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Composites Based on Natural Fibre Fabrics

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1. Introduction

In the latest years industry is attempting to decrease the dependence on petroleum-based fuels and products due to the increased environmental consciousness. This is leading to the need to investigate environmentally friendly, sustainable materials to replace existing ones. The tremendous increase of production and use of plastics in every sector of our life lead to huge plastic wastes. Disposal problems, as well as strong regulations and criteria for cleaner and safer environment, have directed great part of the scientific research toward eco-composite materials. Among the different types of eco-composites those which contain natural fibers (NF) and natural polymers have a key role. Since few years polymeric biodegradable matrices have appeared as commercial products, however their high price represents the main restriction to wide usage. Currently the most viable way toward eco-friendly composites is the use of natural fibres as reinforcement. Natural fibres represent a traditional class of renewable materials which, nowadays, are experiencing a great revival. In the latest years there have been many researches developed in the field of natural fibre reinforced plastics (Bledzki & Gassan, 1999). Most of them are based on the study of the mechanical properties of composites reinforced with short fibers. The components obtained therefore are mostly used to produce non-structural parts for the automotive industry such as covers, car doors panels and car roofs (Magurno, 1999, John at al., 2008) (Fig.1,2).

Fig. 1. Mercedes-Benz A natural fibre composites components (source: DaimlerChrysler AG)

Few studies deal with structural composites based on natural reinforcements. These studies are mainly oriented to the housing applications where structural panels and sandwich beams are manufactured out of natural fibres and used as roofs (Saheb & Jbg., 1999).
Considering the high performance standard of composite materials in terms of durability, maintenance and cost effectiveness, the application of natural fiber reinforced composites as construction material holds enormous potential and is critical for achieving sustainability. Due to their low density and their cellular structure, natural fiber possesses very good acoustic and thermal insulation properties and demonstrate many advantageous properties over glass or rockwool fibre (e.g. handling and disposal).

Fig. 2. Examples of applications of Natural Fibres in the automotive field

Nowadays natural fibre composites are not exploited only in structural and semi-structural applications of the automotive sector, but in other fields too (Fig. 3).

Fig. 3. Examples of use of Natural Fibres in several applications
Natural fibres (Fig.4) can be divided, according to their origin, into: animal, vegetable and mineral. The most used are the vegetable ones due to their wide availability and renewability in short time respect to others, so when we say “natural fibres” We refer here to the vegetables ones. In the past, natural fibres were not taken into account as reinforcements for polymeric materials because of some problems associated with their use:

- Low thermal stability, in other terms the possibility of degradation at moderate temperature (230-250 ° C).
- Hydrophilic nature of fibre surface, due to the presence of pendant hydroxyl and polar groups in various constituents, which lead to poor adhesion between fibres and hydrophobic matrix polymers (John et al., 2008, Kalia et al., 2009). The hydrophilic nature can lead to swelling and maceration of the fibers. Furthermore, moisture content decreases significantly fibre’s mechanical properties.
- Properties variability depending on the quality of the harvest, age and body of the plant from which they are extracted, the extraction techniques and the environmental conditions of the site.

Lack of good interfacial adhesion, low degradation temperature, and poor resistance towards moisture make the use of natural fibre reinforced composites less attractive than synthetic fibre (glass, carbon, aramid, etc.) that have been up to now the only choice for reinforcing polymeric composites, due to their superior mechanical properties. However, the production of composites reinforced with synthetic fibres and matrices requires a large amount of energy which is only partially recovered with incineration of fibre reinforced composites. This has once again drawn the attention towards natural fibres due to their environmental advantages. It has been demonstrated that the energy needed for production of natural fibres is, on average, more than half of the amount needed for synthetic fibres (Fig.5). Thus, the renewed interest in the natural fibers, due to their lightweight, nonabrasive, non irritating, combustible, nontoxic, biodegradable properties (Saheb & Jog, 1999), low energy consumption for production, budget zero CO₂ emissions if burned, low cost (Table 1), main availability and renewability compared to synthetic fibres, has resulted in a large number of applications to bring it at par and even superior to synthetic fibers. Because of such properties natural fibers are fast emerging as a viable choice as reinforcing material in composites (Kalia et al., 2009).

Even if natural fibre has a very low energy consumption for production compared to other synthetic fibre, such as glass or carbon, careful environmental impact evaluation must be
take in consideration in order to make the right choice. In fact, the validity of “green” case for substitution of synthetic fibre by natural ones is dependent on the type of reinforcement and related production processes. A parameter which better describe the environmental impact is the embodied energy calculated with reference to all related agricultural operations (from ploughing to harvest), fibre extraction operations (retting and decortication), fibre preparation operations (hackling and carding), fibre processing operations (spinning or finishing) and materials used for these operations. The use of embodied energy parameter reveals that not any kind of natural fibre reinforcement is “greener” than synthetic ones. Fig. 6 shows that, even if adopting the most environmental friendly option (no-till and water retting) for flax fibre production, only mat fabrics are, in energetic terms, “greener” while flax yarns has a higher embodied energy respect to glass fibre continuous filament production.

