SOUND ABSORPTION CHARACTERISTICS OF NATURAL FIBER FILLED IN FLEXIBLE FOAM UPON UV IRRADIATION EXPOSURE

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This thesis especially dedicated to:

My loving husband,

whose sacrificial care for me and our daughter Amanda

made it possible for me to complete this study

My beloved mother, father and in law’s family

My supportive supervisor

My family members

My dearies friends

~Thanks for all the courage and spirit~

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Thank you.

ABSTRACT

Wood filler- flexible foam composite (FC) has been studied and proved to have ability to absorb sound. In this study, untreated and treated *Meranti Merah* wood dust and Palm Trunk namely as MM and PT respectively was use as the filler. Therefore, this study was developed to compare the behavior and the ability of FC towards sound absorption coefficient based on conditions of the fillers and wood fiber sizes. With the size of <355 and <500 µm, three different percentages has been selected which is 10 wt%, 15 wt% and 20 wt% for both type of woods. Subsequently, these samples have been tested by using Impedance Tube test for determination of sound absorption coefficient (SAC). The morphological structure has been test using Scanning Electron Microscopy (SEM) to observe the pores structure of wood-FC. While, Fourier Transform Infrared (FTIR) was used to determine the functional group of FC and UV-Weatherometer was used to determine the resistance of FC towards UV light up to 1000 hours. As a result, the SAC shows value at achievable frequency range of 2500 to 6000 Hz. The highest SAC is 0.995 while the lowest SAC is 0.600. Nevertheless, a result of FC after UV exposures shows only small decreases in SAC values and frequency absorption for some samples were slightly shifted toward higher range for example sample 15 wt.% untreated <355 µm Meranti-FC curve shift from 4500 to 5000 Hz after 1000 hrs UV exposures. The pores structures after UV light exposures shows slightly increasing in sizes and the SAC values decreased for all samples. As a conclusion, in this study, the SAC behavior of <355µm *Meranti Merah*, <500µm *Meranti Merah*, <355µm Palm Trunk and <500µm Palm Trunk-FC samples show the ability to influence the absorption coefficient of polymeric foam at different frequency levels. Lastly, the MM and PT-FC is highly resistance to UV light and suitable for sound-proofing blanket and curtain systems for outdoor sound applications.

ABSTRAK

Kayu isian- busa berkomposit (FC) telah dikaji dan terbukti mempunyai keupayaan untuk menyerap bunyi. Di dalam kajian ini, kayu meranti merah dan batang kelapa sawit yang tidak dirawat dan dirawat dengan asid masing-masing dinamakan sebagai MM dan PT digunakan sebagai kayu isian. Oleh itu, kajian ini telah dijalankan untuk membandingkan kelakuan dan keupayaan FC terhadap pekali penyerapan bunyi berdasarkan keadaan kayu isian dan saiz serat kayu. Dengan tiga peratusan yang berbeza iaitu 10 wt%, 15 wt% dan 20 wt%, saiz <355 dan <500 µm telah dipilih bagi kedua-dua jenis kayu. Selepas itu, sampel-sampel ini telah diuji dengan menggunakan ujian Tiub Bergalang untuk menentukan pekali penyerapan bunyi (SAC). Bagi ujian struktur morfologi Mikroskopi Pengimbas Elektron (SEM) telah digunakan untuk memerhati struktur liang untuk kedua-dua saiz dan jenis kayu-FC. Manakala, FTIR telah digunakan untuk menentukan kumpulan berfungsi yang wujud di dalam FC dan UV-Weatherometer telah digunakan untuk menentukan ketahanan FC terhadap cahaya ultra-ungu(UV) selepas 1000 jam. Kesan terhadap penyerapan bunyi, SAC menunjukkan nilai pada frekuensi 2500-6000 Hz. Nilai SAC tertinggi 0.995 manakala nilai SAC yang paling rendah ialah 0.6. Walau bagaimanapun, hasil selepas pendedahan FC terhadap UV menunjukkan perubahan kecil pada nilai SAC dan frekuensi penyerapan bergerak ke julat yang lebih tinggi sebagai contoh sampel 15 wt.% tidak dirawat <355 µm MM-FC beralih dari 4500 ke 5000 Hz selepas 1000 jam terdedah pada cahaya UV. Terdapat perubahan pada struktur liang selepas pendedahan cahaya UV dan saiz turut meningkat dan nilai SAC juga menurun bagi kesemua sampel. Kesimpulannya, dalam kajian ini, kelakuan SAC bagi sampel M3, M5, P3 dan P5-FC menunjukkan keupayaan untuk mempengaruhi pekali penyerapan busa polimer pada frekuensi yang berbeza. Akhir sekali, MM dan PT- FC memberi kesan rintangan terhadap cahaya UV dan sesuai untuk langsir luaran penyerap bunyi atau langsir di dinding penghalang bunyi.

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ASTM	-	American Society for Testing Materials
H_2O	-	Water
O_2	-	Oxygen
CO_2	-	Carbon dioxide
OH	-	Hydroxyl group
C-C	-	Carbon to carbon
RH	-	Rice Husk
FTIR	-	Fourier Transform Infrared
H	-	Hours
MDI	-	Methylene diisocyanate
TDI	-	Toluene diisocyanate
HMDI	-	Hexamethylene diisocyanate
PU	-	Polyurethane
PU	-	Polyurethane Foam
FC	-	Foam composite
WPC	-	Wood polymer composite
UV	-	Ultraviolet
MM	-	Meranti Merah
PT	-	Palm Trunk
M3	-	*<355µm Meranti Merah*
M5	-	*<500µm Meranti Merah*
P3	-	*<355µm Palm Trunk*

P5	-	*<500µm Palm Trunk*
UM	-	*Untreated Meranti Merah*
TM	-	*Treated Meranti Merah*
UP	-	*Untreated Palm Trunk*
TM	-	*Treated Palm Trunk*

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CHAPTER 1

INTRODUCTION

1.1 Introduction and research background

As the problem of undesirable and potentially hazardous noise has become serious, the demand of better environment and residential safety is increased and becomes a major requirement. Various studies focusing acoustic properties have been performed. Acoustical material plays important roles in acoustic engineering such as the control of room acoustics, industrial noise control, studio acoustics and automotive acoustics. They are used as interior lining for apartments, automotives, aircrafts, ducts, and enclosures for noise equipments and insulations for appliances (Knapen *et al.*, 2003; Youn & Chang, 2004).

From previous study, polyurethane foams composites made from palm oil were synthesized, crosslink and doped with eco natural filler of rubber waste or sawdust powder (Rus, 2010 & Rus, 2009b). As natural resources become scarce, many researchers and industries are beginning to investigate and utilize various renewable resources such as the abundant and cheap vegetable oils, which represent a major potential source of chemicals (Rus, 2008 & Rus, 2009a).

In the recent years, a number of studies have been carried to develop new materials and technologies improving the sound absorption properties, the sound absorption coefficient of the system improved with the increase of open porosity over the entire frequency range of 125- 4000 Hz. (Zhang, *et al*., 1997) & (Yu, 1999). The sound absorption panel developed by using processed bamboo and oil palm frond has been tested for its sound absorption properties (Koizumi, *et al.*, 2002 &

Sihabut, 1999). Numerous studies had been done on the sound absorption of porous material like wood based material (Wassilieff, 1996), tea-leaf-waste material (Ersoy & Kucuk, 2009), melamine foam (Kino *et al.*, 2009) and fabric (Dias *et al.*, 2007).

