reinforcement phase also called the fiber phase because commonly the material that use as reinforcement is in the fiber form (Callister & Rethwisch, 2010). It is also divided into two types which are synthetic fiber (glass and carbon) and natural fiber (extracted from animal, mineral and plant). The dispersed phase affected the properties of composites which are depends on their amount and geometry such as concentration, shape, orientation, size and distribution (Callister & Rethwisch, 2010).

The reinforcement phase can be classified into three which are whiskers, wires and fibers (Callister & Rethwisch, 2010). These classifications are based on their shape that has been produced. Whiskers are the one example of strongest materials that have very high crystalline degree and large length-to-diameter ratio due to their form that very thin single crystal (Callister & Rethwisch, 2010). Although whiskers provided very high in strength, they is not widely uses as reinforcement material because they are extremely expensive. Examples of whiskers materials are silicon nitride, silicon carbide and graphite. Another examples of reinforcement phase is wires, wires have relatively large diameter which are mostly used in wire-wound high-pressure, in filament-wound rocket casing and in automobile tires as a radial free steel reinforcement (Callister & Rethwisch, 2010). Steel, tungsten and molybdenum are examples for wires materials.

While, for fibers materials can be classified into two either amorphous or polycrystalline (Callister & Rethwisch, 2010). Fibers have small diameter and it also can be called as fibrous materials which are can be either ceramic or polymer. Glass, carbon, boron, aluminium oxide and silicon carbide are examples of ceramic types while example for polymer type is polymer aramids (Callister & Rethwisch, 2010). Fibers can be divided into two types which are continuous fibers (long fiber) and discontinuous fibers (short fiber) (Figure 2.2). These two types of fibers are measured by aspect ratio (length to the diameter ratio) which are $l/d \geq 100$ was identified as long fibers while $l/d < 100$ was short fibers.

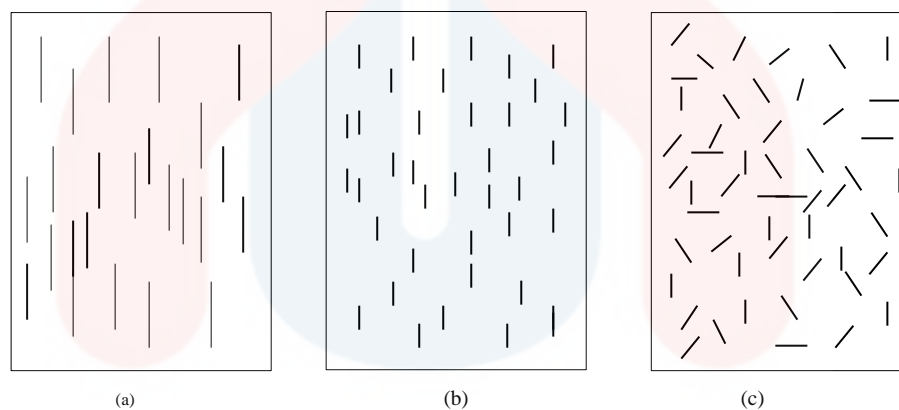


Figure 2.2: (a) Continuous fibers and aligned (b) Discontinuous fibers and aligned (c) Discontinuous fibers and randomly orientated

2.2.3 Classification of Composites

Classification of composite is according to the matrix material used such as metal, polymer and ceramic and it also can be classify based on shape of reinforcement materials which are particles, fibers and laminates (Figure 2.3) (Thomas *et al.*, 2012).

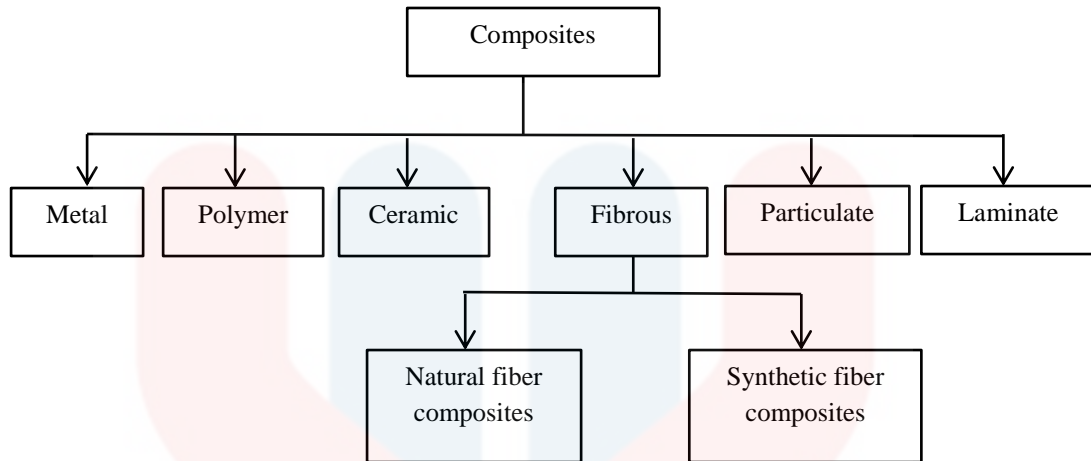


Figure 2.3: Classification of composites (Thomas *et al.*, 2012).

MMCs is more expensive than PMCs because it processing is required high temperature (Chung, 2010; Callister & Rethwisch, 2010). There are two steps in processing of MMCs which are synthesis and shaping process (Callister & Rethwisch, 2010). The shaping process is followed the metal forming process such as forging, rolling and extrusion. Mostly, the aluminium is used as the matrix for lightweight structures and low thermal expansion. It is also can increase the strength and modulus. Other than that, it can reduce the coefficient of thermal expansion. MMCs are widely used in automobile manufacture in engine component due to it lightweight, wear resistance and thermal distortion. It also used in aerospace industry (Callister & Rethwisch, 2010). However, their application is limited due to their high cost (Chung, 2010).

PMCs are the most affordable cost for fabrication process compared to that of MMCs and CMCs due to the low processing temperature (Chung, 2010; Thomas *et al.*, 2012). The thermoplastic and thermoset is famously used as matrix materials in PMCs. Thermoplastic as a matrix is used due to it greater ductility properties and

more faster than thermoset in term of processing. Polyimide (PI), polyethersulfone (PES) and polyetheretherketone (PEEK) are examples for thermoplastic. Epoxy resin, phenolic resin, vinyl ester and polyester are examples of thermoset. Thermoset is harden gradually in presence of heat and pressure due to it polymerization process. It cannot be melted once cured because of their structure that high crosslink (Thomas *et al.*, 2012). It is used for lightweight structures such as wheel chair, aircraft, automotive and sporting good (Chung, 2010).

CMCs can be produced using hot pressing, hot isostatic pressing or liquid-phase sintering technique (Callister & Rethwisch, 2010). It is commonly used because of good resistance to oxidation, high-temperature creep behavior and resistance to thermal shock. This make these composite is attractive for high temperature applications such as aerospace and automotive which is used for engine component (Chung, 2010; Callister & Rethwisch, 2010).

2.3 Unsaturated Polyester Resin-Based Composites

There are two types of matrix which are thermoset and thermoplastic. The matrix is not only acts as a binder that holds the fibers in the desired position thereby transferring the external load to reinforcement (Gupta, 2014) but it also can reduce the crack propagation and damage in the composite (Al-bahadly, 2013). Unsaturated polyester resin (UPR) is one type of thermoset polymer and it is low cost compared to other the thermoset polymer like epoxy and vinyl ester (Abdul Khalil *et al.*, 2011; Maheshwari *et al.*, 2015; Sahari & Maleque, 2016; Barbero, 2011). Moreover, UPR is popularly used in aerospace and marine industries because of their special properties such as transparency (Sahari & Maleque, 2016; Kargarzadeh *et al.*, 2014;

Barbero, 2011) high strength and modulus, good water resistance (Kargarzadeh *et al.*, 2014) and have capability to cured at room temperature (Kargarzadeh *et al.*, 2014; Sahari & Maleque, 2016). According to Abdullah *et al.* (2015), UPR also has been applied as matrix in electronic equipment, container, automotive and culture marble due to excellent corrosion resistance, good chemical resistance and good mechanical properties (Sahari & Maleque, 2016).

Besides that, UPR also has its own disadvantages such as low toughness limit its usage (Kargarzadeh *et al.*, 2014), very sensitive with fire, easily degrade when expose to UV radiation and moisture diffusivity (Maheshwari *et al.*, 2015). These limitations were affected the mechanical and physical properties of products or materials such as low in strength. However, these limitation can be avoid by adding the others materials such as filler (Maheshwari *et al.*, 2015). For example, the aluminium trihydroxide (ATH) was added in UPR to improve the fire retardant and carbon black was added to improve UV resistant (Maheshwari *et al.*, 2015). Unsaturated polyester resin (UPR)-based composite can be reinforced by synthetic and natural fibers. UPR reinforced natural fiber composite become increasingly popular as it offers low cost, ease of processing and environmental friendly.

Most of the journals were discussed about the effect of chemical treatments on mechanical and physical properties of composite. According to Liu *et al.* (2014), chemical treatment was made in UPR-based composites reinforced with bamboo fiber by treated the bamboo fiber with isocyanatoethyl methacrylate (IEM) in the presence of dibutyltin dilaurate (DBTDL) as a catalyst resulting in high tensile strength, impact strength and flexural strength by comparing the untreated bamboo fiber (reference) with treated bamboo fiber. Figure 2.4 shows the SEM images of tensile-fracture surface for untreated and treated bamboo fiber with IEM. In addition,

the composite that was produced become water resistant due to lower in water absorption and these also improved the adhesion between reinforcement (bamboo) and matrix (UPR). Based on the journal of characterization of jute and glass fiber reinforced polyester based hybrid composite by Hassan *et al.* (2016), the mechanical properties of composite is increased after the glass fiber was added in jute fiber with 3 layers of glass fiber and 2 layers of jute fiber which are increased in tensile strength and flexural strength. Then, water absorption was decreased after the glass fiber added. The increasing in mechanical properties and decreasing in water absorption of hybrid composite after added glass fiber are shown in Table 2.1.

