Agricultural residues and the wood of umbrella tree (*Musanga cecropioides*) as raw materials for the development of reduced emission particleboards

Dissertation

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LIST OF ABBREVIATIONS

ANOVA: Analysis of Variance

IB: Internal bond

PB: Particleboard

PBs: Particleboards

MC: Moisture content

TS: Thickness swelling

WA: Water absorption

MOR: Modulus of Rupture

BS: Bending strength

MOE: Modulus of Elasticity

HCHO: Formaldehyde

UF: Urea Formaldehyde

PMDI: Polymeric methylene diphenly diisocyanate

w/w: weight on weight

NAF: None-Added Formaldehyde resins

MUF: Melamine Urea Formaldehyde

PF: Phenol Formaldehyde

OSB: Oriented Strand Boards

BMWI: “Bundesministerium für Wirtschaft und Energie“

DIN: “Deutsches Institut für Normung“

EN: European Norm

CARB: California Air Resource Board

VOC: Volatile organic compounds

BCR: Bean crop residues

UTW: Umbrella tree wood

CTP: Cacao tree prunings

SL: Surface layers

CL: Core layers

Bs: Bean shells

rpm: Rounds per minute

LVDT: Linear voltage differential transducer

ppm: parts per million

TMP: Thermo-mechanical pulp

LLMS: Lignin Laccase-Mediator System

LMS: Laccase-Mediator System

1.0 Introduction

The increasing global demand for wood as an energy resource as a result of the rapid development of the global economy and continues increase in world's population have led to a shortage of wood for the wood processing industry, especially the wood-based panel industry, which relied almost solely on low-grade wood and sawmill by-products. At the level of Germany, the amount of wood required by the wood-based panel industry is no longer available or is too expensive due to the increase in the thermal utilization of the so-called "industrial wood", spurred by the political support on the use of biomass through market incentive programs. These incentive programs are aimed at curbing the dependence on fossil fuel, which would make positive contributions to climate change (Weimer *et al*., 2012, BMWI, 2016). On another level, forest restructuring, measures which would ensure the regeneration of forests in Germany to achieve sustainable and stable mixed forests, which are better able to withstand climate change has also played an important role in the problematic raw material situation of the country's wood-based panel industry. The amount and the area covered by softwoods (spruce and Pine) preferred by the wood-based panel industry has declined over the years following the forest restructuring policy. The restructuring policies are aimed at making the conifers to appear as companion species to the deciduous species (less desired by the wood-based panel industry) such as beech (*Fagus sylvatica* L) (Hapla and Militz, 2008, Polle *et al*., 2008). According to Behrendt and Rupp 2006, the present stock of timber in the forests in Germany will not

be sufficient to meet the demand of the next decades. Bemmann (2013) forecasts a deficit in the provision of wood in Germany of 20 to 30 million cubic meters per year for the coming years.
On the global scale, Buongiorno *et al*. (2012) has estimated that by the year 2060, the global sawn wood and wood-based panel consumption would reach 800 million cubic meters. The overall high demand for the wood resource by both the energy sector and the wood product industry has led to a continuous increase in the wood prices in the global market. These have translated to global deforestation, thus contributed to global warming in the last decades. Consequently, the search for sustainable alternative raw materials for the wood-based panels industry has become the center of focus.

Various measures to guarantee a long-term supply of raw material to the wood-based panel industry have been suggested. For example, the recycling of post and pre-consumer wood products, increasing plantation sizes of fast-growing tree species with short rotation cycles, utilization of hardwood species and fibers from annual plants, etc. (Behrendt *et al.* 2007; Dohrenbusch and Bolte, 2008; Spellmann and Kehr, 2008; Youngquist *et al*., 1994; Bowyer and Stockman, 2001). Significant progress has been made in terms of research and development on the use of agricultural by-products and other non-wood fibers as a substitute for wood in the wood-based panel industry. Some of these include, Maize (Kharazipour *et al*., 2011), Hemp, canola, bagasse (Nikvash *et al*., 2010), rice husk (Gerardi *et* *al*., 1998), cotton stalks (Guler and Ozen, 2004), date palm fronds (Hegazy and Aref, 2010), wheat straw (Azizi *et al*., 2011), wheatgrass (Zheng et al., 2007). Some Plants are currently utilizing materials such as Kenaf, Bagasse, Bamboo, Hemp, Flax, Cotten, etc., in the production process of wood-based panels.

On the European level, about 238 to 311 million tons of agricultural by-products are produced each year. Of these, 46 to 61 million tons are produced in Germany alone (Nguyen *et al*., 2006; Deppe and Ernst, 2000). However, Germany's share is mainly used for renewable-clean energy for one obvious reason; which is the country's commitment to becoming carbon neutral by the year 2050. Nevertheless, the use of these raw materials is conceivable and ecologically sensible. While the wood-based panel industry in Germany is faced with the problematic raw material situation, in other countries, especially the under-industrialized countries in Africa with no established wood-based composite industry, exist huge quantities of alternative lignocellulose-containing raw materials from agriculture and fast- growing tree species. Some of these fast-growing tree species have the potential to be cultivated in plantations and to be used for the production of wood-based composite. To this end, the identification and the use of some of these raw materials for the development of particleboards is the focus of this work. Perhaps in the future investors from Europe and could establish small-scale particleboards plants in these areas where the raw materials are readily available to produce panels aimed for the European and the German Market.

1.1 Aim of the thesis

Given the current raw material situation for the wood-based panel industry in Europe and Germany in particular, the utilization of suitable agricultural residues and fast-growing tree species from other sources could be one of the means by which the composite panels industry in Germany could avoid the high wood prices and competitive situation in wood market. For this purpose, this study aims to investigate the possible feasibility to utilize the agricultural residues; Cacao tree (*Theobroma cacao)* prunings, the residues of the annual plant - common bean (*Phaseolus vulgaris*) and the wood of a fast-growing tree species - umbrella tree (*Musanga cecropioides*) from Cameroon as raw materials for the production of particleboards, with objective being:

- To develop new knowledge through the characterization of raw the materials.
- To investigate their feasibility to serve as raw materials for the production of particleboard for general-purpose applications used under dry conditions (DIN EN 312-2).
- To examine their feasibility to be used as raw materials for the production of particleboards with reduced formaldehyde emissions.

- To determine the extent to which the material could be suitable for the production of particleboards with bulk densities lower than the typical density of industrially-produced particleboards (650 kg/m^3) used for general purpose applications including furniture.

1.2 Problems statement

Increasing global population, rising income levels, the prices of substitute products, trends in consumer preferences and the prices of the wood products themselves are factors that have influenced the demand and supply of wood and wood products (Whitemann *et al*. 1999). These factors are responsible for the continuous increase in the prices of wood as raw material for the wood products industry (Kozlowski and Manys', 1997). On the other hand, the natural forest is a limited resource, home to a vast number of biodiversity, and houses some of the most remarkable ecosystems and habitats on earth. Considering the current threat on earth's life forms by climate change, it is undoubtedly clear that the forest must be protected. In the near future, none or very little industrial and recycling wood will be available for the wood-based panels industry, as many nations of the world will become committed to achieving carbon neutrality by abandoning fossil fuel consumption to stick with bio-energy. Giving the current population growth rate and the projected 9.7 billion inhabitants by 2050 (UN DESA, 2015), it is unarguably clear that the search for sustainable

alternative resources for the wood-based products industry is an inevitable option.
Another problem is the recent shift in consumers' preference towards lightweight furniture products especially in countries with small household sizes like Germany. Nowadays, consumers prefer the easy-to-handle lightweight products of high-quality standards at low prices (Poppensieker *et al*., 2005; Dauvergne and Lister, 2011). Lightweight wood-based products do not only benefit the consumer but also provide benefits for the manufacturer. Larger quantities of panels could be transported within the maximum weight limit on roads and railways, thus fuel and transportation costs savings can be achieved compared to panels of high-density. The reduction in the weight of wood-based panel products continues to have positive effects on all post-production and processing steps (Olhauser, 2005).

Formaldehyde emission from wood-based composites is another major problem that needs to be addressed in its entirety. Formaldehyde containing resins are the most commonly used binder agents in wood-based panels' products. These thermosetting resins such as urea-formaldehyde (UF), hardens to infusible macromolecule upon the application of heat (Kharazipour, 1996). However, the presence of free formaldehyde is obvious due to the reversibility of the aminomethylene link in the presence of water and moisture especially at high temperature, hence subsequent formaldehyde emission when hardened and in service (Dunky, 1997). Sax *et al*.

(2006) and Hun *et al*. (2009) concluded that formaldehyde was the highest contributor to the cumulative cancer risk from exposure to air contaminants that are typically found in residence. Formaldehyde was reclassified in 2004 by the International Agency for Research on Cancer (IARC) from being "probably carcinogenic to humans" to "carcinogenic to humans". Because of this, countries of the world such as USA and Japan have put in place stringent regulations (CARB Phase 2 and F**** standards respectively) to control the formaldehyde emission of wood-based panels used for interior applications. On the European level, formaldehyde was reclassified in 2014 as a carcinogen category 1B. The new classification was enacted into law in June 2014, after being published in the Official Journal of the European Union. This new law allows for the maximum value of 0.1 ppm of formaldehyde emission from wood-based panels used for interior applications when tested by the European chamber method (DIN EN 717-1). In reaction to the strict formaldehyde emission regulations, the resin suppliers, as well as wood-based panel manufacturers, had sought to remain competitive by developing techniques to reduce formaldehyde emission of their products. Some of these techniques include; the application of low molar ratio (F/U) urea-formaldehyde resin, the application of formaldehyde scavengers(e.g. urea), post-treatment of the produced wood-based panels (e.g. ammonia fumigation) or spraying of panels with formaldehyde reactive chemicals (Roffael, 1993). However, some of these techniques have negative effects on the physical-mechanical properties of the boards. For instance, the application of low molar ratio UF-resin decreases formaldehyde emission, internal bonding and bending strength but increases water absorption and thickness swelling of the panels (Sundin *et al*. 1987).

Formaldehyde-free binder systems such as polymeric diphenyl methane diisocyanate (PMDI) have only been largely used in the bonding of annual plants and crop residues in the production process of wood-based composites because of their ability to penetrate the waxy surfaces of these materials. Its cost has been the major setback to its wider industrial acceptance and use in most particleboard plants, except for the production of specialty products such as moisture-resistant boards reserved for niche markets (Sam-Brew and Smith, 2015). For example, PMDI today costs between 1.7 € - 1.8 € per kilogram compared to about 0.3 € per kilogram (based on liquid) for Urea-formaldehyde resin. Based on the huge differences in price of both resin, even when lower dosages of PMDI (3-6% based on dry mass of material) are used for panels manufacture compared to the UF resin (8-12% based on dry mass of material), price-wise, the PMDI bonded boards cannot compete in the market with the UF resin boards.

Table 1 gives an overview of the various standards and the different formaldehyde limit values for raw panels in comparison to the European test chamber DIN EN 717-1 method.

Table 1: Overview on formaldehyde limit values for raw panels (Egger 2014, modified)

Emission class	**E 1**		**CARB Phase 2**		**IOS-MAT 0003**		**F ******	
Testing method	European Emission test chamber **EN 717-1 (ppm)**[2]	Perforator value EN 120 (mg/100 g oven dry board)[3]	American Emission test chamber **ASTM 1333 E (ppm)**[1]	Comparative value European test chamber EN 717-1 (ppm)	American Emission test chamber **ASTM 1333 E (ppm)**	Perforator value EN 120 (mg/100 g oven dry board)	Desiccator value acc. JIS A 1460 **(mg/l)**	Comparative value European test chamber EN 717-1 (ppm)
Particle board	**0.1**	max. 8	**0.09**	0.065	**0.09**	max. 4	**0.3**	0.03-0.04
Thin MDF/HDF (< 8 mm)	**0.1**	max. 8	**0.13**	0.14	**0.13**	max. 5	**0.3**	-
MDF (> 8 mm)	**0.1**	max. 8	**0.11**	0.12	**0.11**	max. 5	**0.3**	-
OSB	**0.1**	max. 8	**-**		**0.09**	max. 4	**0.3**	-

[1]American emission test chamber method (ASTM 1333 E): Chamber volume min. 23 m³, varying loading factors, Temperature: 23°C, 50% rel. humidity, Air exchange rate: 0.5/h.
[2]European emission test chamber method (EN 717-1): uniform loading factor, Temperature: 23°C, 45% rel. humidity, Air exchange rate: 1.0/h.
[3]For factory production control.

1.3 Possible Solutions

The previous section has managed to depict the problems of the particleboard industry. These problems can be categorized under three main points; raw material scarcity, the shift in consumers' preference towards lightweight wood products and formaldehyde emissions. This section suggests possible measures by which these problems could be solved.

1.3.1 Alternative raw materials from other sources.

One of the measures to achieve win-win results of protecting the forest and guaranteeing the continued supply of raw materials to the wood-based panels industry is through the utilization of alternative biomass (agricultural waste). Nonetheless, as Germany pushes towards achieving a carbon-neutral economy in the coming decades, more and more quantities of biomass (wood and agricultural waste) will be demanded by the bio-energy industry. This means that the supply of these materials for the wood-based panel industry will become scarcer compared to today's supply. To overcome this foreseeable obstacle, there is a need to widen the scope of research for agricultural by-products and fast-growing tree species outside of Germany particularly in those countries with no established wood-based panel industry. This would allow the industry to reduce its reliance on it current sources of raw material supply which are local in scope and open up small-scale processing plants in areas where the raw material supply is readily available.

1.3.2 Lightweight particleboards

Great efforts have been made by the wood-based composite industry to reduce the density of panels without compromising the properties of the material. To produce particleboards of reduced bulk density (lightweight), various approaches, which takes advantage of the relatively low bulk densities of the raw materials, bean crop residues and the wood of the fast-growing tree species *Musanga cecropioides*, as wells as and the intrinsic strength properties of cacao tree prunings have been considered. One of these approaches involves the combination of the low bulk density materials (annual plant residues) with the relatively higher strength material (wood) to produce hybrid particleboards.

1.3.3 Particleboards of reduced Formaldehyde emission

Since the enforcement of formaldehyde emission laws in Europe, various resins have been formulated and advertised as low molar ratio urea-formaldehyde resins, and non-added formaldehyde (NAF) resins. This research identifies such resin, UF-resin (340) produced by BASF GmbH and polymeric diphenylmethane diisocyanate (PMDI) produced by the company Huntsman GmbH, which has been proven to effectively bond a wide range of lignocellulose containing materials. These resins have been evaluated in this research for their effectiveness as alternative binders to the higher molar ratio UF-resins to be potentially effective in bonding the new raw materials. In addition, a newly developed binder system based on animal protein, blood albumin, has also been evaluated for the same purpose. Also, reducing the quantities of resin required for wood-based composite production is generally not preferred since it leads to panels that do not meet the required standards (Sundin *et al.,* 1987). However, it is relevant to ascertain if minor reductions in the present dosing levels (2 - 3%) of PMDI used in particleboard manufacture could achieve particleboards that meet the relevant standards.

2.0 Background

2.1 The case of agricultural residues in the wood-based panel industry

The interest in using agricultural residues (annual and perennial crops) in the production of construction panels is not new. It dates back since the early 1900s when the first straw-based panels were first produced in Germany (Bowyer and Stockmann, 2001). Since then, numerous publications have already described the use of various residues from different agricultural plants as raw materials for the development of wood-based panels. In recent years, the diminishing wood resource on one hand and the increasing demand in wood-based panel products on another have spiked the need for agricultural residues as a raw material for the wood-based panel industry. Their use in this industry is even more reasonable because:

- They are fully recyclable and biodegradable; meaning that after completing their service lifespan they could be used to nourish the next generation of plants (Schell *et al*., 2004).

- The by-products of agriculture in many countries around the world especially in developing countries present an environmental challenge through methods of disposal. They are often burnt or dumped in landfills, none of which are eco-friendly. The use of these materials could ameliorate this environmental challenge and at the same time create income for the farmers.

- Annual plants are readily available and abundant worldwide (Wilson 1995, ILSR, 1997). They are renewed each year, thus the name annual plants and produce more cellulose per year than trees growth rings.

- Agricultural residues or annual plant fibers require less energy for drying and processing compared to wood (Mueller *et al.*, 2002), thus saving cost for the manufacturer. Their utilization would reduce the energy requirement of the industry.

- They exhibit good physical and mechanical properties such as high tensile strengths, good structural homogeneity, good thermal insulating properties and good acoustic properties due to their hollow tubular structures (Golbanaie, 2006).

- As already mentioned, lightweight wood products are generally preferred by consumers over their heavier counterparts. The low bulk densities of agricultural residues make them technically ideal for the production of panels suited for this purpose.

However, agricultural residues account for just about 5% of the total raw material used in the wood-based panel industries for particleboards production, the rest is accounted for by wood (Thoemen *et al.*, 2010). This is because the use of agricultural residues especially the seasonal crops is associated with some downsides which include:

- The problem of seasonal change makes it difficult to guarantee the continued supply of agricultural residues of the same quality throughout the year. For this reason, the materials have to be stored in bales for a long time at a moisture content of less than 15% to discourage biological attack. The extent to which the gluing-relevant properties of the raw materials, such as pH, buffer capacity and volatile acid content, are influenced by the duration of storage has not been sufficiently understood.

- Lignocellulose materials of Agriculture, such as straws, are generally harder to glue compared to wood materials. This is especially true for the bonding of straw with conventional condensation resins, like the acid hardening urea-formaldehyde resins (UF resins) and alkaline curing phenol-formaldehyde resins (PF resins). Even when the more expensive Isocyanate based binders such as polymeric diphenylmethane diisocyanate (PMDI) are used, one would need 10 to 20 times more binder to be able to achieve panels with properties compared to those made from wood under the same production condition (Wasyliciw, 2006). Thus, considerably reducing the price advantage when using annual plants or their residues as raw materials for wood-based panels' production.

- The relatively low bulk density of annual plants (e.g. straws) compared to wood makes high-speed transport of the mat along the production line onto the press systems problematic. Because of this, the production of straw particleboards on industrial scale has been limited to small capacity.

- The ash/silica contents of annual plants are generally much higher compared to wood (Nikvash *et al.*, 2013a). This chemical aspect negatively affects the bonding process as well as post-processing steps of the resulting panels, like the quick wear-out of sawblades.

2.2 Chemical characteristics of wood

Wood has always remained one of the most important natural resource to humankind because of its versatile applications, unique characteristics and comparative abundance (Miller, 1999b). Its unique properties such as strength, poor thermal conductivity, and malleability have made it unique for its wide range of applications. Wood is made up of about 50% carbon, 44% oxygen, 6% hydrogen and trace amounts of metal ions (Fungel and Wegener, 1989).

Woods are generally classified into two broad categories, hardwoods, and softwoods. The terms hardwoods and softwoods in the classification are not solely based on the softness or hardness of the wood, rather it refers to the deciduous tree which loses their leaves annually and the coniferous trees that usually remain evergreen throughout the year respectively. Both hardwoods and softwoods differ in their chemical composition. Wood being a complex and inhomogeneous material, its chemical components contained within its cellular structure vary from one tree to another and from one part of the tree to another. The variation in the chemical composition of the tree depends on factors such as age, origin, climate, soil (Han and Rowell 1996). All wood materials are made up of four main components, cellulose, hemicellulose, lignin, and extractives. All woods also contain small amounts of inorganic minerals (ash and silica). Since wood comprises about 90% (by weight) of particleboards, different volumes of these components present in wood (furnish) do have profound influence on the properties of particleboard.

Table 2: Chemical composition of hardwoods and softwoods. Source: Sjöström, and Alén, (1999).

Composition	Hardwoods	Softwoods
Cellulose	40 - 44%	40 - 44%
Hemicellulose	15 - 35%	30 - 32%
Lignin	18 – 25%	25 – 32%
Extractives	2%	5%

2.2.1 Extractives

Generally, extractives refer to the non-structural component of plant materials. They are essentially small molecular weight compounds that can be extracted with a solvent and usually occur in small amounts.They contain both organic and inorganic substances (Miller, 1999b). The organic extractives range from monomeric sugars to polymeric substances such as starch and tannins (Roffael, 2015). The amount and composition of the extractives vary tremendously between and within species, within the various sections or parts of the tree as well as between the harvesting seasons. The age, time-span after harvesting and growth conditions of a given species has a strong influence on the amount and the chemical composition of extractive contained in the species.

Wood extractives are one of the gluing relevant properties of wood. They have a major influence on the bonding process of wood chips and wood fibers with common synthetic adhesives such as urea-formaldehyde and phenol-formaldehyde resin (Roffael, 2015). They have an influence on the acidity and wettability (fatty substances and wax) at the wood-adhesive interface during the bonding process. A chemically-induced effect can occur based on the pH of the furnish which might either accelerate or decelerate the hardening process of adhesives based on polycondensation resins. Furnish of low pH favours the curing of the acid curing urea-formaldehyde resin but retards the curing process of the alkaline curing phenol-formaldehyde resin and vice versa. However, the pH of the furnish is usually monitored to allow for proper adjustment of the hardener level to shorten press time or prevent pre-cure of the mat before achieving optimal consolidation.

In addition, some extractives can act as formaldehyde scavengers by reacting with free formaldehyde in the produced panel, thus, reducing its formaldehyde emission. Nevertheless, some volatile organic compounds

(VOC) of wood can also be responsible for VOC emissions from wood-based panels. Some wood extractives such as tannins can be used as binders in the wood-based composites industry (Roffael, 2015).

2.2.2 Cellulose

Cellulose is the major and most abundant structural units that build up the wood material. It is a complex polysaccharide polymer consisting of many units of monosaccharide glucose made up of the elements Carbon, Hydrogen and Oxygen, thus function as an energy source for living things. Cellulose is the principal structural component of the cell wall of trees as well as other higher plants, most algae and some fungi (UKCES, 1997). It is the most abundant organic compound on the surface of the earth, characteristic of its glycan polymer of D-glucopyranose units, linked together by β-(1-4)-glucosidic bonds (Roger *et al*., 2012).

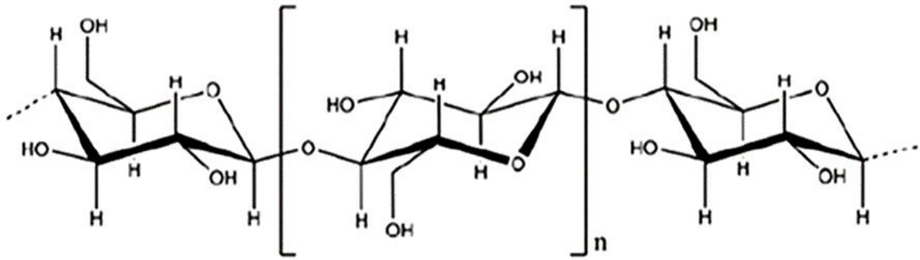


Figure 1: Basic chemical structure of cellulose showing repeating unit of Cellobiose. Source: Devi N. et al., 2016.

It is a linear-polysaccharide consisting of repeating units of cellobiose (the actual building block of cellulose) attached end to end. This long molecular chain structure is the reason why it found to be insoluble in most solvents. The long molecular chain forms into a criss-cross, which gives rigidity and strength to the cell wall of leaves, roots and stems (Schell *et al.*, 2009). According to Sari *et al.*, (2012), higher cellulose contents of wood furnish results in superior mechanical properties particleboards.

2.2.3 Hemicellulose

Hemicellulose is a branched polymer of pentose and hexose sugars found in the plant cell wall (*Huffman, 2003*). It is the second most abundant type of polysaccharide in nature and represents about 20-35 % of lignocellulose containing biomass (Saha, 2003). Hemicellulose is the polysaccharide matrix within which the cellulose microfibrils are embedded. Hemicelluloses, unlike celluloses, are not chemically homogeneous and are composed of pentoses, hexoses and sugar acid (Saha, 2003). The hemicellulose composition varies with species. Softwoods are composed predominantly of hexoses; meanwhile, the hardwoods are rich in pentoses. The pentosan content is significantly lower in softwoods compared to hardwoods (15-20%) and it ranges between 10% and 20% for tropical wood species. Hemicelluloses are slightly branched linear polysaccharides with generally no unique shape. The slight branching is the reason why they are more susceptible to chemical reactions and the action of solvents compared to cellulose. High hemicellulose content has been found to decrease the mechanical properties and increase the thickness swelling of particleboards. Its high contents have also been found to have positive effects on the formaldehyde release of particleboards (Sari *et al*., 2012).

2.2.4 Lignin

Lignin is a three-dimensional polymer of plants cell wall that holds the other structural components (cellulose and hemicellulose) together. It is literarily the binder agent or glue in plants. It is the third most abundant component in wood and other higher plants. Its matrix system binds the polysaccharide microfibrils and fibers (cellulose and hemicelluloses polymers) in plants cell wall; thereby giving plants (stem) their characteristic rigid and firm structure necessary for vertical growth (Feldman, 2002). Lignin is mainly concentrated in the middle lamella and the secondary cell wall of plant tissues. The presence of lignin makes woody tissues more resistant to biological attack (Feldman, 2002). Its structures are complex and diverse, marked by the presence of different functional groups such as phenolic, aliphatic alcohols, aldehydes methoxyls, ketones and ethers (Zakis, 1994). Amolecules of lignin consist of an aromatic system composed of phenylpropane units, linked together mainly by β-O-4 arylether linkages (Wegener, 1982).

Lignin content in wood can reach about 30% (Feldman, 2002) and varies between coniferous and deciduous wood as well as within the different parts of a tree. On average, the lignin content of softwoods ranges between 28% to 30%, and 18% to 22% for hardwoods (Roffael, 2004). In addition, the also exist some structural differences in the lignin of hardwoods, softwoods lignin and lignin of annual plants.

Although, lignin being the most abundant renewable carbon source on earth after cellulose with about 40 to 50 million tons produced worldwide each year, a huge quantity of it is non-commercialized. Between the two classes of lignin, so far only the sulfur-bearing lignin (lignosulphonates and kraft lignin totaling about 600 000 tons per year) is being commercialized (ILI 2000-2017). Sulfur-free lignin is yet to have a market. Currently, it is either used as in-house fuel in the industry to produce energy or discarded as waste. Only a small amount (about 1 to 2%) of this lignin is used to produce other value-added products (Lora and Glasser, 2002). Particleboards of wood furnish with higher lignin contents have been known to show superior physical and mechanical properties compared to those with lower lignin content. This is because being a natural glue by itself polymerizes under high temperatures and act as an additional glue to the particleboards as well as it being hydrophobic help in the moisture resistance of the particleboards.

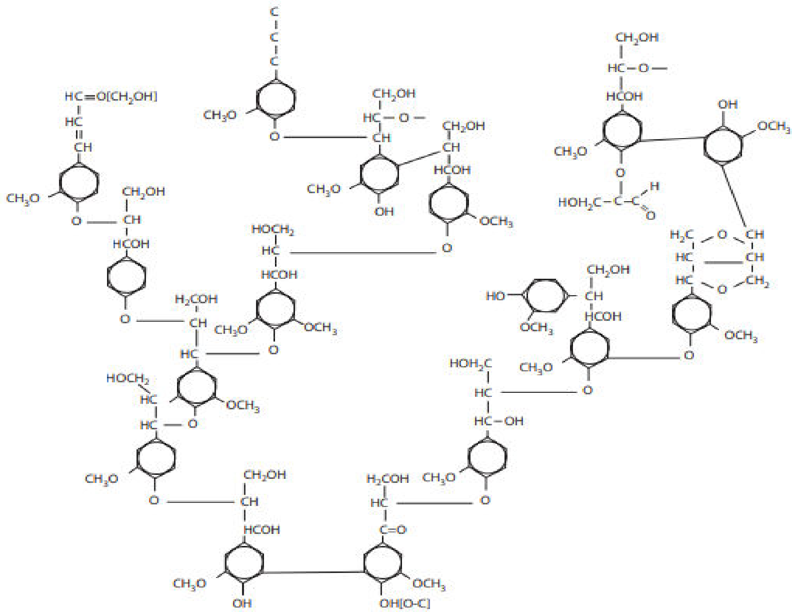


Figure 2: The structure of a segment of softwood lignin (Roger et al., 2012).

2.2.5 Ash

Ash is the low molecular weight inorganic mineral components of wood constituting mainly of calcium, magnesium, manganese, potassium, and silica. Among these mineral components, wood from the temperate zones constitutes predominantly the elements potassium, calcium and magnesium, whereas, woods of the tropics may constitute mainly of silica as its inorganic component (Dietrich & Gerd, 1989). These inorganic components emanate mainly from salts that are deposited in the cell walls and lumen of plants, which usually serves in some biological pathways in the plants.

The ash content of wood typically does not exceed 1% of its dry weight (Sjöström, 1999). In non-wood plant materials such as agricultural residues, values of up to 9% have been reported. The high ash content of agricultural plant materials is one of the factors that have contributed to the production downsides of the use of annual plants (e.g. straws) as raw materials for particleboards production especially when bonded with acid-curing aminoplastic resin. However, this problem could be solved by applying chemo-thermo-mechanical treatments (Markessini *et al.*, 1997) and by sorting out the parts of the plant materials with potentially high ash contents such as leaves and nodes (McKeen *et al.*, 1997).

2.3 Particleboards – Definition

The term particleboard refers to an engineered wood composite made out of lignocellulosic particles or chips from wood and none-wood plants, bonded together with an adhesive at an appropriate temperature (typically 160-200 ^oC) and pressure (typically 2-4 MPa). The production parameter depends on adhesive, raw material, board density, thickness, and product end-use. The wood particles are usually of different shapes and sizes, randomly oriented within the composite matrix, thus accounting for the final board properties. Particleboards generally consist of about 90% (by weight) wood and about 10% (by weight) adhesive and are almost exclusively made from softwood species. Adhesives in liquid form are applied in droplets form on the surfaces of the chips in a blender with the help of spray nozzles.

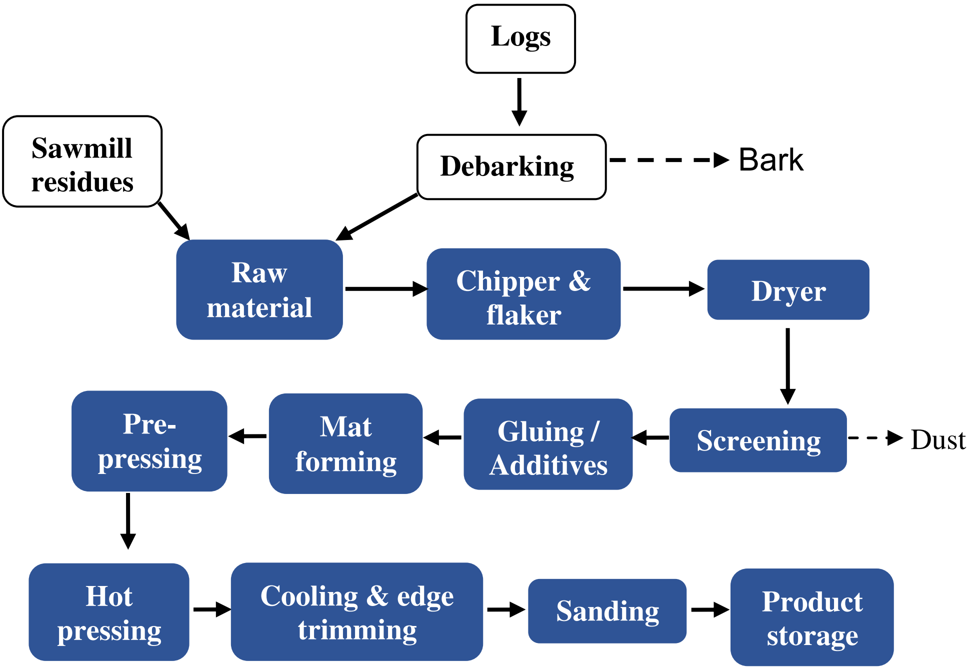


Figure 3: Flowchart of particleboards production process.

Particleboards are typically 3-layered; two surface layers and one core layer as illustrated in figure 4. The surface layers and the core layer usually account for 40% (w/w) and 60% (w/w) respectively of the total mass of the board. The core layer typically comprises of coarser particles whereas the surface layers consist of fine particles, thus providing a smooth board surface for easy lamination and other finishing steps. Particleboards production process in a typical industrial plant involves several steps, which are summarized in the flow diagram in figure 3.

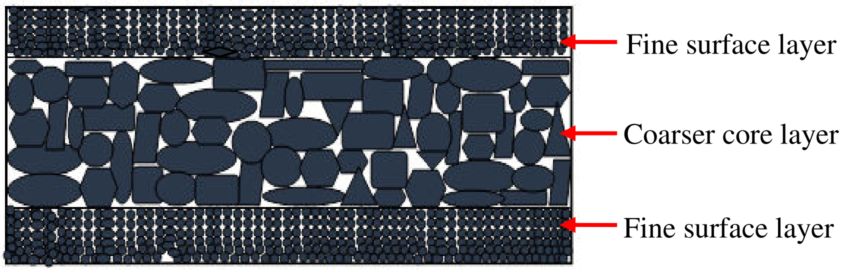


Figure 4: An illustration of 3-layered particleboard configuration.

