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FEASIBILITY OF TANNIN BASED ADHESIVE EXTRACTED FROM *CERIOPS DECANDRA* (GRIFF.) BARK IN WOOD COMPOSITE PRODUCTION

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DECLARATION

I, **Suresh Kumar Nath, Student** ID- 110538, hereby declare that this project thesis is based on my own research work except for quotations and citations, which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at other institutions.

Suresh Kumar Nath

Date: 17.04.2016

Dedicated
To
My Beloved Parents

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ABSTRACT

Ceriops Decandra (Griff.) Ding Hou, locally known as goran is a common mangrove species. The purpose of this study was to assess the technical feasibility of using a tannin based adhesive coming from the bark of goran for the production of particleboard. Tannin was extracted by hot water extraction method for 3 hours at a temperature of 80 ± 5 °C. The extracted tannin was mixed with the commercial grade urea-formaldehyde (UF) with the ratio of 0, 25, 50, 75 and 100% (w/w). The particleboards were produced using wood particles collected from Akij Particle Board Mills Ltd., Manikgonj, Bangladesh, with 8% adhesive (w/w basis), at a temperature of 160 °C with 5 MPa pressure for the period of 8 minutes. The particleboards were analyzed according to different standards for its physical and mechanical properties. Particleboards produced from 100% tannin resulted very good mechanical properties. Average MOR and MOE value for 100% tannin based particleboards were 18.4 and 948.0 MPa, respectively and both these values were the highest among all the treatments. The results thus prove tannin, preferably in pure form to be technically feasible as an adhesive for application in particleboard production.

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1 Introduction

1.1 Background of the study

Wood is bonded in about 70% of its applications or uses, as for like particleboard (PB), fiberboard, oriented strand board (OSB), etc. (Lee, 1991). In most cases petroleum (nonrenewable resource) based synthetic adhesives are being used, which are of high cost and health hazardous (IARC, 2004). Nowadays people are more concern about eco-friendly products. Thus, studies have been carried out to replace these synthetic adhesives with eco-friendly ones produced from natural, renewable resources. According to different studies tannin from wood and bark of some species exhibit good adhesive properties and being used in some woodworking processes even in industrial scale (Pizzi, 1994; Capparucci *et al.*, 2011).

Utilization of bark is one of the utmost pressing problems facing in the wood industries. The quantity of bark available is substantial, since approximately 10-15% of the volume of every log consist of bark (Roffael *et al.*, 2000). Disposal of this enormous quantity of bark creating a problem, particularly with the outcome of more stringent environmental regulations regarding air and water pollution enacted in the recent days (Wang *et al.*, 2015). It has become imperative that the industries no longer consider bark as a waste. Bark is a low cost material with a limited market for diverse applications such as a high-quality mulch or as energy source and even composite products (Blanchet *et al.*, 2000). However, the current uses do not require bark in sufficient quantities to utilize the supply available. Bark and heartwood of limited wood species contain large quantities of water soluble extractives of polyphenolic nature suitable as a starting material for the production of adhesives (Grigoriou, 1997). The extractives are mainly concentrated in the inner layer of the bark. Tannin is the most suitable one to use as an adhesive (Roffael *et al.*, 2000).

The most common commercial condensed tannins are mimosa (*Acacia mearnsii*, or *mollissima*) bark tannin, quebracho (*Schinopsis balansae* and *Schinopsis lorentzii*) wood tannin, pine (*Pinus radiata*) bark tannin and gambier (*Uncaria gambir*) leaf tannin (Carvalho *et al.* 2014). Among the species of the Sundarbans, *Ceriops decandra* (Griff.) Ding Hou, locally known as goran, exhibit high tannin potential (Raju *et al.*, 2008). Traditionally bark decoction of goran is used to arrest hemorrhage, leaves are used to treat skin diseases, leprosy, malaria, etc (Ponglimanont and Thongdeeying, 2005). Roots are used as purgative (Uddin *et* *al.*, 2005). The bark of goran is a rich and cheap source of tannin for fishing community around Sundarbans, extracted tannin is used for treating cotton fishing nets (Raju *et al.*, 2008).

Tannins are phenolic compounds, classified as secondary metabolites, important in plant defense mechanisms against solar radiation, herbivores and pathogenic organisms (Constabel *et al.*, 2014). The concentration of these phenolic compounds in the plant is affected both by plant genetics and by the environmental conditions in which the plant is found (Lattanzio *et al.*, 2008). Condensed tannin–formaldehyde wood adhesives have been used industrially since the 1970s for bonding of interior and exterior wood products such as particleboard and plywood (Pizzi, 1994; Pizzi, 1983). Tannin is the most widely used renewable resource in adhesive production and represents the best substitute for phenol in adhesive preparation (Moubarik *et al.*, 2009).

Adhesives are the most important constituent and play a central role in the production of particleboard (PB). The quality of bonding and hence the properties of the wood-based panels are determined mainly by the type and quality of the adhesive (Dunky and Pizzi, 2002). In the production of PB, the most used adhesive is urea-formaldehyde (UF). Its main application is for production of systems for interior uses. UF is synthetically produced from non-renewable resources, such as petroleum and natural gas (Pizzi and Mittal, 2011). Due to other uses of these non-renewable resources, price of these resources as well as the adhesives is increasing rapidly in the recent days. This creates the need to search for adhesives from natural renewable resources of the same quality as the synthetic adhesives (Jang *et al.*, 2011). Another problem of synthetic adhesives, mainly of the urea-formaldehyde type, which is used in interior environments due to its less resistance to moisture, is formaldehyde emission (Carvalho *et al.*, 2014; Kim, 2009). This causes indoor air quality degradation (IAQ), which can be hazardous to human health since it is a carcinogenic compound (IARC, 2004).

Due to the high price of adhesives of synthetic non-renewable sources, which normally exhibit high formaldehyde emissions, studies that seek for total or partial replacement of synthetic adhesives are of great importance, to produce good quality panels with lower formaldehyde emissions. However, there have been no studies related to the use of tannin based adhesives from goran (*Ceriops decandra*) bark for PB production. Thus this study was designed to extract tannin based adhesive from goran bark and applying the extracted tannin as an adhesive for particleboard production and assessing the technical feasibility of tannin as an alternative to urea-formaldehyde (UF).

1.2 Objectives of the study

Bark of goran (*Ceriops decandra)* from the Sundarbans Mangrove Forest contain a substantial amount (35-40%) of tannin. Bark can be collected from the harvested goran in large amount. In goran average bark volume ranges from 10 to 20%. If the produced PB fulfill the requirements of commercial grade boards, than this could be a great opportunity to produce goran bark tannin based adhesive as an alternative to environmentally harmful urea-formaldehyde (UF) for PB production. It will reduce volatile organic compounds (VOCs) emission from PB industries and households where PB is used. Thus, the specific objectives of this study are:

- To prepare tannin based adhesive from the bark of goran (*Ceriops decandra*) by the hot water extraction method.
- To assess the technical feasibility of using a tannin-based adhesive coming from the bark of goran for the production of particleboard (PB).

2 Literature review

2.1 General information about particleboard

A particleboard is a board (or sheet) constituted from fragments of wood and/or other lingo-cellulosic materials (chips, shavings, flakes, splinters, sawdust, etc.), bonded with organic binders with the help of one or more agents like heat, pressure, humidity, catalyst, etc. (Shrivastava, 1997). It may be classified as a panel product manufactured under pressure and heat from particles of wood or other lingo-cellulosic materials bonded entirely with a binder, generally a synthetic resin, to which other chemicals (e.g., fire retardant, fungicide, water retardant etc.) may be added to improve certain properties (Salehuddin, 1992).

2.1.1 Brief history and development of particleboard

Particleboards are not more than a few decades old production. Before particleboard, modern plywood, as an alternative to natural wood, was invented in the 19th century, but by the end of the 1940s there was not enough lumber around to manufacture plywood affordably. By that time particleboard was intended to be a replacement (Sheng, 2004). But before that scarcity in raw materials of plywood, first efforts were made in the early 1920's for manufacturing of particleboard. But it was unsuccessful as for the lack of suitable adhesives. Then new techniques introduced in the 1930's in resin applications with the growing demand paved the way for the industrial production of particleboard in the early 1940's (Moslemi, 1985). The first commercial piece was produced during World War II at a factory in Bremen, Germany. It used waste material such as planer shavings, off-cuts or sawdust, hammer-milled into chips, and bound together with a phenolic resin. Today's particleboard manufacturer provides high-quality products that consumers require due to up gradation of manufacturing techniques (Moslemi, 1985; Sheng, 2004).