Fig. 5. Energy for production of some fibre (sources: SachsenLeinen; Daimler 1999; BAFA; NOVA; AVB; CELC; REO)

<table>
<thead>
<tr>
<th>Fiber</th>
<th>Price / m³</th>
<th>Specific Gravity Kg/ m³</th>
<th>Price / kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wood</td>
<td>420</td>
<td>1600</td>
<td>0.26</td>
</tr>
<tr>
<td>Flax</td>
<td>600</td>
<td>1500</td>
<td>0.40</td>
</tr>
<tr>
<td>Glass</td>
<td>4850</td>
<td>2600</td>
<td>1.87</td>
</tr>
<tr>
<td>PP</td>
<td>650</td>
<td>900</td>
<td>0.72</td>
</tr>
</tbody>
</table>

Table 1. Cost comparison between natural and synthetic fibre (Source: Georgia Institute of Technology www.me.gatech.edu/jonathan.cotton/me4793/hafiber.pdf)

Natural fibres can be classified according to their origin and grouped into leaf: abaca, cantala, curaua, date palm, henequen, pineapple, sisal, banana; seed: cotton; bast: flax, hemp, jute, ramie; fruit: coir, kapok, oil palm. Among them flax, bamboo, sisal, hemp, ramie, jute, and wood fibres are of particular interest (Kalia et al., 2009). The most important physical and mechanical properties are summarized in Table 2. Physical and mechanical properties depend on the single fibre chemical composition (Cellulose, hemicelluloses, lignin, pectin, waxes, water content and other minors) according to grooving (soil features, climate, aging conditions) and extraction/processing methods.

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conditions. Grooving conditions is recognized as the most influential parameter for the variability of mechanical properties of the fibres. The chemical composition of several natural fibres is summarised in Table 3.

---

Table 2. Natural fibre properties. Source: Natural fibre ’09 Proceedings (University of Bath)

<table>
<thead>
<tr>
<th>Plant fibre</th>
<th>Tensile strength (MPa)</th>
<th>Young's modulus (GPa)</th>
<th>Specific modulus (GPa)</th>
<th>Failure strain (%)</th>
<th>Length of ultimate, l (mm)</th>
<th>Diameter of ultimate, d (μm)</th>
<th>Aspect ratio, l/d</th>
<th>Microfibril angle, φ (°)</th>
<th>Density (kg.m⁻³)</th>
<th>Moisture content (eq.) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cotton</td>
<td>6-10</td>
<td>4-6.5</td>
<td>6-8</td>
<td>20-64</td>
<td>11.5-17</td>
<td>2752</td>
<td>20-30</td>
<td>1500</td>
<td>8.5</td>
<td></td>
</tr>
<tr>
<td>Kapok</td>
<td>4</td>
<td>12.9</td>
<td>1.2</td>
<td>8-32</td>
<td>15-35</td>
<td>724</td>
<td>311-384</td>
<td>5.9</td>
<td>1500</td>
<td></td>
</tr>
<tr>
<td>Bamboo</td>
<td>27</td>
<td>18</td>
<td>-</td>
<td>2.7</td>
<td>10-40</td>
<td>9259</td>
<td>-</td>
<td>1500</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>Flax</td>
<td>500-900</td>
<td>50-70</td>
<td>34-48</td>
<td>27-36</td>
<td>17.8-21.6</td>
<td>1258</td>
<td>5</td>
<td>1400-1500</td>
<td>12</td>
<td></td>
</tr>
<tr>
<td>Hemp</td>
<td>310-750</td>
<td>20-41</td>
<td>2.4</td>
<td>8.3-14</td>
<td>17-23</td>
<td>549</td>
<td>6.2</td>
<td>1400-1500</td>
<td>12</td>
<td></td>
</tr>
<tr>
<td>Jute</td>
<td>200-450</td>
<td>14-39</td>
<td>2-3</td>
<td>1.9-3.2</td>
<td>15.9-26.7</td>
<td>157</td>
<td>8.1</td>
<td>1300-1500</td>
<td>12</td>
<td></td>
</tr>
<tr>
<td>Kena</td>
<td>295-1191</td>
<td>22-60</td>
<td>-</td>
<td>2.6-1</td>
<td>17.7-21.9</td>
<td>119</td>
<td>-</td>
<td>1220-1400</td>
<td>17</td>
<td></td>
</tr>
<tr>
<td>Ramie</td>
<td>915</td>
<td>23</td>
<td>15</td>
<td>3.7</td>
<td>60-250</td>
<td>26.1-35</td>
<td>4839</td>
<td>-</td>
<td>1500</td>
<td>8.5</td>
</tr>
<tr>
<td>Abaca</td>
<td>12</td>
<td>41</td>
<td>-</td>
<td>3.4</td>
<td>4.6-5.2</td>
<td>17.2-24</td>
<td>257</td>
<td>-</td>
<td>1500</td>
<td>14</td>
</tr>
<tr>
<td>Banana</td>
<td>529-914</td>
<td>20-24</td>
<td>1.3</td>
<td>2.3-8</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>11-12</td>
<td>1300-1350</td>
<td>-</td>
</tr>
<tr>
<td>Pineapple</td>
<td>413-1627</td>
<td>42-57</td>
<td>0.4-6</td>
<td>-</td>
<td>20-40</td>
<td>6-14</td>
<td>1400-1500</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Sisal</td>
<td>60-840</td>
<td>6-15</td>
<td>2-14</td>
<td>1.8-3.1</td>
<td>18.3-23.7</td>
<td>115</td>
<td>10-22</td>
<td>1300-1500</td>
<td>11</td>
<td>-</td>
</tr>
<tr>
<td>Coir</td>
<td>106-175</td>
<td>5.2</td>
<td>15-40</td>
<td>0.9-1.2</td>
<td>16.2-19.5</td>
<td>64</td>
<td>39-49</td>
<td>1150-1250</td>
<td>13</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 3. Natural fibre composition (Williams et al., 2000; Bogoeva-Gaceva et al., 2007)

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Natural fibre mechanical properties depend on the type of cellulose and the geometry of the elementary cell. The celluloses chains are arranged parallel to each other, forming bundles each containing forty or more cellulosic macromolecules linked by hydrogen bonds and through links with amorphous hemicelluloses and lignin which confer stiffness to fibre called microfibrils. More interwoven microfibrils form a rope-like structure (Rong et al., 2001) (Fig. 7).

