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Production of rice husks composites with *Acacia mimosa* tannin-based resin

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Abstract Rice husks are amongst the typical agricultural residues, which are easily available in huge amounts. They have been considered as raw material for composites panels' production. However, the major hindrance in rice husks utilization for composite manufacture lies in the lack of direct interaction with most adhesive binders to form the anticipated interfacial bonds. Rice husks are highly siliceous and have poor resistance to alkaline and acidic conditions. Manufacture of rice husks composites panels having good interface bond is difficult and largely dependent on a proper understanding of the interaction between the husks and the binder. This paper presents and discusses results on the production of composites boards from a mixture of rice husks and wattle (*Acacia mimosa*) tannin based resin. The experimental results have shown that the 'as received rice husks' when blended with alkali-catalyzed tannin resin do not result in optimum composite panel properties. However, it was found that a slight physical modification of the rice husk particles by hammer-milling resulted in drastic improvements in the interfacial bond strength and stiffness of the composites panels from 0.041 MPa to 0.200 MPa and 1039 MPa to 1527 MPa, respectively.

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Introduction

One of the most common agricultural residues produced in vast amount is rice husk. Like any other lignocellulosic material, rice husks contain a significant amount of cellulose, hemicelluloses, and lignin. These three components have been known to dictate most of the properties of lignocellulosic materials [1]. However, unlike other residues, rice husks also contain a large proportion of ash and silica, which to a large extent has been reported to affect the property and application of rice husks [2–4]. Table 1 shows the estimated amount of agricultural/plant products residues or waste resulting from harvesting of the main crop. The estimated residue factor gives the factor by which the primary product crop has to be multiplied to get the total residue or waste in the last column in Table 1 generated in producing the said amount of crop. From Table 1 it can be found that rice husks are the only agricultural residues which can easily be obtained in enough amounts for industrial or commercial applications in Tanzania compared to others: This is in terms of logistics and other applications.

Today, the manufacture of composite materials from agricultural residues such as rice husks is becoming a reality. However, in order to successfully achieve this there must be an understanding of several underlying factors pertaining to the manufacture of lignocellulosic composite. One of the most relevant aspects with regard to using lignocellulosics for manufacture of composites is their polar nature with susceptibility to degradation when exposed to aggressive environments like ultra violet radiation, moisture, heat, and chemicals [1]. Protection of these materials against some of these exposures has been enhanced by impregnating lignocellulosic materials with

Table 1 Estimates of amounts of agricultural/plant product residues [5, 6]

Type of Residue	Primary Product (Crop)	Annual Production (000 tones)	Estimated Residue (w/w of Primary Crop)	Total Residues (000 tones)
Corn stalk	Maize	2638.0	2.00	5276.0
Rice straw	Paddy	681.0	3.75	2574.0
Corn cob	Maize	2638.0	0.25	659.5
Bagasse	Sugar cane	16.8	3.30	385.4
Rice husks	Paddy	681.0	0.25	204.3
Coir	Coconut husk	22	0.3	6.6
Coconut dust	Coconut husk	22	0.7	15.4

synthetic or natural adhesive polymers. This can be achieved by formation of strong interfacial bond between lignocellulosic materials and polymeric binders as a result of substantial chemical, physical, absorption interaction mechanisms at their interfaces. To understand whether these materials are suitable for producing quality composites we need to understand the factors affecting the interfacial bonding between lignocellulosic materials and the adhesive polymer.

Tannin-based adhesive resins have been generating a lot of interest in the scientific community [7, 8]. Tannin is one of the naturally occurring plant polyphenols, other than lignin, which is composed of very dense groups of oligomers and polymers. The term ''tannin'' comes from the ancient Celtic word for oak (*Quercus* sp.), which still remains a popular source for tannins used in converting animal skin into leather (tanning) [7]. Today there are many other plant species that are being used to produce commercial tannin. These include *Acacia* sp. (wattle), *Eucalyptus* sp. *Mirtus* sp. (myrtle), *Acer* sp. (maple), *Betula* sp. (birch), *Salix Caprea* (willow) and *Pines* sp. (pine).

