

# Properties and Modification Methods for Vegetable Fibers for Natural Fiber Composites

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## SYNOPSIS

Studies on structure and properties of natural vegetable fibers (NVF) show that composites made of NVF combine good mechanical properties with a low specific mass. The high level of moisture absorption by the fiber, its poor wettability, as well as the insufficient adhesion between untreated fibers and the polymer matrix lead to debonding with age. To build composites with high mechanical properties, therefore, a surface modification of the fibers is necessary. The existing physical and chemical NVF modification methods—e.g., plasma treatment or graft copolymerization—which are used for the development of NVF-polymer composite properties is discussed. It is shown that modified cellulose fiber-polymer interaction mechanisms are complex and specific to every definite system. By using an coupling agent, like silanes or stearin acid, the Young's modulus and the tensile strength increases, dependent on the resin, until 50%. Simultaneously, the moisture absorption of the composites decreases for about 60%. With other surface modifications, similar results are obtained. © 1996 John Wiley & Sons, Inc.

## INTRODUCTION

Fiber-reinforced plastics are structural composites, in which a polymer matrix is combined with fibers, fabric cuttings, or filling webs. In conventional composites, the fibers are made of glass, aramid, or carbon. The advantage of fiber-reinforced plastics results from their structure. The controlled structure in which the fibers are lying in the polymer matrix increases the strength and the stiffness of the composite. At present, natural vegetable fibers (NVF) are used in composites, where high strength and stiffness are not of first priority. NVF reduce the mass of the composite, because they have a low density. Their production is economical, with low requirements on equipment, and they can easily be recycled.

Conventional fibers (glass, aramid, carbon, etc.) can be produced with a definite range of properties. The characteristic properties of NVF vary considerably. This depends on whether the fibers are taken

from the plant stems or leaves,<sup>1</sup> the quality of the plants location,<sup>2</sup> and on the preconditioning.<sup>3,4</sup>

Depending on their origin, vegetable fibers can be grouped into seed, bast, leaf, or fruit qualities. Bast and leaf qualities are so-called hard fibers (e.g., flax, jute, and ramie)<sup>1,5-7</sup> and are the most used ones. In practically all cases, cellulose is the main component of vegetable fibers (Table I).

The elementary unit of a cellulose macromolecule (Fig. 1) is anhydro-*d*-glucose, which contains three alcohol hydroxyls (—OH). These hydroxyls form hydrogen bonds inside the macromolecule itself (intramolecular) and between other cellulose macromolecules (intermolecular) as well as with hydroxyl groups from the air. Therefore, all vegetable fibers are of a hydrophil nature; their moisture content reaches 8–12.6% (Table I).

An important characteristic of vegetable fibers is their degree of polymerization (DP). The cellulose molecules of each fiber differ in their DP and, consequently, the fiber is a complex mixture of polymer homolog (C<sub>6</sub>H<sub>10</sub>O<sub>5</sub>)<sub>n</sub>. Bast fibers commonly show the highest DP among all the NVF (approximately 10,000).<sup>6</sup>

The fibrils of the cellulose macromolecules form spirals along the fiber axis.<sup>8</sup> The strength and stiff-

**Table I** Chemical Composition and Structure Parameters of Vegetable Fibers

Fiber	Cellulose (Wt %)	Cell Length (mm)	Spiral Angle (Deg)	Allied Substances (Wt %)				Moisture Content (Wt %)	Refs.
				Hemi-cellulose	Pectins	Lignin	Wax		
Jute	61.0	2.3	8.0	20.4	0.2	13.0	0.5	12.6	8, 9
Flax	71.0	20.0	10.0	18.6	2.3	2.2	1.7	10.0	8, 9
Hemp	74.4	23.0	6.2	17.9	0.9	3.7	0.8	10.8	8, 9
Ramie	68.6	154.0	7.5	13.1	1.9	0.6	0.3	8.0	8, 10
Sisal	78.0	2.2	20.0	10.0	—	8.0	2.0	11.0	8, 11
Coir	43.0	0.8	45.0	0.3	4.0	45.0	—	8.0	8, 11