2.4 Chemical additives

Besides the main raw material wood, adhesive and other auxiliary additive account for up to about 12% of the total mass of the board. The use of the right adhesive in the right amount is an important factor in wood-based panel production. The properties of wood-based panels can be directly and specifically influenced by choosing the right adhesive system and the auxiliary additives, such as hydrophobic agents and hardeners (Ambrozy and Giertlova, 2005).

2.4.1 Adhesives

Wood adhesives are the indispensable part of wood-based composites, as they are capable of attaching wood pieces together through surface adhesion. Different types of adhesives are used in the wood-based panel industry and their costs are a major factor in the total production cost of wood-based panels. In the production of particleboards, to achieve the inter surface-fixed connection between the individual wood chips, the adhesive must sufficiently wet the surface of the wood chip, heat and a corresponding pressure has to be applied.

Temperature is one of the determining factors in the reactivity of adhesives. Heat has a direct influence on the curing process of glue in particleboards production. Adhesives with low reactivity might require high press temperatures and longer press times (Carroll 1963; Lehmann *et al.*, 1973), to enable the temperature in the core layer of the boards to reach levels where the curing of the glue will be completed. Press temperatures, however, must be optimal to exclude the degradation of the adhesives and material on the surface layers in content with the press plates.

The amount of adhesive used in the production process of particleboards can also affect the properties of the resulting panels. An optimum adhesive level is an important parameter in the production process, as higher adhesive levels do not always lead to a corresponding increase in the strength properties of the particleboards. For example, Kimoto *et al.* (1964) showed that particleboards produced with UF-resin with the resin load of 8%, 10% and 15% (solid), based on dry mass of wood chips were found to show only slight improvements in the strength properties and thickness swelling for the boards with resin load of 15% compared to those with 10%. This means the 5% increase in resin load is not necessary. The adhesive content of particleboards is generally dependent on the desired boards' properties and the economics of production. In the particleboards sector, various studies have been conducted on the different adhesives used in the production process. Formaldehyde-based adhesives such as Phenol-Formaldehyde (PF), Urea-Formaldehyde (UF) and Melamine-Formaldehyde (MUF) resins are the most commonly used adhesives in particleboards production and account for 99% of the total sales volume and 97% of the total value (Huang & Li, 2008). Isocyanate based resins such as pMDI are only used in small amounts for the production of specialty products.

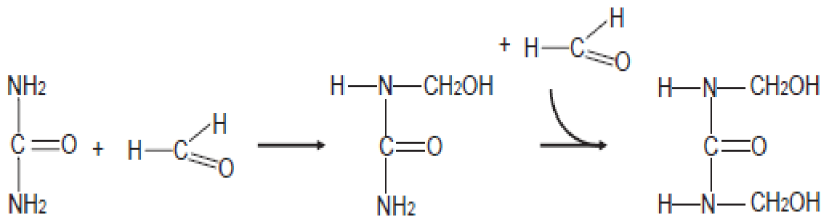
2.4.1.1 Urea-Formaldehyde

Urea-formaldehyde resins are the most important group of amino-plastic resins for the wood-based panel industry (Kharazipour, 1996). They are so-called thermosetting resins or amino-plastics because upon the application of heat they harden to insoluble and infusible macromolecules (Kharazipour, 1996). They are unique in their high reactivity, water solubility and reversibility of the amino-methylene linkages. UF-resins are highly susceptible to moisture and water, especially at high temperatures, which explains why they are extensively used in the manufacture of interior grade hardwood plywood and particleboards.

The synthesis of UF-resins is a two-step process; methylolation and acidic condensation. Methylolation means the addition of three (four in theory) molecules of bivalent formaldehyde to one molecule of urea (at pH 7) to produce the mono- and di-methylolureas as shown in Figure 5. The reaction is stopped at this stage to avoid the formation of high molecular weight methylolureas (Christjanson *et al.*, 2006). At the acidic condensation stage (low temperature and slightly acidic pH), the methylolureas and free formaldehyde still present in the reaction mixture polymerizes to produce urea-formaldehyde resin as shown in Figure 6. The condensation product may be linear or cross-linked. The synthesis of UF-resins is dependent upon certain factors such as the molar ratio between urea and formaldehyde, pH, reaction time and reaction temperature. These conditions can be tailored to produce UF resins for different end uses.

The curing of UF-resins takes place under acidic conditions, which could be achieved through the direct addition of an acid (hardener). The hardeners, ammonium sulfate and ammonium chloride are the most predominantly used in the wood-based panel industry. The hardener reacts with the free formaldehyde in the UF-resin to produce acid, which lowers the pH of the solution. The amount of hardener required to cause the change in pH is dependent on the ability to release the acid.

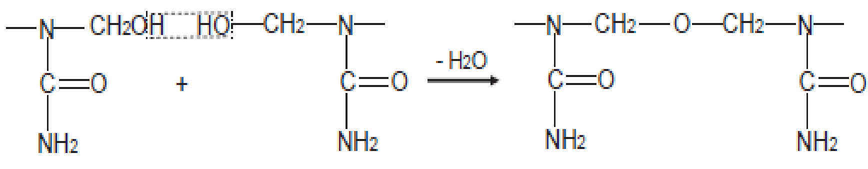
UF-resins, primarily because of their water solubility, high curing rate, good adhesion, short hot-pressing time, clear glue-line, cold tack ability, and low cost are the most widely used adhesives in the particleboards industry. However, they are unstable at higher relative humidity, especially at higher temperatures. This is because the amino-methylene bridges are susceptible to hydrolysis under such conditions (Pizzi and Mittal, 1994). In addition, the emission of the free formaldehyde from UF resin bonded wood-based panels is a matter of great concern, which has been linked to the possible cause of cancer when used for indoor applications.



Urea Formaldehyde Monomethylolurea Dimethylolurea

Figure 5: Methylolation of urea in the synthesis of UF resin (adopted from Christjanson et al., 2006)

Through ether links



Through methylene links

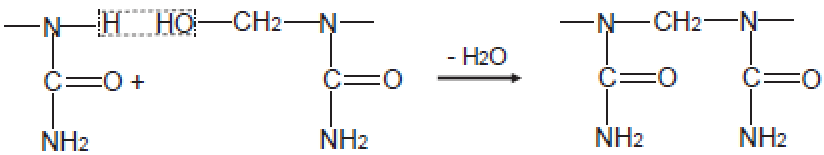


Figure 6: The acidic condensation of methylolureas to form Urea-formaldehyde resin (adopted from Christjanson et al., 2006)

2.4.1.2 Phenol Formaldehyde

Phenol formaldehyde (PF), also known as phenolic resins are a very significant class of resins in the wood-based panels industry. They are synthesized by reacting phenolic alcohol derived from benzene (Phenol) and formaldehyde in the presence of a basic or an acid catalyst to produce resol or novolac resins respectively (see figure 7). The molar ratio between phenol and formaldehyde is usually 1:2.5 (Becker and Braun, 1990; Kloeser *et al*., 2007). Novolac resins are produced in the case where an acidic catalyst is used during the condensation reaction between phenol and formaldehyde.

The polycondensation reaction of the first formed methylol phenol derivatives is allowed to complete with the addition of phenol. The reaction stops when formaldehyde in the reaction mixture is exhausted leaving behind unreacted phenol. Novolac resins cannot cure by themselves. Therefore, in order to use Novolac resin as a wood adhesive, a curing agent is usually added to cross-link the resin. The most frequently used curing agent is hexamethylenetetramine.

PF-resins are used for the manufacture of moisture-resistant particleboards, OSB, MDF, and plywood as well as for the production of very low formaldehyde emission panels. The strong C-C-bond between the aromatic nucleus and the methylol group or methylene bridge account for their high resistance to hydrolysis and low formaldehyde emission. Nevertheless, PF resins require a longer press time compared to UF-resins and cause a dark glue line (Dunky, 2001), which is undesirable for decorative and interior applications.

However, because of the high hygroscopic nature of the catalyst, the equilibrium moisture content of the panel is increased due to PF-resin. Consequently, press time has to be increased compared to other adhesives. The curing of PF-resin finally takes place when the water between the reactive hydroxymethyl groups and the detachable hydrogens at the phenolic core is eliminated (Roffael and Schneider, 1981; Kharazipour, 1996; Kloeser *et al*., 2007).

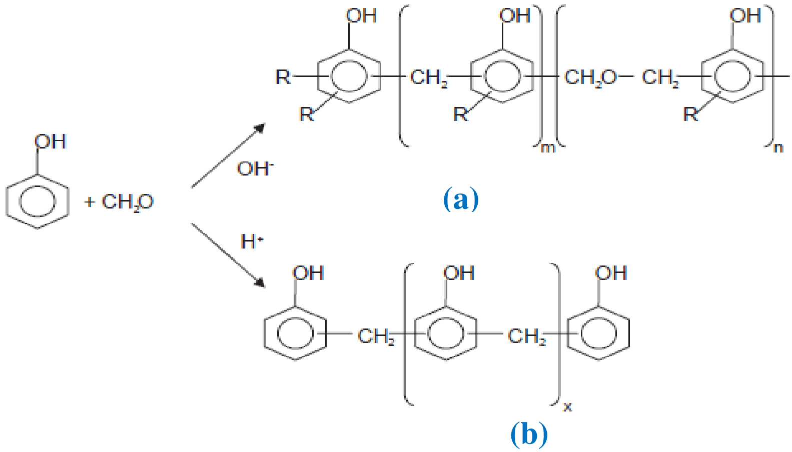


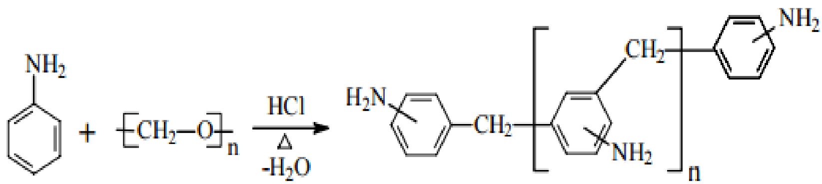
Figure 7: Polycondensation reaction of phenol-formaldehyde resins- (a) Resol is formed when a basic catalyst is used, (b) Novolac is formed when an acid catalyst is used. (Adopted from Kloeser et al., 2007)

When this happens, new methylene bridges are formed. Linear structures arise and, with growing condensation level, cross-linking bridges are created between phenolic groups of different linear structures (Roffael and Schneider, 1981; Kharazipour, 1996; Kloeser *et al*., 2007). PF-resin starts the final curing at about 130 °C. The speed of curing depends on the press temperature. As temperature increases, the curing speed becomes progressively shorter (Umemura *et al*., 1995; Wang *et al*., 1995; Geimer and Christiansen, 1996; Kloeser *et al*., 2007). Hot-staking of PF-resin bonded panels is necessary to achieve full curing of the glue after the hot pressing stage.

2.4.1.3 Isocyanate-based resin - PMDI

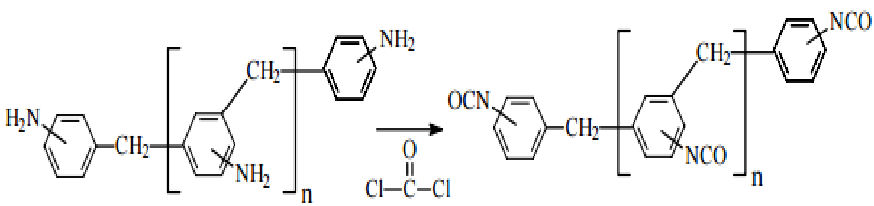
Polymeric diphenyl methane diisocyanate (PMDI) is a compound composed of polyaromatic isocyanates, usually in the form of an organic brown liquid.

PMDI is synthesized through phosgenation of a primary amine. This process involves a two-step reaction: The first step is the acid-catalyzed condensation of Aniline and formaldehyde or paraformaldehyde to produce polyamine (Figure 8). The second step is the phosgenation of primary amine (polyamine) by phosgene to produce PMDI (Figure 9).



Aniline Paraformaldehyde Polyamine

Figure 8: Acid-catalyzed condensation of Aniline and Paraformaldeh- yde to produce Polyamine (Twitchett,1974).



Polyamine Phosgene PMDI

Figure 9: Phosgenation of Polyamine to produce PMDI (Twitchett, 1974).

PMDI, since it was first introduced in the German particleboard market in the 1970s, has widely been used in the production of OSB worldwide and MDF in Europe (Papadapolous *et al*., 2002). It is considered one of the most performing binder systems in the wood-based panel industry. This is because the isocyanate group of PMDI is capable of reacting with the hydroxyl group of wood furnish and moisture to form irreversible urethane linkages (Smith, 2012). Such bonds are highly resistant to hydrolysis, thus giving the PMDI bonded boards a degree of moisture resistance. The irreversibility of the urethane linkages is the reason why PMDI bonded boards perform better in both interior and exterior applications compared to the boards bonded with UF resin. The dosing level of PDMI (usually 2-3%) in the manufacturing process is at least three times lesser compared to UF resin (8-10%) to achieve boards of the same or even better physical-mechanical properties. However, the price of PMDI is significantly higher than that of other synthetic resins. Nevertheless, the fact that PMDI cures at low temperature (Smith, 2012) and can be applied to wood furnish with high moisture content (Conner, 2001), one could argue that the manufacturer could save on energy consumption required for drying and production speed, thus offsetting the high cost of PMDI. In addition, PMDI is not pH sensitive and thus can tolerate a wide variety of lignocellulosic materials with different chemical compositions (Wood-based panels international, 2009). Compared with traditional formaldehyde-containing resins, formaldehyde emission from pressed boards bonded with PMDI is negligible (Kloeser *et al*., 2007).

However, the use of PMDI in the production of wood composite also has some disadvantages. Beside the already mentioned high price, its highly reactive nature causes it to stick firmly to metallic parts of the hot press. Therefore, a technical problem arises when PMDI is used on the surface layers of boards. For this reason, special release agents must be used to coat the metallic parts of the hot press if PMDI is used as surface layer resin. Another negative aspect of PMDI usage is that significant protective measures have to be taken to avoid health risk and occupational diseases from inhalation of its toxic volatile compounds.

2.4.1.4 Bio-based adhesives

Bio-based adhesive refer to substances from natural or renewable sources that are capable of binding individual wood chips or fibers together. These renewable sources include a wide range of materials from plants, animals, fungi and other microorganisms (Nikvash, 2013).

The first attempt to produce wood-based panel-like products of sawmill residues bonded with animal protein-based adhesive was first expressed in 1887 (Hubbard, 1887). Over the years, proteins have been replaced in most wood bonding applications by petroleum-based adhesive because of their low cost, easy-to-use, and most especially because of their higher durability. However, in recent years several factors like the stringent regulations on formaldehyde emissions of wood-based panels, fluctuation in petroleum prices and the improvement of protein adhesive technologies have led to the re-emergence of protein binders in the manufacturing process of wood-based panels.

Binder systems based on enzymes are another interesting group of environmentally friendly alternatives to petrochemical-based adhesives. The Adhesive properties of lignin can be enhanced by activating the lignin on wood fibers surfaces with an oxidase enzyme, laccase (Kües *et al*., 2007). Euring *et al*., (2011, 2013, 2015, 2016), and Euring (2008) showed that by treating thermo-mechanically pulped (TMP) fibers with Laccase-Mediator-Systems (LMS) it is possible to activate the lignin on the surfaces of the fibers and use the bonding forces of the activated lignin on the fibers' surfaces as a natural glue to produce MDF and insulation panels without the addition of conventional adhesives. The mechanical-technological properties of the resulting boards are comparable to those bonded with conventional petrochemical adhesives such as UF-resin. Euring *et al*., (2016) also demonstrated that the addition of technical lignin to the LMS, resulting in the so-called Lignin-Laccase-Mediator System (LLMS), activates additional bonding strengths in the system and results in MDF with superior strength properties compared to those of the LMS.

2.4.1.4.1 Blood albumin

The animal protein, blood albumin, has long been used as an adhesive in wood-based products. Several patents (Henning, 1920; Lindauer, 1923; Cone, 1934; and Gossett *et al*., 1959) indicate that animal protein-based adhesives were popular from early to the mid- twentieth century. These kinds of adhsives became almost non-existent with the invention of synthetic adhesive such as phenol-formaldehyde resin (PF) and urea-formaldehyde resin (UF). This is because protein-based adhesives were not readily available, expensive, and had a shorter shelf-life (Sellers, 1985; Detlefsen, 1988). In recent years, the have been an increasing demand for natural binders in the wood-based panel industry (Adhikari *et al.*, 2017). This is because the widely used formaldehyde-based resins such as UF-resin emits formaldehyde, which has been known to be carcinogenic to humans (IARC, 2006; Pizzi and Mittal, 2011; European Chemical Agency, 2014). In addition to this, the most commonly used synthetic resins, e.g. UF-resin, are based on crude oil, which is non-renewable and are dependent on fluctuating oil prices (Wilson, 2010). Based on this recent trend, several studies have been focused on partially or completely substituting synthetic adhesives with renewable alternatives, enzymes such as laccase (Euring *et al.*, 2015; Kirsch *et al.*, 2016, 2018), plant proteins such as wheat gluten (Lei *et al.*, 2010) or soy (Fan *et al.*, 2016), and blood albumin (Lin and Gunasekaran, 2010). However, except in plywood applications, there has been no appreciable use of blood protein adhesives in the manufacture of wood-based products as well as appropriate research. Li *et al.* (2018) is the only reported study that focused on developing an adhesive that is based purely on blood-meal. In this study, a blood-meal-water-solution of 28% solids content was prepared with different additives to improve its properties.

There are several challenges facing the use of protein-based adhsives in wood-based products. Beside the fact that synthetic resins have led to the establishment of high standards to be met by natural binders, the handling of such binders can be problematic due to their high viscosity, low solids content, long pressing times and low resistances to hydrolysis. These are due to the complex chemical structure of proteins. Also, proteins from different sources e.g., animal protein from pig, beef, sheep, *etc*., are usually different in type and composition as the proteins may be accompanied by other polymers such as carbohydrates, fats, *etc*., thus would have varying properties. Another important challange faced by proteins with regards their use in wood-based panel products is fact that proteins are vulnerable to denaturation. Upstream processing, which may require treatments that changes temperature and pH (in the effort to make them more or less stable) can affect the proteins and their adhesive properties. The overall incentive to overcome these challenges is to create a health-friendly product completely based on natural resources (Frihart and Lorenz, 2017).

In Germany, a huge amount of animal blood from slaughterhouses is available. Bonding particleboards with this NAF bio-based binder would be environmentally friendly and eliminates the health risks associated with the use of conventional synthetic adhesives.

2.4.2 Auxiliary additives

In addition to the right choice of adhesive, auxiliary additives and their amounts are an integral part of the production process of particleboards. They have a direct influence on the properties of particleboards. These include curing agents (hardeners) and water repellents (hydrophobic agents), but also formaldehyde scavenging substances and fire retardants.

2.4.2.1 Hydrophobic agents

Water repellents are used in chipboard and fibreboard production to reduce the rate of water absorption and thus the swelling in the case of short-term exposure to water or increased air humidity. The use is also important with regard to the treatment of fibreboards with aqueous glues or water-based paints, but also when using the materials in the wet or outdoor areas. Paraffin emulsions are the most predominantly used water repellents, with dosing levels of 0.3% to 1.0%, based on the dry weight of fibers (Deppe and Ernst, 1996).

2.4.2.2 Curing agent

Ammonium sulfate $(NH_4)_2SO_4)$ and ammonium nitrate (NH_4NO_3) are the most widely used and effective curing agents (hardeners) in the wood-based panel industry. Their role is to accelerate the curing process of UF-resin. They react with free formaldehyde of the UF-resin to produce their respective acids (sulfuric acid or nitric acid), which eventually lowers the pH of the resin as well as that at the material-adhesive interface, thereby creating a suitable environment for the cross-linking process for acid curing binder system. The reaction speed or the rate at which the acid is produced depends on the amount of the available free formaldehyde and hardener in the reaction mixture. This means that the reaction speed is higher with UF-resins of higher molar ratio compared to the lower molar ratio UF-resins. However, with the current strict regulations on formaldehyde emission of wood-based panels, most manufacturers turn to use lower molar ratio UF resins, whose free formaldehyde is insufficient to cause a significant drop in pH within a short amount of time when these hardeners of ammonium salts are used. This problem can be overcome by using new curing agents that do not depend on the amount of free formaldehyde to cause a decrease in pH (Alexandropoulos *et al.*, 1998). Such special hardeners include aluminium sulfate [$Al_2(SO_4)_3$], ammonium persulfate [$(NH_4)_2S2O_8$], citric acid ($C_6H_8O_7$) and other combined hardener systems (Mantanis *et al.*, 2018). The hardeners of ammonium salts are generally used because they are inexpensive and easy to handle (Pizzi, 1994; Dunky, 1998), and are usually added based on the solid content of the adhesive in the range of between 0. 2% - 2% (w/w).

3.0 The study raw materials

The focus of this work is to investigate the possible feasibility to use the wood of the fast-growing tree species, the umbrella tree (*Musanga cecropoides*), and the prunings of cocao tree (*Theobroma cacao*) for the production of particleboards. In addition to these, the residues of the annual plant, common bean (*Phaseolus vulgaris*), are also investigated for the same purpose. The following chapter describes in detail the various raw materials.

3.1 Bean crop (*Phaseolus vulgaris*) residues

Common bean (*Phaseolus vulgaris L.*) is a herbaceous annual plant of the Fabaceae family. It originated in Central and South America and was cultivated in Peru by 6000 BC. Today, it is widely cultivated as a major crop in Europe, Africa, Asia and the Americas (Wortmann, 2006). Common bean consists of a wide range of cultivars and is one of the most important food legumes for the Eastern and Southern African agriculture. It is cultivated in more than 100 countries worldwide, covering more than 29 million hectares of harvested area (FAOSTAT, 2016) and provides food for more than 100 million people (Wortmann *et al*., 1998).

The name common bean refers to the class of bean that is grown to maturity and harvested purposely for the seeds within the pods. This type of bean typically requires 70 to 120 days from the time of seeding to maturity. They grow as either bushes or vines. The pods (shells) are narrow, with size range between 8 cm to 20 cm by 1cm to 2 cm. Each pod can contain up to 12 seeds, but most varieties have 4 to 6 seeds per pod. The seeds as well as the pods are of various colours, depending on the cultivar (Purseglove, 1968; Wortmann, 2006). Average yields of between 0.5 to 1.5 tons per hectare are common. Also, yields of up to 5 tons per hectare have been reported (Wortmann, 2006). Green biomass annual yield of about 16 tons per hectare have also been reported (CNC, 2004).

In Cameroon, common bean is cultivated and harvested in two seasons; the dry and the rainy seasons. The harvesting and processing methods differ with the various seasons. For the year 2017, about 413,072 tons of dried beans were produced in a harvested area of 310,650 hectares (FAO-STAT, 2017).

Figure 11: An image of common bean shells.

Figure 10: An image of mature common bean plant. Source: Sliver Reef Organicfarms

Upon harvest, huge quantities of the shells and straws remain in the field without any further value-added uses. They are rather subjected to incineration, dumped in landfills, or allowed to rot on spot as means of disposal. None of the disposal practices is sustainable to the environment. In this context, it is of interest to study the usefulness of crop's by-products as a raw material for particleboard production; perhaps this could contribute towards alleviating the problems associated with bean shells disposal.

3.2 Umbrella tree (*Musanga cecropioides*) wood

Umbrella tree (*Musanga cecropioides L.*) is a tropical evergreen tree species of the secondary forest. It is called the umbrella tree because of its characteristic umbrella-like crown. It belongs to the family of Urticaceae, and is one of the most common tree species on forest clearings and abandoned farmland. It is fast-growing and can reach a height of 30 meters and diameters of 30 cm to 91 cm by the end of its lifespan. The species grow mainly in regions with average annual temperatures of between 25 °C to 30 °C and annual precipitation of between 1300 mm to 2500 mm. Under these conditions, it can grow on sites from 700 to 1200 meters above sea level. The umbrella tree has a short lifespan of about 20 years and has shown annual height increments of 5 meters within the first year of planting (Orwa *et al.*, 2009).

The wood is exceptionally lightweight when dry, with densities of 200 ... 250 kg/m³ (Burkill, 1985). Due to its low density and thus the relatively low energy density, the wood is not used as fuel. The wood has so far been used for the production of kitchen utensils, musical instruments, toys, and swimming equipment such as canoes. In general, it is a widely underutilized tree species that grows rapidly and is abundant in Cameroon and other parts of West and Central Africa.

Figure 12: The freshly harvested wood of umbrella tree.

However, there exists no inventory data on the species in Cameroon. Despite its rapid growth, the species is not cultivated in plantations due to its limited uses. It is therefore important to study the suitability of its wood to be used as a raw material for particleboards production.

3.3 Cacao tree (*Theobroma cacao*) prunings

The cocoa plant (*Theobroma cacao L.*) is a valuable tree species that grows to approximately 6.5 million hectares in 57 tropical countries. The global annual cocoa bean harvest is estimated at 4.23 million tonnes (ICCO, 2016). Africa is the largest cocoa producing continent in the world, accounting for 68 percent of global production. Cacao is the main cash crop to more than 75 percent of Cameroon's population (Tcharbuahbokengo, 2005). Cameroon produced about 250,000 tonnes of cocoa beans in the 2017/2018 cacao year (ICCO, 2019).

Cacao prunings refer to the sections (unwanted branches) of cocao tree that are removed annually through thinning operations to improve on the fruit development of the tree. Large quantities of the unwanted branches remain in the fields every year after thinning without any value-added used.

A B

Figure 13: A prunned cacao tree(A) and an unwanted cacao tree branche (B).

It is estimated that cacao thinning produces over 21 kg of organic dry matter per tree, which is about 25 tons per hectare per year (Lim, 1986a). To put this into perspective, about 50 cubic meters of dry prunings (considering the average wood density of cacao at 470 kg/m^3) are generated per hectare per year, which is more than the average wood increment per hectare per year for temperate forests. The efficient use of these residues could contribute to effective and sustainable value creation and increase the incomes of cacao farmers. It is therefore important to study the suitability of cacao tree prunings for the production of particleboards.

4.0 Materials and methods

This work aims to develop three-layered particleboards of reduced formaldehyde emissions using the residues of the common bean (*Phaseolus vulgaris*), the prunings of cacao tree (*Theobroma cacao*) and the wood of umbrella tree (*Musanga cecropioides*) as a raw material. This chapter focuses on the detailed description of the various processing steps and the types of equipment that were involved in transforming the different raw materials into chips that were used in producing the particleboards as well as the characterization of the raw materials. The process techniques used for the development of the particleboards in this study, consistent with the manufacturing process generally employed in the industry are also described in detail. The adhesives, as received from the various manufacturers and used in this research are also described. Finally, this chapter ends with a description of the methods through which the physical-mechanical properties, as well as formaldehyde properties of the fabricated particleboards, were tested and evaluated against the specifics of the standard DIN EN 312-2.

4.1 Preparation of the raw materials

4.1.1 Umbrella tree wood and cacao tree prunings

The wood of the umbrella tree (*Musanga cecropioides*) and the Cacao tree (*Theobroma cacao*) prunings of recent thinning operations were obtained from a farm about one kilometer from Boa Bakundu village of the South-

West region of Cameroon. The umbrella tree wood was supplied as freshly harvested logs of about one meter lengths. The materials were manually debarked and transported to the MDF pilot plant of the Büsgen Institute of the University of Goettingen by means of a shipping container. While at the pilot plant, with the help of a drum chipper, the materials were chipped into sizes of between 15mm to 120mm. To produce particleboard-size chips, the chipped wood chips were further shredded with the help of a laboratory-scale knife ring flaker type PML 1 150/250 DELACHAUX GmbH, Offenbach. With the help of a whirling sieve from the company Allgaier-Werke GmbH, type TSM 1200/2, the resulting wood chips were then screened to separate into core layer and surface layers-size chips.

Surface layers Core layer

Figure 14: Laboratory produced core layer and surface layers wood chips. The ruler at the bottom is marked in cm.

Undersized particles (dust <0.5 mm) were excluded from the materials. The screened wood chips were air-dried to moisture content below 15% (w/w) for further processing and characterization.

4.1.2 The bean crop residues

The residues of the bean crop (*Phaseolus vulgaris*) were collected from different locations in the North-west and South-west regions of Cameroon at two different seasons; the rainy season and the dry season. The materials were also shipped to the MDF pilot plant of the Büsgen Institute of the University of Goettingen. The bean crop residues (BCR) were seasonally differentiated into the bean crop residues of the rainy season and the bean crop residues of the dry season. The beans crop residues of the rainy season are those that were harvested and pre-processed during the rainy season. They consisted of 100 percent bean shells and within the context of this research project will be referred to as bean shells. On the other hand, the bean crop residues of the dry season are those that were harvested and pre-processed during the dry season. They were made up of a mixture of bean straws and bean shells in the ratio of about 50:50 percent (w/w) respectively. This is because the harvesting and the primary processing methods of beans in Cameroon differ in the two seasons. In the dry season, farmers prefer to dry out the entire bean plant in the fields until about 90% of the leaves have fallen off, and then harvest the entire bean plant to separate the beans from the rest of the plant with the help of traditional tools. This makes it difficult to separate the straw from the shells. On the other hand, during the rainy season, the bean pods are harvested without involving other parts of the plant, and then the beans are taken out of the pods by hand.

Figure 16: Beans crop residues of the rainy season (100 percent bean shells).

Figure 15: Beans crop residues of the dry season (a mixture of shells and straws.

While in the pilot plant, the bean crop residues were minced to produce particleboards-size chips using a hammer mill ELECTRA SAS, Poudenas, France, type F3 and then air-dried to moisture content below 15 percent. The harmer-milled chips were then screened to separate into core layer-size and surface layers-size chips with the help of a whirling sieve from the company Allgaier-Werke GmbH, type TSM 1200/2.

4.1.3 The reference materials

To be able to characterize the raw materials used in this work and make meaningful comparisons of their performance, it was necessary to obtain some reference materials. The wood of Norway spruce (*Picea abies*), one of the most common softwood species used in the production of particleboards in Europe was harvested from a forest near Witzenhausen, Germany. The wood was debarked, chipped and air-dried at the MDF pilot plant of Buesgen institut. The chipping of the wood was done by using a laboratory-scale drum chipper. The wood chips were further minced a laboratory-scale knife ring flaker of model PML 1 150/250 from the company DELACHAUX GmbH to produce particleboard-size wood chips. The resulting wood chips were screened to separate into core layer-size and surface layers-size particles with the help of a whirling sieve from the company Allgaier-Werke GmbH, type TSM 1200/2. The materials were air-dried for further processing.

In addition to the wood of Norway spruce, industrially produced particleboard-sized wood chips of the core layer and surface layers were also obtained from the company Pfleiderer Holzwerkstoffe GmbH & Co. KG, D-59759 Arnsberg.

4.2 Physical and chemical characterization of the study raw materials

The development of particleboards requires an in-depth understanding of the physical and chemical characteristics of the raw materials being used. In doing so, the production process can be tailored to achieve boards of desired physical and mechanical characteristics. The following chapters present in detail the various methods and equipment used for determining the physical and chemical properties of the study raw materials described in chapter 3. In addition to the study raw materials, the wood chips of Norway spruce (*Picea abies*) and wood chips from the industry (Pfleiderer Holzwerkstoffe GmbH & Co. KG) were also analyzed for the various physical and chemical characteristics. All of the analyses were conducted based on the respective standards and norm as described in the following chapters.

4.2.1 Determination of fractional composition and Bulk densities of the chips

The particle size distribution of the produced chips of the various materials for both the surface layers and the core layer was determined using the sieve analysis method according to DIN 66165. To do this, a whirling sieve from the company Retsch, D-42781 Haan, of the model AS 400 was used. One kilogram of chips was placed on the top sieve of the whirling sieve. The sieves were arranged such that the sieve with the largest mesh size is at the top of the stack and the smallest at the bottom. The materials were fractionated for a period of 5 minutes at a rotation speed of 250 rpm, with a change in the rotation direction after the third minute. The rotation splits the materials into the fractions of the individual sieves. After sieving was completed the fractions of the individual sieves were determined gravimetrically with the help of a digital scale. The determination was carried out in five repetitions and the average values were considered.

Furthermore, the bulk densities of raw materials for both the surface and the core layers were determined according to DIN EN 12580. Bulk density is the ratio of the mass of loose bulk material to its occupied volume. The determination of the bulk density is significant because, according to Brinkmann (1982), bulk density has a significant influence on the properties of composite panels. Among other things, the bulk density influences the minimum density of materials. It is known that the bulk densities should be well below the targeted bulk densities of the composite panels. To this end, the materials were poured into a vessel with a defined volume (20 liters) and weighed. Five repetitions were performed for each material and the average value was taken. The bulk density of each material was calculated from the weight and the volume of the material according to the formula below.

$$\text{Bulk density } [g / l] = m / V$$

Where

- m is the mass of the sample (air dry) in grams
- V is the mass of the measuring cylinder in in liters

4.2.2 The determination of cold-water soluble extractives content

Wood extractives refer to the non-structural part of wood (Kirker *et al.*, 2013). They are basically the small molecular weight compounds that can be extracted with a solvent and usually occur in small amounts. They contain both organic and inorganic substances (Miller, 1999b). The organic extractives range from monomeric sugars to polymeric substances such as starch and tannins (Roffael, 2015). The amount and composition of the extractives vary tremendously between species, within species and between the various sections or parts of the tree as well as with seasons. The age, time span after cutting and growth conditions of a given species has a strong influence on the amount and the chemical composition of extractive contained in it.

