MODIFIED OIL PALM STARCH AS A POTENTIAL BINDER FOR PARTICLEBOARD

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MODIFIED OIL PALM STARCH AS A POTENTIAL BINDER FOR PARTICLEBOARD

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LIST OF SYMBOLS AND ABBREVIATION

CS_100	Corn starch with the 100 % ratio
DSC	Differential scanning calorimetry
DTG	Derivative thermogravimetric
FTIR	Fourier transform infrared
IB	Internal bonding strength
MC_70:30	70 % of corn starch : 30 % PVOH
MC_80:20	80 % of corn starch : 20 % PVOH
MC_90:10	90 % of corn starch : 10 % PVOH
MCS_70:30	70 % of corn starch : 30 % PVOH: 3 % SiO_2
MCS_80:20	80 % of corn starch : 20 % PVOH: 3 % SiO_2
MCS_90:10	90 % of corn starch : 10 % PVOH: 3 % SiO_2
MOE	Modulus of elasticity
MOP_70:30	70 % of oil palm starch : 30 % PVOH
MOP_80:20	80 % of oil palm starch : 20 % PVOH
MOP_90:10	90 % of oil palm starch : 10 % PVOH
MOPS_70:30	70 % of oil palm starch : 30 % PVOH: 3 % SiO_2
MOPS_80:20	80 % of oil palm starch : 20 % PVOH: 3 % SiO_2
MOPS_90:10	90 % of oil palm starch : 10 % PVOH: 3 % SiO_2
MOR	Modulus of rupture
OPS (S + LA)	Native oil palm starch (Sodium metabisulphite + Lactic acid
OPS (S)	Native oil palm starch (Sodium metabisulphite only)
OPS_100	Oil palm starch with 100 % ratio
PVOH / PVA	Polyvinyl alcohol
SEM	Scanning electron microscopy
SG	Solubility
SiO_2	Nano silicon dioxide
SP	Swelling power
TG	Weight loss
TGA	Thermogravimetric analysis
UF	Urea formaldehyde
XRD	X – ray diffractometry

KANJI KELAPA SAWIT TERUBAHSUAI SEBAGAI PEREKAT BERPOTENSI UNTUK PAPAN PARTIKEL

ABSTRAK

Kajian ini mengenalpasti potensi kanji kelapa sawit diekstrak daripada batang kelapa sawit untuk digunakan sebagai perekat bagi papan partikel. Dua kaedah pengekstrakan kanji telah dijalankan iaitu dengan kaedah merendam sampel kelapa sawit di dalam larutan 1000 ml yang hanya mengandungi 0.5 % *sodium metabisulhite* sahaja dan satu lagi kaedah iaitu merendam sampel kelapa sawit dengan 0.2 % *sodium metabisulhite* dan dicampur dengan 0.5 % asid laktik sebagai bahan katalis untuk mendapatkan hasil kanji yang lebih banyak. Kajian awal turut dijalankan untuk mendapatkan keadaan optimum bagi pembuatan papan partikel. Kanji jagung komersil telah digunakan sebagai perbandingan kepada kanji kelapa sawit di dalam kajian ini. Kedua-dua jenis kanji kemudiannya diubahsuai secara kimia dengan polyvinyl alkohol (PVOH) serta dicampurkan juga dengan asid borik. Nano silikon dioksida (SiO_2) sebanyak 3 % juga telah ditambah secara pengadunan fizikal di dalam kaedah pengubahsuaian kanji tersebut. Kedua-dua jenis kanji telah dinilai pada empat nisbah kandungan yang berbeza, iaitu, 100 % kanji asli; 90 % kanji dengan 10 % polyvinyl alkohol (PVOH); 90 % kanji dengan 10 % polyvinyl alkohol (PVOH) ditambah 3 % Nano silikon dioksida (SiO2); 80 % kanji dengan 20 % polyvinyl alkohol (PVOH); 80 % kanji dengan 20 % polyvinyl alkohol (PVOH) ditambah 3 % Nano silikon dioksida (SiO); 70 % kanji dengan 30 % polyvinyl alkohol (PVOH); 70 % kanji dengan 30 % polyvinyl alkohol (PVOH) ditambah 3% Nano silikon dioksida (SiO_2). Asas pencirian, sifat-sifat fizik dan kimia serta kekuatan papan partikel telah dianalisis dalam kajian ini. Keputusan kajian telah menunjukkan bahawa rendaman partikel kelapa sawit dengan *sodium metabisulfite* dalam masa 48 jam sudah mencukupi untuk mengeluarkan kanji daripada batang kelapa sawit. Pembuatan papan partikel dengan menggunakan tekanan panas yang diperolehi daripada kajian awal ialah dengan menggunakan suhu tekanan panas pada 160 °C selama 15 minit dengan 15 % penggunaan perekat berasaskan kanji. Walaubagaimanapun bagi pembuatan akhir papan partikel suhu 165 °C selama 15 min dengan 15 % perekat telah dipilih bagi mengimbangi perekat berasaskan jagung berdasarkan kepada keputusan analisis terma bagi sampel kanji tersebut. Kandungan lembapan, pH, kandungan kanji, amilosa dan amilopektin, kandungan kanji yang telah diubahsuai didapati telah mempengaruhi ikatan perekat. Kandungan protein, lipid dan kandungan abu yang tinggi telah menjejaskan kualiti kanji yang diekstrak daripada batang kelapa sawit. Kandungan protein yang tinggi juga menjejaskan ujian biodegradasi. Kandungan lipid yang tinggi telah menjejaskan prestasi pembengkakan papan partikel, ketelusan seta mengurangkan ketelarutan kanji. Kandungan amilosa yang tinggi telah meningkatkan keputusan modulus keanjalan (MOE) bagi sampel papan partikel, tetapi ia juga akan menyebabkan papan partikel menyerap air yang lebih tinggi dan memberi kesan terhadap suhu penyediaan perekat. Modulus keanjalan (MOE) juga didapati lebih tinggi pada kedua-dua jenis kanji asli berbanding kanji yang diubahsuai. Kandungan amilopektin yang tinggi telah menghalang kanji daripada mudah untuk dicairkan. Keadaan ini adalah disebabkan perekat berasaskan kanji sukar untuk menembusi ke dalam serat partikel. Pengurangan nisbah kandungan kanji telah berjaya menambahbaik ketahanan papan partikel daripada menyerap air, walaubagaimanapun ia telah mengurangkan sifat mekanikal papan partikel berkenaan. Nano silikon dioksida (SiO_2) telah berjaya meningkatkan sifat-sifat mekanikal dan berjaya mengurangkan masalah kestabilan

