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# Composite building materials from natural fibers/ agro-forest residues

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Interest in using renewable resources such as natural fibers and agro-forest residues as reinforcements/fillers in polymer matrices has grown considerably for making wood alternatives in view of their low cost, easy availability, saving in energy and pollution free-production. In this paper, an overview on the global perspective of natural fiber reinforced composites has been outlined with reference to their applications in buildings. Researches carried out on polymer composites based on natural fibers and agro-forest residues are briefly reviewed in terms of their physico-mechanical properties and performance characteristics. During fabrication, 30-50 wt % sisal/jute fibers in laminates/sheets, 50-60 wt % rice husk in extruded/injection moldings and ~95 wt % pine needles in boards have been used. The sandwich composites used in the doors ( jute laminate face, and polyester-urethane foam core) have a density of 500-610 kg/m<sup>3</sup> and exhibited ~1.5% weight loss when exposed to termites according to ASTM D 3345. Based on the results, sisal/jute composite panels/ door shutters, shuttering plates, rice husk/plastic profile frames and pine needle composite boards have been successfully manufactured at pilot plant scale. Techno-economics and sustainability issues related to these composites have also been discussed in order to develop confidence among users and entrepreneurs. A comparative assessment between the properties of these newly developed products and commercial specification gives a clear guideline for users about their suitability in large scale adaptation in practice.

Keywords: Natural fiber composites, Agro-forest residues, Rice husk/plastic profile frame, Pine needle composite boards, Wood substitutes

## **1** Introduction

The utilization of renewable raw materials such as natural fibers, agro-residues and leaves and stalks of forest plants has been widely recognized in making reconstituted ligno-cellulosic panel products for building industry<sup>1-4</sup>. These types of industrial products can provide suitable wood alternatives, particularly in the face of universal concern for ecological degradation caused by continuing dependence on depleting forest resources. Since natural fibers/agroforest residues are inexpensive reinforcements, their use with polymer matrices would be to provide composite materials that can attain sustainability in terms of their attractive attributes of recyclability, biodegradability and environmental acceptability<sup>5</sup>. These composites are economically cheaper than wood and also to glass fiber composites and could replace them in applications where cost consideration outweighs strength requirement<sup>6-7</sup>. Although, natural fiber/agro residue based composites are gaining interest, the challenge is to produce composites of requisite strength & stiffness, dimensional stability against moisture attacks and non-vulnerability towards other environmental agents for real world applications. Thus, it is essential to adopt three corner approaches of cost-effective surface treatment of natural fibers/agro-forest residues, resin matrix modification through functionalization and blending and selection of appropriate processing techniques to obtain composites of desired properties.

# 2 Natural Fibers and Agro-Forest Residues

Wood fibers and flours have been the traditional source for plastic composites. In view of diminishing forest resources and limitation on enlarging man made forests, non-wood fiber sources have gained importance<sup>1</sup>. Although vast amount of these materials are produced every year (~400 million tonnes), the actual availability of these resources in a condition that can be used by the industry remains a question to many. They vary from essentially forest based sources like bamboo, grasses etc. to agricultural residues such as cereal straws, bagasse, cotton stalks etc. However, the technical characteristics of these materials may be

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more limiting due to presence of high proportion of extractives and hemicellulose that are heat labile particularly their use in melt blend processing. Rice hulls represent one of the useful agricultural processing residues of interest for bio-composites<sup>8</sup>. The hulls contain ~20% silica making them somewhat unique among agricultural residues in melt blending composite processing.

Natural fibers such as jute, sisal, coir, hemp etc. are considered as naturally occurring composites consisting of helically wound cellulose microfibrils in a matrix of lignin and hemicellulose<sup>9</sup>. Presently, India produces ~ 6 million tons of natural fibers annually compared to the world production of ~ 25 million tons<sup>2</sup>. The physico-mechanical and chemical properties of some natural fibers are given in Table 1.

The strength of these fibers varies from plant to plant depending on their origin, physico-chemical and structural properties such as crystalline cellulose content, spiral angle, cell number and cell aspect ratio<sup>10-11</sup>. It is reported that the tensile strength as well as young modulus of natural fibers is lower than that of E-glass fibers commonly used in composites <sup>6,12</sup>. However, the density of E-glass is high (~ 2.5 g/cc) while that of natural fibers is much lower (~1.4 g/cc). This becomes particularly important when the weight of structure needs to be reduced. In view of the low cost of natural fibers, their low density, high elastic modulus and low elongation at break make them an attractive alternative reinforcement to the more expensive glass fibers in polymer composites. However, these advantages have not been fully exploited in the reinforced composites because natural fibers absorb a great deal of moisture and also have poor wettability with the polymer matrix resins<sup>13</sup>. It is, therefore, necessary to modify the fiber surface to render it more hydrophobic and also more compatible with polymer matrices<sup>14</sup>.

# **3 Natural Fiber Composites in Buildings**

Considerable attempts<sup>15-24</sup> have been made on the development of composite materials using jute and allied fibers and polymer matrices such as unsaturated polyester, phenolic, epoxy and alike. The properties of these composites depend on the properties of constituent phases such as fiber, resin and fibermatrix interface <sup>25</sup>. The variation of certain of these parameters yields composites with poor properties. To overcome the drawbacks of moisture absorption and wettability, natural fibers are pre-treated with alkali, coupling agents, isocyanates, plasma, radiation etc.<sup>4,14,26-28</sup>. This results in improved physicomechanical properties and hydro-thermal stability to the resultant composites. Recently, Liu et al.<sup>29</sup> reported a decrease in surface flaws when nano-silica is deposited on to the surface of jute fibers via in-situ synthesis. The surface free energy and tensile strength of these pretreated jute fibers are increased by 11.9% and 17.9%, respectively in comparison to the parent acid/alkali treated jute fibers. The thermal stability