To do this, 5 grams of dry material was weighed into a 250 ml conical flask to which 150 ml of demineralized water was added. The flask and its content were allowed to digest for 24 hours at room temperature (about 20°C) on a mechanical shaker vibrating at about 120 rpm. After 24 hours, the extract (solution) in the flask was filtered off with the help of a tared fritted-glass crucible of G3 porosity.

The aqueous extracts of the various raw materials were used to directly determine the pH and the buffering capacities of the raw materials.

To determine the extractives content of the raw material, the content of the fritted-glass crucible was washed clean with distilled water to remove any leftover cold-water-soluble extractives. The fritted-glass crucible and its content were dried to constant weight at a temperature of 103°C and then cooled to room temperature in a desiccator. The fritted-glass crucible and its content were finally weighed and the cold-water extractives content of the material was calculated using the formula below. The determination was carried out in four replicates for each of the raw materials used in this work.

$$\text{Cold-water extractives content (\%)} = \frac{W_1 - W_2}{W_1} \times 100$$

Where

- W_1 is the mass of absolute dry sample, in grams
- W_2 is the mass of dried sample after extraction with cold water, in grams

4.2.3 The determination of pH-value and buffering Capacity

The aqueous extracts of the various raw materials (extraction procedure described in 4.2.2) were used to directly determine the pH and the buffering capacities of the raw materials. To determine the pH, the pH electrode was kept for 4 minutes in a 50 ml beaker containing 20 ml of the aqueous extract at room temperature and then reading the value.

With the help of a Titrometer from the company Schott, D-55122 Mainz, each of the aqueous extract was analyzed through titration to determine the buffering capacity. In each case, 20 ml of the extract is titrated with a 0.01 molar NaOH solution to a pH of 7 (neutral value). To this end, the buffering capacity of each extract was determined based on the amount of NaOH solution used up in the titration with the help of the following relationship:

1ml of 0.01 mole / literNaOH-solution used-up corresponds to buffer capacity of 1.5 mmolNaOH / 100g of oven dry material to be determined

4.2.4 The determination of hot-water soluble extractives content

This procedure determines the amount of hot-water soluble extractives contained within the raw materials. For this purpose, 2 grams of the material was weighed into a 250 ml Erlenmeyer flask to which 100 ml of demineralized water was added. The Erlenmeyer flask and its content were placed on a heating source and then connected to a reflux condenser. The setup was allowed to cook under reflux for two hours and then cooled to room temperature.

After cooling, the aqueous extract of the flask was filtered using a tared fritted-glass crucible of porosity G3. The extracted material in the fritted-glass crucible was washed clean with hot water and then dried to constant weight at a temperature of 103 ^{o}C in a drying oven. The fritted-glass crucible and its content were finally allowed to cool to room temperature in a desiccator and then weighed. The amount of hot-water soluble extractives content of the raw material (in percent) was determined by the weight loss with the help of the formula below. The determination was carried out in four repetitions for each of the raw materials used in this study.

$$\text{Amount of hot-water soluble extractives (\%)} = \frac{W_1 - W_2}{W_1} \times 100$$

Where

- W_1 is the mass of oven dry sample, in grams
- W_2 is the mass of dried sample after extraction with hot water, in grams

a **b**

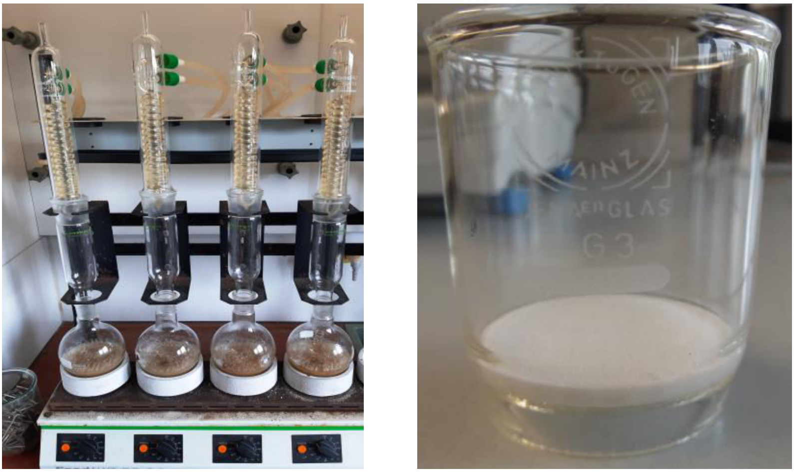


Figure 17: The setup for the determination of the hot water extractive content of the raw materials (a) and the fritted-glass crucible of porosity G3 (b)

4.2.5 The determination of the solvent-soluble extractives content

The determination of the solvent-soluble extractives content of the raw materials was determined by the ethanol-cyclohexane method. To do this,

5 grams of the dry material was weighed into an extraction thimble of dimensions 33 x 100 mm and placed in position inside a Soxhlet extraction apparatus. 150 ml of ethanol-cyclohexane, prepared by mixing ethanol and cyclohexane in the ratio of 1: 2 (v/v) respectively was put into a 250 ml Soxhlet extraction flask containing anti-bombing granules. The flask was placed a serial heating block from the company OMNILAB, FoodALYT RS60 and then connected to the Soxhlet extraction apparatus (see figure 18) and then extracted for six hours.
After the extraction was completed and the setup was cooled to room temperature, the solvent mixture was evaporated to near-dryness in a vacuum rotary evaporator from the company Heidolph Instruments GmbH & Co. KG, D-91126 Schwabach; model Hei-VAP value. The Soxhlet extraction flask and its content were then dried to constant weight in a vacuum oven. Finally, the weight of the extraction flask and its residue was measured and used to calculate the amount of solvent-soluble extractives of the material based on the relationship below. This determination was also conducted in 4 replicates for each of the raw materials.

$$\text{Solvent-soluble extractive content (\%)} = \frac{W_r - W_f}{W_s} \times 100$$

Where

- W_s is the dry mass of specimen or raw material, in grams

- W_r is the dry mass of extraction flask + residue + three anti-bombing granules
- W_f is the dry mass of extraction flask + three anti-bombing granules.

a **b**

Figure 18: The setup for ethanol-cyclohaxane extraction of the raw materials (a) and the vacuum rotary evaporator from the company Heidolph (b).

4.2.6 The determination of pentosan content of the raw materials

Pentosan is the main component of hemicellulose. The determination of the pentosan content of a lignocellulosic material makes it possible to approximate its hemicellulose content. The amount of pentosan is determined by determiningcolorimetrically the amount of furfural in the distillate produced by boiling the material in hydrobromic acid. To do this, 200 ml of 3.2 molar hydrobromic acid was put into a 1000 ml boiling flask containing 2.0 grams of the material (oven dry). The boiling flask, placed on a heating device, was connected onto a three-way connecting tube equipped with a graduated separatory funnel. A two-way connecting tube connects the free end of the three-way connecting tube and the Graham condenser below which a 250 ml volumetric flask was placed with marks added at intervals of 90 ml.

The setup was allowed to distill until the furfural-containing distillate reached the volume of 240 ml. During the distillation process, 90 ml of distilled water was added into the boiling flask through the separatory funnel when the furfural-containing distillate in the volumetric flask reached volumes of 90 ml and 180 ml. The furfural-containing distillate was made up to the 250 ml mark by adding 10 ml of distilled water. After shaking vigorously to ensure a uniform mixture of the solution, 5 ml of the distillate was pipetted into a 100 ml volumetric flask and topped up to the 100 ml mark by adding 95 ml of distilled water thereby obtaining a 1:20 dilution. From the 1:20 dilution solution, 10 ml was transferred into a 50ml volumetric flask and added 40 ml of distilled water to make up to the 50 ml mark, thereby obtaining a total of 1:100 dilution.

$$\text{Pentosan content (\%)} = \frac{0.3438 \text{ x Furfural conc. of distillate (mg/L)}}{0.878 \text{ x mass of dry sample (mg)}} \text{ x } 100$$

- Where furfural concentration (mg/mL) = Absorbance x 6.782 x dilution factor.

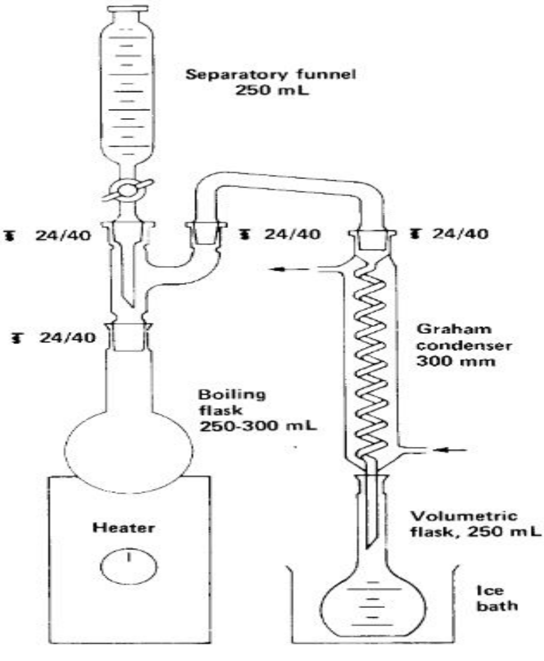


Figure 19: Pentosans distillation apparatus. Source: TAPPI (2001), T223cm-01.

To photometrically determine the pentosan content, 1ml of the 1:100 dilution solution was pipetted into a quartz cuvette of 10 mm thickness and the absorbance of the solution was measured at a wavelength of 277,5 nm (in the UV spectrum) using a spectrophotometer from the company

Beckman Coulter of the model DU 800. The spectrophotometer was first calibrated using distilled water. This determination was conducted in four repetitions for each of the raw material and the pentosans content was calculated using the formula below.

4.2.7 The determination of Klason lignin

Klason lignin usually referred to as the acid-insoluble lignin, is the insoluble residue that remains after the carbohydrate portion of plant tissues have been completely hydrolyzed by concentrated acid. It is the direct approach for determining the amount of lignin contained in plant tissues. For this purpose, 1.0 gram (oven dry) of the material was weight into a 50 ml glass beaker to which 15 ml of 72% concentrated sulphuric acid was added. The content of the 50 ml flask was allowed to digest by incubating it for 2 hours at room temperature. The incubated content was stirred with a glass rod at intervals of 10 minutes. After 2 hours of incubation, the digested content of the 50 ml glass beaker was transferred into a 500 ml Erlenmeyer flask to which 345 ml of distilled water was added, thus diluting it to 3% sulphuric acid and then refluxed for 4 hours. After 4 hours, the insoluble residue was filtered into a tared fritted-glass crucible of porosity G3. The insoluble residue was then washed with cold water, then dried to constant weight in a drying oven at a temperature of 105 ^{o}C. The crucible and its content were then cooled to room temperature in a desiccator and weighed. The determination was conducted in four repetitions for each of the raw material used in this research and the lignin content was calculated using the formula below.

Klason lignin (%) = R*100/W

Where

- R is the mass of the residue in grams.
- W is the mass of the sample in grams.

4.2.8 The determination of ash content

The ash content of plant tissues refers to the mass of the inorganic residue that is left after ignition and complete oxidation of the organic portion of plants tissues. This determination was carried out following DIN EN 14775 standard. To do this 1.0-gram (oven dry) of the material was placed in a tared porcelain crucible and heated slowly to a temperature of 550 ^{o}C for 24 hours in a muffle furnace. After the ignition, the crucible and its content were placed in a desiccator and allowed to cool to room temperature. The weight of the residue was finally measured and the ash content of the material (oven-dried at 105 ^{o}C) was calculated according to the formula below.

$$\text{Ash}\ (\%) = (W_1/W_2) * 100$$

Where: W_1 is the mass of the ash

W_2 is the mass of the oven-dried material before ignition.

4.3 The determination of the physical and mechanical properties of the manufactured particleboards

The manufactured particleboards were tested to determine their physical and mechanical properties. The following chapter presents in detail the various measurements that were conducted based on their respective standards. After producing the particleboards, they were allowed to cool to room temperature for about 24 hours, and then their edges were trimmed, sanded and cut-to-size for the testing of the physical and mechanical properties.

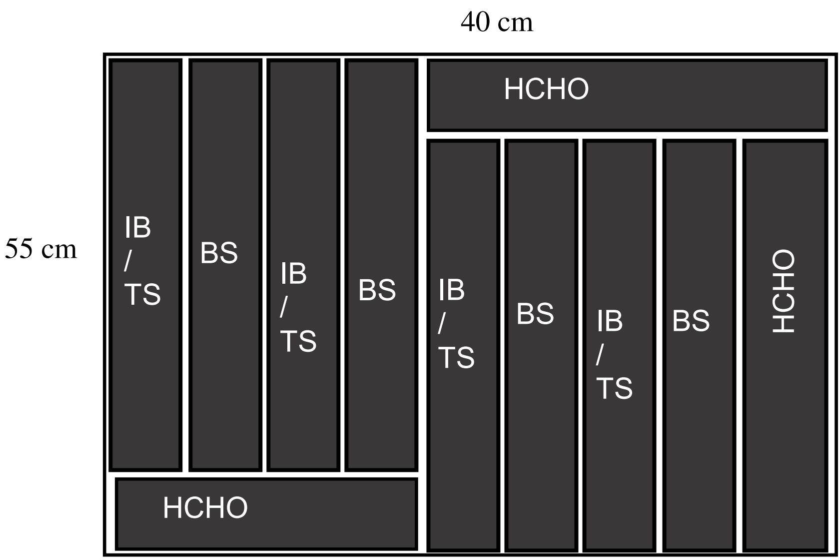


Figure 20: The cutting pattern used to obtain test pieces for the testing of the physical-mechanical properties as well as the formaldehyde emissions of the produced particleboards.

Test pieces were obtained from each of the produced particleboards based on the cutting pattern shown in figure 20. As much as possible, the test samples for internal bond strength and the water-related tests were randomly selected. The selected test pieces from each board were conditioned to constant mass in a climate chamber at a relative humidity of 65 ± 5 percent and a temperature of 20 ± 2 ^{o}C before testing.

4.3.1 The Determination of the bulk density and density profile

The Bulk density of a material is the ratio of its mass to its volume expressed in kg/m^3. According to DIN EN 323:1993, after acclimatization, the bulk density of 40 samples (with side lengths 50 ± 1 mm) from each board was determined with the help of a caliper and a scale. To do this, the length, width, and thickness of each sample were measured to the nearest 0.01 mm and the weight to the nearest 0.01 g. The thickness was measured at the center point of the intersection of the diagonals of the side lengths. The density p of each test piece expressed in kg/m^3 was calculated as shown by the formula below.

$$p = \frac{m}{l_1 \, x \, l_2 \, x \, t} \text{x } 10^6$$

Where: m is mass of test piece, in grams,

l_1 and l_2 are side lengths of test pieces, in millimeters, *t* is thickness of test piece, in millimeters.

The density profile is the variation or differences in density through the thickness of the particleboard due to varying particle characteristics and pressing conditions. This was measured using a laboratory densitometer DA-X from the company GreCon, D-31042 Alfeld –Hannover. To do this, 8 test pieces were selected randomly from each board, placed vertically in the sample holder and the density over the thickness cross-section was scanned individually.

4.3.2 The Determination of the internal bond strength

The internal bond (IB) strength also referred to as the transverse tensile strength is obtained by measuring the cohesive strength of the board by applying a tensile force perpendicular to the plane of the board. In this case, the test samples used in measuring the density profile were used in determining the IB strength of the boards according to DIN EN 319, since the measurement of density profile is a non-destructive test. To do this, each ofthe test samples were glued onto two test plates using hot melt such that the sample appears as a sandwich between the test plates. The glued test samples were then subjected to breakage one after another by loading on to the loading head of the Universal testing machine from Zwick / Roell, D-89079 Ulm. An evenly distributed tensile force was applied on the test specimen at a constant load speed so that the breakage is achieved within 60 ±30 seconds. The internal bond strength expressed in

N/mm^2 of each test sample was calculated as the ratio of the maximum force applied, to the cross-sectional area of the test sample as shown by the formula below.

$$\text{Internal bond strength } (N/mm^2) = \frac{F_{max}}{a \text{ x } b}$$

Where

- F_{max} is the maximum force applied to break the test piece, in Newton.
- a, b are the length and the width of the test specimen in mm

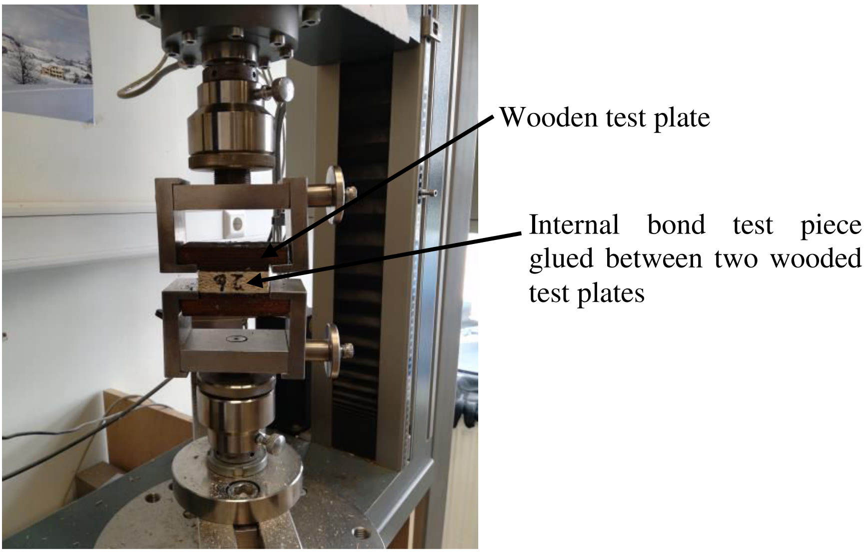


Figure 21: A test sample for internal bond test, mounted in a Universial testing machine Zwick / Roell.

4.3.3 The determination of bending strength and modulus of elasticity of the produced particleboards

The bending strength, also known as the modulus of rupture (MOR) and the modulus of elasticity (MOE) of the produced particleboards were determined by conducting three-point bending tests on test samples of each board according to DIN EN 310. Bending strength is the maximum load-carrying capacity of a material. In this case, the test measures the force required to bend the test specimen under three-point loading.

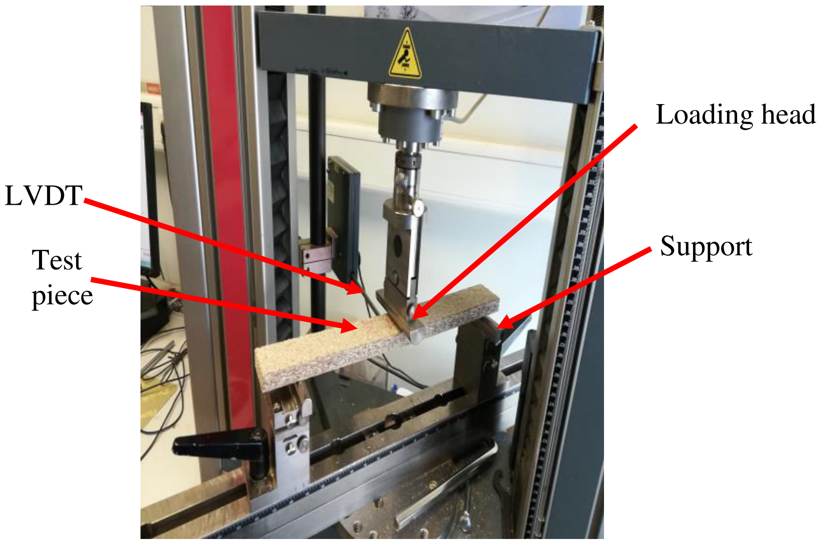


Figure 22: A picture showing the measurement of the three-point bending test.

To do this, rectangular test specimens with width (50±1) mm and length 20 times the nominal thickness of the board plus 50 mm were loaded on to the Universal testing machine from Zwick / Roell, D-89079 Ulm, and the bending strength was determined by applying a force perpendicularly to the length of the test piece at the mid-point of the test piece supported at two points as demonstrated in Figure 22.

The modulus of rupture MOR (in N/mm²) of each test piece was calculated based on the following equation.

$$MOR = \frac{3\,F_{max}\,l_1}{2\,b\,t^2}$$

Where

- F_{max} is the maximum load, in Newtons,
- l_1 is half the distance between the two supports, in millimeters,
- b is the width of the test piece, in millimeters,
- t is the thickness of the test piece, in millimeters

The modulus of elasticity was calculated by taking into account the deflection of the test samples. The “linear voltage differential transducer” (LVDT) positioned at the center point of the test piece directly beneath the loading head registers automatically the load-deflection data. From the slope of the straight-line section of the load-deflection curve (from 10% to 40% of maximum load attained), Young’s modulus is calculated which is then used to calculate the apparent MOE based on the following equation. This does not calculate the true MOE of the test piece because both shear and bending take place during the testing procedure.

$$\text{MOE (N/mm}^2\text{)} = \frac{L^3}{4wt^3} \text{ x } \Delta F/\Delta y$$

Where

- L is the length of the test piece in mm
- w is the width of the test piece in mm
- t is the thickness of the test piece in mm
- $\Delta F/\Delta y$ is the slope of the straight-line portion of the deflection curve in N/mm.

4.3.4 The determination thickness swelling and water absorption

The thickness swelling (TS) and water absorption (WA) is determined by measuring the increase in thickness and increase in mass respectivey of the test pieces after complete submersion in water. The TS and WA were determined according to EN 317: 1993, and DIN 52351 respectively. To do this, eight test pieces (with dimension 50 mm x 50 mm (±1mm) x thickness (mm)) selected randomly from each board were submerged vertically in a water bath filled with clean water (of neutral pH) and stored at room temperature for 2 hours and subsequently for 24 hours. The thickness and the mass of each test piece were measured prior to submersion in water to the nearest 0.01 mm and 0.01 g respectively. The thickness (measured at the intersection of the diagonals) and the mass of each test piece were measured after two hours of immersion to obtain the 2 hours values. The same procedure was repeated for a further 22 hours to obtain the 24 hours values. The TS and WA of the test pieces were determined based on equations 1 and 2 respectively:

$$Gt\ (\%) = \frac{t_2 - t_1}{t_1} \times 100 \text{ ----------(1)}$$

Where

- G_t is the thickness swelling of each test piece expressed in percentage
- t_1 is the thickness of the test piece before submersion in water, in mm
- t_2 is the thickness of the test piece after submersion in water, in mm

$$W_A\ (\%) = \frac{m_2 - m_1}{m_1} \times 100 \text{ ---------- (2)}$$

Where

- W_A is the water absorption of each test piece after submersion in water, expressed in percentage
- m_1 is the mass of test piece before submersion in water, in grams

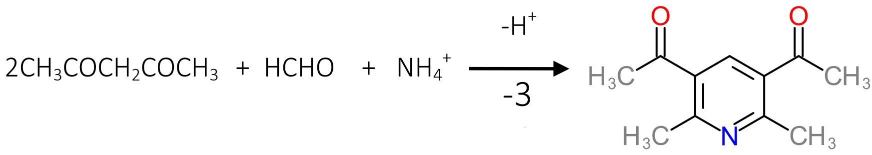
- m_2 is the mass of test piece after submersion in water, in grams.

4.4 The determination of formaldehyde release of the of the produced particleboards

The formaldehyde emission of wood-based composites can be determined using various methods. These methods include the flask method (DIN EN717-3), the perforator method (DIN EN 120), the chamber method (DIN EN717-1) and the gas analysis method (DIN EN 717-2). The chamber method, usually known as the reference method, is the generally accepted method for testing formaldehyde emissions of wood-based panels in Europe. Based on this method, the formaldehyde emission must not exceed 0.1 ppm (0.124 mg/m³) for panels that are used for indoor applications. This maximum limit is known as the E1-limit. However, this method is generally expensive and time-consuming to execute. For expedience purposes, derived methods such as the gas analysis, flask and perforator methods are being used in the industry for production control. It is important to note that to ensure compliance with the E1-limit for particleboards, the rolling average tested by the perforator method over a period of 6 months from the internal factory production control must not exceed 6.5 mg/100 g of the dry board. The following chapters explain in detail the methods used in determining the formaldehyde properties of the produced particleboards.

4.4.1 The determination of formaldehyde release by the Flask method

According to this method, about 20 grams of the test sample of dimensions 25 x 25 mm were made to suspend from the lid of a sealed-500 ml polyethylene flask containing 50 ml of demineralized water. The sealed polyethylene flask and its content were placed in a preheated chamber at a constant temperature of 40 °C for 3 hours. This procedure was repeated with other test samples for the duration of 24 hours to determine the 24 hours flask values. After 3 and 24 hours, the formaldehyde concentration of the aqueous solution was determined by the acetylacetone method in which 10 ml of the solution in the 500 ml flasks were transferred into 50 ml test bottles and added 10 ml each of acetylacetone and ammonium acetate. The 50 ml test bottles were stoppered and placed in a warm water bath at a temperature of 40 °C for 15 minutes and then cooled to room temperature in a dark chamber. The concentration of formaldehyde contained in the aqueous solutions was then determined colorimetrically at a wavelength of 412 nm with the help of a spectrophotometer from Biochrom, Libra S12. The colorimetric determination is based on the Hantzsch reaction, a test highly specific for the presence of formaldehyde. The formaldehyde in the aqueous solution reacts with ammonium ions from Ammonium acetate and acetylacetone to yield diacetyl-dihydro lutidine (DDL) with its characteristic greenish-yellow color (Nash, 1953).



Acetylacetone Formaldehye Ammonium ion DDL (greenish-yellow)

The Hantzsch reaction scheme for the formation of diacetyl-dihydrolutidine. Source: Nash, 1953.

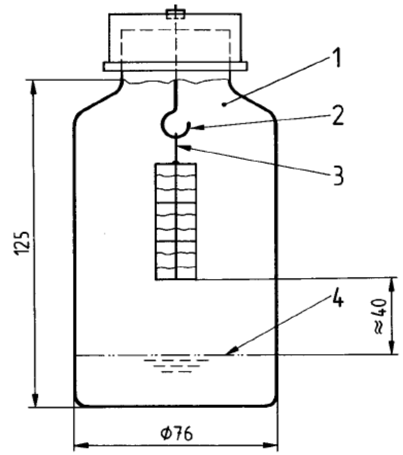
The extinction values (photometer readings) were used to calculate the formaldehyde release per 1000 grams of oven-dried boards according to the mathematical formula below. A blank solution (without test samples) was used to calibrate the spectrophotometer. The determination was conducted in three duplicates for each panel variant.

$$F_V = \frac{(A_S - A_B) \times f \times 50 \times 100\,(100 + H)}{m}$$

Where

- M is the mass of test piece or sample, in grams
- F_v is the flask value in milligrams per 1000g of oven dry board
- A_S is the absorbance or extinction value of the analyzed solution from flask
- A_B is the absorbance or extinction value blank solution
- *f* is the slope of calibration curve of standard formaldehyde solution, in mg/ml.

- H isthe moisture content of test pieces or sample, in percent.



1 500-ml-polyethylene flask with a lid

2 Hook made of stainless steel

3 Gummiband

4 Water surface

Figure 23: Test equipment for the flask method (adopted from EN 717-3: 1996).

4.4.2 The determination of formaldehyde by the perforator method

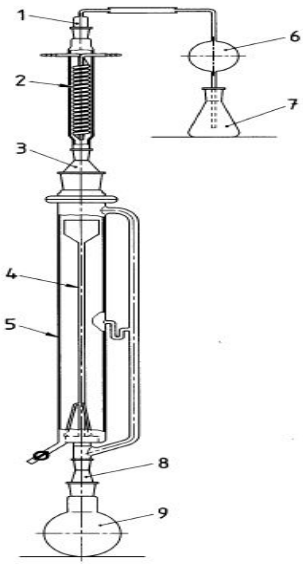
Formaldehyde emission of wood-based panels can be measured by determining the amount of extractable formaldehyde of the panels. To do this, about 110 grams of test samples (of the dimensions of 25 mm x 25 mm x thickness in mm) were placed in a 1000 ml Erlenmeyer flask to which 600 ml of toluene was added. The flask and its content were connected to the lower end of the perforator attachment filled with about 1000 ml of distilled water. The distilled water was filled such that a space of about 30 mm was left between the water surface and the siphon outlet. The Dimroth condenser and the gas absorption tube were then connected, with one end of the absorption tube connected to a 250 ml flask (overflow flask) containing about 100 ml of distilled water as shown in figure 24. The setup was allowed to run for 2 hours commencing from the moment the first bubble passed through the filter-insert. After 2 hours, the setup is cooled to room temperature and the water contained in the perforator and the overflow flask were transferred into a 2000 ml volumetric flask and made up to the 2000 ml mark by rinsing the perforator twice with about 700 ml of distilled water. The extraction was carried out in three duplicates for each panel variant and the formaldehyde concentration of the aqueous solution was determined colorimetrically based on the acetylacetone method. The results are expressed in mg/100 g of oven-dried boards based on the mathematical expression below. A blank value is also determined in parallel by using 10 ml of distilled water and following the steps of the acetylacetone method.

$$P_v = \frac{(A_s - A_B).\,f.(100+H).V}{m_H}$$

Where

- m_H is the mass of test samples, in grams
- P_V is the amount of extractable formaldehyde in milligrams per 100g of oven-dried board

- A_S is the absorbance or extinction value of the analyzed solution from flask
- A_B is the absorbance or extinction value of blank solution
- *f is* the slope of calibration curve of standard formaldehyde solution, in mg/ml.
- H is the moisture content of wood-based panel, in percent.
- V is the volume of volumetric flask (2000 ml).



1: Conical adaptor 29/32

2: Dimroth condenser

3: Conical adaptor, case 45/40, core 71/51

4: Filter insert

5: Perforator attachment

6: Double bulbed tube

7: 250 ml conical flask

8: Conical adaptor, case 29/32, core 45/40

9: Erlenmeyer flask, 1000 ml

Figure 24: Extraction apparatus used for perforator method (adopted from EN 120: 1992).

4.5 Chemical additives used in producing the particleboards

4.5.1 Adhesives

Two different commercial-grade UF-resins of the kaurit group K350 liquid and K340 liquid from the company BASF AG, D-67056 Ludwigshafen were used. In addition to the UF-resins, a commercial-grade Isocyanate-based resin – PMDI (I-Bond PB em 4352) supplied by the company Huntsman Holland BV, 103197 KG Botlek-Rotterdam Netherland was also used. A fourth resin, based on animal protein - blood albumin, was also used as a binder system in the production of the particleboards. Table 3 presents the properties of the various binder systems used in this research.

Table 3: The properties of the binder systems used in this research.

Resin type	**UF**	**UF**	**PMDI**	**Blood albumin**
Product name	K350 liquid	K340 S liquid	I-Bond PB em 4352	Blood albumin
Colour	White	White	Brown	Brown
Solids content	66%	66%	100%	54%
pH	8.5	8.3	n/a	7,4
Viscosity (at shear rate 511.3 s^{-1})	322 mpa.s	667 mpa.s	280 mpa.s	495mpa.s

At a temperature of 25 °C

4.5.2 Auxiliary additives

As mentioned earlier, hardeners are generally used in the production process of particleboards to create an acidic medium, which accelerates the curing process of UF resin and shortens the press-time. Hardeners are generally used in the amount of 1-2 percent depending on the pH and buffering capacity of the furnish. Ammonium sulfate (40% solids content) was used as a hardener for the UF-resins. To improve on the hydrophobicity of the produced particleboards, two types of paraffin-based hydrophobic agents, hydrowax 138 and hydrowax pro A18 both of 50% solids contents, from the company Sasol GmbH were used.

4.6 The requirements of the properties of particleboards

4.6.1 Requirements of the physical and technological properties

As earlier mentioned, the produced particleboards were tested for their physical-technological properties and evaluated in accordance with the German institute for standardization e,V. The table below presents the minimum requirements for the physical and technological properties of particleboards of various types (P1, P2, and P4) according to DIN EN 312: 2010-12.

Table 4: The DIN EN 312: 2010 minimum requirements for particleboards of thickness between 13 mm to 20 mm.

Properties	Test Norm	Units	P1	P2	P4
IB	EN 319	N/mm^2	0.24	0.35	0.35
MOR	EN 310	N/mm^2	10	11	15
MOE	EN 310	N/mm^2	-	1600	2300
Surface soundness	EN 311	N/mm^2	-	0.8	-
Thickness swelling 24 h	EN 317	%	-	-	15

- P 1: particleboards for general purpose application for use in dry condition.
- P 2: particleboards for interior applications (including furniture) for use in dry conditions.
- P 4: Particleboards for load-bearing applications for use in dry condition.