2.1.2 Types of particleboard

There are different types of particleboards depending on:

2.1.2.1 Types of particles used

Flakeboard: A particleboard in which the wood is largely in the form of flakes, giving the surface a characteristic appearance (Shrivastava, 1997). A small wood particle of predetermined dimensions specifically produced as a primary function of specialized equipment of various types, with the cutting action across the direction of the grain (either radially, tangentially, or at an angle between), the action being such as to produce a particle of uniform thickness, essentially flat, and having the fiber direction essentially in the plane of the flakes, in overall character resembling a small piece of veneer (Shrivastava, 1997).

Chipboard: A particleboard made from chips. It is made in varying thickness and may be surfaced with paper, veneers, plastic materials, etc. (Shrivastava, 1997). Gluing together wood particles with an adhesive, under heat and pressure makes chipboard. This creates a rigid board with a relatively smooth surface. Chipboard is available in a number of densities: -normal, medium and high-density. It is often used for kitchen tops (which are laminated with melamine) and fire doors. Medium density is somewhere between normal and high density. There are exterior grades of chipboard available but most are only suitable for internal use. Chipboard with a veneered surface is widely used for flat-pack furniture and work surfaces. High-density chipboard is often used as the carcass for kitchen units and worktops and flooring. This type of chipboard is hardwearing, rigid and heavy (Salehuddin, 1992).

Shavings board: A particleboard in which wood shavings are the chief constituents. (Shrivastava, 1997). Shavings are produced from various kinds and sizes of trees being converted to lumber of different dimensions. Often instigated by the need for reducing costs of disposal of materials that clog production, or by the desire to get some return from material that in the log form has represented a considerable outlay of money. Shavings ordinarily come from air-dried or kiln-dried wood. Shaving produced from machining dry wood of a single species afford their producer the best prospects for marketing waste material. Uniform particle sizes (achieved by screening) are needed for some uses. For most uses only fresh material is acceptable. Shavings, when exposed to the weather, deteriorate very rapidly and lose much of their value (Salehuddin, 1992).

Waferboard: It is a structural material made from rectangular wood flakes of controlled length and thickness bonded together with waterproof phenolic resin under extreme heat and pressure (Salehuddin, 1992). Waferboard is a widely used, versatile structural wood panel. Manufactured from waterproof heat-cured adhesives and rectangular shaped wood strands that are arranged in cross-oriented layers, Waferboard is an engineered wood panel that shares many of the strength and performance characteristics of plywood. Waferboard's combination of wood and adhesives creates a strong, dimensionally stable panel that resists deflection, delamination, and warping; likewise, panels resist racking and shape distortion when subjected to demanding wind and seismic conditions. Relative to their strength, waferboard panels are light in weight and easy to handle and install (Shrivastava, 1997).

Oriented trand board: Oriented strand board, or OSB, or Sterling board (UK) or Smart Ply (UK and Ireland) is an engineered wood product formed by layering strands (flakes) of wood in specific orientations (Salehuddin, 1992). Oriented strand board is manufactured in wide mats from cross-oriented layers of thin, rectangular wooden strips compressed and bonded together with wax and synthetic resin adhesives. The resin types typically used include phenol formaldehyde (PF), melamine fortified urea-formaldehyde (MUF) or isocyanate, all of which are moisture resistant binders. In Europe, it is common to use a combination of binders, typically PMDI would be used in the core and MUF in the face layers and this has the advantage of reducing press cycles whilst imparting a bright appearance to the surface of the panel (Shrivastava, 1997).

2.1.2.2 Particle size distribution in the thickness of board

Single layer or homogeneous board: Single-layer particleboards are made from pressing together wood particles of similar sizes to form a flat, dense board. This type of particleboard is suitable as a base for plastic lamination and veneering, but not for painting. Single-layer particleboards are used commonly for interior applications. They have some water-resistance capabilities, but are not fully waterproof (Shrivastava, 1997).

Three layer board: A three-layer particleboard is made from sandwiching a layer of larger wood particles between two layers of high-density, finer wood particles. The outer layers have a higher amount of resin adhesive than the inner layer. Three-layer particleboards have smooth outer layers that are suitable for painting. These boards are not as dense as single-layer boards and tend to split easily (Shrivastava, 1997).

Graded-density boards: Graded-density particleboards are similar to three-layer particleboards. They have an inner core of coarse wood particles sandwiched between two outer layers of finer particles. However, unlike a three-layer particleboard, the transition between the coarse surfaces to the finer ones is gradual. Graded-density particleboards are used in cabinet construction and for furniture components (Salehuddin, 1992).

2.1.3 Raw materials for particleboard manufacturing

2.1.3.1 Ligno-cellulosic materials

Woody materials, planer savings, sawmill residues, such as slabs, edging, trimmings, etc. Residues from timber cutting in furniture and cabinet manufacturing plants, residues from match factories, veneer and plywood plant residues, saw dusts, Logging residues, such as short logs, broken logs, crooked logs, small tree tops and branches, forest thinning , etc. and bark (Salehuddin, 1992).

2.1.3.2 Non-woody materials

Jute sticks, bagasse, bamboo, flax shaves, cotton stalks, cereal straw, almost any agricultural residue (such as husks, coconut coir etc.) after suitable treatment (Youngquist, 1999).

2.1.3.3 Chemicals

Binder or adhesive

Adhesives or binders are the materials used in the fabrication of timber structures and components offers a neat and efficient method of bonding together the separate pieces of wood, or of board products such as plywood, chipboard, or fiberboard, which comprise the finished product. The bond attained must meet the strength requirements for the structure as a whole and this bond must remain unaffected by the condition to which it will be exposed throughout its life (Youngquist, 1999).

There are mainly two types of adhesive. One originated from natural sources known as natural adhesive and another is synthetic adhesive.

Natural adhesive

Adhesives of natural origin- such as animal, casein, soybean, starch and blood glues are still being used to bond wood in some plants and shops, but are being replaced more and more by synthetics (Vick, 1999).

Synthetic adhesive or Synthetic resin adhesive

Synthetic adhesives are man-made polymers which resemble natural resins in physical characteristics but which can be tailored to meet specific woodworking requirements. Synthetic adhesives can be categorized into two groups, namely thermosetting adhesives and thermoplastic adhesives (Dunky and Pizzi, 2002).

Other additives

Additives are mainly used to improve the certain properties of particleboard such as moisture repellency, fire retardancy etc.

Paraffin wax or wax emulsion: Wax is added as a water repellent in the production of wood-based manufactured composite boards such as particleboard, medium density, oriented strand and other board products.

Fire retardants: The most common and best known fire retardance methods for wood are based on changing the pathway of pyrolysis. In this simple and inexpensive method, wood is treated with a substance that enhances the pyrolysis reaction of cellulose through the pathway leading mainly to char formation. A fire retardant may also slow down pyrolysis reactions and stabilize the chemical structures of wood against decomposition. For instance, aluminium sulphate added to wood creates bonds between cellulose molecules in increased temperatures, thus preventing thermal decomposition (Dunky and Pizzi, 2002).

Curing agents and hardeners: Deliver a complex combination of performance attributes that can vary from high bond strength to good insulating properties depending on end-use application. Provide unique benefits to adhesives such as fast and low temperature cure, excellent thermal shock resistance, and low viscosity for solvent-free systems, hydrophobicity, and good overall chemical and mechanical properties (Lee, 1991).

2.2 Tannin as a natural adhesive

2.2.1 Tannin

The tannin compounds are widely distributed in many species of plants, where they play a role in protection from predation, and perhaps also as pesticides, and in plant growth regulation. They are commonly found in both gymnosperms as well as angiosperms.

There are three major classes of tannin (Fig. 1).