dimensi papan partikel. Ia juga menghalang papan partikel terutamanya papan partikel yang digam dengan 70 % kanji dan 30 % PVOH dari serangan serangga dan kulat. Pelepasan formaldehid yang lebih rendah telah dicapai dalam semua jenis papan berbanding papan kawalan (UF_100). Papan partikel yang digam dengan 15 % perekat, 70 % kanji kelapa sawit: 30 % PVOH serta ditambah 3 % Nano silikon dioksida (SiO_2) telah menunjukkan prestasi yang setanding di dalam ujian modulus keanjalan (MOE), modulus kerapuhan (MOR), pembengkakan ketebalan, penyerapan air, ujian kebolehbasahan, ujian pengorek, lebih tahan terhadap serangan organisma pengorek dan kulat, serta mempunyai nilai yang rendah bagi pelepasan formaldehid. Mikrograf SEM juga membuktikan bahawa struktur dinding yang mampat telah berlaku dalam sampel-sampel papan partikel. Ini menunjukkan bahawa perekat berasaskan kanji telah menembusi ke dalam partikel dan digambarkan mempunyai ciri-ciri kekuatan yang lebih baik terutama bagi sifat mekanikal dan sifat pembengkakan. Penciriran kanji juga menunjukkan bahawa kanji berasaskan jagung mempunyai sifat terma yang lebih tinggi berbanding daripada kanji berasaskan kelapa sawit. Ini dapat disimpulkan bahawa kanji jagung memerlukan tenaga yang lebih tinggi bagi memecahkan interaksi kimia di antara perekat berasaskan kanji dengan partikel berbanding dengan perekat kanji kelapa sawit. Walau bagaimanapun, di dalam kajian ini, tidak terdapat isyarat yang jelas di dalam analisis FTIR yang boleh mengaitkan kehadiran 3 % Nano silicon dioxide (SiO_2) bagi semua sampel kanji kelapa sawit dan kanji jagung yang telah diubahsuai. Modifikasi 80 % kanji jagung: 20 % PVOH: 3 % Nano silicon dioxide (SiO_2) merupakan perekat kanji berasaskan jagung yang paling cemerlang berbanding enam jenis ratio kanji jagung yang lain . Keseluruhan kajian telah mendapati bahawa kedua-dua jenis kanji kelapa sawit dan kanji jagung asli dan terubahsuai berpotensi digunakan sebagai perekat semulajadi untuk pembuatan papan partikel di masa hadapan.

MODIFIED OIL PALM STARCH AS A POTENTIAL BINDER FOR PARTICLEBOARD

ABSTRACT

This study investigates the potential of oil palm starch extracted from oil palm trunk to be used as binder for particleboard. Two types of oil palm samples were prepared, which was soaked with sodium metabisulphite only with 1000 mL of 0.5 % (w/v) in aqueous solution, and another type of sample used 0.2 % sodium metabisulphite and 0.5 % of lactic acid as a catalyst in oil palm starch extraction methods in order to get higher yields of starch collection. Preliminary studies were also carried out to get optimum condition for the particleboard manufacturing. In this study, the commercial corn starch was used as a comparison for oil palm starch performance. Both types of oil palm and corn starch were then chemically modified with polyvinyl alcohol (PVOH) and crosslinked with boric acid, respectively. Physical blending by adding 3 % of Nano silicon dioxide (SiO_2) in the modification method was conducted accordingly. Both types of oil palm and corn starch were evaluated at four different starch and PVOH ratios, such as 100 % native starch; 90 % starch with 10 % polyvinyl alcohol (PVOH); 90 % starch with 10 % polyvinyl alcohol (PVOH) plus 3 % of Nano silicon dioxide (SiO_2); 80 % starch with 20 % polyvinyl alcohol (PVOH); 80 % starch with 20 % polyvinyl alcohol (PVOH) plus 3 % of Nano silicon dioxide (SiO_2); 70 % starch with 30 % polyvinyl alcohol (PVOH); 70 % starch with 30 % polyvinyl alcohol (PVOH) plus 3 % of Nano silicon dioxide (SiO_2). Basic characterizations, physicochemical and particleboard strength properties were analyzed in this study. The results indicated that by steeping oil palm chips in sodium metabisulphite within 48 h is the optimum condition to extract starch from oil palm trunk. Manufacturing condition by hot pressing at 160 °C for 15 min with 15 % resin level showed the best performance in the preliminary study. However, the pressing time was set to 165 °C and selected in order to balance the corn starch sample and oil palm starch sample performance according to their thermal stability analysis. The moisture content, pH, starch content, amylose and amylopectin contents in modified starch were influenced by the adhesive bonding. Higher protein, lipid and ash contents were affected by the purity of starch extracted from oil palm trunk. The protein content was also affected by the biodegradation test. Higher lipid content affected the swelling and solubility performance and also reduced the starch solubility. Higher amylose content enhanced the modulus of elasticity (MOE) of the particleboard samples, but it also caused the particleboard to have higher water absorption and affect to pasting temperature. The higher starch ratio has a significant influence towards the particleboard properties. Modulus of elasticity (MOE) also has a higher value in both types of native oil palm starch and corn starch compared to their modified starch. Higher amylopectin in oil palm starch samples were preventing the starch from dissolving easily. This condition caused the starch based adhesive to be difficult to penetrate into the particles. Decrease in starch ratio successfully enhanced the water resistant of the boards but it also reduced the mechanical strength properties. Nano silicon dioxide (SiO_2) increased the mechanical properties and successfully reduced the dimensional stability of the particleboards especially in thickness swelling performance which was obtained the values below 50 %. Board bonded with 70 % starch and 30 % PVOH is also resistant to insects and fungal attack. Lower formaldehyde emissions were achieved in all types of the board compared to the control board (UF_100). Particleboard bonded with 15 % resin level, 70 % oil palm starch: 30 % PVOH plus 3 % of Nano silicon dioxide showed the optimum performance in MOE, MOR, thickness swelling, water absorption, wettability test, borer test, resistant to borer and fungal attack, and also have a lower value of formaldehyde emission. The SEM micrographs showed compact wall structures that have occurred in the particleboard samples. This indicates that starch based adhesive was well penetrated into particles thus reflected to have better properties especially for mechanical properties and thickness swelling performance. Starch characterizations also show that corn starch was higher thermal stability as compared to oil palm starch samples. This indicates that corn starch samples need more energy to break up their chemical interaction between particles and starch based adhesive compared to oil palm starch samples. However, in this study, there is no clear peaks that relates to the present of 3 % of Nano silicon dioxide in the mixture for all modified oil palm and corn starch samples appeared in FTIR analysis. Modified 80 % of corn starch: 20 % of PVOH: 3 % of Nano silicon dioxide (SiO_2) was shown the best performance amongst the others sample bonded with similar corn starch based adhesives. Overall study indicated that both types of native and modified oil palm and corn starches can be used as a potential green binder for particleboard in the future.