| Table 1 —                                       | - Physical, chemical and | mechanical prope | erties of some natur | al fibers <sup>10</sup> . |           |
|-------------------------------------------------|--------------------------|------------------|----------------------|---------------------------|-----------|
| Property                                        | Jute                     | Banana           | Sisal                | Pineapple                 | Coir      |
|                                                 |                          | (a) Physical     |                      |                           |           |
| Diameter (µm)                                   | 200                      | 80-250           | 50-250               | 20-80                     | 100-450   |
| Fiber length (mm)                               | 180-800                  | -                | 50-150               | -                         | 50-350    |
| Specific gravity                                | 1.02-1.04                | 1.04             | 0.70-0.80            | -                         | 0.89-1.11 |
| Bulk density (Kg/m <sup>3</sup> )               | 120-140                  | -                | 90-160               | -                         | 145-280   |
| Moisture content (%)                            | 12.6                     | -                | 11.0                 | -                         | 8.0       |
| Water absorption (%) (24h)                      | 25-40                    | -                | 60-70                | -                         | 130-180   |
|                                                 |                          | (b) Chemical     |                      |                           |           |
| Cellulose content (%)                           | 61                       | 65               | 67                   | 81                        | 43        |
| Lignin content (%)                              | 12                       | 5                | 12                   | 12                        | 45        |
| Hemicellulose (%)                               | 16.8                     | 16.0             | 17.6                 | 13.3                      | 20.2      |
| Others                                          | 9.62                     | 12.31            | 2.37                 | 3.04                      | 2.63      |
|                                                 | (0                       | c) Mechanical    |                      |                           |           |
| Ultimate tensile strength (MN m <sup>-2</sup> ) | 240                      | 529-754          | 270-350              | 413-1627                  | 131-175   |
| Elongation (%)                                  | 1-1.2                    | 1.0-3.5          | 3-7                  | 0.8-1.6                   | 15-40     |
| Elastic modulus (GN m <sup>-2</sup> )           | -                        | 8-20             | 9-16                 | 34-62                     | 4-6       |
| Tenacity (MN m <sup>-2</sup> )                  | 440-533                  | 529-754          | 568-640              | 413-1627                  | 131-173   |

of treated jute fibers is also improved significantly due to existence of nano-silica on their surface. Fuentes et al.<sup>30</sup> in their studies concluded that moisture content in the environment has limited effect on the fiber-matrix interface during manufacturing of flax/polyester composites. However, the longitudinal composite strength decreased somewhat for composites produced in humid conditions. Anawan et al.<sup>31</sup> reported characterization of wettability and interfacial properties of kenaf fiber and its unsaturated polyester composites fabricated by resin transfer molding. The surface free energy of fibers is found to increase with longer soaking time in 6% NaOH solution. The highest value of inter-laminar shear strength is recorded for 3 h treated kenaf composites. Pereria et al.<sup>32</sup> studied charpy impact tenacity of epoxy matrix reinforced with aligned jute fibers (30 vol%). They observed a significant increase in the absorbed impact energy with increasing volume fraction of jute fibers. Adhikari *et al.*<sup>33</sup> studied mechanical properties of metal oxide dispersed jute fiber reinforced polyester composites. They selected 20 wt% filler content (18% jute and 2%  $ZrO_2/Al_2O_3$ ) as optimized composition. The results indicate that ZrO<sub>2</sub> particle incorporated biocomposites show better mechanical properties than the Al<sub>2</sub>O<sub>3</sub> dispersed biocomposites. Biswas et al.<sup>34</sup> studied influence of ZrO<sub>2</sub> incorporation on the properties of sisal/polyester composites. They found that optimum mechanical properties are obtained at 35 wt% filler content.

Investigations on natural fiber composites 6-7,12,35-36 revealed that their properties can be better utilized in hybrid composites wherein natural fibers are used as core materials with glass fibers as outer skins. The glass skins not only enhance the mechanical properties but also protect the natural fiber composites from weathering effect. Hashemi et al.<sup>37</sup> fabricated six different hybrid composites with different kenaf-glass fiber ratios using pultrusion technique. They reported that radial configuration of kenaf yarns in the core of composites and bilateral configuration of glass fibers in the shell region indicated a noticeable effect on the static flexural modulus, strength and strain at break. The strength properties of these hybrid composites are considerably less than that of the glass fiber composites (Table 2). However, the cost factor makes this composite more attractive in buildings for nonstructural uses.

The construction of primary school buildings in Bangladesh<sup>38</sup> and the fabrication of low cost housing

| Table 2 — Mechanical properties of some natural/glass fiber/unsaturated polyester composites <sup>11</sup> . |                   |                               |                              |                              | /glass                      |
|--------------------------------------------------------------------------------------------------------------|-------------------|-------------------------------|------------------------------|------------------------------|-----------------------------|
| Hybrid<br>composite                                                                                          | Density<br>(g/cc) | Flexural<br>strength<br>(MPa) | Flexural<br>modulus<br>(GPa) | Tensile<br>strength<br>(MPa) | Tensile<br>modulus<br>(GPa) |
| Jute-25%                                                                                                     | 1.23              | 121                           | 7.69                         | 62                           | 1.46                        |
| Glass-7%                                                                                                     |                   |                               |                              |                              |                             |
| Sisal-46%                                                                                                    | 1.20              | 89                            | 3.11                         | 65                           | 5.95                        |
| Glass-8%                                                                                                     |                   |                               |                              |                              |                             |
| Palm-27%                                                                                                     | 1.20              | 265                           | 13.00                        | 95                           | 3.23                        |
| Glass-7%                                                                                                     |                   |                               |                              |                              |                             |
| Coir-30%                                                                                                     | 1.28              | 161                           | 2.90                         | 32                           | 3.05                        |
| Glass-12%                                                                                                    |                   |                               |                              |                              |                             |
| Glass-30%                                                                                                    | 1.30              | 363                           | 16.00                        | 163                          | 26.70                       |
|                                                                                                              |                   |                               |                              |                              |                             |

