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Recent Advances on Lignocellulosic-Based Composites for Performance and Environmentally-Compatibility Improvement

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Recent Advances on Lignocellulosic-Based Composites for Performance and Environmentally-Compatible Improvement

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To write this article, more than 300 articles were overviewed, that covered what has been published in the most important journals of the research area of Wood Science and Technology, from 2002 to 2006. From those, 115 are referred in this article. This review presents the latest results from research on wood composites, and more broadly, on lignocellulosic-based composites, aimed, obviously, at improving performance in service, but with a special emphasis on research progresses that have bringing about environmental benefits, too. Topics covered include chemical modification, nonwood fiber resources, thermoplastic-based composites, composites from waste or recycled lignocellulosics, improvement of biodegradation resistance, adhesives based on renewable chemical resources, low- or near-zero-emission formaldehyde-based adhesives, reduction of volatile organic compounds emissions, structural applications, inorganic binders, LCA – Life-Cycle Assessment, heat treatment, binderless composites.

Keywords: composites; environmental benefits; improvement; lignocellulosics; review; wood

INTRODUCTION

Less Common Wood Species/Raw Materials

Exploring the possibilities of using less common wood and lignocellulosic raw materials may be a means of overcoming the shortage of

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common industrial species, lowering production costs and/or reducing the pressure on natural forests of environmental value. Examples on this can be given, as: LVL from *Pterocarya fraxinifolia* [1], MDF from a mixture of two exotic larch species (*Larix gmelinii* and *Larix sibirica*) [2], PB from Korean thinning logs (*Pinus rigida*, *Pinus densiflora*, *Larix leptolepis* and *Quercus acutissima* [3], PB, MDF and structural flakeboard from Chinese tallow tree (a noxious, invasive plant in the southeastern U.S.A.) [4], MDF from a mixture of wood and bark of hybrid poplar, jack pine, red pine and white spruce grown in Canada [5], OSB from a mixture of quebracho blanco (Argentina) and loblolly pine (this latter to lower density) [6].

INFLUENCE OF FURNISH CHARACTERISTICS ON PRODUCT PROPERTIES

To study the influence of the characteristics of raw materials on the properties of the final product can lead, obviously, to a product improvement, to rise process productivity, to lower raw materials and energy consumption levels, and, as a consequence, to lower production costs.

Preheating MDF fiber mat using microwave energy before hot-pressing increased the overall mat temperature within a very short period of time leading to a shortening of the hot-press cycle [7]. The addition of thermally conductive fillers (for example, synthetic graphite, natural graphite, boron nitride) to strandboard furnish increased IB (an indirect measure of degree of resin cure) [8]. The higher thermal conductivity of synthetic graphite ($600 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$), as compared to the low thermal conductivity of wood ($0.2 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$), as been highlighted [9].

COMPOSITES IN STRUCTURAL APPLICATIONS

The application of special wood composites for structural applications is not a new issue. However, some of the advantages of replacing solid wood by wood composites should be highlighted here once more. Composites can be made by wood residues, their properties are usually more consistent than wood, and such properties can be targeted to specific applications. With the reduction in the availability of big-diameter roundwood, either by an over exploration or by environmental protection regulations, composites enable the use of small roundwood, lower quality wood, etc.

OSB, okoume PY and MDF were found to be viable as an alternative to solid wood (Scotch pine and Turkish beech) for wood chair

frames [10]. In I-beams, PB and MDF showed competent performance in comparison with the most commonly used OSB [11].

ADHESIVES AND WOOD RECYCLING

One of the problems often encountered when one aims at making use of wood residues to make composites is the compatibility of the adhesive with the raw material; i.e., the resin may not cure in an extent enough to give the desired properties of the product. Among other reasons, the furnish might be contaminated or can come from preservative-treated wood. A laboratory synthesized PRF adhesive, with a high resorcinol content, fulfilled the applicable ASTM requirements in bonding CCA-treated pine lumber [12]. MUF, MF and PF resins produced a relatively strong adhesive strength with copper azole-treated pine and larch woods [13].