Fig. 7. Natural fibre hierarchal structure

Among natural fibres the bast fibres, extracted from the stems of plants such as jute, kenaf, flax, ramie and hemp are widely accepted as the best candidates for reinforcements of composites due to their good mechanical properties. Hemp was shown to have very promising tensile properties for applications where mechanical properties are a requisite (Nair et al., 2000)

As many authors agree, the two basic parameters that allow to characterize mechanical behavior of natural fibers are the cellulose content and the spiral angle. In general, the tensile strength of the fibers increases with increasing cellulose content and with decreasing angle of helix axis of the fibers.

The strength of natural fibre composites in on average lower compared to the synthetic fibre reinforced composites, even under optimised fibre-matrix interaction (Heijenrath & Peijs, 1996, Berglund & Ericson, 1995), but their lower density and cost make them competitive in terms of specific and economic properties. This is basically due to the composite-like structure of natural fibres (Van den Oever et al., 1995); they are generally not single filaments as most manmade fibres but they can have several physical forms, which depend on the degree of fibre isolation. Composite strength depends also on fibre diameter (smallest diameter could achieve higher mechanical resistance due to larger specific contact surface with matrix) and fibre length.

2. Natural fibre fabric types

The possibility to have long or short fibres depends on the material under consideration, in fact, for synthetic fibre it is easy and common to have long continuous fibres out of
production plant, while, for natural fibres, the fibre’s length is an inherent limit for the material itself due to their natural origin which limits their length (for example the plant stem). This is a basic reason why natural fibres are usually found as short reinforcements which are used to produce mat fabrics. Discontinuous fibres (chopped) are generally used for a randomly oriented reinforcement (mat) when there is not any preferential stress direction and/or there is a low stress/strain level in the composite (Fig.8). As it will be shown in the case studies mats, due to the random fibre orientation, are non-optimised fabric for mechanical performances.

Fig. 8. Hemp mat

The alternative to the use of short fibres is the manufacture of long yarns. Yarn is a long continuous assembly of relatively short interlocked fibres, suitable for use in the production of textiles, sewing, crocheting, knitting, weaving, embroidery and ropemaking that are twisted with an angle to the yarn axis in order to provide axial strength to the yarn. Spun yarns are made by twisting or otherwise bonding staple fibres together to make a cohesive thread and may contain a single type of fibre or a blend of various types. Two or more spun yarns, if twisted together, form a thicker twisted yarn. Depending on the direction of this final twist, the yarn will be known as s-twist or z-twist (Fig.9). Two or more parallel spun yarns can form a roving. The main advantage of using natural yarns is the possibility to weave them into 2D and 3D fabrics with tailored yarn orientations.

A common measure unit used to classify fibres and yarns is the denier which corresponds to the linear mass density of the yarns. Denier is defined as the mass in grams per 9000 meters. In the International System of Units the tex is used instead, defined as the mass in grams per 1000 meters. The most commonly used unit is actually the decitex, abbreviated dtex, which is the mass in grams per 10000 meters. Similar to tex and denier, yield is a term that helps describe the linear density of a roving of fibres. However, unlike tex and denier, yield is the inverse of linear density and is usually expressed in yards/ lb. Linear mass of twisted yarn is expressed by a fraction where the numerator is the yarn count and the denominator is simply the number of ends (e.g. 30/3).

Spun yarns obtained from natural fibres present usually some short fibres protruding out of the main yarn body (Fig.10). This short fibres are commonly referred to as yarn hairiness. Although not desirable in many cases, the hairiness can lead to better mechanical yarn/resin interlocking in composites. Another advantage of natural yarns is the increased surface roughness of yarns compared to fibres, which increases the interfacial strength due to mechanical interlocking, improving the transverse properties. In addition, twisting localizes the micro damages within the yarn leading to higher fracture strength.
An important control parameter for such natural yarns is the twist level. It has been shown (Goutianos & Peijs, 2003) that very low twisted yarns display a very low strength when tested in air and therefore they cannot be used in processes such as pultrusion or textile manufacturing routes like knitting or weaving. (Fig. 11) where heavy loading is experienced by the yarns while processing. In the case of short staple (length) fibres, higher twist level is necessary to prevent fibre slippage and to develop sufficient strength.

Besides yarn strength, the amount of twist also affects the inter-yarn impregnation while fabricating reinforced composites. With increased twist level yarns become more compact making it difficult for the resin to penetrate into the yarn. Dry yarns lead to lower bonding between yarns and resin thus leading to delamination and lowering of the composite tensile properties. Several authors showed that when highly twisted yarns are impregnated in a polymer resin, their strength may decrease significantly with decreases similar to the drop in strength of an off-axis composite (Goutianos & Peijs, 2003; Baley, 2002). Thus, there is an optimum level of twist, which should be kept as low as possible for optimal composite mechanical properties to allow for proper yarn’s wetting to be achieved.

Fig. 9. Hemp twisted yarn and scanning electron microscope image of hemp twisted yarn

Fig. 10. Hemp and flax fibre rovings
The fibre contribution to composite mechanical properties improvement is emphasized when the stresses have components along the fibre direction (Fig. 12). However, most of the studies reported in literature are focused on the use of mat which are the cheapest alternative (Paiva et al., 2004) among technical fabrics. Several studies showed that the random orientation of the fibres in mat fabrics leads to lowering of the reinforcing efficiency (Baiardo et al., 2004).