There are two main types of tannins: condensed tannin (CT) sometimes called Proanthocyadnidis (PA) and hydrolysable tannin (HT) [9]. The CT consists of flavonoid units (Fig. 1) that have undergone various degrees of condensation. It is a hydroxylated aromatic polymer based on a 15-carbon flavonoid monomer unit. The poly-flavonoid polymers of CT carry either *resorcinolic* (from wattle and quebrancho) or *phloroglucinolic* (from pines). It also has non-phenoilc components like pinitol. HT is a mixture of simple phenols such as Pyrogallol, Ellagic acid, Gallic acid and m-Digallic acid. Figure 2 shows the chemical structure of some of the simple phenols ellagic and *m*-Digallic acid obtained in HT.

Returning to the interface between a lignocellulosic material, for example, rice husks and an adhesive (for example a tannin-formaldehyde based resin), the designer's anticipation is to find physical and chemical coupling taking place between them as they are brought together. In order for this to take place there must be a uniform distribution of the binder over and within the particles.

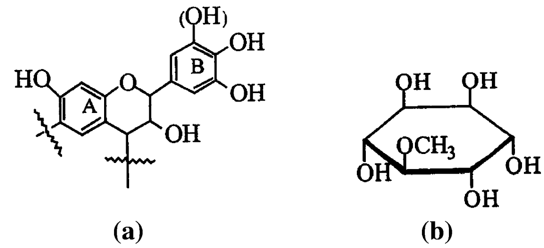

Fig. 1 Condensed tannin products: (a) Condensed Wattle tannin Flavonoids (b) Non phenolic component of wattle tannin—Pinitol

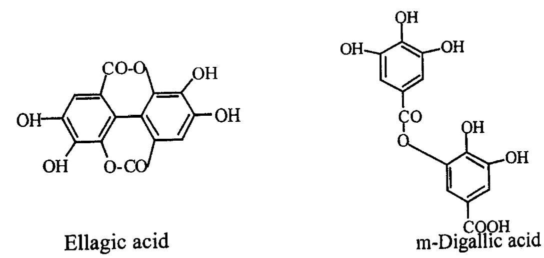

Fig. 2 Some forms of Hydrolyzed Tannin (HT)

However, when the particles furnish has a large proportion of fine particles, smaller particles absorb a considerable amount of resin compared to larger particles. Based on this reason, it has been recommended that the proportion of fine particles in the mixture of particles and resin should not exceed 6% [10]. In addition, the rice husks need to have a lower bulk density in order to reduce the high pressure necessary for their compaction, which has the tendency of inducing internal stresses at the particle–matrix interface. The bulk density can be reduced by crushing the rice husks particles [5]. Concurrently, the moisture content in the mixture after blending should be kept to a minimum level necessary only for aiding the heat transfer from the board face to the core during hot-pressing. A certain amount of moisture in the particles is also important for keeping them flexible and non-brittle [5]. This avoids generation of excessive steam, which forms pockets or voids in the composite as it escapes thus leading to blister and delamination at their interfaces. A combination of the spring-back and the escaping steam becomes damaging to the substrate-adhesive interface bond [11, 12]. However, a high compaction is also beneficial in terms of reducing inherent voids in lignocellulosic materials [13] and generation of the necessary stresses that enhance bonding at the particle-binder interface during compaction [4]. Therefore, the optimum moisture and amount of compaction can be quantified based on a compromise between their merits and demerits during production of composite boards.

As pointed out by Bisanda et al. [7], much of the work on tannin-based adhesives has been focused on developing resins for exterior application as well as lowering formaldehyde emission levels. This paper presents and discusses the possibility of producing composite boards made from a mixture of rice husks and *Acacia mimosa* tannin-based resin.