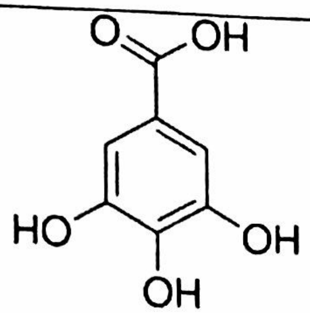
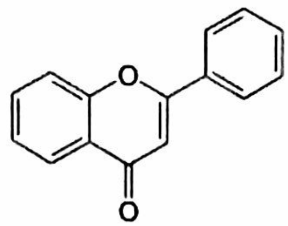
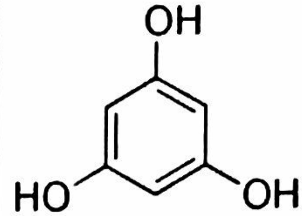
Base unit			
	Gallic acid	Flavone	Phloroglucinol
Class/Polymer	Hydrolyzable tannins	Non-Hydrolyzable or condensed tannins	Phlorotannins
Sorces	Plants	Plants	Brown algae

Fig. 1. Monomers of the tannin and types of tannin based on the monomers (Mole, 1993)

2.2.2 Source of tannin

Tannins are found in leaf, bud, seed, root, and stem tissues. An example of the location of the tannins in stem tissue is that they are often found in the growth areas of trees, such as the secondary phloem and xylem and the layer between the cortex and epidermis. In all vascular plants studied so far, tannins are manufactured by a chloroplast-derived organelle, the tannosome. Tannins are mainly physically located in the vacuoles or surface wax of plants. These storage sites keep tannins active against plant predators, but also keep some tannins from affecting plant metabolism while the plant tissue is alive; it is only after cell breakdown and death that the tannins are active in metabolic effects (Carvalho, 2014).

2.2.3 Goran as a source of tannin

Goran is a mangrove species. A shrub or small tree, upto 4 m high, erect branches formed its canopy conical, leaf-scar marks present on the branches. Aerial roots forming shallow buttress or root knees in the trunk base, leaves simple, opposite, obovate, aduceus, rounded apex, dark green, glabrous. Tannin percentage in the stem bark is 19%, which is largely used by the fishermen for tanning their fishing nets (NIO Bioinformatics Centre, 2016). Goran is valued for its reddish brown colored bark which is known for its high tannin content ranging from 68 to 75% (Raju *et al.*, 2008). The dyeing with this tannin gives brown color, and the fishermen use it to protect cotton fishing nets from decay or extend its life for a longer period (Raju *et al.*, 2008).

2.2.4 Current use of tannin as an adhesive

Lately, there has been considerable industrial interest in the development of natural, or green, wood adhesives to substitute for synthetic thermosetting resins as adhesives for wood panels because of an undue increase in the cost of oil and mounting environmental pressure worldwide (Lei *et al.*, 2008). Amongst the possible alternatives, tannin is an excellent renewable resource which can be used for replacing petroleum-derived phenolic compounds (Kim, 2009). Condensed tannin–formaldehyde wood adhesives have been used industrially since the 1970s (Pizzi 1983) for bonding of interior and exterior wood products such as particleboard and plywood (Pizzi 1994). According to Moubarik *et al.* (2009), tannin is the most widely used renewable resource in adhesive production and represents the best substitute for phenol in adhesive preparation.

2.2.5 Advantages of tannin based adhesive

The natural tannin adhesives are used to replace UF resin in the formaldehyde-based resin system in order to reduce formaldehyde and volatile organic compound (VOC) emissions from the synthetic formaldehyde based adhesives. Formaldehyde is produced worldwide on a large scale. It is used mainly in the production of resins that are used as adhesives and binders for wood products, pulp, paper etc. International Agency for Research on Cancer (IARC), part of the World Health Organization, concluded that formaldehyde is *carcinogenic to humans* (IARC, 2004). So people are trying to replace it with a natural environmentally friendly ones. Amongst the possible alternatives, tannin is an excellent renewable resource which can be used for replacing petroleum-derived phenolic compounds (Kim, 2009).

2.3 Description of goran tree

2.3.1 General description

Goran is a straight columnar tree, usually small to medium-sized. However under favorable conditions it attains a height of 35 m and trunk diameter of 35 cm, with a relatively narrow crown and short basal buttresses which appear to develop from the fusion of stilt roots clusters. The roots are superficial, radially spread, with small knobby and/or pneumatophores loop in wet sites. The bark is whitish or pale grey, and smooth but slightly fissured towards the base. It peels around the buttresses. The branches are conspicuously jointed with swollen nodes. The leaves are oppositely arranged, clustered at the end of the twigs, coriaceous. Obovate to elliptic-oblong, measuring 4.5-10 cmx 2.5-6 cm, wedge-shaped at the base, rounded or subemarginate at the apex, hairless and glossy. The petiole is 1-2.5 cm long, 'with lance-shaped deciduous stipules at the base measuring 1.5-2.5cm long (NIO Bioinformatics Centre, 2016; Grierson, 1991). Pictures of goran tree parts is shown if Fig. 2.