Yarns offer a viable and interesting alternative to the use of short fibres as multiple filament yarns can be weaved into 2- or 3-Dimension textiles. Weaving is a textile production method which involves interlacing a set of longer threads, twisted yarn or roving, (called the warp) with a set of crossing threads (called the weft) (Fig. 13). This is done on a frame or machine known as a loom, of which there are a number of types. Some weaving is still done by hand, but the vast majority is mechanised. The main advantage of using weaved fabrics is the possibility to pre-orient the filaments in the designed directions. Natural yarns differ from multifilament of synthetic fibres (ie. tow) because they are an assembly of short fibre instead of an assembly of aligned continuous fibres. However, the fibres which constitute the yarn have a preferential orientation along an helical trajectory which make the use of natural yarns attractive compared to short fibres because in such yarns fibres are mostly along the load direction.
The manner in which the warp and weft threads are interlaced is known as the weave style. The three basic weave styles or architectures are:

- plain weave
- satin weave
- twill weave

Plain weave is the most basic type of textile weaves. The warp and weft are aligned so they form a simple criss-cross pattern. Each weft thread crosses the warp threads by going over one, then under the next, and so on (Fig. 14, 15). The next weft thread goes under the warp threads that its neighbour went over, and vice versa. In balanced plain weaves the warp and weft are made of threads of the same weight (size) and the same number of ends per inch.

http://en.wikipedia.org/wiki/Plain_weave - cite_note-1

Fig. 13. Warp and weft in plain weaving

Fig. 14. Plain woven yarn and woven roving schemes (0°/ 90° reinforcement directions)

Fig. 15. Examples of plain woven flax yarns. H-181 100% Hemp Canvas weave 18oz/ sq yd Wide 59” 5N/ 2 x 8N/ 2 x23x21. Source: dongpinghemp.com
The satin weave is characterized by four or more weft yarns floating over a warp yarn or vice versa, four warp yarns floating over a single weft yarn (Fig.16).

Twill is a type of fabric woven with a pattern of diagonal parallel ribs. It is made by passing the weft thread over one or more warp threads and then under two or more warp threads and so on, with a "step" or offset between rows to create the characteristic diagonal pattern (Fig.17,18). Because of this structure, twills generally drape well. In a twill weave, each weft or filling yarn floats across the warp yarns in a progression of interlacings to the right or left, forming a distinct diagonal line. This diagonal line is also known as a wale. A float is the portion of a yarn that crosses over two or more yarns from the opposite direction.

Fig. 16. Satin weave with 16 warp yarns floating over each weft yarn.

Fig. 17. Structure of a 3/1 and 2/2 twills

Fig. 18. Examples of plain woven flax yarns. (A) Natural Twill Weave 100% Hemp 12oz Width 57/58" (B) Natural Herringbone Weave 52% Hemp 48% Flax 20oz Width 57/58". Source: EnviroTextile.com
A twill weave can easily be identified by its diagonal lines. and is often designated as a fraction—such as 2/1—in which the numerator indicates the number of harnesses that are raised, in this example, two, and the denominator indicates the number of harnesses that are lowered when a filling yarn is inserted, in this example one. The fraction 2/2 would be read as “two up, two down.” with two warp threads crossing every two weft threads. The offset at each row forms the diagonal pattern. The minimum number of harnesses needed to produce a twill can be determined by totalling the numbers in the fraction.

The fewer interlacings in twills allow the yarns to move more freely, and thus they are softer and more pliable, and drape better. Twills also recover better from wrinkles than plain-weave fabrics. When there are fewer interlacings, yarns can be packed closer together to produce high-count fabrics.

There is an increasing number of producers of natural fibre fabrics around the world which are tailoring their products for composites technology. Table 4 shows some costs for a selection of fabrics commercialized in U.S.A. by the company EnviroTextile LLC.

<table>
<thead>
<tr>
<th>Fabric Descriptions and Specifications</th>
<th>Units = Yards</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural 100% Hemp Canvas Plain Weave, 16oz/yd, 41c/28, Width 57/58&quot; 12oz Semi-</td>
<td>1-49</td>
</tr>
<tr>
<td>Bleached, Preshrunk, Cationic Softener, SBPE 100%</td>
<td>$14.60</td>
</tr>
<tr>
<td>Natural 100% Hemp Canvas Basket Weave, 10oz/yd, 10am/3, 50yds, Width 57/58&quot; 18.5oz</td>
<td>$16.45</td>
</tr>
<tr>
<td>Semi-Bleached, Preshrunk, Cationic Softener, SBPE 100%</td>
<td></td>
</tr>
<tr>
<td>Black 100% Hemp Canvas Basket Weave, 10oz/yd, 10mm/3, 30yds, Width 57/58&quot; 18.5oz</td>
<td>$17.60</td>
</tr>
<tr>
<td>Preshrunk, No Softener, SBPE 100%</td>
<td></td>
</tr>
<tr>
<td>Dark Brown 100% Hemp Canvas Basket Weave, 10oz/yd, 10am/3, 30yds, Width 57/58&quot; 18.5oz Semi-Bleached, Preshrunk, No Softener, SBPE 100%</td>
<td>$17.60</td>
</tr>
<tr>
<td>Sand 100% Hemp Canvas Basket Weave, 10oz/yd, 10am/3, 30yds, Width 57/58&quot; 18.5oz Semi-Bleached, Preshrunk, No Softener, SBPE 100%</td>
<td>$17.60</td>
</tr>
<tr>
<td>Natural 100% Hemp Harringtonweave, 16oz/2x8.5oz, 41c/37, Width 55/56&quot; 10.5oz</td>
<td></td>
</tr>
<tr>
<td>Soft Finish, SBPE 100%</td>
<td></td>
</tr>
</tbody>
</table>

Table 4. Costs of some fabrics sold by EnviroTextile

Other examples of commercial products available on the market are the flax fabric (Fig. 19) manufactured by Biotex (http://www.compositesevolution.com) which are also available as pre-impregnated fabric with PLA (polylacticacid) and PP (polypropylene). Other products available are the pre-impregnated fabrics (FLAXPLY©) produced by Lineo. The products sold by Lineo have pre-treated fibers for increased fiber-matrix adhesion. The FLAXPLY© are proposed to be used for internal layer of mixed carbon/flax design for improved vibration absorption (Fig.20).