ness of hemp, ramie, and jute correlates with the angle between the axis and the fibril of the fiber. The smaller this angle is, the higher are the mechanical properties (Table II). The cellulose of NVF contains different natural substances (Table I). The most important of them are lignin and several waxes. The distinct cells of hard NVF are bonded together by lignin (or lignin/hemicellulose), acting as a cementing material.<sup>15</sup> The lignin content of the bast fibers influences its structure,<sup>6</sup> its properties,<sup>2,6,8</sup> and morphology.<sup>16</sup> The waxy substances of vegetable fibers can be eliminated by extraction with organic solvents. They are the cause of the fiber's wettability and adhesion characteristics.<sup>17,18</sup>

Comparative values of cellulose and conventional reinforcing fibers (Table II) show the high attraction for composites made of ramie, flax, and jute fibers. Their density values make it possible to produce composites which combine good mechanical properties with a low specific mass.

## NVF COMPOSITES

The properties of composites depend on those of the individual components and on their interfacial compatibility. The adhesion between fiber and matrix is obtained by the mechanical anchoring of the fiber ends into the matrix. Polymers for which the chemical bonding with reinforcements made of cellulose is known are methylolfunctional polymers, such as phenol-formaldehyde resins.<sup>19</sup> Methylol groups react with the hydroxyl groups of the cellulose to form stable ether linkages. So, a high compatibility in the system cellulose and polymer is achieved. Unfortunately, this is the only example of a high compatible cellulose-polymer compound. In other cases, the absorption of moisture by untreated fibers, poor wettability, and insufficient adhesion

between polymer matrix and fiber leads in time to debonding.<sup>1,7</sup> Without effective wetting of the fiber, strong interfacial adhesion cannot exist. The lack of interfacial interactions leads to internal strains,<sup>20</sup> porosity, and environmental degradation.<sup>21,22</sup> The wettability of the fiber depends on the viscosity of the polymer and the surface tension of both materials. The surface tension of the polymer must be as low as possible,<sup>23</sup> at least lower<sup>19,24</sup> than the surface tension of the fiber. There are different methods of modification to change the surface energy of the fiber<sup>25</sup> and the polymer. The fibers' minimum value of surface tension must be 43 din/cm in the case of epoxide (EP) and 35 din/cm in the case of unsaturated polyester (UP) resins.<sup>24</sup>

Therefore, the modification of the fiber or/and the polymer matrix is a key area of research at present to obtain optimum fiber matrix properties.

## PHYSICAL METHODS OF NVF MODIFICATION

Reinforcing fibers can be modified by physical and chemical methods. Physical methods, such as stretching,<sup>18</sup> calendering,<sup>26,27</sup> thermotreatment,<sup>4</sup> and the production of hybrid yarns<sup>28,29</sup> do not change the chemical composition of the fibers. Physical treatments change structural and surface properties of the fiber and thereby influence the mechanical bondings in the matrix.

Electric discharge (corona, cold plasma) is another way of physical treatment. Corona treatment is one of the most interesting techniques for surface oxidation activation. This process changes the surface energy of the cellulose fibers,<sup>25</sup> and in the case of wood, surface activation increases the amount of aldehyde groups.<sup>30</sup>