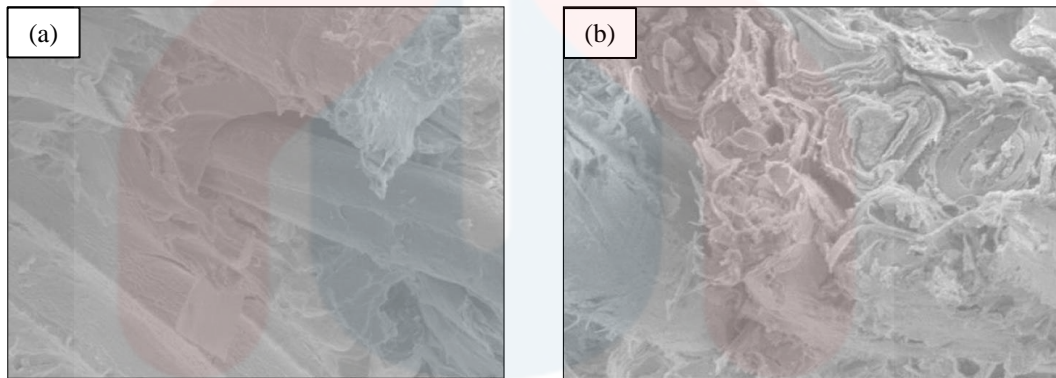


Figure 2.4: SEM image of tensile-fracture surface for (a) untreated and (b) treated bamboo fiber with 3 wt% of IEM (Liu *et al.*, 2014)

Table 2.1: Tensile strength, flexural strength and water absorption of composite (G: Glass fiber, J: Jute fiber)

Sample name	Fiber orientations	Tensile strength (MPa)	Flexural strength (MPa)	Water Absorption (%)
C1	JJJJ	38.6875	54.71	7.61
C2	GJGJG	104.625	134.65	4.84

The mechanical and physical properties of UPR-based oil palm shell composite were discussed by Sahari & Maleque (2016) show that the adding of oil palm shell

(OPS) to UPR matrix was improved these both properties which are the 30 vol% of OPS used show high Young's modulus and tensile strength compared to the 0, 10, 20 vol% of OPS. Besides that, the density of composite is decreased due to increasing in OPS contents which are suitable for automotive and construction components. According to Yunus *et al.* (2015), the adhesion between multiwalled carbon nanotubes (MWCNTs) and UPR is improved by using tetrahydrofuran solvent (THF) resulting in well dispersed as shown in Figure 2.5. At the same time, the THF-MWCNTs-UPR produced a composite with superior mechanical properties such as high tensile strength due to good compatibility between MWCNTs and UPR.

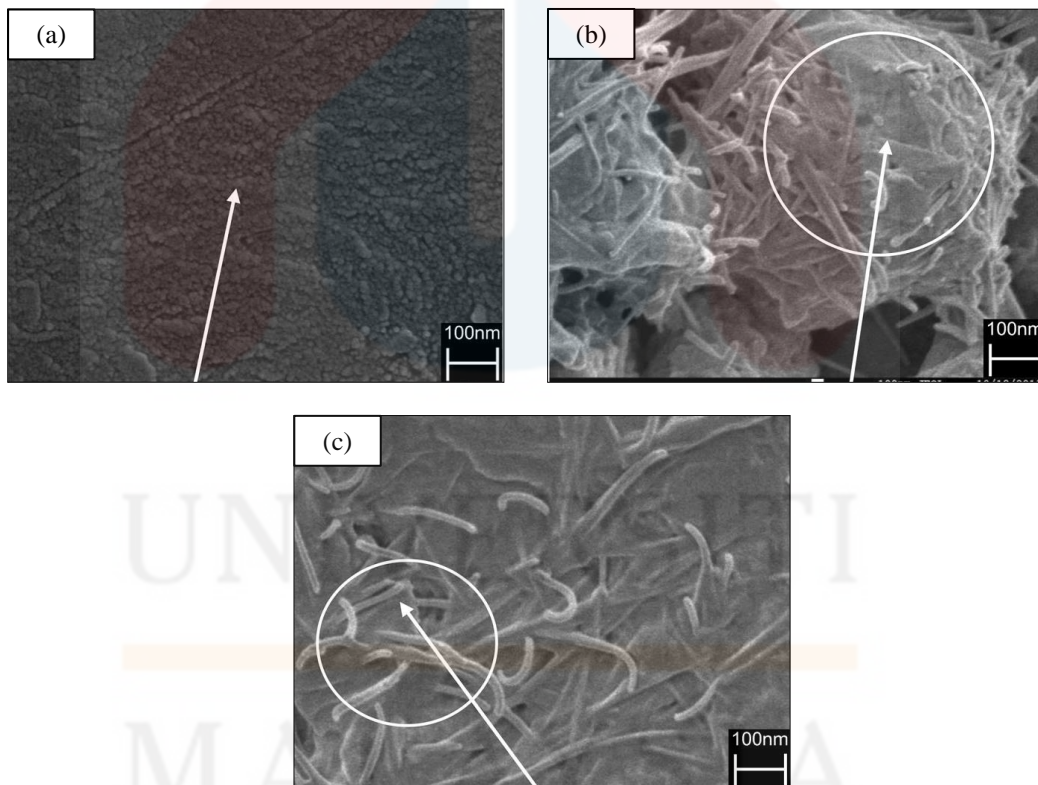


Figure 2.5: FESEM micrographs of non-fracture surface of (a) UPR (smooth surface) (b) MWCNT-UPR (agglomerated MWCNTs) and (c) THF-MWCNT-UPR (well dispersed MWCNT) (Yunus *et al.*, 2015)

2.4 Natural Fiber

Natural fiber reinforced polymer composites (NFPCs) are brought into the competitive market because of wide range of advantages over synthetic fiber based composites such as biodegradable (Faruk *et al.*, 2013; Wei & Meyer, 2016; Soltani *et al.*, 2014; Kale *et al.*, 2016; Boccarusso *et al.*, 2016; Sarki *et al.*, 2011; Al-bahadly, 2013; Thomas *et al.*, 2012; Jayamani *et al.*, 2014; Mysamy & Rajendran, 2011; Sahari & Maleque, 2016), high strength to weight ratio (Gupta, 2014; Jayamani *et al.*, 2014; Kale *et al.*, 2016), lightweight (Gupta, 2014; Mysamy & Rajendran, 2011), high strength at elevated temperatures (Gupta, 2014), high creep resistances (Gupta, 2014) and high toughness (Gupta, 2014; Sarki *et al.*, 2011). Natural fiber is a fiber that was extracted from the natural based material. Natural fiber can be classified into three large groups: animal, vegetable and mineral natural fiber (Soltani *et al.*, 2014) as shown in Figure 2.6.

Natural fiber appear very intriguing for their high specific strength (Foruzanmehr *et al.*, 2015), compostability (Foruzanmehr *et al.*, 2015) and their low cost (Foruzanmehr *et al.*, 2015; Dai & Fan, 2014; Faruk *et al.*, 2013; Kale *et al.*, 2016; Boccarusso *et al.*, 2016; Sarki *et al.*, 2011; Jayamani *et al.*, 2014; Gu *et al.*, 2014; Mysamy & Rajendran, 2011). This make these fiber is chosen to use as reinforced material in structural and semi-structural (Foruzanmehr *et al.*, 2015) as well as automotive industries (Foruzanmehr *et al.*, 2015). Natural fibers are also increasingly being used in bio-derived and bio-inspired materials (Foruzanmehr *et al.*, 2015). Moreover, according to Faruk *et al.* (2013), natural fiber has higher demand especially in automotive, building and construction industries.

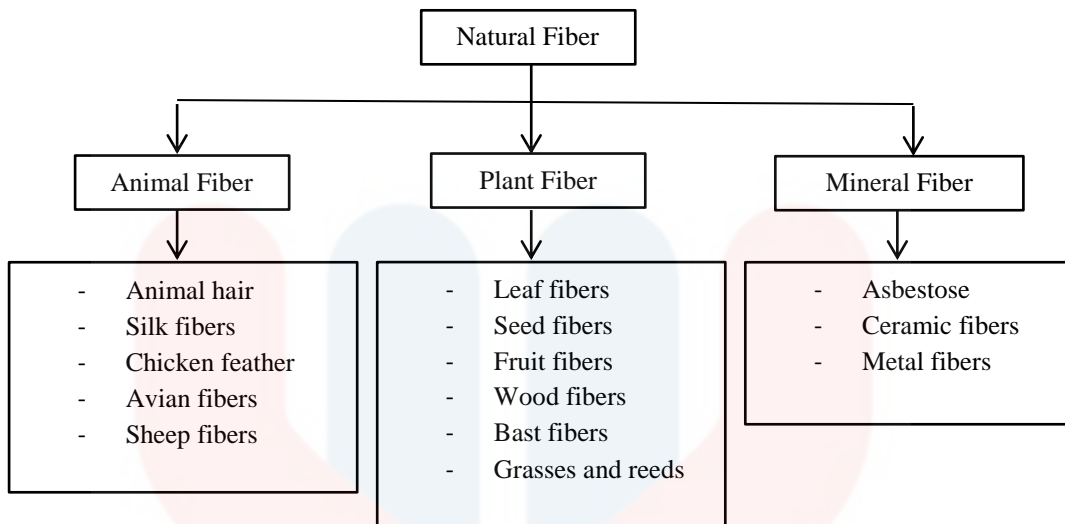


Figure 2.6: Classification of natural fibers based on source of origin (Gupta, 2014; Sparniš, 2006)

Natural fiber consist of three major components which are cellulose, hemicellulose and lignin (Soltani *et al.*, 2014; Wei & Meyer, 2016; Summerscales & Grove, 2014). Cellulose is a homopolymer of glucose (Soltani *et al.*, 2014) whereas subunits linked by β -1,4 bonds and the basic repeating unit is cellobiose (Wei & Meyer, 2016). Cellulose is a semi-crystalline polysaccharide and is responsible for the hydrophilic nature of natural fibers (Soltani *et al.*, 2014). While hemicellulose is a heteropolymer consisting of hexose (glucose, mannose and galactose) and pentose sugars (xylose and arabinose) (Soltani *et al.*, 2014) which is the main chain of hemicellulose is characterized by a β -1,4-linked-D-xylopyranosyl, which carries a variable number of neutral or uronic monosaccharide substituents (Wei & Meyer, 2016).