CHAPTER 1

INTRODUCTION

1.0 Background of the study

Wood based industry is contributing a return investment to worldwide economy. The particleboard industry in Europe and North America was predicted to increase by 1.48 % from 12,270 million m^3 to 12,454 million m^3 in Europe and 6.55 % from 1,098 million m^3 to 1,175 million m^3 in North America for the year 2014 to 2015 (UNECE, 2015). The increasing demand of wood composite will lead to challenge in the timber industry such as increasing demand of raw materials, wood adhesive and energy consumption (Douglas, 2011; Ivan, 2012). Malaysia also is contributing to this product approximately 546.318 m^3 or RM 371,153,495 for 2015 (MTIB, 2015).

Currently, most of the binders used in wood composite manufacturing are made from synthetic resin such as urea formaldehyde (UF), phenol-formaldehyde (PF) and melamine-formaldehyde (MF) (Liu & Li, 2007). Formaldehyde is one of the most widely used chemicals in producing this type of wood adhesive. The formaldehyde (CH2 = O) release especially from wood engineered product is a colorless gas with an irritating nasty odor (Ebrahimi et.al., 2014). These synthetic products are known as strong binders, water resistant and dimensional stable. However the formaldehyde emission released from this product during hot processing emits carcinogenic formaldehyde gas during their service life (Bertolini, 2009; Bertaud et al., 2012; Hashim et al., 2011; Nadhari et al.,2014). Therefore, some countries such as Turkey has set up their general standard for classified formaldehyde release for wood-derived boards stated as E0, E1, E2 and E3. However, the allowable formaldehyde release for dry particle board are stated as E0: 1-2 mg CH2O/100 g; E1: 2-10 mg CH2O/100 g and E2: 10-30 mg CH2O/100 g (Sofuoğlu et al., (2014) as cited in (Bozkurt, 2015).The standard threshold limit of 0.16 – 2.0 mg/l was generally applied in formaldehyde emission test when used the rubberwood particles with low molecular weight phenol formaldehyde (LmwPF) resin (Zaidon et.al., 2015).

Global market value has reported that adhesive and sealants were estimated to have a value approximately $43.36 billion in 2014 and expected to increase up to $59.75 billion by 2020, with an increment approximately 4.72 % between 2015 to 2020 (Anonymous, 2015). Urea formaldehyde (UF) is a common binder used in wood composite for interior purposes. This glue has a high adhesion strength and considered as a low cost binder especially for particleboard fabrication (Hashim et al., 2005; Ebnesajjad, 2009). Eventhough UF is categorized as an inexpensive adhesive and only used between 8% to 10% of dry weight, but it still contributes up to 60% of particleboard production cost (Hashim et al., 2005).

The depleting source of petroleum and awareness of health and environment impact have lead the global trend and forced the market place to move towards in using an adhesive with less or zero formaldehyde emission with a sustainable supply (Paster et al., 2003; Hashim et al., 2009). The continuous development of green technologies worldwide is also encouraging the development of natural based adhesive. Therefore, the wood composite industry has taken action to manufacture the adhesive derived from natural resources (Ashori, 2008). The United State of

America (USA), has started an incentive of 10 % tax credit or up to U.S. $ 500,000 for exporters who produced bio-composite for furniture and building materials from natural resources (MITI, 2011). A pressure from associations such as Greenpeace and NGOs as well as the increasing awareness on preserving and protecting the environment for future generations, has forced the government to take serious legislation on this issue. In 2012, starch world production was approximately 75 million tons, and estimated to increase up to 85 million tons by 2015 (Waterschoot et al., 2015a). Only a small fraction of the crops such as maize (7 %), cassava (4 %), wheat and potato (0.9 %) and 0.007% of rice are used for starch production (Waterschoot et al., 2015a). Starch was produced for non-food products such as paper and board, pharmaceutical, cosmetics and chemical (Chiu & Solarek, 2009). Presently, particleboard binding with bio-adhesives still have disadvantages such as higher moisture content, higher percentage of thickness swelling and water absorption as compared to Urea formaldehyde (UF) adhesive. Hydrophilic characteristics of the nature of starch itself will cause the bonding between particles and starch can easily be broken. Therefore, the panel bond with bio-adhesives is only suitable for interior application due to poor dimensional stability properties (Robyt, 2008). Starch was categorized as a carbohdyrate adhesive (Pizzi, 2006) and is still not well known as binder for wood composite (Thompson, 2005).