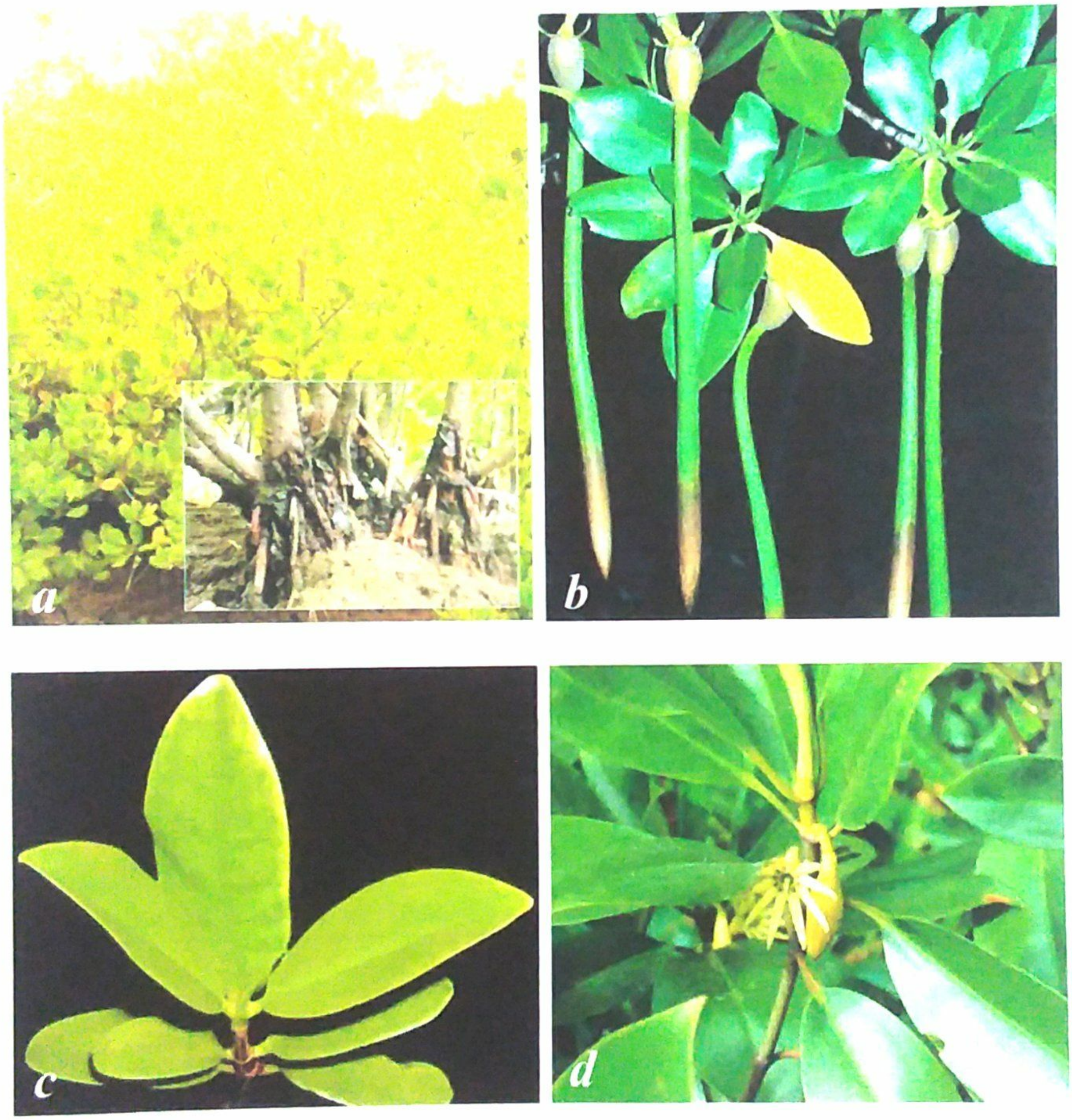


Fig. 2. Goran (a) stem, (b) fruits, (c) leaves and (d) flowers

The flowers are in head-like, condensed, up to 5-flowered cymes in leaf axils at the upper part of a branch. They are 5-6-merous, measure 5-6 mm long and with deeply lobed sepal. The white petals are about 2.5 mm long and divided fringe-like at the apex. The stamens are twice the number of sepal lobes. The anthers are longer than the filaments. The ovary is semi-inferior and 3-celled. The fruit is an ovoid-conical berry, measures 1-1.8 cm long, with persistent erector ascending sepal lobes, blunt basally and warty at the apex. The seeds are viviparous. The hypocotyl is club-shaped, protruding below the fruit measuring 9-15 cm long, occasionally longer (e.g. in New Guinea) and slightly fluted (Globinmed, 2016; Grierson, 1991).

2.3.2 Geographical distribution

The genus *Ceriops*, was once more widely distributed than it is today. For example, it was probably present in Europe in the Eocene: both *Ceriops* and some other *Rhizophoraceous* genera appear in the European fossil record before they appear in that of Southeast Asia. Clearly, the geographical ranges of the species have changed. Thus, although *C. decandra* is now centered in Southeast Asia, it is not certain that it originated in this region. Its current range extends from the Indus delta in Pakistan around the coast of India and across the Bay of Bengal to Burma and thence through Indo-China, Thailand and Southeast Asia to Papua New Guinea. It also occurs locally in north-eastern Australia. In Southeast Asia it is found in Peninsular Malaysia, the Philippines, Borneo, Java, Sulawesi, the Lesser Sunda Islands, the Moluccas, and New Guinea. It has not yet been collected from Sumatra, but has been recently reported there (Globinmed, 2016; Grierson, 1991). According to Duke *et al.* (2010) the range of goran is restricted to the east coast of India and Bangladesh, southwestern Thailand and western part of the Malay Peninsula and native to Bangladesh; India; Malaysia; Myanmar and Thailand. Global distribution of goran is shown in Fig. 3.

Fig. 3. Geographical distribution of goran (Duke et al., 2010)

3 Materials and methods

3.1 Goran bark collection and preparation

Goran bark were collected from Burigoalini forest range of Sundarban West Forest Division. The average age of the trees was between 7-8 years. Bark was collected from the butt portion of the trees, preferably below breast height (1.3 meter). After the collection, bark were chopped to get smaller size. The chopped bark were mixed up and dried in air under the shade. After drying, the bark were grinded and passed through a sieve having the openings of 2 mm^2. Pictures of bark before and after oven drying is shown in Fig. 4.

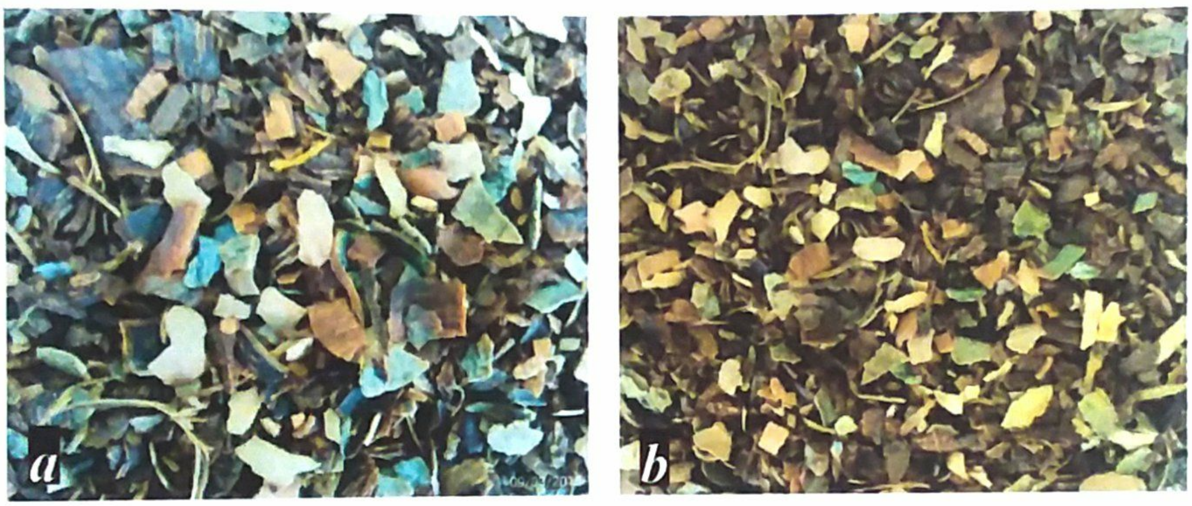


Fig. 4. The bark of goran (a) green) and (b) dried

3.2 Tannin extraction and adhesive preparation

Extraction of tannin was done at pathology lab of FWT discipline of Khulna University, following the hot water extraction method. The grinded bark was soaked in distilled water at a ratio of 15:1 (v/w). The water-bark mixture was then heated at a temperature of 80 ± 5 °C for 3 hours. The mixture was cooled down and filtered using a sieve having 0.1 mm^2 screen openings. The tannin solution was then evaporated until the solid content goes to around 50%. It was then stored in refrigerator at a temperature of -10 °C for later applications.

3.3 Particle collection

Wood particles were collected from Akij Particle Board Mills Ltd., which is situated at Torraghat in Manikgonj District, Bangladesh. The particles were of standard size used in the industry. There were two different sized particles coarse for middle layer of the board and fine for different faces. The coarse particles had the average dimension of 2 × 12 × 0.60 mm^3

(width × length × thickness) having the average slenderness ratio of 6.18. The fine particles were almost powdery.

3.4 Chemicals

Needed urea-formaldehyde (UF) resin was also collected from Akij Particle Board Mills Ltd., Manikgonj, Bangladesh. Solid content of the adhesive was 49.2%. Viscosity was 221 cP. The adhesive was stored in a cool place in an air tight bottle.

3.5 Preparation of particles and adhesive

Before manufacturing of board the particles were dried in an oven for 24 hours with the temperature of 103 ± 2 °C. Laboratory scale blender was used to mix the adhesive and particles. Adhesives composed of different percentage of UF and tannin were prepared separately just before application.

3.1 Treatments used for the manufacture of the particleboards

Different percentages of UF and tannin adhesive were used to produce the boards. The percentage of UF and tannin adhesive were applied according to Table 1.

Table 1. Adhesive treatments used for the manufacture of the boards

Treatments	Tannin (%)	Urea-formaldehyde (%)
T-0	0	100
T-25	25	75
T-50	50	50
T-75	75	25
T-100	100	0

3.6 Particleboard manufacturing

The particleboards were manufactured at the laboratory following the uniform procedures that replicated the industrial production. The wood particles and adhesives (UF and/or Tannin) with a ratio of 92:8 were mixed in a rotary drum blender for 6 minutes for producing homogenous mixture of particles and adhesive. Coarse and fine particles were mixed separately. The mixture was weighed and placed in aluminium caul plate using a forming box to form standardized mat. During heating and pressing the mat, aluminium foil was used to avoid the direct contact between adhesive and metal plates. Pressing was completed in three phases, i.e., at first, manual pressing to reduce the mat height, then shifting into the electrically heated hot press for pressing in high temperature and finally cold pressing phase for final setting of resin or adhesive. Pressing temperature, pressure and time for hot and cold pressing were set according to the Ayrilmis *et al.* (2010). Maximum pressing temperature in the hot press was 160 °C having the pressure of 5 MPa, and pressing time was 8 minutes. After that heating of the machine turned off for cooling down of the boards for 20 minutes. At least three replication of each treatment were performed to produce boards having dimension of 30 × 25 × .7 cm approximately. The manufactured boards were trimmed and put into a conditioning room for 24 hours before testing of properties.