As mentioned before, yarns and rovings can be woven in 3-Dimension fabrics, even if they are not so widespread as plain ones. To date no commercial example of 3D weaved fabric based on natural yarns is available.
3. Fiber surface treatments

The contribution of fibres to the final properties of the composite depends on:
- Mechanical properties of fibres;
- Type (continuous/discontinuous) and orientation of fibres in the composite (anisotropy);
- Volume fraction of fibres;
- Fibre-matrix interface;
- Processing technique used for composite manufacturing.

Shortcomings associated with natural fibres have to be overcome before using them in polymer composites. The most serious concern with natural fibres is their hydrophilic nature due to the presence of pendant hydroxyl and polar groups in various constituents, which can lead poor adhesion between fibres and hydrophobic matrix polymers (Rong et al., 2001, Bledzki & Gassan, 1996). The hydrophilic nature of the fibre surface lead also to high moisture uptake for the natural fibres which can seriously lower the mechanical properties of the fibres themselves.

The natural fibres are inherently incompatible with nonpolar-hydrophobic thermoplastics, such as polyolefins. Moreover, difficulty in mixing because of poor wetting of the fibres with the matrix is another problem that leads to composites with weak interface (John & Anandjiwala, 2008).
There are some physical fibre treatments (e.g. Plasma), but nowadays when we speak about surface treatments we almost mean chemical ones. These treatments can clean the fibre surface, modify the chemistry on the surface, lower the moisture uptake and increase the surface roughness. As the natural fibres bear hydroxyl groups from cellulose and lignin they are amenable to chemical modification. The hydroxyl groups may be involved in the hydrogen bonding within the cellulose molecules thereby reducing the activity towards the matrix. Chemical modifications may activate these groups or can introduce new moieties that can effectively lead to chemical interlock with the matrix. Mercerization, isocyanate treatment, acrylation, permanganate treatment, acetylation, silane treatment and peroxide treatment with various coupling agents and other pretreatments of natural fibres have achieved various levels of success for improving fiber strength, fiber fitness and fiber-matrix adhesion. In the following section we report a review of the main pretreatments techniques.

3.1 Alkali treatment
Alkali treatment of natural fibers, also called mercerization, is the common method to produce high-quality fibers. The scheme of the reaction is:

\[ \text{FIBER-OH} + \text{NaOH} \rightarrow \text{FIBER-O-} \text{Na}^+ + \text{H}_2\text{O}. \]

Mercerization leads to fibrillation which causes the breaking down of the composite fibre bundle into smaller fibres. Mercerization reduces fibre diameter, thereby increases the aspect ratio which leads to the development of a rough surface topography that results in better fibre/matrix interface adhesion and an increase in mechanical properties (Kalia at al., 2009). Moreover, mercerization increases the number of possible reactive sites, allows better fibre wetting and gets an effect on the chemical composition of the hemp fibres, degree of polymerization and molecular orientation of the cellulose crystallites due to cementing substances like lignin and hemicelluloses which were removed during the mercerization process. As a result, mercerization had a long-lasting effect on the mechanical properties of hemp fibres, mainly on fibre strength and stiffness. If the treatment is done at high percentage of NaOH there could be an excessive extraction of lignin and hemicelluloses which can result in damage of the ultimate cells walls. Similar reduction of mechanical properties after alkali treatment have been reported in the literature (Rodriguez at al., 2007). Alkali treatment is recognized to hydrolyses the amorphous parts of cellulose present in fibres so that after treatment the material contains more crystalline cellulose (Le Troedec, 2008). Furthermore, it removes waxes and oils from the surfaces (Sgriccia, 2008).

3.2 Acetylation
Acetylation was originally applied to wood cellulose to stabilize the cell walls against moisture, improving dimensional stability and environmental degradation and to introduce plasticization to cellulose fibers by esterification. Acetylation is based on the reaction of cell wall hydroxyl groups of lignocellulosic materials with acetic or propionic anhydride at elevated temperature (Fig. 21). Pretreatment of fibers with acetic anhydride substitutes the polymer hydroxyl groups of the cell wall with acetyl groups, modifying the properties of these polymers so that they become hydrophobic (Andersson & Tillman, 1989; Murray, 1998; Rowell, 1991). Hydroxyl groups that react with the reagent are those of lignin and hemicelluloses (amorphous material), whereas the hydroxyl groups of cellulose (crystalline material) are being closely packed with hydrogen bonds, prevent the diffusion of reagent and thus result in very low extents of reaction (Rowell, 1998).
3.3 Peroxide treatment

Peroxide treatment of cellulose fibre has attracted the attention of various researchers due to easy processability and improvement in mechanical properties. Organic peroxides tend to decompose easily to free radicals, which further react with the hydrogen group of the matrix and cellulose fibers. In peroxide treatment, fibers are treated with 6% benzoyl peroxide or dicumyl peroxide in acetone solution for about 30 min after alkali pretreatment (Sreekala et al., 2002; Sreekala et al., 2002; Paul et al., 1997) conducted at a temperature of 70°C to support the decomposition of the peroxide.

3.4 Graft copolymerization

Synthesis of graft copolymers by creation of an active site, a freeradical or a chemical group which may get involved in an ionic polymerization or in a condensation process, on the preexisting polymeric backbone is one of the common methods. Polymerization of an appropriate monomer (e.g. benzoyl chloride, maleated polypropylene/maleic anhydride MAH-PP, acrylation, titanate) onto this activated back-bone polymer leads to the formation of a graft copolymer with a higher surface energy and wettability and adhesion interface by polymer matrix. It has been reported that maleic anhydride treatment reduced the water absorption to a great extent in hemp, banana and sisal fibers and their composites (Mysra et al. 2000).

