PROPERTIES OF COCONUT FIBRE BASED PARTICLEBOARD BONDED WITH MODIFIED POTATO STARCH

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by

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LIST OF SYMBOLS AND ABBREVIATIONS

%	percentage
°C	degree Celsius
°C/min	degree Celsius per minute
cm	centimeter
CAMPS	Citric acid-modified potato starch adhesive
FT-IR	Fourier transform-infrared spectroscopy
g	gram
g/cm^3	gram per centimeter cube
GDP	Gross Domestic Product
GH	Gross heat
GMPS	Glutardialdehyde-modified potato starch
h	hour(s)
IB	Internal bonding
JIS	Japanese Industrial Standard
KBr	Potassium bromide
LOI	Limited oxygen index
min	minute(s)
mm	millimeter
MDF	Medium-density fiberboard
MOE	Modulus of elasticity
MOR	Modulus of rupture
MPa	Megapascal
N/mm^2	Newton per millimeter square
NPS	Native potato starch adhesive
SEM	Scanning electron microscopy
TGA	Thermogravimetric analysis
TS	Thickness swelling

UF	Urea-formaldehyde
WA	Water absorption
W.H.O.	World Health Organization
XRD	X-ray diffractometry

SIFAT-SIFAT BOD PARTIKEL BERASASKAN SERAT KELAPA TERIKAT DENGAN KANJI KENTANG TERUBAH SUAI

ABSTRAK

Perekat berasaskan kanji kentang telah dikaji sebagai pengikat alternatif untuk papan partikel oleh kerana sifat reologinya, mesra alam dan murah. Dalam kajian ini, papan partikel dihasilkan daripada gentian kelapa dan diikat dengan perekat hijau yang berasaskan kanji kentang, perekat yang diubahsuai dengan asid sitrik serta glutardialdehid. Papan partikel dihasilkan dengan kandungan perekat yang berbeza iaitu sebanyak 10%, 12% dan 15% daripada berat partikel keringan ketuhar; dengan sasaran ketumpatan 0.8 g/cm^3 menggunakan tekanan 14 MPa dan pada suhu 145 °C selama 15 minit. Kelikatan dan kandungan pepejal perekat juga dinilai dan dibandingkan. Analisis kimia serat seperti kandungan abu, ekstraktif, holoselulosa, alfa-selulosa dan lignin telah dijalankan untuk menganalisis pengaruh mereka terhadap kualiti akhir papan partikel. Analisis FT-IR dan TGA bagi serat kelapa juga dinilai untuk menentukan kumpulan fungsian dan kestabilan haba bahan tersebut. Papan panel yang dihasilkan telah diuji untuk mengkaji sifat-sifat fizikal, mekanikal, terma dan perencat api. Mikrograf bagi pandangan keratan rentas sampel panel diperhatikan dengan Mikroskopi Pengimbasan Elektron (SEM). Indeks Oksigen Terhad (LOI) dan haba pembakaran kasar diguna untuk menilai potensi perencat api pada papan partikel berasaskan serat kelapa. Keputusan menunjukkan bahawa panel yang dihasilkan daripada kanji kentang yang diubahsuai dengan asid sitrik (CAMPS) mempamerkan sifat keseluruhan tertinggi. Contoh partikel yang dihasilkan daripada kanji kentang yang diubahsuai dengan glutardialdehid juga menunjukkan sifat mekanikal yang lebih baik berbanding dengan yang dihasilkan dengan kanji kentang asli tetapi dengan

sifat fizikal yang kurang baik. Beberapa papan partikel yang diuji berjaya mencapai Piawaian Perindustrian Jepun (JIS A-5908) untuk papan partikel Jenis 8, Jenis 13 dan Jenis 18. Berdasarkan hasil keputusan yang diperoleh dalam kajian penyelidikan ini, perekat mesra alam daripada kanji kentang yang telah diubahsuai dan penggunaan serat kelapa sebagai bahan mentah alternatif untuk partikel kayu dalam pembuatan papan partikel dapat memberikan kesan besar terhadap alam sekitar, industri papan partikel dan kelestarian pertanian.

PROPERTIES OF COCONUT FIBRE BASED PARTICLEBOARD BONDED WITH MODIFIED POTATO STARCH

ABSTRACT

Adhesives based on potato starch have been studied as an alternative binder for particleboards due to their rheological, environmentally friendly, and inexpensive properties. In this research, particleboards were manufactured from coconut fibre and bonded with green adhesives produced from potato starch, the adhesive was modified with citric acid as well as glutardialdehyde. The particleboards were produced at varying adhesive contents of 10%, 12%, and 15% of the oven-dry particle weight; at a target density of 0.8 g/cm^3 using a pressure of 14 MPa and a temperature of 145 °C for 15 minutes. The viscosity and solid content of the produced adhesives were evaluated and compared. The chemical analysis of the fibre including the ash, extractives, holocellulose, alpha-cellulose, and lignin contents was conducted to analyse their influence on the final quality of the particleboards. The FT-IR and TGA analysis of the coconut fibre was also evaluated to determine the functional group and thermal stability of the material. The panels produced were tested for their physical, mechanical, thermal, and flame-retardant properties. The micrographs of the cross-sectional view of the panel samples were observed with the Scanning Electron Microscopy (SEM). The Limited Oxygen Index (LOI) and gross heat of combustion are used to evaluate the potential of the coconut fibre-particleboard flame-retardant properties. The results showed that the panels produced with citric acid modified potato starch (CAMPS) exhibited the highest overall properties. The glutardialdehyde-modified potato starch particleboards also exhibited improvement on the mechanical properties compared to those produced with native potato starch but with less

desirable physical properties. A number of the tested particleboards have met the Japanese Industrial Standards (JIS A-5908) for Type 8, Types 13, and Type 18 particleboards. Based on the results obtained in this research work, an environmentally friendly adhesive from modified potato starch and the utilization of coconut fibre as an alternative raw material for wood particles in the production of particleboards will have a great impact on the environment, particleboard industry and sustainability of agriculture.