The oil palm starch yields can be extracted easily up to 20–25 % from oil palm trunk resources (Salleh et al., 2015). A total of 4 $\times 10^5$ t of starch are also reported to be derived from this material. Starch extraction from oil palm trunks not only contributed to the economy, but can also be used as a green adhesive (Noor et al., 1999; Salleh et al., 2015). Oil palm starch (OPS) can be commercialized as a new starch based adhesive that has the potential to be used in the particleboard industry in the future (Sulaiman, et al., 2013). The bonding properties of native starch was reported to have a low strength in gluing of constructional wood material (Imam et al., 1999). Therefore, the modification of starch is neccesary to improve the performance of starch-based wood adhesives thus it allows them to compete with petrochemical-based adhesives (Li et al.,2014). The use of oil palm starch used as binder in particleboard manufacturing was explored by other authors such as Sulaiman et al., (2013); Selamat et al., (2014); and Salleh et al., (2015). Eventhough, they reported that mechanical properties were successfully improved when using the modifed oil palm starch, the dimensional stability for these boards still needs enhancement. Therefore this research was conducted to enhance native and modified oil palm starch as future binders for particleboard.

In this study, it will propose to modify the oil palm starch with polyviynl alcohol (PVOH) crosslinked with boric acid that could enhance the adhesion between particles and starch based adhesive. Modification of other types of starch, PVOH and boric acid was generally applied in packaging product thoroughout the casting method. However, this is a new modification method used for oil palm starch source and applied as wood adhesive for experimental particleboard fabrication. Nano silicon dioxide (SiO_2) was reinforced into adhesive formulation by using only 3 % of this additive that could further enhance the dimensional stability properties such as thickness swelling and water absorption of the panel. Then, the commercial starch such as corn starch was used as a comparison for oil palm starch in this study.

1.1 Hypothesis

Wood composites using native starch as binder have low strength properties compared to synthetic glue. Therefore, through the modification process of oil palm starch it could enhance the physical and mechanical properties of the composites. Secondly, starch content ratio plays a significant influence in board properties. Thirdly, starch based adhesive can be proposed as a bio adhesive that can be applied for interior purposes and other possible application.

1.2 Objectives

This study was aimed to evaluate the potential of native and modified oil palm starch as a binder for particleboard. The specific objectives of this study are;

(1) To evaluate and compare the effect of modification process in oil palm and corn starch through physicochemical properties, adhesive properties and starch characterization.

(2) To evaluate the physical and mechanical strength properties of particleboard made from native and modified oil palm starch and corn starch compared to commercial glue such as Urea formaldehyde.

(3) To determine the optimum parameters of starch ratios when modified with polyvinyl alcohol and Nano silicon dioxide (SiO_2).

(4) To study the effect of Nano silicon dioxide (SiO_2) towards the particleboard strength, dimensional stability and biodegradation activities of such based particleboards.

CHAPTER 2

LITERATURE REVIEW

2.0 Wood adhesive in particleboard manufacturing

Particleboard was first commercialized during the World War II at a factory in Bremen, Germany (Kofi, 2014). This panel is widely used in building construction, furniture and packaging (Baskaran et al., 2015). An adhesive is a substance that adheres or sticks the two surfaces together without deformation or failure (Baumann & Corner, 2003). Adhesives can be categorized into two groups either natural or synthetic (Kennedy, 1989). Urea formaldehyde (UF) is a common binder used in particleboard manufacturing (Hashim et al., 2005). It is colourless, was a fast curing rate, hygroscopic, non-flammability; good thermal properties and easily adaptable to any curing condition (Pizzi et al., 2009). This type of adhesive normally undergoes permanent change when heat was used during hot pressing in particleboard manufacturing. It started being commercialized in 1937 (Kollmann et al., 1975). A good binder characteritics should meet the specific strength and fulfills the durability performance before being used in particleboard construction (Chapman, 2006) .

Factors such as higher temperature and longer curing time will be affected during the overall cost of production (Chapman, 2006). In order to create a greater interfacial contact between particles and adhesive, factors such as density of raw materials used, particles size and geometry (Chapman, 2006; Hashim et al., 2010), surface smoothness, temperature, good wettability and curing rate time should be considered in particleboard fabrication (Chapman, 2006). However, UF adhesive still has a disadvantage in its bonding that is due to moisture and relative humidity that was absorbed from the environment (Pizzi, 1994; Aydin et al., 2006; Baldwin, 1995). Due to this problem, the wood industry has urge that the adhesive suppliers to create a new value of adhesive formulation (Schwartz & Whistler, 2009) to tailor to the manufacturers demand (Gomez-bueso & Haupt, 2010). Adhesive distribution and adhesive properties have strong significant effect to the final product (Kollmann et al., 1975).

Wood adhesive from petroleum based chemicals affected indoor pollutant through utilization of composite panel products such as particleboard, fibreboard and plywood in furniture and building construction. Generally formaldehyde emission (FE) was released from this product. FE is colourless and has a strong smelling gas thus effected the human health and enviroment (Nakos & Athanassiadou, 2010). Therefore, a natural adhesive was found as an alternative to reduce the formaldehyde emission release to the environment (Papadopoulou & Chimar Hellas, 2009).

Natural adhesives also considered as bio-adhesives, can be obtained from tannins, lignin, carbohydrates and proteins (Pizzi, 2006). Previous study claimed that, only carbohydrates and proteins resources were most suitable to be applied as wood adhesives (Yang et al., 2006). Starch is one of the carbohydrate components that is being commercially used in various industries especially in the packaging industry (Mays & Hutchinson, 2005).Starch can be extracted from the grain or crop roots such as potatoes, corn, wheat, rice, yam or cassava (Masamba et al., 2001). Large quantities of starch which derived from various botanical sources are reported to have good technical and economic prospects (Emengo et al., 2002). Starch is also expected to be an excellent material for both food and non-food industries in the future (Ivan, 2012). Starch is an abundant, renewable, and cheap material extracted from multiple plant sources (Wang et al., 2013). Starch is used as binders, sizing materials, glues, and pastes in various products (Kennedy, 1989). Starch based adhesive need a higher temperature when used as a binder but the previous work by Wang et al., (2013) proved that it can be used in room temperature as well. Starch based adhesives with higher cellulose will improve the bonding strength (Moubarik et al., 2009).