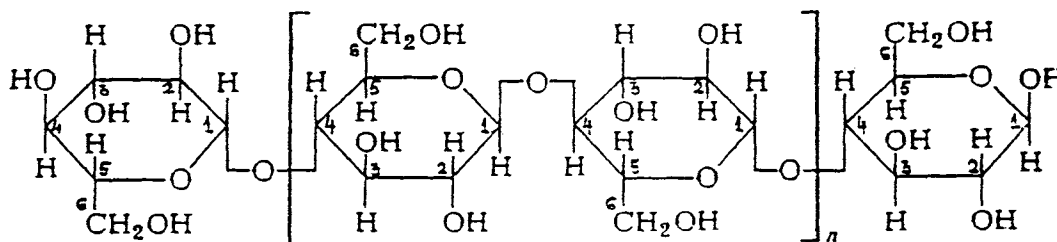


Figure 1 Macromolecules of cellulose.<sup>12</sup>

The same effects are reached by cold plasma treatment. Depending on the type and nature of the used gases, a variety of surface modifications can be achieved. Surface crosslinkings could be introduced, surface energy could be increased or decreased, and reactive free radicals<sup>25</sup> and groups<sup>31</sup> could be produced.

Electrical discharge methods are known<sup>32</sup> to be very effective for “nonactive” polymer substrates such as polystyrene, polyethylene, and polypropylene. They are successfully used for cellulose fiber modification to decrease the melt viscosity of cellulose-polyethylene composites<sup>33</sup> and to improve mechanical properties of cellulose-polypropylene composites.<sup>25</sup>

An old method of cellulose fiber modification is mercerization<sup>8,12,17,18,34</sup>, it has been widely used on cotton textiles. Mercerization is an alkali treatment of cellulose fibers; it depends on the type and concentration of the alkalic solution, its temperature, time of treatment, tension of the material, as well as on the additives.<sup>17,34</sup> At present, there is a tendency to use mercerization for NVF as well. Optimal conditions of mercerization ensure the improvement of the tensile properties<sup>8,12,13,34</sup> and absorption char-

acteristics,<sup>8,12,34</sup> which are important in the composing process.

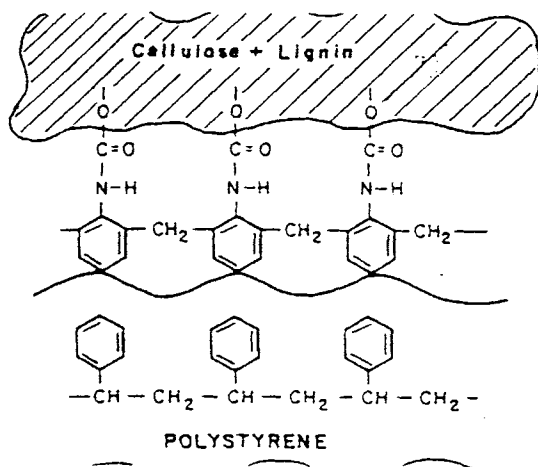
### CHEMICAL METHODS OF MODIFICATION FOR NVF

Strongly polarized cellulose fibers<sup>23</sup> inherently are little compatible with hydrophobic polymers.<sup>20,25,35,36</sup> When two materials are incompatible, it is often possible to bring about compatibility by introducing a third material that has properties intermediate between those of the other two. There are several mechanisms<sup>19</sup> of coupling in materials:

- Weak boundary layers—coupling agents eliminate weak boundary layers.
- Deformable layers—coupling agents produce a tough, flexible layer.
- Restrained layers—coupling agents develop a highly crosslinked interphase region with a modulus intermediate between that of the substrate and the polymer.

Table II Comparative Values of Cellulose and with Conventional Reinforcing Fibers

Fiber	Density (g/cm <sup>3</sup> )	Elongation to Break (%)	Tensile Strength (MPa)	Young's Modulus (GPa)	Cost (\$/kg)	Refs.
Jute	1.45	1.5	550	13	0.3	1, 8
Flax	1.50	2.4	1100	100	—	1, 8
Hemp	—	1.6	690	—	—	8
Ramie	1.50	1.2	870	128	—	8, 13
Sisal	1.45	2.0	640	15	0.36	1, 8
Coir	1.15	15.0	140	5	0.25	1, 8
E-glass	2.50	2.5	2000–3500	70	3.25	14
S-glass	2.50	2.8	4570	86	—	1, 14
Aramid (normal type)	1.40	3.3–3.7	3000–3150	63–67	—	14
Carbon (high tensile strength)	1.70	1.4–1.8	4000	230–240	> 16	14



**Figure 2** Hypothetical chemical structure of the cellulose-PMPPIC-PS interface area.<sup>36</sup>

- Wettability—coupling agents improve the wetting between polymer and substrate (critical surface tension factor).
- Chemical bonding—coupling agents form covalent bonds with both materials.
- Acid-base effect—coupling agents alter the acidity of the substrate surface.

