Some opportunities and challenges of producing bio-composites from non-wood residues

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Abstract Plant-based composites (bio-composites) may in the future, become materials to replace polymer based composites and wood in terms of their attractive specific properties, lower cost, simple processing technologies, eco-friendliness, and ability to be recycled after use. The quality and performance of plant fibre-based composites can further be improved by adopting appropriate engineering techniques. Although plant-based fibres have these advantages, they also have some limitations. One of the serious problems of plant fibres is their strong polar character, which creates many problems of incompatibility with most thermosetting and thermoplastic matrices. Production of bio-composites with high quality and performance is therefore based on adjusting the properties of the constituents to meet the requirements of the composite material i.e. a product with consistent, uniform, predictable, and reproducible properties. Such adjustments involve creating strong interfacial bonds between the lignocellulosic substrates and the binder. Successful development of bio-composites therefore stem from a careful understanding of the influence of these adjustments on the composite properties. This paper suggests some opportunities available in producing bio-composites from non-wood resources, and the challenges that must be overcome to make this technology commercially viable. Gaps in knowledge and information required before full commercialisation of these materials are identified.

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Introduction

The social and economic developments of humans are largely dependent on better utilisation of available resources. From ancient times, *Homo sapiens* have made great use of wood-based products as the most available resource. However, increased environmental consciousness particularly on recycling of traditional materials, unprecedented forest resource degeneration, and global warming, have led to world-wide efforts to develop bio-composites from non-wood resources.

Environmentally sustainable lignocellulosic resources are available in different forms of non-wood based fibres and agricultural residues. Non-wood commercial fibres include jute, sisal, kapok, kenaf, flax, hemp, ramie, etc. Agriculture residues include stalks of most cereal crops, rice husks, coconut fibres (coir, bagasse, maize cobs, peanut shells, and other wastes. Man has a long history of using non-wood lignocellulosic resources for producing composites. Its beginning can be traced back during the ancient times when the Egyptians added straw to mud to make bricks which have proved to be very strong and durable. Today, man-made lignocellulosic composites are becoming attractive in both commercial and non-commercial applications.

Most of the recent developments of bio-composites from non-woody lignocellulosic resources have been aimed at improving the quality and performance of the products. However, apart from attaining these core attributes, the composites produced from these resources offered several other advantages, among them being the achievement of improved mechanical properties [1].

Composites produced from non-wood lignocellulosic resources can be categorised based on the geometry of the non-wood lignocellulosics. Those made from lignocellulosic

particles are generally referred to as particleboards (BS 5669: Part 2, 1989) while those that are made from ligno-cellulosic fibres are called fibreboards or fibre reinforced composites. The type and form of the non-wood lignocell-ulosics used for composite manufacture determine the properties and quality of the composite produced. Properties of the particle-based composites tend to vary considerably throughout the product surface, primarily due to inconsistent properties of the particles. However, the properties of the fibre-based composites have better consis-tency and are more predictable, an attribute highly desir-able for any structural material. Adjustments of the properties of lignocellulosic reinforcements/fillers have been carried out purposefully to improve the bio-composite properties.

Since most of lignocellulosics used for composite manufacture exhibit inconsistent and unpredictable prop-erties, they therefore lack the quality of being regarded as materials. The properties of lignocellulosics vary depend-ing on several factors: among these are agricultural practice and season, extraction method, treatment method, etc [2]. When lignocellulosics are directly used for composite manufacture, they tend to transmit these weaknesses to the composite produced. It is most often found that the com-posite produced also have inconsistent and unpredictable properties. In order to produce a composite with consistent and predictable properties the constituting components (lignocellulosics and binder) must be adjusted to be con-sistent with the requirements of the ''composite material''. Achieving this goal is often not a straightforward under-taking. However, by understanding the properties and requirements of the individual components of the com-posite, the designer is able to improve the quality of the composite product. This is why it is important to first characterise and understand the properties of the constitu-ent components of the composite before producing the composite.