2.1 Starch as new resource

Starch is a plant carbohydrate which is considered as reserve food for plants (Pizzi, 2006). It can be obtained from roots, seeds, tubers, stem, leaves and fruits of the plants (Vilieger, 2003) and widely being used in the food industry (Patil, 2012). Starch is a polymer of glucose units (Frihart, 2009)] consists of amylose [α-(1→ 4)-] and amylopectin [α-(1→6)-]. The amylose is a linear polymer ($M_w 10^5 - 10^6$ g/mol) whereas the amylopectin is a highly branched polymer ($Mw 10^7 - 10^9$ g/mol) (Vilieger, 2003; Thomas and Atwell, 1999). Most of the commercial starch available in the market contains 20 % - 30 % amylose, but this ratio can change from 0 % – 90 % (Swanson et al., 1993).

Starch is being used in various products including binders, sizing materials, glues and pastes (Jarowenko, 1977).The industries prefer starch because of its low price, attractive products, and suitability for any chemical modification and considered as an ideal product to replace synthetic polymers. The starch industry has been continuously increasing at a rate of 6.2 % and it is expected to reach $58.2 billion by 2019 in the global market (Ivan, 2012). The plant carbohydrate consists of polysaccharides, gums, oligomers and monomeric sugar and being used in adhesive formulation over the years (Pizzi, 2006; Wool & Sun, 2011). The carbohydrate based adhesives can be used either directly as an adhesive or as an addition in Urea formaldehyde (UF) / Phenol formaldehyde (PF) to improve their performance (Pizzi, 2003). Biodegradable polymers from starch are expected to be the most attractive research in near future (Ivan, 2012). The food and packaging industries are looking for a natural adhesive and an economical binder that alternatively comes from the starch resources. However starch still has many disadvantages in order to produce a bio-adhesive, and it is still not a popular choice, especially in the wood manufacturing process due to its hygroscopic nature (Thompson, 2005).

2.2 Starch extraction and modification process

Generally starch was modified to enhance its native properties for food and non-food industries. There were various methods in extracting starch from various resources. Sorghum starch was extracted by steeping in 0.25 % of alkaline solution (w/v) with the ratio of 1:5 (w/v) and stored at 4 °C for 24 h. The slurry was sieved with a sieve size 200 µm. The samples were then centrifuged at 3000 rpm within 30 min for four times until reached pH 7 before it was dried at 45 °C for 24 h. Approximately 97 % of sorghum starch purity was successfully obtained from this method (Sun et.al., 2014).

Mbougueng et.al., 2012 extracted tubers and roots by peeling, washing and cut into small sizes before milled by using a cutter (Manurhin, 03300 Cusset, no 426, France). The sample was sieved using a 170 mesh screen and washed for several times and dried at 45 °C before further used.

Awg-Adeni et.al., 2010 reported approximately 66 % of starch was successfully obtained from sago `hampas` after extraction process. Another study was the extraction of the Brazilian starch (BS) from the clean rhizomes by peeling it manually and blended with K2S205 solution (0.5 w/v) at 1:2 ratios. The slurry was then filtered with 60 mesh sieve to segregate the fibrous material. Then the supernatant was passed through a 200 mesh sieve after natural sedimentation for 3 h. The starch was washed twice before drying it on a Teflon tray and stored for flour production (Mahecha et.al., 2012).

Starch was extracted from fresh tubers cassava roots by washing, peeling and mixing it with 100 ml of 1 M NaCI. Then the samples was stirred for 2 min before filtered with muslin cloth and allowed to stand for 1 hour to facilitate starch sedimentation and then discard the liquid from the slurry. The starch was then dried for 24 h to 36 h before stored in a plastic container at a room temperature (Nuwamanya et.al., 2010).

Starch extraction from rice flour was done by soaking it in 5 equivalents (w/w) of 0.4 % NaOH solutions at room temperature for 48 h. The starch precipitate was washed with deionized water until it reaches pH 7. Then the starch samples were vacuum freeze dried until the moisture obtained 9 %. The starch was ground into powder and sieve through 100 μm sieve wire. Approximately 0.44 % protein, 0.61 % fat and 28.9 % of amylose was successfully extracted using this method (Zhang et al., 2014).

Starch extraction process from bun long taro tubers was done by washing the samples with distilled water and cut into small cubes in starch extraction process. Then one aliquot of 1000 g was crushed in a blender with 3000 ml distilled water. The slurry was filtered through four-layer gauze. Proteins and other non-starch materials were removed from the starch samples by repeatedly washing it with distilled water for several times. It was centrifuged at 5000 rpm speed within 10 min for three times until the supernatant became transparent. The starch precipitate was collected and dried at 40 °C. Approximately 8.4 % of amylose was obtained from this process (Zeng et al., 2014).

Wheat bran starch was extracted by blending approximately 5 kg of wheat bran in 10 volumes of water at 40 °C for 15 min in a stirred reactor. Then the starch was filtered twice to obtain a starch content lower than 1 % wt. of dry matter. The slurry was sieved before centrifuged at 3000 rpm. The starch sample was then dried at 45 °C before further used (Zeitoun, et.al., 2010).

Other study has extracted 150 g of legume seeds by soaking it in 500 mL water containing 0.01 % sodium metabisulphite for 18 h at 50 °C. The soaked seeds were then rinsed with distilled water before being grinded in a blender for 5 min. Then the grounded seeds were sieved through 100 µm before neutralized with 0.1M hydrochloric acid and centrifuged at 1500 x g for 10 min. The residue was dried at 40 °C and grinded before being sieved again with a 100 µm sieve wire size (Güzel & Sayar, 2010).

After the extraction process, starch was normally modified to enhance the positive attributes and eliminate the weakness of their native starch for various purposes. Most of the studies were used four types of starch modifications such as chemical, physical, enzymatic and genetical alteration of starch physicochemical properties. Some researchers did dual modification process by combining two methods for their starch conversion (Ashogbon & Akintayo, 2014).