The development of a definitive theory for the mechanism of bonding by coupling agents in composites is a complex problem. The main chemical bonding theory alone is not sufficient. So, consideration of other concepts appears to be necessary, which include the morphology of the interphase, the acid-base reactions at the interface, surface energy, and the wetting phenomena.

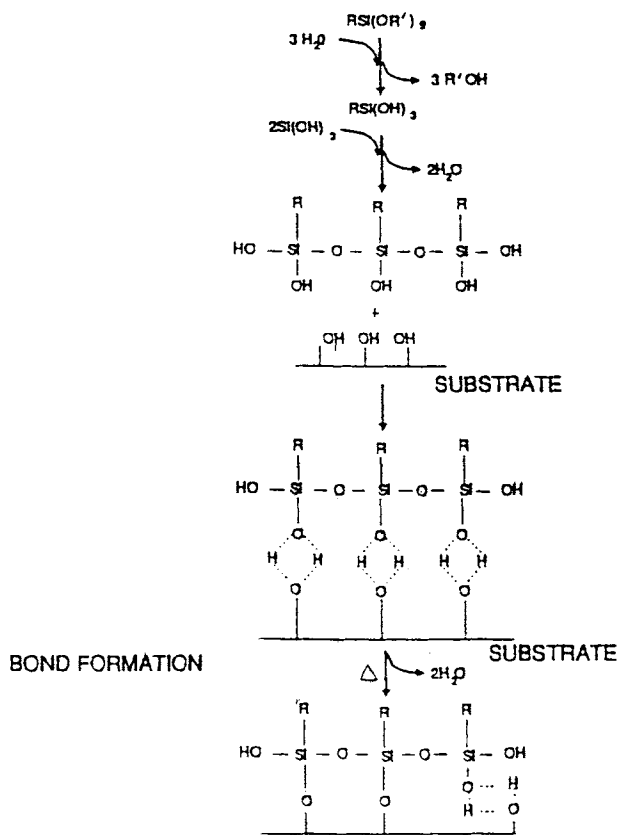
### The Change of Surface Tension

The surface energy of fibers is closely related to the hydrophilicity of the fiber.<sup>23</sup> Some investigations are concerned with methods to decrease hydrophilicity. The modification of wood cellulose fibers with stearic acid<sup>37</sup> hydrophobizes those fibers and improves their dispersion in polypropylene. As can be observed in jute-reinforced unsaturated polyester resin composites, treatment with polyvinylacetate increases the mechanical properties<sup>27</sup> and water repellence.

Silane coupling agents may contribute hydrophilic properties to the interface, especially when amino-functional silanes, such as epoxies and urethanes silanes, are used as primers for reactive polymers. The primer may supply much more amine functionality than can possibly react with the resin