3.7 Determination of physical properties

Samples having the size of 50 × 30 × 5 mm were cut according to the EN 323 (BSI, 1993) standard from the boards it was then oven dried and then weighted. The samples were consequently immersed in distilled water at 25 °C for 2 and 24 hours, respectively with a change of water. Green weight and dimensions of the samples were measured at the beginning of the test. Determination of water uptake (%) of each sample was expressed as weight absorbed divided by the dry weight of sample. Dimensional swelling (%) of each sample was calculated based on the dry weight. Density of the specimens was evaluated according to the test method and requirements of EN 323 (BSI, 1993). At least three replicates were used for each treatment.

3.8 Determination of mechanical properties

The laboratory test for characterization of mechanical properties was carried out using the Unconfined Compression Test Apparatus (SIMSUCA-02) according to the ASTM D 1037-93 (ASTM, 1995) standard in the laboratory of Civil Engineering Department of Khulna University of Engineering and Technology, Khulna, Bangladesh. Samples were cut according to ASTM D 1037-93 (ASTM, 1995) standard, with the dimension of 220 × 180 × 7 mm.

3.8.1 Modulus of rupture (MOR)

The MOR was calculated by Equation 1.

$$MOR = \frac{3PL}{2bd^3} \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots Eq. 1$$

Where, MOR = the modulus of rupture (N/mm^2), P = load (N), L = span length (mm), b = width of test sample (mm), d = Thickness of test sample (mm).

3.8.2 Modulus of elasticity (MOE)

The MOE was calculated by Equation 2.

$$MOE = \frac{P'L^3}{4\Delta bd^3} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots Eq.2$$

Where, MOE = the modulus of elasticity in (N/mm^2), P' = load (N) at the limit of proportionality, L = the span length (mm), Δ = the deflection (mm) at the limit of proportionality, b = the width of sample (mm), d = the thickness/depth of sample (mm).

3.9 Analysis of data

All the data, obtained during the laboratory tests for characterization of physical and mechanical properties of each type of particleboards were analyzed by using Microsoft Office Excel 2013 (USA) and Minitab 17 (USA). ANOVA (Analysis of Variance) and LSD (Least Significant Difference) were done to analyze the data ($\alpha \leq 0.05$).

4 Results and discussion

4.1 Properties of the adhesives

The data for viscosity and solids content of the tannin, commercial grade urea-formaldehyde (UF) and mixed adhesives are shown in Table 2. In this study, the tannins were hydrated aiming at an adhesive with a solids content of 50%, a percentage which was successfully achieved for the tannin adhesive (Table 2). In general, both the mixtures and the pure UF and Tannin adhesives showed values near 50%, this being an adequate range for production of particleboard.

Table 2. Properties of the adhesives having different tannin percentage

Tannin (%)	Viscosity (cP)	Solids content (%)	pH
0	221	49.2	9.0
25	272	54.4	6.4
50	349	52.3	5.1
75	398	56.8	4.8
100	512	57.3	4.2

The viscosity of the tannin based adhesive was high in comparison to the urea-formaldehyde adhesive (UF). Addition of the tannin adhesive to the UF adhesive caused an increase in viscosity. However, at standards acceptable for application by spraying in the production of particleboard, which is at most 1,000 cP (Iwakiri, 2005). Addition of tannin also decreased the pH of adhesives gradually, with the increased percentage of tannin in the adhesive. Decreased pH affect the internal bonding of particleboard negatively, lower adhesive pH hamper the curing of adhesive and thus results lower mechanical properties but after a certain limit it sometimes promote the curing (Carvalho *et al.*, 2014).

4.2 Visual features of particleboards

Higher percentage of tannin resulted in darker color boards. This caused because of the darker color of tannin and may be because of lesser penetration of tannin having higher viscosity. Pictures of particleboard of different treatments are shown in Fig. 5.

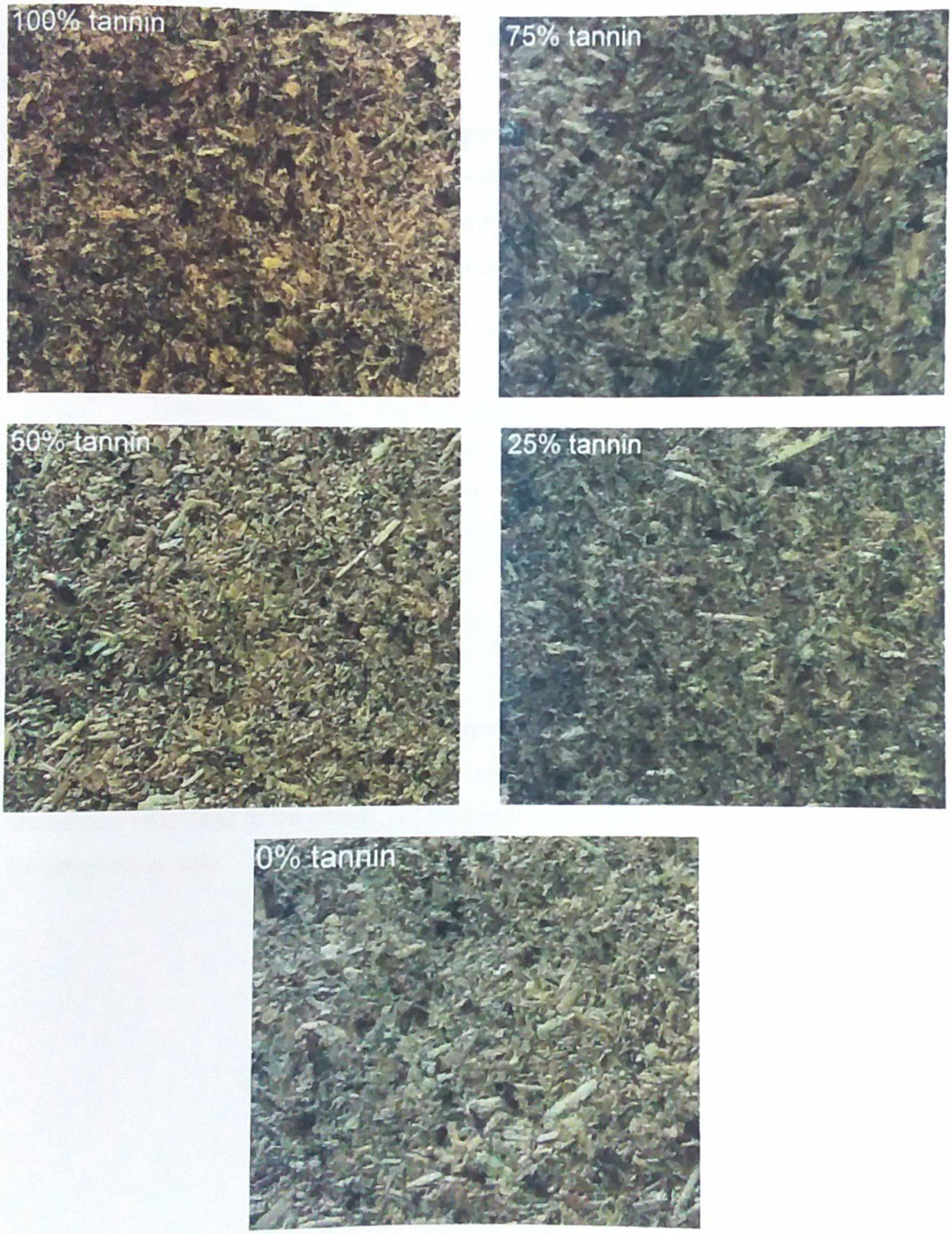


Fig. 5. Particleboards produced by different adhesive treatments

4.3 Physical properties of the particleboards

4.3.1 Density and compaction ratio

Mean values of composite density and compaction ratio is shown in Table 3. From the result it was observed that there was no statistical difference among the densities obtained from different treatments (Appendix II). The basic density obtained for collected particles was 0.60 g/cm^3.