Properties of the non-wood lignocelulosics

Wood and non-wood fibres and particles are all lignocell-ulosics that are three-dimensional composites, principally composed of macromolecules of cellulose, hemicelluloses, and lignin. The three polymeric components, which occur in the cell wall of lignocellulosics, determine most of the properties of lignocellulosics [3]. The crystalline cellulose acts as the reinforcement while lignin predominantly acts as a matrix, making the cell wall matter a composite in nature.

Although lignocellulosics differ in chemical composi-tion from one another as shown in Table 1, their charac-teristics are similar. All lignocellulosics have a tendency to

swell and shrink when external moisture changes below the point of saturation of moisture in fibres. The presence of the hydrophilic OH$^-$ groups in the structure of the cell wall polymers of the lignocellulosic components make the lignocellulosics more susceptible to hydrolytic degradation when exposed to moist environment. Lignocellulosics burn easily when are in contact with fire and degrade under acidic, basic, and ultra violet radiation conditions. In order to successfully develop composite materials from lignocellulosic substances we must first consider these deficiencies to come up with an appropriate modifica-tion scheme for the composite material manufacture requirements.

Table 2 shows the susceptibility of each type of the cell wall polymer components on various environmental deg-radation mechanisms. Lignocellulosics with high amounts of crystallinity have better resistance to UV while high lignin content in these materials gives resistance to bio-degradation and thermal degradation.

Properties of adhesive binders or matrix materials

Adhesive binders are materials that connect one substrate to another in the composite. Binders must be hydrophobic substances which when mixed with hydrophilic lignocell-ulosics help to shield them from the aggressive environ-ments including moisture. The most common binders used for binding lignocellulosic substances are synthetic phenol formaldehyde (PF), urea formaldehyde (UF), and diphenyl-methane di-isocyanate (MDI). Recently, natural binders like tannin and cashew nut shell liquid (CNSL) have been successfully used [4, 5]. In joining one substrate to another

Table 1 Chemical composition of some plant fibres

Fibre type	Cellulose (%)	Lignin (%)	Pentosan (%)	Ash (%)	Silica (%)
Rice	28–48	12–16	23–28	15–20	9–14
Wheat	29–51	16–21	26–32	4.5–9	3–7
Oat	31–48	16–19	27–30	2–5	0.5–4
Bagasse	32–48	19–24	27–23	1.5–5	0.7–3.5
Bamboo	26–43	21–31	15–26	1.7–5	0.7
Seed flax	43–47	21–23	24–26	5	—
Kenaf	44–57	15–19	22–23	2–5	—
Jute	45–63	21–26	18–21	0.5–2	—
Hemp	57–77	9–13	14–17	0.8	—
Ramie	87–91	—	5–8	—	—
Kenaf	37–49	15–21	18–24	2–4	—
Jute	41–48	21–24	18–22	0.8	—
Abaca	56–63	7–9	15–17	1–3	—
Sisal	43–62	7–9	21–24	0.6–1	—
Cotton	85–96	0.7–1.6	1–3	0.8–2	—
Coniferous	40–45	26–34	7–14	< 1	—
Deciduous	38–49	23–30	19–26	< 1	—

Source: [2]

Table 2 The order of degradation of different cell wall polymer components

Fastest biodegradation	Fastest ultra violet degradation	Fastest hydrolytic degradation	Fastest thermal degradation
Hemicelluloses	Lignin	Hemicelluloses	Hemicelluloses
Accessible cellulose	Hemicelluloses	Accessible cellulose	Cellulose
Non-crystalline cellulose	Accessible cellulose	Non-crystalline cellulose	
Crystalline cellulose	Non-crystalline cellulose	Lignin	
Lignin	Crystalline cellulose	Crystalline cellulose	Lignin

Source: [3, 23]

in the composite, binders must form interfacial bonds with lignocellulosic substrates. The degree of interfacial bonding between these substances varies depending on the compatibility of the binders to the surface properties of lignocellulosics.

There are several factors that influence the interfacial bonding:

(i) Chemical affinity between the polymer binder and the lignocellulosic fibre,
(ii) The surface condition of the lignocellulosic fibre,
(iii) Processing parameters especially pressure and temperature.