ADHESIVES FROM RENEWABLE CHEMICAL RESOURCES

To find chemicals from renewable sources, mainly from plants, has been and still is an orienting principle for research in many fields, as well as environmental protection is. The advantages have been so far and often stated elsewhere, that they are not highlighted here once more. In general, radiata pine bark tannin-epoxide combined resin systems behave similarly to tannin-paraformaldehyde systems, especially at basic pH values [14]. RTF copolymer resins, prepared by using bark extracts of Taiwan acacia and China fir to substitute part of the resorcinol, showed bonding strength the same as RF resin, but the RTF had worse stability and shorter shelf life than RF resin [15]. In the presence of ammonia, both quebracho and radiata pine bark extracts showed accelerated adhesive cure behavior with formaldehyde, in a similar manner to PRF adhesive. But adhesive blends of quebracho extract and PRF failed to attain satisfactory bond strength at full cure. Of the pine bark components, only a pure tannin fraction produced acceptable bond strength when blended with PRF [16]. Blends of hydrolyzed tannin, cashew nut shell liquid (CNSL), and UF cured faster and resulted in PB made from coffee husks with better water and moisture resistance when compared to UF alone [17]. Kraft pine lignin was successful as a phenol substitute in LPF resins, as it presented higher amounts of activated free ring positions, higher MW and higher thermal decomposition temperature [18]. Softwood bark pyrolysis oils could be mixed at ratios as high as 40% with pMDI and enabled then to manufacture acceptable interior grade particle-board [19].

MDF boards bonded with blood-based PF-cross-linked protein resin were comparable to PF-bonded boards. Soy-protein-based phenolic resin enabled to meet requirements for interior MDF; and with flake-board, this was inferior to PF-bonded board but outperformed PF-bonded board in accelerated aging tests [20].

VOC'S – VOLATILE ORGANIC COMPOUNDS

VOC emissions to the atmosphere are of concern because, in urban air, they can become involved in, or lead to, the generation of ozone (O_3), a secondary air pollutant. This happens as a consequence of the so-called photochemical reactions, that need also nitrogen oxides, and are promoted by intense sunlight and high air temperatures. The forest product industries are just one of the many sources of such air pollutants. But they should be far from being the most important one. Measurement of VOC emissions with OSB samples from a new house constructed using structural engineered panels (samples uncoated, coated with an oil-based primer, with topcoat latex or with both) showed that the highest total VOC emissions were measured with the primer-coated OSB [21]. Emissions from OSB made of Scots pine measured 24 h after hot-pressing over a period of 2 months, resulted in that terpene emissions decreased continuously, whereas aldehyde concentrations initially increased and subsequently decayed: It was concluded that for a reduction in VOC emissions from wood-based materials, wood properties, manufacturing process, and storage conditions need to be considered [22].

The most prominent factor affecting the emissions of TVOC's during hot-pressing of radiata pine MDF was pressing temperature, followed by pressing time. Resin content appeared to have no effect on TVOC emissions although it was the most important factor influencing formaldehyde emissions [23]. Terpene emissions from OSB made of Scots pine were lowered with elevated pressing times, whereas the formation of volatile aldehydes was accelerated. In this study it was found that a sustainable reduction in aldehyde emissions by adjusting the relevant process parameters did not seem to be feasible [24]. Furthermore, if the surface consisted of fine particles, terpene emissions would be lowered and the course of aldehyde formation would be altered [25].

For panels pressed with both UF and PF resins, mat MC significantly affected formaldehyde emission, but mat resin content and panel density did not have a significant effect on formaldehyde emissions. An additive in the PF resin decreased formaldehyde emissions, so that pressing panels with the PF resin yielded lower formaldehyde

emissions than pressing with pMDI resin, and application of pMDI caused a significant decrease in HMwVOC emissions [26].

Epoxy powder coatings used to finish the MDF samples achieved 99% emission reduction in formaldehyde and up to 94% reduction of TVOC emissions. The respective formaldehyde/TVOC emission reductions were 99%/88% for phenolic paper laminates [27].

OTHER EMISSIONS TO THE ATMOSPHERE

PM10 emissions derived from wood processing in Switzerland's wood-working industry were estimated: the upper extrapolation limits estimated for national PM10 wood dust emissions were 110 t/a and 318 t/a, which corresponds to a maximum of 1% or 3% of the total national industry emissions [28].

FORMALDEHYDE-SPECIFIC ISSUES

For sure that formaldehyde is a VOC. However, having a chapter dedicated to it can be justified by the concerns brought by this chemical, very common in natural products, even many kinds of food products, but that is at the basis of discussions on the effects on health of the application of formaldehyde-based adhesives in many kinds of wood composites, when used indoors, on commercial and market competition issues, etc. It should not be forgotten that many wood species are, naturally and just by themselves, a measurable source of formaldehyde.

