

Multi-component Polymer Systems Comprising Wood as Bio-based Component and Thermoplastic Polymer Matrices – An Overview

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The production of wood-based polymer composites has gained increasing interest in recent years, especially regarding sustainability issues, aiming at the recovery, reuse, and up-cycling of by-products from natural resources exploitation, as well as plastics. Due to their reduced cost, low density, and availability, wood components (fibers, flour) are attractive fillers for thermoplastic polymer matrices used in multi-component systems. Performance of wood-based thermoplastic materials mainly depends on the type and strength of interactions at the polymer-wood interface. Different low polarity polymers (high/low density polyethylene, polypropylene, polyvinylchloride) can be successfully used as matrices in such formulations. Various methods may be applied in order to obtain specified performance attributes of wood-based composites. Addition of appropriate compatibilizing agents, chemical and/or physical modification of the filler in order to improve its compatibility towards the matrix, or a judicious combination of these approaches may be employed. This paper briefly reviews some recent literature data, as well as research results by the authors, aiming at a comparative assessment of the materials properties (structure, thermal, mechanical and water sorption behavior) in correlation with the nature and type of components, processing, recycling options, and environmental impact.

Keywords: Composites; Thermoplastic polymer matrix; Wood filler; Properties; Investigation methods

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In memory of our dear friend and colleague, Dr. Ruxanda Bodîrlău, who passed away in June 2016.

WOOD-BASED FILLERS AND THERMOPLASTIC MATRICES FOR SUSTAINABLE MULTI-COMPONENT POLYMER SYSTEMS

Sustainable polymer materials have attracted much interest from both academics and industrial technologists, given the opportunities that they can offer in terms of properties and cost effectiveness. By combining high-cost and low-cost materials, with a special mention for the recycled materials, mainly polymer waste, it is possible to achieve new multicomponent materials with properties superior to their precursors, sometimes recyclable *per se*. Thus, using complex formulations that brought together various raw (virgin or secondary) materials, such as wood, plastics, gypsum, clays, cements, as well as compatibilizers, plasticizers, UV stabilizers, flame retardants, and other additives, the interfacial interactions have been enhanced so as to enable novel materials having special

properties fit to meet specific demands (Youngquist 1995; Rowell *et al.* 1996; Youngquist 1999).

From the perspective of the sustainable and environmentally friendly development, and taking into consideration society's increased awareness and drive toward the use of green products as substitutes for many other classic materials, it appeared that tailoring new composites using bio-based components offers a valuable alternative. As an example, natural bio-based fillers (*e.g.* fibers from lignocellulose resources) are considered interesting and valuable environmentally friendly materials for the production of emerging biodegradable (bio-) composites mainly when the recovery, reuse, and re-/up-cycling of various by-products from agriculture and food industry is envisaged. Apart from technological considerations, the embodied energy and carbon footprint of the materials, as well as their life cycle (cradle-to-gate and cradle-to-grave assessments), are important issues to be considered when designing new composite materials based on natural, renewable resources.

The main advantages conferred by the composites comprising bio-based materials (*e.g.* natural fibers and thermoplastic bio-based polymers/thermosetting bio-resins) are their lightweight character, improved and/or new functional properties (*i.e.*, damping/impact absorption), reduced occupational health hazards, and obviously, the environmental friendly features including low embedded energy, CO₂ retention, reduced use of fossil-based resources, and a favorable impact on the sustainability of agriculture, as different agriculture residues are re-/up-cycled (Weager 2010). Wood fibers sources continue to play a significant role in the human society, since they are involved in a large range of applications from packaging materials to buildings and transportation infrastructures (Ross 2010). Other natural fibers, such as hemp, flax, jute, and kenaf, are currently used in significant amounts mainly in the automotive industry for interior panels and components, where they play a role as reinforcement for synthetic polymers, such as polypropylene (PP); the fibers are useful in such applications because they are strong and stiff, and have lower density than conventional glass or carbon fibres (Bismarck *et al.* 2006; Ashori 2008).

Wood polymer composites (WPCs) are versatile hybrid materials. They usually comprise wood waste materials and by-products, in concordance with the sustainable principle of cascading exploitation and resource efficiency (Teuber *et al.* 2016), as well as plastic components, including thermosets (*e.g.* once cured, thermoset resins such as epoxies and phenolics cannot be repeatedly melted by heating) or thermoplastics (that can be repeatedly melted). The last-mentioned property is more advantageous for mixing wood materials with different plastics to form composite products. The most used thermoplastics for WPCs production are virgin polymers such as polypropylene (PP), polyethylene (PE) and polyvinyl chloride (PVC), and these combinations mainly have been utilized in building, construction, furniture, and automotive products applications (Ashori 2008; La Mantia and Morreale 2011; Clemons *et al.* 2013; Gurunathan *et al.* 2015). Nevertheless, recycled thermoplastics may confer great opportunities for WPCs production (Kazemi-Najafi *et al.* 2006; Kazemi-Najafi 2013; Sommerhuber *et al.* 2015), even if these polymer components may exhibit different performance properties as a function of their degradation level. Their chemical composition can be affected depending to a large extent on their source and exposure under variable storage and reprocessing conditions. They may also impose additional costs for separation when recycled plastics are used as blends.

Wood-thermoplastic composites (WPCs), which also have been called wood-plastic composites, have experienced significant development in the recent decades

(Clemons *et al.* 2013). They are usually manufactured through mixing the natural bio-based component with the thermoplastic polymer or its addition to the polymer matrix, followed by processing under high pressure and temperature conditions. Thus, composite materials may result through extrusion, injection, or thermoforming processing methods, and they can be used in structural and non-structural applications. Wood-based materials can be used in composite formulations both as filler or reinforcement depending on their amount, shape and size: long or short fibers or fiber bundles, particles (flour, sawdust). The process may also use additives including colorants, coupling agents (for compatibility purposes), stabilizers (*e.g.* against photo-degradation processes), foaming agents, other reinforcements, and lubricants for sustainable final products in concordance with their envisaged area of application. Thus, the most important applications are considered decking and the automotive industry, followed by exterior siding and fencing (Carus *et al.* 2014), although the highway construction sector is an emerging market due to the high potential of WPCs to replace classic materials (*e.g.* virgin plastic and steel). The same is the case for the preservative based products (treated wood) (Thompson *et al.* 2010). The development of a design value for WPCs cladding is also feasible, and it can be derived from normative natural and accelerated weathering trials (Friedrich and Luible 2016). The environmental conditions experienced by WPCs used for cladding exterior sides of buildings should be considered through monitoring degradation of WPCs properties by means of specific durability tests such as fungal decay, weathering exposure, and freeze-thaw cycling (EN15534-1, 2014).

In the following, an overview of some recent advances is presented in a systematic manner, considering the nature and type of matrix and filler, the polymer-wood interfacial interactions in correlation with modern compatibilization approaches, the processing methods, and recycling protocols able to meet the environmental challenges, as fundamental discriminating criteria. Using wood and plastic waste may result in lower environmental impact, as the amount of wood in WPCs can be increased and the plastics recycling industry already provides high quality materials (Miller *et al.* 2015; Wager and Hischier 2015; Sommerhuber *et al.* 2015, 2016, 2017; Wenker *et al.* 2016). Some considerations on the life cycle assessment (LCA) studies of WPCs and future perspectives are included here. Such approaches are strongly required for investigating the presence of the potentially hazardous organic compounds in relation with the effective resources use. The cradle-to-grave LCA approach can be important, as it aims not only at the service life, but also at the final disposal of WPCs. In terms of sustainable production, it is important to use recycled materials (the cascading principle) not only for wood, but also for the polymer matrices (Sokka *et al.* 2015).

Thermoplastic Polymer Matrices

Generally, a thermoplastic polymer softens upon heating and hardens during cooling. Thermoplastics commonly selected for use in composite applications along with lignocellulosics materials, including wood and agricultural fibers, must melt or soften at or below the minimum degradation temperature of lignocellulosic component, which is usually in the range 200 to 220°C (Youngquist 1999; Klyosov 2007; Borrega and Kärenlampi 2008).