Density of boards produced by 100% tannin resulted the highest. This occurred may be because of higher solid content and higher compaction of the particles. Density decreased with the decrease of the percentage of tannin content. Higher percentage of tannin adhesive resulted higher compaction ratio and solid content was also higher in tannin.

Table 3. Density and compaction ratio of particleboards produced by different adhesive treatments

Treatments	Density (g/cm^3)	Compaction ratio
T-0	0.76	1.27
T-25	0.76	1.27
T-50	0.77	1.29
T-75	0.79	1.32
T-100	0.79	1.32

According to Maloney (1993), the ideal compaction ratio of particleboard is from 1.3 to 1.6 which leads to adequate compaction of the particles and consequently good physical and mechanical properties of the boards. All the treatments were within the cited adequate range for compaction ratio.

4.3.2 Moisture content (MC)

Variation of the mean values of moisture content percentage is shown in Fig. 6. It was observed that there was no statistical difference among the density obtained for the different treatments (Appendix II).

Among five treatments, 50% tannin resulted in highest MC% value. 100% tannin treatment resulted almost similar MC%. Higher tannin content resulted in higher moisture content. This occurred may be because of water absorbing nature of tannin.

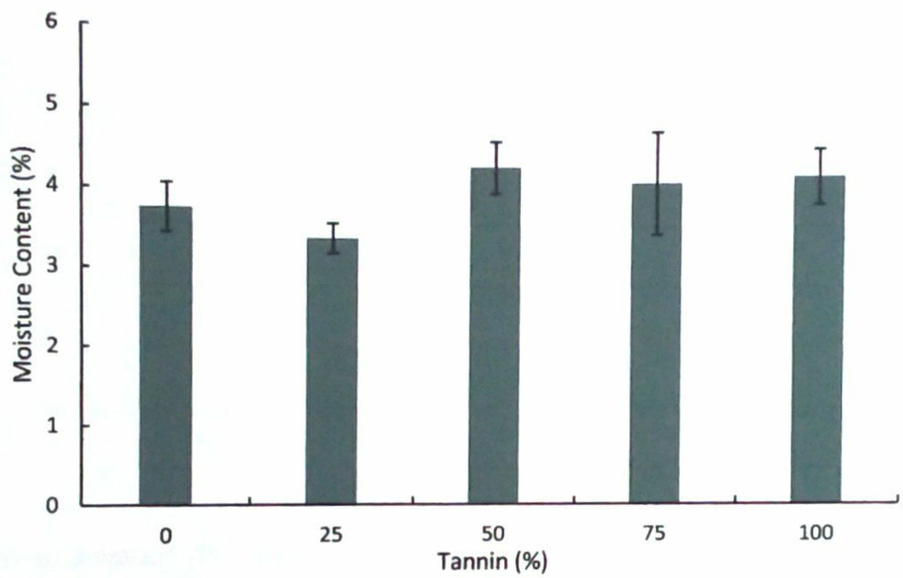


Fig. 6. Moisture content (%) of the boards produced by different adhesive treatments

Espert *et al.* (2003) stated that wood consists mostly of vessels in which moisture is absorbed. Tannin is also water soluble. Different percentage of tannin and urea-formaldehyde and the different pH values may be the cause of variation in moisture content (%).

According to ANSI (1999) standard, the mean moisture content of particleboard shall not exceed 10% (based on the oven dried weight of the board). All the treatments are within the cited adequate range for moisture content (%).

4.3.3 Water absorption

Variation of the mean values of water absorption (%) after 2 hours of immersion (WA_2h) and after 24 hours of immersion (WA_24h) is shown in Fig. 7. The greatest mean value was obtained from the board produced with 100% tannin and gradually decreased with the decreased percentage of tannin. This fact leads to conclude that the combination of two types of adhesive promoted a certain interaction which directly affects this property.

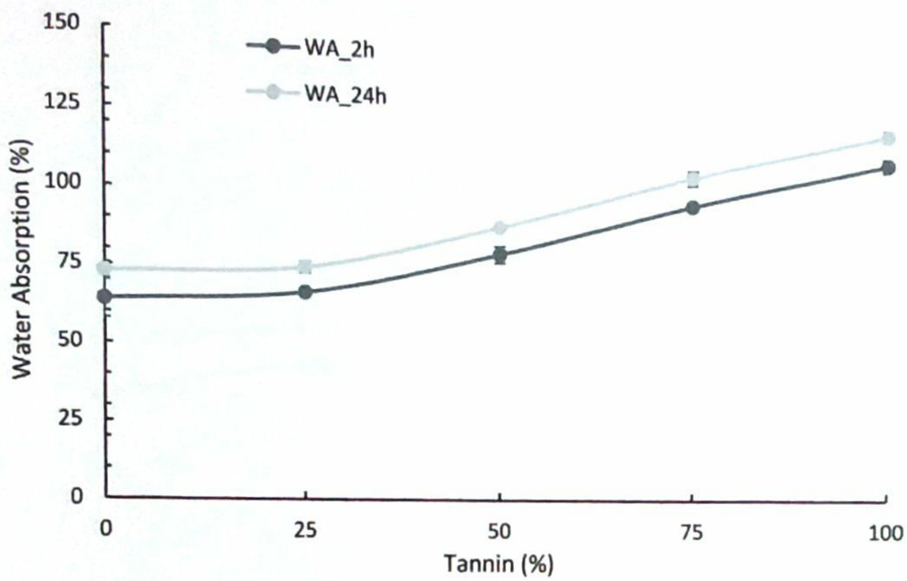


Fig. 7. Water absorption (%) of the boards after 2 and 24 hours of immersion in water.

Carvalho *et al.* (2014) produced OSB panels with *Pinus oocarpa* wood with 8% adhesive. Urea-formaldehyde and tannin from *Stryphnodendron adstringens* in different ratio were used as adhesive. The authors found WA_2 values of 77, 95, 103, 96 and 80% and WA_24 values of 86, 131, 113, 100 and 89% for 0, 25, 50, 75 and 100% tannin content in the adhesive, respectively.

Vital *et al.* (2004) produced panels from flakes, with 8% tannin from *Eucalyptus pellita*, obtained mean values of 88 and 108.9 % for WA_2h and WA_24h, respectively.

The mean values of water absorption for higher percentage of tannin obtained in this study proved to be higher than the studies cited. One of the factors that affected the property could be the pH of the adhesives (Table 2), where the mixture of the tannin adhesive led to a significant reduction of the pH of the UF adhesive, which led to incomplete curing of UF, and thus facilitated water absorption by the wood particles. Other factor could be the higher moisture absorbing property of goran tannin, which increased the amount of moisture absorption.

4.3.4 Thickness swelling

Variation of the mean values of thickness swelling (%) after 2 hours of immersion (TS_2h) and after 24 hours of immersion (TS_24h) is shown in Fig. 8. The greatest mean value was obtained from the board produced with 100% tannin. This fact leads to conclude that the combination of two types of adhesive promoted a certain interaction which directly affects this property.

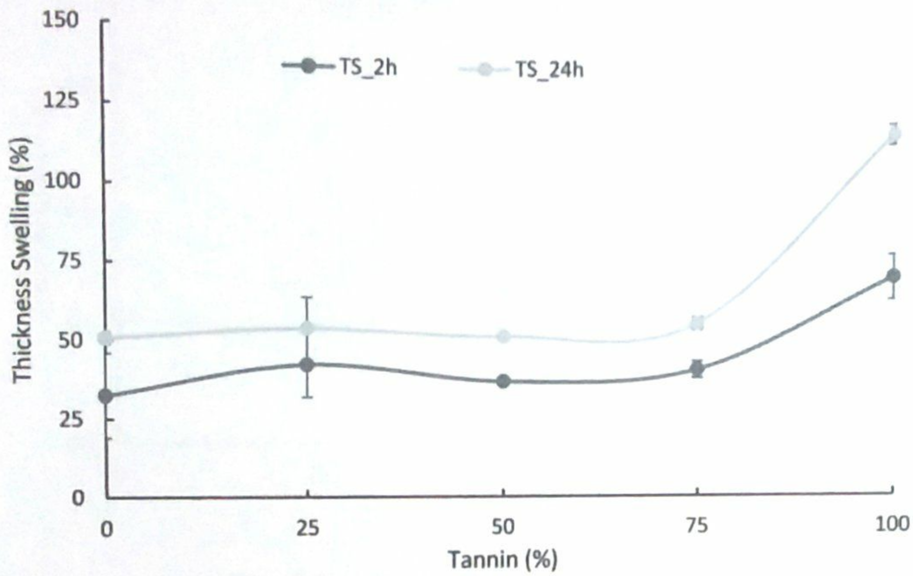


Fig. 8. Thickness swelling (%) of the boards after 2 and 24 hours of immersion in water.