A formaldehyde-free wood adhesive system has been developed, consisting of soy protein (SP) and 1,3-dichloro-2-propanol (DCP), this latter serving as a crosslinking agent for SP [29]. An emission of formaldehyde equal to that of unbounded wood could be obtained by the use of glyoxal as hardener in radiata pine tannin extract-based adhesives [30]. Mimosa tannin hardened with hexamine at pH 10 has shown both at the laboratory and industrial level to be a formaldehyde-free system [31].

Release of formaldehyde from PY was greatly reduced by treatment of PY with microwave radiation: concentration of formaldehyde released increased with microwave irradiation and, after the microwave treatment, it decreased to a level below that of nontreated material [32]. A non-formaldehyde cross-linking system composed of citric acid (CA) and 1,2,3,4-butanetetracarboxylic acid (BTCA) with sodium hypophosphite (SHP) as catalyst was developed. Further intensification of the wood (fir and beech) modification process and

an increase in the quality of wood-based materials were achieved using microwave treatment [33]. Formaldehyde emissions from particleboards bonded with pine and wattle tannin-based adhesives, using paraformaldehyde, hexamethylenetetramine and TN (tris(hydroxyl)-nitromethane) as hardeners, satisfied grade E₁ [34]. A Versar mathematical model, developed by US-EPA, is available as a tool for making home projections of formaldehyde concentrations [35].

INORGANIC BINDERS

Composites made with inorganic binders, like cement and gypsum, instead of organic synthetic ones, like PF, MUF, PF, or adhesives incorporating natural products, like polyphenols, have many advantages and some disadvantages: formaldehyde emissions are only originated in the raw material and, therefore, are minimal; usually they present high resistances to moisture, fire and biodeterioration; inorganic binders many times are compatible with wood or lignocellulosic residues, more than the organic binders. On the other hand, if not special techniques are applied they need a long pressing time, to allow cement or gypsum hardening, and density is higher.

Injection of pure CO₂ gas as well as diluted (about 25% CO₂ concentration in air) greatly reduced (by two orders of magnitude) the press time required to yield dimensionally stable products of sufficient strength for initial handling, thus lowering production costs [36]. Accelerated processing techniques involving carbonation reactions were successfully employed to produce cement-based board products incorporating wheat straw, by overcoming hydration inhibitory effects [37]. Also, suitable homogeneous cement-bonded particleboards were successfully made from maize stalk with Portland cement and CaCl₂ has catalyst [38].

Cork granules are of low density and may be used as lightweight aggregates for making concrete. Beyond the most valuable cork product, that is cork stoppers, a percentage of about 87% cork is left, when calculated taken as a basis all grades of cork taken from cork oak stands. But that high percentage cannot be called a residue, but instead a sub-product, because it is applied extensively in agglomerated cork stopper (ex., for champagne bottles) and in many kinds of cork particleboards. Nevertheless the influence of extractives, particle size and density on hydration, cork granules were found to be compatible with cement [39]. Wood pulp fiber was responsible for the greatest replacement of asbestos in the Australian fiber cement industry [40].

HEAT TREATMENT OF FURNISH FOR WOOD COMPOSITES

Heat-treatment of solid wood to increase its dimensional stability and durability is well known and established in the industry. Now such technique is reaching wood composites manufacture. Heat treatment of strands of Scots pine before hot-pressing OSB reduced TS, resulting in increased dimensional stability; IB was not affected [41]. Steam pre-treatment, with temperatures below 200°C, improved the dimensional stability of panel products such as MDF and PB [42]. Heat treatment of MDF fiber resulted in an important reduction in TS and WA, although TS increased after repeated cycles of adsorption and desorption; no significant differences in MOR, MOE and IB of panels following heat treatment [43]. OSB panels from *Pinus taeda* wood manufactured with PF resin, without wax, and subjected to a thermal post-treatment, improved OSB dimensional stability by decreasing wood hygroscopicity and releasing hot-pressing stress [44]. Thermal treatment of spruce wood led to strong improvement of the adhesion between the modified wood surface and polyethylene; the contact angle of an applied water droplet increased significantly [45].