Materials and methods

Resin preparation

The resin with a solid content of 58% was prepared and supplied by the Tanzania Industrial Research and Development Organisation (TIRDO) which is based in Dar es Salaam, Tanzania. Dry tannin powder from Tanganyika Wattle Company in Njombe was dissolved in water to give a 44% tannin in water dispersion. This was hydrolyzed accordingly with 33% sodium hydroxide and castor oil for 3 h at 90 °C. The solution was then cooled to 70 °C and 4 g of 99.5% acetic acid was added, after which the mixture was cooled to 25 °C prior to any cross-linking reaction in the resin. For every 1,800 g of hydrolyzed tannin, 160 g of Urea was added to assist in breaking of hydrogen bonds and 320 g of formaldehyde (40%) were added for condensation reaction. In addition 160 g of Cashew nut shell liquid (CNSL) were added as an emulsifier. The mixture was thoroughly stirred and its pH adjusted to 5 by adding small amounts of acetic acid and sodium hydroxide.

Fabrication of the composite boards

The raw materials used in this study were crushed and uncrushed rice husks. The rice husks were obtained from Fidahussein & Company LTD based in Dar es Salaam, Tanzania. Saw dust was added to the uncrushed rice husk. The rice husks were mixed with tannin-based resin in different weight ratios according to the targeted density using a simple mechanical blender. The mat was prepared in a wooden mould-box having internal dimension 380 mm × 380 mm × 225 mm and was pre-compressed before hot-pressing. The composite panels were produced by varying the pressing temperature, time, and pressure until the optimized processing conditions were achieved. This was found to be a pressure of 27.6 MPa, temperature of 180 °C, and press closing and opening cycle of 12 min. The resulting panels were trimmed and specimens for different tests were machined from them. Three replicated samples were used for each test and the average value was used to characterize the properties of composite boards produced.

Mechanical tests

The internal bond strengths (IBS) of the husk boards were determined by measuring the tensile strength of the board perpendicular to its surface. This was done according to BS 5669: Part 1. Three samples 65 mm by sample thickness (varied from 11.1 mm to 12.3 mm) were each bonded between two rigid wooden blocks of the same surface dimensions by white wood glue. After curing of the glue for 24 h the samples were tested in a Zwick/Z010 universal testing machine at a speed of 4 mm/min.

The impact resistance of the rice husk particle board was determined according to the method described in BS 5669: Part 1: 1989, Section three. The procedure for the determination of the impact strength involved dropping a 340 g body of spherical end shape (60 mm diameter) from a series of increasing heights at a fixed point on a rigidly supported body until a visible penetration or observation of failure was observed. Sample sizes were 152 mm by 152 mm by board thickness. The impact strength was calculated from the ratio of the height producing failure to the board nominal thickness.

Flexural bending tests were done to determine the modulus of rupture (MOR) and the modulus of elasticity (MOE) of samples on a three point bending test. Four specimen cut from the pressed rice husk boards were used. The specimen had variable lengths as recommended by standards, with a constant width of 76 mm. The samples were conditioned at 60% relative humidity (RH) and 20 °C, before testing in the Zwick Z010 machine at a loading speed of 4 mm/min. The span length was 250 mm.

Results and discussion

Interfacial bond strength

The interfacial bond strength, which is often referred to as the IBS was determined by conducting the tensile strength test perpendicular to the thickness of the composite board. The results for this test are shown in Fig. 3 and in Table 2. Figure 3 shows a comparison of the behaviour of the different rice husks i.e. BRH (broken rice husks), URH (unbroken rice husks) and URB + 30 SD (Unbroken rice husks with 30% saw dust) with varying resin content.