The most used thermoplastics as polymer matrices for the manufacture of wood plastic composites are: polyethylene PE (Liao *et al.* 1997; Bengtsson *et al.* 2005; Mamunya *et al.* 2006; Adhikary *et al.* 2008) in its high-density polyethylene (HDPE), low-density polyethylene (LDPE), and linear low-density polyethylene (LLDPE) forms, as well as

polypropylene PP (Raj *et al.* 1989a,b; Takase and Shiraishi 1989; Myers *et al.* 1991; Oksman and Lindberg 1995; Joly *et al.* 1996; Kazayawoko *et al.* 1999; Stark and Rowlands 2003; Chowdhury and Wolcott 2007; Wang *et al.* 2011; dos Santos *et al.* 2015), poly(vinyl chloride) (PVC) (Matuana *et al.* 1998a,b, 1997; Mengeloglu *et al.* 2000; Bodîrlău *et al.* 2009), and polystyrene (Maldas and Kokta 1991a; Rizvi *et al.* 2000). Usually, the thermoplastics such as PE and PP, with low-melting temperature values, are preferable in combination with wood materials as fillers or reinforcements (Bledzki *et al.* 1998), in order to avoid significant degradation of wood, which presents low thermal stability. For the European WPC-production market, PVC is considered of major importance (Carus *et al.* 2015), despite the fact that other thermoplastics already have emerged and are extensively used.

Wood Materials as Fillers or Reinforcements in WPCs

When they are to be used as filler or reinforcement in WPCs, wood-derived materials can be prepared in particulate form (*e.g.* wood flour) or in the form of very short fibers and bundles of fibers. Such fillers are sometimes preferred in order to alter the mechanical performance of WPCs (*e.g.* stiffness, heat deflection temperature) without an excessive increasing of density (Clemons *et al.* 2013), and to avoid processing difficulties. In general, wood flour is obtained as a by-product of wood processing (lumber mills and furniture factories, millwork, window and door profiles manufacturing, including recycling), being made from sawdust, planar shavings, sanding dust, and scraps. Longer fibers from hardwood and softwood species, even medium density fiberboard (MDF) waste (Chavooshi *et al.* 2014) are also used. Wood flour is very attractive for WPCs production and their end-applications, as compared to wood fibers, given its availability, low cost, relatively high bulk density, and free-flowing character. It adds hardness to the composites, but not considerably affecting their strength, and it improves the interfacial interaction between components, facilitating their easy processing. For WPCs production in the United States, the common wood species used include softwood (pine) and hardwood (maple and oak) species (Caulfield *et al.* 2010). Increasing the content of bio-based component in biodegradable composites yields immediate effects related to the modification of mechanical properties. Improvement of mechanical performance is notable mainly when fibers, such as wood, pulp, or paper fibers, are added as reinforcements in WPCs compared to wood flour (Clemons *et al.* 2013). Recently, even the potential use of decayed wood in production of WPCs consisting of wood flour and PP, using MAPP as coupling agent, was considered (Ayrilmis *et al.* 2015). For high level content of decayed wood flour (40% and 50%, respectively), thermal stability of composites was improved, while the tensile and flexural properties were diminished, comparatively with WPCs consisting of sound wood (wood free from decay) flour.

METHODS APPLIED FOR SOLVING COMPATIBILITY ISSUES IN WPCs

Numerous studies have focused upon finding and recommending effective strategies in order to improve the fiber–matrix interfacial interaction in both natural fiber composites (NFCs) and wood-polymer composites. A recent literature survey presented a comprehensive overview of the current state of the art of methods for solving compatibility issues related to the polyolefin-based NFCs and WPCs (Sobczak *et al.* 2013). These methods are usually fiber-based strategies (*e.g.* fiber surface modification), and matrix-

based strategies, as well, both of them including the use of coupling agents. Combinations of these two approaches have been also proposed (Wu *et al.* 2000; Bledzki *et al.* 2008; Bettini *et al.* 2010).

The interfacial adhesion between a wood component and a thermoplastic polymer matrix is essential for WPCs in order to achieve better performance properties (Kazayawoko *et al.* 1999). While there can be some advantages (such as low cost, low density, renewable nature, high specific strength, and high modulus), bio-based materials (*e.g.* including those derived from wood sources) are hydrophilic because of their main polysaccharide constituents, cellulose and hemicelluloses, which are dominant in their chemical structure. When used as filler for wood-polymer composites (WPCs), their hydrophilic nature causes poor interfacial adhesion between wood surface and the hydrophobic polymer matrix, thus decreasing the WPCs toughness (Klason *et al.* 1984; Dalvag *et al.* 1985). Moisture sorption inside the structure of wood that remained uncovered by the polymer can also promote swelling and water uptake, the biological attack and, subsequently, degradation.

Thermal treatment of wood aims at its modification based on the chemical degradation of wood polymers by heat transfer; it was shown that such treatment can improve wood's resistance to decay, while also providing dimensional stability (Kamdem *et al.* 2002; Esteves and Pereira 2009; Allegretti *et al.* 2012; Pelaez-Samaniego *et al.* 2013a; Candelier *et al.* 2016). Such improvement in properties is of real significance for production of WPCs comprising thermally treated wood, and these issues were extensively investigated under both wet and dry conditions (Liu *et al.* 1994; Follich *et al.* 2010; Hosseinaei *et al.* 2012a; Pelaez-Samaniego *et al.* 2013b). Thermal pre-treatment of wood in moist environments (hydrothermal methods) in the presence of water or steam prior to blending with the polymer is one of the most interesting approaches and can be considered as a feasible strategy to reduce the tendency of WPCs to take up water (Pelaez-Samaniego *et al.* 2013a). This is accomplished through the removal of hemicelluloses from wood by processes such as dissolution and hydrolysis to free sugars (Amidon *et al.* 2008; Liu 2010). This fractionation of wood chemical components with significant solubility and susceptible to be easily hydrolyzed was able to reduce the equilibrium moisture content of wood (Hill *et al.* 2012). At the same time, alongside considerable degradation of hemicelluloses, a condensation process of lignin component may occur, which might be attributed to the cleavage of ether bonds and subsequent structural rearrangements. All of these processes occurring during thermal treatment of wood are related to the lower pH value reached through releasing of carboxylic acids, such as acetic - from the hemicelluloses component, namely glucuronoxylan from hardwood species, and formic - from degradation of hexoses (Lai 2001). The resulting pre-treated wood under heat conditions showed an improved durability and dimensional stability (Sundqvist 2004; Hosseinaei *et al.* 2012b).

Use of coupling agents in WPCs represents a very effective and important approach to achieve compatibility and adhesion between polar wood surfaces (as fibers or flour) and non-polar polymer matrices (Lu *et al.* 2000). Organic coupling agents are more efficient than inorganic ones, a fact that has been made evident by the stronger adhesion occurring at the interface. Despite the large number of coupling agents that are used or have been tested at the laboratory and industrial levels, the most known and applied to a large extent are organic {*e.g.* isocyanates, anhydrides, anhydride-modified copolymers such as poly[methylene(polyphenyl isocyanate)] (PMPPIC) and maleated polypropylene (MAPP)} and hybrid (organic-inorganic) agents (*e.g.* silanes). In the case of MAPP, the

anhydride component reacts with a hydroxyl group of the wood cell wall, forming an ester bond, as presented in Fig. 1.