At the present time, green technology is proposed used to be manufacture materials from agricultural as substitute to synthetic fiber and wood based material for noise absorption purposes. Natural fibers (rice husk) have many advantages compare to synthetic fibers, for example low weight, low density, low cost, acceptable specific properties and recyclable or biodegradable. These materials have demonstrated good distinctive features from the both of aspect of sound qualities and mechanical. Zaidi *et al.,* (2009) has studied, at low frequency, 0 – 500Hz, sound absorption of rice husk was higher than virgin PU with the value of 0.899 at 250Hz. whereas, the virgin PU recorded higher absorption at a higher frequency, 2000Hz with a value of 0.679. This shows that mineral fibers have the potential as filler material of sound absorbent material (Zaidi *et al.*, 2009).

Sound absorbing materials absorb most of the sound energy striking them and reflect very little. Therefore, sound-absorbing materials have been found to be very useful for the control of noise (Arenas, *et al*. 2010); Materials that have high value of sound absorption coefficient which is above 0.900 are usually porous (Crocker, *et al*., 2007). The absorption coefficient is a useful concept when using geometrical acoustic theory to evaluate the growth and decay of sound energy in a room (Rus, *et al*., 2012).

Nowadays, in agro-industrial and plantation of timber industry, high value of hardwood saw mill residue, which is currently treated as solid waste were produced. In practice this residue is burned in incinerators which may be causes of environmental pollution problems in nearly localities and offers limited value to the industry (Rahman *et al*., 2006). Furthermore, saw mill residue uses is still limited. Basically, it is used as animal feed or simply as landfills (Musatto *et al*., 2003).

By considering this scenario, an alternative practice should be considered by the sawmill industry to commercialize the residue from hardwood species to recycle back without causing environmental pollution and produce valuable product. This practice will requires less energy, and diminishes pollutants in industrial effluents, as well as being more economically advantageous due to its reduced costs.

According to the several research groups on the hydrolysis of saw mill residue, it is only focus on the softwood species such as corn, rice husk, sugarcane

baggase and so on (Lavarack *et al*., 2002, Mussatto & Roberto, 2005 and Téllez-Luis *et al*., 2002).

In this study *Meranti Merah* wood (MM) and Palm Trunk (PT) was used because its abundant sources of the furniture manufacturing. The acoustical properties (sound absorption coefficient) of the composite were determined to investigate the possibility of untreated and treated MM and PT as filler of polymer foam composites. Both woods were chosen as raw material because of its availability. Wood sawdust is a lignocellulosic byproduct of sawmill that is available at low cost throughout the year. It is produced in enormous quantities by sawmills and the economical disposal of them is a serious problem to the wood based industries.

Roberto *et al.,* (2003), have studied that the hydrolysis of wood to produce xylose solution could be a good alternative use for this abundant resource. Xylose is a hemicellulosic sugar which can be used as a raw material for the manufacture of a wide variety of compounds or fuels by chemical and biotechnological processes The hemicellulosic fraction of wood can be easily and selectively extracted with dilute sulfuric acid under mild treatment conditions to obtain xylose-rich hemicellulosic hydrolysates which can be an economical substrate to produce xylitol. Under optimized treatment conditions, the dilute acid hydrolysis of lignocellulosic mainly produces xylose from hemicellulose, leaving a solid residue containing the cellulose and lignin fractions almost unaltered (Islam & Mimi Sakinah 2011). The remaining wood after hydrolysis treatment containing acidic cellulose-lignin and before acid hydrolysis treatment was tested as filler.

In this present study, a comparison between before and after acid hydrolysis treatment of MM and PT contributed as filler to composite foam to measure the sound absorption ability and understanding the quality of fibrous material in composite foam.

During outdoor exposure, polymers degrade chemically due to the action of short wavelength Ultraviolet rays present in the solar spectrum. The service-life of polymers in outdoor applications becomes limited due to weathering (Davis, 1977). Polymer degradation has been investigated for many years. Ultraviolet accelerated weathering or exposure testing evaluates the test for FC reaction to photo-induced degradation. Photolysis is a chemical reaction that can affect color fastness,

brittleness, fading, cracking and other forms of deterioration as a result of exposure to ultraviolet radiation.

In this study, an ultraviolet weathering chamber was used to conduct polymer foam degradation in ultraviolet exposures. Ultraviolet radiation and water condensation were cycled periodically in the chamber, simulating sunlight and rain or dew.

1.2 Problems statement

Wood-based production industries has expended and yielding a huge amount of wood. Sawdust or wood fiber is a by-product of cutting, grinding, drilling, sanding, or otherwise pulverizing wood with a saw or other tool; it is composed of fine particles of wood. Han, *et al.*, (1998) has reported that the properties of particleboard produced from fine particle were better than board that made from coarse particles.

From previous study Wassilieff, 1996, has reported that most practical sound absorbing products used in the building industry consist of glass- or mineral-fibre materials. However, the growing concern about the potential health risks as being associated with fibre shedding from glass- or mineral-fibre materials provides an opportunity for wood-based sound absorbers to be developed for use in many applications. Natural fiber reinforced polymer composites materials are almost replacing materials such as ceramics, metals, glasses, etc. From the previous research, rubber waste or sawdust powder was used as eco natural filler. The acoustical behavior of FC may vary due to the different particle sizes, foam composition and type of woods.

This treated wood is by-product from acid hydrolysis treatment of wood mainly produces xylose from hemicelluloses, leaving a solid residue containing the cellulose and lignin fractions almost unaltered in controlled condition. Cellulose is one of the most important natural polymers produced in the biosphere, and it is considered the most abundant renewable polymer on earth. Production of cellulose-lignin is estimated to be over 7.5×10^{10} tons per year (Habibi *et al.*, 2010).

Cellulose is structural materials in plants and the most abundant biomass in earth However; there are lacks of studies regarding the utilization of wood in polymer foam composite. Most of the prior studies are only focusing on the

alternative polyol from renewable natural source from rice husk, castor oil and soybean oil. Other than that, there are few studies regarding the by-product (cellulose and lignin) with the different particle sizes and wood species reinforce in polymer foam composite. Hence, this research is come out to focusing on the studies of the effect of foam composition, particle sizes and type of wood in acoustical behavior of FC. Thus, the urgency of this study is to produce an economical and high quality of alternative natural filler for polymer composite from biomass (mainly plant-wood).

Although polymer properties can be slightly affected during their processing, storage, and transportation, the most significant degradation occurs during exposure to the environment. The ultimate objective of testing is to predict the component lifetime under service conditions. The appropriate exposure conditions are therefore those that match exactly the service environment. The main motivation for artificial aging is to accelerate the weathering processes so that a reliable prediction of the service lifetime can be obtained in an acceptably short test period. Acoustical Blankets is great for use in outdoor environments where an extended lifespan is needed for the blankets. Typical uses include curtain material in acoustical enclosures where weather resistance and excellent durability is required as well as maximum longevity and noise reduction is required.