Besides that, hemicellulose is a fully amorphous polysaccharide with a lower molecular weight compared to cellulose (Soltani *et al.*, 2014). The amorphous nature of hemicelluloses comes from the fact that it is partially soluble in water and alkaline solutions (Soltani *et al.*, 2014). Then, lignin is consists of the β -O-4 ether linkages,

followed by other types of ether and C\C linkages such as α -O-4, β - β , β -5 and 5-5 (Wei & Meyer, 2016) which provides rigidity to the structure of the plant (Soltani *et al.*, 2014).

2.5 Coconut Shell as Reinforced Materials in Composites

CS is one example of plant fiber that extracted from the coconut tree which is it is classified as fruit fiber. The SEM image of CS and three layers of coconut fruit which are endosperm, endocarp and mesocarp (Agunsoye *et al.*, 2015; Sarki *et al.*, 2011) shown in Figure 2.7. The endocarp is a scientific name for the CS and this layer is called as hard lignocellulose composite. The common name for mesocarp is coir with their ductile properties because of spongy structure (Agunsoye *et al.*, 2015). While, the endosperm is a thick albuminous testa that is adhering to the inside wall of the endocarp (Sarki *et al.*, 2011). The CS can be used as a filler or reinforce material in composite based on their good thermal properties and CS is a brittle material that easily to form into powders, chips or particles (Sarki *et al.*, 2011). The properties of CS is depend on the content of chemical composition that present such as 21% of hemicellulose, 34% of cellulose and 27% of lignin (Bledzki *et al.*, 2010).

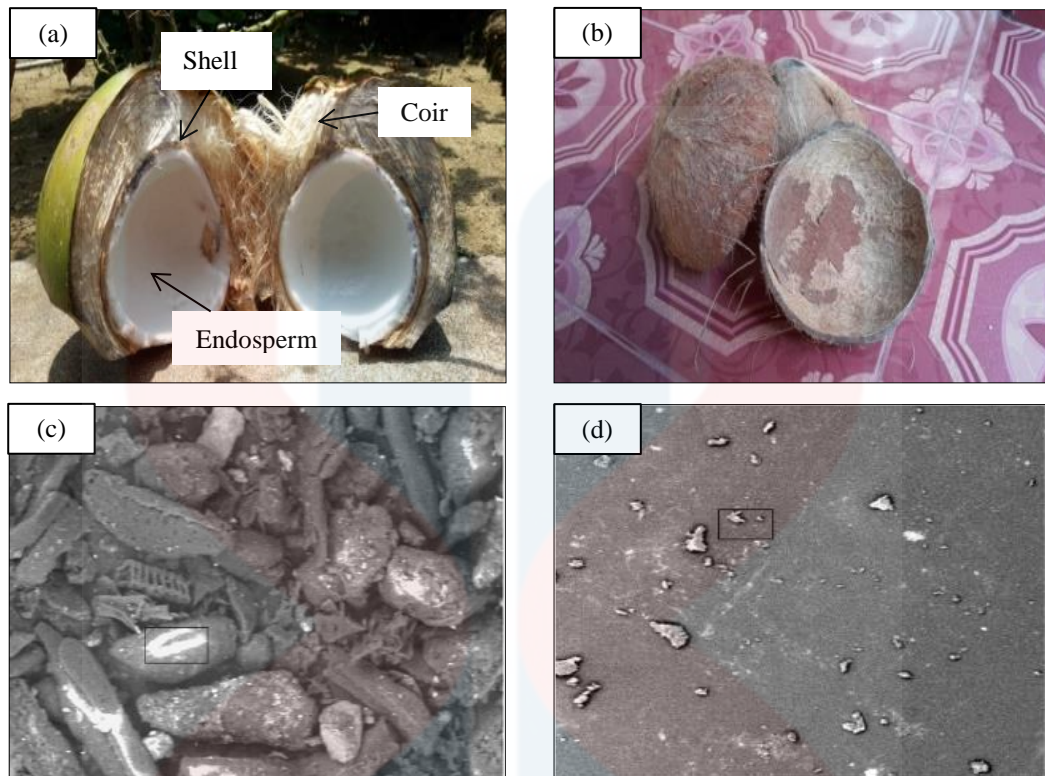


Figure 2.7: (a) Coconut (b) Coconut shells (c) SEM microstructure of the coconut shell particles (Sarki *et al.*, 2011) (d) SEM microstructure of the coconut shell fillers reinforced epoxy (Sarki *et al.*, 2011).

According to Sarki *et al.* (2011), CS is use as filler in biodegradable polymer composite due to good thermal stability. Moreover, the mechanical properties of CS particles reinforced epoxy composites improved Young modulus and ultimate tensile strength due to the addition of percentage of CS. Based on the study of activated carbon by Jain & Tripathi (2013), the CS is used as electrode material due to their mesoporous structure which is provide large surface area, high conductivity and well connected after carbonization process. In addition, the mesoporous structure of CS can be suitable use as filler because of the compatibility with matrix whereas the CS particles will be well dispersed or mixed between the matrix materials. This will be improve the matrix properties such as improve the strength and adhesion of the matrix.

2.6 Filler

Filler is added to composite to reduce cost of consumption of reinforcement. Addition of filler may alter composite physical and mechanical properties. Filler is often used in polymer to improve the tensile strength, toughness, thermal stability and provide abrasion resistance (Callister & Rethwisch, 2010). Filler can be divided in two types that are organic and inorganic. Organic filler is derived from lignocellulosic biomass waste streams such as walnut nutshell (*Juglans regia*), red alder (*Alnus rubra*) bark and furfural production residues such as corn cob residue and the walnut shell was reported that have a potential in increasing the adhesive performance (Yang & Frazier, 2016). Inorganic filler is produced from silicon derivatives and consist of particles such as glass, quartz, pyrogenic silicon dioxide and colloidal silica via a sol gel process (Salazar, 2011). Examples of inorganic filler are alumina, kaolin and aluminium hydroxide (Yang & Frazier, 2016).

According to Salazar (2011), the mechanical performance of the composite is depend on the filler content and particles size. In addition, it has reported that, reducing the filler particles size was increased the rate of wear, increase tensile strength, increase compressive strength, resistance to fracture and was reduced the shrinkage of polymerization. This can be conclude that the smaller the size of the particles, the better mechanical performance.

2.6.1 Titanium Dioxide

Titanium dioxide (TiO_2) also known as Titanium (IV) Oxide or Titania, it is very famous material that has been applied in many application because of their

properties such as high photocatalytic activity (Salim *et al.*, 2015; Wang *et al.*, 2016; Luo *et al.*, 2014), non-toxicity (Wang *et al.*, 2016; Luo *et al.*, 2014; Zhao *et al.*, 2016), chemical stability (Foruzanmehr *et al.*, 2015; Salim *et al.*, 2015), low density, high specific surface area and superior delivering ability (Deivanayaki *et al.*, 2013). TiO₂ widely used in many application not only because of their excellent properties but also because of their cost that very affordable (Salim *et al.*, 2015; Deivanayaki *et al.*, 2013; Luo *et al.*, 2014) and can be provide in higher quantities in a time (Wang *et al.*, 2016).

According to Zhao *et al.* (2016), TiO₂ have potential to become a shell for microcapsules due to its excellent properties especially non-toxicity, chemical inert, high thermal conductivity and good mechanical properties. TiO₂ also has been apply in composite material in order to improve the strength of the interface between the fibers and the polymeric matrices (Foruzanmehr *et al.*, 2015). In addition, TiO₂ is material used in photocatalysts application (Wang *et al.*, 2016; Maria *et al.*, 2015; Nguyen *et al.*, 2013) due to its excellent photocatalytic activity. Besides that, it is also has been used in other application such as cosmetics, gas sensors (Wang *et al.*, 2016), self-cleaning coating (Maria *et al.*, 2015; Nguyen *et al.*, 2013), antibacterial coating on medical device (Wang *et al.*, 2016; Maria *et al.*, 2015; Nguyen *et al.*, 2013), energy storage device (Maria *et al.*, 2015), dye-sensitized solar cells (Maria *et al.*, 2015) and photoelectric conversion in solar cell (Nguyen *et al.*, 2013).

According to Foruzanmehr *et al.* (2015) and Wang *et al.* (2016), TiO₂ can be found in three forms such as rutile, anatase and brookite. Rutile is the most thermodynamically stable of TiO₂ phase (Foruzanmehr *et al.*, 2015) while anatase

and brookite will be transform into thermodynamic stable phase of rutile at high temperature this is the reason it called as metastable phases (Wang *et al.*, 2016).

2.7 Fabrication of Composites

2.7.1 Compression Molding

Compression molding is a method for fabrication of thermoplastic and thermoset polymer (Callister & Rethwisch, 2010). However, this method is commonly used for thermoset material (Al-bahadly, 2013). There are three types of compression molding which are sheet molding compound (SMC), bulk molding compound (BMC) and liquid molding compound (LMC). This process is involved two heated mold die that will compress the material using vertical press such as cold press and hot press method, but only one mold is moveable during the process (Al-bahadly, 2013). Figure 2.8 is shown the schematic diagram of a compression molding. The material is placed between two heated mold and the pressure is applied, the material was flow to conform to the mold shape (Callister & Rethwisch, 2010). According to Callister & Rethwisch (2010), the preform process is the first before the molding process in order to reduce the molding time and pressure used. Preform is the process which material may be mixed and cold-press into a disc. Based on Al-bahadly (2013) thesis, the curing temperature for cold press and hot press are 40-50°C and 80-100°C for one to two hours respectively. The thickness of the product can be between 1-10 mm.