unit prototypes and food grain silos using jute/polyester composites in India, the production of low cost building materials based on the composites of henequen / palm/ sisal fibers and polyester resin in Mexico<sup>39</sup>, installation of building panels and roofing sheets made from baggase/phenolic composites in Jamaica, Ghana and Philippines<sup>40</sup> and manufacturing of jute/polyester based items for low cost housing applications in Iran<sup>41</sup> have been widely recognized for the appropriate utilization of natural fibers in building industry. Currently, the interest has diverted to use latest composite molding techniques to make familiar with the response of natural fibers for the production of composite products. Rowell<sup>42</sup> reported the fabrication of composite boards using jute fibers and phenolic resin by steam stabilization technique. The physico-mechanical properties such as thickness swelling, water absorption and strength properties of these boards are reported to be adequate. Singh *et al.*<sup>43</sup> used pultrusion molding technique to produce profile sections utilizing jute fibers as reinforcement in unsaturated polyester/phenolic resin. These profiles are considered as framing materials for doors & windows alternatives to wooden frames in buildings. Saxena et al.<sup>44</sup> developed door shutters using jute, red mud/fly ash and unsaturated polyester resin and reported that these door shutters exhibited comparable properties as wooden door shutters. Efforts are also made to use resin transfer molding for making automotive panels using jute fibers and unsaturated polyester resin<sup>4</sup>. The results are quite encouraging in terms of productivity and cost-effectiveness.

The use of natural fibers in thermoplastics such as poly (vinyl chloride), high density polyethylene, polypropylene and polystyrene has also been explored for making panels by adopting either film stacking or melt impregnation process<sup>45-47</sup>. Nabila et al.<sup>48</sup> studied the effect of weight fraction of jute fibers on the properties of jute/polypropylene composites. At 40 wt% jute content, an increase of 19.7% in tensile strength, 79.8% in tensile modulus and 143% in heat distortion temperature is observed in comparison to parent polypropylene. The fiber pull-out is the prime mode of failure as observed in fracture surfaces of the specimens due to the poor fiber-matrix interface. Pererio et al.<sup>49</sup> used jute reinforcement in two types of matrices- polypropylene and polylactic acid for making eco-composites. An infra-red imaging device is used for on-line monitoring of specimens under impact or cyclic and quasi static bending. The results of thermal effect visualization coupled with mechanical stresses may be helpful to understand the reaction of materials to applied forces and their damaging modes. Da Luz et al.<sup>50</sup> studied hydrothermal carbonization of sisal fiber and its use in making sisal/recycled polypropylene composites. The addition of 20 wt% carbonized sisal fibers caused an increase of tensile and impact properties of composites. Chen et al.<sup>51</sup> reported the properties of polymer composites maximized at 70 wt% rice husk loading. As the rice husk content increased, water absorption, tensile strength and fire retardency of composites increased. The compatibility between the natural fibers and thermoplastic matrices and difficulty to achieve good dispersion of fibers in the matrix are reported to be improved through polymer grafting (maleic anhydride based polypropylene) onto the fiber surface 47. The chemical bonding between the grafted polymer and hydroxyl groups of natural fibers caused a better stress transfer from the matrix to the fiber. Inspite of a continuous thrust given on the research and development, natural fiber based products have not been exploited commercially to a large extent and their practical applications are still limited to low value products. Acceptability of these products in buildings requires fresh efforts to meet the requisite minimum performance that conform to the existing standard specifications.

# 4 Research and Development at CSIR-CBRI

A study was undertaken at the CSIR-Central Building Research Institute, Roorkee over the last few years to develop building materials based on the composites of natural fibers and agro-forest residues which could be cost-effective showing better performance in end use applications <sup>2,11,43,52-59</sup>.

Some of them being pursued currently are briefly described.

#### 4.1 Materials Development

#### 4.1.1 Sisal fibers

The use of sisal fiber - a leaf of Agave sislana ( physical properties: specific gravity 1.10; moisture conent 12.77; water absorption 123.73; tensile strength 280-370 MPa; elongation 3-5%; and modulus of elasticity 34-62 GPa; chemical composition: cellulose 66-78%; lignin 8-10%; hemicelluloses 10-12%; pectin; other materials 2-4%) as a reinforcement in unsaturated polyester resin has been investigated aiming at to mitigate some of their bottlenecks such as unsuitability for hand lay-up techniques, lack of good fiber-matrix interface and poor dimensional stability against hygro/hydrothermal conditions<sup>52-54</sup>. Solving the problem of hygroscopicness of sisal fibers. organophilites are introduced on their surfaces during chemical modification with coupling agents such as organosilane, organotitanate, organozirconate and Nsubstituted methacrylamide <sup>54</sup>. The efficacy of these coupling agents onto the sisal fibers indicates that Nsubstituted methacrylamide performs better as compared to other coupling agents in terms of its surface wettability. The adsorbed coupling agent is not only minimizing the further ingress of moisture into the fibers but also provides good coupling with the resin matrix. The polymer composites are fabricated from treated sisal fiber chopped stand mats and unsaturated polyester resin and evaluated their physico-mechanical properties as a function of fiber content, fiber aspect ratio, number of fiber plies and sisal/glass combination. The void contents of surface treated composites (6-13%) are lower than the untreated ones (16.11%). The optimum strength of laminates is noticed at the fiber aspect ratio of 323 used in the mat, ~ 50 wt % fiber content and four number of fiber plies. Under humid conditions (95% RH & 95% RH  $/50^{\circ}$ C), the reduction of 30-40% in the tensile strength is observed with respect to the unaged samples. However, the strength retention of surfacetreated sisal composites (except silane) is fairly high as compared with the untreated sisal composites. Microstructural examinations of failed untreated and treated sisal composites are also carried out to know the failure behavior in terms of fiber fracture/pull-out, debonding, resin yielding and fiber swelling. The results of this study can be successfully translated to other similar natural fibers. Based on these findings, an optimum system for preparation of composite panels has been developed.