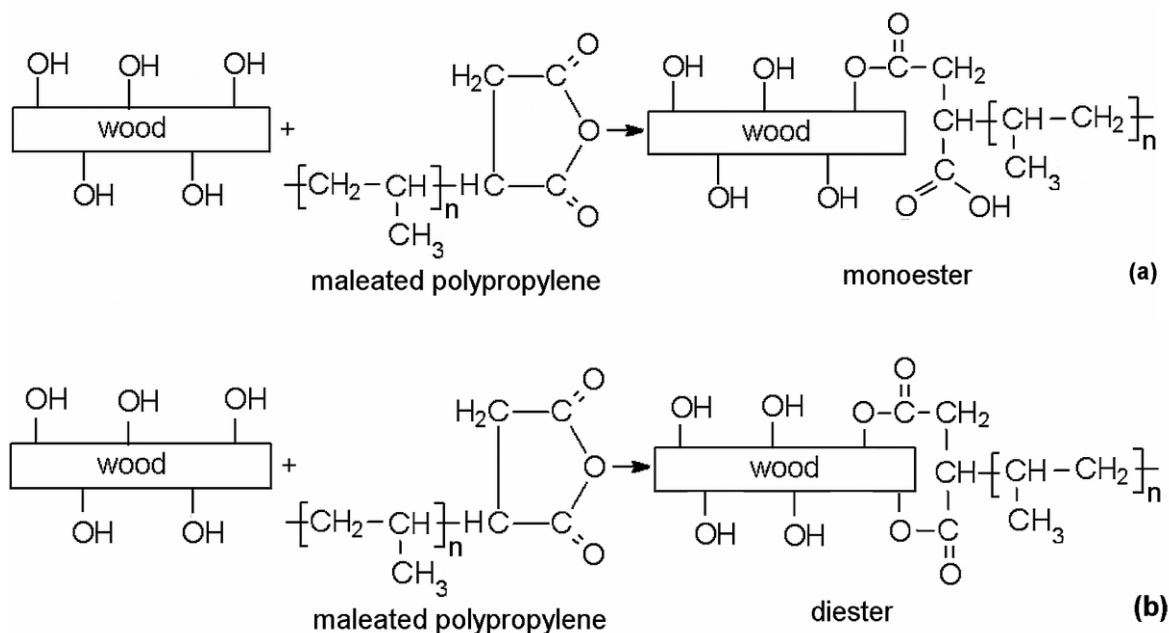


Fig. 1. A schematic representation of reaction between wood and maleated polypropylene with formation of corresponding wood monoester (a) and wood diester (b)

At the same time, one can incorporate polypropylene segments of the MAPP into the bulk polypropylene matrix (Lu *et al.* 2000; Caulfield *et al.* 2010). This effective promoter for a better adhesion was earlier demonstrated to act as a real coupling agent in composites based on PP and HDPE filled with wood and cellulose flour (Dalvag *et al.* 1985). A reinforcement effect was evidenced by the increased strength, even in a molten state, and ductility. MAPP as a compatibility agent in composite formulations is effective through improving mechanical properties (mainly the tensile strength) as evidenced by Bledzki and co-workers (Bledzki *et al.* 2002), the composites made of hardwood fibers showing better performance than those with softwood fibers in formulation. This positive effect is related to the reduction of the total wood fiber surface free energy, a better impregnation of fibers by the polymer matrix, an improved dispersion and orientation of fibers, and an enhanced interfacial adhesion through mechanical interlocking process (Kazayawoko *et al.* 1999).

Coupling efficiency is strongly related to the molecular weight of MAPP, which influences the interfacial adhesion in resulting composites more than the content of maleic anhydride in MAPP (Lu *et al.* 2005). However, MAPP tends to be less effective when polymer matrices other than PP are used in WPCs (*e.g.*, in PE/wood flour or PVC/wood flour composites). This has been attributed to incompatibility between the PP backbone of MAPP and the other polymer matrices in composites (Matuana *et al.* 1998a,b). Other effective compatibilizers for improvement of WPCs properties are represented by styrene ethylene/butylene styrene (SEBS) copolymers (Oksman and Lindberg 1995; Lai *et al.* 2003). Such a compatibilizing effect appears to overcome the problem of the different

solubility parameters of cellulose component and styrene, which may cause repulsion effects or by chemical bonding (grafting) as a consequence of processing at high temperature values. The mechanical properties of WPCs consisting of linear low density polyethylene (LLDPE) and wood flour (WF) were improved by using SEBS triblock copolymer modified with maleic anhydride and a ionomer polymer as compatibilizers (Oksman 1996).

Coupling agents that have high molecular weight, moderate acid number, and are able to provide compatibilization at a low concentration level, such as maleated polyethylene (MAPE), are often preferred to improve interfacial adhesion in WPCs consisting of wood fibers and HDPE and manufactured in the presence of dicumyl peroxide as initiator (Lu *et al.* 2005) by formation of a switch structure at the interface between wood component and thermoplastic polymer matrix.

Earlier studies (Raj *et al.* 1989; Maldas and Kokta 1991b; Matuana *et al.* 1998a) have provided evidence that coupling agents such as aminosilanes and anhydride-modified copolymers such as poly[methylene(polyphenyl isocyanate)] (PMPPIC) can effectively enhance the bonding between wood fibers and different thermoplastic polymer matrices and improve mechanical properties of the resulting composites, such as tensile strength and modulus.

Due to the polarity differences between wood components (as fillers or reinforcements) and thermoplastic matrices, their interfacial interactions are often weak. This feature often leads to undesirable mechanical failures. Many studies have been carried out in order to improve the adhesion between phases. There are two main approaches: (a) modification of the thermoplastic polymer matrix (*e.g.* by graft copolymerization method) and (b) modification of the wood component before mixing. Bio-based fibers have a good potential for modification treatments (Bledzki *et al.* 1998; Kalia *et al.* 2009; Adekunle 2015) due to the presence of hydroxyl groups in the structure of their constituent polymers, namely lignin and cellulose. Reaction of hydroxyl groups can change the surface energy and polarity of wood surfaces, improving their interfacial interactions with non-polar polymer matrix. Some of the treatments concerning wood component modification include both physical, such as electric discharges – corona, cold plasma treatments (Sakata *et al.* 1993; Belgacem *et al.* 1994), thermal treatment (Pelaez-Samaniego *et al.* 2013a), mercerization (Borysiak 2012), and chemical methods, *e.g.* pre-treatment of wood fibers surface, grafting, use of coupling agents and functionalized polyolefin-coupling agents (Lu *et al.* 2000).

Chemical methods usually applied to wood substrates include isocyanate treatment (Raj *et al.* 1988, 1990; Kokta *et al.* 1990a; Maldas and Kokta 1991a), acrylation (Li and Matuana 2003), benzoylation (Dominkovics *et al.* 2007; Xie *et al.* 2009; Farsi 2010), organic anhydride treatment – acetylation (Dawson *et al.* 1999; Haque and Hill 2000; Hill 2006; Rowell 2012), succinylation (Doczekalska *et al.* 2007; Bodirlău and Teacă 2009), and silane treatment (Kokta *et al.* 1990b; Ichazo *et al.* 2001). Reaction of wood subjected to organic anhydride treatment (*e.g.* acetic anhydride, succinic anhydride) can be exemplified as presented in Fig. 2.

These methods may contribute to the improvement of the fiber-matrix adhesion, acting upon features such as strength, size, and shape. For example, the mercerization process can be quite effective. The alkali treatment tends to enhance the reactivity of wood component surface for an increased adhesion toward the polymer matrix by a fibrillation process and increasing surface roughness, thus allowing the better response of wood to chemical modification.