4.6.2 The requirements for formaldehyde emissions

Besides the physical and mechanical properties, particleboards can be classified according to their formaldehyde emission levels. On the European level, the formaldehyde emission from particleboards has been standardized. Wood-based panels that are to be used for indoor applications must conform to the European emission class E1 requirement. According to the E1 emission class, the formaldehyde emission must not exceed the limit of 0.1 ppm when tested by the European chamber method (EN 717-1). However, the derived method for factory production control, the Perforator method (EN 120) has a direct correlation with the E1 limit. The moisture content of the board must be corrected to 6.5% for the correlationof the perforator values to be true. Table 4.3 shows the formaldehyde limit values for the emission class E1 and the correlation value of the derived method.

Table 5: The limit values of formaldehyde emission class E1 tested by the EN 717-3 and EN 120.

Emission class	Test Methods	
	Chamber method (EN 717-1)	Perforator method (EN 120)
E1	≤ 0.1 ppm	≤ 8 mg/100 g

4.7 The manufacture of particleboards on a laboratory scale

The following chapters describe procedure used in the development of the particleboards on a laboratory-scale. The chapters include a detailed description of the types of equipment, production parameters, and the processing steps that were involved in the manufacturing process of the particleboards of the various raw materials. The process sequence for the production of the particleboards in this study is consistent with the manufacturing sequence commonly used in the industry.

4.7.1 The development of the particleboards based on cacao tree prunings and umbrella tree wood as raw materials

The development of particleboards of umbrella tree wood and cacao tree prunings was carried out in two phases.The first phase focused on the feasibility of producing PB based on the raw materials and the second phase was focused on investigating the effect of the coarser core layer chips of the various raw materials on the properties of the boards. For this purpose, the wood chips of the surface and the core layers of the various raw materials were blended separately with UF-resin of reduced molar ratio of the group K 340 in a laboratory-scale rotating drum mixer with the help of an air-pressure atomizer. The wood chips of the Norway spruce (*Picea abies*) were also blended to serve as a reference. The resin load was based on the oven-dry weight of the furnish. As much as possible, the rotating drum mixer equipped with an air pressure atomizer uniformly blended the wood chips for about 10 minutes. A hardener, ammonium sulfate (40% solid content v/v) in the amount of 1.5% based on the solid content of the resin was added to the glue. The hydrophobic agent, hydrowax 138 (50% solid content v/v) from the company Sasol GmbH in the amount of 1.5% based on the oven-dry weight of the wood chips was also blended with the wood chips. Table 6 shows the production parameters for the development of the particleboards.

After the blending was completed, the resinnated furnish was formed into a mat by hand with the help of a transparent-plastic laboratory-scale mat former of dimensions 605 mm x 455 mm. The surface layers and the core layer were of the ratio 40:60 percent the total mass of each of the board. The mat was formed on metal press plates of dimensions 800 mm x 600 mm overlaid with PTFE (Polytetrafluoroethylene) film to serve as a release agent to allow for easy removal of the boards after the press opens.

Table 6: The production parameters for the development of the particleboards based on cacao tree prunings and umbrella tree wood in a laboratory scale.

Raw material	**UF-resin load [%] SL / CL**	**Hardener content [%]**	**Hydrophobic agent content [%]**	**Target thickness [mm]**	**Target densities [kg/m^3]**	**Press time [min]**	**Press temp. [ºC]**
UTW, CTP, Spruce wood	10 / 8.5	1.5	1.5	20	650/550/450	4	200

Umbrella tree wood (UTW), Cacao tree prunings (CTP), Surface layer (SL), Core layer (CL).

The resulting mat was evenly pre-pressed with a wooden board to reduce its thickness and then hot-pressed to a target thickness of 20 mm to achieve panels of the targeted densities (see table 6). The boards were pressed at a temperature of 200 °C and a pressure of 200 bar for 4 minutes. Three boards were produced for each board variant, giving a total of 27 boards. The resulting particleboards were conditioned overnight to cool down to room temperature, then sanded and sawn to the dimension as required by the testing standard for the physical-mechanical properties as well as formaldehyde emissions.

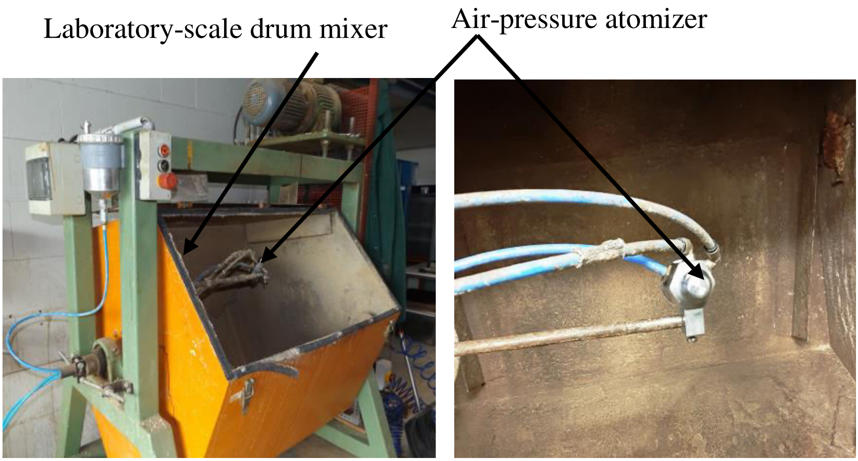


Figure 25: Laboratory-scale drum mixer equipped with an air-pressure atomizer for blending wood chips for particleboards.

The dependence of the physical-technological properties on the coarseness of the wood chips of the core layer of the raw materials; cacao tree pruning and umbrella tree wood was the focus of the next phase of the PB manufacturing process. It investigates the effect of increasing the coarseness of the core layer chips on the physical and technological properties of the particleboards of umbrella tree wood and cacao tree prunings.

Figure 26: Hot press from the company Siempelkamp, D-47803 Krefeld (Left), and pre-pressed mat ready for hot-pressing (Right).

It has been scientifically established that the dimensions of the wood chips used in particleboard production significantly affect the physical-mechanical properties, surface quality and processing properties of the resulting Particleboards (Istek *et al*., 2018; Youngquis J.A., 1999; Frybort *et al*., 2008).

For this purpose, detailed sieve analysis of the wood chips of the core layer was conducted as described in chapter 4.2.1. The fractional compositions of the chips of cacao tree prunings and umbrella tree wood were compared to the fractional composition of the industrially produced core layer wood chips from the company Pfleiderer GmbH. From the analysis, it was found that the laboratory-produced wood chips of the core layer for both cacao tree prunings and umbrella tree wood were not as coarser as the industrially-produced wood chips. For this reason, the fractional compositions of the chips of the study materials (umbrella tree wood and cacao tree prunings) were adjusted to contain about 10 percent of chips with sizes greater than 6 mm (see Figures 27 and 28). Finally, the core layer chips with the adjusted fractional composition were used to produce another series of three-layered particleboards with the same production parameters as shown in Table 7. Three boards were produced per board variant thus giving 18 boards in total. The resulting boards were also conditioned overnight to cool to room temperature, then were sanded and sawn to the dimensions for evaluating their physical-technological properties as well as formaldehyde emissions.

Table 7: The production parameters for the development of the particleboards based on coarser core layer chips of cacao tree prunings and umbrella tree wood in a laboratory scale.

Raw material	**UF-resin load [%] SL/CL**	**Hardener content [%]**	**Hydrophobic agent content [%]**	**Target thickness [mm]**	**Target densities [kg/m³]**	**Press time [min]**	**Press temp. [°C]**
UTW, CTP	10/8.5	1.5	1.5	20	650/550/450	4	200

Umbrella tree wood (UTW), Cacao tree prunings (CTP), Surface layer (SL), Core layer (CL)

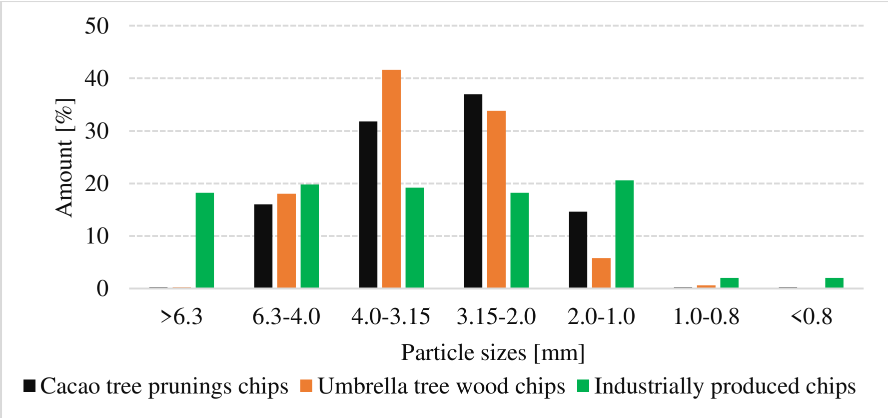


Figure 27: A comparison of the fractional composition of the core layer wood chips of cacao tree prunings and umbrella tree wood with the industrially produce core layer wood chips.

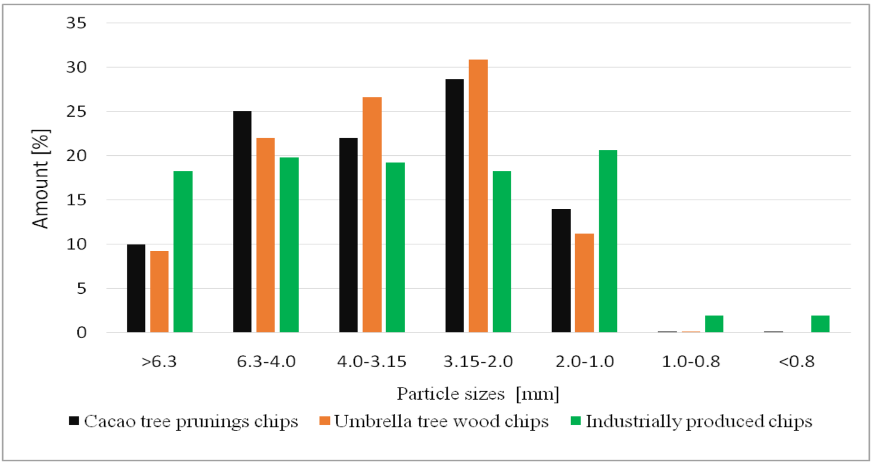


Figure 28: The fractional composition of the adjusted particle sizes of the chips of cacao tree prunings and umbrella tree wood compared with the industrially produced wood chips of the core layer.

4.7.2 The development of non-formaldehyde bonded particleboards based on cacao tree prunings and umbrella tree wood as raw materials

As discussed in chapter 1, the formaldehyde emission of wood-based composites is a serious concern to the manufacturers as well as the consumers. Regulations on the formaldehyde emissions of wood-based panels in Europe is becoming more and more stringent. For this reason, an investigation into the possible feasibility of producing particleboards with non-formaldehyde containing binder systemsby utilizing umbrella tree wood and cacao tree prunings as raw materials was the focus of this manufacturing series.

To do this, three-layered particleboards of the chips of umbrella tree wood and cacao tree prunings were produced on a laboratory scale. In addition to these, particleboards of a mixture of the chips of both materials were produced. The combination ratio of both materials was 50:50 percent (w/w) on both the surface and the core layers. On the one hand, an isocyanate-based resin – PMDI (I-Bond PB em 4352) supplied by the company Huntsman Holland BV, 103197 KG Botlek-Rotterdam Netherlands was used to glue the chips. On the other hand, an animal protein-based binder system, blood albumin, was used. The blood albumin was supplied by Fritz Häcker GmbH & Co. KG in the form of whole blood powder (Vollblutpulver). Hydrowax 138 (in the amount of 1.5% based on the oven-dry weight of the wood chips) was also blended with the chips. The process steps and equipment involved in the manufacturing process are the same as described in chapter 4.7.1. The mats were hot-pressed at a temperature of 200 °C and a pressure of 200 bars. Particleboards of six variants were produced, thus 18 boards in total.

Table 8: The manufacturing conditions for the non-formaldehyde bonded particleboards produced in a laboratory scale. Surface layer (SL), Core layer (CL).

Variant	**Resin type**	**Resin load [%] SL CL**	**Hydrowax 138 [%]**	**Target thickness [mm]**	**Target densities [kg/m³]**	**Press time factor [s/mm]**
1. UTW 2. CTP	Blood albumin	10.0 8.0	1.5	20	650, 550	12
3. UTW-CTP mixture	PMDI	3.0 3.0	1.5	20	650, 550	12

Umbrella tree wood (UTW), Cacao tree prunings (CTP) and Umbrella tree wood-Cacao tree prunings (UTW-CTP mixture)

The produced boards were sanded and then sawn to size after conditioning to room temperature overnight.The boards were finally evaluated for their physical-mechanical properties and formaldehyde emissions according to the standards for the respective tests. The production conditions of the particleboards are shown in Table 8.

4.7.3 The development of hybrid particleboards based on the chips of the various wood materials and the chips of bean shell

The experimental work presented in this chapter evaluates the technical possibility of the annual plant residue of the rainy season (bean shells) to serve as a substitute raw material for wood in both the core and surface layers of particleboards. It also evaluates how the properties of the particleboards of the various wood materials in combination with the bean shells differed from one another. To do this, three-layered particleboards of three variants were produced. For the variants 1 and 2, the surface layers of the particleboards of the chips of the raw materials; cacao tree prunings, umbrella tree wood and the industrially produced wood chips (from the company Pfleiderer GmbH) were completely replaced with the chips of bean (*Phaseolus vulgaris*) shells. Thus, a 40 percent substitution of the various wood chips as illustrated in Figure 29.

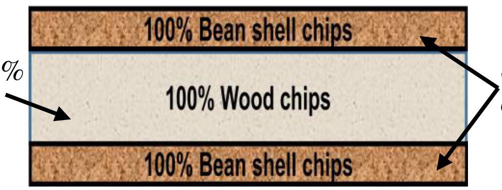


Figure 29: The schematic illustration the substitution of wood chips with the chips of beans shells on the surface layer of the particleboards of variant 1 and variant 2.

The chips for the manufacture of the particleboards of variant 1 were bonded with PMDI (I-Bond PB em 4352) supplied by the company Huntsman Holland BV. The resin loads of 5% and 3% were used on the surface layer chips (bean shells) and the core layer chips of the various wood materials respectively. For the manufacture of the boards of the second variant, UF- resin of the group K340 was used to glue the chips. The resin load was 10% on the chips of the surface layers and 8.5% on the chips of the core layer. The amount of resin used (resin load) was calculated based on the oven-dry weight of the chips.

In the manufacture of the third variant of the hybrid particleboards, 20% of the wood chips of the core layer and surface layers were substituted with the chips of bean shell as illustrated in figure 30. Thus, a 40 percent (w/w) substitution of the various wood chips with the chips of bean shells. The chips were then blended with PMDI (I-Bond PB em 4352 supplied by the company Huntsman Holland BV). In this case, the resin load was 4% for the chips of the surface layers and 4% for the chips of the core layer. After the gluings, the mats were formed, pre-pressed and then hot-pressed to a density of 650 kg/m^3 at a temperature of 200 ^oC and pressure of 200 bar following the process steps and apparatus described in chapter 4.7.1. A total of 27 boards were produced for all of the panel variants and raw material combinations. The detailed experimental design for the production of each of the panel variants is presented in Table 9.

Figure 30: Schematic illustration of the substitution of the wood chips with the chips of bean shells in both the core and surface layersof the particleboards of variant 3.

Finally, the manufactured particleboards weresanded and cut-to-size for the evaluation for their physical and mechanical properties as well as their formaldehyde release.

Table 9: Detailed production conditions for the hybrid particleboards based on chips of the annual plant residue, bean shells and the chips of the various wood materials.

Variant	Bean shell chips substitution level [%]		Resin type	Resin load [%]		Hydrowax 138 [%]	Target thickness [mm]	Press time [min.]
	SL	CL		SL	CL			
Variant 1	40 %	0.0 %	PMDI	5.0	3.0	2.0	20	4
Variant 2	40 %	0.0 %	UF-resin	10	8.5	2.0	20	4
Variant 3	20 %	20 %	PMDI	4.0	4.0	2.0	20	4

Surface layers (SL), Core layer (CL), the wood materials of each variant: Cacao tree prunings, umbrella tree wood and industrially produced wood chips.

4.7.4 The manufacture of UF-resin bonded particleboards based on the chips of umbrella tree wood and bean crop residues of the dry season

Within the context of this work, the beans crop (*Phaseolus vulgaris*) residues (BCR) of the dry season are the residues that are generated through the harvesting and primary processing of common bean during the dry season in Cameroon. Unlike the bean crop residues of the rainy season, which are composed of 100 percent bean shells, the residues of the dry season are composed of a mixture of the bean straws and bean shells because the harvesting and primary processing methods differ with the two seasons of Cameroon as explained in chapter 4.1.2. The work presented in this chapter examines the technical possibility of producing particleboards of the DIN EN 312-2 (2010) standard by substituting 10%, 15% and 20% of the chips of umbrella tree wood with the chips of bean crop residues of the dry season in the core layer of the boards. To this end, a higher molar ratio UF-resin of the group K350 was used to separately glue the core layer and the surface layer chips.The chips were also blended with a hydrophobic agent, hydrowax 138 from Sasol GmbH. The resinated chips were formed into three-layered mats, the mats were pre-pressed and then hot pressed to a density of 650 kg/m^3 at a press-temperature of 200 ^{o}C following the process steps as described in chapter 4.7.1. For comparison purposes, one variant of particleboard was produced exclusive-ly from the bean crop residues according to the experimental design summarized in Table 10.

Table 10: The experimental design for the development of the UF-resin bonded particleboards based on the chips of umbrella tree wood and bean crop residues of the dry season.

Board type	% of BCR	% of UTW	Resin load (K350) (%) SL / CL	Target density (kg/m^3)	Target thickness (mm)	Press time factor (s/mm)
100% BCR	100	0	11 / 9	650	20	12
10 % BCR	10	90	10 / 8.5	650	20	12
15 % BCR	15	85	10 / 8.5	650	20	12
20 % BCR	20	80	10 / 8.5	650	20	12

*Core layer (CL), surface layer (SL), Bean crop residues (*BCR) *of the dry season, umbrella tree wood (UTW.*

Three board replicates were produced for each of the substitution levels giving 12 boards in total. The final boards were conditioned to room temperature prior to edge trimming and sanding. Finally, the produced boards were cut-to-size and evaluated for their physical and mechanical properties as well as formaldehyde release.

4.8 Statistical evaluation of the results

The particleboards produced in this research were evaluated for their physical and mechanical properties, as well as their formaldehyde release. The results of the physical-mechanical properties are presented as the arithmetic mean with standard deviation (STD) in the bar diagrams.

Meanwhile, the results for the formaldehyde properties are only presented as mean values. With the help of the analysis of variance test (ANOVA) and the Tukey HSD test, the results were tested for the significant differences between the variants of the particleboards. All statistical analyses were conducted with STATISTICA 13.0 software package at a 95% significance level.

5.0 Results and discussion

This work investigates the technical possibilities of using the residues of the annual crop; bean (*Phaseolus vulgaris*), cacao tree (*Theobroma cacao*) prunings, and the wood of the fast-growing tree species; umbrella tree (*Musanga cecropoides*) as raw materials for the production of three-layered particleboards with physical-technological properties that meet the requirements of the standard DIN EN 312-2010. To accomplish this, a detailed investigation into the physical and chemical characteristics of the raw materials was essential. The produced panels were evaluated for their physical-technological properties as well as their formaldehyde emission and the results of the systematically collected data are presented in the following chapters.

5.1 The physical and chemical characterization of the raw materials

5.1.1 Fractional composition and bulk density

The bulk density of the chips of raw material is a direct reflection of its natural density and can influence the physical and technological properties of particleboards. The measurement of bulk density determines the minimum density to which the material can be compressed for the formation of the board to occur. For example, to produce particleboards with high enough strength the bulk density of the raw material must be well below the target density of the panel. The raw material should be able to compress to at least 5% (in practice about 50%) above its natural density

(Thoemen *et al.*, 2010). A material with high bulk density has a lower degree of compaction during the pressing process, which has an adverse influence on the tensile strength of the resulting panel (WKI, 2008).

Table 11 presents the bulk densities of the chips of the core layer and the surface layers of the study's raw materials. For comparison purposes, the bulk densities of spruce wood and the industrially produced wood chips from the company Pfleiderer GmbH were also measured. The residual moisture content of the materials ranged from 2.8% to 5.0%.

Table 11: The bulk densitiesof the chips of the study's raw materials in comparison to spruce wood and the industrially produced wood chips. Bean crop residue (BCR).

	Bulk density (kg/m³)	
Raw materials	**Core layer**	**Surface layer**
Umbrella tree wood	92	127
Cacao tree prunings	124	177
BCR (rainy season)	70	93
BCR (dry season)	71	96
Spruce wood	119	170
Industrially produced wood chips	139	138

In terms of the core layer chips, it can be seen that the industrially produced wood chips had the highest bulk density (139 kg/m^3), followed by the chips of cacao tree prunings (124 kg/m^3), the chips of spruce wood (119 kg/m^3) and the chips of umbrella tree wood (92 kg/m^3).

The measurement of the surface layers chips, on the other hand, revealed the highest bulk density (177 kg/m^3) for cacao tree prunings, followed by spruce wood (170 kg/m^3), industrially produced wood chips (138 kg/m^3), and the umbrella tree wood (127 kg/m^3). The chips of the bean crop residues of both seasons for both the surface layers and the core layer yielded the least bulk densities compared to the chips of the wood materials. This means that for the manufacture of particleboards of a given density, more chips (volume-wise) of beans crop residues and umbrella tree wood will be required compared to the cacao tree prunings and the reference materials to achieve particleboards of the same thickness. A higher chip-to-chip contact can be achieved even at low panel density for the bean crop residues and the umbrella tree wood, which means the manufacture of lightweight particleboards may be possible with these materials compared to the cacao tree prunings.

Particleboard properties such as IB, MOR, MOE and TS are significantly influenced by the size characteristics of the wood chips (Istek *et al*., 2018; Youngquis J.A, 1999; Frybort *et. al*., 2008; Lias *et al*., 2014). Shorter but thick particles will significantly reduce the MOR and MOE of particleboards (Mundy and Bonfield, 1998). On the other hand, such particle sizes will increase the IB and reduce the TS of the particleboards (Rackwitz, 1963; Hutschneker, 1975; Nazerian *et al*., 2011). For this reason, the size distribution of the laboratory-produced core layer and surface layers chips of cacao tree prunings, spruce wood, and umbrella tree wood were determined and compared with the size distribution of the industrially produced wood chips. The size distributions of the chips of the bean crop residues of both seasons were also determined and compared against the size distribution of the industrially produced wood chips.

Figure 31 compares the size distribution of the laboratory-produced wood chips of the core layer with the industrially produced core layer wood chips. It can be seen that the industrially produced wood chips were more or less uniformly distributed within the various size ranges (≥6.3 to 1.0 mm) whereas the laboratory-produced chips of the core layer of the various wood materials had at least 65% of their chips within the size range from 4.0 mm to 2.0 mm. In addition, the laboratory-produced wood chips of the core layer had no chips with sizes equal to or greater than 6.3 mm, except for the chips of spruce wood, with only about 3%.

In the same light, the size distribution of the wood chips of surface layers (figure 32) revealed that the industrially produced wood chips had a much higher amount of finer particles (58%) with sizes less than ≤ 0.8 mm) compared to the laboratory-produced wood chips.

The laboratory-produced wood chips of the surface layer were mostly of the sizes between 1 mm to 2 mm, with about 58% for the chips of cacao tree pruning, 71% for the umbrella tree wood chips and 72% for the chips of spruce wood. The percentage of chips with sizes greater than 2 mm was negligible for all the materials.

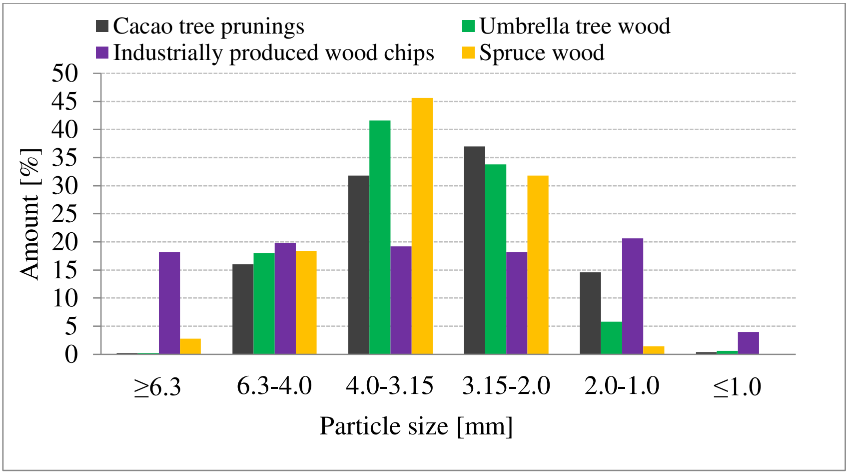


Figure 31: The size distribution of the laboratory-produced wood chips of the core layer compared with the industrially produced wood chips (Core layer).

Figure 33 presents the fractional composition of the core layer chips of the bean crop residues of the rainy season (bean shell) and the bean crop residues of the dry season compared against the industrially produced wood chips. It was found that the chips of the bean crop residues of both seasons were mostly of sizes between 4.0 mm to 6.3 mm, which is 45% of the chips of bean shells and 62% of the chips of the bean crop residues of the dry season. Their distributions within the other size ranges were less than 20% except for the chips of bean shells, in which 31% of its chips were found to be within the size range of 3.15 mm and 4.0 mm. It was also found that the sizes of the chips of the bean crop residues of both seasons were all less than 6.3 mm.

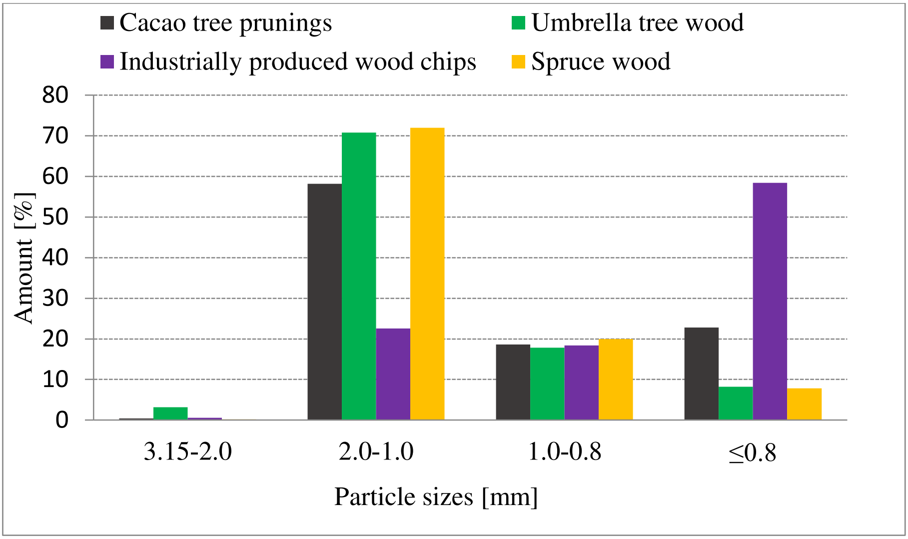


Figure 32: The size distribution of the laboratory- produced wood chips of the surface layers compared with the industrially produced wood chips.

Figure 34 shows the fraction composition of the surface layers chips of the bean crop residues of rainy season (bean shells) and the dry season compared against the industrially produced wood chips. It can be seen that the fractional composition of the chips of the beans crop residues of the both seasons was identical in all of the size ranges. Unlike the industrially produced wood chips, more than 70% of the chips of the bean crop residues of both seasons were of sizes between 1.0 mm – 2.0 mm. The industrially produced wood chips of the surface layer were much finer than the surface layer chips of the bean crop residues.

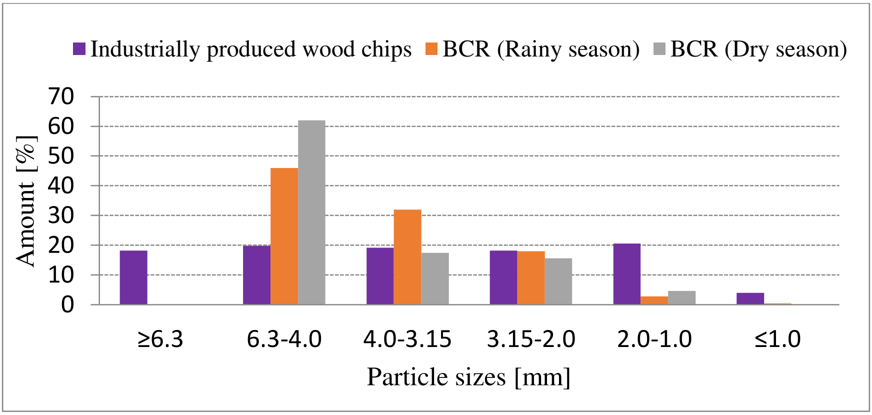


Figure 33: The fractional composition of the laboratory-produced core layer chips of the bean crop residues (BCR) of the rainy season and the dry season compared with the industrially produced wood chips of the core layer.

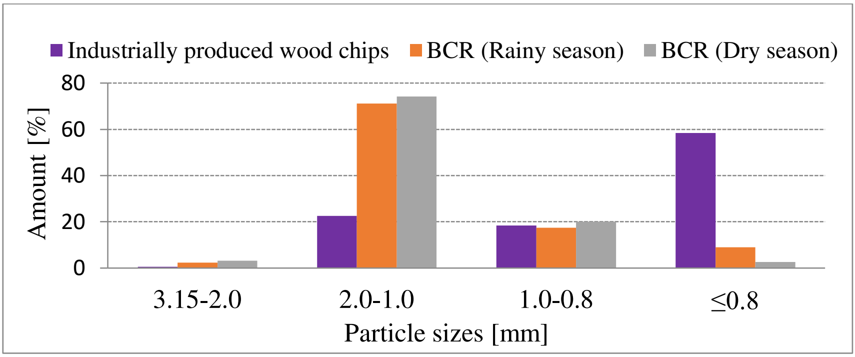


Figure 34: The fractional composition of the laboratory-produced chips of the surface layers of the bean crop residues (BCR) of the rainy season and the dry season compared with the industrially produced wood chips of the surface layer.

It is important to note that the chips with sizes less than 0.8 mm for the laboratory-produced chips were considered as dust and thus were excluded from further analysis in this dissertation.

5.1.2 pH and buffering capacities of the raw materials

The pH and buffering capacity of the raw material are important factors in the production of particleboards as they are fundamental to the gluing process. For example, pH-sensitive adhesives such as UF-resin cure faster in acidic pH conditions compared to an alkaline environment. The pH of raw material may alter the pH of the glue at the interface between the raw material and the adhesive and change the curing process of the adhesive (John and Niazi, 1980; Maloney, 1977). Extreme pH-values of wood have been known to inhibit the formation of strong adhesive bonds in the production process of wood-based panel (Bryant, 1968; Chen, 1970). For these reasons, the acidic properties, the pH and buffering capacities of the cold-water extracts of the raw materials were determined (see chapter 4.2.3). Table 12 presents the pH and buffering capacities of the cold-water extracts of the raw materials in comparison with spruce wood. It can be seen that the pH-values of umbrella tree wood, cacao tree prunings, and the bean crop residues of the dry season were close to neutral. The pH-value of the bean crop residues of the rainy season was 5.6 and differed only slightly from the pH-value (5.4) of the reference material, spruce wood. However, the pH-value of the chips of spruce wood was not consistent with the values of between 4.5 to 4.9 reported in the literature by Roffael *et al.*, 1992. This difference might be as a result of the difference in the growth condition of the tree or the part of the tree from which the samples were obtained.

The umbrella tree wood and cacao tree prunings exhibited lower buffering capacities of values 0.54 mmol NaOH/100 g and 0.57 mmol NaOH/100 g respectively compared to the rest of the raw materials. The bean crop residues of both seasons showed very high buffering capacities, 14.27 mmol NaOH/100 g for the bean crop residues of the rainy season and 6.24 mmol NaOH /100 g for the dry season bean crop residues.

Table 12: The pH and buffering capacities of the cold water extracts of the raw materials compared with the reference material, spruce wood. Bean crop residues (BCR).

Raw materials	**Cold water extractives**	
	pH values	**Buffering capacity** [mmol NaOH/100 g of material]
Umbrella tree wood	6.2	0.54
Cacao tree prunings	6.8	0.57
BCR (rainy season)	5.6	14.27
BCR (dry season)	6.4	6.24
Spruce wood	5.4	0.74

The buffering capacity of the bean crop residues of the rainy season (bean shells) was more than two times higher than that of the bean crop residues of the dry season. Given the pH-values of the bean crop residues of the both seasons, couple with their high buffering capacities, the bonding process of their chips with UF-resin might be problematic or delayed which may result in unfavorable bonding strengths. However, this could be solved by adding a suitable catalyst or hardener and extending the hot pressing time (Dunky and Niemz, 2002).