Carvalho *et al.* (2014) produced OSB panels with *Pinus oocarpa* wood with 8% adhesive. Urea-formaldehyde and tannin from *Stryphnodendron adstringens* in different ratio were used as adhesive and found TS_2 values of 14, 22 and 18% and WA_24 values of 25, 25 and 24% for 25, 50 and 75% tannin content in the adhesive, respectively.

Salari *et al.* (2013) upon evaluating the quality of OSB panels produced with 10 % urea–formaldehyde adhesive and with apparent density of 0.70 g /cm^{-3}, obtained mean values of 43.2 and 93.5 % for TS_2h and TS_24h, respectively.

The mean values of thickness swelling for higher percentage of tannin obtained in this study proved to be higher than the studies cited. One of the factors that affected the property could be the pH of the adhesives (Table 2), where the mixture of the tannin adhesive led to a reduction of the pH of the UF adhesive, which led to incomplete curing of UF, and thus weakened the internal bonding of the boards and facilitated water penetration absorption. Other factor could be the higher moisture absorbing property of goran tannin, which increased the amount of moisture absorption.

4.3.5 Linear expansion

Variation of the mean values of linear expansion (%) after 2 hours of immersion (LE_2h) and after 24 hours of immersion (LE_24h) is shown in Fig. 9. Found similar pattern as thickness swelling. The greatest mean values were obtained from the boards produced with higher percentage of tannin as adhesive.

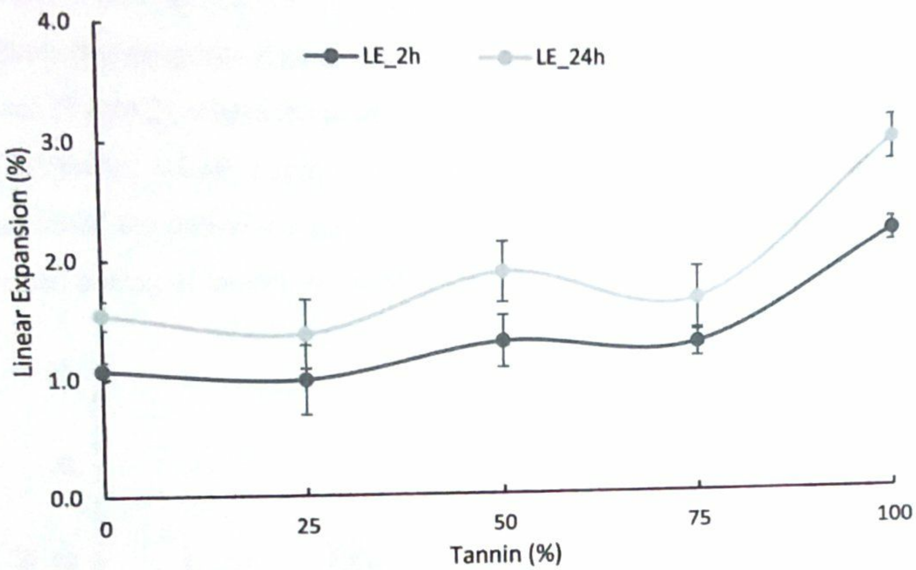


Fig. 9. Linear expansion (%) of the boards after 2 and 24 hours of immersion in water.

Salari *et al.* (2013) upon evaluating the quality of OSB panels produced with 10 % urea–formaldehyde adhesive and with apparent density of 0.70 g /cm^{-3}, obtained mean values of 1.2 and 1.8 % for LE_2h and LE_24h, respectively.

The greater values of linear expansion in case of higher percentage of tannin in the adhesives might be associated with lower internal bonding of these treatments. This may be associated with the curing reaction caused by the mixture of the adhesive, creating a situation in which the low pH of the tannin (Table 2) led to the incomplete curing of UF, thus promoting lower penetration of the adhesive in the anatomical structure of the particles and, consequently, a lower resistance of the internal bond. And in case of 100% tannin highest value might be obtained because of higher water absorbing property of goran tannin.

4.4 Mechanical properties

4.4.1 Modulus of rupture (MOR)

Variation of the mean values of MOR is shown in Fig. 10. The lowest mean values were obtained by the boards produced with the mixture of adhesives, which is certainly associated with lower resistance of the bond of the particles of these treatments. This fact leads to conclude that the combination of two types of adhesive promoted a certain interaction which directly affects this property. One of the factors that affected the reaction could be the pH of the adhesives (Table 2), where the mixture of the tannin adhesive led to a reduction of the pH of the UF adhesive, which might lead to incomplete curing of UF. The later accelerated polymerization of the adhesives, also might be occurred due to tannin, once in reduced pH, the accelerated curing of tannin occurred.

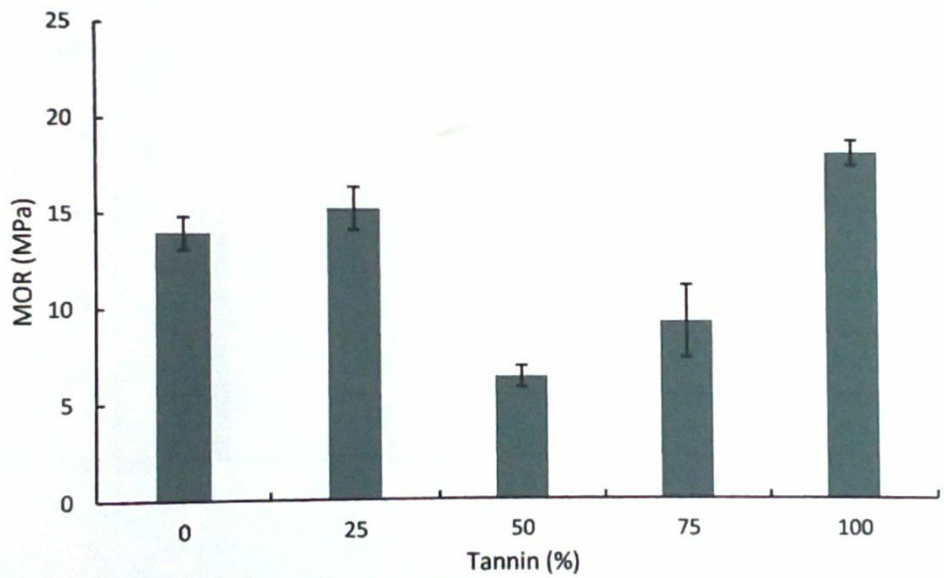


Fig. 10. Average MOR values of the boards produced by different adhesive treatments.

Carvalho *et al.* (2014) produced OSB panels with *Pinus oocarpa* wood with 8% adhesive. Urea-formaldehyde and tannin from *Stryphnodendron adstringens* in different ratio were used as adhesive. The authors found MOR values of 27.4, 24.4, 12.3, 11.5 and 20.8 MPa for 0, 25, 50, 75 and 100% tannin content in the adhesive, respectively. Variation is almost similar with this study.

4.4.2 Modulus of elasticity (MOE)

Variation of the mean values of MOE is shown in Fig. 11. Found similar pattern as the results for MOR values. The lowest mean values were obtained from the boards produced with the mixture of adhesives, which could associated with lower resistance of the bond of the particles of these treatments. One of the factors that affected the reaction could be the pH of the adhesives (Table 2), where the mixture of the tannin adhesive led to a reduction of the pH of the UF adhesive, which might lead to incomplete curing of the adhesives. The later accelerated polymerization of the adhesives, also might be occurred due to tannin, once in reduced pH, the accelerated curing of tannin occurred.

