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Mechanical and physical properties of particleboards made from Ailanthus wood and UF resin fortified by Acacias tannins blend

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Abstract
In this study the influence of adding a blend of tannins extracted from the bark of two Acacia species on the mechanical and physical properties of laboratory particleboards made from UF resin and underutilized raw materials was investigated The comminuted bark of Acacia seyal var. seyal (Ass) and Acacia nilotica spp. tomentosa (Ant) was extracted with hot water (initial temperature was 90°C) using a ratio of 1:6 powdered bark to water (w/v). Their spray dried tannins powder was blended (BT, 1:1) and was added as concentrated solution (35%) to UF resin at three different levels (5%, 10% and 15%, w/w) The obtained panels were evaluated for their mechanical and physical properties according to the BS EN relevant standards. The obtained results revealed an increase in the modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond strength (IB) of these panels when small amount (5%) of BT was added. In contrast addition of higher percentages (10&15%) was found to decrease the mechanical properties. It was also observed that the addition of BT to UF did not improve the physical properties (Thickness swelling (TS, 24 h) and water absorption (WA, 24 h), which remained comparable to the control panels.

Keywords: Tannins, UF, Ailanthus excelsa, Acacia nilotica subsp tomentosa, Acacia seyal, particleboard

1. Introduction

The high demand for wooden materials and the rise in agricultural areas and forest fires have increased the importance of composite particleboards versus solid wood. The large amount of wood consumption could mean a high worldwide deforestation rate that can cause negative impacts on the environment [1]. Developing countries, especially in Africa face an increasing demand for building materials due to fast growing urban populations and subsequent need for better shelters [2].
Currently, particleboard provides industrial users the consistent quality and design flexibility needed for the rapid and efficient production of quality consumer products [3]. The global production of particleboard is increased in the last years to more than 23 million cubic meters per year as a result of increasing in global consumption [4]. Particleboard defined as a panel manufactured from small pieces of wood or other lignocellulosic materials bonded together by the use of an organic binder together with one or more of the following agents: heat, pressure, humidity, a catalyst, etc [4].
Particleboards are among the most popular materials used in interior and exterior applications in floor, wall, ceiling panels, office dividers, bulletin boards, cabinets, furniture, counter and desk tops [5]. The major type of adhesives used to produce particle board and other wood based panels industries is Urea formaldehyde resins [6], it is estimated to about 90% or more of the world's board production [7]. Formaldehyde-containing resin is widely used as an adhesive in the manufacture of particleboard; several researches are looking for alternative adhesive systems due to the highly toxic nature of formaldehyde [8].
Phenolic-type compounds obtained from natural resources have been used for the production of wood adhesives since 1970s [9].
One of them is Tannins which are naturally occurring phenolic compounds, which have been a subject of extensive research leading to development of a wide range of industrial applications [10]. Many attempts with

varying degrees of success have been made to use these materials either totally or partially for the production of wood adhesives [11].

Tannins are used in producing formaldehyde based adhesives due to the high cost of synthetic phenolic adhesives [12]. For example, Mimosa tannin adhesives are commercially produced and used in South Africa and Brazil while Quebracho tannin adhesives are used in Argentina [13]. Mangrove tannin and other polyflavonoids have been used for leather manufacture in Nigeria but no detailed research has been done on its utilization as wood adhesive [14].

Several articles have been published that report scientific findings on using tannins as an adhesive. The tannins extracted from white Oak (Quercusalba) added to UF at different portions for decreasing the free formaldehyde content of MDF with reporting that free formaldehyde value decreased when the portion of tannins increased. However, the panels mechanical properties i.e. MOR, MOE and IB were a little lower than controlled MDF panels [15]. Tabarsa(2011) [16],produced wheat straw particleboard using tannin modified PF resin with accepted properties. On the field of composite sheet the using of Eucalyptus tannin as partial substitution of PF to produce resin with acceptable mechanical properties [17]. Kim 2009 [18] used the natural wattle tannin adhesive to replace UF resin in order to reduce formaldehyde and volatile organic compound emissions from the adhesives used between plywoods and fancy veneers. And for producing low formaldehyde emission plywood suitable for interior and exterior uses using Acacia mangium tannin and PF [19]. In a study by Pizzi, A and coworkers [20], the Tannin from larch (Larix gmelini) bark extracts was used to prepare tannin–urea–formaldehyde (TUF) resin. Larch tannin has an adverse effect on the resin curing. However, it promoted the rigidity and flexibility of the glued system and upgraded the properties of plywood. Therefore, larch tannin could be applied in the modification of urea–formaldehyde resin. Bisandaet al 2003 [2] produced particle boards made from coffee husks, and bonded using the tannin resin blend that include UF and cashew nut shell liquid to obtain particleboards with superior properties to those made using UF alone. Furthermore, as additive with UF in particleboard production to reduce the formaldehyde emission and they reported increasing in bonding strength in the case of high tannin additive content [21].