Modification of cellulosic fibers by etherification enhances certain new ranges of properties and makes it more useful and acceptable in diversified applications. Sodium hydroxide plays an important role in forming a charged intermediate species with the fiber, which allows the faster nucleophilic addition of epoxides, alkyl halides, benzyl chloride, acrylonitrite, and formaldehyde (Matsuda, 1996). Benzoyl chloride is the most often used benzoylation pretreatment. Benzoyl (C₆H₅C=O) groups react with the cellulosic OH group of fiber decreasing hydrophilic nature of the treated fiber (Joseph et al., 2000) after a 30 min pre-soaking with NaOH solution to activate the hydroxyl groups of the cellulose and lignin in the fiber, followed by filtration and washing with water (Fig.22).

![Fig. 21. Scheme of acetylation reaction](image1)

![Fig. 22. Possible reaction between cellulosic-OH and benzoyl chloride (Joseph et al., 2000)](image2)
and enzymatic grafting. The conventional techniques of grafting of natural fibers require significant time and energy. It has been found that grafting under microwave radiations is the best method in terms of time consumption and cost effectiveness. Microwave radiation technique reduces the extent of physicochemical stresses to which the fibers are exposed during the conventional techniques (Kaith & Kalia 2008).

3.5 Coupling agents
Coupling agents usually improve the degree of crosslinking in the interface region and offer a perfect bonding. Among the various coupling agents, silane coupling agents were found to be effective in modifying the natural fiber-matrix interface. Silane grafting is based on the use of reactants that bear reactive end groups which, on one end, can react with the matrix and, on the other end, can react with the hydroxyl groups of the fiber (Fig. 23). The alkoxy or ethoxy are the end groups which can form stable covalent bonds reacting with the hydroxyl groups of the fiber. The end groups which can react with the matrix vary according to the polymer matrix type. If unsaturated polyester is used silanes bearing methacryl-, amine- and vinyl- can be used (Soo-In et al., 2001; Li Hu et al., 2009). Efficiency of silane treatment was high for the alkaline treated fiber than for the untreated fiber because more reactive site can be generated for silane reaction. Therefore, fibers are pretreated with NaOH for about half an hour before its coupling with silane. Fibers are then washed many times in distilled water and finally dried. Silane coupling agents may reduce the number of cellulose hydroxyl groups in the fiber-matrix interface minimizing fibre sensitivity to humidity. In the presence of moisture, hydrolizable alkoxy group leads to the formation of silanols. The silanol then reacts with the hydroxyl group of the fiber, forming stable covalent bonds to the cell wall that are chemisorbed onto the fiber surface (Agrawal et al., 2000). Therefore, the hydrocarbon chains provided by the application of silane restrain the swelling of the fiber by creating a cross-linked network because of covalent bonding between the matrix and the fiber.

![Fig. 23. Reaction of silane with OH groups of natural fiber](image)

Silanes are effective in improving the interface properties (Coutinho at al., 1997; Gonzales et al., 1997). Alkoxy silanes are able to form bonds with hydroxyl groups. Fiber treatment with toluene disocyanate and triethoxyvinyl silane could improve the interfacial properties.
Silanes after hydrolysis undergo condensation and bond formation stage and can form polysiloxane structures by reaction with hydroxyl group of the fibers. Silane grafting can modify the mechanical performances of fiber as a consequence of the use of acid solution for the treatment.

Isocyanate has –N=C=O functional group, which is very susceptible to reaction with the hydroxyl group of cellulose and lignin in the fibers and forms strong covalent bonds, thereby creating better compatibility with the binder resin in the composites (Kokta et al. 1990).

### 3.6 Permanganate treatment

Pretreatments with permanganate are conducted by using different concentration of potassium permanganate \( \text{(KMnO}_4 \text{)} \) solution in acetone with soaking duration from 1 to 3 min after alkaline pretreatment. As a result of permanganate treatment, the hydrophilic tendency of the fibers is reduced, and thus, the water absorption of fiber-reinforced composite decreases with increase in \( \text{KMnO}_4 \) concentration (Sreekala et al., 2000; Paul et al., 1997). Permanganate treatment is indicated as one of the best method to improve the bonding at the fiber-polymer interface.

### 3.7 Physical plasma treatment

Plasma treatment is an effective method to modify the surface of natural polymers without changing their bulk properties. The plasma discharge can be generated by either corona treatment or cold plasma treatment. Both methods are considered as a plasma treatment when ionized gas has an equivalent number of positive and negative charged molecules that react with the surface of the present material. The distinguishing feature between the two categories of plasmas is the frequency of the electric discharge. High-frequency cold plasma can be produced by microwave energy, whereas a lower frequency alternating current discharge at atmospheric pressure produces corona plasma. The type of ionized gas and the length of exposure influenced the modification of the wood and synthetic polymer surfaces (Young et al., 1992; Goring & Bolam, 1976).

### 3.8 Chemical treatments on natural fibre: effect on mechanical properties

Chemically treated fibers can show a considerable decrease in tensile properties and this decrease is attributed to the substantial delignification and degradation of cellulose chains during chemical treatment. The extension at break of these fibers does not change much. Most of the chemical treatments have been found to decrease the fiber strength due to breakage of the bond structure, and disintegration of the noncellulosic materials but silane and acrylation treatment leave to strong covalent bond formation and the stiffness is enhanced marginally due to the crystalline region (cellulosic) of the fiber. The alkali treatment can produce a drop in both tensile strength and Young’s modulus of the fibers if a very high percentage treatment is adopted. This result is attributed to the damage induced in the cell walls and the excessive extraction of lignin and hemicellulose, which play a cementing role in the structure of the fibers. Morphological studies showed that the silane, benzoylation and peroxide pretreatment of flax fiber improved the surface properties. Silane and peroxide treatment of flax led to a higher tensile strength than that of untreated flax (Wang et al., 2007).
4. Case study: hybrid glass/natural fibre composites for curved pipes

4.1 Case study outline
The case study presented here refers to the analysis of the hybridization of glass fibres with natural fibres for applications in the piping industry (Cicala et al, 2009). The natural fibres studied were hemp, flax and kenaf. The pipe selected for the study was a curved fitting (90°) flanged at both ends designed to withstand an internal pressure of 10 bar and in the presence of acid aqueous solutions. This type of fitting is widely used in chemical plants which bear acid solution. The actual fittings are manufactured by hand layup with a complex sequence of glass mats and fabrics impregnated with epoxy vinyl ester resins. The problem was how to save cost without significant loss in mechanical properties and solvent resistance. Natural fibres mats were investigate as an alternative to glass mats.

