PROPERTIES OF PARTICLEBOARDS MADE FROM *ACACIA SEYAL VAR. SEYAL* USING UF–TANNIN MODIFIED ADHESIVES

M. ELBADAWI,[*] Z. OSMAN,[*] T. PARIDAH,[**] T. NASROUN[***] and W. KANTINER[****]

[*]*Institute for Technological Research, National Center for Research, Khartoum, Sudan*
[**]*Institute of Tropical Forestry and Forest Products, University Putra Malaysia (UPM), Serdang, Malaysia*
[***]*Forestry Research Centre, Agricultural Research Corporation, Sudan*
[****]*Dynea, Krems, Austria*
✉ *Corresponding author: Z. Osman, zeinabosm@yahoo.com*

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The aim of this study was to investigate the effect of adding a blend of tannins to commercial urea formaldehyde (UF) on the properties of particleboard made from wood particles of *Acacia seyal var. seyal*. The tannins were extracted from the bark of *Acacia seyal var. seyal* (Ass) and *Acacia nilotica subsp. tomentosa* (Ant) with hot water (initial temperature was 90 ºC), using a ratio of powdered bark to water of 1:6 (w/v). The tested Acacia species (Ass and Ant) exhibited high tannin contents (82.18% and 73.09%, respectively). A blend from the two tannin types (BT) was made (1:1 w/w) and added to UF in the form of a concentrated solution (35%) at three different percentages (5%, 10% and 15%, weight/weight). The different UF–BT formulations were used to produce particleboards (340 mm × 340 mm × 10 mm in size). The obtained panels were tested according to the BSEN relevant standards and showed high mechanical properties, compared to the ones produced by solely UF. It was also observed that the addition of BT to UF did not improve the physical properties of the panels (thickness swelling (TS) and water absorption (WA)), but the results obtained were slightly higher than the ones for the UF panels.

Keywords: tannins, UF, *Acacia nilotica subsp. tomentosa*, *Acacia seyal*, particleboard

INTRODUCTION

Particleboards are used extensively in the construction, furniture, cabinet and joinery industries, while the ability to 'engineer' these products to meet specific performance requirements has been a significant factor in their growth. The technology for producing these panels has developed significantly, in particular with the introduction of continuous pressing, providing new levels of product uniformity.[1] Urea–formaldehyde (UF) has been the major adhesive for wood-based particleboards. Although formaldehyde-containing resin is widely used today as an adhesive in the manufacture of particleboard, scientists are seeking alternative adhesive systems due to the highly toxic nature of formaldehyde.[2]

UF adhesives have several strong positive aspects, such as their very low cost, non-flammability, very rapid cure rate and light color. However, they also present some disadvantages: the bonds are not water-resistant and formaldehyde continues to evolve from the adhesive. UF adhesives are the largest class of amino resins, and the predominant adhesives for interior grade plywood and particleboard.[3,4] UF adhesive is used in the production of an adhesive for bonding particleboard (61% of the urea-formaldehyde used by the industry), medium density fiberboard (27%), hardwood plywood (5%), and a laminating adhesive for bonding (7%), for example, furniture case goods, overlays to panels, and interior flush doors.[5]

A number of natural phenolic-containing materials are also available, including lignins and tannins. Many attempts with varying degrees of success have been made to use these materials either totally or partially for the production of wood adhesives.[6] The word tannin has been used loosely to define two different classes of chemical compounds of mainly phenolic nature: hydrolyzable tannins and condensed tannins. Their chemical behavior towards formaldehyde is analogous to that of simple phenols of low reactivity and their moderate use as phenol substitutes in the above-mentioned resins does not present difficulties. Their lack of macromolecular structure in their natural state, the low level of phenol substitution they allow, their low nucleophilicity, limited worldwide production, and their higher price somewhat decrease their chemical and economical interest.[3]