CHAPTER 1

INTRODUCTION

1.0 Research background

The high depletion rate of deforestation and petroleum resources affects the wood-based panel industry as the wood are inadequately available and synthetic resins are made from crude oil and gas. The price increase or fluctuation indicates a shortage of raw materials for particleboard production. Alternatives based on renewable sources are being sought. One of the alternatives is to use the natural bonding properties of lignin, a polymer found in almost any tissue of the plant. The coconut fibre contains a remarkably high lignin content (Jústiz-Smith et al., 2008). Thus, researches are being carried out on how to utilize this natural raw material in particleboard production.

Coconut, a botanical name of *Cocos nucifera L.*, is a multipurpose tree because of its exceeding features that have produced an extensive variety of usage in food, cosmetics, industrial goods. All over Asia, coconut is broadly spread, and Indonesia is its major producer in the world, accounting for more than 60 million metric tons (MT) (Burton, 2018; Yon, 2017). Malaysia is also one of the top 10 coconut-producing countries in the world with a statistic of 517,589 MT; it is the 4th significant industrial produce in Malaysia after oil palm, rubber, and rice (Burton, 2018). Due to its numerous importance, coconut received its description as a ‘tree of life’, and this made Malaysia put more efforts in boosting and growing the coconut industry (Ahmad et al., 2017).

Coconut fibre is an extract from the external shell of the coconut fruit, the plantations in Malaysia generates a considerable quantity of this waste without any application and are often dumped as agricultural wastes. Daily, these fibres generated are dumped in an unrestrained method causing environmental pollutions by increasing the already high volume of garbage in the dumpsite and occupying the much-needed landmass. (Kadir et al., 2016).

The methods used for removal of these agricultural wastes are by disposal into the landfill and by the combustion process, i.e. burning the wastes which is very dangerous to the environment and the ecosystem because of the carbon monoxide and the volatile organic compounds emitted from the burning process (Pariatamby et al., 2020). Burning these wastes is a very harmful and dangerous method, like open burning, in which during combustion, carbon monoxide and other toxic gases will be released to the air, and after the combustion process, the resulting ash will release gas discharge into the environment. Unrestrained combustion of agricultural wastes on the farm is illegal, as it could result in loss of lives and property, it can also lead to illnesses and diseases form pollution which might spread rapidly in communities (Angell et al., 2016).

Alternative methods must be used to control these problems. In the search for alternative methods to properly dispose of coconut fibre, various researches have been done for optimum utilization of coconut fibre into useful materials. One is the use of coconut fibre in the production of particleboard as an alternative for wood particles, making use of agriculture waste materials (coconut fibre) to improve the properties of the particleboards. (Fiorelli et al., 2019; Panyakaew et al., 2011).

Particleboard is a wood composite which is produced by compressing particles of wood and concurrently bonding them with a binder (adhesive) (Sarı et al., 2013).The particleboard furnish is obtained from a multiple of sources, the evolution industry resulted as the struggle for solid wood and its residues increased, to meet the demand of settling the large quantities of sawdust, planer shavings, plant residues and other waste that are generated from other various wood industries, as well as wood species not previously considered.

During the production process, the use of urea-formaldehyde as a synthetic adhesive has often been previously used, this adhesive is often produced using hazardous formaldehyde which is very toxic and carcinogenic to the detriment of the human health (Lamaming et al., 2019; Marutzky, 2018).

Coconut fibre was used to make particleboard with the aid of urea-formaldehyde resin as the adhesive by Khedari et al. (2004), this resin emits formaldehyde which is dangerous. Therefore, green adhesives (starch) is being introduced as a potential to replace the use of formaldehyde in particleboard production. However, from literature, particleboard produced with starch has been reported to have less desirable dimensional stability, which has resulted in the need to modify starch adhesives with materials that will improve their properties and be free from formaldehyde emission (Neelam et al., 2012). Therefore, research on the use of renewable natural materials to produce effective formaldehyde-free adhesive to produce the particleboard is increasing daily. The use of coconut fibre and modified potato starch-based adhesives in the study is proposed to see their potentials as alternatives to replace wood particles and synthetic adhesives; respectively. As they are both natural materials, a synergic effect is expected from the product.

1.1 Problem statement

Particleboards are supposed to be ecologically safe and cheap (Pawar et al., 2015). However, most of them are not; this is due to the use of urea-formaldehyde and other formaldehyde-based resins as a synthetic binder in a wood-based panel board. These resins had been reported by World Health Organization (WHO) as human carcinogen which has health-related risks such as cancer, burning sensation of the eyes, nose, and throat, coughing, nausea, and skin irritation on the end-users or customers of synthetic adhesives (Khanjanzadeh et al., 2019; Pang et al., 2018). The cost of using these synthetic adhesives further resulted in a high cost of particleboard production. Furthermore, the methods for disposal of coconut fibre are a great concern because the two most commonly used methods have lots of disadvantages that are dangerous to the environment, the human system, and the global warming phenomenon (Halvarsson et al., 2008).