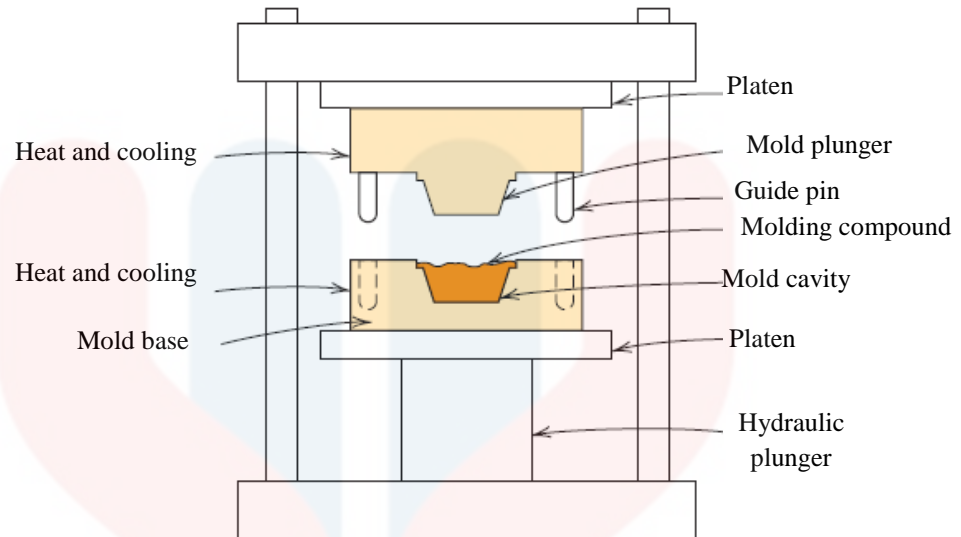


Figure 2.8: Compression molding (Callister & Rethwisch, 2010).

2.7.2 Pultrusion

Pultrusion is used to produce vary of the component shape that has continuous lengths and a constant cross-sectional shape such as rods, tubes and beams. It is a continuous process that is easily automated and the production rate is relatively high, this is making it very cost effective. Figure 2.9 show the pultrusion process which is in this process, the product is pulled from die rather than forced using pressure (Callister & Rethwisch, 2010; Thomas *et al.*, 2012).

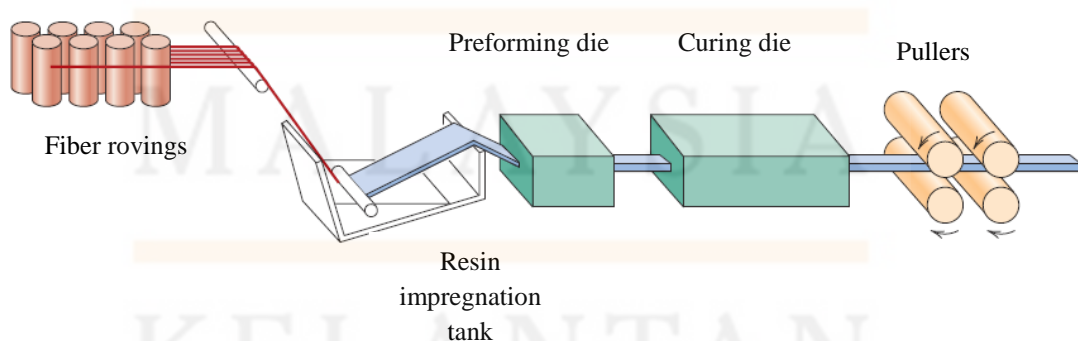


Figure 2.9: Pultrusion process (Callister & Rethwisch, 2010)

2.7.3 Prepreg

Prepreg process is shown in the Figure 2.10. This process is for the continuous fiber reinforcement pre-impregnated with a polymer resin that is only partially cured. The final product for this process is thin tape composite which is consisting of the continuous and aligned fiber embedded in a partially cured resin. The thickness of the thin tape is range between 0.08 and 0.25 mm and the widths range between 25 and 1525 mm (Callister & Rethwisch, 2010).

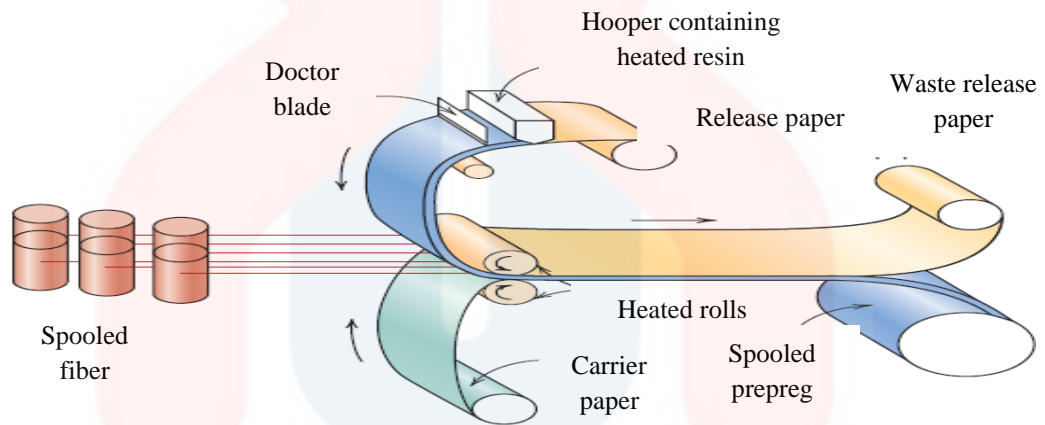


Figure 2.10: Prepreg process (Callister & Rethwisch, 2010).

2.7.4 Filament Winding

Filament winding process is a process for produce the hollow (usually cylindrical) shape of composite. Figure 2.11 show the filament winding process. In this technique, the product was produced by high speed precise lay down. The examples of the composite product using this process are pipes, storage tank and rocket motor casings (Callister & Rethwisch, 2010; Thomas *et al.*, 2012).

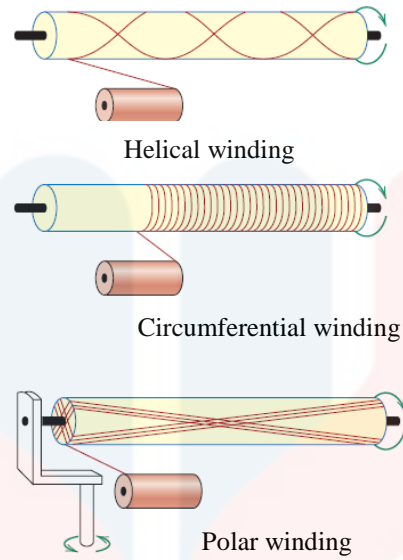


Figure 2.11: Filament winding process (Callister & Rethwisch, 2010).

CHAPTER 3

MATERIALS AND METHOD

3.1 Introduction

This chapter was explained the materials and methods that used in order to produce a composite material which can be presented in Figure 3.1. The processes are involved the preparation of raw material such as coconut shell (CS), composites preparation and characterization of physical and mechanical properties of the composites.

3.2 Preparation of Raw Materials

In this study, composite was produced using polyester, coconut shell (CS) and titanium dioxides (TiO_2). The type of polyester that was used as a matrix in this study are unsaturated polyester resin (UPR) and TiO_2 (99.8% trace metals basis with molarity value 78.87g/mol) was used as filler. Both UPR resin and TiO_2 powder was purchased by Sigma Aldrich. The UPR was mixed with 1 wt% of methyl ethyl ketone peroxide (MEKP). CS was obtained from the coconut milk seller in Jeli area. The coconut husk that still left behind on the surface of CS was removed using knife and it was cleaned with pipe water to remove any dust and dirty on their surface due to the husk removing process. Then, CS was allowed to dry under sunlight for one day to remove any moisture contents in order to produced pure CS. This is to obtain the accurate result. After one day, the shell was crushed into chips form and the chips was grounded into powder form with 150 μm . CS was divided in variety of composition for samples CS1, CS2, CS3, CS4 and CS5 which are 35 wt%, 30 wt%, 25 wt%, 20 wt% and 40 wt%.

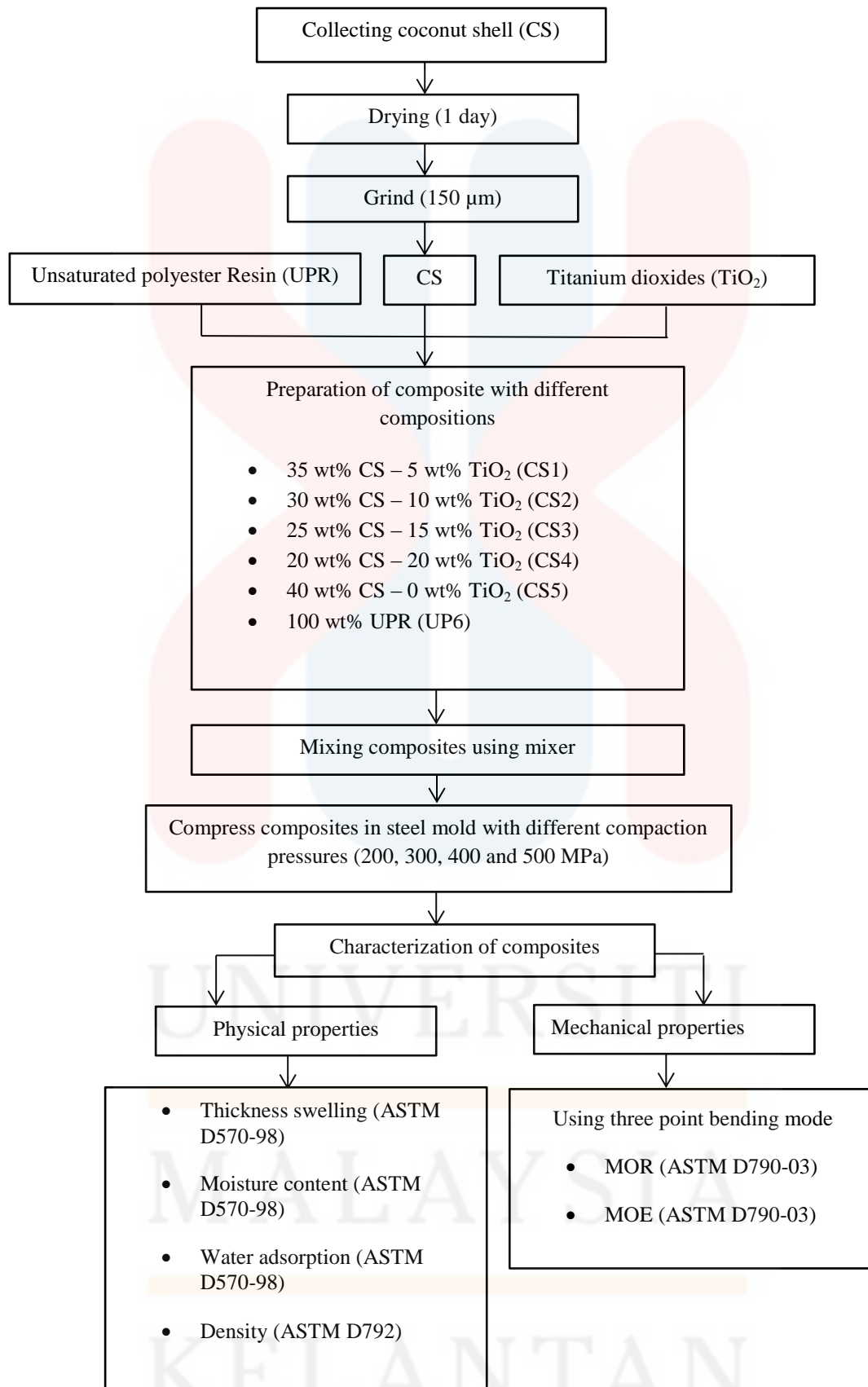


Figure 3.1: Methodology process for UPR-CS-TiO₂ composites preparation and characterization.