5.1.3 Water-soluble extractives content of the raw materials

Wood extractives play significant roles in the bonding process of wood-based panel as they determine the pH and the wettability of the surfaces of the wood chips. For example, wood with high amounts of hydrophobic extractives such as oil, fats, and resins can adversely affect the wettability of the wood with adhesives (Roffael and Schäfer, 2002). In addition, extractives of low pH can accelerate the curing of acid curing UF-resin and prolong the curing of PF-resin (alkaline curing adhesive) (Roffael, 2015). For this reason, the hot and cold-water extractives content of the raw materials were determined (see chapters 4.2.2 and 4.2.4) and the results are presented in Table 13.

The results indicated that the umbrella tree wood contains lesser amount of water-soluble extractives compared to the rest of the raw materials including the reference material, spruce wood. That is 0.28% for the cold-water and 1.1% for the hot-water extractions as opposed to 1.8% and 3.2% for the cold-water and hot-water extractions respectively, for the cacao tree prunings. The hot-water extractives content of the bean crop residues of the rainy season was seen to be 17.5% compared to 14.25% for the dry season bean crop residues. The difference in the cold-water extractives content of the bean crop residues of both seasons was insignificant. However, the extractives content of the bean crop residues was seen to be much higher compared to the wood materials. Annual plants, in general, are known to have higher amounts of extractives as compared to woody plants (Martunis, 2008; Nikvash, 2013). The relatively high extractives content of the chips of the bean crop residues may inhibit the bonding process, which would be detrimental to the physical and technological properties of the particleboards.

Table 13: The water-soluble extractives contents of the raw materials compared with the reference material, spruce wood. Bean crop residues (BCR).

Raw materials	**Extractives content**	
	Cold water [%]	**Hot water[%]**
Umbrella tree wood	0.28	1.1
Cacao tree prunings	1.8	3.2
BCR (rainy season)	14.7	17.5
BCR (dry season)	14,58	14.25
Spruce wood	1.1	3.5

5.1.4 Solvent-soluble extractives content of the raw materials

Beside the water-soluble extractives, wood also contains the water-insoluble extractives such as waxes, resins, fats, etc, which can only be extracted by solvents. The solvent-soluble extractives content of the raw materials as determined by the ethanol-cyclohexane method (see chapter 4.2.5) is presented in Table 14. The results indicated that the umbrella tree wood contained the highest amount of ethanol-cyclohexane extractives (3.6%), followed by the bean crop residues of the dry season (3.1%), the bean crop residues of the rainy season (3.0%) and then spruce wood (2.7%). The ethanol-cyclohexane extractives content of cacao tree prunings (1.4%) was the lowest amongst the study raw materials. The solvent extractives content of the bean crop residues of both seasons did not differsignificantly.

Table 14: The solvent-soluble extractives contents of the raw materials compared with the reference material, spruce wood. Bean crop residues (BCR).

Raw materials	Extractive contents [%]
Umbrella tree wood	3.6
Cacao tree prunings	1.4
BCR (rainy season)	3.0
BCR (dry season)	3.1
Spruce wood	2.7

5.1.5 Pentosan content of the raw materials

The determination of the pentosan content of wood makes it possible to estimate the hemicellulose content as it is the major component of hemicellulose. According to Kurschner (1966), pentosan accounts for about 50% of softwoods' hemicellulose.

Based on the analysis, the chips of the cacao tree prunings were found to contain the highest amount of pentosan (16.7%), followed by the umbrella tree wood with a value of 13.0%. Amongst the bean crop residues, those of the dry season had a higher amount of pentosan (10.2%) as opposed to the residues of the rainy season with a value of 8.2%. The pentosan content of spruce wood (8.5%) as determined in this study was found to be lower than the value of 11% reported by Zeitsch (2000). Pentosans are more abundant in hardwoods than softwoods (TAPPI, 2001; Antczak *et al*., 2013); hence, the higher pentosan contents of the umbrella tree wood and the cacao tree prunings compared to the spruce wood.

Table 15: The pentosan contents of the raw materials compared with the reference material, spruce wood. Bean crop residues (BCR).

Raw materials	Pentosan content [%]
Umbrella tree wood	13.0
Cacao tree prunings	16.7
BCR (rainy season)	8.2
BCR (dry season)	10.2
Spruce wood	8.5

5.1.6 The klason lignin content of the raw materials

Lignin is the natural glue that binds together the other structural elements of plants. Klason lignin measures the amount of the acid-insoluble lignin contained in wood and pulp. The Klason lignin contents of the research raw materials and spruce wood as determined by the TAPPI Standard T 222 (see chapter 4.2.7) are presented in Table 16. Based on the analysis, the cacao tree prunings had the highest lignin content (39.2%) compared to umbrella tree wood (37.0%) and spruce wood (29.4%). The 29.4% lignin for spruce wood is somewhat close to the 30.15% reported by Markessini *et al*. (1997). The lignin content of the cacao tree prunings and umbrella tree wood are higher than the range of 18 to 30 percent expected for wood materials as reported in Roffael (2004). Also, the lignin content of the bean crop residues of the rainy season (19.6%) was higher than that of the bean crop residues of the dry season (16.8%). It can also be seen that the lignin contents of the beans crop residues of both seasons are substantially lower than the lignin contentsof the wood materials. This is in line with the findings of Prasad *et al*., 2007 and Markessini *et al*., 1997, who showed that the lignin contents of annual plants are generally lower compared to the lignin contents of wood materials.

Table 16: The Klason lignin contents of the raw materials compared with the reference material, spruce wood. Bean crop residues (BCR).

Raw materials	Klason lignin content [%]
Umbrella tree wood	37.0
Cacao tree prunings	39.2
BCR (rainy season)	19.6
BCR (dry season)	16.8
Spruce wood	29,4

5.1.7 The ash content of the raw materials

Ash is the inorganic mineral component of wood and biomass. The ash content of wood usually does not exceed 1% of the wood's dry mass, but may vary from species to species and also depend on the growth condition of the tree. The ash contents of the research raw materials including the reference material, spruce wood, as determined according to the DIN EN 14775 standards (see chapter 4.2.8) are presented in Table 17. The analysis showed that the bean crop residues of the dry season and rainy season had substantially higher ash contents in the values of 8.9% and 8.5% respectively compared to the woody materials. Annual plants are known to generally contain higher ash contents compared to wood (Markessini *et al.*, 1997). There was no marked difference in the ash contents of umbrella tree wood (1.85%) and cacao tree prunings (2.1%). Spruce wood had the least ash content value (0.36%).

Table 17: The ash contents of the raw materials compared with the reference material, spruce wood. Bean crop residues (BCR).

Raw materials	Ash content [%]
Umbrella tree wood	1.85
Cacao tree prunings	2.1
BCR (rainy season)	8.5
BCR (dry season)	8.9
Spruce wood	0.36

5.1.8 Summary of the physical and chemical characteristics of the study raw materials

The results of the investigations into the physical and chemical properties of the study raw materials indicate that

- The bulk density of surface and the core layers chips of the bean crop residues of both the dry season and the rainy seasons are well below the bulk densities of the wood materials. Amongst the wood materials, the chips of umbrella tree wood chips had lower bulk densities, which are 127 kg/m3 and 92 kg/m3 for the surface layer and the core layers chips respectively compared to the rest of the materials. Based on the relatively low density of umbrella tree wood, the production of lightweight particleboards may be feasible.

- The fractional composition of the laboratory-produced chips of the umbrella tree wood, cacao tree prunings, and spruce wood, as well as the chips of the bean crop residues, differed profoundly from the industrially produced wood chips of both the surface and the core layers. Slight variations in fractional composition were also observed between the chips of; umbrella tree wood, cacao tree prunings, and spruce wood.
- Despite the comparatively higher pH-values of umbrella tree wood (6.2) and cacao tree prunings (6.8), they exhibited lower buffering capacities of values 0.54 mmol NaOH/100 g and 0.57 mmol NaOH/100 g respectively. The bean crop residues of the rainy season and the dry season had pH-values of 5.6 and 6.4 respectively but showed substantially higher values of buffering capacities compared to the wood materials. This means that the chips of the bean crop residues might be problematic in bonding with pH-sensitive binder systems.
- The umbrella tree wood contained the least amount of water-soluble extractives, 0.28% for cold water and 1.1% for hot water, but contained the highest amount of ethanol-cyclohexane extractives (3.6%) compared to the rest of the wood materials. The water-soluble extractives content of the bean crop residues ranged from 14.58% to 14.7% for cold water and 14.25% and 17.5% for hot water.

- The umbrella tree wood and cacao tree prunings showed higher values of pentosan content, 13%, and 16.7% respectively compared to the rest of the materials. The pentosan content of the bean crop residues of the rainy season (8.2%) was not any different from that of spruce wood (8.5%).
- The klason lignin contents of cacao tree prunings and umbrella wood, 39.2%, and 37% respectively, were observed to be substantially higher compared to the values of the bean crop residues and the reference material, spruce wood (29.4 %).
- As expected, the values of the ash contents of the bean crop residues of both seasons were seen to be substantially higher in comparison to the values of the wood materials, 8.5% for the bean crop residues of the rainy season and 8.9% for the bean crop residues of the dry season. Amongst the wood materials, the cacao tree prunings and umbrella tree wood had higher ash contents in the values of 2.1% and 1.85% respectively compared to spruce wood (0.36%).

5.2 The properties of the UF-resin bonded particleboards based on cacao tree prunings and umbrella tree wood as raw material

5.2.1 The physical and mechanical properties

The following chapter presents and discusses the results of the physical and mechanical properties of the investigation pertaining to the development of particleboards based on the raw materials, umbrella tree wood and cacao tree prunings in comparison to the particleboards of spruce wood. Within the context of this work, the particleboards were manufactured in two series. In the first series, the panels were manufactured with the chips as they were originally screened (unaltered chip sizes). In order to investigate the influence of coarser core layer chips on the properties of the particleboards of umbrella tree wood and cacao tree prunings, another series of particleboards with coarser core layer chips was produced and tested for their physical-technological properties (see chapter 4.7.1). The results obtained were evaluated and compared against the standard for particleboards used for interior applications under dry conditions (DIN EN 312:2010). This is to allow for a qualitative assessment of the boards of the research raw materials in comparison with panels that are already established on the market.

5.2.1.1 The internal bond strength

The internal bond (IB) strengths as a function of the mechanical strength of the boards represented by the test samples are shown in figure 35. As seen from the figure, particleboards made out of cacao tree prunings had significantly higher IB strength ($p<0.001$) in comparison to the umbrella tree wood for the PB series with unaltered chips sizes as well as for the board series with coarse core layer chips. A similar trend was observed for the boards of umbrella tree wood of all the densities compared to the boards of spruce wood. The IB strength of the PB of each of the raw materials reduced with decreasing board density.

Comparing between the particleboards series with the coarser core layer chips and the series with unaltered chips sizes, it was found that by increasing the coarseness of the core layer chips of cacao tree prunings resulted in a significant decrease ($p<0.05$) in the IB strength of the boards of the densities 650 kg/m^3 and 550 kg/m^3. A percent decrease in IB strength of 16.9% and 22% for the densities 650 kg/m^3 and 550 kg/m^3 respectively was observed. However, but no significant difference ($p=0.9996$) in IB strength was observed between both panel series of cacao tree prunings for a density of 450 kg/m^3.

Similarly, for the umbrella tree wood particleboards, the board series with coarser core layer wood chips had significantly lower values of IB strength ($p<0.0002$) for each of the densities compared to the particleboard series with unaltered chip sizes. That means by increasing the coarseness of the core layer chips of umbrella tree wood resulted in a significant decrease in IB strengths. By increasing the coarseness of the core layer chips of umbrella tree wood, a percent decrease in internal bond strengths of 41%, 23%, and 30% was observed for the densities 650 kg/m^3, 550 kg/m^3, and 450 kg/m^3 respectively. The decrease in internal bond strength with an increase in coarseness of the middle layer chips as observed in this study is in line with the findings Lias *et al*., (2014).

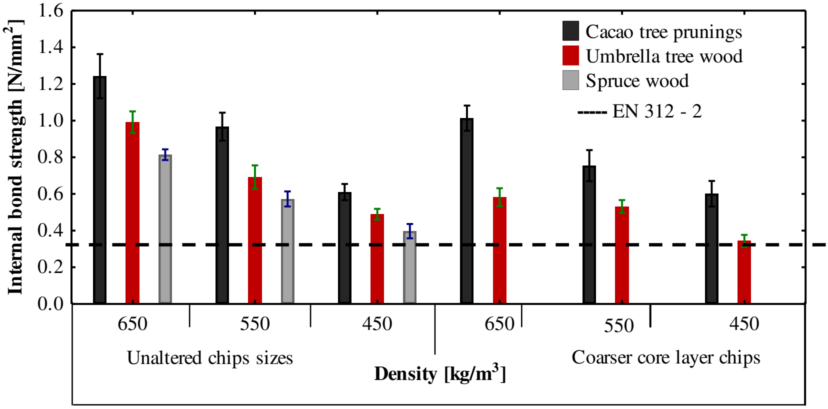


Figure 35: The internal bond strengths of the UF-resin bonded particleboards, based on umbrella tree wood and cacao tree pruning, in comparison with the boards of spruce wood. Comparison of the IB strength between the panel series with unaltered chip sizes and the series with coarser core layer chips of umbrella tree wood and cacao tree prunings (Board thickness 20 mm).

According to DIN EN 312-2 (2010), 0.35 N/mm^2 is the minimum IB strength required for particleboards of thickness 13 mm to 20 mm. As seen in figure 35, all the particleboards achieved the IB strength requirement for this standard except for the 450 kg/m^3 boards of umbrella tree wood of the panel series with coarser core layer chips.

5.2.1.2 The Modulus of Rupture

The modulus of rupture (MOR) is an important property for the application area of particleboards, and it has been known to be influenced by the amount and the type of adhesive used in gluing the chips, the density of the panels and the composition of the raw material. Figure 36 presents the MOR as a function of the density, 650 kg/m^3, 550 kg/m^3, and 450 kg/m^3 of the various raw materials, as well as a comparison between the board series with unaltered chip sizes and the boards series with coarser core layer chips of umbrella tree wood and cacao tree prunings. For the panels with unaltered chip sizes, it can be seen that the spruce wood boards achieved significantly higher MOR ($p<0.0001$) in each of the densities compared to the boards of umbrella tree wood and the boards of cacao tree prunings. Also, the cacao tree prunings boards had comparatively higher MOR (11.53 N/mm^2) than the umbrella tree wood boards (10.27 N/mm^2) for the density of 650 kg/m^3, whereas no significant difference between the two was observed for the panels of the densities 550 kg/m^3 and 450 kg/m^3.

For the panels with coarser core layer chips, the particleboards of cacao tree prunings achieved significantly higher MOR ($p<0.00061$) in each of the densities compared to the boards of umbrella tree wood.

It is also noticeable that the panels of both the cacao tree prunings and umbrella tree wood of the panel series with coarser core layer chips had significantly higher MOR ($p<0.001$) in each of the densities compared to the boards of the panel series with unaltered chip sizes. At the density of 650 kg/m^3, the boards of umbrella tree wood and cacao tree prunings for the panel series with coarser core layer chips had 27% and 50% higher MOR values respectively compare to the panels series with unaltered chip sizes. A percent increase of 9% and 25% for the boards of umbrella tree wood and cacao tree prunings respectively for the density 550 kg/m^3 was also observed for the panel series with coarser core layer chips relative to the panel series with unaltered chip sizes.

The increases in MOR could be explained by the fact that the coarser wood chips possesses higher stiffness compared to the chips of lesser coarseness. The improvement in the MOR characteristic observed in this study confirms the findings in the literature by Lias *et al*., (2014), Istek *et al*., 2018; Youngquis J.A, 1999 and Frybort *et al*., 2008, who reported that by increasing the sizes of the wood chips used in producing PB significantly increase the MOR. Nevertheless, The PB of cacao tree prunings of both panel series at the density of 650 kg/m^3 had high enough MOR to fulfill the minimum requirement of DIN EN 312-2 (11 N/mm^2 for board thickness of 13 mm to 20 mm). The boards of cacao tree prunings of the panel series with coarser core layer chips of the 550 kg/m^3 also fulfilled the MOR requirement of the EN 312-2 (2010).

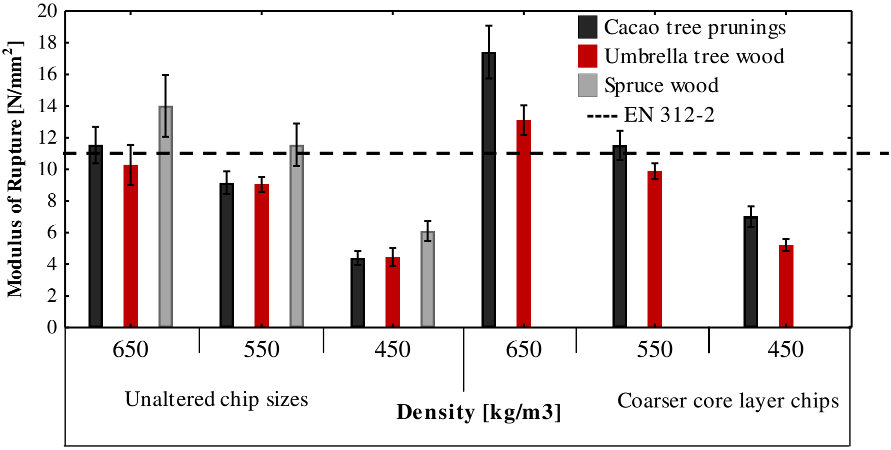


Figure 36: The modulus of rupture of the UF-resin bonded particleboards, based on umbrella tree wood and cacao tree pruning, in comparison with the boards of spruce wood. Comparison of the MOR between the panelsseries with unaltered chip sizes and the series with coarser core layer chips of umbrella tree wood and cacao tree prunings (Board thickness 20 mm).

5.2.1.3 The Modulus of Elasticity

Figure 37 presents the modulus of elasticity (MOE) of the manufactured particleboards of the various raw materials for the densities 650 kg/m^3, 550 kg/m^3 and 450 kg/m^3. It also presents a comparison between MOE of the panels' series with unaltered chips sizes and the series with coarser core layer chips of both the umbrella tree wood and cacao tree prunings. The results showed that, for the panels with unaltered chip sizes, the MOE ofthe boards of spruce wood in each of the densities were at least 37%

higher compared to the MOE of the boards of umbrella tree wood and the boards of cacao tree prunings. Unlike the modulus of rupture, the MOE of the PB of umbrella tree wood was significantly higher ($p<0.05$) as compared to the boards of cacao tree prunings for both the panel series. This indicates that the test samples of the boards of umbrella tree wood had a lesser degree of deflection during breakage as compared to test samples of the boards of cacao tree prunings, as MOE (Young's modulus) measures the resistance of a material to undergo elastic deformation under a load. This means that the boards of the umbrella tree wood had higher stiffness compared to the the boards of of cacao tree prunings, which could be accounted for by the low bulk density of the umbrella tree wood which is give it a greater degree of compaction compared to the cacao tree prunings.

It can further be seen from Figure 37 that in each of the densities, the MOE of the boards of both cacao tree prunings and the umbrella tree wood of the panel series with coarser core layer chips is significantly higher ($p<0.00051$) compared to the MOE of the panel series with unaltered chip sizes.

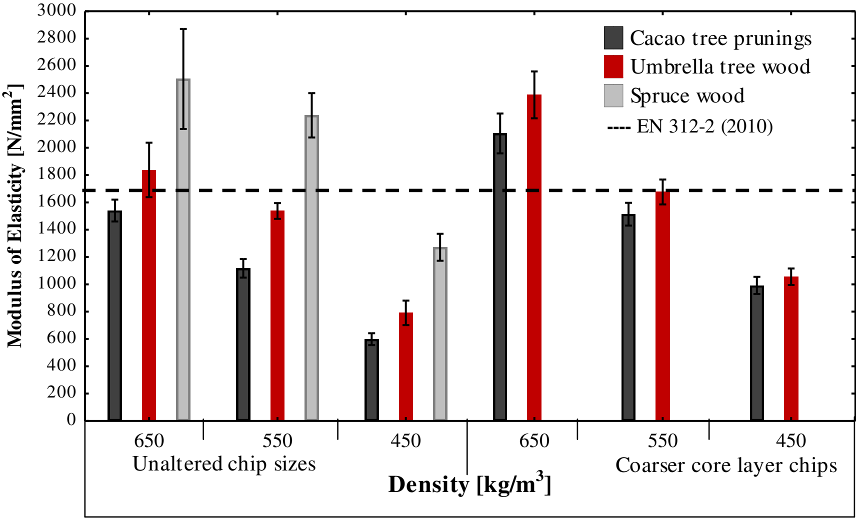


Figure 37: The modulus of elasticity of the UF-resin bonded particleboards, based on umbrella tree wood and cacao tree pruning, in comparison with the boards of spruce wood. Comparison of the MOE between the panel series with unaltered chip sizes and the series with coarser core layer chips of umbrella tree wood and cacao tree prunings (Board thickness 20 mm).

5.2.1.4 The thickness swelling and water absorption

The 24 hours thickness swelling (TS) and water absorption (WA) properties of the produced particleboards are present in figure 38 and figure 39 respectively. Significant differences were observed between the boards of umbrella tree wood and cacao tree prunings in each of the densities, 650 kg/m^3, 550 kg/m^3, and 450 kg/m^3, for the panel series with unaltered chips sizes as well as the series with coarser core layer chips.

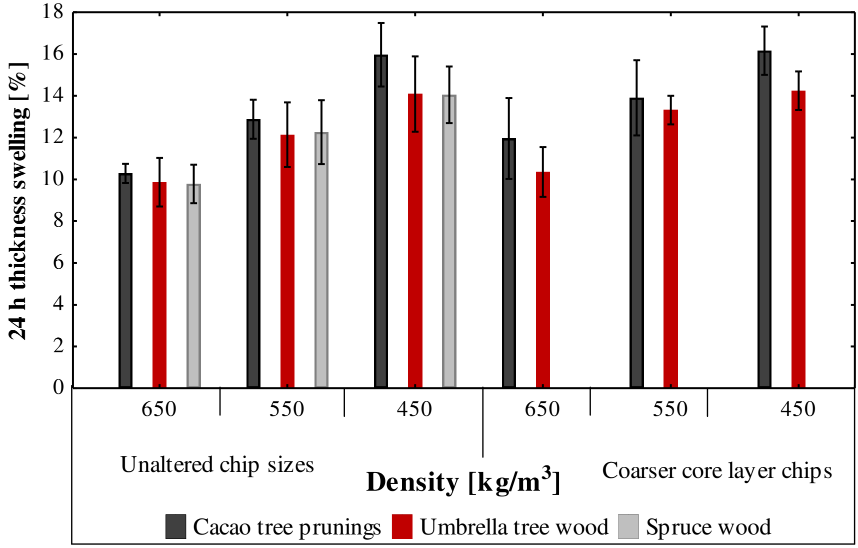


Figure 38: The thickness swelling (after 24 hours) of the UF-resin bonded particleboards, based on umbrella tree wood and cacao tree pruning, in comparison with the boards of spruce wood. Comparison of the TS between the panel series with unaltered chip sizes and the series with coarser core layer chips of umbrella tree wood and cacao tree prunings (Board thickness 20 mm).

For the production series with unaltered chip sizes, the highest swell in thickness (15.9 %) corresponding to the highest water absorption (85.7%) was consistently observed for the boards of cacao tree prunings at the density 450 kg/m^3. At the density 550 kg/m^3, TS of 12.8% and WA of 56.9% was also observed for the boards of cacao tree prunings of the same panel series. In addition, the thickness swelling of the boards of umbrella tree wood did not differ significantly from the TS of the boards of thereference material, spruce wood. However, the water absorption values of the boards of spruce wood in each of the density 650 kg/m^3 (25.7%), 550 kg/m^3 (47.5%), and 450 kg/m^3 (74.2%) were observed to be significantly higher (p=0.00019, p=0.00316, and p=0.00012 respectively) than the boards of umbrella tree wood.

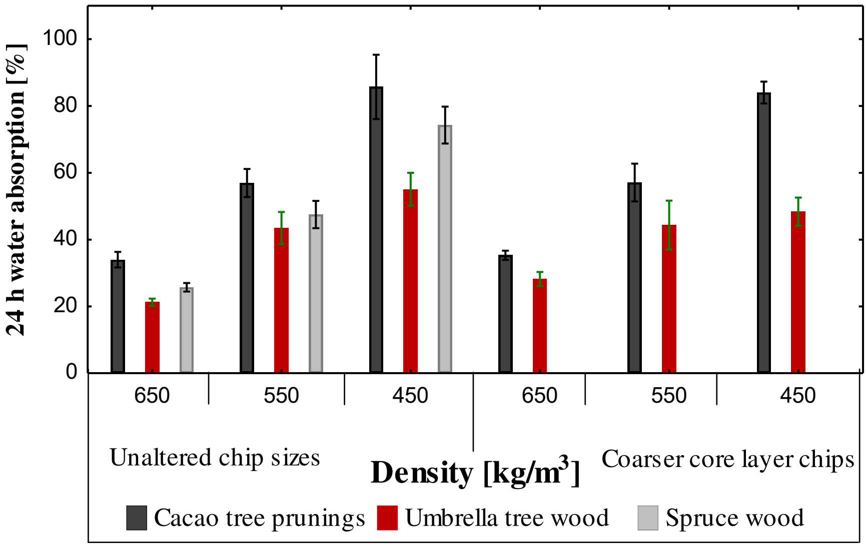


Figure 39: The water absorption (after 24 hours) of the UF-resin bonded particleboards, based on umbrella tree wood and cacao tree pruning, in comparison with the boards of spruce wood. Comparison of the WA between the panel series with unaltered chip sizes and the series with coarser core layer chips of umbrella tree wood and cacao tree prunings (Board thickness 20 mm).

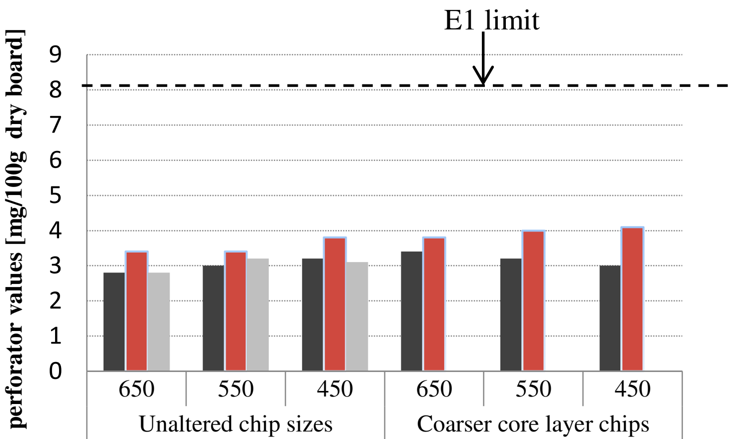
Generally, the panel series with the coarser core layer chips showed less dimensional stability in terms of thickness swelling compared to the panel series with unaltered chip sizes for the materials umbrella tree wood and cacao tree prunings. However, for the density, 450 kg/m^3, no significant differences in TS were observed between the boards of both panel series. At the density 650 kg/m^3, the water absorption of the boards of the umbrella tree wood of the panel series with coarser core layer chips was observed to be slightly higher (p=0.00021) compared to those of the panel series of unaltered chip sizes. The boards of cacao tree prunings of the panel series with coarser core layer chips did not differ significantly (p=0.058) from the same boards of the panel series with unaltered chip sizes. Nevertheless, the boards of both panel series of each of the raw materials did not differ significantly in terms of water absorption, for the densities 550 kg/m^3, and 450 kg/m^3. However, the DIN EN 312 (2010) has no requirement for TS and WA of particleboards that are used for interior application under dry conditions.

5.2.2 The formaldehyde releaseof the UF-resin bonded particleboards based on cacao tree prunings and umbrella tree wood as raw materials

The formaldehyde properties of the manufactured particleboards measured according to the perforator method (DIN EN 120: 1993a) and the flask method (DIN EN 717-3) are presented in the flowing chapter. Due to the small number of repetitions (4 repetitions per variant), the results of formaldehyde properties were not evaluated statistically.

5.2.2.1 Perforator values

The perforator method (EN 120) measures the amount of extractable formaldehyde of wood-based composites. Figure 40 presents the perforat- or values of the UF-resin (K 340) bonded boards, of the panel series with coarser core layer chips and the series with unaltered chip sizes, for various raw materials in each density. For the panel series with unaltered chips sizes, it can be seen that the perforator values of the boards of umbrella tree wood in each of the densities, 650 kg/m^3, 550 kg/m^3, and 450 kg/m^3 were higher compared to the values of boards of cacao tree prunings. The same trend was observed for the panel series with coarser core layer chips, with the highest perforator value of 4.1 mg/100 g recorded for the density of 450 kg/m^3. Also, no actual differentiable differences were observed in the perforator values of the boards of cacao tree prunings and the boards of spruce wood. According to the European emission class E1 standard, the formaldehyde emission of panels for indoors applications must not exceed 0.1ppm (0.124 mg/m^3) when tested according to the DIN EN 717-1. This maximum limit correlates with the perforator value, 8.0 mg/100 g oven-dry board. It can be seen from the results that the maximum formaldehyde release of the manufactured boards was merely half the maximum allowable limit of 8.0 mg/100 g.

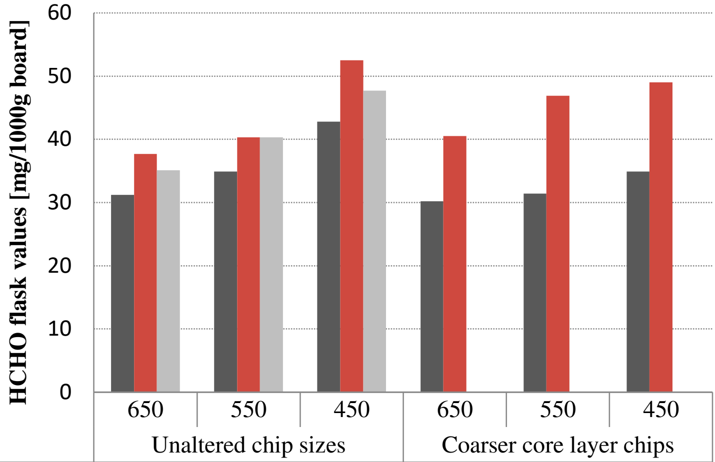


	650	550	450	650	550	450
Cacao tree prunings	2.8	3	3.2	3.4	3.2	3
Umbrella tree wood	3.4	3.4	3.8	3.8	4	4.1
Spruce wood	2.8	3.2	3.1			

Figure 40: The perforator values (at 6.5% MC) of the UF-resin bonded particleboards, based on umbrella tree wood and cacao tree pruning, in comparison with the boards of spruce wood. Comparison of the perforator values between the panels' series with unaltered chip sizes and the series with coarser core layer chips of umbrella tree wood and cacao tree prunings (Board thickness 20 mm).

5.2.2.2 Flask values

Another method of determining the formaldehyde release of wood-based panels is by measuring the formaldehyde content of the aqueous solution contained in a flask containing the test samples at 40 °C. Figure 41 shows the results of the flask values (after 24 hours) for the boards of the various materials for the respective densities and panel series.



	Unaltered chip sizes 650	550	450	Coarser core layer chips 650	550	450
Cacao tree prunings	31.2	34.9	42.8	30.2	31.4	34.9
Umbrella tree wood	37.7	40.3	52.5	40.5	46.9	49
Spruce wood	35.1	40.3	47.7			

Figure 41: The flask values of the UF-resin bonded particleboards, based on umbrella tree wood and cacao tree pruning, in comparison with the boards of spruce wood. Comparison of the flask values between the panels' series with unaltered chip sizes and the series with coarser core layer chips of umbrella tree wood and cacao tree prunings (Board thickness 20 mm).

The flask values of the PB of umbrella tree wood in each of the densities, 650 kg/m^3, 550 kg/m^3, and 450 kg/m^3, for both panel series, were observed to be higher compared to the flask values of the boards of cacao tree prunings. Unlike the perforator values, the flask values of the boards of the reference material, spruce wood, were slightly higher than the flask values of the boards of cacao tree pruning. This may be because the perforator method is more sensitive as compared to the flask method and because the flask method accounts mostly for the formaldehyde from certain areas (surfaces and edges) of the boards. The highest flask value, 52.5 mg/1000g, was recorded for the boards of umbrella tree wood of the density 450 kg/m^3 of the panel series with unaltered chip sizes. It is also noticeable that the flask values for the panel series with the coarser core layer chips were lower compared to the values of the panel series with unaltered chip sizes in each of the densities, except for the boards of umbrella tree wood of the densities 650 kg/m^3 and 550 kg/m^3.