Acacia seyal is a small to medium-sized tree, growing to 17 m tall.[7] It has thin red-brown bark. The inflorescence is a bright yellow, axillary penduculate, globose head. Pods are slightly curved, thin, and 7-20 cm long.[8] *Acacia seyal* is native to Sudan, Egypt, Eritrea, Ethiopia, Ghana, Iran, Kenya, Malawi, Mali, Mozambique, Namibia, Niger, Nigeria, Saudi Arabia, Senegal, Syrian Arab Republic, Tanzania, Uganda, Yemen, Republic of Zambia, and Zimbabwe. Besides, it is exotic to Afghanistan, Bangladesh, Bhutan, India, Nepal, Portugal, Sri Lanka and the US.[7]

Acacia seyal has been used for food as Gum talha, which can be eaten when fresh, and in combination with some local foods. The bark is extensively used for feeding cattle, sheep and goats during the dry season. The pods and leaves are nutritious and palatable to livestock. Moreover, *A. seyal var. seyal* is an important source of rural energy as both firewood and charcoal, being widely used throughout its range. In Sudan, it is used to make a fragrant fire over which women perfume themselves.[8]

Acacia nilotica subsp. tomentosa is widely distributed along the Nile banks and its tributaries, and can withstand inundation for 3 or more months.[9] In Sudan, *Acacia nilotica* forests manage to produce wood for railway sleepers, furniture and firewood. The tree bark, which contains a high percentage of tannin, is considered as waste in the area of saw mills, while the pods, which also contain high content of tannin, are used as tanning agent in tanneries and for alternative medicine.

The idea of using tannins as a bioadhesive and as additive in composite manufacture was investigated in a number of previous studies.[10] The tannin extracted from *Acacia mangium* was added to phenol formaldehyde resin to produce plywood with low formaldehyde emission, suitable for both interior and exterior uses.[11] Also, Eucalyptus tannin was used to partially substitute PF to produce resins with feasible mechanical properties for manufacturing composite sheets.[12] In addition, tannins were used with urea formaldehyde (UF) in particleboard manufacture, as additive to reduce the formaldehyde emission level, and the study on the adhesion properties of the urea formaldehyde resin for particleboard led to the conclusion that curing the high tannin additive content in this adhesive system indicated that the bonding strength increased.[13] However, F. A. Cameron and coworkers[14] reported that the addition of tannin extract to UF resin appeared only to be a "stop-gap" short-to medium-term measure and the board reverted to emissions similar to those of the UF controls once the free formaldehyde was scavenged by the tannins.

In another study, in order to determine the effect of the tannin content in the urea formaldehyde (UF) resin on the panel properties of medium density fiberboard (MDF), tannin extracted from the bark of white oak (*Quercus alba*) was added to the UF resin in different ratios (based on the resin) to decrease the free formaldehyde content of MDF panels, and it was determined that the free formaldehyde values of MDF panels decreased when the ratio of the tannin content in UF resin increased. However, the MOR, MOE, and IB of these panels were a little lower, and the thickness swelling (24 h) and water absorption (24 h) values were a little higher compared to the control MDF panels.[15]

The aim of this study was to investigate the possibility of adding tannins to urea formaldehyde resin in order to improve the mechanical and physical properties of particleboard made from the wood of *Acacia seyal* – an indigenous tree growing in Sudan with wide distribution in Kordfan and Blue Nile states.

EXPERIMENTAL

Particle preparations

The logs of *Acacia seyal var. seyal* were collected from the Blue Nile state, air dried, debarked and crosscut into about 3 cm thick disks. They were then manually chipped to about 3 mm thickness and hammer milled using a Yoshida Seisakusho hammer mill to achieve a 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.

Determination of wood pH

The pH of wood particles was determined according to the method described by T. Lebw *et al*.[16] Particle samples of one gram were soaked in 20 ml distilled water. The solution was kept at 20±1 °C, shaken for 30 min, and then moved to a water bath for 10 min at 100 °C before the pH was measured.