The physical modification process is the simple and cheaper method compared to other conversion methods. It can be produced without any chemical or biological agents. Example of physical modification methods are superheated starch, iterated syneresis, thermally inhibited treatment, osmotic pressure treatment, multiple deep freezing and thawing, instaneous controlled pressure, drop (DIC) process, mechanical aviation with stirring ball mill, micronization in vacuum ball mill, pulsed electric fields treatment and corona electrical discharges (Ashogbon & Akintayo, 2014).

However chemical modification was used the chemical interaction with hydroxyl groups in starch granules to tune the specific applications due to alteration in their physicochemical properties. Chemical modifications can generally cause reconstruction of starch proximate compositions, gelatinization, retrogradation and pasting characteristics. It also promotes intra and inter-molecular bonds at random locations in the starch granules for their stabilization. There are five methods used in starch chemical modification which are acetylation, cationization, oxidation acid hydrolysis and cross-linking process (Ashogbon & Akintayo, 2014).

However, in this thesis the main focus is a crosslinking method which has generally altered the physicochemical properties of the native starch. The most common crosslinking agents are sodium trimetaphosphate (STMP), sodium tripolyphosphate (STPP), epichlorohydrin (ECH), and phosphoryl chloride (POCL3) being the most additive materials used in starch modification (Ratnayake & Jackson, 2008). Crosslinking process also reduced the moisture, protein and fat contents of the native starch when applied in banana starch modification (Garcia, Rivera, Montealvo, Montoya, & Pérez, 2009). Koo, Lee, & Lee, (2010) was crosslinked corn starch with STMP/STPP with the ratio of 99:1 (w/w) and found that crosslinking process has reduced the solubility and paste clarity of the corn starch samples. However, the crystallinity index in both native and modified corn starch samples shows a non-significant effect. This may be due to the crosslinking method that took place in the amorphous region and did not surrogate the crystalline pattern of both starches. The SEM results for this study showed a significance difference in starch granules shape size.

Lower solubility and swelling power were also found in wheat starch after the crosslinking process (Majzoobi et.al., 2009). Larger starch granules were also found in this study after modification process. Other researchers reported that modified starch have lower paste clarity and was more compacts when using the crosslinking method. Crosslinked native starch with hydroxypropylation and acetylation had increased the granule surface size and porosity (Singh et.al., 2004; Hung & Morita, 2005).

The viscosity of crosslinked rice starch decreased when the content of the crosslinking agent such as Epichlorohydrin (EPI), Sodium trimetaphosphate (STMP) and the mixing of STMP/STPP (Sodium tripolyphosphate) were used as crosslinking agents in this study. The tensile strength of rice starch films was reported to increase in line with the decrease in elongation at break and water vapor permeability. The crystalline peak structure and percentage of crystallinity of rice starch films also decreased when the crosslinking reaction decreased. Fourier Transform infrared spectroscopy (FTIR) analysis also confirmed that the crosslinking between starches molecules and crosslinking agents has occurred. The glass temperature (Tg) of crosslinked rice starch films obtained a higher temperature when compared to untreated rice starch films (Detduangchan et.al., 2014).

The high quality edible films produced, based on 80:20 waxy: normal corn starch which has demonstrated a valuable potential as a coating or packaging material in the food industry. The crosslinking method has successfully improved the interaction between the plasticizer and the modified starch. Lower moisture content, water activity, higher crystallinity, greater elongation under stress and higher thermal resistance were also observed in this study (Gutiérrez et.al., 2015).

2.3 Starch based adhesive

Starch based adhesive is a type of adhesive that comes from various starch resources such as corn, sago and potato starch. These types of starch was commercially used in the adhesives industry (Kennedy, 1989), however their bonding capacity is still a disadvantage for gluing wood surface (Zhang et al., 2015). Starches are inherently unsuitable for most applications and, therefore, must be modified physically and or chemically to enhance their positive attributes and/or to minimize their defects. Starch based adhesives can be prepared by either physical modification or chemical modification. Physical modification is a simple method by mixing in additive materials into the mixture whereas chemical modification involves the manipulation of hydroxyl group in the starches that can be utilized for specific purposes. All these approaches alters the physicochemical of starch properties and this modified starch can be attributes for food and non-food industries (Din et al., 2015). Therefore, Table 2.1 summarized the starch modification by various methods for the preparation of composite products and wood adhesive has been proposed by other researchers. This table also listed a few examples of physical and chemical modifying agents including nanosilica particles (SiO_2) polyvinyl alcohol, boric acid, and sodium dodecyl sulfate. Commercial types of starches such as corn starch, sago starch and potato starch have been studied extensively by using all these modifying agents in starch modification process.

Table 2.1: Previous studies on modified starches in composite manufacturing and wood adhesives.

Types of Starch	Type of Modification	Summaries	Reference
Corn starch	Polyvinyl alcohol(PVA)	- Improved mechanical properties but still poor performance in water resistance. - Considered as an economical and as an alternative adhesive in wood industry.	(Basta et al., 2013)
Corn starch	Silica nanoparticles (SiO_2)	- 10% of Nano silica is an optimum value to improve bonding strength and water resistant in rice straw composites. - Considered as an adhesive with excellent performance.	(Wang et al., 2011)
Sago starch	Polyvinyl alcohol(PVA)	-Successfully improved the young modulus but decreased in the tensile strength of polymer composites. -However it can reduce the cost of materials when only 40 – 50 % of sago pith bio filler was blended with polyvinyl alcohol.	(Yee et al., 2011)
Corn starch	Polyvinyl alcohol(PVOH)/ Latex and hexamethoxy-methyl-melamine (Cymel 323)	-Provide excellent mechanical properties in plywood product. -Successfully improved the moisture resistance. -Reduced the viscosity which allows easy application. -Resistance to fungi and microorganism. -Presence of an additive material such as wax could improve the performance in the future.	(Imam et al., 1999) (Imam et al., 2001)
Corn starch	Sodium-dodecyl sulfate	-Addition of sodium dodecyl sulfate (SDS) decreased the shear strength in wood adhesive. -Viscosity, mobility and storage stability could only enhanced when 1% and above of SDS was added to starch.	(Li et al., 2014)