The mechanisms of interfacial bonding and the measures to improve it have been subjects of much interest due to the returns expected in terms of consistent material properties. The interface controls the composite material properties because this is where stress is transferred from fibre to fibre through the matrix. Processing of bio-composite has therefore focused on ways to facilitate formation of efficient mechanical and chemical bonds.

Potentials of the non-wood biocomposites

Wood-based materials are the most widely used and there is little doubt that they will continue to dominate as solid wood and composite panels in terms of market and quality. But the questions are, for how long will the wood-based materials survive the consumption pressures? Which particular lignocellulosic should be used? The answers depend on the change of pattern of use for the available fibre resources shown in Table 3.

Bolton [6] outlined that there are different factors, which influence the industrial pattern use of different fibres worldwide. He pointed out that some of these factors may be economical, political, social, and technological. However, the use patterns of lignocellulosic fibres vary from one country to another depending on which of these factors is significant. The mismatch between social and economic development in different parts of the world, influences the amount of fibre consumption and demands. However, the world consumption of wood fibres as illustrated in Table 3,

suggests that there is a need to diversify into other non-wood fibres. With dwindling forest reserves and imminent desertification of many tropical countries, it is very likely that non-wood fibres will become potential fibre resources to supplement wood-based fibres in the future.

Plant based fibres have long been used as cost-cutting fillers in the plastics industry. Recently there is growing interest in using plant fibre reinforced composites as replacement to glass fibre reinforced composites in some parts such as car interior panels, etc.

There are numerous advantages of using non-wood lignocellulosics for composite manufacture. They include their ease to process into composite, which requires relatively low energy input. The low density of these residues is an added advantage to the weight reduction of the produced composite in terms of high specific properties, which are comparable to some synthetic fibres [1] as shown in Fig. 1. The other most outstanding characteristic of bio-composites is their ability to be recycled without causing potential threat to the environment. However, although plant based fibres have these advantages over glass fibre, they are not totally free of problems. A serious problem of the natural fibres is their strong polar character, which creates many problems of incompatibility with many thermoplastic matrices, especially polyolefins. Surface treatments, although having a negative impact on economics, can potentially help to overcome the problem of incompatibility [7, 8].

Table 3 Annual world production of selected fibres

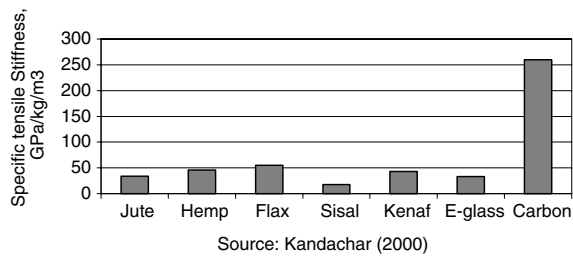| Fibre	Quantity ($\times 10^3$ tonnes)	% of the total
Wood fibre	1,750,000	68.5
Rice straw	700,000	27.4
Rice husks	70,000	2.8
Cotton	18,645	0.75
Bamboo	10,000	0.39
Jute	3,630	0.14
Kenaf	970	0.04
Flax	830	0.03
Sisal	380	0.01
Hemp	220	0.009
Ramie	110	0.004
Coir	100	0.0039

Sources: [1, 6]

Fig. 1 Comparison of the specific tensile properties of some natural and synthetic fibres

Designing of biocomposites from non-wood particles

When we speak of man-made bio-composites we refer to a combination of lignocellulosic fibres/particles of various sizes and shapes held together in a matrix of primarily natural binder or sometimes synthetic binders. However, when we need to produce a composite that has consistent and predictable properties, we must first consider several underlying factors. These factors include the following:

Physical properties

The most important physical properties of the non-wood particles include the particle size and particle size distribution, particle shape, fibre length, and moisture content. For example, the particle size and shape differentiates the quality and type of the particleboards.

Very small particles increase furnish surface area and absorb more resin than larger particles. A mixture of the fine and large particles should therefore be avoided during blending as they can lead to a disproportionate absorption of the resin [9], poor resin distribution and incidental flaws in the composite. However, oversized particles tend to subject the composite to poor quality, i.e. increase flaws in the product due to large inter-particle distances. Smaller particles give smooth surface texture and have smaller inter-particle distances. It is based on this fact that smaller particles are normally used for surfacing while larger particles are for the core.