3.3 Composite Preparation

The unsaturated polyester resin (UPR)-CS-TiO₂ composite was produced using compression molding. UPR, CS fiber and TiO₂ was mixed together by commercial mixture that has been shown in Figure 3.2 to achieve controlled and uniform dispersion of the all constituents and make sure there are no air bubbles presence during the mixture because it can cause in presence of void that can affect the physical and mechanical properties of composite. There are six samples with different composition of CS particles and TiO₂ was produced which are CS1, CS2, CS3, CS4, CS5 and UP6 this different composition was calculated using rule of mixture. Variation of composition is shown in Table 3.1.

The mixture was poured into a 150 mm x 150 mm x 3 mm steel mold as shown in Figure 3.3 to produce a composite with various compaction pressures (200, 300, 400 and 500 MPa). Before the mixtures is pour into the mold, the mold was first polished to prevent the composite sticking in the mold wall and it make the removing of composite from the mold become easier. In order to cure, the composite was left it for one day at room temperature. After the mixture is hardened, it was removed from the mold. Then, the sample was cut into desire size for physical and mechanical properties followed the standards of American Society for Testing and Materials (ASTM).



Figure 3.2: Commercial mixture

Table 3.1: Different compositions for preparation of composite

Sample	Unsaturated polyester resin (UPR) (wt%)	Coconut shell (CS) (wt%)	Titanium dioxide (TiO ₂) (wt%)
CS1	60	35	5
CS2	60	30	10
CS3	60	25	15
CS4	60	20	20
CS5	60	40	0
UP6	100	0	0

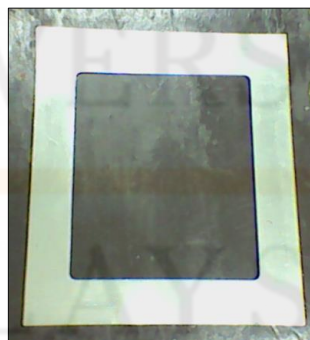


Figure 3.3: Mold

3.4 Compression Molding

The sample was cold pressed in a shaping machine as shown in Figure 3.4. The composites were placed on a disc (preform) in order to decrease the process time. Then, the sample was pressed between two mold die and the different compaction pressure (200, 300, 400 and 500 MPa) was used for each composite composition.



Figure 3.4: Compression molding machine

3.5 Physical Properties

The physical properties of unsaturated polyester resin (UPR)-CS-TiO₂ composite such as density, percentages of thickness swelling, percentages of water absorption and percentages of moisture content was determined. The samples were

cut into desired size according the standards of American Society for Testing and Materials (ASTM).

3.5.1 Density

The density of UPR-CS-TiO₂ composite was calculated using Eq. 1. The density was measured using Archimedes principle using the electronic balance with density kit. The initial mass of the composite was measured as m_0 and the final mass of the sample after sink in the distilled water was measured as m_1 . The mass of the sample was measured using analytical balanced that capability to reading up to 0.0001g. Before the mass was measured, the sample was placed into an oven for one day at 105°C to eliminate the effect of moisture and water absorption. The sample with the size 10 mm x 10 mm x 1 mm followed the ASTM D792 standard.

$$\rho = \frac{m_0}{m_0 - m_1} \times \rho_w \quad \text{Eq. 1}$$

where m_0 is mass of composite in the air, m_1 is a mass of composite in the water and ρ_w is density of water.

3.5.2 Thickness Swelling

The thickness swelling of composite sample was measured using digital vernier caliper and the sample was cut into 50 mm x 50 mm x 3 mm size according to the ASTM D570-98. The thickness of the sample before and after soaking in the distilled water was measure as T_0 and T_1 . The percentage of thickness swelling was measured using Eq. 2:

$$\text{Percentage of thickness swelling}(\%) = \frac{T_1 - T_0}{T_0} \times 100 \% \quad \text{Eq. 2}$$

where T_0 is initial thickness of sample before immersion in distilled water and T_1 is final thickness of sample after immersion in distilled water (mm).

3.5.3 Water Absorption

The percentage of water absorption was measured using Eq. 3 after soaking the sample into distilled water for one day. The composite sample was cut into dimension of 50 mm x 50 mm x 3 mm according to the ASTM D570-98. Then, the sample was weighed as W_0 before being immerse in the distilled water and the sample will be weighed as W_1 after being immersed in the distilled water.

$$\text{Percentage of water absorption}(\%) = \frac{W_1 - W_0}{W_0} \times 100 \% \quad \text{Eq. 3}$$

where W_0 is initial weight of sample before immersion in distilled water and W_1 is final weight of sample after immersion in distilled water.

3.5.4 Moisture Content

The percentage of moisture content was measured by Eq. 4. The composite was cut into dimension of 50 mm x 50 mm x 3 mm according to the ASTM D570-98. The samples were heated in an oven for one day at 105°C. Before heating the samples, the weight of composite was measured as M_0 and the composite sample after being heated will be measure as M_1 .

$$\text{Percentage of moisture content(\%)} = \frac{M_1 - M_0}{M_0} \times 100 \% \quad \text{Eq. 4}$$

where M_0 is initial weight of sample before heated in an oven and M_1 is final weight of sample after heated in an oven.

3.6 Mechanical Properties

The modulus of elasticity (MOE) and modulus of rupture (MOR) were determined using three point bending test method in order to investigate the mechanical properties of UPR-CS-TiO₂ composite. The samples were cut into desired size followed the standards of American Society for Testing and Materials (ASTM). Figure 3.5 show the flexural machine that used to determine the MOE and MOR information.



Figure 3.5: Flexural machine

3.6.1 Modulus of Elasticity

Modulus of elasticity (MOE) was determined using Testometric with five tonnes load. The sample was cut into 110 mm x 50 mm x 3 mm size. The sample was determined by ASTM test method D790-03.

3.6.2 Modulus of Rupture

The sample was cut into dimension of 110 mm x 50 mm x 3 mm for modulus of rupture (MOR) test. Sample was determined according to ASTM test method D790-03 using Testometric with 5 tonnes load.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Introduction

This chapter explains the results and discussion on the effect of different composition and compaction pressure of UPR-CS-TiO₂ composite. Mechanical properties and physical properties were discussed in this chapter are flexural which was presented by modulus of rupture (MOR) and modulus of elasticity (MOE), density, thickness swelling, water absorption and moisture content.

4.2 Mechanical Properties

4.2.1 Modulus of Rupture

Figure 4.1 shows MOR of UPR-CS-TiO₂ composite with different composition of fibers and fillers and compaction pressure. MOR for each composite exhibited different trends at all the compaction pressures due to different composition the fibers and fillers addition. UP6 with 100% of UPR show the highest MOR compared to the others composites as a result of hydrophobic properties. This property prevents the water and moisture uptake within the composite. If water and moisture absorption is higher it will lead to the poor composite mechanical properties (Kabir *et al.*, 2012). The highest MOR of UP6 also can be because of polyester polymer is a ductile phase and least brittle in composite (Thomas *et al.*, 2012). However, at 300 MPa MOR of UP6 decreased due to void and crack present during inhomogeneous mixing, casting and hardened process.

Composite with 40 wt% CS show the highest MOR at 200 and 400 MPa as supported by Liu *et al.* (2016) which the flexural properties of composite is increased due to increase of fiber content. This is also due to minimum of void present inside composite which the highest MOR was obtained when the lowest porosity present. Higher void content lead to sudden failure on composite and results in low mechanical properties (Kabir *et al.*, 2012). It is observed lower MOR was exhibited by CS3 and CS4.

Meanwhile, composite with equal fiber and filler (20 wt%) contents show lower MOR compared to that higher fiber content. According to Jawaid *et al.* (2011), incomplete wettability of fiber and filler within matrix causes void formation in the composite. Therefore, higher TiO₂ content may lead to void generation as a result of lack of good dispersion within matrix and reinforcement. However, the composite such as CS1, CS3, CS4 and CS5 decreased in MOR at 300 MPa due to poor compatibility between fiber, filler and matrix. This is because the hydroxyl group in fiber content and the formation of porosity. The porosity present is due to the improper mixing and casting process.