1.3 Hypothesis of research

The hypotheses of this research are:

i. To prove that by-products of xylose production from *Meranti Merah* wood and Palm Trunk can be used as an alternative filler for FCs.
ii. To prove that untreated and treated of wood filler affect the sound absorption behavior of FCs.
iii. To prove that the wood filler FC have high resistant against weathering for prolonged ultraviolet irradiation exposures.

1.4 Objectives of research

The objectives of this study are:-

i. To characterize the use of palm trunk and *Meranti Merah* wood as alternative filler in sound absorption.
ii. To study the acoustical behaviour of FC filled with untreated and treated MM and PT woods.

1.5 Scope of research

In this study, polymer foam composites (FC) were developed based on crosslinking between polyol, flexible isocyanates with MM and PT wood as filler. MM wood was obtained from *Tukang Kayu A.Hamid Sdn Bhd*, furniture industry from Peserai, Batu Pahat. Meanwhile, PT was taken from Sime Darby *Berhad* from Kuala Lumpur. With a size of <355µm and <500µm for both woods, the FC were fabricate based on three percentages of filler loading which is 10, 15, and 20 wt % of polyol. These samples were then exposed using ultraviolet Weatherometer for 250, 500, 750, 1000 hours. The resistance of FC after ultraviolet exposures was detailed in sound absorption coefficient band. The color changing of FC was observed after ultraviolet exposures. Meanwhile, Fourier Transform Infrared (FTIR) was used to provide valuable information of functional groups present in FC. In addition, density and porosity was tested to measure the pore sizes and its effect on the sound absorption of FC samples. All instruments for testing an measurement have been done in laboratory at Universiti Tun Hussein Onn Malaysia (UTHM).

1.6 Significant of research

The significances of the research study included:

i. This research was done to improve FC with alternative filler for sound absorption especially for indoor and outdoor applications. This research

will determine whether this alternative is able to increase the physico-mechanical properties of FC that serves as sound absorber.

ii. The FC was developed using filler from biomass which is found abundantly on earth. This alternative was selected to replace the existing filler which mostly sources from chemicals which may effect on user's health.

iii. These UV irradiation exposures were conducted to determine the durability of FC after exposing in UV simulator for 1000 hrs in order to enhance the FC lifetime.

1.7 Thesis organization

CHAPTER 1 has highlighted the general introduction on this research, background of study, problem statement, hypothesis, objective, scope and its significance of research. It discussed the reason of research aimed in developing of FC using wood filler which can provide better sound absorption.

In **CHAPTER 2**, reviews of literature were focusing on wood filler, polymer, foam production and its parameter and sound absorption coefficient.

CHAPTER 3 shows the methodology that used to conduct the whole study. The technique of FC preparation, the physical and mechanical test is described in details.

CHAPTER 4 and **CHAPTER 5** cover the results and discussion of the experimental carried out from this research. The particles of FC are discussed in **CHAPTER 4**. It also shows the physical properties of the FC characterized using FTIR, SEM and density measurement. In **CHAPTER 5**, the results of sound absorption before and after fabricated of FC were discussed in details.

Table 1.1: Thesis organization description

CHAPTER	Description
CHAPTER 1	Introduction
CHAPTER 2	Literature Review
CHAPTER 3	Research Methodology
CHAPTER 4	Foam Production, Filler Characteristics and Physical properties
CHAPTER 5	Sound Absorption Behavior
CHAPTER 6	Conclusions and Recommendations

Lastly, **CHAPTER 6** summarized the results and discussion of all the experimental testing. At the end of this chapter, the recommendations are list out for future study. The entire chapter is illustrated as tabulated in Table 1.1.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

The literature review is relevant to formation of polymer foam composites, wood filler, as well as acoustical properties of polymer foam composite were reviewed in this chapter. It helps to point out some useful information regarding the research topic such as the development of foam with wood filler, the characteristics of polymer foam composite and the testing methods used for evaluated the potential of the FC for soundproof of indoor and outdoor applications.

2.2 Chemical structure of biomass

In general, utilization of biomass in lignocellulosic composites has been attributed to several advantages such as biodegradability and low cost. Biomass is composed of three major polymers- Cellulose, Hemicelluloses and lignin- and their ratio, composition and structure determine biomass properties (Kato,1981).

Cellulose is the most abundant renewable polymer resource available today, and it is considered an almost inexhaustible source of raw material for the increasing demand for environmental friendly and biocompatible products (Brinchi,*et al.,* 2013). Cellulose-based fibers are the most widely used, as biodegradable filler. Other researcher (Yang *et al.,* 2003) studied theabsorption coefficient of four fiber assemblies,cashmere, goose down and kapok, which are bothnatural and acrylic

fibers. The natural fibers haddistinctive internal structures that would influence thesound absorption coefficient. Woodsis one type of renewable biomass that widely use in furniture manufacturing and so on. Type of woods that been used is referring to Palm Trunks wood and *Meranti Merah* wood that generate during the maintenance and pruning of trees and plants. It is the most common type of woody waste available in Malaysia sawmills industry.

Biomass one of the renewable sources and attracts research interest in development of various process technologies. Wood is a biomass containing lignocelluloses will become one of the renewable resources for needs of mankind (Sun, *et al*., 2002). From previous study, the *Meranti* wood biomass contains cellulose, hemicelluloses and lignin as the major biopolymer (Roberta *et al.,* 1995). Figure 2.1 show the three major polymer structures.

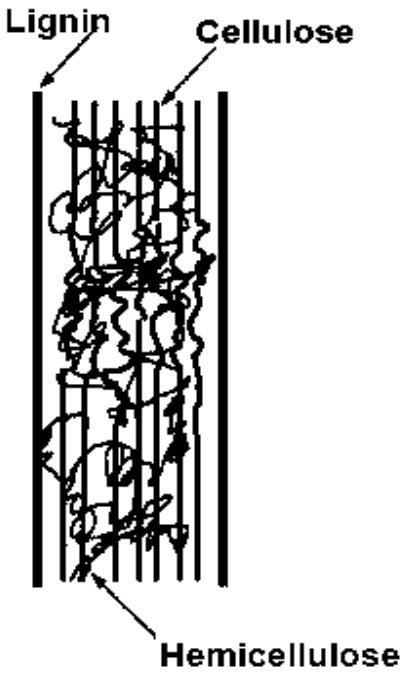


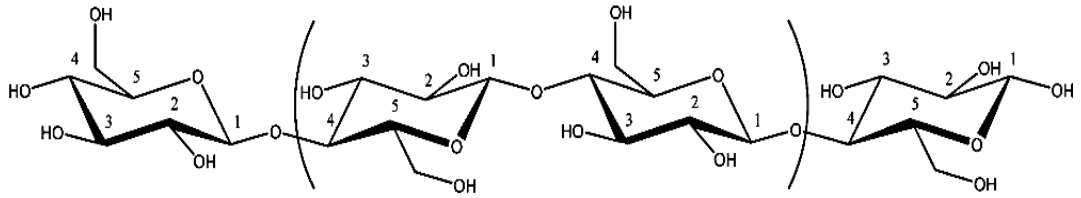
Figure 2.1: Structures of cellulose, hemicellulose and lignin in biomass.
(Roberta *et al.,* 1995)

The proportions of the constituent vary between different species. Hardwood has a content of cellulose and hemicelluloses around 80% of total feedstock dry matter while softwood contains around 70% of total dry matter (Balat, 2010).