Also, starch because of its chemical structure requires slight modification to produce a bioadhesive of desired qualities, to improve the physical and mechanical properties of the particleboards produced with it (Ferdosian et al., 2017). A lot of studies and researches have been done on finding alternatives to the use of synthetic adhesives and a potential replacement for wood particles, to the best of my knowledge and study of literature, no research has been done on the utilization of coconut fibre and modified potato starch for particleboard production. Thus, in this research, a study on the properties of particleboard produced using coconut fibre and potato starch-based bioadhesive.

1.2 Objectives

The principal objective of this study was aimed at the general characterization of the coconut fibre-based particleboard made using bioadhesive produced from the native and modified potato starch and their effect on the quality of the prepared particleboards.

The specific objectives are:

i. To evaluate the chemical analysis of the coconut fibre.

ii. To evaluate the properties of the bioadhesives produced from potato starch modification with citric acid and also with glutardialdehyde.

iii. To characterize the particleboards produced using the bioadhesives, in accordance with the standard. The characterizations include the physical properties (water absorption and thickness swelling), mechanical properties (internal bonding, modulus of rupture and modulus of elasticity), Fourier transform-infrared spectroscopy (FT-IR), scanning electron microscopy (SEM), x-ray diffractometry (XRD), differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), flame retardant properties (limiting oxygen index (LOI), gross heat of combustion), vertical density profile (VDP), dynamic mechanical analysis (DMA).

CHAPTER 2

LITERATURE REVIEW

2.0 Wood-Based Industry in Malaysia

In Malaysia, the wood-based industry encompasses four key divisions, namely the sawn timber, veneer, and panel products which consist of plywood and other rebuilt panel products like particleboard, chipboard, fireboard, moldings and builders' joinery and carpentry (BJC). The wood industry is predominantly owned by Malaysians and it is estimated that 80 to 90 percent of the companies comprise small and medium-sized establishments (Pang et al., 2015). They produce a wide-ranging array of furniture from office, kitchen, bedroom, dining room, sporting, living room, sofa, outdoor and garden furniture. The level of technology employed by the Malaysian furniture industry is similar to other countries that manufacture furniture because the country's manufacturers have invested considerably in machinery and equipment (Ng et al., 2017).

Before 1995, the furniture industry was branded by activities like logging, sawmilling, and plywood manufacturing. The industry has advanced further to embrace value-added processing such as furniture, medium density fiberboards (MDF), panel products, and veneer. The annual growth rate of the industry as well as its exportation of major wood-based products has recorded a steady increase since then (Nurkomariyah et al., 2019). Although, the existence of substantial growth in wood-based export was achieved, Malaysia is still a small producer in the global market for these downstream products, denoting that there is considerable room for expansion in the value-added products category. Malaysia is dynamically promoting the wood-based industry to ensure continued development in the future and as such, various strategies are being developed to assist the industry in all form feasible (Chandran et al., 2017; Welfle et al., 2018).

Malaysia is involved in outstanding economic growth and development, which can be attributed to the sales of natural resources and industrial products from them. These exported products are majorly inclusive of forest produce, confirming the major role the wood product industry plays in growing the economy.

2.1 Particleboard

Particleboards are merged composites defined as woody material that is bonded together using an adhesive. They are produced as boards from dried wood (lignocellulosic materials) particles mixed with a binder or other types of adhesive and are bonded together by the aid of pressure and heat in a hydraulic press machine. During the hot-pressing process, the internal bond by the particle in the form of adhesion is produced by the aid of the included binder, and other additives that were added during the manufacturing process to improve specific properties of the particleboard produced (Akinyemi et al., 2019).

The escalating demand for rubberwood, which is the most commonly utilized wood raw material for particleboard making, has caused the particleboard industry to seek alternative means to source for raw material supply (Selamat, 2014). New research and developments on the use of the abundant agricultural residues that are rich in lignocellulosic content will ease the total dependence of sourcing for raw material in the forest.

Taking advantage of the self-bonding property of lignin when the heat is applied to it at a high temperature, the lignin melts to the surface of the fiber; and with the aid of the adhesives used and pressure applied, the resulting particleboard is presumed to be highly dimensionally stable with good mechanical properties (Tajuddin et al., 2016).

2.1.1 History of particleboard

The history of particleboard begins from Germany, its originated place. In 1887, (Hubbard) made the purported "artificial wood" using wood flour and an albumin-based adhesive; this mixture was fused under high heat and pressure was applied. During the 2nd World war, Luftwaffe pilot and inventor Max Himmelheber played a role in making the first sheets of particleboard, phenolic resin was widely accessible in Germany, and was mixed with combinations of floor sweepings, wood chips, and ground-up off-cuts and glue (Timber, 2014). The first marketable piece was manufactured during World War II at a factory in Bremen, using waste material - such as planer shavings, offcuts, or sawdust- combined and milled with a hammer into chips and merged with phenolic resin. Hammer-milling requires smashing material into smaller and smaller pieces until they can pass through a screen (Lubi et al., 2007).