5.2.3 Summary of the results of the UF-resin bonded particleboards based on cacao tree prunings and umbrella tree wood as raw materials

The study presented in this chapter evaluated the technical feasibility of producing UF-resin bonded particleboards of reduced formaldehyde emissions, from the raw materials, umbrella tree wood, and cacao tree prunings, with physical and mechanical properties that are comparable to the products that are already established in the market. It also investigated the effect of the fractional composition of core layer chips on the properties of the particleboards. The results of the properties of the manufactured particleboards revealed that:

- The boards of cacao tree prunings of both panels series for all the densities, 650 kg/m^3, 550 kg/m^3, and 450 kg/m^3, had higher enough internal bond strength to meet the minimum IB requirement of the DIN EN 312-2(2010). The same was true for the boards of umbrella tree wood except for the density of 450 kg/m^3 of the panel series with coarser core layer chips.
- The MOR and the IB strength of the boards of cacao tree prunings were generally higher compared to the boards of umbrella tree wood. Meanwhile, the PB of umbrella tree wood had higher MOE compared to the boards of cacao tree prunings.
- Although the internal bond strength of the panels of both raw materials was decreased by increasing the coarseness of the core layer chips, the MOR and the MOE, on the other hand, were significantly increased by increasing the coarseness of the core layer chips. The PB of cacao tree prunings for the densities 650 kg/m^3 and 550 kg/m^3 had high enough MOR to satisfy the minimum MOR requirement for DIN EN 312-2 (2010). The MOE of the panels of umbrella tree wood for the same densities exceeded the minimum MOE value stipulated by the DIN EN 312-2 (2010).
- Higher IB strength values were observed for the PB of umbrella tree wood and cacao tree prunings compared to the boards of

spruce wood in each of the board densities. However, the PB of spruce wood performed better in terms of MOR and MOE in comparison with the board of umbrella tree wood and the boards of cacao tree prunings.

- The boards of cacao tree prunings, in each of the densities, showed higher values of TS and WA compared to the boards of umbrella tree wood and the boards of spruce wood.
- As for the formaldehyde release of the manufactured panels, the boards of umbrella tree wood showed higher perforator and flasks values, in each of the panel densities, compared to the panels of cacao tree prunings and the boards of spruce wood. The Perforator values of all the boards were seen to be at least 2 times below the maximum allowable limit of 8.0 mg/100g.
- The results of this study suggest that at the densities of 650 kg/m^3 and 550 kg/m^3, it is possible to produce particleboards of reduced formaldehyde emissions from the raw materials, umbrella tree wood, and cacao tree prunings, when bonded with UF-resin 340. However, caution should be taken to ensure that the coarseness of the core layer chips is appropriate for achieving high enough MOR and MOE.

5.3 TheProperties of the non-formaldehyde bonded particleboards based on cacao tree prunings and umbrella tree wood as raw materials

The following chapter presents and explains in details the results of the investigation into the feasibility of producing non-formaldehyde bonded particleboards, based on the raw materials; umbrella tree wood and cacao tree prunings. For the qualitative assessment of the manufactured particleboards to be meaningful, the results are compared with the standard of DIN EN 312-2 (2010). These are particleboards that are used for interior applications (such as furniture and interior fittings) under dry conditions.

5.3.1 Physical and mechanical properties

The particleboards which results are discussed in this chapter were produced in two series. In one production series, the formaldehyde-free binder, PMDI, and in another series, blood albumin (animal protein), were used to glue the wood chips. The produced chipboards were evaluated for their physical and technological properties and the results are presented in the following chapters.

5.3.1.1 Internal bond strength

Figure 42 presents graphically the results of the internal bond (IB) strengths of the manufactured panels of each of the densities 650 kg/m^3 and 550 kg/m^3. The values of the IB strength of the PB of cacao tree prunings of both binder systems; blood albumin, and PMDI, were seen to be significantly higher ($p<0.0001$) in each of the densities compared to those of umbrella tree wood and the hybrid boards of both raw materials (umbrella tree wood and cacao tree prunings mixed in the ratio 50:50 percent w/w). This result is consistent with the results of the UF-resin bonded particleboard discussed in section 5.2.1.1, where the PB of cacao tree prunings attained the highest IB strengths compared to their umbrella tree wood counterparts. Within the density of 550 kg/m^3, the PB of the mixture of umbrella tree wood and cacao tree prunings (UTW & CTP hybrid) showed significantly higher IB strength values compared to the panels of umbrella tree wood for both binder systems, PMDI ($p=0.00004$) and blood albumin ($p=0.04007$).

In contrast to the density 550 kg/m^3, the hybrid boards and the boards of umbrella tree wood bonded with PMDI did not differ significantly from each other with regard to their IB strengths for the density 650 kg/m^3 ($p=0.4795$). The same applies to the series bonded with blood albumin ($p=0.6366$). The IB strengths of the PMDI bonded PB in each of the densities were significantly higher in comparison to the PB bonded with blood albumin. Nevertheless, the IB strength of the produced panels of each density exceeded the DIN EN 312-2(2010) minimum requirement of 0.35 N/mm^2 for particleboards used for interior purposes under dry conditions.

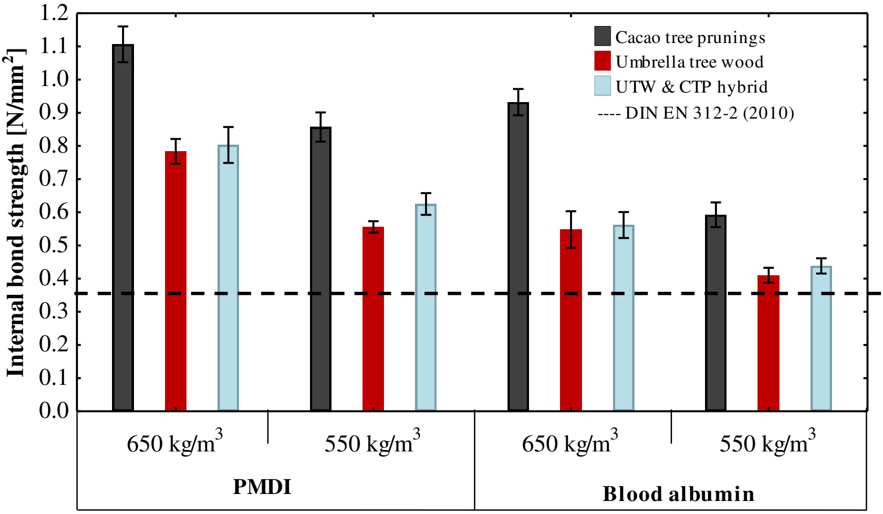


Figure 42: The internal bond strength of the PMDI and blood albumin bonded particleboards, based on the raw materials umbrella tree wood and cacao tree prunings. Particleboards of the 50:50 percent (w/w) mixture of umbrella tree and cacao tree prunings (UTW-CTP hybrid). Horizontal line indicates the minimum value stipulated by DIN EN 312-2(2010).

5.3.1.2 Modulus of rupture

Figure 43 shows the results of the modulus of rupture (MOR) of the manufactured particleboards. Amongst the PMDI bonded PBs of the density 650 kg/m^3, it can be seen that the MOR of the boards of cacao tree prunings was significantly lower ($p<0.0001$) compared to that of the boards of umbrella tree wood and the boards of the hybrid between umbrella tree wood and cacao tree prunings (UTW-CTP hybrid). A similar trend was also observed for the blood albumin bonded particleboards of the same density 650 kg/m^3. Also, amongst the blood albumin bonded PBs of the density 650 kg/m^3, the MOR of boards manufactured from the mixture of umbrella tree wood and cacao tree prunings (UTW-CTP hybrid) was significantly higher (p=0.0015) compared to the MOR of the boards of cacao tree prunings. Meanwhile, the MOR of boards of umbrella tree wood did not differ significantly (p=0.0950) from the boards of cacao tree prunings. In addition, for the density 650 kg/m^3, the MOR of the boards of umbrella tree wood did not differ significantly from the MOR of the hybrid particleboards (UTW-CTP hybrid) of both binder systems, PMDI (p=0.9617) and blood albumin (p=0.2785).

In contrast to the density 650 kg/m^3, the MOR of the panels of the density 550 kg/m^3 did not differ significantly (p>0.05) from one another for both binder systems. No significant differences in terms of MOR exist between the PMDI and blood albumin bonded boards of each of the raw materials in each of the densities, except for the boards of umbrella tree wood at the density 650 kg/m^3 (p=0.02875). This indicates that for the various raw materials used in this study, the PMDI bonded boards (with 3% resin load) were not superior in terms of their modulus of rupture than the boards bonded with the natural adhesive, blood albumin at resination levels of 8,5% and 10% on the middle layer and surface layers respectively. The hybrid boards (UTW-CTP hybrid) of both binder systems of the density 650 kg/m^3 attained MOR values which are greater than the minimum value of 11N/mm^2 stipulated by the DIN EN 312-2 (2010).

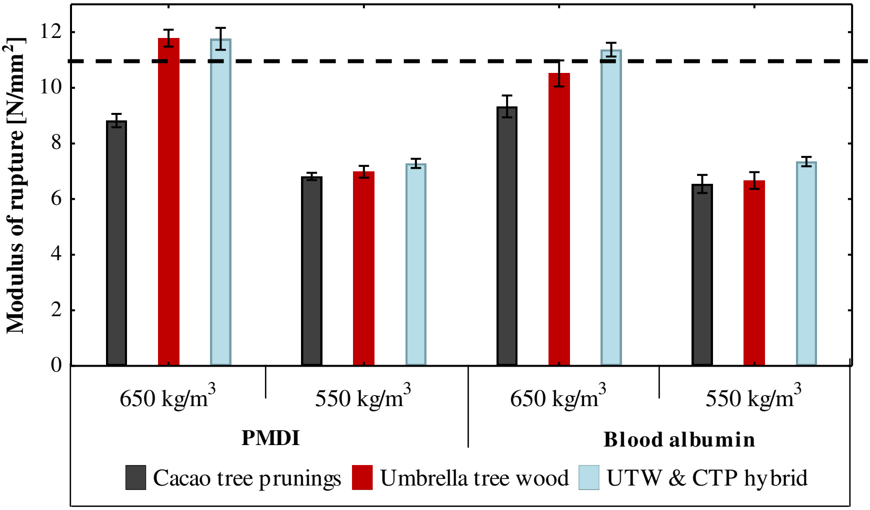


Figure 43: Modulus of Rupture of the PMDI and blood albumin bonded particleboards, based on the raw materials umbrella tree wood and cacao tree prunings. Particleboards of the 50:50 percent (w/w) mixture of umbrella tree and cacao tree prunings (UTW-CTP hybrid). Horrizontal line indicates the minimum value stipulated by DIN EN 312-2(2010).

The particleboards of umbrella tree wood of the density 650 kg/m^3, bonded with PMDI also achieved high enough MOR. The results suggest that a substitution of the chips cacao tree wood with the chips of umbrella tree wood could significantly improve the modulus of rupture of the particleboards made predominantly from cacao tree prunings.

5.3.1.3 Modulus of Elasticity

Figure 44 presents the results of the modulus of elasticity of the produced particleboards. As it was prviously seen in the results of the Modulus of rupture, the modulus of elasticity of the particleboards of cacao tree wood bonded with PMDI as well as the particleboards bonded with blood albumin (for the density 650 kg/m^3), were seen to be significantly lower ($p<0.0001$) compared to the modulus of elasticity of the boards of umbrella tree wood and the hybrid boards. The same trend was observed for the PMDI bonded boards of the density and 550 kg/m^3.

The MOE of the blood albumin bonded particleboards of cacao tree prunings of the density 550 kg/m^3 did not differ significantly ($p=0.1739$) from that of the hybrid boards. In addition, the MOE of the boards of umbrella tree wood of each of the binder systems did not differ significantly from the MOE of the hybrid boards (UTW-CTP hybrid). For the density 650 kg/m^3, the MOE of the hybrid boards and the boards of umbrella tree wood bonded with PMDI were seen to be significantly higher compared to their blood albumin bonded counterparts. It can be seen from the results that, the MOE of the boards of umbrella tree wood and the hybrid boards (UTW-CTP hybrid) of the density 650 kg/m^3 bonded with PMDI, exceeded the minimum required of 1600 N/mm^2 as stipulated by the DIN EN 312-2(2010).

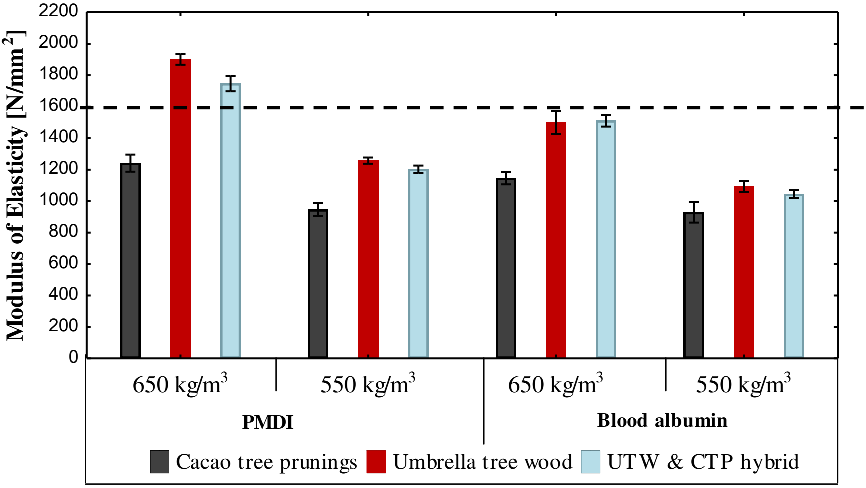


Figure 44: Modulus of Elasticity of the PMDI and blood albumin bonded particleboards, based on the raw materials umbrella tree wood and cacao tree prunings. Particleboards of the 50:50 percent (w/w) mixture of umbrella tree and cacao tree prunings (UTW-CTP hybrid). Horrizontal line indicates the minimum value stipulated by DIN EN 312-2(2010).

5.3.1.4 Thickness swelling and water absorption

The 24 hours thickness swelling (TS) and water absorption (WA) properties of the manufactured particleboards are of the various raw materials are presented in Figure 45 and figure 46 respectively. The water-absorbent nature of cacao tree prunings is evident from the TS and WA values of its particleboards. For the boards of the density 650 kg/m^3, of both binder systems, the TS of the boards of cacao tree prunings was significantly higher ($p<0.0001$) than the TS of the hybrid boards and the boards of umbrella tree wood. Surprisingly, at the density of 550 kg/m^3, the TS of the boards of cacao tree prunings did not differ significantly from the TS of the boards of umbrella tree wood of both binder systems; PMDI ($p=0.9919$) and blood albumin ($p=0.2676$). The 24 hours TS values of the boards of umbrella tree wood, as well as the boards of cacao tree prunings, were observed to be significantly higher ($p<0.0001$) in each of the densities and glue type compared to the TS of the hybrid boards. The results also revealed that the TS values of the boards bonded with PMDI were significantly lower ($p<0.001$) compared to the boards bonded with blood albumin. However, for the density 650 kg/m^3, the TS of the hybrid boards did not differ significantly with respect to the glue type.

Like the TS, the WA values of the boards of cacao tree prunings were seen to be significantly higher ($p<0.0001$) in each densities compared to the boards of umbrella tree wood and the boards of the hybrid of both material.

However, the WA values of the boards of cacao tree prunings of the density 650 kg/m^3, bonded with blood albumin, did not differ significantly ($p=0.7784$) from the WA values of their umbrella tree wood counterparts. In all cases, the 24 hours WA values of the hybrid boards were observed to be significantly lower ($p<0.001$) than the WA values of the boards of umbrella tree wood and the boards of cacao tree prunings.

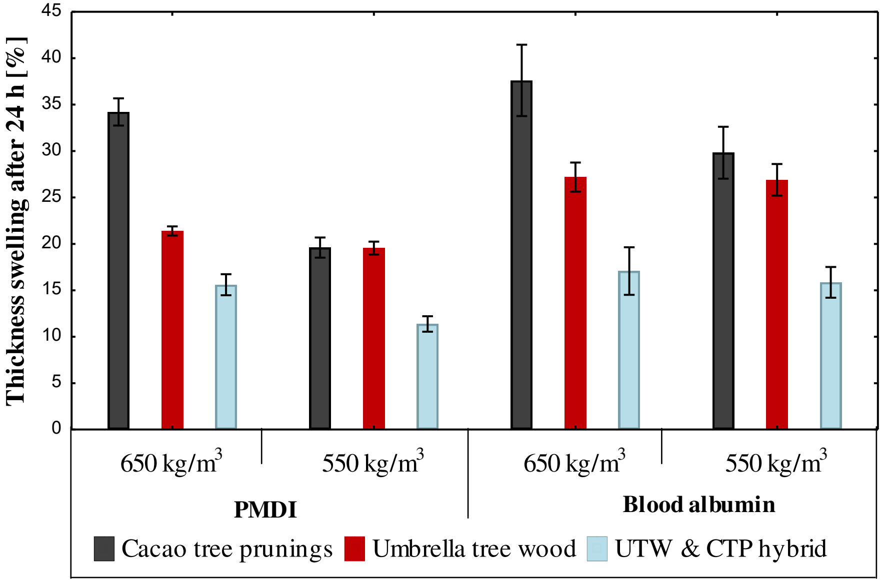


Figure 45: The thickness swelling after 24 hours of the PMDI and blood albumin bonded particleboards, based on the raw materials umbrella tree wood and cacao tree prunings. Particleboards of the 50:50 percent (w/w) mixture of umbrella tree and cacao tree prunings (UTW-CTP hybrid).

This suggests that one of the raw materials might be acting as filler to the other thereby reducing the amounts of voids within the test samples through which water can penetrate and result in higher water absorption. The WA values of the boards bonded with blood albumin were generally higher compared to the boards bonded with PMDI. The results of the TS and WA of the boards of cacao tree prunings relative to the boards of umbrella tree wood in this study are consistent with the findings of the UF-resin bonded particleboards reported in section 5.2.1.4.

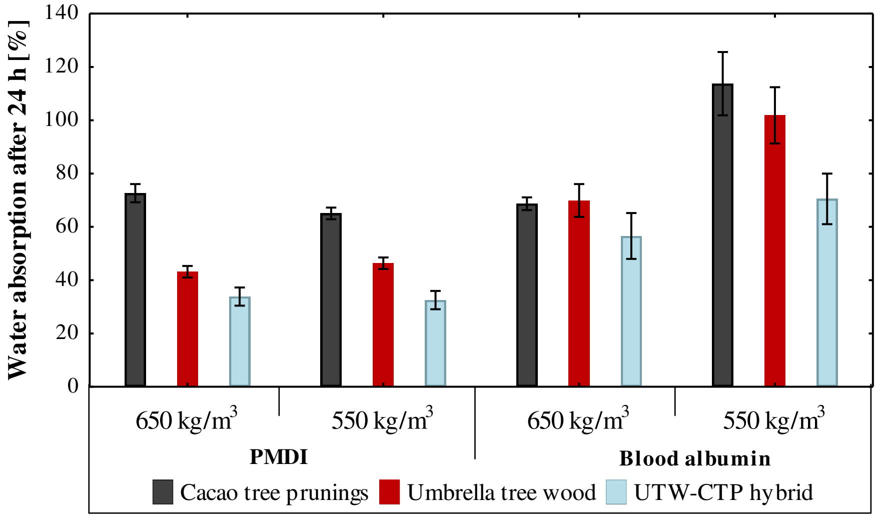


Figure 46: The water absorption after 24 hours of the PMDI and blood albumin bonded particleboards, based on the raw materials umbrella tree wood and cacao tree prunings. Particleboards of the 50:50 percent (w/w) mixture of umbrella tree and cacao tree prunings (UTW-CTP hybrid).

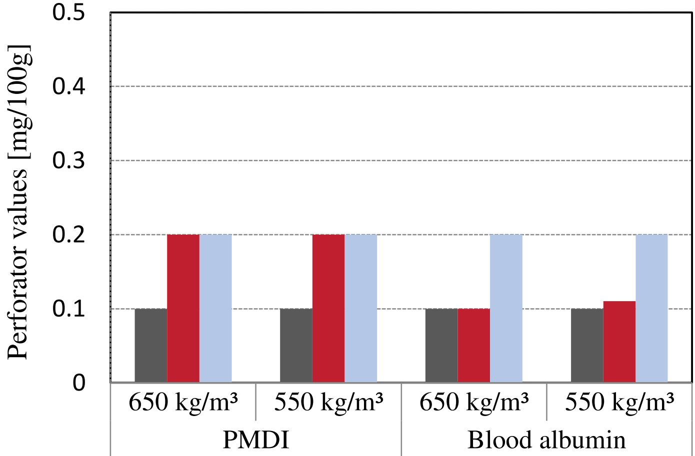
5.3.2 Formaldehyde release of the non-formaldehyde bonded particleboards, based on cacao tree prunings and umbrella tree wood as raw materials

The formaldehyde properties of the particleboards manufactured in this study were examined using the perforator methods (EN 120: 1999a) and the flask method (EN 717-3). The following chapters present the non-statistical results (due to small data set of four repetitions) of the measurements.

5.3.2.1 Perforator values

Figure 47 shows the results of the extractable formaldehyde (Perforator values) of the particleboards of the various raw materials, manufactured on a laboratory scale with the non-formaldehyde containing binder, PMDI and blood albumin. The boards of umbrella tree wood and the boards of the mixture of the wood chips of umbrella tree and cacao tree (UTW-CTP hybrid) bonded with PMDI show the highest perforator values of 0.2 mg/100 g in each of the densities.

The same values (0.2 mg/100 g) were observed for the hybrid particleboards (UTW-CTP hybrid) bonded with blood albumin of both densities. The boards of cacao tree prunings bonded with PMDI, as well as blood albumin, in each of the densities, showed comparatively lower perforator values of 0.1 mg/100 g. It can be said from the results that, umbrella tree wood contain a higher amount of the so-called natural formaldehyde than the wood of cacao tree prunings. It is also noticeable that the perforator values of the boards of umbrella tree wood bonded with blood albumin were half the values of the boards bonded with PMDI. However, the perforator values of the boards manufactured in this study were negligible, about forty times lower than the maximum value permitted for E1 boards (8.0 mg/100 g ~ 0.1 ppm).

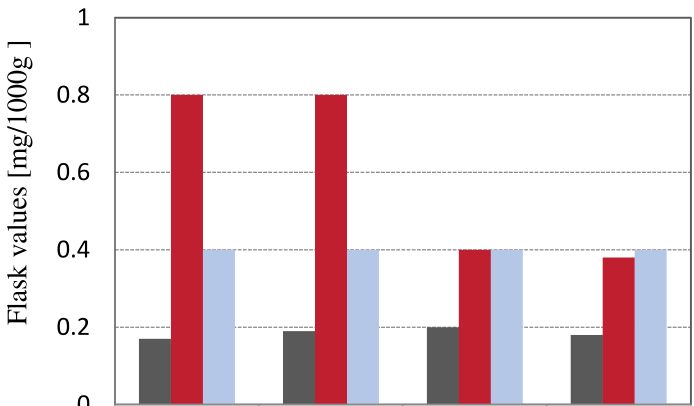


	PMDI 650 kg/m³	PMDI 550 kg/m³	Blood albumin 650 kg/m³	Blood albumin 550 kg/m³
Cacao tree prunings	0.1	0.1	0.1	0.1
Umbrella tree wood	0.2	0.2	0.1	0.11
UTW & CTP hybrid	0.2	0.2	0.2	0.2

Figure 47: The perforator values (at 6.5 % MC) of the PMDI and blood albumin bonded particleboards, based on the raw materials umbrella tree wood and cacao tree prunings. Particleboards of the 50:50 percent (w/w) mixture of umbrella tree and cacao tree prunings (UTW-CTP hybrid).

5.3.2.2 Flask values

Figure 48 presents the flask values of the formaldehyde release of the manufactured particleboards. The flask values of the particleboards of umbrella tree wood, bonded with PMDI (0.8 mg /1000 g), were substantially higher compared to their counterparts of cacao tree prunings as well as those of the hybrid of both materials (UTW-CTP hybrid). The flask values of the hybrid boards and boards of umbrella tree wood bonded with blood albumin, were also observed to be higher than the flask values of their cacao tree prunings counterparts. The hybrid boards of both binder systems had the same flask values of formaldehyde release but the flask values of the boards of umbrella tree wood bonded with PMDI were two times higher than the values of the same boards bonded with blood albumin.



	PMDI		Blood albumin	
	650 kg/m³	550 kg/m³	650 kg/m³	550 kg/m³
Cacao tree prunings	0.17	0.19	0.2	0.18
Umbrella tree wood	0.8	0.8	0.4	0.38
UTW & CTP hybrid	0.4	0.4	0.4	0.4

Figure 48: The flask values of the PMDI and blood albumin bonded particleboards, based on the raw materials umbrella tree wood and cacao tree prunings. Particleboards of the 50:50 percent (w/w) mixture of umbrella tree and cacao tree prunings (UTW-CTP hybrid).

5.3.3 Summary of the results of the non-formaldehyde bonded particleboards based on cacao tree prunings and umbrella tree wood

This chapter investigated the possibility of producing non-formaldehyde bonded particleboards by using the by-product of cacao thinning operations (cacao tree prunings) and the wood of a fast-growing tree species, umbrella tree (*Musanga cecropoides*) as raw materials. The experimental work was based on the use of non-formaldehyde containing binder systems, PMDI, and a newly developed adhesive based on animal protein, blood albumin, for the production of three-layered particleboards of the densities 650 kg/m^3 and 550 kg/m^3. An evaluation of the properties revealed that:

- The particleboards of the umbrella tree wood and cacao tree prunings, as well as the the boards of the mixture of both materials (hybrid boards), of both binder systems of each of the densities, had high enough internal bond strength that surpassed the minimum value (0.35 N/mm^2) required stipulated by DIN EN 312-2 (2010).
- The MOR of the particleboards of umbrella tree wood of the density of 650 kg/m^3, manufactured with PMDI, exceeded the DIN EN 312 requirement for type P2 particleboards. Also, the hybrid particleboards of the density 650 kg/m^3, bonded with PMDI, as well as those bonded with blood albumin, achieved high enough bending strengths to satisfy the requirement of particleboards that are used for interior applications (such as furniture and interior fit-

tings) under dry conditions. A similar trend was also observed for the MOE (for the density 650 kg/m^3), except that the hybrid particleboards bonded with blood albumin did not achieve the minimum MOE value as required by DIN EN 312-2 (2010).

- The thickness swelling and water absorption values of the boards of cacao tree prunings were generally higher compared to theTS and WA of the hybrid boards and the boards of umbrella tree wood. This could be attributed to the water-absorbent nature of agricultural residues as opposed to wood material.
- The formaldehyde release (perforator and flask values) of the produced boards were observed to be negligible compared to the maximum limit of E1. The formaldehyde release recorded from each board can be tied to the so-called natural formaldehyde of the raw materials since the binder systems contained no free formaldehyde.
- The findings indicate that the manufacture of non-formaldehyde containing particleboards of the density 650 kg/m^3 from the raw materials, umbrella wood, and the mixture (50:50 percent by weight) of umbrella tree wood and cacao tree prunings is technically feasible with PMDI. The particleboards can be used for interior applications including furniture.

5.4 The properties of the particleboards based on a hybrid between the chips of bean shells and various wood materials

The following chapter presents the results of the experimental work aimed to assess the performance of the annual crop residue, bean shells (bean crop residues of the rainy season) as a substitute raw material for wood chips in both the core layer and the surface layers of particleboards. The bean shells as already discussed in the previous chapter (see section 3.1), are readily available around the world especially in Africa with no value-added uses and posses waste disposal problem in many of the localities where the crop cultivated especially in Cameroon.

5.4.1 Physical and mechanical properties

The produced particleboards were evaluated for their physical and mechanical properties and the results obtained were compared against the standard for particleboards used for interior applications (including furniture and interior fittings) under dry conditions (DIN EN 312:2010)

5.4.1.1 Internal bond strength

Figure 49 presents the internal bond (IB) strength of the manufactured particleboards. The boards of the panel series with 40 percent (w/w) substitution of the wood chips with the chips of bean shells (Bs) on the surface layer (40% Bs in SL), showed higher IB values for the PMDI (5% SL, 3% CL) bonded boards compared to their UF-resin bonded counterparts. Like most annual crop residues, the bean shells were observed to contain waxy substances on their surfaces. Because of this, the UF-resins could not effectively wet the surfaces of the chips of the bean shell (Roffael *et al.*, 2004; Li *et al.*, 2010; Sauter, 1996), thus inhibiting the formation of sufficient bond between the glue and the hydroxyl group of the chips. In addition, the high pH and high buffering capacities (see chapter 5.1.2) of the bean shells may have inhibited the proper curing of the pH-sensitive UF-resin.

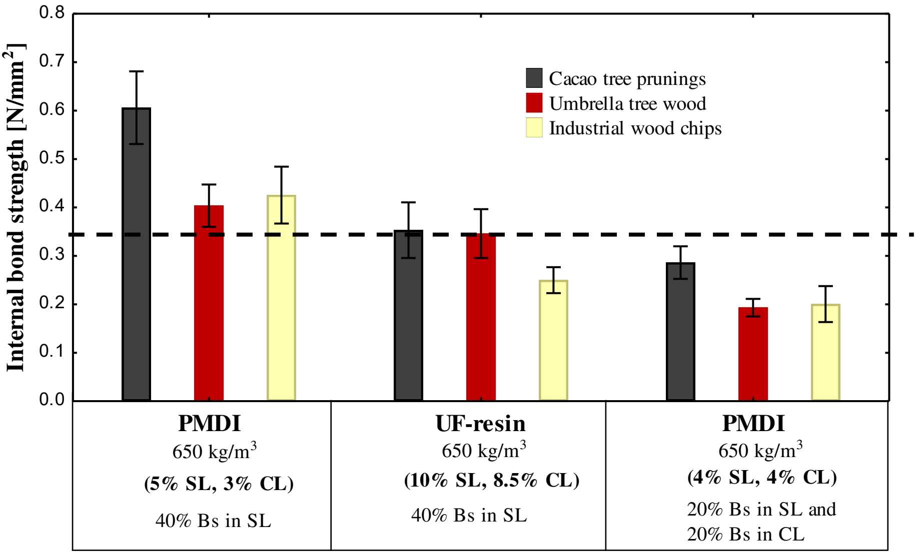


Figure 49: The internal bond (IB) strength of the UF-resin and PMDI bonded particleboards manufactured from umbrella tree wood, cacao tree prunings and industrial wood chips with the various substitutions of bean shells (Bs) on the surface layer (SL) and the core layer (CL) of the boards. Horrizontal line indicates the minimum value stipulated by DIN EN 312-2(2010).

Whereas, PMDI could effectively wet the surfaces of the bean shells despite the hydrophobic wax on their surfaces and form stronger chemical bonds (H-bond) as well as polyurethane bonds (covalent bond) with the bean shell chips. Besides, the PMDI is not pH sensitive as UF-resin, thus less likelihood of curing inhibition. Amongst the boards of the PB variant manufactured with a 40% substitution of the surface layers with the chips of bean shells (40% Bs in SL), bonded with PMDI (5% SL, 3% CL), the boards of cacao tree prunings achieved significantly higher ($p=0.00013$) IB values (0.61 N/mm^2) compared to the boards of umbrella tree wood (0.40 N/mm^2) and the boards of the industrial wood chip (0.42 N/mm^2). Comparing also amongst the UF-resin bonded particleboards with 40 percent bean shell substitution in the surface layer, no significant difference ($p=0.9092$) was observed in terms of the IB strength between the boards of umbrella tree wood and cacao tree prunings. However, both achieved significantly higher values of IB strengths ($p<0.001$) compared to the panels of industrial wood chips (40% Bs in SL).

Also, amongst the panels of the particleboard variant manufactured with 20:20 percent (w/w) bean shells substitution in both the surface and the core layers, bonded with PMDI (40% SL and 4% CL), the boards of cacao tree prunings showed superior IB strength (0.28 N/mm^2) compared to the boards of umbrella tree wood (0.19 N/mm^2) and boards of the industrial wood chips (0.20 N/mm^2). The result of this panel series indicates that the substitution of the wood chips in the middle layer with the chips of bean shells is not suitable for achieving particleboards with high enough IB strength, even when bonded with PMDI (resin load of 4%). Probably because the lower temperatures in the core layer compared to surface layers is not enough to cause the formation of effective bonds between the adhesive and the chips of the bean shell. Nevertheless, the IB strengths values of the panels with 40% bean shells substitution in the surface layer (40% Bs in SL) bonded with PMDI (5% SL, 3% CL) exceeded the 0.35 N/mm^2 minimum values as stipulated by the DIN EN 312-2 (2010) for particleboards of 13 mm -20 mm thickness. The boards of cacao tree prunings bonded with UF-resin (40% Bs in SL) also achieved the 0.35 N/mm^2 minimum IB value.