# 4.1.2 Jute fibers

Jute composites are produced using alkali treated non-woven jute mats (weight  $350 \text{g/m}^2$ ; needle density 250 punches/cm<sup>2</sup>; 12 mm needle penetration; thickness 2 mm; jute: polypropylene ratio 80:20; jute mat content 45-47 wt %)<sup>55</sup> and modified polyester resins (unsaturated polyester-urethane hybrid polymer networks and polyester-polyurethane interpenetrating networks)<sup>56</sup>. The physico-mechanical polymer properties of these composites are given in Table 3. It is found that the water absorption and thickness swelling of jute composites based on hybrid polymer network matrix are reduced by ~50% and ~66% respectively when compared with the parent unsaturated polyester. An improvement of ~23% in the tensile strength is observed over the parent ones. When samples are aged in boiling water for 2 h, a decrease of 13-25% in the tensile strength and ~54% in the tensile modulus are observed. It is noted that the strength retention for hybrid polymer network matrix composites is more than the parent polyester matrix composites. It is believed that isocyanate terminal groups of the hybrid polymer networks react with the surface hydroxyl groups of jute fibers to form a chemical bond which could be responsible for such improvement.

The sandwich composites are constructed by casting in-situ unsaturated polyester-urethane foam core onto

| Table 3 — Physico-mechanical properties of unsaturated polyester and unsaturated polyester-urethane matrix jute composites <sup>56</sup> . |                            |                      |        |
|--------------------------------------------------------------------------------------------------------------------------------------------|----------------------------|----------------------|--------|
| Property                                                                                                                                   | Polyester matrix composite | IPN matrix composite |        |
| Density (g/cc)                                                                                                                             | 1.20                       | 1.24                 | 1.25   |
| Water absorption (%)<br>(24 h)                                                                                                             | 6.28                       | 5.51                 | 3.96   |
| Thickness swelling (%) (24 h)                                                                                                              | 4.61                       | 3.58                 | 1.5    |
| Tensile strength<br>(MPa)                                                                                                                  | 57.50                      | 66.60                | 71     |
| Elongation (%)                                                                                                                             | 5.86                       | 6.30                 | 5.40   |
| Tensile modulus<br>(MPa)                                                                                                                   | 1568                       | 1737                 | 1862   |
| Energy to break (J)                                                                                                                        | 4.16                       | 4.57                 | 4.2    |
| Toughness (MPa)                                                                                                                            | 25                         | 30                   | 29     |
| Flexural strength (MPa)                                                                                                                    | 70.40                      | 99.60                | 112.48 |
| Flexural modulus<br>(MPa)                                                                                                                  | 3634                       | 4597                 | 4750   |
| Impact strength (J/m <sup>2</sup> )                                                                                                        | 64                         | 70                   | 74     |

the jute laminate faces<sup>57</sup>. Two types of jute sandwich panels (13 mm and 23 mm) are produced and their physico-mechanical properties are given in Table 4. The density of samples lies in the range of 500-610 Kg/m<sup>3</sup>. The low value of thermal conductivity (~0.15-~0.18 W/mK) indicates that the composites have adequate thermal insulation. During tapping test, lack of de-bonded area in the panel supportives of good bonding between its face and core. Impact indentation values of the samples are negligible (< 0.2 mm) when 500 g of steel ball of 50 mm diameter is dropped on the sample from a height of 750 mm at various places. The flexural strengths of sandwich panels for 13 mm and 23 mm are ~15 MPa and ~32.50 MPa respectively while the edgewise compressive strengths of panels are ~26 MPa for 13 mm and ~19 MPa for 23 mm, respectively. The core shear strengths of panels for both thicknesses are 0.35 MPa and ~0.5 MPa. The Izod impact test of sandwich panels tested as per ASTM D 256 is  $\sim 21 \text{J/m}^2$ . The screw withdrawal load of panels is ~1500 N as against the specified value for wood (1000N) mentioned in the standard. These values on panels are adequate to resist against operational loads for use as panel insert/door shutter materials. Viewing the importance of the biological attacks, termite resistance and natural decay tests on the samples are also carried out as per ASTM D 3345-74 and ASTM D 2017-81, respectively. The composite samples (untreated and treated with copper chrome arsenic compound) are exposed to Microcerotermus bessoni termite colony in both accelerated lab and terrestrial conditions. After 2 months, the samples appear to be sound and free from termite tunneling. Only cling of termites onto the surfaces of samples is observed. The maximum weight loss in the samples is ~1.5 % only. All termites are survived at the end of exposure of samples. During natural decay in Aspergillus niger strain, the weight loss in the samples is  $\sim 1\%$  only when subjected

| Table 4 — Physico-mechanical<br>composites using hybrid polyr | 1 1 5          |          |  |
|---------------------------------------------------------------|----------------|----------|--|
| Property                                                      | Sandwich panel |          |  |
| Thickness (mm)                                                | 13             | 23       |  |
| Density (g/cc)                                                | 0.61           | 0.50     |  |
| Compressive strength (MPa)                                    |                |          |  |
| Edgewise                                                      | 26.2           | 19.10    |  |
| Flatwise                                                      | 105.23         | 66.85    |  |
| Bending strength (MPa)                                        | 32.51          | 14.73    |  |
| Bending modulus (MPa)                                         | 1451.16        | 13173.31 |  |
| Core shear strength (MPa)                                     | 0.51           | 0.35     |  |
| Flatwise tensile strength (MPa)                               | 4.30           | 1.50     |  |
| Impact strength (J/m)                                         | 21             | -        |  |

to potato dextrose agar / broth culture. The surfaces of samples are smooth and exhibiting no residual locus of fungus even after cleaning. It is concluded that the developed sandwich jute composites can be considered as potential building materials for wall panels, partition, door shutters, door panel insert etc.