The particle geometry is a very important physical property that has a remarkable contribution to the properties of the composite. Flaked particles (high flake length) have been found to have significant influence on the maximum strength of the composite. Similar findings have been published for particles with high aspect and slenderness ratio [10].

Modified physical properties

Generally, for the purpose of producing a ''composite material'' with uniform, consistent, and predictable prop-

erties, lignocellulosics are reduced to small and relatively consistent sizes and shapes, so that the negative effects of differences in morphology are minimised [11]. Size reduction in lignocellulosics is accomplished by chemical or mechanical modification, or by steam treatment. The choice of the method depends on the geometry and composition of the lignocellulosics. For example, rice husks modification might be slightly different from wood particles because of their differences in physical and chemical properties. Studies conducted on rice husks particleboard development have revealed that addition of sawdust in the mixture improves the mechanical and physical properties of the particleboard [12]. Similar studies have demonstrated that crushing of rice husks increases the particleboard performance though at the expense of high resin absorption [12–14]. These findings suggest that it is not easy to quantify the optimum particle size required for all lignocellulosics except for individual materials. Chemical modification has been primarily employed to increase the hydrolytic stability of lignocellulosics, and in some cases, chemical modification also makes the surface reactive to binders.

Compatibility with binders

In order for the particle and matrix to perform their functions in the composite, they must be joined together through the interfacial bond. In order to accomplish this, the particle surfaces must be chemically compatible with the matrix, so as to allow formation of a strong chemical bond. However, the tensile and shear strengths of the matrix must be good enough to facilitate the transfer of the applied stress from one particle to another through an interfacial bond [9].

The interfacial bonding starts when the liquid resin is dropped on the solid surface of the substrate [15] and it continues to develop until it reaches the maximum strength after the matrix has cured. The amount of adhesive bond formed depends on the amount of resin used, resin viscosity and flowability, resin pH, and the surface wettability, particle surface roughness and compatibility with the resin. Resin penetration is a function of resin viscosity, chemical compatibility of the resin with the particle surface, and physical condition of the particle surface. The chemical compatibility of the resin with the particle is a function of several aspects. One of these features is the resin pH, which has a greater contribution on the interfacial bonding. UF resins prepared under alkaline condition have been noted to perform poorly in lignocellulosic composites [16]. When phenolic resins prepared under acidic conditions [17] are used to bind lignocellulosic substrates they enhance the reaction between phenol and lignin present on the substrate

surface. The implication is that phenol is covalently bonded to lignin [18]. In order to achieve better interaction, the binder should also be physically compatible with the particle surfaces. The resin can flow over the substrate surface if its surface tension is less than that of the substrate. It has been established that the rough surfaces introduced on the particle tend to create chemically reactive crevices [15]. This is also some form of mechanical interlock. The chemically reactive crevices formed there on increase the likelihood of the particle and resin interaction. The implication is that the physical properties of the particle surface have significant contribution to the interface bonding in terms of the chemical and physical compatibility with the resin.

Dinwoodie [9] later redefined the requirements of the particles and matrix by taking into account the properties of composites that depends on the type of substrates and binder used as well as the interaction between them. He noted that excessive particles overhang might lead to poor strength if the interface bond is confined to a small portion between two particles. The binder should cover as much surface area of the particles overlaps so that it can resist the internal stresses induced in it as the particles move in response to moisture stress changes. And the surface chemistry and topography should facilitate the formation of strong interface bonding. Above all the binder must be able to penetrate the particle cell wall to be able to produce stronger bonds.