The tannins used in this study were extracted from two indigenous Sudanese trees species which is Acacia seyal var. seyal and Acacia nilotica subsp. Tomentosa.

A. seyal is a small to medium-sized tree, growing to 17 m tall. The bark of this tree is recognizable even at a distance; it has thin red-brown bark and it is native in Sudan, Egypt, Eritrea, Ethiopia and other countries in Africa and Middle East. It is considered as an important source of rural energy as both firewood and charcoal that is used widely throughout its range. In Sudan it is used to make a fragrant fire over which women perfume themselves [22]. The other source of tannin is Acacia nilotica subsp. tomentosa trees and it is widely distributed along the Nile banks and its tributaries and can withstand inundation for 3 or more months [23]. Acacia nilotica forests in Sudan manage to produce wood to use it for railway and furniture or to produce fire wood. As a result of sawing operations the bark of the tree which contains high percentage of tannin considered as waste. On the other hand the pods of Acacia nilotica which contain high percentage of tannin used as tanning agent in tanneries and for alternative medicine.

The lignocellulosic material used in this study due to their light weight (low density) and their availability was Ailanthus excelsa wood particles.

Ailanthus excelsa is a large, fast-growing tree which can reach a height of 20-24m [24]. Native to central, western and southern India, but is now being spread to other semi-arid and subtropical areas. It is fairly common in Central Sudan and is found planted on riverine and sandy soils [25]. In India perhaps its most important use is as a fodder tree. It is harvested twice a year, producing a very potable and nutritious fodder which is commonly sold in markets and given to stallfed goats [24]. However, the use of Ailanthus excelsa as timber; the wood is easily worked but is perishable and subject to insect attack and stain. Nevertheless, it is used in boxes, crates, poles, fishing floats, tool handles, matches and drums [24, 26].

Density of the raw material furnish is the most critical parameter when determining the potential of a given species for particleboard manufacture [27]. A lower density wood will lead to form a thicker mattress. The density most commonly used in particleboard production vary from 0.30 to 0.50 g/cm3. Higher density species can be used in combination with lower density ones. Wood density is the most important variable to take into account, since the quality of the particleboard depends on it. A low density wood provides a high density compaction rate and, therefore, a higher contact surface between the particles than high density wood. This leads to a more uniform product with a greater capacity to transmit loads between the particles, resulting in higher flexural and internal bond properties in particleboards made of low-density wood [28].

The effect of wood density has been extensively studied and the general conclusion reached that species of lower density than the final particleboard density are required to economically produce high quality particleboard [29, 30].
The aim of this study was to investigate the feasibility of adding tannins to Urea Formaldehyde resin in order to improve the mechanical and physical properties of particleboard made from the Ailanthus excelsa wood particles which regarded as low density wood species and low quality and underutilized invasive species with wide distribution in South Kordfan and Blue Nile states in Sudan.

2. Materials and Methods

2.1. Particles Preparations

Ailanthus excelsa wood raw material was collected from South Kordoufan State. A tree of 15 meter height was felled and crosscut into logs of 1.5 m length. The logs were further chopped into small disks of 3 cm in thickness to avoid rotting and decay. They were air dried, debarked and then manually chipped to about 3 mm thickness and hammer milled using Yoshida Seisakusho hammer mill to achieve suitable particle size. They were then sieved using a mesh (850 µm) to eliminate powder and small particles. The particles retained on the sieve were used for particleboard preparation.

2.2. Determination of wood pH

The pH of wood particles was determined according to the method described by [31]. One gram of particles sample of was soaked in 20 ml distilled water. The solution was kept at 20±1 C^o, shaken for 30 min, and moved to water bath for 10 min at 100 C^o before the pH was measured.

2.3. Extraction of the Tannins

Tannins were extracted according to a method reported by Osman Z., 1995[32]. However the initial extraction temperature was increased from 70 to 90°C. A ratio of 1:6 (bark: water w/v) was used to prepare the extract which left for overnight. The resultant solution was filtered using special cloth. The filtrates volumes were determined and spray dried using a laboratory spray dryer (Bowe Engineering) at 175 °C and a rate flow of 10 ml/min.