Since the early 1950s, particleboard because a huge sensation that influenced furniture designing as it was used in constructing cabinets, kitchen furniture, table-tops, shelves. It was however, more expensive than solid wood at that time because of the raw materials and technology in its production. As research and development of methods and techniques of particleboard were sought, it became cheaper as different strategies were applied by using less expensive materials and equipment.

The different grades and types of particleboards are based on the raw materials (wood particle and adhesive) and the technology used for production. Veneered, laminated, cement-bonded, melamine particleboards are the different particleboard types with differences in applications.

Till date, there are still safety alarms on the use of synthetic adhesives that are based on formaldehyde in producing particleboards, they are the main cause of formaldehyde emissions and are classified as a human carcinogen by World Health Organization (W.H.O.) (Khanjanzadeh et al., 2019; Kim et al., 2011).

2.1.2 Raw material of particleboard

Particleboards consist mainly of the wood particle and adhesive. It is differentiated from the other types of wood composites due to the raw materials used for its production. To produce particleboard, the most common type of wood particles used are softwood and medium density hardwood shavings, flakes, chips, sawdust, strands, fibers, and agricultural wood-wool (Tosi et al., 2019).

For the binder, synthetic adhesives, e.g. urea formaldehyde and phenol formaldehyde, are the most used, the adhesive level is often varying from 5-15% of the oven-dried weight of the board. The formaldehyde contained in the synthetic adhesive has been declared hazardous to the human system and the environment in totality, prompting the search for an alternative method of a renewable, formaldehyde-free, and environmentally friendly bioadhesive (Rathi et al., 2019). Green adhesives are being developed to serve as a potential to replace these synthetic adhesives, and starch is one of the natural materials that is being studied (Gonenc et al., 2019; Opara I. J, 2017; Salleh, 2014).

In a review paper, adhesives produced using different starches, such as corn, potato, wheat, rice, cassava and tapioca; and the various forms of modification used which includes, blending with urea-formaldehyde (UF), modification with acids (boric acid, citric acid) and other replacement forms for improving the starch rheological properties (Gadhave et al., 2017).

Also, the consistent deforestation resulting from rubberwood harvest by almost all sectors of the wood-based industry has led the particleboard industry to look into other alternative supply of raw material. Coconut fibre which is abundant in Malaysia has high lignocellulosic content, which seems to be a good potential as raw material for the particleboard industry (Khedari et al., 2004). This alternative should ease the demand from the forest and present an adequate utilization of this lignocellulosic waste that causes environmental pollution due to improper disposal methods (Mahieu et al., 2019; Pawar et al., 2015).

2.2 Particleboard manufacturing process

The general manufacturing process for particleboard is represented in Figure 2.1. The major stages of the production include drying, mixing with adhesives, mat formation, hot pressing, and finishing (Cunha et al., 2019).