An important field of cellulose application is in composites materials as reinforcement of engineering polymer systems (Biagiotti,*et al.,* 2004).Moreover,

properties of cellulosic fibers are strongly influenced by many factors, which differ from different part of a plant as well as from different plants (Siquera, *et al.*, 2010).

The chemical formula of cellulose is $(C_6H_{10}O5)_n$; n, calledthe degree of polymerization (DP), represents the number of glucose groups,ranging from hundreds to thousands or even tens of thousands. In the twentiethcentury, it was proved that cellulose consists of pure dehydrated repeating units ofD-glucoses (as shown in Fig. 2.2), and the repeating unit of the cellulose is called Cellobiose (Habibi *et al.*, 2010).



Cellobiose

Figure 2.2: Chemical structure of cellulose nanocrystals. (Habibi *et al.*, 2010)

Previous study by Rozman *et al.*, (2003), has reported that rice husk (RH) with different particle size shows filler tightly embedded in the PU matrix. It is clearly seen that the filler possesses a higher surfacearea in contact with the matrix. This indicates the existence of interaction between the RH surfaces with the PU matrix. This might be the result of the reaction between OH groups of RH with the NCO groups from MDI.

Miléo, *et al.*,(2011) reported that after the stage of alkaline delignification it is possible to observe a great number of free cellulose fibers, showing that the stage of pretreatment followed by delignification can provide a better availability of cellulosic fibers for subsequent processes, such as the use of the fiber as reinforcement in bio-composites.

Figure 2.3 shows the SEM for fracture surfaces of composites; fibers were welldistributed and aggregated to the matrix.Better performance of composites is attained due to homogeneous distribution of fibers in the matrix; as aresult the stress transference between fibre and matrix is more effective, affecting positively theperformance.

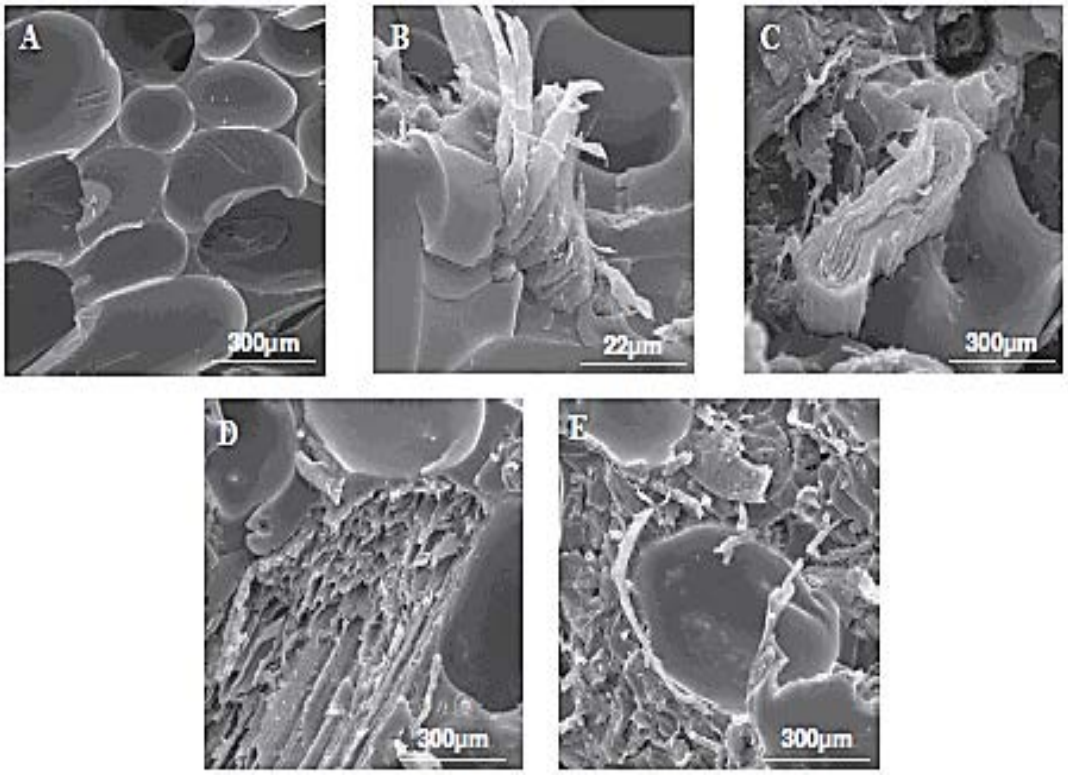


Figure 2.3: SEM of fractured surfaces of composites: (A) pure matrix; (B) 5% cellulose/ PU composite; (C) 10% cellulose/PU composite; (D) 15% cellulose/ PU composite; (E) 20% cellulose/ PU composite. (Miléo,*et al*., 2011)

2.2.1 Palm Tree

According to researchers, Malaysia is currently the world's largest producer and exporter of palm oil (Wahid *et al.*, 2004).Malaysian palm oil industry generates an enormous quantity of palm tree biomass, including palm frond, palm trunk, empty fruit bunch and excessive of fibers from palm oil production (Rasat *et al.,* 2011).

Palm tree is currently Malaysia largest agricultural plantation with the total of 4.69 million hectares with the life cycle of 25-30 years for productive oil palm. It is estimated that in dry weight there would be more than seven million tonnes of oil palm trunk wastage available per year (Chin,*et al*., 2011).

As referring Figure 2.4, Rahman*et al*.,(2014) has study the utilization of palm fibers for acoustic characteristics and proves that both fibers arepromising for use as sound absorber materials to protect against environmental noise pollution.

Figure 2.4: (a) Crude oil palm fibre, (b) chopping fiber. (Rahman *et al.,* 2014)

The comparison between acoustic properties of coir and oil palm fiber has proved the potential use of coir fibre in a sound absorption panel (Zulkifli *et al.,* 2009). In addition, the organic fibre is used in applications to decrease noise transmission in the space interior and external transmission (Zulkifli *et al*., 2009; Ayub *et al*., 2009).The advantages of these fibers are their renewability, abundance and low cost. These fibers are also more effective than industrial materials in terms of their reduced health hazards and protection during processing. Furthermore, this crude fibre can be used to reduce the noise emitted from power plants (Rahman *et al.*, 2014).

2.2.2 *Meranti Wood*

The studies on the use of *Meranti* wood as adsorbent are limited. Wood from Red *Meranti* species was chosen as raw material in this study because it is one of the most common and popular hard- wood species present in all tropical countries such as Malaysia and Indonesia (Rafatullah *et al.*, 2009 and Rafiqul *et al.*, 2012). To be name a few, Rafatullah *et al.,* (2009) and Ahmad *et al.,* (2009) has study the utilization of *Meranti* Wood as adsorbent for metal ions. Figure 2.5 shows the morphology structure of *Meranti* wood.