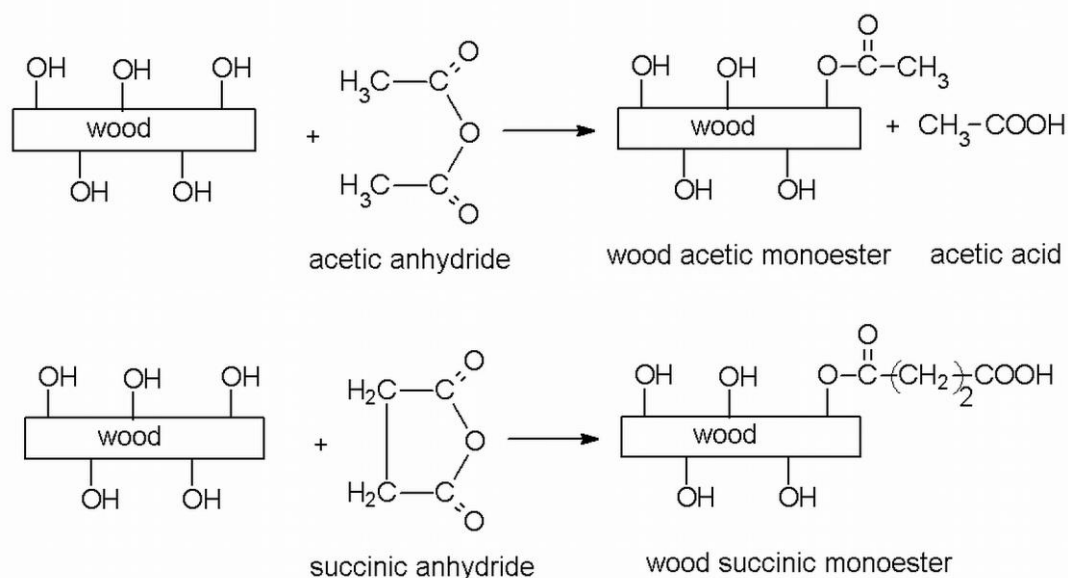


Fig. 2. A schematic representation of reaction between wood and organic anhydrides – acetic (a) and succinic (b) – with formation of corresponding wood monoester

Combined methods can also be successfully applied for solving compatibility issues in WPCs, *e.g.* heat treatment and acetylation (Liu *et al.* 1994), use of silane coupling agent and addition of compatibilizer such as maleic anhydride modified polyolefins (Wang *et al.* 2011), and maleic anhydride modified block copolymers as styrene-butadiene-styrene SBS (Wu *et al.* 2000).

A novel reactive processing method of simultaneously co-refining wood and polyolefins (POs)—polyethylene and polypropylene—by steam-explosion was proposed and investigated in the manufacture of wood-thermoplastic composites (Rennekar 2004). This method was effective through modification of wood fibers by addition of “non-reactive” polyolefins without the need for further coupling agents. The resulting wood fibers exhibit modified sorption properties (Rennekar *et al.* 2006) related to the migration of oxygen-containing species, resulting from the polyolefin degradation during processing into wood cell walls.

ASSESSMENT OF PROPERTIES

The properties of WPCs are determined by the properties of components, formulation, chemical structure and supramolecular architecture of the final material, and the nature and strength of the interfacial interactions. The content of wood materials used in WPCs influences to a large extent their resulting properties, ranging from mechanical characteristics and dielectric properties, to thermal stability (Djidjelli *et al.* 2002). The later mentioned is essential for an easy processing by using the conventional techniques (extrusion, calendering, often as a method of choice for processing PVC, and injection), which require temperature values below 210 °C. The wood component has no significant influence on the glass transition temperature as content. Instead, it may increase the

temperature of decomposition with positive effect on thermoplastic matrix behavior, conferring a good thermal stability for processing purposes.

Usually, wood is added to modify mechanical performance of the resulting composites, as wood fibers are stronger than the wood from which they are obtained (Rowell 1992). Nevertheless, different marketing strategies may consider the natural appearance of wood very important, while others focus on the increasing wood content in composite formulations (Clemons *et al.* 2013). The improved mechanical performance can be attributed to the higher strength and aspect ratio of wood fibers when they are used, compared to wood flour, but also to the effective stress transfer from the thermoplastic polymer to the wood fiber when a well-bonded (the polymer is suitably wetting the filler) composite structure is formed. Wood fibers can present favorable aesthetics and low abrasiveness during processing; they can have a large impact on the reinforcing ability depending on the preparation methods (Stark and Rowlands 2003; Klyosov 2007; Caulfield *et al.* 2010; Clemons *et al.* 2013), alongside reduced density. Long and semi-flexible wood fibers (high quality pulp fibers) give a lighter color and are more effective in composites than thermo-mechanical pulp (TMP) fibers, which are stiff and poorly bonded due to their surface rich in lignin and extractives that contribute to the weak adhesion. Much of these wood components must be removed from the TMP fibers surface with increasing energy costs. The main disadvantages of using long fibers in composites production are related to their low bulk density and entanglement, which may cause further processing difficulties, such as feeding and metering into conventional polymer processing equipment like extruders, thus limiting their use. For WPCs, wood acts as extender to reduce polymer use, and it is generally in the form of short fibers, particles, or flour. These compressible small sizes can integrate well into the processing technologies used by the plastics industry.

A recent thesis emphasizes that the inclusion of thermally extracted wood distillates, which were obtained from wood processing, into WPCs can improve the mechanical performance and water resistance (Väisänen 2016). Moreover, they can contribute to a sustainable development by using raw materials and secondary products from the wood-processing industries (Väisänen *et al.* 2016a,b,c).

WPCs are susceptible to water absorption phenomena, which further induce thickness swelling and the occurrence of micro-cracks in the composite materials. All these conditions contribute to mold growth, volume changes, and subsequently to fungal attack. These factors can lead to weight loss and decreases in mechanical performance. Different strategies have been considered to eliminate or, at least, reduce these disadvantages. Current methods to improve the moisture resistance of WPCs include changing the morphology of the composites surface by changing the processing method, and/or processing parameters, and in this manner, changing the WPCs degradation pathways (Stark and Matuana 2006). Other methods involve treating the wood filler with acetic anhydride (Rowell 1992; Rowell 2012), adding a maleated polyolefin into the composite formulation (Clemons *et al.* 2013), and coating the WPCs surface using co-extrusion method (Stark and Matuana 2007).

The effective methods to modify wood fibers include heat treatment (Ayrilmis *et al.* 2011), extraction of hemicelluloses (Hosseinaei *et al.* 2012a), and treatment with coupling agents (Müller *et al.* 2012). Thus, modified wood fibers granted significantly increased water resistance to the resulting WPCs with further positive impact on the mechanical properties.

The mechanical durability of WPCs can be influenced to a significant extent by incorporation of different additives, such as MAPP or MAPE (Hristov *et al.* 2004; Adhikari

et al. 2012; Pérez *et al.* 2012), waste charcoal or biochar, (Li *et al.* 2014; Das *et al.* 2015), or inorganic fillers (Zhong *et al.* 2007; Gwon *et al.* 2012; Beigloo *et al.* 2017) into the composites. Use of recycled polymer matrices in composite formulations in place of pristine materials can improve their mechanical performance (Adhikary *et al.* 2008).

The effects of exposure to strong light also have been a topic of much investigation. In general, WPCs tend to become lighter in color over time during exposure to light. An efficient method to slow down this effect is the addition of pigments (Butylina *et al.* 2012a). There have been less noticeable color changes observed during weathering mainly when darker pigments (brown or grey) were added. The addition of mineral fillers can be especially beneficial for wood-polymer composites that are intended to be used in outdoor applications, mainly through reduction of water absorption. Given their favorable mechanical properties and low rates of weathering, wood-polypropylene composites that contain talc as mineral filler seem to be suitable for outdoor uses (Butylina *et al.* 2012b).

The weathering behavior of WPCs is very important for their mechanical performance mainly when outdoor applications are considered. The biopolymers forming wood chemical structure (namely cellulose, lignin, hemicelluloses) exhibit different abilities for interaction with UV light to generate radicals (Teacă *et al.* 2013). Lignin is very sensitive to UV irradiation, and this can lead to the generation of aromatic type free radicals (phenoxyl radicals). Such radical species can further react with oxygen from the atmosphere to form carbonyl and carboxyl groups (Pandey 2005), and these can lead to wood discoloration due to the presence of unsaturated carbonyl compounds (quinones). The addition of lignin when formulating WPCs can accelerate their photochemical degradation under UV weathering conditions (Chaochanchaikul *et al.* 2012). Besides this aspect, there are many other factors affecting the photochemical degradation of WPCs, such as wood flour content, use and type of coupling agents, manufacturing methods, and weathering conditions. Thus, researchers have studied the influence of accelerated artificial weathering conditions on the strength characteristics and color changes of PVC/wood-flour composites over a range of wood flour contents (Matuana and Kamdem 2002). Composites presented severe color changes, which were expressed as darkening after 2600 h of weathering exposure, but they retained their initial tensile properties. At relatively low wood content in formulation (15%), PVC/wood composites were darkened to a large extent comparatively with those comprising higher amounts of wood in the composition (30%, and 45%, respectively).