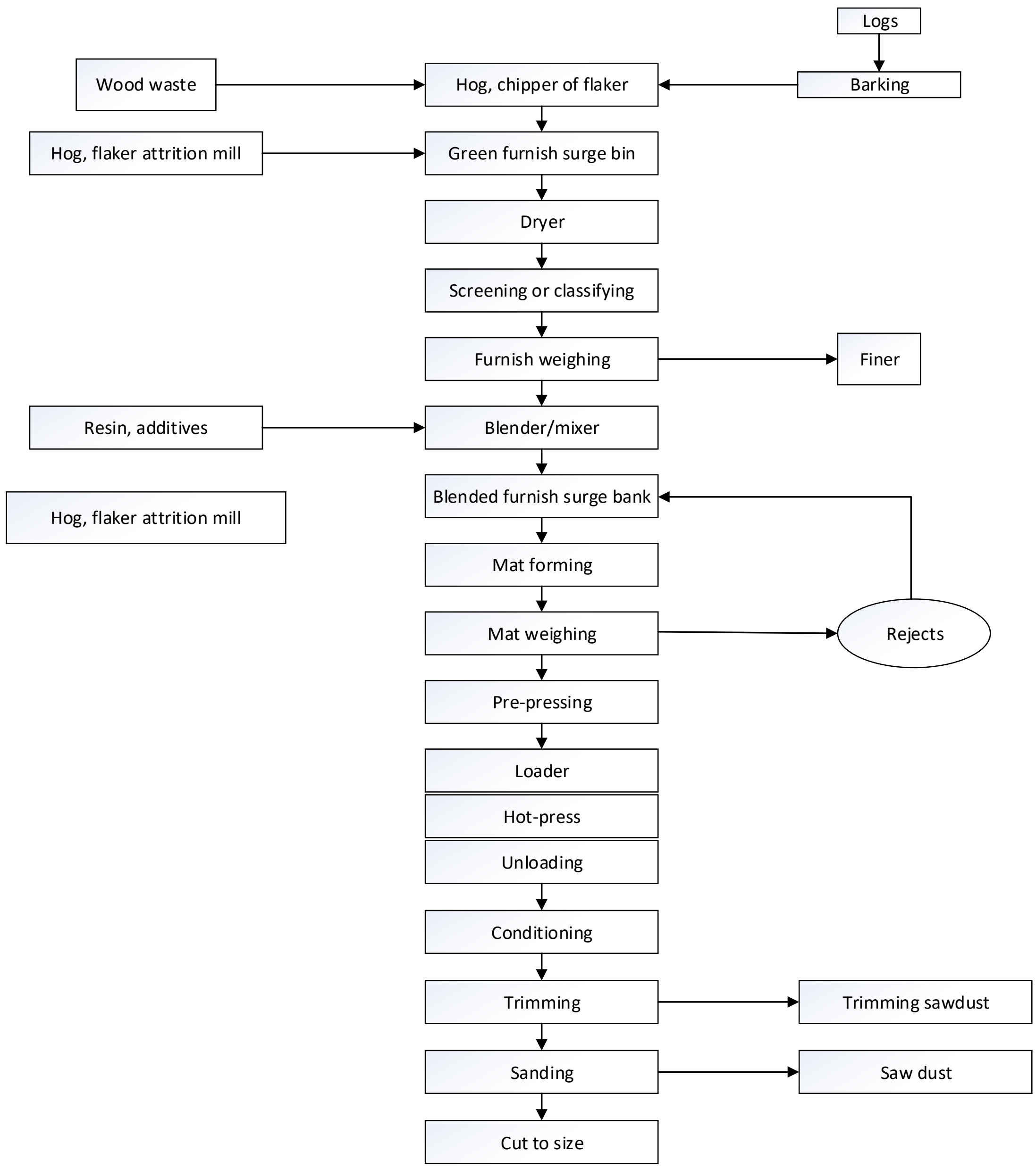

Figure 2.1: Process flow of particleboard manufacturing (Chapman, 2006).

2.2.1 Particle preparation and blending

The manufacturing process begins by mixing wood or (non-wood) particles and an adhesive and pressing the particle mix to form a panel. The particle furnishes to be used in the production often have gone through series of preparation before it can be used, it is obtained from different sources low-grade residues, in the form of hogged mill waste, sawdust, planer shavings. The furnish is separated by size. The bark is detached from logs of wood, stones, and metal particles are also removed with the aids of stone-traps and magnetic separators, respectively. Equipment such as chippers, flakes, hammer mills, knives, disc plates is used to reduce the sizes and geometry of the particle furnish to fit the desired core and surface layers of the particleboard to be produced. The particles undergo continuous drying to reduce the moisture content and are further screened by sizes (Hoque et al., 2007; Tosi et al., 2019).

The binder or resin is sprayed or mixed with the dried particles, in the production, numerous categories of binders are used. Urea-formaldehyde-based adhesives are the best from the view on cost and ease of use, however, melamine resins offer better resistance towards water absorption (Dodiuk et al., 2013). Adhesives based on the dry weight of particles are used and blended rapidly with the particle. For the production of the panel with a unique usage or specific qualification, additives such as wax, dyes, wetting agents and release agents, are added to aid processing or make the final product highly water-resistant, fire-proof, or termite-proof. (Hashim et al., 2011; Nemli et al., 2002; Pawar et al., 2015).

2.2.2 Mat forming and pressing

Upon thorough blending of the particles with the resin enough to bind the particleboard successfully, the particle mix is then made into a mat which will be flattened in a cold press (it is also referred to as pre-pressing) to reduce the thickness and release any air in it. After the cold compression, they are further compressed at a high temperature and pressure to reinforce the glue into the particle walls. The hot press temperature and pressing time differs based on the type of raw material used and particleboard being produced (Komah, 2018). The whole process is precisely monitored to certify the accurate physical and mechanical properties of the produced particleboard.

2.2.3 Board finishing

After the hot pressing, the particleboards are removed, they undergo further curing in the form of stacking, before cooling. They are then trimmed with the use of trimming saw to cut the particleboard to the specified size. They particleboards are further sanded with a knife planer to the specified thickness and good surface value (Stark et al., 2010).

Other finishing processes such as flame-retardant proof and moisture resistant application, decorative finish (lamination), and other value-added operations are done on the particleboard before packaging takes place (Waelaeh et al., 2017).

2.2.4 Properties of particleboard

The quality of the board is subject to factors like the size and nature of fibre or wood particle used, the quantity and efficiency of adhesive used, the density of the board, both preceding and or post-manufacturing treatments used, and so on. For the type of adhesive used, particleboards made using melamine-formaldehyde and phenol-formaldehyde are said to have higher dimensional stability than the particleboards produced using urea-formaldehyde resin as an adhesive (Wei et al., 2015).

The application of the particleboard is influenced by its density, low-density boards are less than 0.60 g/cm^3, while medium-density ranges from 0.6 g/cm^3 to 0.90 g/cm^3 and high density is higher than 0.9 g/cm^3 (Bao et al., 2016). Particleboards with advanced densities are simultaneous with higher strengths but with more problematic machining features, they exhibit poor dimensional stability, but with a higher cost per volume. For the low-density board, improved insulating characteristics are exhibited, better dimensional stability, lower strength, and lesser unit cost per volume (Ashok et al., 2018; Hashim et al., 2012).

2.2.5 Application of particleboard

Particleboard is regularly used as a construction material in building, furniture making, and other fields. They are of low cost and are used for both interior and exterior purposes. They exist in different grades as a factor of the particle sizes and types of adhesive used in the production. The intended application often determines the grade and type of particleboard to be acquired (Davies et al., 2017).

Particleboard can be applied in providing smooth and equal floor underlayment, to make particleboard interior stair treads. It is also used in constructing mobile home floor decking, cabinets, shelves, tabletops, kitchen furniture, speakers, tables, ceilings, bookshelves, and partitions (Monteiro et al., 2018).