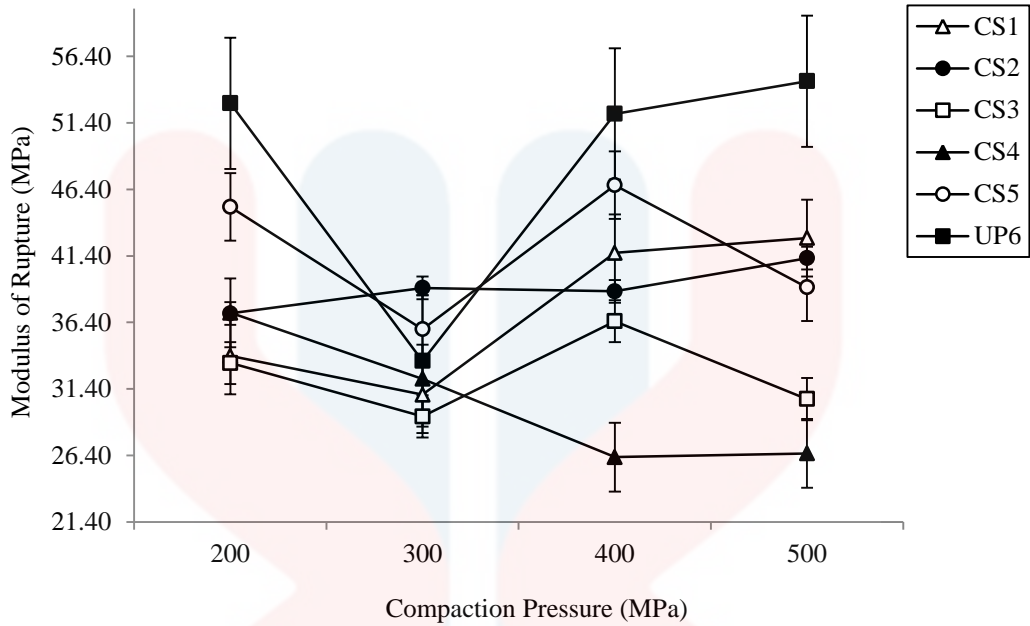


Figure 4.1: MOR of UPR-CS-TiO₂ composite

4.2.2 Modulus of Elasticity

Modulus of elasticity (MOE) of UPR-CS-TiO₂ composite is shown in Figure 4.2. UP6 composite had the lowest MOE among the composite different composition. This composite had lower deformation and thus, prone to failure as a result of no fiber additions to prevent crack propagation. The fiber or reinforcement addition is important in structural composite to support any loading on the composite (Al-bahadly, 2013). The highest MOE compared to the other composite was CS5 because CS is the highest. This is in agreement with Bledzki *et al.* (2010) where higher content CS in polymer matrix resulting in better elongation break.

Moreover, CS3 show the lowest of MOE at 200 and 300 MPa while at 400 MPa, the lowest of MOE was CS4 this is due to the CS fiber that tend to has high amount of moisture when expose to aqueous environment. Poor MOE properties and caused by the weak interfacial bonding between fiber-matrix interface (Zamri *et al.*, 2016). Other than that, the lowest MOE is also depends on the void formation within composite. Void formation is caused by bubbles during mixing and casting process. Similar to MOR, higher content of filler also lead to lower MOE. Moreover, TiO₂ was not well dispersed within matrix and reinforcement.

CS1 with the lowest content of TiO₂ show the MOE is increased with increased in compaction pressure. This can be concluded that, the structure of composite components became highly packed due to high compaction pressure lead to better MOE properties. This is because the chance of void formation within composite is minimum.

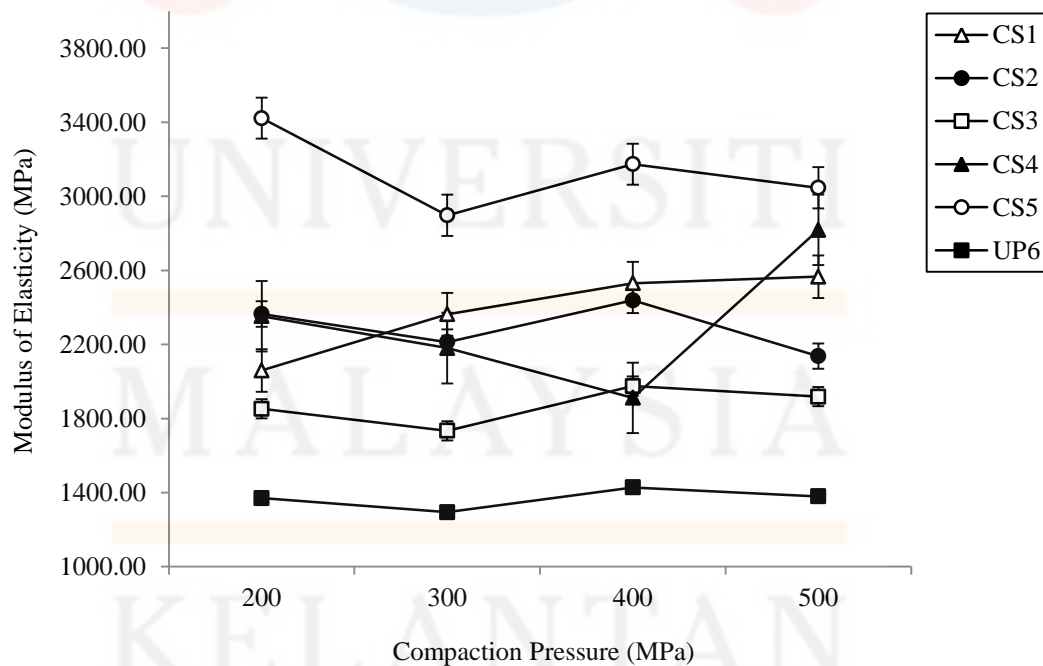


Figure 4.2: MOE of UPR-CS-TiO₂ composites

4.3 Physical Properties

4.3.1 Density

Figure 4.3 shows the density of different compositions and compaction pressure of UPR-CS-TiO₂ composite. In composite industry, the lowest density is the most desirable factors for composites products. In this study, the change of density at different compaction pressures is not significant. All composites show the density higher than the reference composite (UP6) due to different composition of fibers and fillers contents.

CS5 with the highest fiber content (40 wt%) show the lowest density compared to the composite that was added with the lower filler (TiO₂) content such as CS1, CS2, CS3 and CS4. According to Biswas *et al.* (2016), high density is induced by void formation due to increasing fiber content. This is vice versa with this study which the composite with highest fiber content shows the lowest of density although during the void present. This is may be due to the agglomeration occur between TiO₂ and CS fiber is the highest compared to the void present resulting in highest density.

This can be proved in this study that the addition of TiO₂ content with 5, 10, 15 and 20 wt% had increased the density of composite. The agglomeration of TiO₂ inside composite is caused by the inhomogeneous is mixing. Besides that, the lumpy that produced by TiO₂ before and after the mixing process may be due to it long term exposure to the atmosphere environment. In addition, the gap present between the matrix and filler interface lead to the holes present with increase in the filler content (Biswas *et al.*, 2016). Although CS5 shows the lowest density among composites but

density of CS5 was higher than UP6 due to higher fiber content that also supported with the study of (Liu *et al.*, 2016) where the density is increased when fiber content is increased.

Besides that, with the increased of fiber content also affects the density of composites. Higher fiber content may lead to water easily filled the void within composite. The lowest density was between $1.27-1.29\text{gcm}^{-3}$ while the highest density was 1.47gcm^{-3} . In this study, it can be concluded that the value of density is directly proportional to the TiO_2 content but somehow, the density are inversely proportional to the CS content.

Density for CS5 is increased with the increased of compaction pressure from 200 to 400 MPa however density is decreased back when 500 MPa apply. The composite which was compacted by high pressure produced minimum of void or porosity due to having high point contact number and contact area. The relative density is increased when the compaction is increased. This is also supported by Sridhar & Fleck (2000) study. After the density is remain constant at 200 to 400 MPa, CS1 and CS4 composites density were decreased at 500 MPa. According to Sahari & Maleque (2016), the density of UPR/oil palm shell (OPS) composite are decreased with addition of fiber content (OPS) and similar in this study where the density of UPR-CS- TiO_2 composite decreased due to addition of CS content ranging from 20 to 40 wt%.

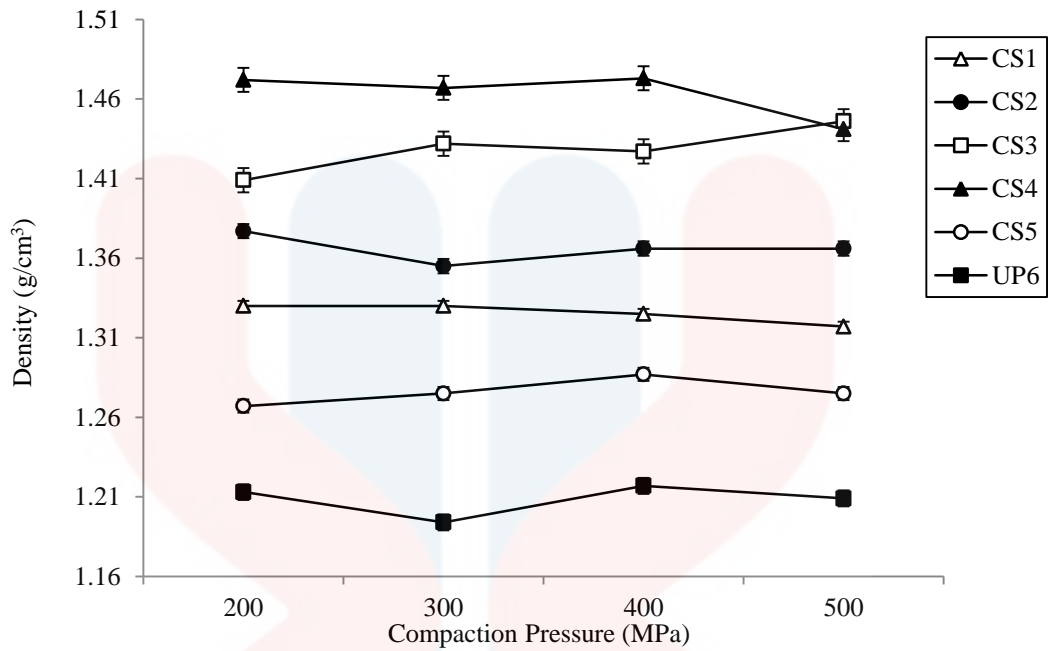


Figure 4.3: Density of UPR-CS-TiO₂ composites

4.3.2 Thickness Swelling

Thickness swelling of UPR-CS-TiO₂ composite is shown in Figure 4.4. In the most composite, the percentage thickness swelling was decreased when the pressure was increased. The thickness swelling is directly proportionate to the water and moisture absorption. This is due to the water and moisture built up in the cell wall and fiber/matrix interface (Abdul Khalil *et al.*, 2011) that caused dimension change and thickness swelling of the composites. As shown in Figure 4.4, CS4 has the highest thickness swelling with the equal percentage content of CS fiber and filler contents (20 wt%) at 200 to 400 MPa. This is due to lower dispersion between matrix, fibers and fillers that can lead to the agglomeration. The presents of lumpy of TiO₂ in composite causes the porosity formation also can be the reason of the thickness swelling is higher.