**Table III** Characteristics of Representative Commercial Silane Coupling Agents

Organofunctional Group	Chemical Structure	OSi-Specialties Germany GmbH Product	Critical Surface Tension of Glass with Silane Treatment (dyne/cm)	Applied Polymers (Abbreviations According ASTM 1600)
Vinyl	$\text{CH}_2=\text{CHSi}(\text{OCH}_3)_3$	A-171	25.0	UP
Vinyl	$\text{CH}_2=\text{CHSi}(\text{OC}_2\text{H}_5)_3$	A-151	30.0	UP, PE, PP, DAP, EPDM, EPM
Chloropropyl	$\text{ClCH}_2\text{CH}_2\text{CH}_2\text{Si}(\text{OCH}_3)_3$	A-143	40.5	EP
Epoxy	$\begin{array}{c} \text{O} \\ \diagup \quad \diagdown \\ \text{CH}_2\text{CHCH}_2\text{O}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3 \end{array}$	A-187	38.5–42.5	EP, PA, PC, PF, PVC, PUR
Methacrylate	$\begin{array}{c} \text{CH}_3 \\   \\ \text{CH}_2=\text{C}-\text{COO}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3 \end{array}$	A-174	28.0	UP, PE, PP, DAP, EPDA, EPM
Primary amine	$\text{H}_2\text{N}(\text{CH}_2)_3\text{Si}(\text{OC}_2\text{H}_5)_3$	A-1100	35.0 } 33.5 }	UP, PA, PC, PUR, MF, PF, PI, MPF
Diamine	$\text{H}_2\text{N}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$	A-1120		
Cationic styryl	$\text{CH}_2=\text{CHC}_6\text{H}_4\text{CH}_2\text{N}^+\text{H}_2(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3\text{Cl}^-$	—	—	All polymers
Phenyl	$\text{C}_6\text{H}_5\text{Si}(\text{OCH}_3)_3$	—	40.0	PS, addition to amine silanes
Mercapto	$\text{HS}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$	A-189	41.0	EP, PUR, SBR, EPDM



**Figure 3** Scheme of the alkoxy silanes bond formation with hydroxyl groups containing the surface.<sup>19</sup>

at the interphase. Those amines which could not react are hydrophilic and therefore responsible for the poor water resistance of the bonds. An effective way to use hydrophilic silanes is to blend them with hydrophobic silanes such as phenyltrimethoxysilane. Mixed siloxane primers also have an improved thermal stability, which is typical for aromatic siloxanes.<sup>19</sup>

**Impregnation of Fibers**

A better combination of fiber and polymer is achieved by an impregnation<sup>38</sup> of the reinforcing

fabrics with polymer matrices compatible with the polymer. For this purpose, polymer solutions<sup>20,39</sup> or dispersions<sup>40</sup> of low viscosity are used. For a number of interesting polymers, the lack of solvents limits the use of the method of impregnation.<sup>38</sup> When cellulose fibers are impregnated with a butylbenzyl phthalate plasticized poly(vinyl chloride) (PVC) dispersion, excellent partitions can be achieved. In polystyrene (PS), this significantly lowers the viscosity of the compound and the plasticator and results in a co-solvent action for both PS and PVC.<sup>40</sup>

**Chemical Coupling**

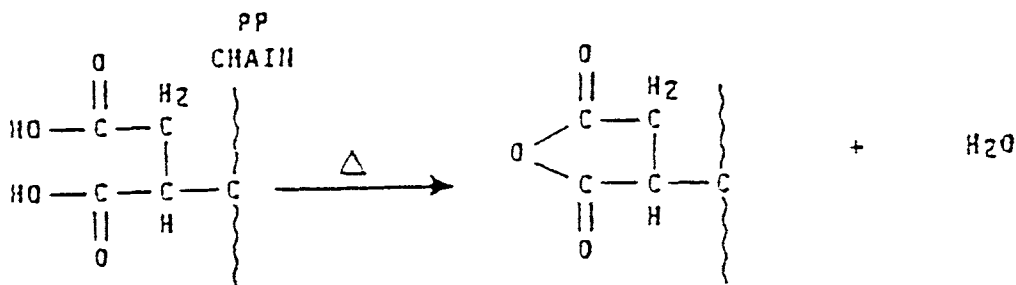
An important chemical modification method is the chemical coupling method. This method improves the interfacial adhesion. The fiber surface is treated with a compound that forms a bridge of chemical bonds between the fiber and matrix.