When HDPE was used as thermoplastic polymer matrix in WPCs, an opposite trend was noticed under weathering conditions. Composites with higher amount of wood flour tended to be lightened to a greater extent than those with lower wood content (Stark and Mueller 2008). Investigation upon the effects of wood flour content and the use of compatibility agent on the durability of WPCs, comprising HDPE and PP as thermoplastic polymer matrices (Ndiaye *et al.* 2008), evidenced that increasing the wood content will increase the rate of oxidation process, while the addition of compatibility agent decreased the intensity of oxidation process by dispersing of wood flour more uniformly within the thermoplastic polymer matrix.

Researchers have investigated the effects of weathering process under natural and accelerated (artificial) conditions on the visual appearance, chemical changes, and moisture sorption properties of WPCs comprising HDPE and PP as thermoplastic polymer matrices (Matuana *et al.* 2001; Matuana and Kamdem 2002; Stark *et al.* 2004; Stark and Matuana 2004a,b; Muasher and Sain 2006; Stark 2006; Stark and Matuana 2006; Stark and Matuana 2007; Fabiyi *et al.* 2008; Stark and Mueller 2008; Segerholm *et al.* 2012). An increase of

significant parameters such as lightness, total color change, and wood loss was noticed in the course of increasing exposure time. WPCs with PP as the thermoplastic polymer matrix exhibited an increased rate of photochemical degradation process with respect to lightness and the loss of wood. The increased lightness is related to the delignification and oxidation processes that occur in composites under weathering conditions.

The discoloration of WPCs with HDPE under exposure to natural weathering conditions was investigated (Muasher and Sain 2006), and the occurrence of two competing redox reactions after UV exposure was evidenced, namely: the formation of *p*-quinone-type chromophores through the lignin oxidation reaction that lead to a yellowing process, and the reduction of these *p*-quinone oxidation products to hydroquinones, which is manifested as photochemical bleaching. The oxidation reaction (yellowing phenomenon) was dominant during the first 250 h of exposure, preceding the reduction reaction (photochemical bleaching), which was more evident along with the increasing exposure time.

Light stabilizers (or photostabilizers) are usually used to protect polymers in WPCs against UV degradation in order to minimize fading. Such compounds are classified in concordance with their mechanism of action as ultraviolet absorbers (UVAs)—acting as a shield against UV light; and hindered amine light stabilizers (HALS)—acting as quenchers and free-radical scavengers for radical intermediates, resulting in the photo-oxidation process (Stark and Matuana 2004a).

Pigments or mineral fillers (*e.g.* talc, calcium carbonate) are often added as photo-blocking agents to combat the polymers photo-degradation (Butylina *et al.* 2012a,b). Such addition can result in improved color stability of the WPCs in outdoor applications. Besides this effect, mineral fillers can be effectively used to improve mechanical and flame retardancy properties. Additives such as fire retardants and light stabilizers can also be effectively added to produce WPCs with better fire retardancy and durability performance (Garcia *et al.* 2009). In this case, a combination of HALS, UV filter, and antioxidant properties resulted in no cracking or fracture occurrence in the weathered WPCs, and there was only some fading.

Coating the WPCs samples with a polymeric layer through co-extrusion, compression, or thermoforming can also be an effective method to improve WPCs durability in outdoor applications, even with occurrence of the inherent difficulty to promote adhesion in polyolefin substrates (Garcia *et al.* 2009; Stark and Matuana 2009; Stark and Matuana 2007).

The interfacial interactions and the functional performance of WPCs can be effectively controlled by reaching a certain degree of compatibility between components (employing an appropriate technique for the modification of wood fibers surface, use of a specific compatibilizer and/or other additives), while the selection of materials and processing parameters depends on the specifications of different end-users.

Processing parameters (*e.g.* residence time, temperature, pressure, shear rate, shear stress, cooling rate) vary with processing method and determine different final WPCs properties (Michaeli and Menges 1989). When comparing the processing methods, injection molding and extrusion, for the same applied processing data, the pressure level and shearing are significantly higher for the first mentioned method than for the second one. Besides this, the structural parameters such as orientation or boundary layers are more distinct for the injection molding processing method. The morphology and internal stress type are related to the processing parameters such as cooling rate, pressure, and deformation occurrence. When better mechanical performance and dimensional stability

are envisaged for resulting WPCs, the injection molding processing method is considered (Migneault *et al.* 2009).

When the same wood fibers were considered for WPCs formulations comprising different polymer matrices, it became evident that the resulting alterations in the type and strength of interfacial interactions, as well as the performance of the material, can be attributed, to a large extent, to inherent materials limitations and processing challenges that finally yielded three undesirable conditions. The first is fiber damage during processing and, consequently, a poor wettability of fibers. The second is weak bonding between matrix and filler, entailing degradation processes at the interface. The third is high moisture absorption.

These challenges and their potential solutions have been seriously addressed, and the available literature data (Olakanmi and Strydom 2016) confirmed that the results can effectively contribute to the improvement of the WPCs performance and, subsequently, promote the development of sustainable WPCs on competitive criteria.

Surface Chemical Properties

Interfacial characterization techniques can provide information pertaining to the chemical composition and the morphology of the surfaces, as well as the interactions between the wood fiber and polymer matrix (Hristov *et al.* 2004; Stark *et al.* 2004; Stark and Matuana 2004a,b, 2006; Dominkovics *et al.* 2007; Stark and Matuana 2007; Fabiyi *et al.* 2008; Adhikari *et al.* 2012; Pérez *et al.* 2012; Segerholm *et al.* 2012).

WPCs surface properties can be evaluated using chemical investigation methods such as contact angle analysis, inverse gas chromatography (IGC), infrared (IR) spectroscopy, scanning electron microscopy (SEM), and X-ray photoelectron spectroscopy (XPS). Thus, the contact angle analysis and IGC provide information on the surface thermodynamics and adhesion. Compatibility of chemically modified wood fibers in WPCs can be evaluated by means of contact angle measurement methods (Felix and Gatenholm 1991; Chen *et al.* 1995). This feature is improved in composites with thermoplastics when modified wood surfaces present smaller surface energy (*i.e.*, larger contact angles) with a probe liquid (*e.g.* water). Characteristics of fiber wettability at the wood fiber-polymer interface also have been considered (Lu *et al.* 2002; Lu 2003). IGC was used to estimate the interfacial adhesion between wood fiber and PP matrix in composites (Kazayawoko *et al.* 1999), evidencing that the thermodynamic work of adhesion due to the physical forces may not be relevant in explaining the improvement in the tensile strength of the composites. The influence of secondary interactions (*e.g.* hydrogen bonding) on the development of interfacial structure in composites made of wood and amorphous thermoplastic polymers can be also investigated by using inverse gas chromatography (Rials and Simonsen 2000).

Infrared spectroscopy provides information on the functional group composition at the composite surface, while XPS reveals the elemental composition at the material surface. Both investigation methods are useful for quantifying the surface chemical changes in WPCs as a measure of the accelerated UV degradation. XPS can also be used to obtain an in-depth profile (up to 10 nm) of the functional groups at the material surface.

Another highly effective method employed to investigate the specific interactions between polymers is the Fourier Transform Infrared (FT-IR) spectroscopy. FT-IR can be used for qualitative and quantitative analysis of the mechanisms of interactions among the polymeric materials through hydrogen bonding because the absorbance peaks are characteristic to chemical bonds rather than atoms (Lu *et al.* 2000; Marcovich *et al.* 2001;

Matuana *et al.* 2001; Kim *et al.* 2004; Cofta *et al.* 2006; Bodîrlău *et al.* 2009). Attenuated total reflectance (ATR)-FTIR spectroscopy is often used for monitoring the occurrence of WPCs surface oxidation through the carbonyl group change and the structural changes in the polymer matrix by following vinyl group formation and the crystallinity changes of polymer matrix after weathering (Stark and Matuana 2004b).