Meranti tree is widely used for furniture making and the waste wood fiber produced is generally used for heating in the boiler. Meanwhile, Saiful *et al.*, (2012) has stated that a small number of works has been carried out on Malaysian tropical Red *Meranti* wood and wood plastic composites(WPCs)fabrication.

A study of *Meranti* wood composition has been done by Shukla *et al.*,(2002). Lignocellulosic of *Meranti* wood consists of lignin, cellulose, hemicellulose and many hydroxyl groups such as tannins. All those components are active ion exchange compounds. Lignin, the third major component of the wood cell wall is built up from the phenylpropane nucleus; an aromatic ring with a three carbon side chain is promptly available to interact with cationic metal ions.

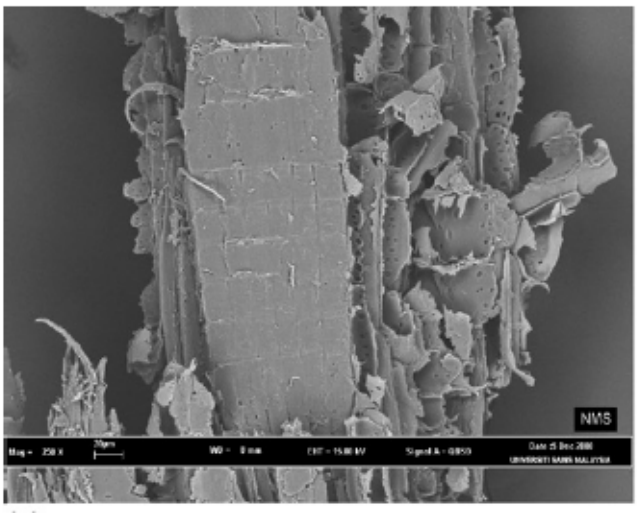


Figure 2.5: SEM of natural *Meranti* Wood. (Rafatullah,*et al.*, 2009)

2.3 Porous fibrous materials

Most of the porous sound-absorbing materials commercially available are fibrous. Fibrous materials are composed of a set of continuous filaments that trap air between them. According to Crocker & Arenas(2007), Fibers can be classified as natural or synthetic (artificial). Natural fibers can be vegetable (cotton, kenaf, hemp, flax, wood, etc.), animal (wool, fur felt) or mineral (asbestos). Fibrous materials consist of a series of tunnel-like openings that are formed by interstices in material fibers. Natural fibers are essentially completely biodegradable and modern technical developments have made natural fiber processing more economical and environmentally friendly.

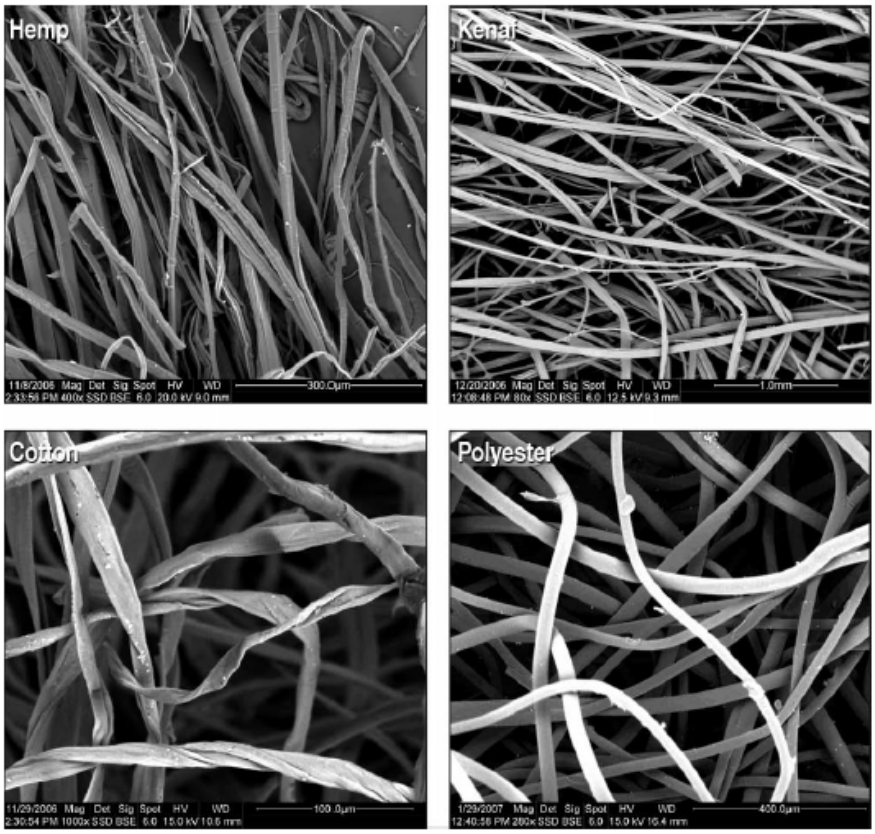


Figure 2.6: Scanning electron microscope images of samples of hemp, kenaf, cotton and polyester fibers. Courtesy of Dr. J. Alba (Polytechnic Univ. of Valencia, 2007).

Figure 2.6 shows SEM images of samples of hemp, kenaf, cotton and polyester fibers. It is evident that natural fibers have more irregular shapes and variable diameters compared to synthetic fibers.

Fibrous materials include those made from natural or synthetic fibers such as glass and mineral fibers Kazragis *et al.*,(2002).Synthetic fibrous materials made from minerals and polymers are used mostly for sound absorption and thermal isolation. However, since they are made from high-temperature extrusion and industrial processes based on synthetic chemicals, often from petrochemical sources, their carbon footprints are quite significant.

Zulkifli *et al.*,(2009) has studied the usage of natural fibers in manufacturing sound absorbing materials. Fiber materials have acoustical properties equivalent to those of glass wool where the SAC value obtained for both materials is above 0.980 at frequency range of 1000 to 4000 Hz, which is a good sound absorber (Koizumi *et*

al., 2002). These new methods may result in increased use of high-quality fiber at competitive prices for industrial purposes.

Studied by Yang & Li, (2012) and Allard, (2009) has proved that sound absorption mechanisms of the composite were the following three aspects: (1) when acoustic waves propagated into the fibrous material, the air in the fiber pores vibrated and rubbed against cell walls. The generated viscous resistance turned the acoustic energy into thermal energy attenuation. (2) The air in the pores was heated when compressed and it cooled when expanded. The thermal conduction in the materials made acoustic energy transform into thermal energy gradually and it was irreversible. (3) The vibration of fiber itself could also cause the dissipation of acoustic energy. These three aspects cooperated and worked together on the acoustic waves so that the acoustic energy was transferred.

Meanwhile, Green *et al.,* (1999) has study the wood materials lie in their microscopic structure i.e. fibre orientation, size, length and structure. On the other hand, Enamul *et al.,* (2014) stated that fiber is used to significantly improve many mechanical properties of polymer, but it may cause processing difficulties due to the nature of the fiber.