4.2 Experimental
A commercial epoxy vinyl ester resin was used as thermost set matrix. Several glass fabrics were used varying from E-glass woven to E-glass random mat and C-glass liner (Table 5). The hemp mat was purchased by Hempcore Ltd., United Kingdom. Kenaf and Flax mats were kindly offered by Sachseinleinen GmbH.

<table>
<thead>
<tr>
<th>Component</th>
<th>Cost (€/m²)</th>
<th>Areal weight (g/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>E-glass woven</td>
<td>2.80</td>
<td>600</td>
</tr>
<tr>
<td>E-glass mat</td>
<td>2.13</td>
<td>600</td>
</tr>
<tr>
<td>C-glass liner</td>
<td>0.50</td>
<td>30</td>
</tr>
<tr>
<td>Hemp mat</td>
<td>0.31</td>
<td>600–650</td>
</tr>
<tr>
<td>Flax mat</td>
<td>0.37</td>
<td>750</td>
</tr>
<tr>
<td>Kenaf mat</td>
<td>0.33</td>
<td>630–650</td>
</tr>
</tbody>
</table>

Table 5. Technical data of the fabrics used

The lamina for mechanical testing were impregnated by hand lay-up and cured at room temperature for 48 h. The fittings were also manufactured by hand lay-up by wrapping the fabric onto a steel mandrel which is shown for reference in Fig. 24.

Fig. 24. (A) Steel mandrel used and (B) example of the fabric wrapping step

Tensile tests of single fibres (free fibre length was 15 mm), manually extracted from each mat, were carried out with a speed of 1 mm/ min.
The cured laminas were tested accordingly to EN ISO 527 either on laminas obtained from a single fabric or on laminates obtained with a lay-up similar to those used for the fitting. Some laminate specimens were also conditioned in different HCl solutions with pH varying from 1 to 7. The specimens were immersed for 40 days and then tested to analyze the effect on mechanical properties. This test was designed to predict the mechanical behaviour of the specimens in real working conditions. All the specimens were wrapped with C-glass liner to simulate the real surface of the interior of the fittings which is usually exposed to acid solutions.

4.3 Results and discussion
The results of tensile testing on single ply lamina are summarized in Fig. 25 for the tensile strength and modulus respectively normalized with respect to the density of each lamina. Bending showed similar results. The lamina reinforced with glass woven fabric showed the best performances in terms of tensile strength and modulus. This result is the consequence of the presence of long and aligned continuous glass fibres. The glass mat showed better mechanical properties compared to the natural fibre mats. The decrease of tensile strength compared to neat resin was observed for the lamina obtained from natural fibre mats. However, slight improvements of tensile modulus were observed compared to neat resin for the same samples. This behaviour can be explained as a consequence of the low fibre volume fraction ($V_f$) achieved for the lamina reinforced with the natural fibres and of the scarce adhesion between fibre and matrix. The latter and matrix were due to the absence of surface treatment on the fibres used in the present study. The natural fibre surface was not treated because this choice avoids to increase the price of the natural fibre. Measurements of $V_f$ were performed on the natural fibre mat samples and an average of 8–11% was obtained. The reason for such low $V_f$ are twofold: the hand lay-up method does not allow to achieve high compaction pressure and poor control on resin quantity is obtained; the natural fibres have a porous structure that increase the amount of resin adsorbed when lamina are impregnated. Moreover, the architecture of the natural fibre mats is quite open and thus higher percentages of resin are allowed to impregnated the mat. If liquid molding techniques like RTM (Resin Transfer Moulding) were employed for the manufacturing a $V_f$ of 30% could be achievable. Table 6 reports the mechanical data of Fig. 25 after normalization to a $V_f$ of 30%. The data clearly show that natural fibres can compare to glass fibres also in terms of mechanical performances if higher volume fraction of natural fibres are achieved.

The laminate sequence leads to a thickness of 11.92 mm and a cost for the fittings of 15.74€ in terms of raw materials cost) with a weight of 2.97 kg. The laminates for fittings which are currently manufactured present the following ply sequence: [C/ C/ M/ W/ M/ W/ M/ M/ W/ M/ W/ M] where C stands for C-glass liner, M for E-glass mat and W for E-glass woven. The resistance of the laminate sequence was verified accordingly to the Tsai–Hill criterion and to the maximum tension criterion using the data from single lamina testing for the calculations. The calculations were carried out for each single ply considering the relative position in the lay-up sequence (table 7).

Accordingly to this finding and taking into account the cured ply thickness of the hemp mat the following alternative design was proposed for the fittings in order to achieve a pipe thickness similar to the original pipe construction: [C/ C/ $M_n$/ W/ W/ $M_n$] where $M_n$ stands for the natural fibre mat.