2.3 Wood adhesives

Wood adhesives are principally applied in every of furniture production, there are used in more than 70% of wood products (particleboard, medium density fiberboards, OSB, plywood) in the world, they are employed in attaching furniture joints and wood composite materials and are important to retain the furniture structures (Ferdosian et al., 2017).

These wood adhesives are basically designed to bind wooden material of the same materials or with similar or different materials. The rise in the quantity of furniture required by humans influencing the particleboard needed to be produced which further leads to the high use of adhesive (resin). Thus, an efficient and qualitative adhesive that will hasten the drying time and fasten the production period to save the production time to meet up with the high demand of the particleboard (Ayrilmis et al., 2017).

Adhesive usage has more advantages than other fastening techniques, the advantages involve binding different material together, its cost-effective characteristics, and more artistic design methods. However, the drawbacks of adhesive usage involve diminishing stability when the heat is applied, and higher effort is required to detach objects while undergoing analysis (Solt et al., 2019). Adhesives can be classified based on the origin of the raw stock, either natural or synthetic origin i.e. adhesives are obtained naturally (organic) or produced synthetically.

2.3.1 Synthetic adhesive

Synthetic adhesives are binders (resin) created by man and are not found in natural forms. Synthetic adhesives are produced based on thermoset, thermoplastics. Common synthetic adhesives are epoxies, polyurethanes, polyimides, silicones, acrylics, polyamides, cyanoacrylates, polyacrylates, polyvinyl acetate (PVA), nitrile, neoprene polymers (Sancaktar, 2018). In 1920, Karlson Axis produced the first commercial synthetic adhesive (Sandström, 2014).

Synthetic adhesives comprise of urea-formaldehyde, melamine-formaldehyde, melamine urea-formaldehyde, phenol-formaldehyde, and other aminoplastics. They are more expensive than naturally-based adhesives because of their ability to give particleboard good mechanical strength, dimensional and thermal stability (Pizzi et al., 2017).

However, when they are used as adhesives in particleboard production, the products are affected by increased temperature, as formaldehyde gas is emitted from them (Khanjanzadeh et al., 2019). Formaldehyde was classified to be a carcinogenic substance by the International Agency for Research on Cancer (IARC), and it is therefore, a possible destructive material to the atmosphere and human body (Chong et al., 2019).

2.3.1(a) Urea-formaldehyde (UF) resin

Urea-formaldehyde (UF) is an unclear thermosetting polymer with a refractive index of 1.55. Urea-formaldehyde resins were commercialized in the 1930s, it is manufactured by heating urea and formaldehyde at 115°C for 5 hours (Dodiuk et al., 2013).

They are generally employed in particleboard, chipboard, MDF, and plywood manufacturing. Urea-formaldehyde resin has several characteristics that are advantages to producers in the industry, these include: low cost, curing conditions, low energy consumption during production, ability to dissolve in water, colorless, high tensile strength, microorganisms and scratch resistance, high heat distortion temperature and high water resistance (Tay et al., 2016).

Some of its disadvantages despite its numerous outstanding advantages; includes a deficiency in resistance to moist conditions, this moistness weakens the glue bonds, thereby diminishing the adhesion characteristics of the UF. Especially when combined with heat (increased temperature), the moist condition leads to a reversal of the bond-forming reactions (hydrolyzation), resulting in the release of formaldehyde gases which is the main disadvantage of using this resin as it is hazardous to the ecosystem and the environment (Marutzky, 2018). Therefore, particleboard produced using UF resin is suitable for indoor use only. It was also reported that during the processing of wood-based particleboards with urea-formaldehyde, one of the major difficulties faced is that uncured urea-formaldehyde is irritating and rather toxic, thus, it has to be used in a well-ventilated area (Ejiogu et al., 2018).

2.3.1(b) Melamine urea-formaldehyde (MUF) resin

Melamine-formaldehyde (MF) resin is a product of the polycondensation reaction of melamine and formaldehyde, resulting in methyl groups that may be up to six (Dodiuk et al., 2013). MF resins are significantly more expensive than phenol-formaldehyde (PF) or urea-formaldehyde (UF) resins (Conner, 2001; Norström, 2018). Melamine formaldehyde resins are mostly used in highly water-resistant applications, because of its advanced water resistance property, this feature which differentiates it from UF resins. MF is more expensive before the water-resistance characteristics, its price is about 2.5 times that of urea-formaldehyde. Consequently, the entrance of melamine urea-formaldehyde came from the need to negotiate within cost and performance; this resin is produced by varying the quantity of urea is added to the MF resin (Pizzi, 2003; Silva et al., 2015).

The MUF resins have valuable characteristics such as; hardness, rigidity, and abrasion-resistance, with exceptional water and solvent resistance property. The resins are self-extinguishing and have superior electrical properties. Like urea-formaldehyde, melamine-formaldehyde leads to formaldehyde gas emission which is hazardous to the ecosystem and the environment (Marutzky, 2018).

2.3.2 Natural environmental-friendly adhesive

In the furniture industrial sector, the second most important raw material is the binder used. Previously before the 1930s, the various adhesives used in the industry were derived from the crop (plant) and animals. The plant and animal glues were used before the synthetic resins came into existence (Norström et al., 2018; Pizzi, 2006).