Regression analyses of the results in Fig. 3 show a predominantly linear correlation between the resin content and the interfacial bond strength. The increase in interfacial bond strength with the resin loading attests the proposition that there is better particles–resin interaction as the resin content is increased [12]. The difference in the slopes of the three curves suggests further that the interaction is also dependent on the particle geometry as well as on the type of particles. Crushing the rice husk particles was apparently found to stimulate the interaction with the resin at higher resin loading than any of the other particles furnish. However, addition of sawdust in the rice husk particle furnish was seen to slow down the interaction as suggested by a less steep slope probably due to presence of a large proportion of very fine particles which tend to cause uneven resin distribution [12]. Despite a significant improvement in the interfacial bond strength of the composite board after crushing the rice husks, the maximum value obtained (0.2 MPa) was still less than the minimum value (0.34 MPa) recommended in BS 5669 Part 2 British Standard, 1989 [14] as shown in Table 2. This suggests that crushing the particles could improve the properties of the resulting rice husk panel if it is accompanied by a removal of the very fine particles from the husk-saw dust furnish.

Impact strength

Figure 4 shows the results for the impact strength test of the composite boards as a function of the board density. The results show a strong linear correlation ($R^2 \sim 0.9$) between the impact strengths and the board densities for the three composite boards produced (BRH, URH and URH + 30%SD). It can be deduced from these results that the increase in the impact strength with the board density could be attributed to the effect of uniform binding stresses occurring at the particles and binder interface when the boards are pressed at higher densities [4]. In other words the magnitude of the impact strength of the composite board depends on the strength of the interface bond. However, the magnitude of the bonding stresses induced is largely dependent on the magnitude of the counter strains as suggested by the differences in the three curves. Higher spring backs caused by the post-stressed particles probably affected the binding stresses in some composite boards, which led to lower values of the impact strength in those boards. The maximum values of the falling ball impact strengths of the rice husk composite boards were much greater than the minimum value (34-mm/mm of the board thickness) recommended in the British Standard [14].

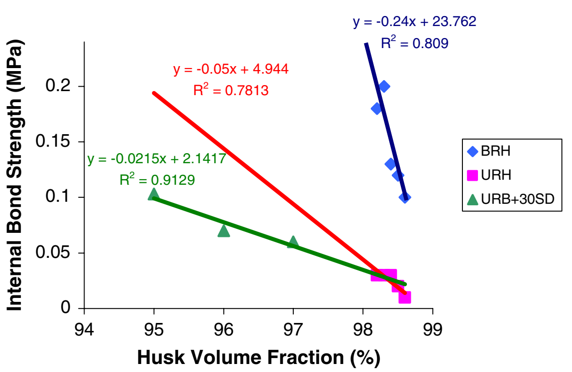

Fig. 3 IBS as a function of rice husks volume fraction: BRH = broken rice husk; URH = unbroken rice husks and URB + 30 SD = Unbroken rice husk with 30% saw dust addition

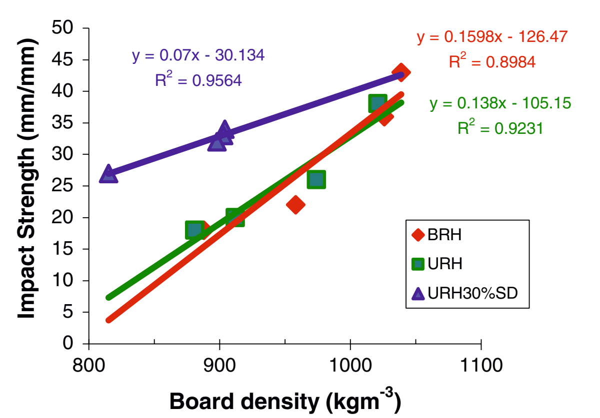

Fig. 4 Impact strength as a function of board density for broken rice husk (BRH), unbroken rice husk (URH) and unbroken rice husks with 30% saw dust (URH + 30%SD)