All of the composites shows the percentages of thickness swelling were decreased with the increased of compaction pressure. This is because of the composite structure became highly compact due to increase in compaction pressure that cause only small chance for void present and water or moisture absorbs inside composites. While, at 400 and 500 MPa the lowest of thickness swelling was CS3 composite with 0.15 and 0.26 wt%. This may be due to the good mixing between fiber and filler.

Different content of CS fiber and TiO₂ produced different in thickness swelling. The main reason is the present of polar group in fibers component that lead the composite to attract with water molecule due to its poor resistance to water uptake (Kabir *et al.*, 2012; Fuentes *et al.*, 2016; Abdul Khalil *et al.*, 2011). Addition of high filler content also leads to high thickness swelling due to less disperse between CS and TiO₂. Those problems usually occur during the mixing process and this also can cause the agglomeration within composite. In addition, the way of measurement of thickness swelling taken also affected the reading of thickness swelling percentage. In this study, thickness swelling was taken in three different points on composite. However, the measurement may differ from each point because some parts may have void content and some part have less void content.

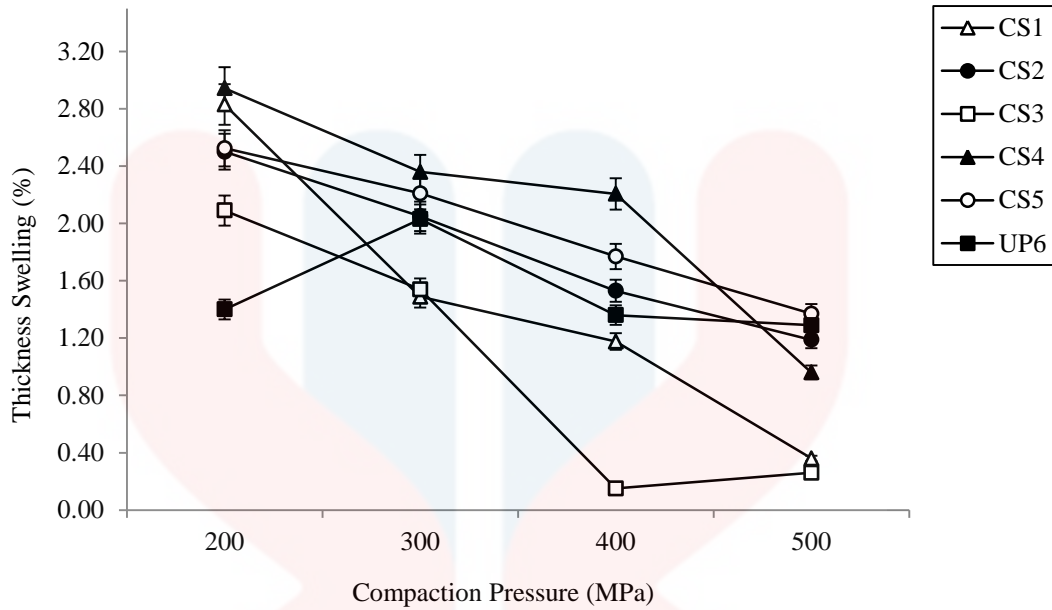


Figure 4.4: Thickness swelling of UPR-CS-TiO₂ composites

4.3.3 Water Absorption

Water absorption of UPR-CS-TiO₂ composites is illustrated in Figure 4.5. The percentages of water absorb was different due to the different fiber and filler compositions and pressures. UP6 composite shows water absorption percentages decreased with increasing in compaction pressures. This is because the hydrophobicity of UPR and lower void.

In fiber and filler addition composite, the lowest water retention was belonged to CS4 composite (between 0.88-1.33%) whereas the water uptake is increased with increased of the compaction pressures from 200 MPa to 400 MPa. This is mainly due to the hydrophilicity behavior of natural fiber (CS) that caused by the presence of hydroxyl group which lead to the increasing in water absorption (Patel & Parsania, 2016). In addition, the presence of void in composites also can be identified as factor contributing to the increased in water uptake (Abdul Khalil *et al.*,

2011). The void formed due to the highest composition of TiO₂ (20 wt%). However, the percentages of water retention were decreased rapidly at 500 MPa as a result of high compaction pressure which caused the composites become more compacted and reduced void formation.

In CS1 composite, the highest water uptake was exhibited at compaction pressures of 200 and 300 MPa. The fiber contents (35 wt%) in CS1 composite was higher than the other composites. This lead to more water absorbed inside the composites due to presence of polar group and waxy substance in fibers structure (Omole & Dauda, 2016). As a general, CS1 shows the water retention are decreased with increased the compaction pressure and the percentages of water absorbs remains constant at 500 MPa this may be due to the cell walls of fibers are saturated with water resulting in no water uptake (Patel & Parsania, 2016).

According to Dhakal *et al.* (2006), three mechanism on how the moisture or water diffuse inside the composite material. First, the absorption of water molecules within the micro gaps between polymer chains. Second, involves the diffusion of water into gaps and flaws at the interface between fiber and matrix. The third mechanism is involved the transport of micro crack in the matrix that arising from the swelling of fiber. When the composite with these weaken properties was exposed to moisture environment this may result to higher water absorption which this leading to composite failure.

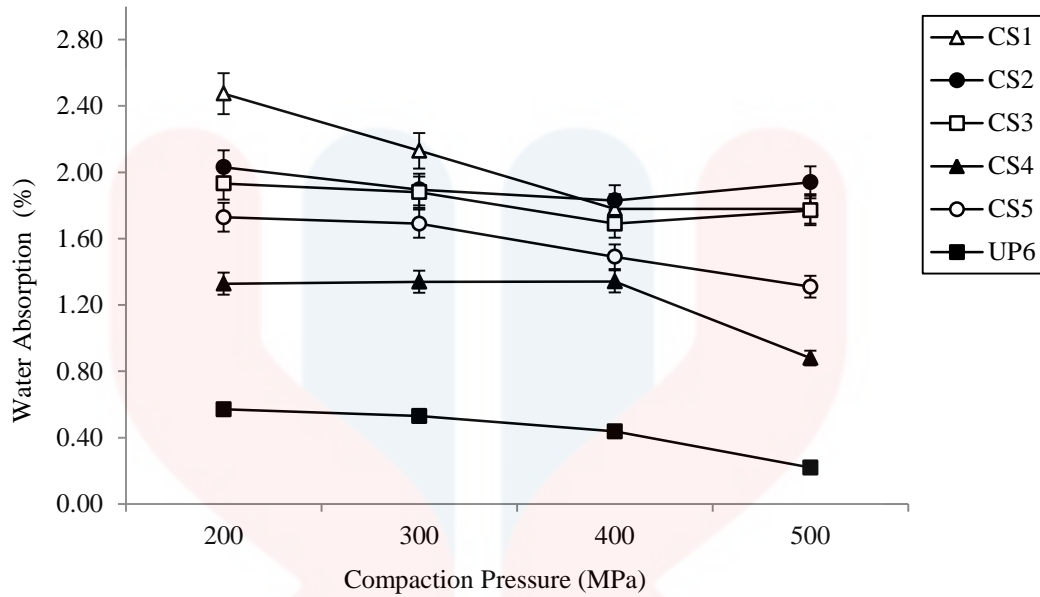


Figure 4.5: Water absorption of UPR-CS-TiO₂ composites

4.3.4 Moisture Content

Figure 4.6 presents the percentages of moisture content for UPR-CS-TiO₂ composites at different compaction pressure and compositions of fiber and filler contents. Similar to that of water absorption, the highest moisture content in composites can lead to poor the mechanical properties of composites due to the poor stress transfer efficiencies caused by degradation of fiber matrix interface region (Dhakal *et al.*, 2006). UP6 composites show the percentages of moisture content are dramatically decreased with the pressure increased due to reduce in gap between C=C double bonds which the moisture uptake is difficult to diffuse through it (Chawla, 2012).

CS4 with 20:20 wt% of CS and TiO₂ contents shows the least moisture absorption percentages compared to CS1, CS2, CS3 and CS5. This is due to well dispersed between matrix, fibers and fillers contents. Starting at 300 MPa, the

moisture uptake was decreased when compaction pressure is increased this is because the applied pressure can remove the air inside the composites during the compaction which this resulting in minimum of void present. The lowest moisture uptake is at 500 MPa with 1.64%. However, the lignocellulosic fibers (CS) as reinforcement in composites was lead to high moisture content due to its hydrophilic and absorb moisture properties (Kabir *et al.*, 2012). This can be proven at Figure 4.6 which shows the highest of fibers content with 40 wt% (CS5) of CS lead to the highest of moisture uptake by 2.85 and 2.79% at 400 and 500 MPa.

Although the CS5 shows the highest in moisture absorbs, the percentages of moisture absorb is decreased due to high compaction pressure (500 MPa). This can be conclude that, the composites is well compact caused the low in moisture retention. In addition, the high compaction pressure leads to lower void present inside the composites. According to Sahari *et al.* (2011), the present cellulose, hemicellulose and lignin in natural fibers also lead to high percentage of moisture absorption. CS1 with 35 wt% of CS fibers show the high moisture uptake at 200 and 300 MPa due to the moisture from the atmosphere came in contact with cellulose, hemicellulose and lignin of the CS fiber which the fibers became the main access of moisture penetration lead to the increased in moisture content absorption.

Besides that, the present of porosity or void inside composites also one of the reason of high moisture retention due to TiO₂ that used in this study became lumpy during the mixing process which lead to air trapped inside the lumpy during the harden process and the not uniform in cutting of composite also can affected the moisture uptake.