Considering the non-destructive surface analytical techniques, XPS is very useful when it comes to investigating the oxidation state of elements or their chemical bonding. XPS has been used in several applications in wood science to investigate the surface of different wood species (Liu and Rials 1998; Sinn *et al.* 2001; Nzokou and Kamdem 2005; Nguila Inari *et al.* 2006; Bouafif *et al.* 2008), chemically modified pulp, paper, and wood (Chen and Tanaka 1998; Matuana *et al.* 2001; Li and Matuana 2003; Hultén *et al.* 2006; Bodîrlău *et al.* 2013), residual lignin presence on the surface of cellulose fibers (Johansson *et al.* 1999), morphological changes and surface reactions of wood fibers in WPCs (Gu *et al.* 2013), and weathering of wood and their WPCs (Matuana and Kamdem 2002; Stark and Matuana 2004; Stark and Matuana 2007; Kocafe *et al.* 2013).

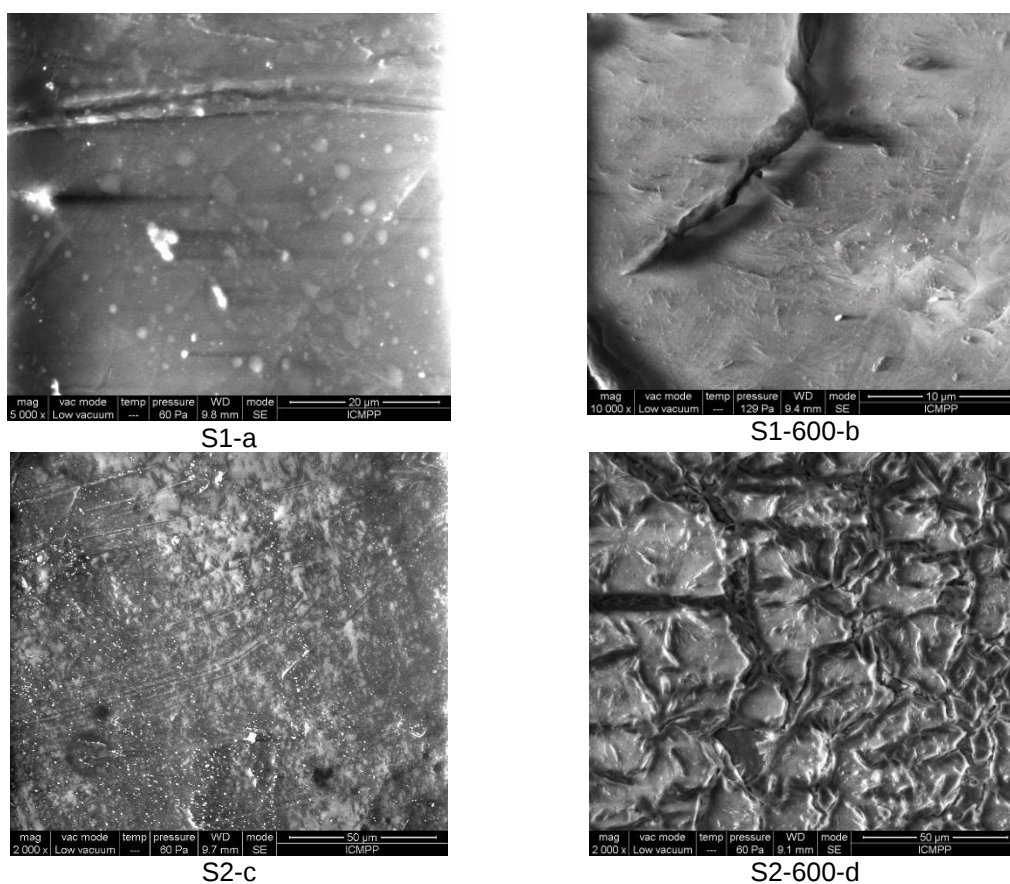


Fig. 3. Micrographs of the fractured surface composites after exposure to accelerated weathering conditions (initial: S1-a, S2-c; after 600h exposure: S1-600-b, S2-600-d)

Scanning electron microscopy (SEM) is used to assess the interfacial adhesion and phase dispersion of wood fiber in the polymer matrix. Usually, the WPCs surfaces present an increased roughness by adding wood fiber, resulting in different surface topographies with evidence of polymer-filler adhesion. SEM is also an effective method for monitoring the WPCs surface morphology changes that occur under different accelerated weathering

tests. This is exemplified in Fig. 3. a-d (Bodîrlău *et al.* 2011), where such changes are evidenced for two series of composites including samples comprising spruce wood fibers (15%), polypropylene (83%), and MAPP (2%) – coded as S1, and samples with the same composition with 15% polylactic acid (PLA) in addition – coded as S2, before (Fig. 3a, Fig. 3c), and after 600 h exposure to accelerated weathering conditions (artificial light from a mercury lamp) in a commercial climate chamber (Fig. 3b, Fig. 3d), respectively.

As one can observe, the WPCs surfaces presented an increased roughness with the addition of wood fibers, these showing a non-uniform distribution in the PP polymer matrix. Some inherent processing problems are related to the occurrence of some fine pin holes in the micrographs. Under exposure to accelerated weathering conditions, the WPCs surfaces evidenced significant visible changes such as an increased roughness leading to a fading appearance, and formation of small cracks through loss of degraded wood components in combination with the degradation of polymer matrix with negative impact on the adhesion between WPCs components.

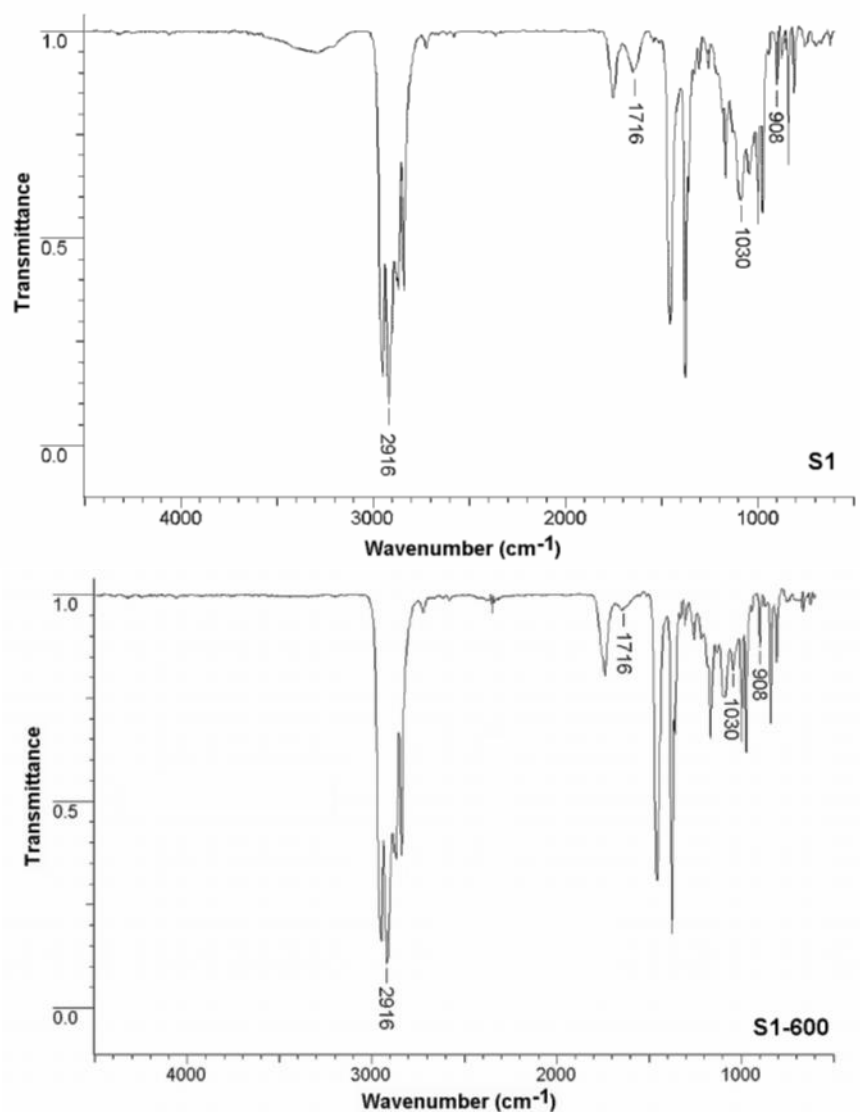


Fig. 4. FTIR spectra recorded for wood thermoplastic polymer composite: initial (S1) and

after 600 h exposure to accelerated weathering (S1-600) – (re-drawn from Bodîrlău *et al.* 2011)

The main indicators of the level of WPCs degradation upon weathering exposure under natural or controlled conditions are represented by the changes of wood index, crystallinity of polymer matrix, carbonyl index, and vinyl index, respectively (Stark *et al.* 2004; Stark and Matuana 2006). Such changes that occur after weathering exposure can be investigated by using FTIR spectroscopy.