In addition to the particle topography and chemistry, addition of more resin improves its distribution within the substrates and hence resulting into better interaction. This however has twofold implications, cost and performance. Increasing the amount of resin is an added cost that increases the cost to the final product [12] especially when high cost synthetic binders are used. For the case of high furnish phenolic resins, increasing the amount of resin increases steam evaporation, which is destructive to the curing interfacial bond during hot-pressing. Another important aspect affecting the composite quality are the restrictions imposed on the use of some conventional binders. Use of methyl di-isocyanate (MDI) and UF are somewhat restricted due to occupational health concerns and environmental pollution. Isocyanate contained in MDI has been associated with occupational bronchial asthma and death to people working in plastic industries that use MDI. On the other hand, restriction of UF is based on its continuous emission of toxic formaldehyde during the lifetime of the composite bound by this resin. The performance of UF resin in lignocellulosic composites depends on the molar ratio of formaldehyde to urea. However, the CH_2–NH bond in the UF resin is easily hydrolysable by water. Therefore, increasing the amount of UF resin in the composite without increasing the molar ratio of

formaldehyde to urea greater than 1 does not improve the stability of UF resin in water. Some studies are being carried out to develop natural resins that are formaldehyde free and less toxic. Cimteclab in Italy is developing CNSL resin by cardanol functionalisation [19]. The Biocomposite Centre at the University of Wales is also working on a similar project.

Adjustments of the adhesive binders

Similar to lignocellulosics, modification of the properties of adhesive binders should also take into account the type of the binder, type of the end product, the mixing method employed, and the required properties of the end product, amongst others.

The most important adhesive properties of the binder which improve the interfacial bonding between it and the substrate includes: viscosity, pH, pot and shelf lives, penetration, wetability, etc [20, 21].

Processing parameters

The combination of the interaction between the material properties and the processing parameters complicates further the prediction of the quality and performance of the composite. Phenolic resins, for example, are high moisture furnish resins. They must be pressed at high pressures and high temperatures for longer periods in order for the curing process to be completed. If we also consider the low densities of the particles, apart from being beneficial to the composite in terms of the specific properties, they also disrupt the interface bond strength. Low-density particles require high compaction in order to reduce the porosity. A combination of the locked-in internal stresses with an evaporating steam from high moisture furnish resins leads to a catastrophic delamination of interface bonds. This may be more devastating when resins of poor wet strengths such as UF and unfortified tannin are used. Therefore an optimum amount of moisture in the particle–resin mixture needs to be established for an effective hot pressing process. A certain amount of moisture in the particles is important to keep them flexible and non-brittle. Moisture in the particle–resin mixture assist in heat transfer from the surface of platen to the board core during hot pressing [12] by means of gas and vapour pressures formation. However, excess moisture in the mixture can be very destructive to the composite interfacial bonds due to excessive steam generation during hot pressing which leads to catastrophic delamination of the interface bonds [12, 22]. Methods for chemical modification of agricultural fibres are being perfected [23], and are the main focus for researchers in this area.

Gaps hindering commercialisation

In order to be able to produce non-wood based composites having high quality we require the following minimum information.

1. Database on the physical properties of various non-woody residues such the optimum particle size and shape, the optimum moisture content, density, particle orientation, and surface topography.
2. Database for the mechanical properties of the non-woody residues and matrix such as the tensile strength, elongation, hardness, and stiffness.
3. Database for the availability and cost of raw materials.
4. Thermal properties of both the non-woody residues and matrix such as the glass transition, melting temperatures, relaxation, and the decomposition temperatures must be known to help in establishing the optimum processing and operating conditions of the composite.
5. The chemical properties of the substrates and binders, which have synergetic influence on the stability of these components and the quality of the resulting composite. Reaction of the composites when in contact with various environments must also be established and data made available.
6. The amount substrates, the optimum particle size, particle shape and orientation in relation to the direction of the property being measured, for optimum performance must be quantified.
7. The optimum amount of resin and conditions as well as type of resin that ensures resin economy and uniform distribution within the particles (critical resin weight fraction) must be quantified.
8. The processing parameters such as compaction ratio, pressure, press temperature and cycle that to a large extent affect the strength of interfacial bonds and rheological properties of the matrix and particles must be established.
9. Resistance to biodegradability and micro-organism attack of the composites need to be established, especially in tropical environments where microbiological activity is most active.