5.4.1.2 The bending properties

The modulus of rupture (MOR) of the manufactures particleboardss of the various production variants (bean shells substitutions), is presented in Figure 50. The particleboards of cacao tree prunings in each of the panels' variants achieved higher MOR compared to the boards of umbrella tree wood and the boards of the industrial wood chips. Amongst the variants with 40 percent substitution of their SL chips with the chips of bean shells, the MOR of the PMDI bonded boards (5% SL, 3% CL) was significantly higher ($p<0.005$) compared to the boards of the variant bonded with UF-resin.

Despite the low IB values of the panel variant with the 20:20 percent substitution of bean shell in the core layer and surface layers of the panels of the various wood materials, they achieved unexpectedly higher MOR values, with the boards of cacao tree prunings exceeding the 11 N/mm^2 minimum value for DIN EN 312-2 (2010). The MOR values of the PB variant manufactured by substituting the surface layer chips with the chips of bean shell (40% BS in SL), bonded with PMDI (5% SL, 3% CL), also exceeded the 11 N/mm^2 value for particleboards of 13 mm – 20 mm as required by DIN EN 312-2 (2010). None of the boards of the variant bonded with UF-resin could achieve the bending strength value of 11 N/mm2.

Figure 51 presents the bending modulus (MOE) as a function of sample stiffness of the manufactured particleboards of the various variants. The values of the MOE of the particleboards of the variant bonded with PMDI (5% SL, 3% CL), and the surface layers substituted by bean shell chips (40% BS in SL), did not differ significantly ($p>0.1801$) from one another. A similar trend was also observed for the UF-resin bonded boards (40% BS in SL). The results also indicate that amongst the panel variant with the surface layers substituted by bean shell chips (40% BS in SL), the stiffness properties of those bonded with PMDI (5% SL, 3% CL) did not differ significantly ($p>0.05$) from their UF-resin bonded counterparts. For the PMDI bonded variant (4% SL, 4% CL), with the 20:20 percent substitution of the surface layers and the core layer by beans shells, shows that the boards of industrial wood chips achieved higher MOE (2072 N/mm^2) followed by the boards of cacao tree prunings (1905 N/mm^2).

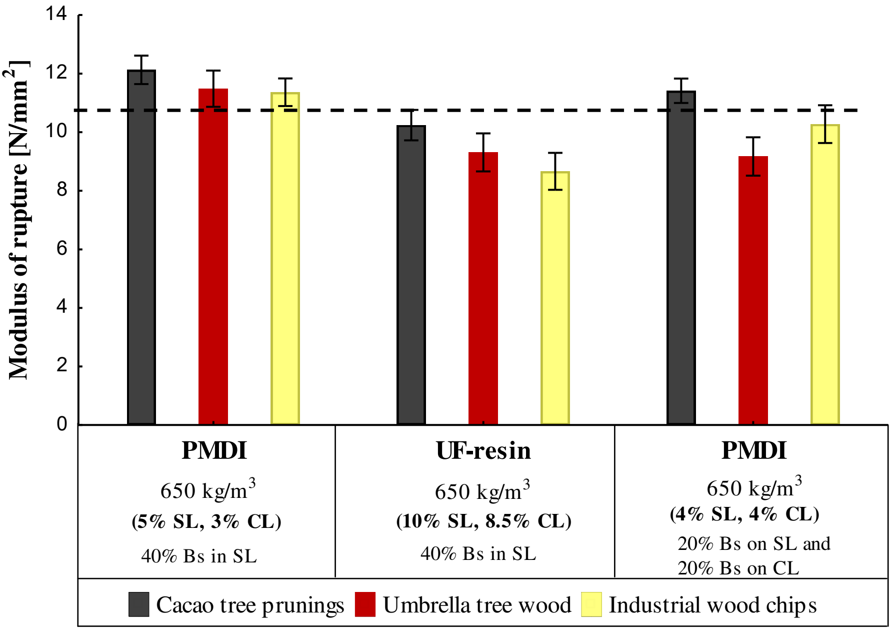


Figure 50: The modulus of rupture (MOR) of the UF-resin and PMDI bonded particleboards manufactured from umbrella tree wood, cacao tree prunings and industrial wood chips with the various substitutions of bean shells (Bs) on the surface layer (SL) and the core layer (CL). Horrizontal line indicates the minimum value stipulated by DIN EN 312-2(2010).

The umbrella tree wood boards of this variant had the least value of MOE (1765 N/mm^2), making them the only boards that failed to achieve the

1800 N/mm^2 minimum value for MOE as stipulated by the DIN EN 312-2 (2010) for particleboards of 13 mm - 20 mm thickness.

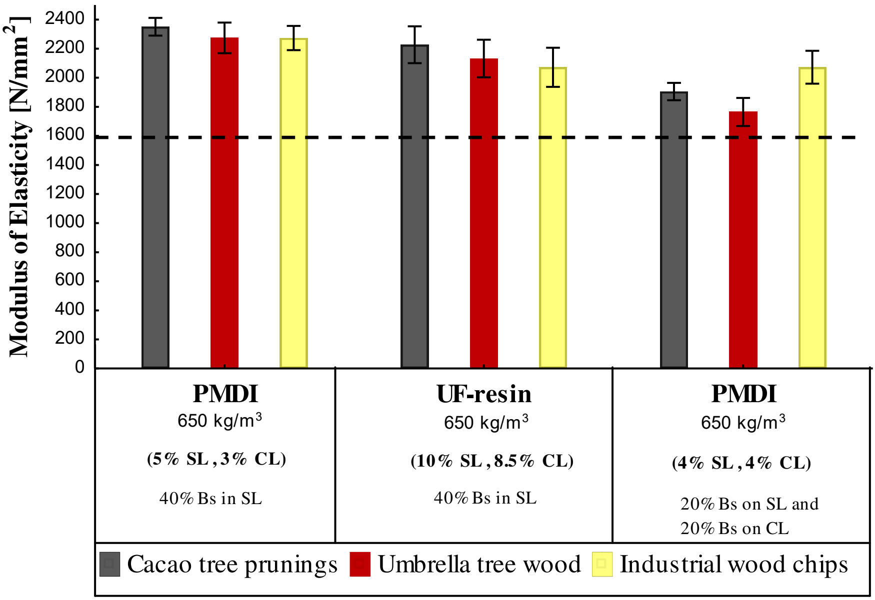


Figure 51: The modulus of elasticity (MOE) of the UF-resin and PMDI bonded particleboards manufactured from umbrella tree wood, cacao tree prunings and industrial wood chips with the various substitutions of bean shells (Bs) on the surface layer (SL) and the core layer (CL). Horrizontal line indicates the minimum value stipulated by DIN EN 312-2(2010).

5.4.1.3 Thickness swelling and water absorption

Thickness swelling (TS) and water absorption (WA) are some of the key parameters in determining the dimensional stability of wood-based composites. The TS and WA (after 24 hours) of the manufactured hybrid particleboards of the different wood materials with the various substitutions of the surface layers and core layers with the chips of bean shell bonded with the various binder systems are presented in Figure 52 and Figure 53 respectively.

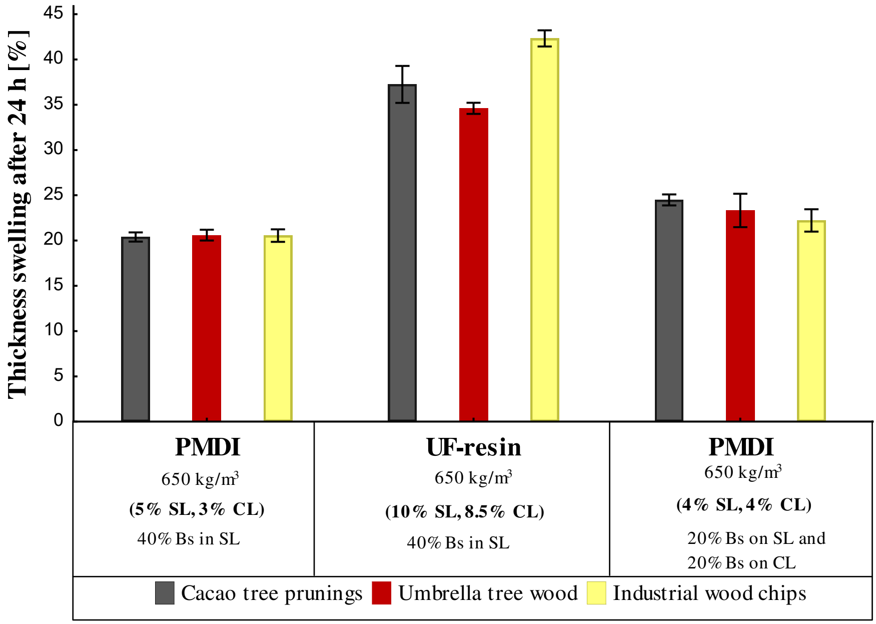


Figure 52: The thickness swelling (TS) after 24 hours of the UF-resin and PMDI bonded particleboards manufactured from umbrella tree wood, cacao tree prunings and industrial wood chips with the various substitutions of bean shells (Bs) on the surface layer (SL) and the core layer (CL).

As can be seen from Figure 52, the values of the 24 hours TS of the particleboard variants bonded with PMDI were significantly lower than the variant bonded with UF-resin (p<0.001). This is because the methylene bonds formed between the UF-resin and the furnish are easily hydrolyzed compared to the urethane bridges formed between PMDI and the furnish.

For the particleboards variant bonded with PMDI (5% SL, 3% CL) with their surface layers substituted by bean shells (40% BS in SL), the 24 hours thickness swelling of the boards of the various materials did not differ significantly from one another. A similar trend was also observed for the boards of the variant bonded with PMDI (4% SL, 4% CL) with 20:20 percent substitution of the core layer and the surface layers by the chips of bean shell.

Conversely, for the PB variant bonded with UF-resin (with 40% BS in SL), the 24 hours thickness swelling values of the panels of industrial wood chips (42.3%) were significantly higher (p<0.001) compared to the TS of the boards of umbrella tree wood (34.6%) and and the TS of the boards of cacao tree prunings (37.2%). The TS of the PB variant with the surface layers substituted by bean shells (40% BS in SL), bonded with PMDI (5% SL, 3% CL), showed the lowest TS rate compared to the other two variants. However, there is no required limit for TS and WA according to the DIN EN 312-2(2010).

Similar to the thickness swelling, the 24 hours WA of the PB of the variant bonded with PMDI were significantly lower than the WA of the boards of the variant bonded with UF-resin (p<0.001). Also, amongst the variants bonded with PMDI, the boards of the variant with the surface layers substituted by bean shell chips (40% BS in SL) had significantly lower WA values ($p<0.001$) compared to the boards of the variant manufactured by substituting both the surface layers (20% w/w) and the core layers (20% w/w) with bean shells chips.

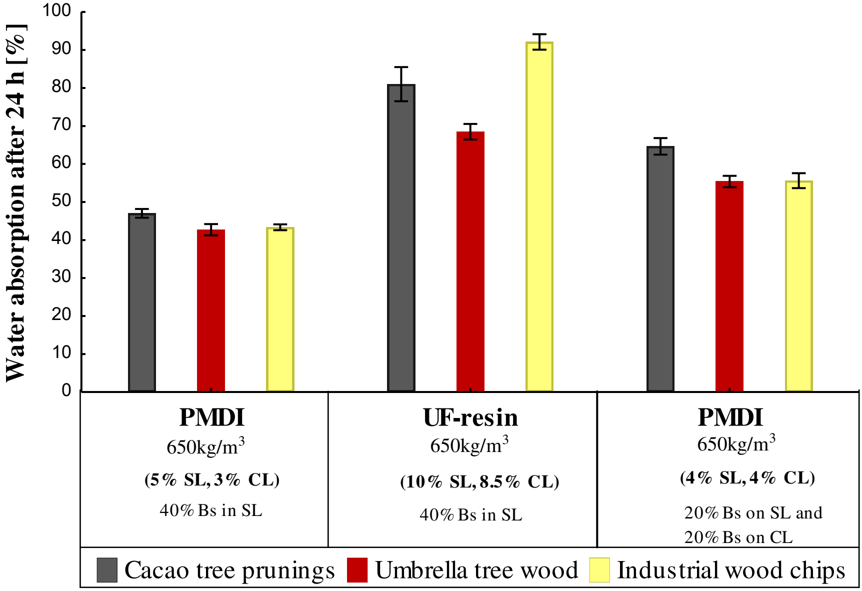


Figure 53: The water absorption (WA) after 24 hours of the UF-resin and PMDI bonded particleboards manufactured from umbrella tree wood, cacao tree prunings and industrial wood chips with the various substitutions of bean shells (Bs) on the surface layer (SL) and the core layer (CL).

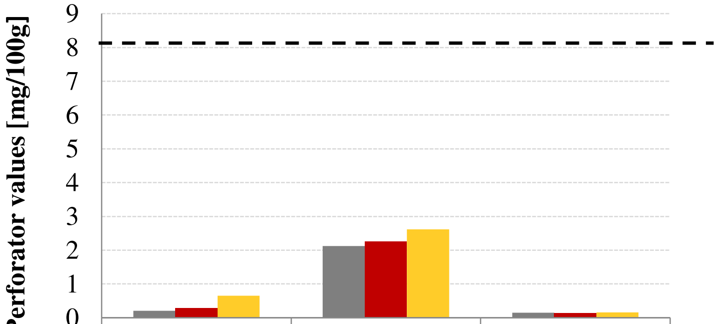
In each case, the WA of the boards containing the chips of cacao tree prunings were observed to be slightly higher compared to the boards containing the chips of umbrella tree wood. The 24 hours WA values of the boards of umbrella tree wood did not differ significantly from the boards of industrial wood chips in each of the variants bonded with PMDI ($p>0.5000$). However, for the variant bonded with UF-resin, the WA of the boards containing industrial wood chips was much higher than the WA of the boards containing umbrella tree wood

5.4.2 Formaldehyde release of the bean shells-wood chips hybrid PB

The formaldehyde release of the experimental particleboards manufactured in this study were evaluated based on two methods; the perforator method (EN 120) and the flask method (EN 717-3). The following chapters present the results of the evaluations patterning to the methods used. Due to the small number of measurements, the results presented here were not subject to statistical analysis.

5.4.2.1 Perforator values

The perforator method of evaluating the formaldehyde release of wood-based panels measures the amount of extractable formaldehyde present in a test sample (Xiong and Zhang, 2010; Salem *et al*., 2012a). Figure 54 shows the perforator values of the manufactured boards of the different variants. The boards of the PB variants bonded with UF resin showed the highest perforator values compared to the boards of the two variants bonded with PMDI. This is because unlike UF-resins, PMDI contains no free formaldehyde. Amongst the boards of the variant bonded with UF-resin, the boards containing the industrial wood chips had the highest perforator value (2.62 mg/100 g) compared to 2.26 mg/100 g for the boards containing the umbrella tree wood and 2.12 mg/100 g for the boards containing cacao tree prunings.



	PMDI 650 kg/m³ (5% SL, 3% CL) 40% Bs in SL	UF-resin 650kg/m³ (10% SL, 8.5 % CL) 40% Bs in SL	PMDI 650 kg/m³ (4% SL, 4% CL) 20% Bs on SL and 20% Bs on CL
Cacao tree prunings	0.21	2.12	0.15
Umbrella tree wood	0.28	2.26	0.14
Industrial wood chips	0.65	2.62	0.16

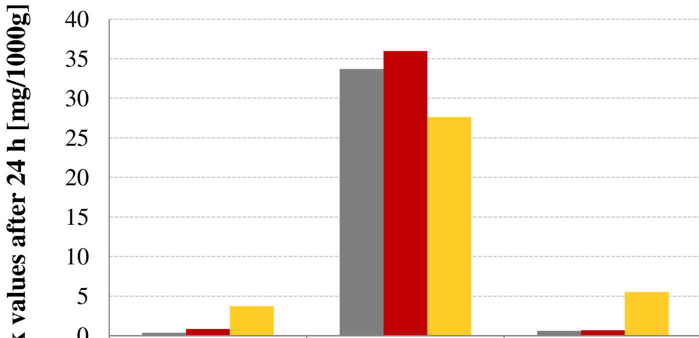
Figure 54: The perforator values (at 6.5% MC) of the UF-resin and PMDI bonded particleboards manufactured from umbrella tree wood, cacao tree prunings and industrial wood chips with the various substitutions of bean shells (Bs) on the surface layer (SL) and the core layer (CL). Horrizontal line indicates the maximum limit for E1emissions standard (8.0 mg/100g).

It is also noticeable that in each of the variants, the perforator values of the hybrid boards containing industrial wood chips were higher compared to the hybrid boards containing the chips of umbrella tree wood as well as the hybrid boards containing the chip of cacao tree prunings. The presence of recycling materials in the industrial wood chips might explain the reason for the higher perforator values of the PB containing these wood chips. Based on the results, one essential statement can be made; the perforator values of the manufactured panels of all PB variants were negligible compared to the E1 maximum limit of 8.0 mg/100 g, especially for the variants bonded with PMDI.

5.4.2.2 Flask values

Unlike the perforator method, the flask method measures the amount of formaldehyde that is emitted from the surfaces and the edges of the test sample (Xiong and Zhang, 2010; Salem *et al*., 2012a). Figure 55 shows the flask values of formaldehyde of the experimental particleboards presented in this study. Like the perforator values, the flask values of the boards of the PB variant bonded PMDI were observed to be negligible as compared to the boards of the variant bonded with UF-resin. It can also be seen that the flask values of the boards containing the industrial wood chips (for variants bonded with PMDI) were higher compared to the boards containing of umbrella tree wood and cacao tree prunings.

Contrary to the perforator values of the UF-resin bonded boards, the flask value of the boards containing the industrial wood chips (27.62 mg/1000 g) was seen to be lower compared to the values of the boards containing the chips of umbrella tree wood (35.96 mg/1000 g) as well as those containing the chips of cacao tree prunings (33.7 mg/1000 g).



	PMDI 650 kg/m³ (5 % SL, 3 % CL) 40% Bs in SL	UF-resin 650kg/m³ (10% SL, 8.5% CL) 40% Bs in SL	PMDI 650 kg/m³ (4% SL, 4% CL) 20% Bs on SL and 20% Bs on CL
Cacao tree prunings	0.38	33.7	0.6
Umbrella tree wood	0.86	35.96	0.7
Industrial wood chips	3.72	27.62	5.5

Figure 55: The 24 hours flask values of the UF-resin and PMDI bonded particleboards manufactured from umbrella tree wood, cacao tree prunings and industrial wood chips with the various substitutions of bean shells (Bs) on the surface layer (SL) and the core layer (CL).

The residual moisture content of the boards containing the industrial wood chips (4.4%) was observed to be lower compared to the boards containing the chips of the umbrella tree wood (7.4%) and the chips of cacao tree prunings (7.3%). According to Aydin *et al*., 2006, the flask values of formaldehyde are significantly affected by the board's moisture content. The lack of proportionality in the flask and perforator values of the boards of industrial wood chips is because the flask values were not corrected to the same moisture content as were the perforator values (6.5%).

5.4.3 Summary of the results of the bean shells-wood chips hybrid PB

The study presented in this chapter investigates the technical possibilityof producing particleboards of comparable performance to the particleboards of the standard DIN EN 312-2 (2010) by substituting the different wood chips with the chips of the annual crop residues, bean shells (rainy season residues), in both the core and surface layers of the boards. An evaluation of the physical and mechanical properties revealed that:

- The production of particleboards of the density 650 kg/m^3 for interior applications used in dry conditions is technically possible by substituting wood chips in the surface layers with bean shell chips (40% w/w) when bonded with PMDI. This means, it is possible to replace up to 40% (w/w) of wood with bean shells in the manufacturing process of particleboards.
- Urea-formaldehyde resin (K340) is not suitable as a bonding agent for particleboards involving bean shells. The presence of the waxy substances on the surfaces of bean shell prevents the penetration of UF-resin beyond the surfaces of the chips to form strong

interlocks with the hydroxyl group of the bean shells, hence poor adhesive bonds.

- The substitution of the wood chip in the core layer and the surface layers of the particleboards (20% and 20% w/w respectively) with bean shells did not result in boards with high enough mechanical strengths (IB) to meet the requirement of DIN EN 312-2 (2010).
- The formaldehyde emissions of the produced panels are at relatively lower levels especially for the PMDI bonded boards compared to the maximum permitted value for E1 (8.0 mg/100 g) when tested by the perforator method.

5.5 The results of the UF-resin bonded particleboards based on the chips of umbrella tree wood and bean crop residues of the dry season.

The following chapter presents the results of the experimental work to investigate the possibility of producing UF-resin bonded particleboards of the standard DIN EN 312-2 (2010) by substituting 10%, 15% and 20% of the chips of the umbrella tree wood with chips of the residues of bean crop (dry season residues) in the core layer of the boards. For the purpose of comparison, the results of the particleboards manufactured with 100% bean crop residues are also presented and described.

5.5.1 The physical and mechanical properties

The manufactured particleboards were evaluated for their physical and mechanical properties and the results obtained were compared against the standard for particleboards used for interior applications (including furniture and interior fittings) under dry conditions (DIN EN 312:2010).

5.5.1.1 Internal bond strength

Figure 56 illustrates the results of the internal bond (IB) strength of the manufactured boards. The IB strength was observed to decrease with increase in the substitution of umbrella tree wood chips with the chips of bean crop residues (from 10% BCR to 20% BCR). The particleboards made by substituting 10% of the umbrella tree wood with bean crop residues (10% BCR) achieved the highest IB strength (0.71 N/mm^2) compared to the boards manufactured by substituting 15% (0.55 N/mm^2) and 20% (0.45 N/mm^2) of th wood chips with bean crop residues.The boards manufacture exclusively from the bean crop residues achieved significantly lower ($p<0.0002$) IB strength (0.20 N/mm^2) compared to the rest of the boards.

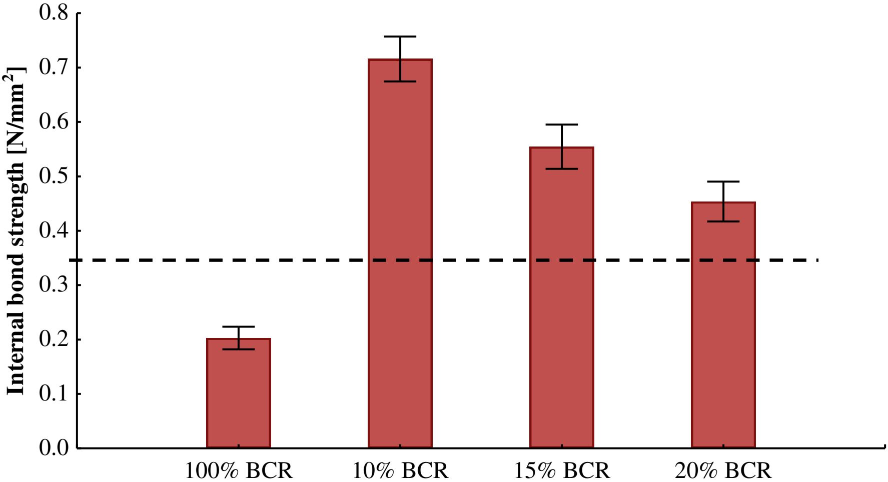


Figure 56: The internal bond (IB) strengths of the UF-resin bonded particleboards manufactured by the substitution of 10% 15% and 20% of umbrella tree wood chips in the core layer with the chips of bean crop residues(BCR), and the IB strength of the boards containing 100% bean crop residues. Horrizontal line indicates the minimum value stipulated by DIN EN 312-2(2010).

As earlier explained, the bean crop residues like most annual crops contained waxy substances on their surfaces, which make it difficult for the chips to form strong bonds with conventional UF-resin (see chapter 5.4.1.1). According to the DIN EN 312-2 (2010), the minimum requirement for IB strength for particleboards of the thickness, 13 mm - 20 mm, is 0.35 N/mm^2. From the results, the IB strengths of the PB manufactured by substituting 10%, 15%, and 20% of the umbrella tree wood chips of the core layer with the chips of the bean crop residues met the requirement of this standard. The boards made exclusively from bean crop residues (100% BCR) fell short.

5.5.1.2 Bending strength

The bending strengths (BS), also known as the modulus of rupture, of the manufactured particleboards measured according to EN 319 are presented in figure 57. The BS of the boards made by substituting 10%, 15% and 20% of the umbrella tree wood chips of the core layer with bean crop residues did not differ significantly from one another ($p>0.500$). However, the highest BS (12.25 N/mm^2) was observed for the boards made by substituting 10% of the umbrella tree wood chips of the core layer with the chips of bean crop residues (10% BCR). The other two variants, 15% BCR and 20% BCR had bending strength values of 11.47 N/mm^2 and 11.41 N/mm^2 respectively. The least bending strength (7.48 N/mm^2) was observed for the particleboards made exclusively from bean crop residues (100% BCR).

The minimum requirement for bending strength according to DIN EN 312-2 (2010) is 11 N/mm^2 for particleboards of 13-20 mm thickness. It can be seen from the results that the bending strengths of the boards manufactured by substitution 10%, 15% and 20% of the umbrella tree wood chips of the core layer with bean crop residues slightly exceeded the minimum value required by DIN EN 312-2 (2010).

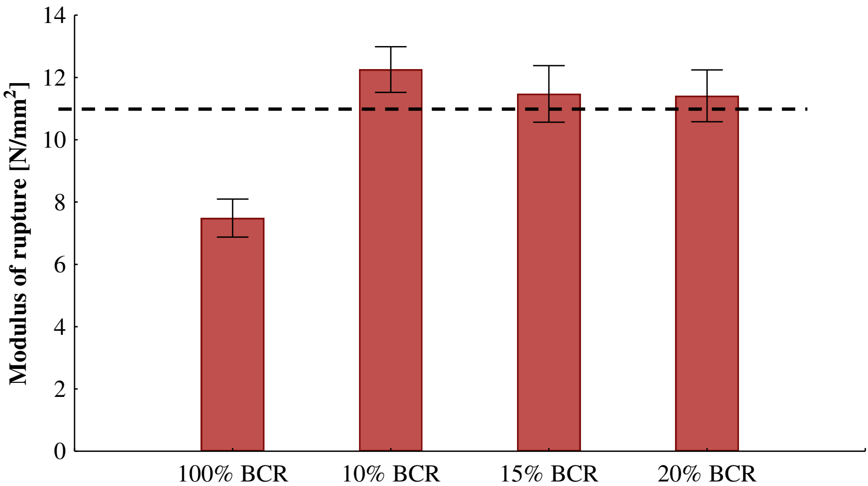


Figure 57: The Bending strength (BS) of the UF-resin bonded particleboards manufactured by substitution 10% 15% and 20% of umbrella tree wood chips in the core layer with the chips of bean crop residues(BCR), and the BS strength of the boards containing 100% bean crop residues. Horrizontal line indicates the minimum value stipulated by DIN EN 312-2(2010).

5.5.1.3 Modulus of Elasticity

Figure 58 shows the modulus of elasticity (MOE) of the particleboards of the various substitution levels of umbrella tree wood with the chips of bean crop residues. Like the results of the bending strength, the MOE of the boards manufactured by substituting 10%, 15% and 20% of the chips of umbrella tree wood of the core layer with the chips of the bean crop residues (10% BCR, 15% BCR, and 20% BCR) did not differ significantly from one another ($p>0.300$).

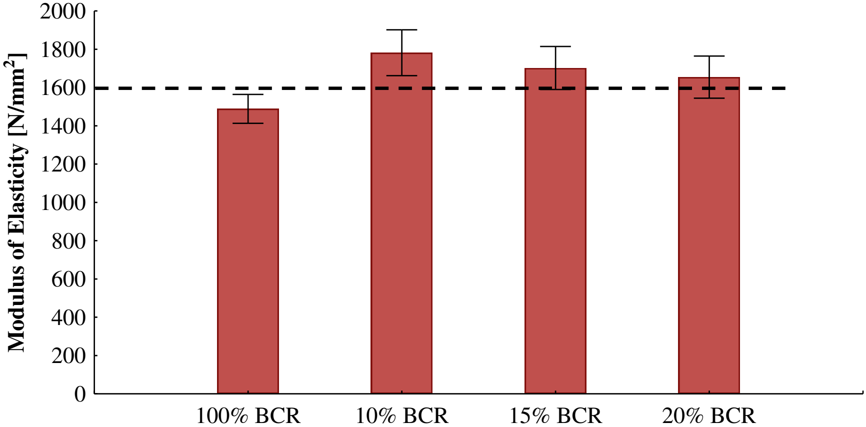


Figure 58: The modulus of elasticity (MOE) of the UF-resin bonded particleboards manufactured by substitution 10%, 15% and 20% of umbrella tree wood chips in the core layer with the chips of bean crop residues (BCR), and the MOE of the boards containing 100% bean crop residues. Horrizontal line indicates the minimum value stipulated by DIN EN 312-2(2010).

Nevertheless, the highest MOE (1782 N/mm^2) was observed for the boards made by substitution 10% of the wood chips with the chips of the bean crop residues (10% BCR). The board variants, 15% BCR and 20% BCR had MOE values of 1702 N/mm^2 and 1654 N/mm^2 respectively. The least MOE (1488 N/mm^2) was observed for the particleboards made exclusively from the chips of the bean crop residues (100% BCR). According to DIN EN 312-2 (2010), the minimum requirement of MOE for particleboards of 13 mm - 20 mm thickness is 1800 N/mm^2. From the results, the values of MOE of all the boards were lower than the minimum value required by DIN EN 312-2 (2010).

5.5.1.4 Thickness swelling and water absorption

The 2 hours and 24 hours thickness swelling (TS) and water absorption (WA) of the manufactured particleboards are summarized in Figure 59 and Figure 60 respectively. The TS values (39.2% and 52.3% for 2 hours and 24 hours respectively) of the boards made exclusively from bean crop residues (100% BCR) were seen to be significantly higher ($p<0.0001$) than the boards made from the various substitutions of umbrella tree wood chips with the chips of the bean crop residues.

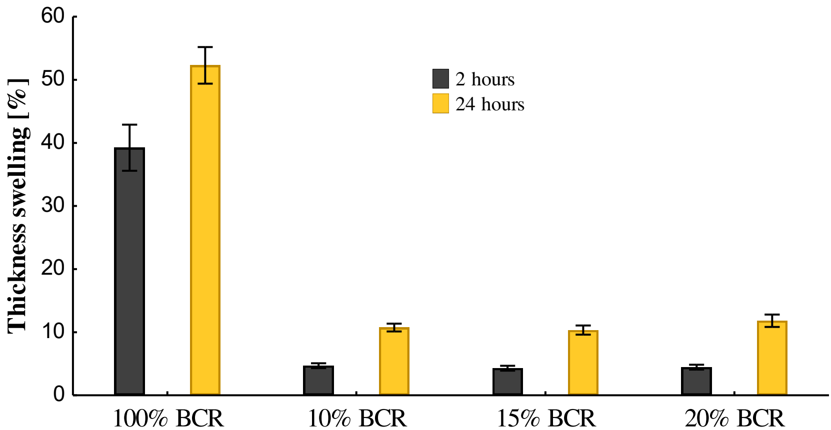


Figure 59: The thickness swelling (TS)after 2h and 24 h of the UF-resin bonded particleboards manufactured by substitution 10%, 15% and 20% of umbrella tree wood chips in the core layer with the chips of bean crop residues (BCR), and the TS of the boards containing 100% bean crop residues.

No significant differences in TS (P>0.0900) were observed amongst the boards manufactured by substituting 10%, 15% and 20% of the wood chips with the chips of the bean crop residues (10% BCR, 15% BCR, and 20% BCR).

Like the TS, the highest WA values (149.5% and 182.8% for 2 hours and 24 hours respectively) were observed for the boards manufactured exclusively from bean crop residues (100% BCR), which were also significantly higher than the values of the boards made by substituting 10%, 15% and 20% of the wood chips with the chips of bean crop residues.

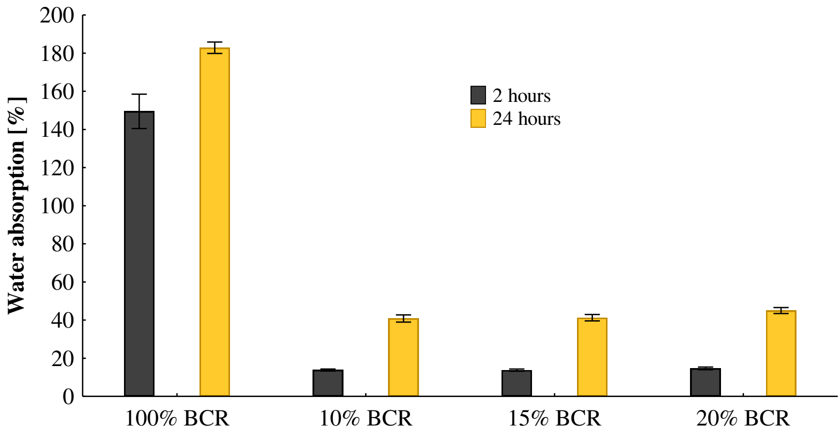


Figure 60: The water absorption (WA) after 2h and 24 h of the UF-resin bonded particleboards manufactured by substitution 10%, 15% and 20% of umbrella tree wood chips in the core layer with the chips of bean crop residues (BCR) and the WAof the boards containing 100% bean crop residues.