NONWOOD-FIBER RESOURCES

Nonwood fiber resources, for the scope of this article, are mainly annual, or short rotation, fast grown plants, as hemp, flax, kenaf, sisal, bamboo, etc. But agricultural residues can be also included, like straw, bagasse, cotton stalks, etc. Advantages that can be mentioned by making use of such fiber sources are the promotion of the replacement of food crops in developed countries, many of them, particularly in Europe, need to import wood and cellulosic fiber; recycling of agriculture residues and value addition to them; creation of conditions for new industries and jobs in developing countries based on such raw materials; less pressure on natural forests, by means of alternative raw materials.

Decomposed products from the pyrolysis of water bamboo husks fiber were found of interest for potential use as organic resources [46]. Treatment with hydrogen peroxide on bamboo surfaces improved the adhesion of a nitrocellulose lacquer coating [47]. Bending properties and IB increased with increasing density of random strandboards made using a moso bamboo strands and PF resin [48]. When chemistry reagents such as NaHCO_3 are applied as a bamboo softening treatment, for the manufacture of veneer, hemicelluloses are destroyed. A new softening technology at 120°C for 30 min in a closed container was developed, which imparts no effect on the composition of

bamboo [49]. Hemp fibers modified with NaOH solutions in order to partially extract noncellulosic substances, and to separate the fiber bundles, became finer, with lower content of lignin, increased flexibility, and in some cases tensile properties were improved [50]. Bamboo flooring has a lower specific gravity than red oak flooring but similar equilibrium MC; but bamboo flooring is more dimensionally stable than red oak flooring and laboratory-made laminated bamboo lumber [51]. Calcutta bamboo was found to have similar surface characteristics to commercial timber species used in North America intended for producing a structural composite [52].

Wheat straw particleboard bonded with a resin based on epoxidised oil and manufactured using a compression molding machine, resulted in a high compatibility between straw particles and oil-based resin [53]. Rice straw-wood particle composite boards manufactured as insulation boards were superior to insulation board in strength; sound absorption coefficients were higher than the other wood-based materials [54].

Free-radical content of wheat straw increased after it was treated with cellulases from *Trichoderma* sp. and *Aspergillus niger*. Wheat straw benzene-ethanol or ether extracts content decreased after wheat straw treatment with lipases from *Candida rugosa*, indicating that the surface wax of wheat straw was reduced, leading to an improvement of adhesion with a UF resin [55].

Wheat straw fibers prepared by chemical process exhibited better mechanical, physical and thermal properties, in comparison to those prepared by mechanical process. Polypropylene composites reinforced with that fiber showed enhanced properties compared to virgin polypropylene [56]. MDF made from wheat straw fiber and soy straw fiber had weaker mechanical and water resistance properties than those made from softwood fiber; in this case, wheat straw fiber and soybean straw fiber should be physically or chemically treated to increase their water resistance property for MDF production [57]. Aspen-kenaf boards made from a combination of commercial liquid and powder PF adhesives boards, with 25% kenaf and 75% aspen, exhibited MOR and MOE values comparable to commercial OSB [58].

Three-layer mixed comrind (CRD) and hardwood OSB manufactured using PF resin with comrind used in the core layer had linear expansion and TS improved by using comrind to replace part of the wood material in the core layer. At lower RH levels pure CRD OSB showed lower equilibrium MC compared to wood OSB [59]. Hardboard, MDF, and bagasse core panel (BCP), were made from bagasse/bamboo combinations with a formulation of 1% pMDI/4% UF as a binder. However, hardboard appeared to be a better panel type for

this kind of agro-based composites [60]. Three-layer particleboards were produced from mixtures of sunflower stalks and Calabrian pine particles, with UF adhesive, suitable for general purposes as well as furniture for interior environments [61]. Cotton stalks as a cheap raw material compared to woody sources in Turkey could be used to produce particleboards with technological acceptable properties [62].

CHEMICAL MODIFICATION

Rowell [63] defined chemical modification of wood as a process of bonding a reactive simple chemical to a reactive part of a cell wall polymer, with or without catalyst, to form a covalent bond between the two. This excludes chemical impregnations (dipping or soaking non-bonding chemicals in carrier solvents), polymer inclusions, coatings, and heat treatments. Such change in the chemical properties of wood usually imparts higher resistance to moisture and biodeterioration, and the chemicals applied are not necessarily toxic nor it is the final product.

Chemical modification of wood chips using propionic anhydride resulted in improved dimensional stability of PB: TS values of the UF bonded boards were lower than controls; IB decreased significantly, but was still substantially higher than the requirements of the reference standards [64].