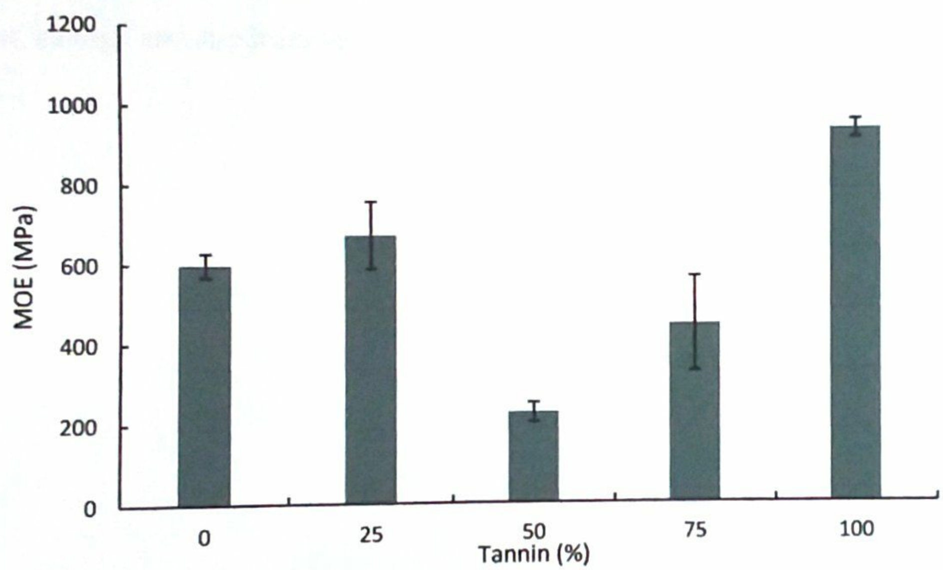


Fig. 11. Average MOE values of the boards produced by different adhesive treatments.

Carvalho *et al.* (2014) produced OSB panels with *Pinus oocarpa* wood with 8% adhesive. Urea-formaldehyde and tannin from *Stryphnodendron adstringens* in different ratio were used as adhesive. The authors found MOE values of 1402.1, 1233.6, 1457.5, 1151.1 and 1384.3 MPa for 0, 25, 50, 75 and 100% tannin content in the adhesive, respectively. Variation is almost similar with this study.

Salari *et al.* (2013), upon evaluating the quality of OSB panels produced with 10% urea–formaldehyde adhesive with apparent density of 0.70 g cm-3, obtained mean values of MOR and MOE of 19.3 and 2,385 MPa, respectively.

5 Conclusion

Huge amount goran bark can be collected from the Sundarbans mangrove forest for the extraction of tannin. About 35% of the goran bark (weight) is water soluble tannin. This tannin can be extracted by simple hot water extraction method. Extracted tannin having 50% solid content can easily be mixed with commercial grade UF resin and applied in particleboard production. According to the results of this study, tannin in pure and mixed form with urea-formaldehyde can be a good alternative to traditional formaldehyde based adhesives. Technical performance of pure tannin based boards is very much similar to the properties of industrial grade urea-formaldehyde based particleboards. The extracted tannin can be used in commercial production of particleboard to save the environment from the formaldehyde emission of synthetic adhesives like UF. However, further studies on tannin extraction, storage and applications are needed to use the tannin as an adhesive in industrial scale.

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Appendix I: Laboratory test results

Table A- 1. Physical properties of composites of different adhesive treatment

Treatment	Density (gm/cc)	Moisture content (%)	Water absorption (%)	
			2h	24h
0% tannin	0.71	3.76	63.99	72.93
25% tannin	0.77	3.39	67.23	75.32
50% tannin	0.75	4.31	80.54	89.36
75% tannin	0.89	4.11	96.58	105.71
100% tannin	0.92	4.19	109.14	118.62

Table A- 2. Physical properties of composites of different adhesive treatment

Treatment	Thickness swelling (%)		Linear expansion (%)	
	2h	24h	2h	24h
0% tannin	32.50	50.82	1.07	1.54
25% tannin	42.80	54.40	1.00	1.40
50% tannin	37.52	52.10	1.33	1.93
75% tannin	41.21	56.15	1.31	1.69
100% tannin	70.54	116.06	2.26	3.05

Table A- 3. Mechanical properties of composites of different adhesive treatment

Treatment	MOR	MOE
0% tannin	14.01	598.72
25% tannin	15.39	676.37
50% tannin	6.49	228.58
75% tannin	9.45	452.69
100% tannin	18.43	948.00

Appendix II: ANOVA with LSD (Tukey)

Table A- 4. ANOVA for density

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	0.00276	4	0.00069	2.180584	0.144732	3.47805
Within Groups	0.003165	10	0.000316			
Total	0.005925	14				

Table A- 5. ANOVA for moisture content (%)

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	1.672677	4	0.418169	0.876347	0.511413	3.47805
Within Groups	4.771731	10	0.477173			
Total	6.444408	14				

Table A- 6. ANOVA for water absorption (%) (2 hrs)

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	4448.104	4	1112.026	37.37332	0.00001	3.47805
Within Groups	297.5454	10	29.75454			
Total	4745.65	14				

Table A- 7. LSD for water absorption (%) (2 hrs)

Treatments	N	Mean	Grouping
T-100	3	109.14	A
T-75	3	96.58	A
T-50	3	80.54	B
T-25	3	67.23	B C
T-0	3	63.99	C

Table A- 8. ANOVA for water absorption (%) (24 hrs)

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	4633.693	4	1158.423	105.8764	3.84E-08	3.47805
Within Groups	109.4128	10	10.94128			
Total	4743.106	14				

Table A- 9. LSD for water absorption (%) (24 hrs)

Treatments	N	Mean	Grouping
T-100	3	118.62	A
T-75	3	105.71	B
T-50	3	89.36	C
T-25	3	75.32	D
T-0	3	72.93	D

Table A- 10. ANOVA for thickness swelling (%) (2 hrs)

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	2649.77	4	662.4426	3.150228	0.064231	3.47805
Within Groups	2102.84	10	210.284			
Total	4752.61	14				

Table A- 11. ANOVA for thickness swelling (%) (24 hrs)

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	9484.641	4	2371.16	33.23166	0.00001	3.47805
Within Groups	713.5246	10	71.35246			
Total	10198.17	14				

Table A- 12. LSD for thickness swelling (%) (24 hrs)

Treatments	N	Mean	Grouping
T-100	3	116.06	A
T-75	3	56.1	B
T-25	3	54.4	B
T-50	3	52.1	B
T-0	3	50.82	B

Table A- 13. ANOVA for linear expansion (%) (2 hrs)

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	3.040059	4	0.760015	7.278097	0.005158	3.47805
Within Groups	1.044249	10	0.104425			
Total	4.084308	14				

Table A- 14. LSD for linear expansion (%) (2 hrs)

Treatments	N	Mean	Grouping
T-100	3	2.26	A
T-50	3	1.33	B
T-75	3	1.31	B
T-0	3	1.07	B
T-25	3	1	B

Table A- 15. ANOVA for linear expansion (%) (24 hrs)

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	5.218123	4	1.304531	7.664345	0.004294	3.47805
Within Groups	1.702077	10	0.170208			
Total	6.9202	14				

Table A- 16. LSD for linear expansion (%) (24 hrs)

Treatments	N	Mean	Grouping
T-100	3	3.05	A
T-50	3	1.93	B
T-75	3	1.69	B
T-0	3	1.54	B
T-25	3	1.4	B

Table A- 17. ANOVA for MOR

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	272.6226	4	68.15564	17.24527	0.000175	3.47805
Within Groups	39.52136	10	3.952136			
Total	312.1439	14				

Table A- 18. LSD for MOR

Treatments	N	Mean	Grouping
T-100	3	18.43	A
T-25	3	15.39	A
T-0	3	14	A B
T-75	3	9.45	B C
T-50	3	6.5	C

Table A- 19. ANOVA for MOE

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	854282.4	4	213570.6	14.95027	0.000319	3.47805
Within Groups	142854	10	14285.4			
Total	997136.4	14				

Table A- 20. LSD for MOE

Treatments	N	Mean	Grouping
T-100	3	948	A
T-25	3	676.4	A B
T-0	3	598.7	B
T-75	3	453	B C
T-50	3	228.6	C