An example of such indexes evaluated from FTIR spectra (shown only for one composite in Fig. 4) recorded for composite samples presented above is shown in Table 1 (Bodîrlău *et al.* 2011).

Table 1. Some Indexes Evaluated from Monitoring WPCs Weathering Through FTIR Spectroscopy Investigation: Vinyl Index (1a) and Carbonyl Index (1b)

(1a)	Sample/ Index	PP matrix/ Vinyl index	S1/ Vinyl index	S2/ Vinyl index
	initial	0.09203	0.0539	0.0689
	after 300h weathering	0.09	0.059	0.0752
	after 600h weathering	0.09141	0.0628	0.08

(1b)	Sample/ Index	PP matrix / Carbonyl index	S1/ Carbonyl index	S2/ Carbonyl index
	initial	0.21887	0.02154	0.0281
	after 300h weathering	0.2076	0.083	0.0797
	after 600h weathering	0.1926	0.1188	0.14

where: I denotes the peak intensity; vinyl index $= (I_{908}/I_{2916}) \times 100$;
carbonyl index $= (I_{1716}/I_{2916}) \times 100$

The peak intensity is normalized to the peak at 2916 cm^{-1} as reference, which corresponds to alkane C-H stretching vibrations of methylene ($-\text{CH}_2-$) groups that changed the least during weathering. The absorbance at 1030 cm^{-1} is attributed to C–O groups in primary alcohols of lignin. The carbonyl index is the most used parameter to evaluate the extent of degradation of polymer matrix, its increase for composites after weathering being known to be proportional to the number of chain scissions occurring in polymer matrix (Wypych 2018). Vinyl group formation is also an indication of polymer chain scission through carbonyl degradation mainly from polymer matrix as a component of wood-based composites (Stark and Matuana 2004b).

Thermal Properties

Fundamental conclusions regarding the thermal stability of composite materials can be successfully stated based on the thermogravimetry (TGA) and differential scanning calorimetry (DSC) data. Investigation related to the decomposition and glass transition temperatures is essential in establishing significant parameters, such as processing conditions and the application envisaged for the final product.

Thermogravimetric analysis (TGA) can be successfully used to assess the changes in the mass of the sample, its thermal stability and stages of thermal decomposition, if appropriate (Hatakeyama and Quinn 1994; Cofta *et al.* 2006).

TGA was extensively applied to investigate the thermal behavior of various WPCs under certain heating/cooling conditions (Oksman and Lindberg 1995). Thermal stability of the WPCs strongly depends on the wood species (Fabiya and McDonald 2010);

therefore, it has to be considered an important comparison criterion when it comes to evaluating different materials.

For example, the final stage of decomposition of hemicelluloses from hardwoods occurs at higher temperatures as compared to softwoods. So, wood species with a higher content in lignin grants their corresponding WPCs a higher thermal stability at temperatures close to their processing limit (around 200 °C).

The thermal decomposition behavior of WPCs primarily depends on their constituents. Simultaneous TG-DTG analysis evidences the differences in thermal behavior of the composite materials. Usually, an initial mass loss related to the loss of the absorbed moisture is noticed below 100 °C, and another mass loss stage appears in the temperature domain 150 to 550 °C due to the thermal decomposition process of the major constitutive polymers present in the wood: cellulose, hemicelluloses, and lignin. Hemicelluloses are known to be the least thermally stable wood components due to their chemical structure that includes thermo-sensitive acetyl groups (Bourgois *et al.* 1989). Consequently, the thermal decomposition of hemicelluloses and lignin, that are amorphous polymers, begins before cellulose degradation (Hill 2006). In the case of lignin, the thermal decomposition process starts at relatively low temperatures (Nassar and Mackay 1984). Therefore, after the initial mass loss recorded below 100 °C due to the progressive evaporation of water, the second stage registered between 150 and 500 °C is caused by the successive decomposition of the three main constitutive polymers, as follows: hemicelluloses (150 to 350 °C), cellulose (275 to 350 °C), and lignin (250 to 550 °C).

Increase of wood content may cause a slight increase of the decomposition temperature of the composites, whereas the glass transition temperature value remains practically unaffected (Valle *et al.* 2007). The resulting composites may become less rigid with the increasing wood content, this aspect being evidenced by investigation of the molecular dynamic behavior using a nuclear magnetic resonance spectroscopy (NMR) technique when proton spin-lattice relaxation time values are determined for molecular mobility evaluation. At higher wood filler content, a better dispersion of phases and an improved interaction between components have been observed, but only up to a certain threshold, above which the WPCs become brittle due to a rigid structure of wood being formed inside the polymer matrix.

Wood-polyolefins composites

In a previous study (Bodîrlău *et al.* 2009a), wood from hardwood species was modified using maleic anhydride in order to improve its compatibility (in terms of dispersion, wettability and adhesion) towards thermoplastic polymer matrices (HDPE, LDPE) in composite formulations. It was found that chemical treatment was effective in decreasing the hydrophilic character of the hardwood sawdust. The thermal properties of composites based on this type of chemically modified wood and thermoplastic polymers (LDPE, HDPE) in different formulations were investigated (Bodîrlău *et al.* 2009b). The temperature domain (T_i - T_f) for the main thermal decomposition process and the activation energy for the main temperature domain are different as in concordance with the polymer matrix and wood ratio in composite samples (Table 2).

As one can observe, thermal properties of the composites are influenced by the amount of the modified wood in the composition. The thermal stability of the polymer matrix was slightly reduced by the addition of modified wood due to its afore-mentioned complex structure. Weight loss at T_m depended on the type of thermoplastic polymer and wood content in composite samples.

Table 2. Thermal Parameters for WPCs Comprising Organic Anhydride Modified Wood (MW) and HDPE/LDPE as Thermoplastic Matrices

Composite Sample	T_i (°C)	W_{Ti} (%)	T_m (°C)	W_{Tm} (%)	T_f (°C)	W_{Tf} (%)	$\ln A$	n	E_a (kJ/mol)
MW5 / HDPE	283	2.0	484	91.0	505	91.5	11	0.0	57.8
MW10 / HDPE	292	3.5	483	89.0	502	91.8	11	0.0	56.6
MW15 / HDPE	305	10.2	478	93.0	507	93.6	10	0.0	52.6
MW20 / HDPE	335	12.0	472	94.6	508	94.8	9	0.0	51.2
MW25 / HDPE	362	17.0	467	95.0	512	97.0	10	0.0	47.5
MW5 / LDPE	370	20.6	459	72.3	488	91.3	16	0.0	49.4
MW10 / LDPE	375	18.2	454	73.5	485	92.0	16	0.0	46.8
MW15 / LDPE	370	23.4	457	73.2	485	90.6	17	0.0	48.1
MW20 / LDPE	375	25.2	455	73.8	482	90.1	18	0.0	40.1
MW25 / LDPE	371	27.6	452	73.4	479	88.6	18	0.0	37.4

where: T_i – initial temperature; W_{Ti} – initial weight loss; T_m – maximum decomposition temperature; W_{Tm} – weight loss at T_m ; T_f – final temperature, W_{Tf} – final weight loss; $\ln A$ – pre-exponential factor; n – reaction order, and E_a – activation energy

The weight loss recorded at the end of the thermal decomposition decreased along with the increasing content of the chemically modified wood in the LDPE-based composite. An opposite behavior was noticed when HDPE was used as the matrix: the increase of the modified wood content from 5% to 25% determined an increase of the onset decomposition temperature, a slight decrease of the maximum decomposition temperature (T_m), and an increase in the weight loss at maximum decomposition temperature (T_m) for all WPCs samples. Hence, this indicated a slight decrease of their thermal stability (Bodîrlău *et al.* 2007). This thermal behavior is more evident when HDPE is used as polymer matrix in composites.