Flakeboard panels manufactured with the inclusion of discrete layers of furnish that had been treated with acetic anhydride exhibited improved TS, keeping with an acceptable IB [65]. Acetoacetylation of wood meal (AAWM) proceeded almost quantitatively with diketene under optimal reaction conditions; AAWM with a high weight percentage gain inhibited the fungal growth of *Fomitopsis palustris* [66]. Chemical modification of wood chips and strands did not significantly affected the bonding efficiency of MDI, but the bonding efficiency of formaldehyde resins was strongly influenced [67].

Fixation of copper ions to acetoacetylated wood meal (AAWM) conducted in $\text{Cu}(\text{OAc})_2$ -MeOH solution resulted in a maximum amount of copper fixed to AAWM of $147.9 \text{ mg} \cdot \text{g}^{-1}$. A chelate was formed between copper and the acetoacetyl group of AAWM, and half of the copper in AAWM-Cu was retained, even at pH 4.0, after a leaching test [68].

BINDERLESS LIGNOCELLULOSIC COMPOSITES

For some years ago, not so few as that much, formaldehyde-based resins, and more generally synthetic resins, have been seen by the public opinion as harmful and derived from non-renewable chemical

sources. This gave an impetus to develop natural chemical-based adhesives. More recently, this general concern about wood adhesives (we do not discuss here how far there is fundament in it) can be probably overcome by the development of techniques to glue wood without any binders; i.e., by exploring the chemical characteristics of wood itself for that purpose.

Fiberboard could be successfully manufactured without any binder from spruce and beech fibers activated by treatment with Fenton's reagent ($\text{H}_2\text{O}_2/\text{FeSO}_4$). The improved adhesion was ascribed to interfiber bonds formed by reaction of radicals or other reactive groups generated in the fibers [69]. Binderless panels could be made from recycled corrugated containers. At both humidity levels of 10 and 20%, they were stronger and stiffer than those made from lodgepole pine [70].

When wood fibers were oxidatively treated with a chelator produced by *Gloeophyllum trabeum* (a brown-rot fungus), in the presence of hydrogen peroxide (H_2O_2) and ferric iron [Fe(III)], free radicals are produced in the lignin-rich fiber obtained from thermo-mechanical pulping, and lignin gains the functionality of a self-bonding adhesive. By this process, wet and modified dry-process softwood fiberboard was manufactured and exhibited increased IB, MOR and MOE after pre-treatment compared to that of fiberboard without pre-treatment [71].

Welding of wood is a wood joining procedure that offers several advantages over traditional mechanical fasteners or gluing: during welding, extensive solid-state transformation phases occur in the so-called melting zone and the heat-affected zone. The nature and the extension of such transformations are correlated to the energy input and thus to the heat generated during the process at the wood joint interface.

Joined wooden work pieces were welded by friction without any additional welding deposit. Microstructure of the welded joint revealed the manner in which the thermally decomposed wood forms the connection between the welded pieces [72]. The width of the welded zone varied as a function of the maximum temperature reached during welding, and the maximum temperature reached at the ends of the specimens was lower than that obtained in the central part of the specimens [73]. Non-destructive evaluation with infrared thermography allowed measurement of the maximal and average peak temperature, temperature profile and distribution, and rate of temperature increase [74]. This technique can also be used to detect welding defects and to provide information on material modification during welding. Panel products, such as PB, OSB and MDF, were joined to levels satisfying the requirements of the relevant standards for metallic connector assemblies by rotation welding of beech dowels through them [75].

Thermal alteration of wood and the formation of a viscous layer acting as adhesive has been studied: cellulose was found less altered than the other essential wood components; polyoses were found less stable under the conditions of friction welding; lignin suffered distinct changes, with an increase of free phenolic groups and a decrease of the typical bonds between the phenylpropane units; furan derivatives were detected within the volatiles of the smoke gas; reactions between furfural and other furan derivatives with lignin were seen as belonging to the main reactions in the friction zone leading to cross-linking [76].

Binderless particleboards were successfully developed from kenaf core using steam-injection press. Although kenaf binderless boards showed good IB strength, durability was relatively poor [77]. Low-density binderless particleboards made from kenaf core were successfully developed using steam injection pressing; thermal conductivity was similar to that of insulation material (i.e., rock wool), and they exhibited a high sound absorption coefficient [78]. Binderless boards prepared from finely ground powders of kenaf core revealed and increase of MOR, MOE, and IB with increasing board density. However, panels exhibited a low water-resistance [79].