2.4. Determination of stiasny number and pH of the tannins

The pH of aqueous tannins extracts were prepared from the spray dried powder at a concentration of 35% was measured using pH meter. The Stiasny number test was carried out in order to determine the percentage of the total polyphenolic materials using a reported method by Yazaki *et al*.[33]. A sample of the tannins spray dried powder (0.1 g) was dissolved in water (10 ml) and reacted at 100 C^o with 36 – 38% formaldehyde (2 ml) catalyzed by 10N HCl (1 ml) for 30 minutes. The resultant precipitate (Stiasny precipitate) was accurately weighed and the Stiasny value calculated as a percentage of the weight of the dried extract.

2.5. Urea Formaldehyde resins

Commercial Urea Formaldehyde adhesive with the following properties; viscosity = 2.24 p, specific gravity = 1.269, solid content = 65.3% and 0.32% free formaldehyde, was used to produce the particleboards.

2.6. Preparation of UF - Tannins modified resins

Using a ratio of 1:1 Acacia seyal var. seyal tannin / Acacia nilotica tannin were thoroughly mixed in order to form the blended tannins (BT). Then an aqueous tannins solution of 35% concentration (w/w) was prepared from the BT. Five percent of Paraformaldehyde based on the oven dry weight of tannin was added immediately to the tannins solution before mixing with UF resin, and then the mixture was immediately sprayed on the wood particles. Three types of tannins modified UF resins (95% UF: 5% BT, 90% UF: 10% BT and 85% UF: 15% BT) were used to produce the particleboards.

2.7. Particleboard preparation

12% (w/w on the oven dry weight of wood particles) of each type of adhesives formulation was used to produce panels of 340 mm × 340 mm × 10 mm in size using Carver hot press at 180 C^o and 150 bar pressing pressure; a pressing time of 7 min was applied. The obtained particleboards were placed in the conditioning room (20 ºC and 65 % relative humidity) for 2 – 3 days before testing.

2.8. Particleboard Evaluation

To evaluate the mechanical properties of the panels, Tthe modulus of elasticity (MOE) and modulus of rupture (MOR) were tested in accordance with BS EN 310: 1993. The internal bond (IB) test was carried out using BS EN 319: 1993. Physical properties such as thickness swelling after two hours soaking in cold water (2hrs TS), thickness swelling after twenty four hours soaking in cold water (24hrs TS) and water absorption (WA) were tested in according to BS EN 317: 1993. Six samples specimens were prepared for each test. All the obtained results were statistically analysed.

2.9. Test Specimens Preparation

The test specimens were prepared according to BS EN 1993 standards.

3. Results and Discussions

3.1. Determination of wood and tannins pH

As the pH value could influence the panels' properties by interfering with the adhesive hardening, the pH of Ailanthus excelsa was found to 6.5 and the pH of all types of tannins ranged between 5.2 to 5.4.

3.2. Tannins Stiasny number

Stiasny number or catechin numbers which indicates the percentage of condensed tannins and hence tannins reactivity was determined for the two types of the tannins. Ass shows higher value of Stiasny number (82.18%) when compared to Ant (73.08%). It could therefore, expected to be more reactive than Ant against formaldehyde.

3.3. Mechanical Properties

The results of the mechanical properties tests for Ailanthus excelsa particleboards were shown in Table 1. Among the studied adhesives formulations, it has been noticed that the formulation [95%UF:5%BT] showed the highest performance, by producing the best values for all mechanical properties (MOR, MOE and IB) and it is recorded (10.35 N/mm2, 1578.85 N/mm2, and 0.83 N/mm2 respectively) when compared with the values obtained by UF alone which were 7.79 N/mm2 , 1051.82 N/mm2 and 0.40 N/mm2 respectively. Furthermore, it indicate that the addition of small amount of tannins (5%) to the UF resin could improve its performance resulting in better mechanical properties. Similar results had been observed by Osman and coworker, 2007 [34], who reported that the UF Tannins modified adhesives could successfully be applied as binder for the manufacturing of particleboard with superior strength properties compared to those made from pure UF adhesive. Formaldehyde emission could also be reduced by addition of tannins, which should further be studied in a future work.

When the percentage of BT increased it decreased the mechanical properties of the produced panels. Similar observation was reported by Nemli G ,et al, 2004 [35], who stated that the mechanical properties (MOR, MOE and IB) were decreased when the wood particles were impregnated by the mimosa bark extract. It was also noticed that when the tannin solution concentration was increased in the UF resin, the MOE values were slightly decreased for MDF panels produced with UF resin containing tannin solution [15]. The same observation was also noticed by Tabarsa et al 2011 [16] who recorded that the increase of tannins percentage on PF resin lead to a drop in the mechanical and physical properties of particleboards produced with wheat straw. Also it has been observed that, the usage of an excessive formaldehyde scavenger such as tannin solution leads to some changes in fiber structure, and thus the mechanical properties of MDF panels were accordingly decrease [15]. The decreased in mechanical properties induced by the addition to tannins to UF could be attributed to the decrease in the UF molar ratio which lead to decrease in the formaldehyde as only 5 percentage of paraformaldehyde was added to the tannins. It could also be explained by the fact that, the added tannins was reacted with the whole of the free Formaldehyde rendering the resin with unbalanced molar ratio