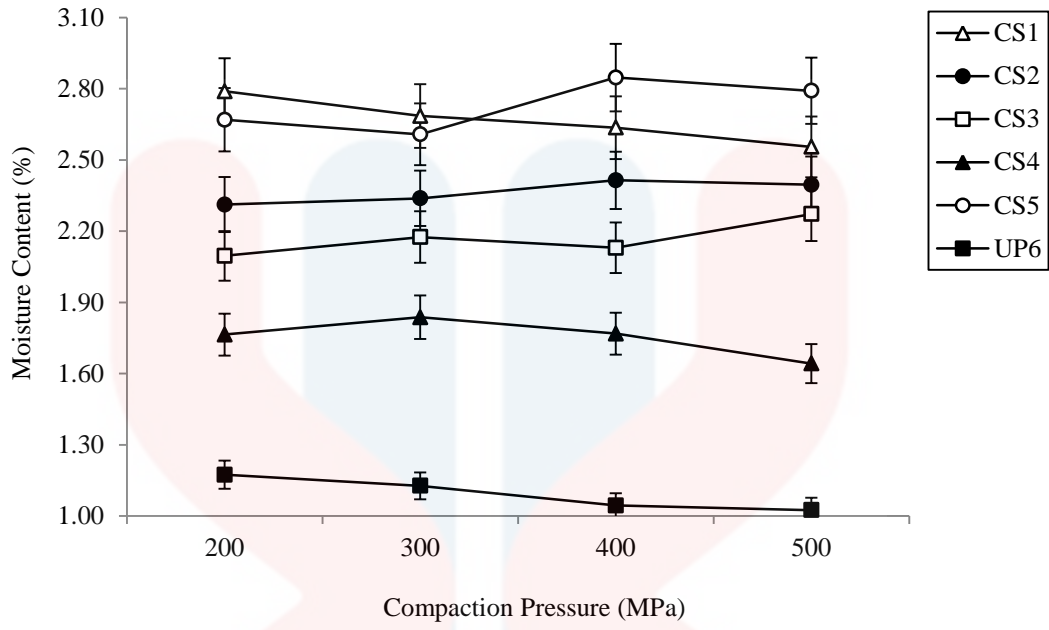


Figure 4.6: Moisture content of UPR-CS-TiO₂ composites

CHAPTER 5

CONCLUSION AND SUGGESTIONS

5.1 Conclusion

In this study, the UPR-CS-TiO₂ has been successful fabricate using compressing molding. The objectives of this study were been investigated by mechanical and physical testing. The effect of variations consolidation pressure, CS composition and TiO₂ contents has been obtained as shown in chapter 4. This can be concluded that:

1. CS5 without addition of TiO₂ show the highest of MOR and MOE compared to the composites that content TiO₂.
2. The density of UPR-CS-TiO₂ composites increased due to the increased in TiO₂ content which caused by higher agglomeration and void content present between CS and TiO₂ particles.
3. Thickness swelling, water absorption and moisture content decrease due to increment of compaction pressure. This was due to the gap between matrix and fiber interface is small caused the water or moisture cannot diffuse inside the composite.
4. CS4 show the lowest of water uptake and moisture uptake while thickness swelling of CS4 show the highest as a result of lack of dispersion between fiber and filler.

5.2 Suggestions for Future Work

There are a few of problem presents either before or after the composite preparation which this problem can lead to the poor physical and mechanical properties of composite. In order to avoid from this problem happen, some suggestion for future work should be considered such as:

1. Pre-treatment of natural fiber must be carried out to remove the hydroxyl group in the natural fiber in order to enhance the interface between polymer matrix and reinforcement.
2. Natural fiber should be dry before the impregnation of matrix and composite manufacturing must be performed under controlled humidity environment to avoid the composite from attacked by water or moisture diffusion.
3. Nano size of TiO_2 need to use as filler in composite compared the micro size of TiO_2 . This is because the micro size of TiO_2 leads to higher porosity and agglomeration present.
4. The variations of thermal, UV and mechanical testing should be considered.

In addition, the surface morphology of composite must observed to know the dispersion between polymer matrix, fiber and filler.

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APPENDIX A

TABLES

A.1 Tables of MOR, MOE, density, thickness swelling, water absorption and moisture content results for UPR-CS-TiO₂ composites.

(a) MOR

Compaction pressure (MPa)	MOR (MPa)					
	CS1	CS2	CS3	CS4	CS5	UP6
200	33.88	37.08	33.34	37.12	45.08	52.87
300	30.96	38.99	29.33	32.15	35.89	33.51
400	41.63	38.73	36.49	26.27	46.71	52.08
500	42.75	41.23	30.64	26.55	39.04	54.53

(b) MOE

Compaction pressure (MPa)	MOE (MPa)					
	CS1	CS2	CS3	CS4	CS5	UP6
200	2059.55	2364.95	1852.52	2352.44	3422.06	1370.31
300	2362.79	2212.81	1733.80	2180.42	2879.13	1293.99
400	2530.33	2437.94	1974.81	1911.56	3173.49	1427.87
500	2565.96	2136.82	1917.96	2818.80	3045.96	1379.58

(c) Density

Compaction pressure (MPa)	Density (gcm ⁻³)					
	CS1	CS2	CS3	CS4	CS5	UP6
200	1.33	1.38	1.41	1.47	1.27	1.21
300	1.33	1.36	1.43	1.47	1.28	1.19
400	1.33	1.37	1.43	1.47	1.29	1.22
500	1.32	1.37	1.45	1.44	1.28	1.21

(d) Thickness swelling

Compaction pressure (MPa)	Percentages of thickness swelling (%)					
	CS1	CS2	CS3	CS4	CS5	UP6
200	2.83	2.50	2.09	2.94	2.53	1.40
300	1.49	2.05	1.54	2.36	2.21	2.03
400	1.18	1.53	0.15	2.21	1.77	1.36
500	0.36	1.19	0.26	0.96	1.37	1.29

(e) Water absorption

Compaction pressure (MPa)	Percentages of water absorption (%)					
	CS1	CS2	CS3	CS4	CS5	UP6
200	2.48	2.03	1.93	1.33	1.73	0.57
300	2.13	1.90	1.88	1.34	1.69	0.53
400	1.78	1.83	1.69	1.34	1.49	0.44
500	1.78	1.94	1.77	0.88	1.31	0.22

(f) Moisture content

Compaction pressure (MPa)	Percentages of moisture content (%)					
	CS1	CS2	CS3	CS4	CS5	UP6
200	2.79	2.31	2.10	1.76	2.67	1.17
300	2.69	2.34	2.18	1.84	2.61	1.13
400	2.64	2.41	2.13	1.77	2.85	1.04
500	2.56	2.40	2.27	1.64	2.79	1.03

APPENDIX B

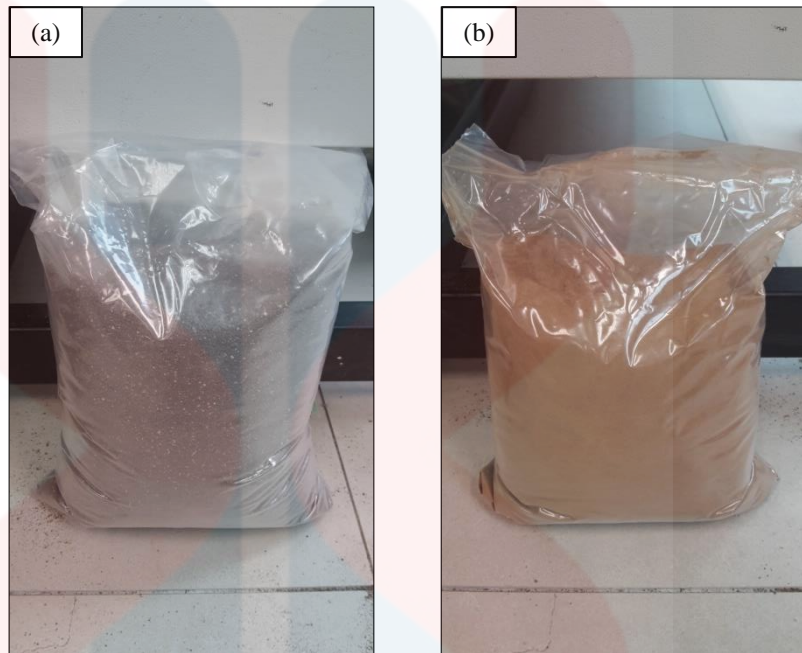


Figure B.1: (a) Coconut shell before sieve (b) Coconut shell after sieve (150 μm)



Figure B.2: 35 wt% of CS and 5 wt% of TiO₂ (CS1)

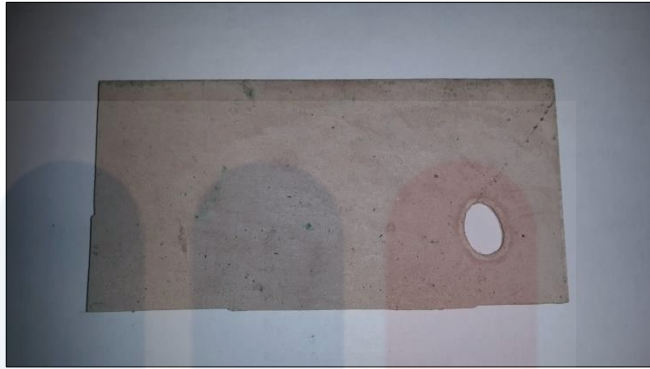


Figure B.3: 30 wt% of CS and 10 wt% of TiO₂ (CS2)



Figure B.4: 25 wt% of CS and 15 wt% of TiO₂ (CS3)



Figure B.5: 20 wt% of CS and TiO₂ (CS4)



Figure B.6: 40 wt% of CS (CS5)

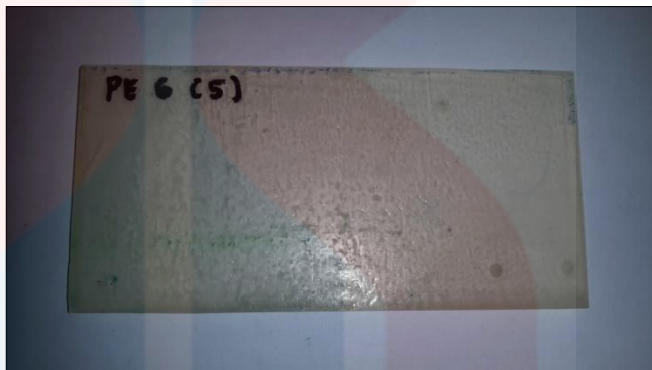


Figure B.7: 100 wt% of UPR