Extraction of tannins

Tannins were extracted according to a method reported by Z. Osman.[7] However, the initial extraction temperature was modified from 70 to 90 °C. A ratio of 1:6 (bark:water w/v) was used to prepare the extract, which was allowed to stand overnight. The resultant solution was filtered using a special cloth. The filtrate volumes were determined and spray dried using a laboratory spray dryer (Bowe Engineering) at 175 °C and a flow rate of 10 ml/min.

Chemical analysis of tannins

In order to determine the Stiasny number (catechin number, indicating the percentage of condensed tannins), the chemical analysis of the tannins was carried out and the pH was determined on an aqueous solution prepared from the spray dried powder at a concentration of 35% as described below.

Determination of Stiasny number

A sample of the tannin spray dried powder (0.1 g) was dissolved in water (10 ml) and reacted at 100 °C 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 was calculated as a percentage of the dried extract weight. The percentage polyflavanoid contents in the bark were calculated by a method described by Y. Yazaki *et al*.[18]

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.

Preparation of UF: tannin modified resins

Acacia seyal var. seyal tannin and *Acacia nilotica* tannin in a ratio of 1:1 were thoroughly mixed in order to form blended tannins (BT). Then an aqueous tannin 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 tannin solution before mixing with UF resin without any change in the molar ratio. The mixture was immediately sprayed on the wood particles. Three types of tannin modified UF resins (95% UF: 5% BT, 90% UF: 10% BT and 85% UF: 15% BT) were used to produce the particleboards.

Particleboard preparation

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

Particleboard evaluation

The evaluation of particleboard was carried out according to BS EN 1993 standards for mechanical properties: Module of Rupture (MOR), Module of Elasticity (MOE) and Internal Bond (IB). The physical properties tested were thickness swelling after two hours of soaking in cold water (2hTS), thickness swelling after twenty four hours of soaking in cold water (24hTS) and water absorption (WA).

Test specimens preparation

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

RESULTS AND DISCUSSION

Determination of wood and tannins pH

The pH of *Acacia seyal var. seyal* was 7, which is considered neutral, and the pH values of all the studied tannins were between 5.2 and 5.4. It was found that the pH of the system (glued particles) could influence the overall properties of the panels by interfering with adhesive hardening. Therefore, when phenolic types of resins are used to make panels, the final pH of the system should be carefully studied and adjusted according to the resin types and setting requirements.[19]

Stiasny number of tannins

Stiasny number, or catechin number, which indicates condensed tannins, and may also show tannin reactivity was determined for the two types of tannins. The Ass tannins showed a higher value of Stiasny number (82%), when compared to Ant tannins (73%). It could be therefore expected that Ass tannins would be more reactive than Ant ones.

Mechanical properties

The results of the mechanical properties testing for *Acacia seyal var. seyal* particleboards are presented in Table 1. Among the studied formulations, it has been noticed that the formulation [95%UF:5%BT] showed high performance, by producing the best values for MOR and MOE (8.21 N/mm^2 and 1140.20 N/mm^2, respectively), when compared with the values obtained for UF, which were 7.18 N/mm^2 and 906.19 N/mm^2, respectively. It is worth noting that the addition of a small amount of tannins to the UF resin could upgrade its performance. This observation is also economically viable. Similar results have been observed by Z. Osman and coworkers,[20] who reported that the UF tannin modified adhesives could successfully be applied as binder for the manufacture of particleboard with superior strength properties, compared to those made from pure UF adhesive. The addition of tannins could also reduce formaldehyde emission, which should further be studied in a future work.