The curing rates of formaldehyde-based resins were very dependent on the pH of the environment in which they cure. If the pH is low then pre-curing may result. When an adhesive pre-cures, the board's layer is weak and flaky. This is because the binder cures before the particles have been compressed and so when the press closes the precured resin bonds were broken [35]. Decreasing pH of the particles to the acceptable levels for good adhesion and successful curing of urea formaldehyde adhesive caused better physical and mechanical properties. Low pH of the particles (below 4) causes precuring of the adhesive before hot pressing process [36].

It has also been reported that the UF adhesives, which are the most common wood adhesives used in the manufacture of particleboards, cure in acidic environment, and wood acidity can affect the rate at which UF adhesives harden [37]. Furthermore the IB value obtained by all tannins modified UF except [95%UF:5%BT] formulation was not significantly different from each other and pure UF adhesive.

Table 1. Mechanical properties of *Ailanthus excelsa* particleboards.

Adhesives	MOR N/mm^2	MOR SD	MOE N/mm^2	MOE SD	IB N/mm^2	IB SD
UF	7.79 [b]	0.463137	1051.82 [cd]	110.590429	0.40 [b]	0.060732
[95%UF:5%BT]	10.35 [a]	2.112220	1578.85 [a]	300.909078	0.83 [a]	0.061738
[90%UF:10%BT]	9.60 [ab]	0.782381	1443.80 [ab]	141.526044	0.46 [b]	0.056668
[85%UF:15%BT]	8.80 [ab]	0.591008	1279.76 [abc]	132.898032	0.42 [b]	0.071601

(a indicate best group followed b,c and so on, Values within the same column followed by the same letters are not significantly different at P =0.05.)

3.2. Physical properties of Ailanthus excelsa particleboards

Table 2 summarizes the obtained results of thickness swelling after two hours (2hrsTS) and after twenty four hours (24hrsTS), water absorption after 24 hours (WA) and the boards' densities. The UF adhesive gave 37.31 %, 42.85 % and 113.02 % respectively for 2hrs TS, 24hrs TS and WA respectively. These values were considered the best values among all tested adhesives formulations used in this study. Furthermore, there were no significant differences between the UF, and the all tannins modified adhesives. All UF: BT formulations produced comparative values

It is worth noting that the addition of tannins to UF adhesive reduced the physical properties of the boards. Similar observation was recorded by Osman and coworkers, 2007[34], who stated that the mixing of different proportions of tannins with UF lead to a decrease in the TS when the tannins percentage increased. These results were also in line with what Boran et al, 2012 reported [15].They observed that as the concentration of tannins content increased in UF resin , the 24hrs water soak test values of the produced panels were higher than the control [15]. Also Tabarsa *et al.* 2011[16] had the same observations with wheat straw particle board using tannins modified PF adhesives. This observation could also be correlated to the board density. The increase of board density and press time resulted an improvement in all of the mechanical properties while decreasing their physical properties [35].

Table 2. Physical properties of *Ailanthus excelsa* particleboards.

Adhesives	2hrsTS %	2hrsTS SD	24hrsTS %	24hrs TS SD	WA %	WA SD	Density g/cm^3	Density SD
UF	37.31 [a]	1.293029	42.85 [a]	3.043818	113.02 [a]	2.991267	0.640 a	0.01251
[95%UF:5%T]	37.11 [a]	3.096256	42.48 [a]	2.941765	104.37 [a]	6.726756	0.658 a	0.02155
[90%UF:10%T]	38.96 [a]	1.754568	44.98 [a]	1.396206	112.75 [a]	2.960589	0.639 a	0.00689
[85%UF:15%T]	41.80 [a]	3.725732	47.63 [a]	3.364610	113.87 [a]	5.637185	0.658 a	0.03671

(a indicate best group followed b,c and so on, Values within the same column followed by the same letters are not significantly different at P =0.05.)

Conclusion

The study concluded that Ailanthus excelsa wood particles could be a promising raw material for particleboard production in Sudan due to their availability and limited usage (as fodder in dry seasons). The addition of tannins to Urea formaldehyde adhesives could improve the mechanical properties of particleboards (MOR, MOE and IB) when it added in small amount. However, it also decreases the physical properties of the produced panels. The pH of the glued particles and boards' density are a very important variable which should be investigated in future studies together with the possible reduction of the formaldehyde emission

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