However, animal glue has a difficulty against dimensional stability and resistance to hot water and the actions of microorganisms on it. The natural adhesives classified based on animal glue that is derived from animal skin and remainders of fish; raw blood which is obtained from the abattoirs; casein which is gotten from animal milk protein; and some mineral material in form of amber, sulfur, and paraffin (Ebnesajjad et al., 2014).

Naturally, adhesives gotten from botanical plants, these glues are attained generally by processing starchier plants and other gums of wood species. Now, formaldehyde is being added to both the animal and vegetable glue, to prevent microbes (Hemmilä et al., 2017).

Binders obtained from plants are categorized into groups of starch-derived and cellulose-derived adhesives. The starch-derived adhesives are, extracted from plants like corn, rice, potatoes, and wheat, while the cellulose-derived adhesives obtained from trees, cellulose, and shrubs. Natural materials that have the potential to serve as a basis for bioadhesives are basically of three types of polymer; starch, protein from plants, and lignin (Ferdosian et al., 2017).

2.3.2(a) Starch

Starch is a natural polysaccharide material that is gotten from seeds, roots, and leaves of plants. It is an advantageous raw material for developing bioadhesives because of its accessibility, cheap cost, excellent adhesion, and film formation properties. Potato, corn, wheat, sago, and rice are various plants that starch can be obtained from in large quantities. (Zhang et al., 2015). The chemical structure of starch is depicted in Figure 2.2.

However, the quality of the adhesives to be produced from it is determined by the quality of the starch. Thus, it is a natural polymer that is the mixture of two different polysaccharide parts, namely; amylose and amylopectin which are both are comprised of glucose with varying sizes and shapes. The amylose to amylopectin ratio varies from starch to starch and it is based on their originating plant species. The amylose forms a linear α-(1→4)-linked glucan, while amylopectin has an additional 4.2–5.9% α-(1→6) branch linkages to the α-(1→4)-linked glucan (Salleh et al., 2015). Amylose is insoluble in water while amylopectin is soluble in water. The amylose content has an effect on the expansion potential, crystallinity of amylopectin, and absorption rate of the starch (Zhang et al., 2015).

Due to the existence of hydrogen bonds among the starch molecules, the chemical structure of starch is a combination of loose, amorphous regions in highly crystalline regions. The crystalline regions inhibit the dissemination of water and other chemical materials into the structure, this leads to increased gelation temperature and a decreased reactivity of the starch. Thus, it is suggested to make some alterations to the crystalline region of starch or to reduce the size of the crystalline segments (James et al., 2009; Zhang et al., 2015).

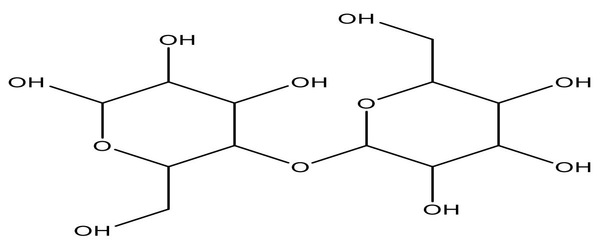

Figure 2.2: Chemical structure of starch (Robyt, 2008)

Several ways chemical and physical reaction that will reduce the crystallinity of starch is being studied and developed. The physical methods will majorly influence the granular structure, and they transform the natural starch to a cold-water-soluble starch. Such treatments like heat-moisture treatment (Boonstra et al., 2006; da Rosa Zavareze et al., 2011), annealing (da Rosa Zavareze et al., 2011), retrogradation, freezing, gelatinization (Hemmilä et al., 2017), thermal inhibition (Azwa et al., 2013), glow discharge plasma treatment (Zhu, 2017), osmotic-pressure treatment (Zia-ud-Din et al., 2017), and ultra-high-pressure treatment are not detrimental to the human application since no chemical was used for the modification.

For treatment such as esterification, crosslinking (Gonenc et al., 2019), oxidation, hydrolysis, acid treatment, dual modification (Neelam et al., 2012), there are chemical methods of modifications, and as such, both the physical and chemical characteristics of the natural starch changes because other functional groups will be included to the starch granules.

2.3.2(b) Protein

Plant protein is the third type of polymer that is a basis for adhesive, it is the type of protein obtained from plants or crops, the nitrogen-rich biological compounds which have large molecules composed of amino acids (Rathi et al., 2019). They are highly viscous with brief pot life. Due to their high sensitivity to water, they produce adhesives with weak water resistance. Therefore, it is essential to modify the protein's structure to obtain better mechanical and physical properties of the bioadhesives. Examples of plant proteins are soy, canola, cottonseed, wheat gluten, zein, peas (Chen et al., 2014).

Some researches were studied on developing protein plants, such as soy as an adhesive used in particleboard production. Various modification methods and chemicals such as; citric acid, urea, glutardialdehyde (Zhang et al., 2019), boric acid (Chalapud et al., 2020), NaOH, dicyandiamide (Chen et al., 2012), urea was used to improve the properties (physical and mechanical) of the produced particleboards (Cheng et al., 2004).

2.3.2(c) Lignin

Another polymer used to produce bioadhesive is lignin; it is a natural and green biological material that is readily derived from spent pulping liquors. It is an essential part of the cell walls of most landed plants and it is used as a raw material in making wood adhesives. Lignin has numerous properties that are suitable to be used in the production of adhesives and where high hydrophobicity is the most desired of its qualities (Ferdosian et al., 2017).