It has been observed that when the percentage of BT increased, it decreased the mechanical properties of the produced panels. This could be attributed to the decrease in the molar ratio of UF; hence less formaldehyde, i.e. only 5% paraformaldehyde, was added to the tannins. Another explanation could be that the added tannins reacted with the whole of the free formaldehyde, rendering the resin with an unbalanced molar ratio. A similar observation was made by G. Nemli *et al.*,[21] who stated that the mechanical properties (MOR, MOE and IB) decreased when the wood particles were impregnated by a 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] In addition to this observation, it has been recorded that the usage of an excessive formaldehyde scavenger, such as a tannin solution, leads to some changes in the fiber structure, so the mechanical properties of MDF panels decrease accordingly.[15]

The panels made from BT:UF modified adhesives showed lower IB values than the one achieved by UF alone. This could be due to the final pH of the glued particles, which was probably not acidic enough for the UF setting. Nevertheless, the higher IB value (1.12 N/mm^2) for BT:UF adhesives was obtained by the formulation [85%UF:15%TB] and this could be possibly attributed to the high percentage of tannins, which increased the acidity of the system and hence resulted in a good setting for the UF resin. It has been noticed that the curing rates of formaldehyde-based resins were very dependent on the pH of the curing environment. If the pH is low, then pre-curing may occur. When an adhesive pre-cures, the board layer is weak and flaky. This is because the binder cures before the particles have been compressed and so when the press closes the pre-cured resin bonds are broken.[21]

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.[22] Furthermore, the IB value obtained by the [85%UF: 15%BT] formulation (1.21 N/mm^2) was not significantly different from that of the pure UF adhesive (1.14 N/mm^2).

Table 1
Mechanical properties of *Acacia seyal* particleboards*

Adhesives	MOR (N/mm^2)	MOE (N/mm^2)	IB (N/mm^2)
UF	7.18	906.19	1.14
[95%UF:5%BT]	8.21	1140.20	0.96
[90%UF:10%BT]	7.42	992.40	0.88
[85%UF:15%BT]	7.95	1091.70	1.12

*BS EN standard required values for boards for interior fitments and furniture are: 14 N/mm^2 for MOR, 1800 N/mm^2 for MOE, and 0.4 N/mm^2 for IB

Table 2
Physical properties of particleboards made from *Acacia seyal*

Adhesives	2hTS, %	24hTS, %	WA, %	Density (g/cm^3)
UF	19.24	22.14	78.58	0.657
[95%UF:5%BT]	21.16	25.36	83.12	0.659
[90%UF:10%BT]	21.92	25.99	82.11	0.663
[85%UF:15%BT]	20.27	24.18	87.05	0.631

Physical properties of *Acacia seyal var. seyal* particleboards

Table 2 summarizes the obtained results for thickness swelling after two hours (2hTS) and after twenty four hours (24hTS), water absorption after 24 hours (WA) and the board densities. The UF adhesive gave 19.24%, 22.14% and 87.58% for 2hTS, 24hTS and WA, respectively. These values were considered the best values among the three BT:UF adhesive formulations used in this study. Furthermore, the values obtained were slightly higher than the ones achieved by UF only. All BT:UF formulations produced comparative values.

It is worth noting that the addition of tannins to UF adhesive reduced the physical properties of the boards. A similar observation was recorded by Z. Osman and coworkers,[19] who stated that the mixing of different proportions of tannins with UF led to a decrease in the TS when the tannins percentage increased. These results are all in line with those reported by Boran *et al.*,[15] who observed that as the concentration of tannins content increased in the UF resin, the 24 h water soaking test values of the produced panels were higher than those for the control.

This observation could also be correlated to board density. The increase in board density and press time caused an increase in the mechanical properties and a decrease in the physical properties.[21]

CONCLUSION

The study leads to the conclusion that the addition of tannins to urea formaldehyde adhesives can improve the mechanical properties of particleboards (MOR, MOE and IB), when a small amount is added. However, the addition of tannins also decreases the physical properties of the produced particleboards. The pH of the glued particles is a very important variable, which should be investigated in future studies, together with the possible reduction in the rate of the formaldehyde emission.

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