## 4.1.3 Rice husk flour

Rice husk - an agro-processing residue (density 410 kg/m<sup>3</sup>, moisture content 8-10 %; cellulose 28-40%; hemicelluloses 16-21%; lignin 12-16%; ash 16-20%) is selected as a reinforcing material alternative to conventionally used wood fibers in the polymer melt blend composite processing because of its huge availability (~20 million tons/annum in India) and adequate heat stability due to high silica content (~20 wt%). Various issues such as poor compatibility with non-polar thermoplastics, hygroscopicness and difficulty of mixing in the conventional equipment have been properly addressed<sup>58</sup>, the compounding of recipes based on rice husk flour, polypropylene random copolymer and additives is made and rice husk flour loading in the recipe is optimized at a level of 50-60 wt%. The main idea behind the development is to provide plasto-elasticity in the homo polypropylene to make it carpenter friendly (screwable & nailable) and to improve interfacial strength without or minimum level of compatibilizer through processing. Steam exploded rice hull is used instead of mechanically milled unsplitted rice hull in melt blending to obtain adequate wetting and good dispersion in the matrix The tensile strength and tensile modulus of injection molding sheets are 16.30 MPa and 754 MPa, respectively. Dynamic mechanical spectra indicate that the storage modulus of samples decreases with the increase of temperature and become insignificant at tan delta peak (105°C). The softening point of molded sheet is  $\sim$ 74°C showing its satisfactory thermal stability. Fracture surface morphology of the tensile samples shows that rice husk flours are uniformly distributed in the polypropylene matrix. Matrix adherence on the surface of rice husk flour is also viewed showing good bonding between the rice husk and the polypropylene random matrix.

The injection molded sheets are aged under different humidity such 80% RH, 95% RH and 95% RH at 50°C and immersed water conditions for 30 days to know their dimensional stability. The weight gain in the exposed samples is ~1.5% only. The thickness swelling of exposed samples is

negligible. When samples are subjected for hydrothermal aging, the strength reduction in an accelerated water aging (2 h in boiling water) is 8-10% only while 15-20% strength loss is noticed under alternate wetting and drying cycle (72 h immersion in the cold water followed by 24 h in an air drying and then 72 h in an oven at 70° C: total 3 cycles). Termite resistance test is carried out in both laboratory and terrestrial conditions using Microcerotermus bessoni termite colony upto 9 months as per ASTM D 3345. It is found that the weight loss in samples is ~1% and ~1.5% at the end of 6 months and 9 months exposures respectively. No tunneling of termite on to the samples and its mortality in the culture are noticed. As per ASTM classification, the samples belong to Class I category (sound). Fire behavior of the samples is assessed in terms of their fire propagation index (BS: 476-part 6), smoke density (ASTM E 662) and rate of burning (ASTM D 635). It is found that the initial fire propagation index of samples is ~5.06 and overall index is ~16.97 only contrary to the kail wood index of ~16.14 and ~41.15. This shows that samples exhibited satisfactory behavior against growth of fire. The smoke density of samples is ~299.32 Ds in the flaming mode while ~261.20 Ds in the non-flaming mode. The rate of burning of samples tested as per ASTM D 635 is 25 mm/min. These results indicate that reaction to fire characteristics of samples is adequate for use in buildings. The comparative properties of rice husk/plastic profiles and natural wood are given in Table 5. Based on these results,

| Table 5 — Comparative properties of rice husk plastic profiles<br>and natural wood.          |                    |                           |  |
|----------------------------------------------------------------------------------------------|--------------------|---------------------------|--|
| Property                                                                                     | Rice husk profiles | Natural wood <sup>*</sup> |  |
| Density (g/cm <sup>3</sup> )                                                                 | 1.20               | 0.404-1.116               |  |
| Moisture content (%)                                                                         | 0.53               | 8-20                      |  |
| Water absorption (%)                                                                         | <1                 | -                         |  |
| Modulus of rupture (MPa)                                                                     | 30.66              | 12-26                     |  |
| Modulus of elasticity (GPa)                                                                  | 10.5               | 9-17                      |  |
| Compressive Strength (MPa)                                                                   |                    |                           |  |
| Parallel to grain                                                                            | 29.6               | 3.3-11                    |  |
| Perpendicular to grain                                                                       | 52.5               | 8-18                      |  |
| Screw withdrawl power (N)                                                                    |                    |                           |  |
| (min)                                                                                        |                    |                           |  |
| Parallel to grain                                                                            | 2327               | >2300                     |  |
| Perpendicular to grain                                                                       | 3273               | >2700                     |  |
| Nail holding power (N) x 102                                                                 | 2.7-4.3            | 2-10                      |  |
| Hardness (Rockwell A)                                                                        | 49                 | -                         |  |
| Flammability (rate of burning)                                                               | 25                 | -                         |  |
| ASTM D 635 (mm/min)                                                                          |                    |                           |  |
| *Safe permissible stress of structural timber, National Building<br>Code, 2005, sec 3 Timber |                    |                           |  |

rice husk flour can be considered as an alternative to wood fibers/particles in thermoplastic composites for making building profile panels.

#### 4.1.4 Pine needles

Pine needle - leaf of Pinus radiata (length 300-380 mm: density 160 kg/m<sup>3</sup>: 'moisture content 20%: absorption 45%; cellulose 40-43%; water hemicellulose 20-24%; lignin 36-40%; ash content 2-4%) is a renewable natural resource material produced annually in large quantity in the Western part of Himalayas (Indian forests ~ 2.7 million tones/annum). The most critical factor related to utilization is the existence of wax coating on the surface of pine needles which affects their bonding with the resin matrix. Because of their inefficient utilization, every year forest fire causes substantial damage to the flora and fauna of the region. Keeping this in view, a systematic study is initiated at the CSIR-CBRI on the utilization of pine needles for making building boards/panels<sup>59</sup>. Prior to use, the pine needles are treated with alkali (2% NaOH aqueous solution) to remove their outer waxy surface. The composite panels are produced from treated pine needles and isocyanate prepolymer on a hydraulic press at 140°C and 10 MPa pressure for 8 min retention. The physico-mechanical properties of these panels are evaluated as a function of pine needle treatments, resin adhesive content and pine needles/wood particle ratio. It is found that composite panels prepared at ~5% resin content gave best results. It is also noted that the boards made with pine needles blended with eucalyptus wood particles in 50-50 ratio exhibited superior properties to pine needle composite boards.