2.4 Chemistry in polyurethane (PU) foam

2.4.1 Polyurethane

Polyurethanes are all around us, playing a vital role in many industries because of their widely ranging mechanical properties and their ability to be relatively easily machined and formed as plastics, foams and elastomers. In particular, urethane materials, such as foams and elastomers, have been found to be well suited for many applications. Automobiles, for instance, contain a number of components, such as cabin interior parts, that comprise urethane foams and elastomers. Urethane foams are also used as carpet backing. Such urethane foams are typically categorized as flexible, semi-rigid, or rigid foams with flexible foams generally being softer, less dense, more pliable, and more subject to structural rebound subsequent to loading than rigid foams.

2.4.2 Flexible polyurethane

Polyurethane (PU) today account for the largest percentage (by weight and volume) of any plastics material used in automotive industry and their growth rate also faster than that of other plastics. Polyurethane is a synthetic polymer which formed by a reaction between a monomer containing at least two isocyanate functional groups and another monomer containing at least two hydroxyl groups in the presence of catalyst. The basic raw materials in producing of polyurethane are polyol and polyisocyanate. According to Hatakayema *et al.*,(1993), natural polymer having more than two OH group per molecule could be used as a polyol for polyurethane preparation if the groups could be reacted with isocyanate. Flexible and rigid PU foams are two predominant application forms of PU with coatings, sealants, elastomers, and adhesives being other common forms of applications.

The reaction is exothermic and then reaction heat can be used to form a cellular structure by evaporating the physical blowing agent. The foaming agent process can be explained by the nucleation and growth mechanism (Woods, 1990), Oertel, 1993), (Klempner*et al*., 1991). PU can be customized to its applications (Patrizia.*et al*., 2013). In the blow reaction (Figure 2.7), Water first reacts with isocyanate to produce a carbamic acid intermediate that quickly decomposes to give an amine and carbon dioxide. Carbon dioxide is the blowing gas which fills the cells. The amine reacts with a second isocyanate to form a urea linkage as a hard segment. Both the gel and blow reactions build molecular weight and therefore cause the polymer to gel, but the blowing gas is only produced from the water reaction(Triwulandari *et al*,. 2007).

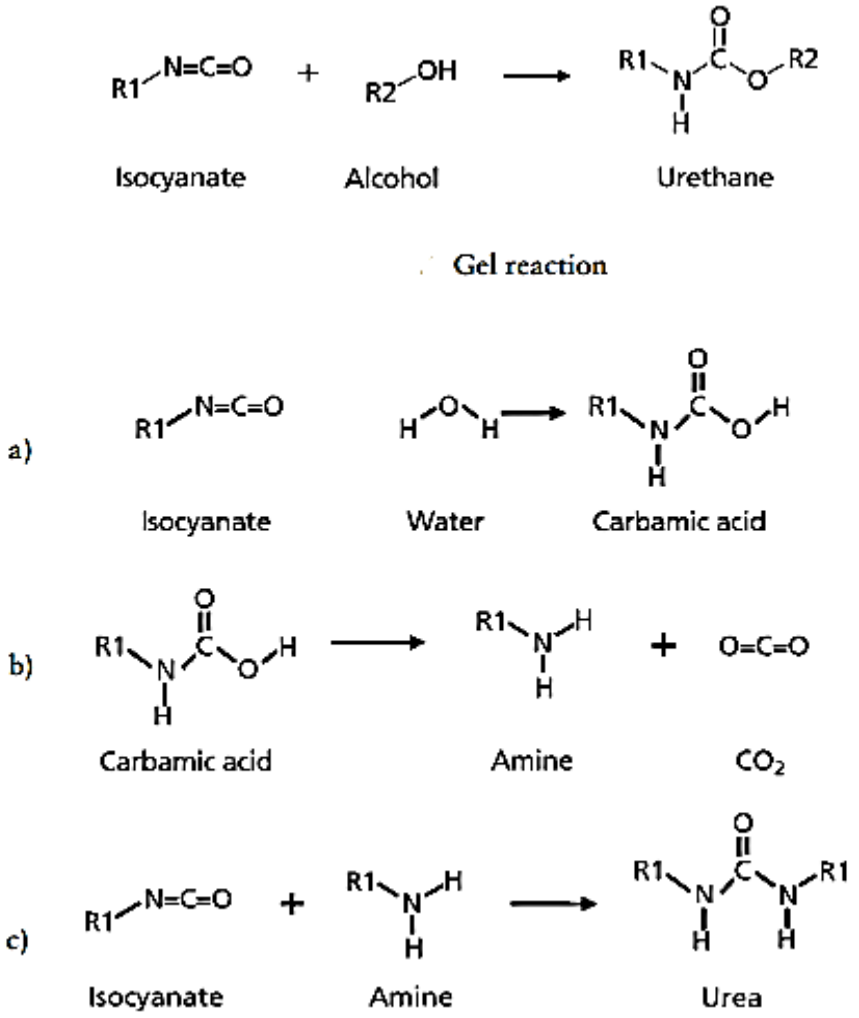


Figure 2.7: a) Blow reaction - first stage; b) blow reaction - decomposition step; c) blow reaction - urea formation.(Triwulandari *et al*., 2007)

Meanwhile, Ogunniyi & Fakayejo (1996) has proved that water, which is present in the formulations, reacts with diisocyanate to liberate carbon dioxide and gives the foam a cellular structure. It is also act as the activators are used to speed up the reaction.Water is added to these formulations to react with the isocyanate. This reaction ultimately produces polyurea, carbon dioxide and heat. This carbon dioxide diffuses to existing gas bubbles in the polyol and so expands the mixture into foam. Control of the amount of air contained in the polyol raw material is one way that manufacturers control the number of nucleation sites in the reacting mixture. These initially small bubbles quickly grow through either gaining gas from the diffusing carbon dioxide or by coalescing with other bubbles.

Previous study by Juan *et al.*,(2013), has indicates that PU is synthetic polymer which has molecule structure consist of block copolymer from hard segment

and soft segment. Hard segment and soft segment is produce from blow and gelation reaction of isocyanate. Hard segment are rigid structures that physically cross-linked and give the polymer its elasticity. By adapting the composition and the ratio of the hard and the soft segment, Figure 2.8 show the segments exist in polyurethane foam.

The soft phase segmentalrelaxation is strongly affected by changing hard segment content,akin to the role of increasing crystallinity in semicrystallinepolymers. Its strength is reduced and the relaxation broadensconsiderably with increasing hard segment fraction, demonstratingthat the relaxing segments in the soft phase exist in anincreasingly heterogeneous environment (Alicia *et al.,* 2011). A studied by Holden *et al.,* (1996) has proved that the hard domains serve as physicalcross-links in the soft matrix, leading to mechanical propertiescharacteristic of elastomers for materials with relatively low hardsegment contents.