Sago starch	2-dodecen-1-yl succinic anhydride/ Propionic anhydride	-When starch content increased, tensile modulus and water absorption also increased in polyester composites. -However, the tensile strength and elongation at break showed an opposite trend.	(Abdul Khalil et al., 2001)
Corn starch	Glutaraldehyde	-The modification had successfully improved the mechanical properties of particleboard but still need some development in dimensional stability by adding a water repellent material.	(Amini et al., 2013)

2.3.1 Corn starch adhesive

Corn starch is the most common type of starch studied for adhesive applications. Imam et al., (1999) has developed an environmentally friendly wood adhesive by crosslinking cornstarch and poly vinyl alcohol) (PVOH) with hexamethoxymethyl-melamine (Cymel 323). Citric acid and latex (UCar 443) were used as a catalyst and moisture resistance agent, respectively. The plywood prepared from this adhesive exhibits excellent mechanical property and comparable to Urea formaldehyde based plywood adhesives, and was suitable for interior applications. The prepared adhesive was easy to apply and needs optimum curing temperature and curing time of 175 °C and 15 min, respectively. The plywood exhibits improved moisture resistance and only 10 % failure was observed in glue line when completely immersed in water for 2 h (Imam et al., 2001).

Previous study by (Chiellini et al., 2001) was successful in fabricating packaging materials from agricultural waste fiber and corn starch adhesive modified by adding polyvinyl alcohol, hexamethoxymethyl-melamine (Cymel 303) and citric acid. The role of hexamethoxmethyl melamine (Cymel 303) is very effective in increasing resistance to water at high relative humidity (95 %). Corn starch adhesive was prepared by modification, using polyvinyl alcohol and nanosilica particles (SiO_2) as fillers to increase the water resistance of the packaging film (Huali et al. 2009). The nanosilica particles, were evenly dispersed in starch formed an intermolecular hydrogen bond (Huali et al., 2009; Xiong et al.,2008).

Previous work by Wang et al., (2011) reported that starch adhesive added with silica nanoparticle exhibits improved molecular structure, enhanced thermal stability, and beneficial changes in rheological properties of starch based wood adhesive as compared to starch adhesive without Nano silica cellulose. Addition of 10 % silica nanoparticles in corn starch increased bonding strength of the composite by 50.1 % in dry state and 84.0% in wet state, whereas the water resistance has increased by 20.2 %. In other studies, the corn starch adhesive using polyvinyl alcohol (PVOH) treatment, and fabricated with an economical rice straw based composite was prepared and the findings show that starch based rice straw composite has improved bending strength such as modulus of elasticity, and thickness swelling when compared to commercial Urea formaldehyde based composite (Basta et al., 2013).

Whereas other researchers reported the shear strength was decreased, while storage ability and viscosity of corn starch adhesive only increased when it was modified with 1 % sodium dodecyl sulfate (SDS). However, with the addition of 1.5–2 % SDS the shear strength was slightly reduced, while the mobility and storage stability of adhesive were significantly enhanced. It was concluded that SDS formed amylose–SDS complex which inhibited the aggregation of latex particles and starch retrogradation (Li et al., 2014).

Modification of corn starch with glutardialdehyde was prepared for particleboard made from rubberwood fibres. The panels exhibits enhanced modulus of rupture and internal bonding strength and satisfied acceptable limit of Japanese standard. However, the dimensional stability of the panels still needs an improvement such as by adding water repellent material (Amini et al., 2013). Boric acid was being used to crosslink starch and poly (vinyl alcohol) (PVA). The results exhibited that the Starch-PVA film has excellent film forming properties, with improved mechanical and water resistance properties (Yin et al., 2005).

Polyvinyl alcohol has proven a strong crosslinking agent with starch; however, addition of glycerol into corn starch paste reduced the tensile properties drastically (Sreekumar et al., 2012). While in another studies, with similar material replacing glycerol with glycerin and nano-SiO_2 exhibits improvements in the mechanical properties such as tensile strength, elongation at break and water resistance (Tang et al., 2008). Blending methylated cornstarch with PVA was prepared for packaging material. The result indicated that the products from these two mixing materials gave high water resistance compared to only starch/polyvinyl alcohol films. All the properties such as physical, mechanical and biological properties were improved significantly when the starch content was kept more than 50 % in the mixture (Guohua et al., 2006).

A packaging material was prepared in previous work by Follain et al., (2005) using a sol-gel method. In this method corn starch was added with polyvinyl alcohol and stirred at a constant speed of 500 rpm and constant temperature at 90 °C for 30 min. Glycerin was added to the mixture and was kept stirring for next 90 min before being casted in a plexi glass plate. The biodegradability analysis revealed that nanosilica (SiO_2) did not affect the biodegradability but it was related to the starch content in the mixture.

2.3.2 Sago starch adhesive

Sago starch is a stem starch extracted from various sago palm stems, especially *Metroxylon sago*. It is a major staple food of poor population in South East Asia such as New Guinea and Moluccas (Ahmad et al., 1999). However sago starch is cheap and economical, (Schirmer et al., 2013) and it has very limited commercial application. Recently, sago starch was studied to be developed as binder for cellulosic material or biodegradable packaging materials (Jamaludin et al., 2014; Bhat et al., 2013). Previously blended sago pith waste added with polyvinyl alcohol was used for the preparation of a polymer composite. The tensile properties of the composites were reduced, but showed an improvement in water resistance (Yee et al., 2011).

A biodegradable polymer of modified sago starch was prepared by esterified method using 2-dodecen-1-yl succinic anhydride (DSA)and propionic anhydride (PA) with 3 different types of solvent: N, N dimethylformamide (DMF), triethylamine (TEA) and toluene (TOU).The results showed that DMF was a better swelling agent in combination of PA and DSA for starch modification as compared to TEA and TOU. In general the esterified starch improved tensile properties of composites compared to the non-modified sago starch. The tensile modulus of prepared biodegradable polymer was improved when the starch content increased in unmodified starch or linear low-density polyethylene composites (Abdul Khalil et al., 2001). In another study, a polymer film for packaging material was prepared by blending the sago starch and polyvinyl alcohol which was cured under ultra violet (UV) radiation. Improved tensile strength was observed when sago starch and polyvinyl alcohol were blended in the ratio of 1: 2 with improved biodegradibility. However the high starch content increased the brittleness of polymer film, and promotes biodegradability of the product (Khan et al., 2006).