The performance of pine needle composite boards/panels is assessed for their dimensional stability, flammability, biological attacks and thermoacoustic behavior. At lower humidity, the pine needle composite boards/panels exhibited 2-7% thickness swelling at equilibrium moisture content whereas at higher humidity, the thickness swelling in the panels ranged between ~13% and ~23%. Under hydrothermal aging, the internal bond strength of panels is reduced by 41-67% in accelerated 2 h water aging and 54-78% in cyclic wetting/drying exposure respectively. Cone calorimetric results indicate that the optimum flammability characteristics of pine needle furnish is obtained at the retention of 7.48 kg/m<sup>3</sup> fire retardant additive. When the panel is tested as per BS 476-1981 (part 6), the fire propagation index is  $\sim 17.52$  only

indicatives of its contribution toward slow fire growth. As per the specified criteria of flame spread (limit:  $165 \pm 25$ mm), the sample belongs to Class I category according to surface spread of flame test (BS 476-1981, Part 7). During natural decay test, the composite boards treated with wood preservatives exhibited 4-8% weight loss after 8 weeks exposure in Aspergillus niger strain using potato dextrose broth culture compared to 9-13% weight loss for the untreated boards categorizing it under "highly resistant class" according to ASTM D 2017-2005. Termites caused ~6% less weight loss in the treated samples than the untreated samples after 10 weeks exposure in Microcerotermus bessoni colony showing their moderate resistance behavior as per ASTM D 3345-2008. The thermal conductivity and sound transmission loss of samples are ~0.136 W/mK and ~26.51 dB respectively showing their adequate insulation properties. Toxicity index based on gases emitted during the burning of composites is found to be 2.2-3.7 according to test conducted as per NES-713 standard which is considered to be safe with respect to other similar materials. The physicomechanical properties of pine needle composite boards are given in Table 6. It is suggested that pine needles in an average size of ~3 mm with desired surface characteristics can be gainfully utilized as supplementary raw material either alone or in combination with wood particles in the manufacturing of composite building panels.

| Table 6 — Physico-mechanical properties of pine needle composite boards. |                           |                                                                |  |  |
|--------------------------------------------------------------------------|---------------------------|----------------------------------------------------------------|--|--|
| Property                                                                 | Requirement<br>(IS: 3087) | Pine needle composite<br>board (~5% resin<br>adhesive content) |  |  |
| Density variation (%)                                                    | ± 10                      | $\pm 0.9$                                                      |  |  |
| Water absorption (%)                                                     |                           |                                                                |  |  |
| 2 h soaking                                                              | 25                        | 19.11                                                          |  |  |
| 24h soaking                                                              | 50                        | 45.44                                                          |  |  |
| Linear expansion 2 h<br>soaking (%)                                      |                           |                                                                |  |  |
| Length                                                                   | 0.5                       | 0.19                                                           |  |  |
| Width                                                                    | 0.5                       | 0.27                                                           |  |  |
| Thickness swelling 2<br>h water soaking (%)                              | 10                        | 12.6                                                           |  |  |
| Modulus of rupture (N/mm <sup>2</sup> )                                  | 11                        | 20.20                                                          |  |  |
| Tensile strength<br>perpendicular to<br>surface (N/mm <sup>2</sup> )     | 0.8                       | 1.12                                                           |  |  |
| Screw withdrawl strength face (N)                                        | 1250                      | 1270                                                           |  |  |

#### 4.2 Fabrication of Composites at Pilot Plant Level

#### 4.2.1 Jute/Sisal fiber composites

The commercial viability of developed composites is assessed at pilot plant scale for making composite panels of sizes 2000 mm x1000 mm and 1000 mm x 1000 mm (thickness: 3-8 mm). The main processing steps include: washing, surface modification of fibers, mat preparation, resin impregnation, compression moulding, curing and finishing 52. Initially, jute/sisal fibers are washed with running water followed by their treatment with ~5% aqueous sodium hydroxide to remove maximum water soluble extractives from their surfaces. Molding grade chopped strand mats are prepared by randomly arranged sisal fibers or nonwoven jute fabric (350 g  $m^{-2}$ ) in the desired size and spread uniformly with the poly(vinyl acetate) emulsion (5-7 wt% of fiber). After evaporation of water to a complete dryness, the resulting entanglements are compressed on a hydraulic press at  $80^{\circ}$ C and 1MPa pressure for ~2 min. Thereafter, the mats are cut to the desired size and wetted with catalyzed unsaturated polyester resin. The wetted mats are stacked over each other in a desired thickness and sandwiched between resin rich/gel coated brass plates. The stacked mats are compressed in a hydraulic press (300 ton capacity) equipped with takeoff unit for ~5 min at a pressure of ~2 MPa (Fig. 1). Glass fabric surface mat (weight  $\sim 30 \text{ g m}^{-2}$ ) is used on the exterior side of sample to get its good surface finish. The demolded sheet is cured at room temperature for 24 h and then post-cured at 80 °C for 2 h under contact pressure. The molded sheets are then saw-cut and trimmed to form accurately sized panels. Corrugated sheet (pitch length ~75 mm, pitch depth ~10.25 mm, thickness ~2.5 mm) is produced from three layers of resin wetted fiber mats using wooden block moulds with corrugated face.