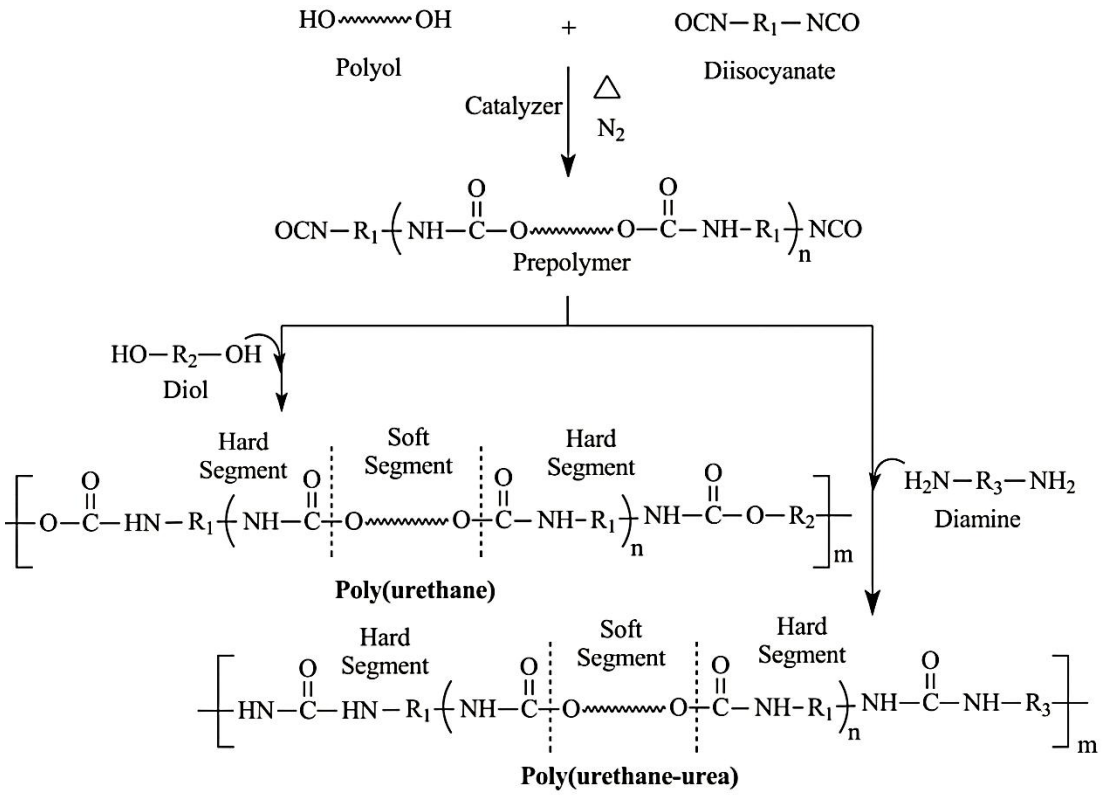


Figure 2.8: Schematic representation of PU foam. (Juan *et al.,* 2013)

PUs is the single most versatile family of polymers. Polyurethanes can be solid or microcellular elastomers (both cross-linked rubbers and thermoplastic

elastomers), foams, paints, fibers or adhesives. They can also be processed with most processing methods known at present.

PU foams account for the largest market among polymeric foams, estimated at nearly two billion kilograms in the US alone (Khemani, 1997).The versatility of polyurethane chemistry permits the production of a great variety of materials such depending on the initial ingredients used in the synthesis (Ligoure,*et al.,* 2005).

Furthermore, PU is one of the most important classes of polymeric materials due to its excellent mechanical properties, good abrasion resistance, high flexibility and damping property. The property of PU can be tailored chemically during the synthesis. Flexible PU porous foams with the open cells are generally considered to be good sound absorbers and vibration isolators and are therefore often used to improve the noise vibration and harshness comfort, commonly in automotive applications (Zhiping. *et al.,* 2012, Bakare *et al.,* 2012, Casati. *et al.,* 2008, Wang.*et al.,* 2004, Verdejo.*et al.,* 2009).

2.5 Raw materials of PU foam

2.5.1 Polyol

Polyol are either polyester, such as ethylene glycol, 1, 2-propanediol, 1,4- butanediol and diethylene glycol combined with glycerol or polyether, such as propylene glycol (PG) and trimethylolpropane (TMP) combined with sucrose. Polyether is used to produce flexible and rigid and polyester are used to produce elastomers, flexible foam and coatings. Structures of polyols made with either ethylene or propylene oxides are illustrated in Figure 2.9shows the common polyol used in polyurethane foam development(Oertel, 1985).

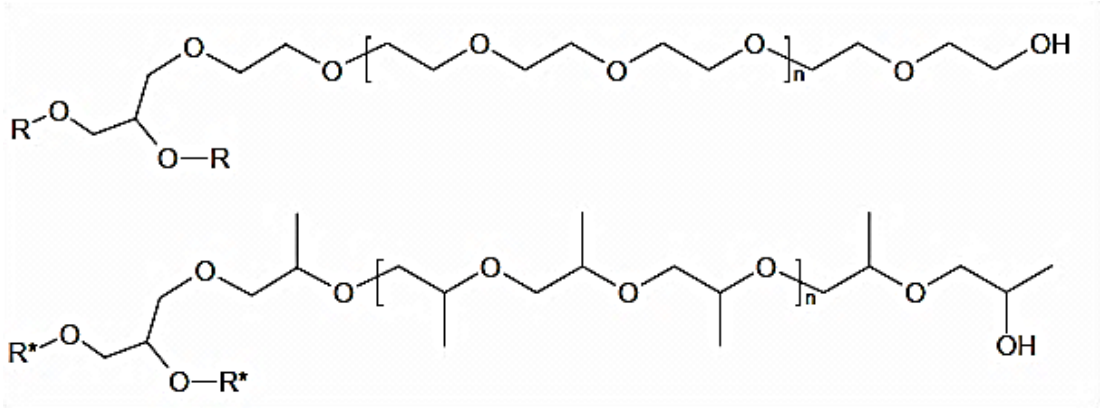


Figure 2.9: Polyether polyols made from ethylene oxide (top) and propylene oxide (bottom). The initiator used, for illustration purposes, is glycerol and R and R* represent the same structures as shown. (Oertel G, 1985)

Polyol from petrochemical derivative are commonly used until now. There are several polymer industry especially industry involve in production of polyurethane has gain interested to substitute this petroleum-based polyol with type of polyol based on renewable resources because of decreasing petrochemical supply while the price is increasing (Triwulandari *et al*., 2007). (Petrovic,2008, Narine, *et al.*,2007, Das, *et al*.,2009) has reported that since 1960s, a wide range of vegetables oils have been considered for the preparation of polyurethane; the most important oils are highly unsaturated oils, whereby using various chemical reactions, the double bonds are transformed into hydroxyl groups including, sunflower, palm, rapeseed, but mainly castor.

Other vegetable oils such as soy bean oil (Garrett. *et al*.,) peanut oil, and canola oil - contain carbon-carbon double bonds, but no hydroxyl groups. There are several processes used to introduce hydroxyl groups onto the carbon chain of the fatty acids, and most of these involve oxidation of the C-C double bond. Treatment of the vegetal oils with ozone cleaves the double bond, and esters or alcohols can be made, depending on the conditions used to process the ozonolysis product (Figure 2.10) (Narayan. *et al*., 2005).