There are different types of lignin; there are; lignosulfonates, kraft lignin, organosolv lignin, enzymatic hydrolysis lignin, and soda lignin, they have been studied to determine their possibility in the synthesis of phenol-formaldehyde (PF) resin for wood adhesives (Ang et al., 2019; Ghaffar et al., 2014; Guo et al., 2015). An adhesive system for wood composites consisting mainly of lignin has yet to be fully developed.

2.4 Starch as an adhesive

Starch adhesives are inexpensive and readily available; they are can easily be applied after being mixed with water. They are produced and purchased as a powder, dissolved in water to form a moderately thick slurry. The adhesive gelatinizes and thickens when hot water or heat is applied to it and can be applied while hot or cold, and it is cured by the loss of moisture, i.e. removal of water, which is done often by evaporation. They have several advantages like readily available, generally of low cost, stable quality, no toxicity, readily biodegradable, excellent heat resistance, and insoluble in oil and fats.

Starch adhesives have disadvantages that include poor moisture resistance, mold growth, and a very slow curing rate. Table 2.1 gives the properties of a few commercial starches, giving their plant sources, their gelatinization temperature, and their amylose and amylopectin content. The values are obtained from the past investigation of Jane et al. (1999).

Table 2.1: Properties of commercial starches

Starch	Potato	Corn (maize)	Wheat	Rice	Tapioca
Source	Root	Seed	Seed	Seed	Root
Gelatinization Temp., °C	59-68	62-72	58-64	68-78	9-70
Amylose, %	25	28	25	19	20
Amylopectin, %	75	72	75	81	80

(Skeist, 2012)

Amylose which is one of the components of starch, is a polysaccharide composed of α-D-glucose units, and it is linked together through α(1→4) glucosidic bonds, it makes up approximately 20-30% of the starch composition. The chemical structure for amylose can be seen in Figure 2.3. For the industrial purpose, the amylose is an essential thickener, and it serves as an emulsion stabilizer, and gelling agent (Copeland et al., 2009).

A significant disadvantage is a decrease in stability and release of water when it undergoes crystallinity. Thus, the increase in amylose concentration decreases the gel stickiness while the gel firmness rises (Wang et al., 2017). Thus, starches with high amylose content, such as amylomaize, is planted because of the gel strength.

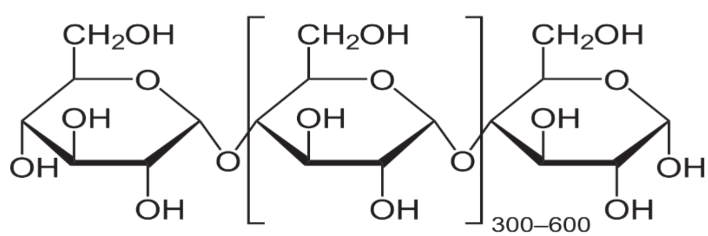

Figure 2.3: Chemical structure of amylose (Robyt, 2008)

Amylose is the smaller component of the starch molecule. The number of amylose molecule is almost 150 times of the other component (amylopectin). However, it accounts for only a quarter of the mass of starch granules in plants. The size of starch granules varies amongst the different plant species, rice starch has a reasonably small granule size of about 2 µm, while potato starches have a granule size of almost 100 µm (Bertoft, 2017).

Amylopectin is the other component of the starch; it is a highly branched polymer of α-glucose units found in the starch. The chemical structure of amylopectin can be found in Figure 2.4. The glucose units of the amylopectin are bonded directly with the α(1→4) glycosidic bonds, it can be dissolved in water and degrades faster than the amylose present (Pfister et al., 2016).

Some plants such as waxy starches like waxy maize, waxy potato starch, and glutinous rice are plant varieties that are void of amylose; they are entirely amylopectin. These starches have decreased retrogradation and produce a stable slurry.

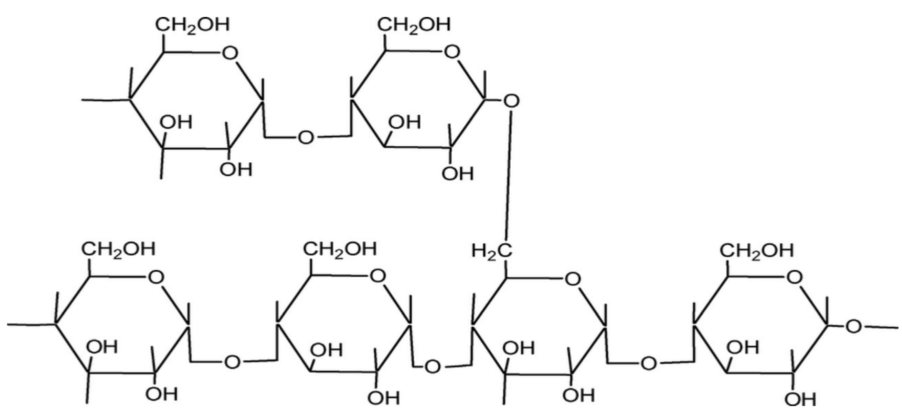

Figure 2.4: Chemical structure of amylopectin (Robyt, 2008)

2.4.1 Starch modification

Although starch has numerous advantages of being abundant, naturally renewable cheaply resourced, no emission of formaldehyde, with better adhesion and the most important its biodegradability. Of all the natural sources of adhesive, the starch adhesive has the most probability for advancement in the aspect of wood adhesives. However, the