Effects of chemical changes in binderless kenaf particleboards have been disclosed: mild steam-injection treatments of kenaf core caused significant degradation of hemicelluloses, lignin, and cellulose; conventional hot pressing caused a lower degree of degradation of the chemical components; hot-pressed kenaf core board without any binders showed poor bonding performance. Thus, it was concluded that partial degradation of the three major chemical components of the kenaf core by mild steam-injection treatment increased the bonding performance and dimensional stability of the binderless boards [80]. Furthermore, parts of lignin and hemicelluloses were decomposed during the hot-pressing process of kenaf core binderless boards. Progress of condensation reactions in lignin and the formation of chemical bonds by low molecular weight conjugated carbonyl compounds in methanol extractives were observed. Thermal softening of lignin also suggested to play an important role in board performance [81]. Kenaf core binderless fiberboards with densities of 0.3 and 0.5 g/cm³ were developed from kenaf core material using the conventional MDF dry-manufacturing process. Binderless fiberboards manufactured with high steam pressure and long cooking time during the refining process had high IB, low TS, but low bending strength values. Binderless fiberboards made from 30% MC fibers showed better mechanical and dimensional properties than those from air-dried fibers [82]. In a less conventional manner, kenaf composite panels were manufactured

with kenaf bast fiber-woven sheets as top and bottom surfaces, and kenaf core particles as core material; no binder was added to the core particles, while MDI resin was sprayed to the kenaf bast fiber-woven sheet. Compared with single-layer binderless particleboard, the bending properties and the dimensional stability were improved, especially at low densities [83].

Lignin analysis showed that steam-treated kenaf core composites had a lower proportion of syringyl- to guaiacyl-derived moieties and also cinnamic acids to guaiacyl-derived moieties than its native counterpart; some parts of the ester-linked cinnamic acids were also cleaved due to the degradation of hemicelluloses and lignin during steam treatment. In addition to the three main components, cinnamic acid was also suggested to participate in the self-bonding [84]. Binderless particleboards were also manufactured from sugarcane bagasse by steam-injection pressing and by hot pressing as a reference method. Inner layer (core/pith) and outer hard fibrous layer (face/rind) of bagasse were both used as raw materials. Bagasse pith particles provided better board properties than bagasse rind particles; steam-pressed boards showed relatively higher board properties than hot-pressed boards [85]. *Miscanthus sinensis* was steam exploded with a thermomechanical aqueous vapor process in a batch reactor to produce fiberboard with no synthetic binders. Increase in the dimensional stability was ascribed to the decrease in hemicelluloses [86].

PROTECTION AGAINST BIODETERIORATION

Wood, and more generally, lignocellulosic composites, have often the drawback of a relatively high susceptibility to biodeterioration. Water swelling, to start, opens avenues for fungi, and other microorganisms to enter into the material and start to degrade it. Furthermore, many wood composites are made of non-durable wood species. Developing wood composites that can withstand in a reasonable extent weathering conditions is a challenge for the industry to expand its market. After an evaluation of various water-repellent preservative systems as furnish treatments for a single-layer strandboard, one of the water-based treatments substantially improved TS and WA. Physical and fungal resistance properties were enhanced without decreasing their mechanical properties [87]. To work with no chemicals, better performance against decay was generally obtained for MDF, hardwood PY and PB, treated at high temperatures and pressures, whereas softwood PY and OSB were most protected at low temperatures under any of the pressure levels tested [88].

Supercritical carbon dioxide (SC-CO₂), when tested for its potential as a carrier solvent for preservative treatment, was not promising for refractory timber species such as *Larix leptolepis* Gordon. In contrast, a 3-iodo-2-propynyl butylcarbamate (IPBC)/SC-CO₂ treatment resulted in enhanced decay resistance without any detrimental physical or cosmetic damage in all structural-use of wood-based composites such as MDF, hardwood PY, softwood PY, PB, and OSB [89]. A combination of IPBC and silafluofen, using supercritical CO₂ as a carrier solvent, enhanced the resistance of the treated wood-based composites against fungal and termite attacks [90].

Weight losses by the brown rot fungus *Gloeophyllum trabeum* of all WPC's treated with a boron compound were generally lower than those of solid wood. Higher wood content was associated with greater weight losses; borates markedly reduced weight losses at all wood/plastic ratios [91]. After 10 years in field (stake and ground proximity) tests of WPC's at a tropical site, it was observed some decline in surface hardness over time: wood particles on the exposed surfaces were degraded in both soil contact and ground proximity samples, but the damage was relatively shallow. Wood content was significantly reduced, relative to plastic content, on weathered WPC surfaces [92].