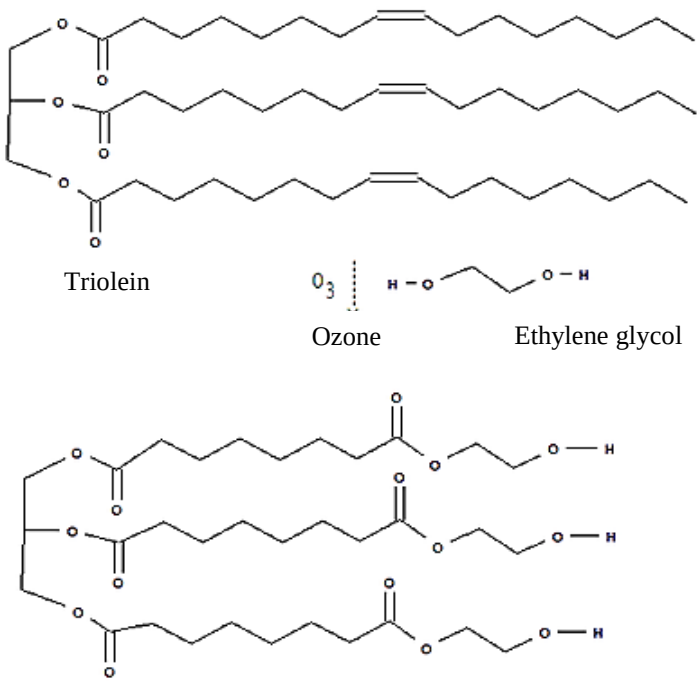


Figure 2.10: Reaction of triolein with ozone and ethylene glycol.(Narayan. *et al*., 2005).

2.5.2 Flexible isocyanate

Isocyanate is the functional group with a formula R –N= C=O. Two isocyanates that attached in a group are known as a diisocyanate. An isocyanate may exist one or more in groups. Isocyanate is commonly used in the manufacture of foams, electrical insulation and paints. Isocyanate is a basic ingredient for polyurethane foam production with the addition of water as a blowing agent. Common diisocyanate in foam formation is shown as Figure 2.11.

At present, for foam applications, only the production of polyol from renewable resources has been reported. Petrovic, (2008), claim that, although aliphatic di-isocyanate from dimerized fatty acids is commercial, they do not have sufficient reactivity for applications in foams, but they could be used for coating and other applications. Thus, isocyanate for PU foams must be aromatic.

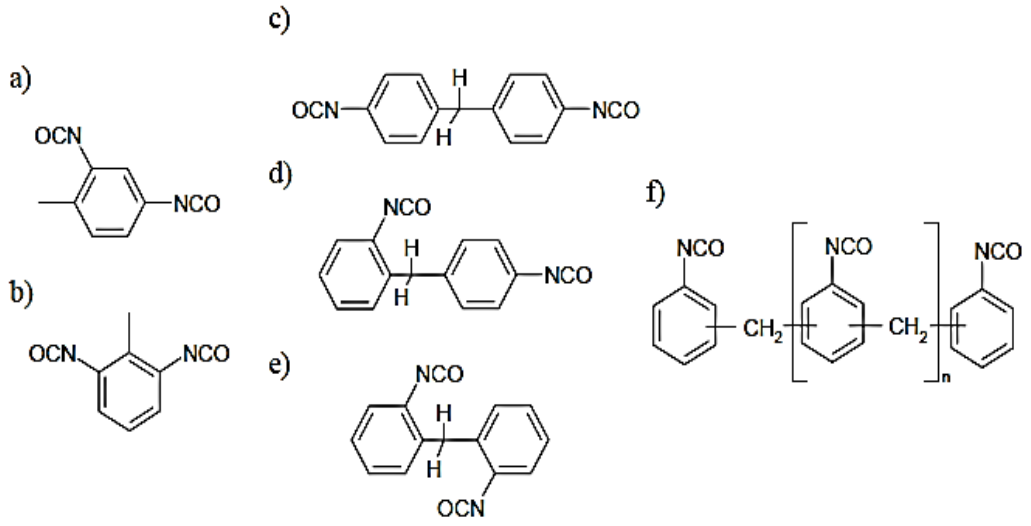


Figure 2.11: Diisocyanates commonly used in foaming: a) 2,4-toluene diisocyanate, b) 2,6-toluene diisocyanate, c) 4,4-methylene diphenyl diisocyanate, d) 2,4-methylene diphenyl diisocyanate, e) 2,2-methylene diphenyl diisocyanate, and f) polymeric MDICommon used diisocyanate.(Oertel G, 1985)

2.6 Covalent crosslink

Polyurethane foams, and most castable elastomers and reactive systems, are crosslinked systems. The chemical crosslinking in polyurethane is one of the following three types (Wirpsza, 1993):

(i) Crosslinking using mainly tri-functional compounds (polyols with more than two hydroxyl groups, low molecular weight triols) with di- or tri-isocyanate compounds. The isocyanate index is then closed to unity. The branching points are present in the carbon chain of the flexible segments, if this is a polyol chain or of the rigid segments, if this is a low molecular weight triol. The branching point may also occur at the nitrogen atom.

(ii) Crosslinking due to reaction of excess isocyanate groups (isocyanate index >100) with the urethane and urea groups of the polyurethane to form some branching allophanate and biuret groups in the rigid segments (Figure 2.12).

(iii) Crosslinking resulting from trimerization of excess isocyanate groups to give branching isocyanate rings.

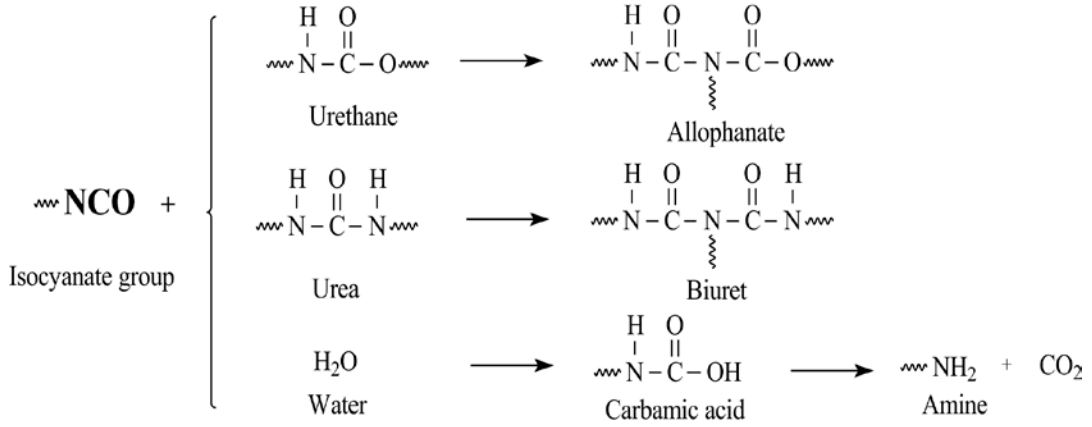


Figure 2.12: The secondary reactions of isocyanate. (Juan *et al.*, 2013)

Both reactions are potential crosslinking reactions, but the biuret linkage is of greater practical importance because urethanes are less reactive to isocyanate than are ureas due to hydrogen on the urea nitrogen is slightly more reactive than thehydrogen in urethane resulting in the biuret linkage having slightly more thermal stability than the allophanate linkage. Regardless, the biuret linkage is thermo-reversible and has temperature-dependent effects on the overall polymer properties (Juan *et al.*, 2013).

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