**Graft Copolymerization**

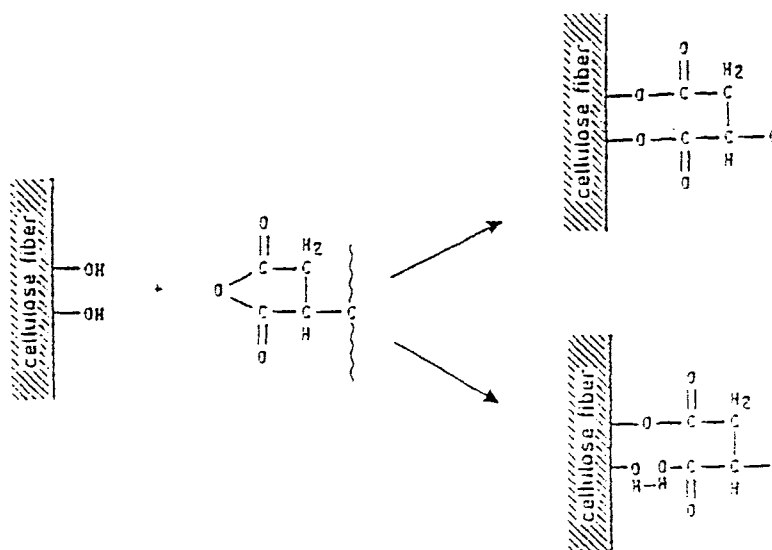
An effective method of NVF chemical modification is graft copolymerization.<sup>8,12</sup> This reaction is initiated by free radicals of the cellulose molecule. The cellulose is treated with an aqueous solution with selected ions and is exposed to a high-energy radiation. Then, the cellulose molecule cracks and radicals are formed. Afterward, the radical sites of the cellulose are treated with a suitable solution (compatible with the polymer matrix), e.g., vinyl monomer,<sup>12</sup> acrylonitrile,<sup>8</sup> methyl methacrylate,<sup>41</sup> and polystyrene.<sup>36</sup> The resulting copolymer possess properties characteristic of both fibrous cellulose and grafted polymer.

For example, the treatment of cellulose fibers with hot polypropylene-maleic anhydride (MAH-PP) copolymers provides covalent bonds across the interface.<sup>20</sup> The mechanism of reaction can be divided into two steps:

- Activation of the copolymer by heating ( $t = 170^\circ C$ ) (before fiber treatment):



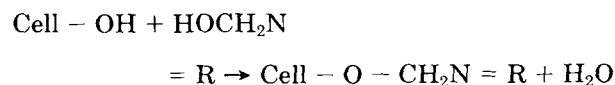
- Esterification of cellulose:



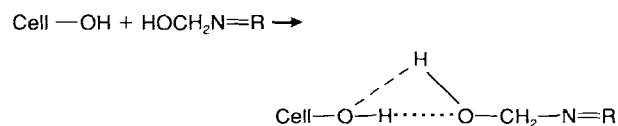
After this treatment, the surface energy of the fibers is increased to a level much closer to the surface energy of the matrix. Thus, a better wettability and a higher interfacial adhesion is obtained. The PP chain permits segmental crystallization and cohesive coupling between the modified fiber and the PP matrix.<sup>20</sup> The graft copolymerization method is effective, but complex.

#### Treatment with Compounds Which Contain Methylol Groups

Chemical compounds which contain methylol groups ( $-\text{CH}_2\text{OH}$ ) form stable, covalent bonds with cellulose fibers. Those compounds are well known and widely used in textile chemistry:



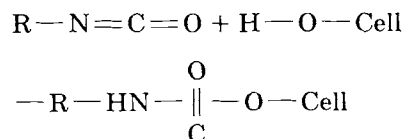
Hydrogen bonds with cellulose can be formed in this reaction as well:



The treatment of cellulose with methylolmelamine compounds before forming cellulose unsaturated polyester (UP) composites decreases the water intake and increases the wet strength of reinforced plastic.<sup>22,42</sup>