In contrast to the TS, the 24 hours WA value (44.9%) of the boards made by substituting 20% of the core layer wood chips with chips of the bean crop residues (20% BCR), were seen to be significantly higher ($p<0.001$) than the TS of the variants with 10% BCR (40.8%) and 15% BCR (41.3%).

5.5.2 Formaldehyde release

The formaldehyde release of the experimental particleboards manufactured boards in this study were evaluated using two methods; the perforator method (EN 120) and the flask method (EN 717-3). The perforator values (Figure 61) of the boards made by substituting 15% (12.2 mg/100 g) and 20% (12.4 mg/100 g) of the wood chips with the chips of the beans crop residues were observed to be higher compared to the boards of the variant with 10% BCR (8.7 mg/100g). The boards made exclusively from the chips of the bean crop residues (100% BCR) had the perforator value of 9 mg/100 g.

Conversely, the 3 hours and the 24 hours flask values were seen to be higher for the board variant made exclusively from the bean crop residues (15.2 mg/1000 g and 111.3 mg/1000 g respectively) compared to the boards made 10%, 15%, and 20% substitution of the wood chips by the chips of the bean crop residues. While the perforator method measures the extractable formaldehyde present in the test sample, the flask method measures the amount of free formaldehyde emitted from its surfaces. The higher the resin load the higher the amount of free formaldehyde. The disparity in the results between the two methods may be caused by the difference in resin load between the boards of 100% BCR and the other board variants (see Table 10). Based on the results, the perforator values of all the boards exceeded the 8.0 mg/100 g limit of E1.

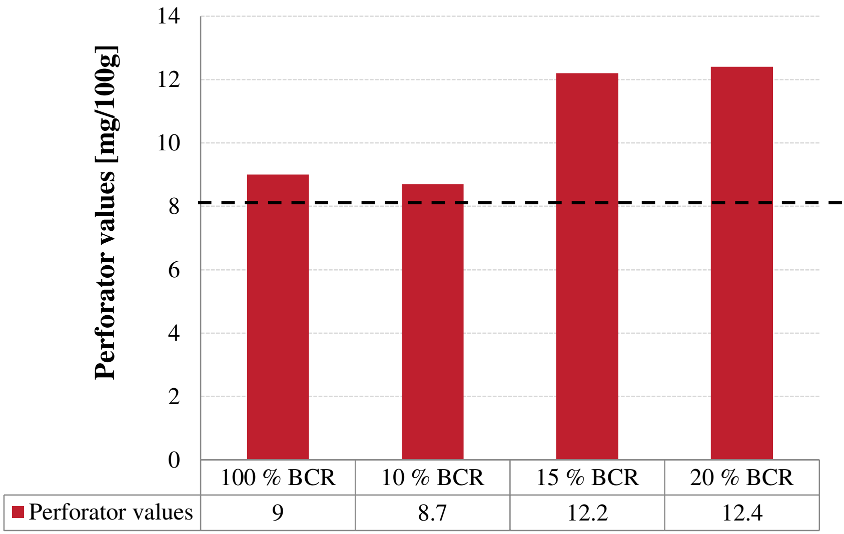
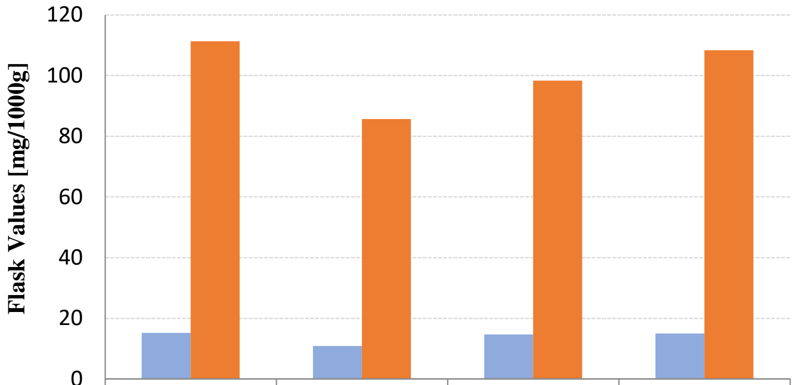


Figure 61: The perforator values (at 6.5% moisture content) of the UF-resin bonded particleboards manufactured by substitution 10%, 15% and 20% of umbrella tree wood chips in the core layer with the chips of bean crop residues (BCR), and the perforator values of the boards containing 100% bean crop residues. Horrizontal line indicates the maximum limit for E1 emissions standard (8.0 mg/100 g).



	100% BCR	10% BCR	15% BCR	20% BCR
3 hours	15.2	10.9	14.7	15
24 hours	111.3	85.7	98.3	108.4

Figure 62: The flask values of the UF-resin bonded particleboards manufactured by substitution 10%, 15% and 20% of umbrella tree wood chips in the core layer with the chips of bean crop residues (BCR), and the flask values of the boards containing 100% bean crop residues.

5.5.3 Summary of the results of the UF-resin bonded particleboards based on the chips of umbrella tree wood and bean crop residues of the dry season

The results of the experimental work to investigate the possibility of producing UF-resin-bonded particleboards of the standard DIN EN 312-2 (2010) by substituting 10%, 15% and 20% of the chips of the umbrella tree wood with chips of the residues of bean crop (dry season residues) in the core layer of the boards revealed the following:

- By using UF-resin K350 it is possible to produce particleboards with high enough mechanical strengths (IB and MOR) to satisfy the requirement of DIN EN 312-2 (2010) by replacing up to 20% of the wood chips in the middle layer of the board made predominantly of umbrella tree wood with bean crop residues.
- by increasing in the amount of bean crop residues (from 10%, 15% to 20%) in the composite matrix results in a significant deterioration of transverse tensile strengths (IB) of the resulting boards, but the MOR, MOE and the physical properties did not differ significantly from one another.
- The perforator values of the formaldehyde release of all the boards exceeded the maximum limit of 8.0 mg/100 g as required by the E1 standard.
- It is technically impossible to produce particleboards of the standard used for interior applications under dry condition (DIN EN 312-2 (2010) exclusively from the bean crop residues (100% BCR) when bonded with the UF-resin K350.

6.0 Summary

Against the background of the soaring wood prices, the existing wood and waste wood scarcity in the wood-based panel industry, the demand for a more efficient use of the raw material wood, and the fact that wood materials with high formaldehyde emission are no longer accepted by consumers, eco-friendly alternatives for wood and formaldehyde-containing binders should be extensively researched. The focus of this thesis was to investigate the possibility of producing particleboards with low formaldehyde emission using alternative materials. This goal and additionally the reduction of the panels' density shall be achieved by using the wood of fast-growing tree species, the umbrella tree (*Musanga cecropoides*), and the prunings of the cocoa tree (*Theobroma cacao*). In addition to its fast growth, the umbrella tree stands out because it places few demands on the location and is available in large quantities. Cocoa tree prunings (25 t/ha yearly) accumulate regularly and are usually left in the fields to rot, and in some cases burned. In this work, a meaningful material use is ensured.

In addition to the umbrella tree wood and the prunings of cacao tree (wood materials), it also investigated whether the residues of the annual crop bean (*Phaseolus vulgaris*) are suitable for particleboards production.

This study was conducted in two phases, the raw material characterization phase (first phase) and the particleboards manufacturing phase. The first phase involved a comprehensive investigation of the physical and chemical properties of the raw materials. In addition to the study raw materials, wood chips of Norway spruce (*Picea abies*) were used for comparison.

The bean crop residues were divided into two categories based on the harvesting and processing season; the rainy season bean crop residues (referred to in this study as bean shells) consisted of 100 percent bean shells and the dry season bean crop residues, consisted of a mixture of bean shells and bean stems.

Based on the results of the physical and chemical analysis of the study raw materials, the following conclusions can be made:

- The evaluation of the bulk densities revealed that the bulk densities of the bean crop residues of both seasons (for both the surface and core layer chips) were well below the bulk densities of the wood materials. Amongst the wood materials, the chips of umbrella tree wood showed comparatively lower bulk densities for both the surface and core layers chips (127 kg/m^3 and 92 kg/m^3 respectively) compared to the chips of cacao tree prunings.
- Analysis of the pH and buffering capacities revealed a higher pH value (6.4) for the bean crop residues of the dry season compared to 5.6 for the bean shells. Amongst the wood material, the cacao tree prunings and umbrella tree wood had similar pH values of 6.8 and 6.2 respectively, compared to spruce wood (5.4).
- The umbrella tree wood and cacao tree prunings, despite their high pH values, exhibited the least buffering capacity values of 0.54 mmol NaOH/100 g and 0.57 mmol NaOH/100 g respectively. The buffering capacity values of the bean shells (14.27 mmol

NaOH/100 g) and the bean crop residues of the dry season (6.24 mmol NaOH/100 g) were substantially higher than the rest of the study raw materials.

- Examination of the extractives content of the research raw materials revealed that the umbrella tree wood contained the least amount of water-soluble extractives (0.28% for cold water and 1.1% for hot water) but contained the highest amount of ethanol-cyclohexane extractives (3.6%) compared to the rest of the wood materials. The water-soluble extractives content of the bean crop residues of both seasons ranged from 14.58% to 14.7% for cold water and 14.25% to17.5% for hot water.
- The analysis of pentosan content showed values of 13% and 16.7% for umbrella tree wood and cacao tree prunings respectively, which were seen to be higher than the values of the bean crop residues as well as the value of spruce wood. The pentosan content of the bean shells (8.2%) was not any different from that of spruce wood (8.5%); rather, it was seen to be relatively lower than the value of the dry season bean crop residues (10.2%).
- The klason lignin contents of cacao tree prunings (39.2%) and umbrella wood (37%) were substantially higher compared to the klason lignin contents of the bean crop residues of both seasons, and the spruce wood (29.4%).

- Finally, the ash contents of the bean crop residues (8.5 -8.9%) were much higher in comparison to the ash content of the wood materials. Amongst the wood materials, cacao tree prunings and umbrella tree wood contained higher amounts of ash (2.1% and 1.85% respectively) compared to spruce wood (0.36%).

The particleboards' manufacturing phase of this study was divided into four parts. The first part investigated the possibility of producing UF-resin (K340) bonded particleboards of reduced formaldehyde emissions based on the raw material; umbrella tree wood, and cacao tree prunings. For comparison purposes, particleboards of spruce wood were also produced. In addition to this, the first part also investigated the effects of coarser core layer chips on the properties of the particleboards of umbrella tree wood and cacao tree prunings. The results of the mechanical-technological properties suggest that it is technically feasible to produce particleboards of reduced formaldehyde emissions of the density 650 kg/m^3 and 550 kg/m^3 of the standard DIN EN 312:2010 (type 2) from the raw materials, umbrella tree wood, and cacao tree prunings when bonded with UF-resin K340. However, the coarseness of the core layer chips must be appropriate for the boards to achieve high enough bending and stiffness strengths.

The possibility to produce non-formaldehyde containing particleboards based on the raw materials, umbrella tree wood, and cacao tree prunings, was the focus of the second part of the particleboards' manufacture. To achieve this, an isocyanate-based adhesive (PMDI) and a natural binder based on animal protein, blood albumin, were used. The wood chips of the core layer and the surface layers were separately blended with the different adhesives. Particleboards of the densities 650 kg/m^3 and 550 kg/m^3 were produced in a pilot plant scale. Evaluation of the mechanical-technological properties revealed that despite the higher internal bond (IB) strengths of the boards of cacao tree prunings compared to the boards of umbrella tree wood, their bending strengths (BS) and stiffness characteristics (MOE) on the other hand were considerably lower compared to the boards of umbrella tree wood. It was also revealed that combining the chips of umbrella tree wood and cacao tree prunings in the ratio of 50:50 percent (w/w) on both the surface and the core layer will results in particleboards with superior bending strengths and MOE compared to the boards made exclusively from cacao tree prunings. In addition, the formaldehyde release of the boards of both binder systems was observed to be negligible compared to the limit of E1. The recorded formaldehyde values of the board could be attributed to the so-called natural formaldehyde of the raw materials because the binder systems contained no free formaldehyde. The overall findings indicated that the production of non-formaldehyde containing particleboards of the density 650 kg/m^3 for general purpose applications including furniture (DIN EN 312:2010) is technically possible with umbrella wood as raw material when bonded with PMDI. Also, a mixture of umbrella tree wood and cacao tree prunings in the ratio of 50:50 percent (w/w) will results in particleboards with high enough mechanical strengths to meet the requirements of DIN EN

312:2010 when PMDI is used as the glue. The IB values of all the boards bonded with blood albumin exceeded the minimum value specified by the DIN EN 312:2010 standard. However, their MOR (except for the boards made from a mixture of both materials, of the density 650 kg/m^3) and MOE failed to achieve the required values.

The third part of the project focused on investigating the feasibility of bean shells (bean crop residues of the rainy season) to serve as a substitute raw material for wood in both the core and surface layers of particleboards. Three-layered particleboards (density 650 kg/m^3) of umbr- ella tree wood, cacao tree prunings, and industrial wood chips were produced in three production series'. In the first and the second series, the proportions were 100 percent bean shells on the surface layers and 100 percent of the various wood materials in the core layer. That is a 40% substitution of the wood chips of each board with bean shells. The panels of the first series were bonded with PMDI and the second series by UF-resin (340). In the third production series, however, 20 percent (w/w) of the surface layers and 20 percent (w/w) of the core layer of each board were substituted with bean shells. The boards of the third series were also bonded with PMDI (resin load of 4% on the SL and the CL). The substitution level of the wood chips with bean shells was also 40%. The results of the evaluation of the physical and mechanical properties were compared to the required minimum values for commercial particleboards (DIN EN 312:2010 standard for type 2 PB). The particleboards of first production series (100% bean shells on the surface layer and 100% wood materials on the core layer) were able to exceed the minimum required values of internal bond strength, bending strength and modulus of elasticity. In addition, the formaldehyde release (evaluated by the flask and perforator methods) of the boards of the first series was also negligible. The panels bonded with UF-resin (series 2), as well as the particleboards manufactured with bean shells substituted in both the core layerand the surface layers (third series) did not have high enough mechanical strength to satisfy the requirement for type 2 particleboards of the DIN EN 312:2010 standard. Overall, based on the mechanical performance of the boards of the first production series, bean shells can be considered as an alternative surface layer material for particleboards of satisfactory performance for use in interior fittings and furniture applications.

The fourth part of the experimental work was to investigate the possibility of producing UF-resin bonded particleboards of the standard DIN EN 312:2010 by substituting 10%, 15% and 20% of the chips of the umbrella tree wood with chips of bean crop residues of the dry season in the core layer of the boards. In addition to these boards, another variant of particleboards made exclusively from the bean crop residues was manufactured. The results revealed that increasing the amount of bean crop residues (from 10%, 15%, and 20%) in the composite matrix significantly reduces the internal bond strength of the particleboards but bending strength, MOE, and the water-related properties are not significantly affected. It is technically possible to produce particleboards with high enough IB and BS to satisfy the requirement of type 2 particleboards of the DIN EN 312:2010 standard by replacing up to 20% of the chips of umbrella tree wood with chips of the bean crop residues in the core layer of the board when bonded with UF-resin (350). However, the MOE values of the boards were slightly lower than the minimum required value (1800 N/mm^2). The properties of the particleboards manufactured exclusively from the bean crop residues did not meet the requirements of this standard. The formaldehyde release of all the boards (perforator values) exceeded the E1 limit value of 8.0 mg/100 g of dry board.

The overall findings of this project indicate that umbrella tree wood, cacao tree prunings and the bean crop residues of both seasons are valuable and potential alternative biomass that could be used wholly or as a substitute for wood in particleboards production. In the future, there could be a possibility to use these previously unused materials for chipboard and to establish them locally in products, for example in Cameroon.

7.0 Zusammenfassung

Vor dem Hintergrund der steigenden Holzpreise, der bestehenden Holz- und Altholzknappheit in der Holzwerkstoffindustrie, der Forderung nach einer effizienteren Nutzung des Rohstoffes Holz und der Tatsache, dass Holzwerkstoffe mit kritischem Formaldehydausstoß keine Verwendung mehr finden, sollten von den Verbrauchern akzeptierte umweltfreundliche alternative Bindemittel mit geringem Formaldehydgehalt oder ohne eingehend untersucht werden. Der Schwerpunkt dieser Dissertation lag auf der Untersuchung der Möglichkeit, Spanplatten mit geringer Formaldehydemission unter Verwendung alternativen Rohstoffen und Bindemitteln herzustellen. Dieses Ziel und die Reduzierung der Rohdichte soll erstmals durch den Einsatz von Holz der schnell wachsenden Baumarten Schirmbaum (*Musanga cecropoides*) und Kakaobaum (*Theobroma cacao*) erreicht werden. Schirmbaum zeichnet sich neben seiner Schnellwüchsigkeit dadurch aus, dass er wenig Ansprüche an den Standort stellt und in Afrika in hohen Mengen vorkommt. Holz aus Kakaobaumabschnitten fallen in Kamerun in Höhe von 25 t/ha jährlich an und werden üblicherweise zum Zwecke die Energiegewinnung verbrannt. In dieser Arbeit ist somit eine sinnvolle stoffliche Nutzung angestrebt. Neben dem Schirmbaum- und Kakaobaumholz wurde auch untersucht, ob die Rückstände der einjährigen Gartenbohne (*Phaseolus vulgaris*) ebenfalls für die Herstellung von Spanplatten geeignet sind.

Diese Dissertation wurde in zwei Teilabschnitten durchgeführt. Im ersten Teil wurde die Rohstoffcharakterisierung und im zweiten Teil die Entwicklung der Spanplatten vorgenommen. Zunächst wurden die physikalischen und chemischen Eigenschaften der Rohstoffe umfassend untersucht. Als Referenz zu den o.g. Rohstoffen wurden Späne aus Fichtenholz (*Picea abies*) herangezogen. Die Ernterückstände der Bohnenfrüchte wurden je nach Ernte- und Verarbeitungszeit in zwei Kategorien eingeteilt: die Ernterückstände der Regenzeit bestanden zu 100 Prozent aus Bohnenhülsen und die Rückstände der Trockenzeit aus einer Mischung aus Bohnenhülsen und Bohnenstängeln. Basierend auf den Ergebnissen der physikalischen und chemischen Analyse der untersuchten Rohstoffe können folgende Schlussfolgerungen gezogen werden:

- Die Auswertung der Schüttdichten ergaben, dass die Späne der Bohnenernterückstände beider Jahreszeiten deutlich unter den Schüttdichten des Schirmbaum- und Kakaobaumholzes lagen. Bei den Spanplatten zeigten die Schirmbaumspäne sowohl für die Deckschicht als auch für die Mittelschicht eine geringere Schüttdichte (127 kg/m^3 bzw. 92 kg/m^3) im Vergleich zu den Kakaobaumspänen.

- Die Analyse der pH-Werte ergab für die Bohnenernterückstände der Trockenzeit einen höheren pH-Wert von 6,4 im Vergleich zu 5,6 für die Bohnenhülsen aus der Regenzeit. Die Späne aus Kakaobaum- und Schirmbaumholz wiesen ähnliche pH-Werte von 6,8 bzw. 6,2 auf. Die Fichtenholzspäne hatten einen einem pH-Wert von 5,4.

- Die Schirmbaum- und Kakaobaumholzspäne zeigten trotz ihrer hohen pH-Werte die niedrigsten Pufferkapazitätswerte von 0,54 mmol NaOH/100 g bzw. 0,57 mmol NaOH/100 g. Die Pufferkapazitätswerte der Bohnenhülsen (14,27 mmol NaOH/100 g) und der Bohnenernterückstände der Trockenzeit (6,24 mmol NaOH/100 g) waren wesentlich höher als die der übrigen Rohstoffe.

- Die Untersuchung der Extraktstoffgehalte ergab, dass die Schirmbaumspäne im Vergleich zu den restlichen Spänen die geringste Menge an wasserlöslichen Extrakten enthielt (0,28 % nach Kaltwasser- und 1,1 % nach Heißwasserextraktion), jedoch die höchste Menge an Ethanol-Cyclohexan-Extrakten (3,6 %). Der Gehalt an wasserlöslichen Extrakten in den Bohnenernterück- ständen beider Erntezeiten lag nach Kaltwasserextraktion zwischen 14,58 % und 14,7 % und bei Heißwasserextraktion zwischen 14,25 % und 17,5 %.

- Die Analyse der Pentosangehalte ergab Werte von 13% bzw. 16,7 % für Schirmbaum- und Kakaobaumspäne. Damit lagen die Werte über den Werten der Bohnenernterückstände sowie der Fichtenholzspäne. Der Pentosangehalt der Bohnenhülsen (8,2 %) unterschied sich nicht von dem der Fichtenholzspäne (8,5 %), lag aber unterhalb des Wertes der Ernterückstände aus der Trockenzeit (10,2 %).

- Die Lignin-Gehalte nach Klason der Kakaobaum- (39,2 %) und Schirmbaumspäne (37 %) waren erheblich höher als der Lignin-Gehalt der Bohnenernterückstände beider Erntezeiten (19.6 % und 16.8 %) und des Fichtenholzes (29,4 %).

- Die Aschegehalte der Bohnenernterückstände (8,5 - 8,9 %) lagen im Vergleich zum Aschegehalt der untersuchten Holzspäne erheblich höher. Die Kakaobaum- und Schirmbaumspäne verfügen über höhere Aschemengen (2,1 % bzw. 1,85 %) als die Fichtenholzspäne (0,36 %).

Die Entwicklung der Spanplatten in dieser Arbeit gliederte sich in vier Abschnitte. Im ersten Abschnitt wurde die Möglichkeit untersucht, UF-Harz (BASF K340) gebundene Spanplatten aus Schirmbaum- und Kakaobaumspänen herzustellen, um die Formaldehydemissionen zu reduzieren. Zu Vergleichszwecken wurden auch Spanplatten aus Fichtenspänen hergestellt. Darüber hinaus wurden im ersten Abschnitt die Auswirkungen von gröberen Mittelschichtspänen untersucht, um die Biegefestigkeit der Spanplatten zu erhöhen.

Die Ergebnisse zeigen, dass die Spangröße der Mittelschicht eine bestimmte Größe aufweisen müssen, um ausreichend hohe Biegefestigkeiten und E-Module zu gewährleisten. Die Überprüfung der mechanisch-technologischen Eigenschaften zeigte, dass die Herstellung von Spanplatten mit reduzierten Formaldehydemissionen bei Rohdichten von 650 kg/m^3 und 550 kg/m^3 der Norm DIN EN 312: 2010 (Typ 2) entsprechen. Die Herstellung von Spanplatten aus Schirmbaum- und Kakaobaumspänen ist unter Verwendung des formaldehydarmen UF-Harzes BASF K 340 technisch machbar.

Die Möglichkeit, formaldehydfreie Spanplatten auf Basis von Schirmbaum- und Kakaobaumspänen herzustellen, stand im Mittelpunkt des zweiten Abschnitts dieser Dissertation. Um dies zu erreichen, wurden ein Bindemittel auf Isocyanatbasis (PMDI) und ein natürliches Bindemittel auf Basis von tierischem Protein, Blutalbumin, verwendet. Im Technikumsmaßstab wurden Spanplatten in Rohdichten 650 kg/m^3 und 550 kg/m^3 produziert. Die Auswertung der mechanisch-technologischen Eigenschaften ergab, dass die Spanplatten aus Kakaobaumspänen trotz der höheren Querzugfestigkeiten im Vergleich zu Schirmbaumholz, niedrigere Biegefestigkeiten und E-Module aufwiesen. Es wurde ebenfalls festgestellt, dass das Kombinieren der Schirmbaum- und Kakaobaumspäne im Verhältnis von 50:50 (Gew./Gew.) in den Deck-und Mittelschichten der Spanplatten zu höheren Biegefestigkeiten und höheren Elastizitätsmodulen führte, als bei alleiniger Verwendung von Kakaobaumspänen. Ausserdem wurde beobachtet, dass die Formaldehydemissionen der Platten beider Bindemittelsysteme im Vergleich zum Grenzwert der Norm E1 sehr geringfügig waren. Die getesteten Formaldehydwerte der Platten konnten auf den sogenannten natürlichen Formaldehyd des gewachsenen Holzes zurückgeführt werden, da die verwendeten Bindemittelsysteme keinen freien Formaldehyd enthielten. Die Ergebnisse zeigten, dass die Herstellung von formaldehydfreien Spanplatten aus Schirmbaumholz mittels PMDI in einer Rohdichte von 650 kg/m^3 für allgemeine Anwendungen, einschließlich Möbel (DIN EN 312: 2010), in der Praxis möglich ist. Eine Mischung aus Schirmbaum- und Kakaobaumspänen im Verhältnis 50:50 (Gew./Gew.) und PMDI als Bindemittel ergab Spanplatten mit ausreichenden mechanisch-technologischen Eigenschaften, um die Anforderungen der DIN EN 312: 2010 zu erfüllen. Die Querzugfestigkeit aller mit Blutalbumin beleimt Spanplatten lag über dem in der Norm DIN EN 312: 2010 festgelegten Mindestwert. Die Biegefestigkeit (mit Ausnahme der aus einer Mischung beider Späne hergestellten Spanplatten mit einer Rohdichte von 650 kg/m^3) und Elastizitätsmodul erreichten jedoch nicht die erforderlichen Werte.

Der dritte Abschnitt dieser Dissertation konzentrierte sich auf die Untersuchung der Eignung von Bohnenhülsen (Ernterückständen währen der Regenzeit) als Ersatzrohstoff für Holz sowohl in den Mittel- als auch in der Deckschicht von Spanplatten. In drei Versuchsreihen wurden dreischichtige Spanplatten (Rohdichte 650 kg/m^3) aus Schirmbaum-, Kakaobaum- und Industriespänen hergestellt. In der ersten und zweiten Versuchsreihe betrugen die Anteile 100 % Bohnenhülsen in den Deckschichten und 100 % der verschiedenen Holzspäne in der Mittelschicht. Hieraus ergibt sich somit eine 40 %ige Substitution der Holzspäne durch Bohnenhülsen. Die Spanplatten der ersten Versuchsreihe wurden mit PMDI und die zweite Versuchsreihe mit UF-Harz (BASF 340) gebunden. In der dritten Versuchsreihe wurden 20 % (Gew./Gew.) der Deckschicht und 20 % (Gew./Gew.) der Mittelschicht jeder Spanplatte durch Bohnenhülsen ersetzt. Die Spanplatten der dritten Versuchsreihe wurden ebenfalls mit PMDI gebunden (Beleimungsanteil von 4 % auf DS und MS, atro). Der Substitutionsgrad der Holzspäne mit Bohnenhülsen betrug ebenfalls 40 %. Die mechanisch-technologischen Eigenschaften der in dieser Dissertation entwickelten Spanplatten wurde mit den geforderten Mindestwerten für handelsübliche Spanplatten verglichen (Norm DIN EN 312: 2010 für Typ 2 Spanplatten). Die Spanplatten der ersten Versuchsreihe (100 % Bohnenschalen in der Deckschicht und 100 % Holzspäne in der Mittelschicht) konnten die geforderten Mindestwerte für Querzugfestigkeit, Biegefestigkeit und Elastizitätsmodul übertreffen. Darüber hinaus war die Formaldehydemission (getestet nach der Flaschen- und Perforatormethode) der Platten der ersten Reihe ebenfalls sehr gering. Die Spanplatten, die mit UF-Harz (Versuchsreihe 2) gebunden wurden, sowie die Spanplatten, die mit Bohnenhülsen sowohl in der Mittelschicht als auch in der Deckschicht (dritte Versuchsreihe) hergestellt wurden, wiesen keine ausreichenden mechanisch-technologischen Eigenschaften auf, um die Anforderungen von Spanplatten für allgemeine Zwecke Typ 2 zu erfüllen (DIN EN 312: 2010). Insgesamt können Bohnenhülsen, basierend auf den mechanisch-technologischen Eigenschaften der untersuchten Spanplatten als alternatives Deckschichtmaterial für den Einsatz im Innenausbau und in der Möbelherstellung angesehen werden.

Im vierten Abschnitt der experimentellen Arbeit wurde untersucht, ob der Ersatz von 10 %, 15 % und 20 % der Mittelschichtspäne des Schirmbaumholzes durch Späne von Bohnenernterückständen der Trockenzeit möglich ist und die Anforderungen nach DIN EN 312: 2010 erfüllt werden können. Neben diesen Spanplatten wurde eine weitere Variante hergestellt, die ausschließlich aus Bohnenernterückständen bestand. Die Ergebnisse zeigten, dass durch steigenden Einsatz der Mittelschichtspäne durch Bohnenernterückständen (10 %, 15 % und 20 %) die Querzugfestigkeiten der Spanplatten signifikant verringert werden. Dagegen wird die Biegefestigkeit, der E-Modul und die hygroskopischen Eigenschaften jedoch nicht negativ beeinflusst. Wird UF-Harz (BASF 340) als Bindemittel verwendet, ist es bis zu einem Einsatz von 20 % Schirmbaumspänen durch Bohnenernterückstände technisch möglich. Die so hergestellten Spanplatten verfügen über eine ausreichende Querzugfestigkeit und Biegefestigkeit, um die Anforderungen von Spanplatten des Typs 2 gemäß DIN EN 312: 2010 zu erfüllen. Die E- Module der Spanplatten lagen jedoch geringfügig unter dem erforderlichen Mindestwert (1800 N/mm^2) der EN-Norm. Die Eigenschaften der ausschließlich aus Bohnenernterückständen hergestellten Spanplatten entsprachen nicht den Anforderungen dieser Norm. Die Formaldehydemissionen aller Platten (Perforatorwerte) überschritten den E1-Grenzwert von 8,0 mg/100 g atro Platte.

Die Ergebnisse dieser Arbeit zeigen insgesamt, dass Schirmbaum-, Kakaobaumholz und die Bohnenernterückstände sinnvolle und potentielle alternative Rohstoffe zur Herstellung von Spanplatten sind. Diese nachwachsenden Rohstoffe können vollständig oder als Teil zur Produktion von Spanplatten eingesetzt werden. Zukünftig wird somit ermöglicht, bisher für Spanplatten ungenutzte Materialien einzusetzen und vor Ort, beispielsweise in Kamerun, in Produkten zu etablieren.

8.0 Outlook

Within the scope of the investigations carried out in this dissertation, the finding has indicated that umbrella tree wood, cacao tree prunings, and bean crop residues could be valuable renewable resources for the production of particleboards. It has been shown that it is technically feasible to produce particleboards with high enough strengths to be used for general purpose applications including furniture with the chips of umbrella tree wood and cacao tree prunings. The bean crop residues of both seasons could be used in combination with wood materials to achieve the same purpose. However, there are still some gaps in terms of research that need to be filled to have a complete understanding of these raw materials.

The first phase of the study was focused on investigating the physical and chemical characteristics of the raw materials.The availability of the bean crop residues like most annual plants is not always guaranteed throughout the year in a uniform quality. Like straw, they must therefore be stored for a long time in bales with a moisture content of less than 15 % in order to avoid a biological attack. The extent to which the gluing-relevant properties of the bean crop residues, such as pH, buffer capacity and volatile acid content, are influenced by the storage time should be investigated.

The study on the effect of coarser core layer wood chips of umbrella tree wood and cacao tree prunings on the properties of the resulting particleboards indicated that an increase in the coarseness of the core layer chips have a significant effect on the mechanical-technological properties of the boards. The results are indicative of a trend but cannot be generalized to include all fractional compositions of core layer wood chips with higher amounts of coarser materials. Therefore, it would be important to identify which fractional composition of the chips of the various raw materials is optimal for particleboards production.

The technique of substituting wood with annual crop residues such as the bean crop residues used in this study is a novel approach with significant potential for producing particleboards with comparative properties as the boards made exclusively from wood. Nevertheless, the major challenge faced when using annual crop residues in the production process is the presence of the waxy substances on their epidermis which adversely affects bonds formation between the glue and the raw material. Developing methods by which these waxy substances could be removed from the surfaces of the annual crop residues without altering their cellular structures would be helpful in establishing them as effective raw materials in the wood-based panel industry.

The investigation into the possibility of producing particleboards of DIN EN 312:2010 standard by substituting 10 %, 15 %, and 20 % (w/w) of umbrella tree wood chips of the core layer with the chips of bean crop residues of the dry season should be extended to substitution levels of 25 %, 30 %, and so on. Also, experimental particleboards should be manufactured with the bean crop residues of the dry season as the surface layer material. The particleboards involving the bean crop residues of the dry season should also be repeated with isocynate-based glue such as PMDI.

Finally, more work need to be done to optimize some of the production variables such as press time, press temperature and resin load.

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Statutory declaration

I, Travolta Achalle Achale, hereby assure that I have independently and scientifically edited and authored the present dissertation titled "Agricultural residues and the wood of umbrella tree (*Musanga cecropioides*) as raw materials for the development of reduced emission particleboards" and have used no sources other than those specified in the literature. Also, I assure that all statements that were taken literally or analogously from other writings have been identified and the work in the same or similar version was not yet part of any other study or publication.

Travolta Achalle, Achale