Table 2 Mechanical properties of different composite boards

Composite Composition	IS mm/mm board thickness	MOR (Mpa)	MOE (Mpa)	IB Strength (Mpa)
HT-CNSL-sawdust (30%) -rice husks composite panels	34	3.40	989	–
HT-CNSL-uncrushed RH composite panels	36	5.41	1039	0.041
HT-CNSL-crushed RH composite panels	43	7.00	1527	0.200
BS 5669: 1989 (Part 2)	34	13.00	2500	0.340

Note: IB means internal bond, RH means rice husks, MOR means Bending Strength, MOE means Bending Stiffness and IS means Impact Strength

Flexural properties

Figures 5 and 6 show the results of the three-point bending test namely the bending strength (MOR) and stiffness (MOE) as a function of the board density. The results in Fig. 5 show no significance differences in the value of MOR for the three composite boards. There is also no strong linear correlation between the MOR and MOE as the board density is varied. High values of the MOR and MOE observed at higher panel densities could also be attributed to the effect of bonding stresses at the particle-binder interface. The differences seen could be attributed to the differences in the magnitudes of the interface bonds formed in the three materials. The values of the MOR and MOE obtained were lower that the minimum values recommended in the British Standard [14] for general-purpose panel boards.

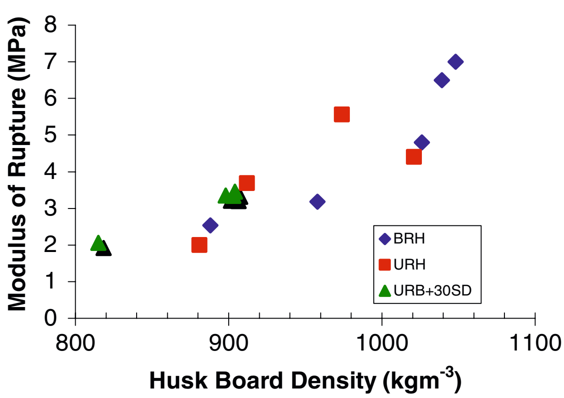

Fig. 5 MOR as a function of board density

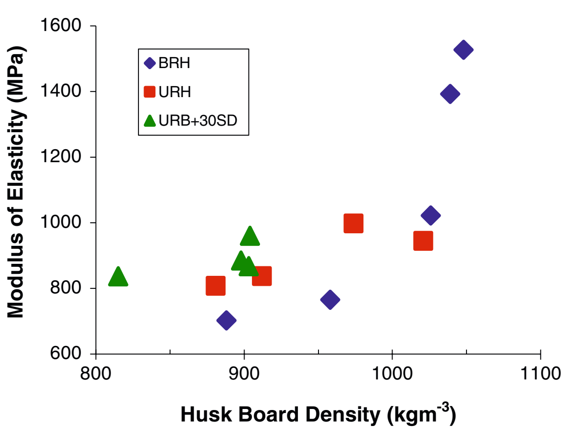

Fig. 6 MOE as a function of board density

Conclusion

The results have indicated that it is possible to produce composite boards from a combination of rice husks and a tannin-based resin, although most of the properties investigated so far fall short of those stipulated in BS 5669. It was only the impact strength which was able to reach or exceed the minimum stipulated values in BS 5669. To successfully achieve the minimum requirements or exceed them, the interfacial bond strength between the components in the rice husk—tannin based resin board must be increased.

This study has revealed that the interfacial bond strength between rice husks particles and a tannin-based resin can be improved by increasing the interaction between rice husks particles and tannin. This interaction can be improved by:

(a) modifying the rice husks particles (crushing and removal of fine particles from the particle furnish),
(b) increasing the tannin resin content or the densities of the composite board and
(c) modifying the adhesive properties of tannin-based resin (reducing the moisture content, and increasing its physical and chemical interaction with the rice husks particles).

However, these improvement methods should be accompanied with a balanced understanding of their effects on the processing parameters, the short term and long term composite boards' behaviour and other particle board properties like screw and nail withdrawal as well termite and fungal resistance.

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