#### 4.2.2 Rice husk/ plastic composites

The manufacturing of rice husk/plastic composites consists of three major steps: i) preparation line of rice husk flour, ii) compounding line for pallet production and iii) extrusion line for profile manufacturing<sup>58</sup>. The steam exploded rice husk is produced by its delignification under an optimized steam pressure (~25 psi for 45 min) and temperature (140 °C) in a digester followed by its washing in the running water, drying in a rotary drier and hammer milling to a desired size (100-300  $\mu$ m). Thereafter, the compounding of various ingredients such as rice husk



Fig. 1 — Manufacturing of jute fiber/unsaturated polyester composite panel/sheets.

flour, polypropylene random copolymer, coupler and additives is made in two step processes: kneading and forced feed extrusion at a temperature range of 180-200 °C. The extruded stands were granulated into 2-4 mm pellets. The pellets (moisture content, <1%) so obtained are fed into hopper of a twin screw extruder equipped with venting and degassing systems and shaped the extruded materials into rectangular sheets/ profiles of various cross sections such as 400 mm x 400 mm, 800 mm x 400 mm, 150 mm x 15 mm sheets and 40 mm dia round bars (Fig.2). The surface appearance of the extruded profiles can be controlled by the adjustment of vacuum venting system. The profiles are surface finished by sanding and embossing operation to obtain their aesthetic appearance.

#### 4.2.3 Pine needle composite boards

Pine needles collected from the forests are chipped and hammer milled preferably to a size of  $\sim$ 3 mm.



Profile sections

Fig. 2 — Manufacturing of rice husk plastic composite profiles.

Subsequently, the pine needles are digested in an autoclave with aqueous NaOH solution (2 wt %) for 30 min at room temperature. The resulting mass is washed with running water to remove its extractives and residual alkali and then dried in a rotary dryer. The dried pine needle furnishes are mixed with isocyanate prepolymer (~5 wt %) along with additives in a rotary drum mixer for 3-5 min. The coated needle furnish is layered down on the silicone paper lined mould plates in the form of loose mat and loaded into a press. The samples are compressed in a single daylight press at 140°C for a period of 8 min under 10 MPa pressure. The demoulded panels are trimmed and surface finished (1000 mm x 1000 mm in 12 mm thick) <sup>59</sup>. The photo view of some manufacturing step is shown in Fig. 3.



Fig. 3 — Manufacturing of isocyanate bonded pine needle composite panel/boards.

#### 4.3 Composite Building Products

#### 4.3.1 Natural fiber panels and door shutters

Various natural fiber based building products <sup>2,11,56</sup> such as panels, partitions, door frame & shutters, shuttering plates (alternatives to plywood) and roofing sheets have been produced (Fig. 4) and standardized as per the existing specifications. Some of these are briefly outlined.

The composite panels are made from sisal/jute nonwoven fabrics and unsaturated polyester/phenolic resins. Their physico-mechanical properties satisfy the requirements of IS 12406-88 laid down for medium density fiber boards<sup>60</sup>. It can be nailed and screwed for easy installation and also accommodative to any kind of paints and varnishes. To reduce their weight and improve specific strength and stiffness, the sandwich panels are produced using sisal/jute laminate as a face and corrugated structure/ unsaturated polyester-urethane hybrid resin foam as a core material. The developed panels can be used as alternatives to single walled pressed boards for door panel and walling applications.

The composite door shutters are produced by bonding the jute/polyester laminate faces (2-3 mm) with unsaturated polyester-polyurethane rigid foam core (28-30 mm). These door shutters satisfy the requirements as mentioned in IS:  $2202^{61}$  when tested as per IS: 4020-1998<sup>62</sup> for slamming, shock, impact indentation, edge loading, misuse etc. Since material possesses desirable screw holding and nailing properties, no wooden reinforcement is needed inside the door to fix the fixtures. The weight of door shutters is 12-14 kg/m<sup>2</sup>. Fixtures such as handles, locks and hinges can be easily fixed in the finished door shutters.

The shuttering plate of sisal/glass fiber-epoxy composites (sisal fiber content ~30 wt %) has been made (8 mm) as an alternative to plywood for concrete slab casting. The tensile strength and tensile modulus of these composite plates are ~57 MPa and ~2.6 GPa respectively whereas, the flexural strength and flexural modulus are ~98 MPa and ~4.7 GPa respectively. The water absorption of composite plates is in the order of ~4% only. Modular shuttering panel (weight, 12.50 kg/m<sup>2</sup>) has been designed as per IS 14687 <sup>63</sup> for casting of 120 mm thick concrete slab with supports at 0.90 m distance (load: 600 kg/m<sup>2</sup>).



Fig. 4 — Photo view of sisal/jute based building products (a) Composite panels, (b) Shuttering plate, (c) Door shutters and (d) Roofing sheets.

The deflection in the plate is  $\sim 1.95$  mm at initial loading and  $\sim 2.57$  mm after 7 days. It has been observed that the deflection in composite plate (8 mm) is under permissible limit (3 mm) and also comparable with the 12 mm plywood. More research is needed on natural fiber based shuttering plates for their large scale usages.

#### 4.3.2 Rice husk extruded profile & door frame

The composite profiles are produced from steam exploded rice husk flour and modified polypropylene by an extrusion molding (Fig.5). The resulting profiles meet the requirements of National Building Code, 2005 when tested as per IS: 1708<sup>64</sup> (Table 5). The sheets can be easily screwed and nailed. The sheets can easily be painted in any colors. The door frames made out of extruded profiles are installed in the school building in the premises of the Institute. The performance is satisfactory since 2014. In order to know the performance of sheets in packaging, 300 mm x 300 mm box is made out of these sheets by nail jointing and filled with sand. The filled box is



Fig. 5 — Rice husk plastic composite profiles and door frame.

dropped from 3 m height. It is found that the sheets are intact and show sufficient rigidity. It is noted that the cost of packaging box of rice husk composites is comparable with conventionally used plywood and timber in terms of material necessity, distribution necessity and reuse & recyclability necessities.