Fiberboard panels made from pine and beech treated with N'-N-(1,8-Naphthalyl) hydroxylamine sodium salt (NHA-Na), borax, and boric acid, with UF resin as binder, showed increased resistance against the decay fungi *Fomitopsis palustris* and *Trametes versicolor* and the Formosan subterranean termite (FST) *Coptotermes formosanus* Shiraki [93]. Loading of zinc borate (ZB) and calcium borate (CB), in OSB from southern mixed hardwoods and southern yellow pine provided sufficient protection from severe structural damage Formosan subterranean termites (FST) [94].

Boards were treated with copper boron tebuconazole amine water-based preservative at different points during the manufacture process: best results were achieved when the preservative was applied by vacuum treatment of dry strands or by diffusion of green strands before board manufacture. Increasing preservative retention had minimal effects on board properties with these two methods, but significant deterioration was noted when the preservative was applied by spraying dry strands or by post-board-manufacture heat and cold quench [95]. Mill location significantly affected most property values of industrially produced North American composites, including LVL, OSB and MDF, treated by vapor boron technology, while treatment caused only significant reductions at the highest treatment level. The significance of mill location was attributed mainly to wood species differences, since industrial wood species varied among locations [96]. Acetylated

OSB stakes tested in ground contact in Western Greece for three years showed that acetylation imparts excellent protection against decay [97].

By exploring the inherent variability of wood, more specifically with regard to resistance against biodeterioration, MDF panels made from black spruce juvenile wood (1–20 annual rings) were found more resistant to mold infection than panels made of fibers taken from black spruce transition zone (21–40 annual rings) and mature wood zone (over 40 annual rings), and top, middle and bottom logs, or hybrid poplar clones, larch or a mixture of spruce, pine and fir (S-P-F) [98].

RECYCLING INVOLVING LIGNOCELLULOSIC COMPOSITES

As it happens with any kind of human activity, residues are generated. The point to highlight here is that wood and lignocellulosic composites offer a great potential to recycle them, including those of other industries than forest products, to make commercial products. The principle to achieve this principle is that wood composites, mostly, consist in particle of organic nature sticked together. One-layer boards were made of various ratios of waste paper (newspaper, office paper or magazine paper) flakes to wood particles mixtures with PMDI resin; newspaper was the most appropriate for board manufacture intended for interior uses, with a substitution of wood particles up to 50% [99]. Base sheets of aluminum foil-laminated and polyethylene (PE) plastic-laminated liquid packaging paperboard could be made into composite boards with adequate properties, either alone or mixed with wood particles [100].

Rice straw-waste tire particle composite boards were manufactured for use as insulation boards in construction, with commercial polyurethane adhesive for rubber used as the composite binder. WA and TS properties of the composite boards were better than those of wood particleboard. The flexibility and flexural properties were superior; also, good acoustical insulation, electrical insulation, anti-caustic and anti-rot properties were obtained [101]. Paper deinking sludge (PDS) was added to MDF furnish as an alternative to the current practice of disposal of PDS in landfill: flammability decreased by increasing clay; decay by the brown-rot fungus *G. trabeum* increased with clay content and decreased with increased calcium carbonate content [102]. PB panels and WPC's could be successfully manufactured with chips from ash trees infested by emerald ash borers (EABs) [103]. PB was also made from black locust, but which furnish contained soundwood, partially decayed wood, branchwood, and bark. Panels made from soundwood had the highest mechanical properties, while

branchwood panels resulted in the lowest. But mixtures from different parts of black locust trees could be used to manufacture particleboard panels [104]. Melamine impregnated paper offcuts and waste, in finely powdered form, could be applied as a binder for particle board and as a melamine substitute during the formulation and preparation of liquid MUF resins, based on the residual activity of the MF resin present [105].