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Fig. 25. Specific tensile strength and modulus on single lamina

Table 6. Mechanical properties of single lamina after normalization to $V_f$ of 30%

<table>
<thead>
<tr>
<th>Material</th>
<th>$V_f$ (%)</th>
<th>Tensile strength (MPa g$^{-1}$ cm$^2$)</th>
<th>Tensile modulus (GPa g$^{-1}$ cm$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>E-glass woven</td>
<td>35</td>
<td>127.64</td>
<td>8.22</td>
</tr>
<tr>
<td>E-glass mat</td>
<td>20</td>
<td>83.78</td>
<td>7.26</td>
</tr>
<tr>
<td>Hemp mat</td>
<td>12</td>
<td>42.95</td>
<td>7.00</td>
</tr>
<tr>
<td>Flax mat</td>
<td>10</td>
<td>60.24</td>
<td>8.04</td>
</tr>
<tr>
<td>Kenaf mat</td>
<td>9</td>
<td>49.50</td>
<td>9.17</td>
</tr>
</tbody>
</table>

Table 7. Calculations according to the maximum tension and the Tsai-Hill criterion

The novel hybrid lay-up has been used to predict the cost (raw material) and the weight of the fittings produced using natural mat as replacement of glass mat. The results are summarized in Figs. 26 and 27 where the data for the original lay-up (named Glass) is reported for comparison purposes.
The comparison shows that the novel lay-up allows for the reduction of cost and weight for all the types of the natural fibres selected. The best performances were obtained with the hemp mat. A prototype of the fitting was build with the proposed laminate sequence using the hemp mat and it was tested under pressure up to 16 bar without any significant deformation or fluid leakage.

Finally some laminates were tested after immersion in aqueous acid solutions for 40 days. In order to have significant data that laminates were wrapped with C-glass liner impregnated with the resin. This construction of the test lamina allows reproducing the conditions of the internal layer of the pipe which is usually exposed to the acid solution. The mechanical test showed that only small variations of the mechanical properties after immersion were obtained. The resistance to acid solution is a consequence of the barrier effect of the liner wrapping.


5.1 Case study outline
The objective of this case study is to compare the mechanical properties of twisted hemp fabric with hemp mats as viable reinforcement for composites. It has been mentioned
previously that hemp mats do not represent a fabric with optimised properties for composites reinforcement due to their random fibre orientation. To overcome the limitation offered by mats the use of fabric made with aligned yarn has been investigated. Two fabric architectures were considered: unidirectional and twill 2x2.

5.2 Experimental

The general purpose unsaturated polyester resin ECMALON 4411, purchased by Ecmass Resins Pvt. Ltd, India, was used as thermoset matrix. Methyl ethyl ketone peroxide (MEKT) and cobalt naphthenate were purchased by Aldrich, Italy, and used as catalyst and accelerator respectively. 3-aminopropyltriethoxysilane (A1100) was purchased from Aldrich, Italy, and used without further purification.

Several hemp fabrics were used in this study, varying from random mat fabric, purchased by Hempcore Ltd., United Kingdom, to unidirectional [0°] and bidirectional [0°/90°] woven fabrics purchased by Canipificio Italiano, Italy. The woven fabrics were obtained weaving yarns of natural fibres made of stable filaments twisted together.

Methyl ethyl ketone peroxide (MEKT) and cobalt naphthenate were added at room temperature at percentages of 1.5 wt% and 0.07 wt% respectively. Hand layup was used to prepare the laminates for mechanical testing. Each composite was cured at room temperature for 48 h.

The cured laminas were tested accordingly to EN ISO 527 for tensile test. Five replicas for each specimen were tested. Tensile test was carried out with a Zwick universal testing machine (model Z050) equipped with a 50 kN load cell. The experiment was performed in displacement-control mode at a stroke rate (i.e. cross-head displacement rate) of 2 mm/s. All output data (strain, displacement of cross-head, and load) were collected by an acquisition system and transferred to the PC.

5.3 Results and discussion

The mechanical properties of the laminates reinforced by mat are reduced by a factor of about 3/8 because of the random distribution of the fibres. To overcome this limitation the use of weaved fabrics made of twisted yarns has been considered here. Two architectures, namely, unidirectional (UD) and 0°/90° were considered (Fig.28). The laminates were obtained by hand layup. The results of tensile testing obtained for laminates prepared with these fabrics are summarized in Fig. 29.

Fig.29 clearly shows that both modulus and strength are greatly enhanced when twisted yarns are used despite their low mechanical properties in dry form compared to single fibres extracted from hemp mats. This finding is the outcome of the impregnation of the yarns with the resin which, upon curing, stabilizes the yarn reducing the sliding effect of the filaments. The good properties measured for the composites reinforced with hemp is the results of the favourable orientation, along the loading direction, of the staple fibres of the yarns. As it can be expected the 0°/90 fabrics present lower mechanical performances compared to unidirectional fabrics. This result is due to the presence in the 0°/90 fabric of yarns directed transversely compared to loading tensile direction. The modulus and strength reported in Fig. are slightly lower than the values found in literature because of the manufacturing method (ie. hand layup) selected and of the low fibre volume fraction achieved.
6. Conclusions

The present chapter was focused on the use of natural fibre fabric as reinforcement for composite materials. The environmental and cost benefits connected with the use of natural fibre based fabrics are at the basis of their wide success. However, several limitations must be overcome in order to exploit the full potential of natural fibres. At first proper fibre surface treatment should be developed and implemented at industrial scale. Secondly, the use of mats should be investigated and the hybridization of mats with different textile further improved by analysing the effects of different layup and manufacturing techniques. Finally, the use of advanced textile based on twisted yarn should be developed further by optimising the yarn manufacturing and realising 3D architectures which are still missing from the market.

7. References


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The main goal in preparing this book was to publish contemporary concepts, new discoveries and innovative ideas in the field of woven fabric engineering, predominantly for the technical applications, as well as in the field of production engineering and to stress some problems connected with the use of woven fabrics in composites. The advantage of the book Woven Fabric Engineering is its open access fully searchable by anyone anywhere, and in this way it provides the forum for dissemination and exchange of the latest scientific information on theoretical as well as applied areas of knowledge in the field of woven fabric engineering. It is strongly recommended for all those who are connected with woven fabrics, for industrial engineers, researchers and graduate students.

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