Other factors hindering the commercial application of non-wood fibre composites are:

(i) Relatively high cost of synthetic resins.
(ii) Lack of indigenous technology and capacity to produce composite processing equipment (presses, blenders, etc.) in developing countries.
(iii) Absence of technology entrepreneurs who can risk to venture in production of these new materials.

Conclusions

Non-wood lignocellulosics have potential to become future raw materials in bio-composite manufacture. However, their physical and chemical properties, which vary significantly, pose great challenges in composite development. It is possible to homogenise the properties of the composites through size reduction and chemical modification of the lignocellulosics.

There are still many gaps in information and knowledge of biocomposites from non-woody resources, which must be closed in order to stimulate commercial production of these novel materials.

References

1. Kandachar PV (2000) In: Proceedings of the International Workshop on the Development of natural Polymers and Composites in East Africa, Arusha Tanzania, September 2000, ICS-UNIDO pp 187–202
2. Rowell RM, Han JS, Rowell JS (2000) Natural polymers and agrofibres composites. San Carlos, Brazil, pp 115–134
3. Rowell RM (1983) Commonw Forest Bureau Oxford England 6(12):363
4. Bisanda ETN, Ansell MP (1992) J Mater Sci 27:1690
5. Ogola WO, Bisanda ETN, Tesha JV (2000) In: Proceedings of the 4th European Panel Products Symposium, Llandudno, Wales, UK, 11–13 October 2000, University of Wales, pp 175–185
6. Bolton AJ (1995) Outlook Agr 24(2):85
7. Zafeiropoulos NE, Williams DR, Baillie CA, Matthews FL (2002) Appl Sci Manuf 33(8):1083
8. Bisanda ETN, Ansell MP (1991) J Compos Sci Technol 41:164
9. Dinwoodie JM (1997) In: Proceedings of the 1st European Panel Products Symposium, Llandudno, Wales, 9–10 October 1997, University of Wales, pp 1–9
10. Nishimula T, Amin J, Ansell M (2001) In: Proceedings of the 5th European Panel Products Symposium, Llandudno, Wales, Oct. 2001, University of Wales, pp 155–166
11. English BP, Chow P, Bajwa DS (1996) In: Roger M, Raymond A, Young, Judith K Rowell (eds) Paper and composites from agro-based resources. Lewis Publishers, NY, pp 269–299
12. Ndazi B (2001) M.Sc. Thesis, University of Dar es Salaam
13. Vasishth RC (1971) In: Interregional Seminar on Industrial Processing of Rice, UNIDO Document. ID/WG/89/23 Joint UNIDO, FAO/ECAFE
14. Vasishth RC (1974) In: Assignee: Cor Tech Research Ltd. Patent, P.N. US 3850677, I.D.: **741126**
15. Houwink R, Salomon G (1965) Adhesion and adhesives, 2nd edn., vol 1, chapter 1. Elsevier Publishing Company
16. Gerardi V, Minellin F, Viggiano D (1998) Biomass Bioenerg 4(3):295
17. Ono HK, Inoue A (1994) In: Hse CY, Tomita B, Branham SJ (eds) Adhesives and bonded wood products. Forest Products Society Proceedings, No. 4735 pp 330–343

18. Bolton J (1997) In: The Burgess-Lane Memorial Lectureship in Forestry, March 5, 1997. www.forestry.ubc.ca/burgess/bolton.html

19. Ferri U (2000) In: Proceedings of the International Workshop on the Development of natural Polymers and Composites in East Africa, Arusha Tanzania, Sept. 2000, ICS-UNIDO, pp 43–61

20. Bisanda ETN, Ogola W, Tesha JV (2003) Cement Concrete Comp 25(6):593

21. Bisanda ETN (2001) J Appl Comp Mater 7:331

22. Dunky M (2000) In: Proceedings of the 4th European Panel Products Symposium. Llandudno, Wales, Oct. 2000, University of Wales, Bangor, pp 1–14

23. Rowell RM (1995) In: Proceedings of a Seminar on Research in Industrial Application of Nonfood Crops, I: plant fibers, May 1995, Copenhagen, Denmark. Denmark Academy of Technical Series, pp 49–70