LCA – LIFE-CYCLE ASSESSMENT

Knowledge of the environmental impact of the materials and processes typically used in a specific industrial sector is a key factor in enabling companies to improve their products from an environmental perspective and thus expedite their introduction into the growing market for “green” products. Environmental factors should be taken into account at the earliest possible stage of product development and design. The establishment of criteria for selecting materials with low environmental impact during the design of wood-based furniture design is an important aspect for furniture manufacture, for example. In the wood-based furniture manufacturing sector in Turkey, standard particleboard had an environmental impact lower than standard fiberboard; for surface and edge finishes, a low-density laminate is preferred to a high-density laminate [106]. Life-cycle inventories (LCI) revealed that, regarding lumber, 53 and 41% of the log volume leaves the mill as planed, dry dimension lumber, respectively in the western and southern U.S.; a much greater portion of the energy used for production in the South is produced on site from wood fuels [107]. In the production of glued-laminated timbers (glulam) in the U.S.-Pacific Northwest (PNW) and Southeast (SE), wood drying and adhesive manufacturing made major environmental contributions to the glulam process; fuel sources, either biomass or fossil-based, have significantly different emission impacts to the environment [108]. Concerning laminated veneer lumber (LVL), data were allocated based on their contribution to the mass sum of all product and co-products produced in manufacturing; carbon flow data are also given [109].

For wood composite I-joists, in addition to LCI data, transportation distances for delivery of materials were also provided. Data are useful for generating cradle-to-gate product LCI's when combined with the LCI's to produce logs for the mills and material transportation impacts, and for conducting LCA of floor and roof assemblies and residential structures using I-joists [110].

U.S.-Southeast OSB manufacturing: of roundwood raw material input, 71% ends in final OSB product. The remaining wood input ends

as wood residue for fuel, wood residues sold as co-products, and wood waste sent to the landfill; heat energy is the largest energy need, about 90% of which is generated from combustion of wood residues [111].

By gathering all results referred above (CORRIM 2004 Reports), it was concluded that, in the U.S. context, the production of wood products uses a third of their energy consumption from renewable resources and the remainder from fossil-based resources, when the system boundaries consider forest regeneration and harvesting, wood products and resin production, and transportation life-cycle stages. When the system boundaries are reduced to a gate-to-gate (manufacturing life-cycle stage) model, the biomass component of the manufacturing energy increases to nearly 50% for most products and as high as 78% for lumber production from the Southeast of USA. The manufacturing life-cycle stage consumed the most energy over all the products when resin is considered part of the production process; extraction of log resources and transportation of raw materials for production had the least environmental impact [112].

RESEARCH CLOSER TO PROCESS APPLICATION

Black spruce tops may be a good alternative raw material for MDF. Considerable positive effects of steam pressure of thermomechanical refining on MOE, TS, WA and linear expansion (LE) were obtained; retention time of preheating affected IB and MOR [113]. Strawboard bonded with UF resins: straw particles were found much more compressible and therefore required less platen pressure for pressing, compared to wood. Hammer milled straw mats have low permeability and subsequently showed high core gas pressure and high maximum core temperature during hot pressing, in addition to large differential densities between surface and core layers in the final pressed boards [114].

FINAL REMARKS

Rosenberb and coworkers highlighted that generalizations should not be made too broadly or be too precise in the forest products research area. Wood, or lignocellulosics in general, are very variable materials. Research activities are seldom guided by an overall theoretical framework that is applicable in all cases, which makes more difficult the technological progress in the forest products industry [115]. These statements can be easily confirmed after reading this paper. Almost every set of research results are applicable as such to the exact conditions of the study.

ANNEX

Glossary

- US-EPA – United States Environmental Protection Agency
- WPC – Wood-plastic composites
- CCA – Chromated copper arsenate
- LPF – Lignin-phenol-formaldehyde resin
- MW – Molecular weight
- HM_w – High molecular weight
- VOC's – Volatile organic compounds
- TVOC's – Total volatile organic compounds
- HD – Hardboard
- IB – Internal bond
- LPF – Lignin-phenol-formaldehyde resin
- LVL – Laminated veneer lumber
- MC – Moisture content
- MDF – Medium density fiberboard
- MDI – Methylene diphenyldiisocyanate resin
- MOE – Modulus of elasticity
- MOR – Modulus of rupture
- MUF – Melanime-urea-formaldehyde resin
- OSB – Oriented strand board
- PB – Particleboard
- PF – Phenol-formaldehyde resin
- pMDI – polymeric MDI resin
- PRF – Phenol-resorcinol-formaldehyde resin
- PY-Plywood
- RTF – Resorcinol-tannin-formaldehyde resin
- TS – Thickness swelling
- UF – Urea-formaldehyde resin
- WA – Water absorption

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