2.3.3 Potato starch adhesive

Potato starch is extracted from tubers of *Solanum tuberosum*, mainly consists of amylose and amylopectin (Schirmer et al., 2013). In comparison to other starches, it is rich in phosphorus found in the form of phosphate monoesters. Although, the modern developments in processing industry have increase in various application of starch, the traditional application of starch as adhesives still maintained their position (Qiao et al., 2014). Other researcher observed an improvement in thermal stability of potato starch treated with boric acid as compared to without treatment in packaging film. The microwave-assisted boration process caused esterification of starch with acid and new hydrogen bond formation which ultimately changed the macrostructure of film. The thermal stability of the modified borated starch increased with increase in degree of esterification (Staroszczyk, 2009). Potato starch film was prepared by incorporating a Nano silica dioxide with the help of plasticizing agent sorbitol/glycerol. The result showed that addition of silicon dioxide nanoparticles improves the overall functional properties of potato starch films with higher resistant to moisture (Torabi, 2013).

2.4 Oil palm trunk as a starch resource

An oil palm tree (*Elaeis guiness Jacq*.) originated from South Africa (Abdullah, 2003 as cited in Lamaming et. al., 2014). Oil palm trunk can be categorized as a monocotyledonous species (Lamaming et al., 2014). The five leading countries in producing oil palm products are Indonesia, Malaysia, Thailand, Colombia and Nigeria (Gourichon, 2013). In Malaysia, it was first planted in 1871 (Basiron et al., 2000). As the largest plantation crop in Malaysia (Shuit et al., 2009), this commodity contributed to Malaysia economic growth (Awalludin et al., 2015). Shuit et al., (2009) also reported that 1 hec of oil palm plantations can contribute approximately 50 – 70 tonnes of biomass waste, thus it makes Malaysia one of the largest biomass resource. The growth of oil palm plantations in Malaysia has increased exponentially until 20th century (Sulaiman et al., 2013). The statistic showed that Malaysia produced 42.3% of the worldwide oil palm production (Hosseini & Wahid, 2013). Oil palm plantation areas in Malaysia increased from about 3.0 % to reach 5.23 million hectares compared to 5.08 million hectares in previous year due to new plantation areas in Sarawak (MPOB 2013). Large areas of oil palm plantations have also contributed a singnificant amount of oil palm biomass especially oil palm trunk (Tay et al., 2013).

The giant government companies such as FELDA, FELCRA, RISDA and private estates such as Guthrie, IOI Plantations, Genting Plantations, and Sime Darby was contributed as top leading oil palm producers for Malaysia experiencing in new plantations and palm oil mills. Nowadays, the total area of oil palm cultivations by 2014 is 5.39 million hectares where the private estate has contributed a bigger percentage, approximately 62 % from overall total palm cultivations in Malaysia (Faizi et.al., 2016). As the industry becomes wider, oil palm biomass waste was overloaded and started creating problems such as disposal difficulties and increased operating cost. Increasing operating cost occurred when the operator has to convert oil palm waste to various products. Currently, this waste was converted to products such as an organic fertilizer, animal feedstock and soil conditioner to oil palm plantation (Awalludin et al.,2015).

Malaysia also promotes `green technology` by providing 4.69 million hectares of oil palm plantations in order to achieve this objective (Ng et al., 2011). The 25 years of oil palm trees or above is no longer considered as economical and is less suitable for cultivation (Hafizah et al., 2012; Jumhuri et al., 2014). These, uneconomical oil palm trees are cut down and being replaced by new plantations. The oil palm trees are either left abandoned or burned in the field but not utilized for any valuable products (Sulaiman et al., 2013; Tay et al., 2013; Hashim et al., 2012). Open burning contributed to significant pollution to the environment. Therefore, several work have been carried out to utilize abandoned oil palm biomass by converting it into other valuable products (Sulaiman et al., 2009; Hashim et al., 2010).

The starch based wood adhesive for composite industries has gain the interest of researchers to manufacture green products (Salleh et al., 2015; Amini et al., 2013; Sulaiman et al., 2013; Dastidar & Netravali, 2012). Eventhough, starch has been studied extensively for wood adhesive purposes but very few researches were done on oil palm starch (Selamat et al., 2014; Sulaiman et al., 2013). Characterization and analysis of physicochemical properties of raw starch are important parameters to be studied in order to understand the potential application for various purposes (Mweta, 2009). This will help to find the most effective method in adhesive application and reflect in enhancing productivity in the wood industry (Chow,1983). Recently, chemical modifications are a common method used in developing the starch properties (Waterschoot et al., 2015b).

Starch with appropriate properties is needed to produce renewable and biodegradable products (Ellis et al., 1998). Various research have been done to synthesise and analyse the physicochemical properties of starch obtained from oil palm trunk (Noor et al., 1999; Sulaiman et al., 2013; Tay et al., 2013). The chemical composition in oil palm trunk such as cellulose, hemicellulose and lignin differ from other hardwood and softwood species (Akmar & Kennedy, 2001). The starch of oil palm trunk generally stored inside in parenchyma cells of vascular bundles, which contained high lignin content (Sulaiman et al., 2009). After all, it still has a significant amount of starch in the oil palm trunk (Hashim et al., 2011; Noor et al., 1999). Approximately, 58 % - 81 % of starch was obtained from oil palm trunks (OPT) (Sulaiman et al., 2013; Tomimura, 1992). Normah et al., (1994) also supported the findings that starch content is variable along the oil palm trunk.

Highest starch content was found in the apical region of young oil palm trunk. The study also suggested using the samples that are closer to the trunk base to obtain higher starch content (Normah et al., 1994). Extraction of oil palm starch is