#### Treatment with Isocyanates

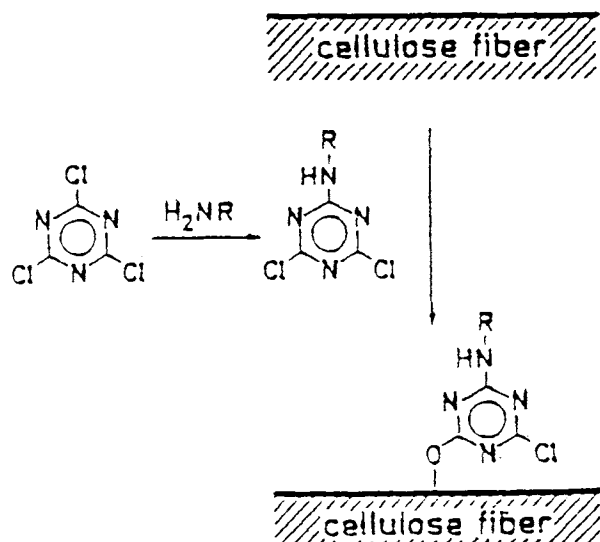
The mechanical properties of composites reinforced with wood fibers and PVC or PS as the resin can be improved by an isocyanate treatment of those cellulose fibers<sup>36,43</sup> or the polymer matrix.<sup>43</sup> Polymethylene-polyphenyl-isocyanate (PMPPIC) in the pure state or in solution in the plasticizer can be used. PMPPIC is chemically linked to the cellulose matrix through strong covalent bonds:



Both PMPPIC and PS contain benzene rings and their delocalized  $\pi$  electrons provide strong interactions, so that there is an adhesion between PMPPIC and PS (Fig. 2). Comparing both methods, treatment with silanes or treatment with isocyanates, it is obvious that the isocyanatic treatment is more effective than is the treatment with silane. Equal results are obtained when PMPPIC is used for the modification of the fibers or the polymer matrix.<sup>36</sup>

#### Triazine Coupling Agents

Triazine derivatives form covalent bonds with cellulose fibers schematically:

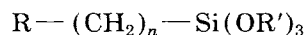


The reduction of the water absorption of cellulose fibers and their composites treated with triazine derivatives is explained by<sup>7,44</sup>

- Reducing the number of cellulose hydroxyl groups which are available for water uptake,
- Reducing the hydrophilicity of the fiber's surface, and
- Restraint of the swelling of the fiber, by creating a crosslinked network due to covalent bonding, between the matrix and fiber.

### Organosilanes as Coupling Agents

Organosilanes are the main group of coupling agents in glass-reinforced polymers. They have been developed to couple virtually any polymer to the minerals, which are used in reinforced composites.<sup>19</sup> Most of the silane coupling agents can be represented by the following formula:



with  $n = 0-3$ ; OR', the hydrolyzable alkoxy group; and R, the functional organic group.

The organofunctional group (R) in the coupling agent causes the reaction with the polymer. This could be a copolymerization and/or the formation of an interpenetrating network. This curing reaction of a silane-treated substrate enhances the wetting by the resin (Table III).

The general mechanism of how alkoxy silanes form bonds with the fiber surface which contains hydroxyl groups is depicted in Figure 3. Alkoxy-

silanes undergo hydrolysis, condensation (catalysts for alkoxy silane hydrolysis are usually catalysts for condensation), and the bond-formation stage, under base- as well as under acid-catalyzed mechanisms. In addition to these reactions of silanols with hydroxyls of the fiber surface, the formation of polysiloxane structures also can take place.

Analogous to glass fibers, silanes are used as coupling agents for natural fiber-polymer composites. For example, the treatment of wood fibers with product A-175 improves wood's dimensional stability.<sup>45</sup> In contrast, a decrease of mechanical properties was observed for coir-UP composites after a fiber modification with dichloromethylvinylsilane.<sup>46</sup> The treatment of mercerized sisal fiber with aminosilane 1100 (Ref. 16) before forming sisal-epoxy composites markedly improves the moisture resistance of the composite. These examples show that theories used for the silane treatment of NVF are not perfect and are contradictory; therefore, further studies are necessary.

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