## 4.3.3 Pine needle composite boards

The composite boards are produced from the processed pine needles (furnishes) and isocyanate prepolymer based adhesive on a single daylight platen press (Fig. 6). The developed boards meet the requirements of IS: 3087<sup>65</sup> (Table 6). The salient features of the composite panels are: dimensionally stable, sufficient internal bond strength, adequate retention of properties under wet/humid conditions, carpenter friendly, good sound and thermal insulation and adequate flammability characteristics. The cost of boards is comparable to commercially available particle boards.

## **5** Techno-economics

The techno-economics of sisal/jute composite panels, rice husk plastic extruded profiles and pine needle composite boards have been prepared. These composites can compete economically in their present state with the existing materials. Market is still in an opening phase for building sector. As far as the market for these composites increases, the reduction of cost and improvement of the quality will be achieved. Since these are a new kind of products, the initial high productivity may pose some marketing



Fig. 6 — Photo view of pine needle composite boards.

problem but the product has got inherent merit for successful marketing.

A process for the manufacturing of sisal/jute composite panels/door shutters has been developed with the production capacity of ~6000 doors/annum. Major raw material includes: sisal, jute and others allied fibers, unsaturated polyester resin and additives. The power consumption of the project is small (4-6 KVA) as compared to corresponding building materials. The cost of developed products (~Rs.2500/door) is comparable with the existing products (FRP door, Rs.3000/- and wooden door, ~Rs.4000/-).

Manufacturing know-how for the production of rice husk plastic profile panels has been developed with the installed production capacity of ~100 kg/h for pellets and ~15 m/h for extruded profiles. The main raw materials used are: rice husk flour, modified polypropylene and additives. The power requirement during production is 1.5unit/kg. The cost of extruded profiles is Rs. 2700 per cubic feet which is comparable to wood (Rs.1500-3200 per cubic feet) depending on the quality of timber. The cost of door frame made out of these profiles (~Rs.2150/-) is comparable to Malaysian timbers (~Rs.2100/-) and Indian sal (~Rs.2200/-).

A process know-how for the manufacturing of pine needle composite boards has been developed. The major raw materials used are: pine needle, isocyanate prepolymer and additives. The salient features of manufacturing unit are: capacity,  $\sim 3000 \text{ m}^3/\text{annum of } 300 \text{ working days, electricity requirement, 15 units/h and cost of production, Rs.25/sq.ft for 12mm thickness compared to particle/fiber boards of 24-30/sq.ft.$ 

#### **6** Sustainability Issue

Environmental sustainability as indicated by the life cycle assessment (LCA) is also one of the advantageous attributes of the natural fiber/agroforest residue based composites. The composites with natural fibers/agro-forest residues 50-60% are considered promising for sustainable development because they contribute to the substitute of renewable resources and help to reduce greenhouse gas emission. Several studies<sup>66-71</sup> have been conducted on the LCA analysis of natural fiber composites vis-à-vis glass fiber composites using commonly used environmental indicators such as resource depletion, global warming, cumulative energy demand, eutrophication etc. The extent to which natural fibers can replace glass fibers (which is heavy and energy intensive to produce) mainly determines the net environmental benefits.

Joshi et al.<sup>68</sup> reported superior environmental impact of natural fiber composites to glass fiber composites in terms of energy consumption during production. reduced emissions during service life of the products and carbon and energy credits from natural fibers incineration. Alves et al.<sup>69</sup> carried out LCA on the replacement of glass fibers with jute fibers in automotive structural parts. Taking into account of recycling, incineration and landfill disposal modes, they observed that jute fiber composites exhibited improved environmental performance. Vidal et al.<sup>70</sup> studied the environmental impact of composites made of recycled polypropylene/high density polyethylene and rice husk/ recycled cotton in comparison to virgin ones considering global warming, non-renewable energy depletion, acidification and eutrophication as evaluation criteria. They found that composites had reduced environmental impact during the materials supplying and processing in all of the impact categories considered. Luz *et al.*<sup>71</sup> compared the LCA of sugarcane bagasse/polypropylene composites and talc/ polypropylene composites. They suggested that sugarcane has a positive contribution to minimize the environmental impacts in terms of carbon credits and reuse of the end- of- life material. Based on these studies, it is recognized that natural fibers/agro-forest residues based products is more environmentally sustainable in comparison to glass fiber composite counterparts.

#### 7 Conclusions

Results indicate that polymer composites made from natural fibers/agro-forest residues present immense opportunities to play an increasing role as alternate building materials especially wood substitutes in low cost housing/buildings. Sisal/jute based composites, rice husk profiles and pine needle boards have been manufactured at pilot plant scale. Their use with adequate strength and dimensional stability perform satisfactorily for door shutters, shuttering plates as alternatives to plywood, door frames, decking materials, roofing and panel products. The performance of resultant composites under accelerated water aging, termites, natural decay and fire is satisfactory after their necessary treatments with chemical additives. Based on the technoeconomic studies, these composites are economically viable and also compete with the existing materials. Environmental benefits assessed by LCA supported superiority of natural fibers based products over glass fiber composites.

It is suggested that the availability of natural fibers in various forms such as long fiber strands, chopped strand mats, prepegs etc. is necessary as similar to glass fibers for utilization of their full potential in the composite industry. Problems related to natural fibers/agro-forest residues such as inconsistency in product performance due to fiber variability, use of fiber in a partially prepared state, wettability and poor fiber-matrix interface needs to be carefully tackled for their industrial exploitation. A state of the art data base should be established on the properties of natural fibers and their composites as a guide line for potential users. This is essentially needed because of variability in the properties of fiber from its origin, age, growing conditions, extraction methods, extent of vulnerability towards various environments etc. which may contribute products unusual performance.

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