Wood-PVC composites

TGA analysis was also used for another previous investigation (Bodîrlău *et al.* 2009c) upon composites comprising chemically modified hardwood sawdust (WS treated with maleic anhydride as different amounts as percentage) / PVC obtained by a melt blending process. The thermal analyses data of these composites in comparison with data recorded for wood filler and PVC polymer matrix are summarized in Table 3.

Table 3. Thermal Behavior of WPCs Comprising PVC and Chemically Modified Wood Sawdust (WS) Comparatively with PVC and Wood Filler

Specimen Code	T_i - T_f (°C)	T_m (°C)	W_{Tm} (%)	W_{Ti-Tf} (%)	W_{600} (%)	T_{10} (°C)	T_{50} (°C)	E_a (kJ/mol)	n
WS(10)	210-335	290	39.11	60.8	85.3	273	287	122.5	0.0
WS(25)	200-350	295	41.15	60.0	83.2	266	287	127.4	0.9
WS(50)	200-350	290	35.67	57.0	97.0	257	286	138.9	1.3
WS	200-355	320	38.4	50.28	97.3	252	303	90.4	1.0
PVC matrix	200-345	283	31.6	66.22	85.8	271	285	149.3	1.4

where: T_{10} , T_{50} – temperature corresponding to 10 and 50% mass loss

T_m – temperature corresponding to the maximum rate of mass loss

T_i – temperature corresponding to the beginning of the decomposition

T_f – temperature corresponding to the ending of the decomposition

W_{Tm} , W_{Ti-Tf} , and W_{600} – mass loss at T_m , T_i - T_f , and $T=600^\circ\text{C}$

E_a – activation energy, n – reaction order

Thermal decomposition behavior of modified WS/PVC composites was almost similar to wood, mainly for the sample containing 10% WS. All composite samples presented a progressive thermal degradation. The temperature corresponding to 50% mass loss (T_{50}) did not vary with the increasing amount of wood. Mass loss values significantly decreased in the temperature range of 200 to 350 °C for composite samples in comparison with PVC. The higher the wood content, the higher is the mass loss.

Nevertheless, thermal degradation of PVC involves the cleavage of some chlorine atoms, yielding extended polyene sequences that enhance the interfacial interaction with WS through intense hydrogen bonding. Relatively high values for T_m for the main decomposition process (Table 3) demonstrated the higher thermal stability of composite WS/PVC comparatively with the raw polymer.

Mechanical Properties

A WPC typically is composed of wood (40 to 65%), polyolefin homopolymer (25 to 60%), coupling agent (0 to 5%), and up to 3% of processing additives. Among these additives, lubricants are employed in order to reduce the friction between the die of the extruder and the processing melt (usually the common means to manufacture WPCs) and to reduce the viscosity of the composite melt. Although such additives can provide benefits, such as preventing edge-tearing, they may have a detrimental impact on the mechanical performance of WPCs and may interfere with other additives (such as coupling agents), as has been pertinently demonstrated for the case of PE-based composites (Wolcott *et al.* 2001).

When composites were prepared using a semi-crystalline polymer as matrix, a three-phase structure was noticed. The interphase is formed around the wood filler, where the morphology of the polymer is severely altered (*e.g.* changes in mobility and orientation of polymer chains, modification of crystallinity). The use of compatibilizers and/or toughening agents aims at an interphase with increased thickness and improved adhesion characteristics. These features are determinants for the mechanical performance of the composites (Harper 2003). This factor illustrates the intensity of the stress transfer across the interphase. In addition, the filler's strength and modulus, as well as the chemical stability and strength of the polymer matrix are of major importance for the composites mechanical properties (Gao and Tsou 1999).

Wood particles are usually considered a low-cost filler, and they are commonly employed in WPCs formulation based on thermoplastics of general use, such as PP and PE, polymers with low melting temperature, mainly when it comes to alter their mechanical properties, due to their addition to the material stiffness without increasing their density in excess (Clemons and Caulfield 2005). Furthermore, wood fibers can nucleate and initiate the crystal growth in PP and PE, with formation of a trans-crystalline layer that affects the WPCs mechanical behavior. Aside from their achieved increased stiffness, WPCs acquire an enhanced brittleness along with a reduction in all mechanical properties governed by the elastic characteristics of the polymer matrix, such as elongation and un-notched impact energy. It also has been observed that the tensile and flexural strengths may decrease as well, unless a compatibilizer is added to the formulation (Lu *et al.* 2000).

In the case of wood fibers-based WPCs, it was demonstrated that the theoretical concept of critical wood fiber length is a key factor that strongly influences the interfacial adhesion and, subsequently, the mechanical properties (Pukanszky 2005; Renner *et al.* 2010; Sobczak *et al.* 2013).

Wood fibers have a rough surface that enables the mechanical interlocking to contribute to the stress transfer at the interface, alongside adhesion or chemical coupling. Different stage methodologies for producing WPCs, *e.g.* compounding, involve mixing of both molten and solid compounds, leading to the breakage of wood fibers and decreases the initial aspect ratio. For fibers with increased length, there is an obvious reduction in the melt flow properties of the resulting WPCs that has been noticed. Furthermore, when wood particles were considered for WPCs manufacture, they were compressed during processing.

Mechanical properties of WPCs are dependent to a large extent on the interfacial adhesion (a reduced adhesion is expression of the incompatibility at the interface) and on the characteristics of wood fibers/wood particles. Composites with satisfactory properties are produced by the use of coupling with functionalized polymers if wood particles are large and have a reduced aspect ratio. Competitive micromechanical deformation processes, such as polymer matrix yielding, debonding, wood fiber pull-out and fracture, both parallel and perpendicular to the wood fiber axis, may occur simultaneously and/or consecutively (Renner *et al.* 2010a). These deformation processes determine WPCs properties. The prevalent deformation process is strongly influenced by the changes in adhesion and wood particle characteristics. An efficient approach to determine the debonding stress as a function of interfacial adhesion is based on acoustic emission measurements (Renner *et al.* 2010b).

Adhesion implies both physical (mechanical interlocking, diffusion, adsorption, and electrostatic interactions) and chemical (reactions between functional groups at the surface of matrix and filler) mechanisms (Stokke and Gardner 2003). In the case of composites with PVC, a high molecular weight polymer that presents mechanical steric hindrance, the wood component interacts through mechanical interlocking, while PVC acts as an anchor in the porous structure of wood particles, and by adsorption (hydrogen bonding and van der Waals interactions) and chemical bonding (acid-base reactions).

These theoretical considerations were supported by experimental data obtained for WPCs based on chemically modified wood (MW) and thermoplastic polymers (LDPE, HDPE) as matrices (coded as MW/polymer matrix), in different formulations (Bodîrlău *et al.* 2009a), when mechanical properties were studied. It was evidenced that strength properties of the composites can be significantly improved by adding different amounts of modified wood. Table 4 summarizes the tensile (as tensile strength, tensile modulus and elongation at break) and impact strength characteristics of these composites.

Higher tensile strength was observed along with the increasing amount of the modified wood, while there was lower elongation at break with increasing wood content for all samples, mainly when LDPE was used as polymer matrix. The Young modulus varies as a function of the wood content and the type of the polymer matrix used for production of composites. As can be seen from Table 4, this parameter increased along with the increasing content of wood for both polymers used as matrices, presenting higher values in the case of HDPE-based WPCs.

The effects of wood content on the composites mechanical properties might be explained by taking into consideration the chemical composition of wood as well. The major component of wood is cellulose, a crystalline polymer in which the fibrils are aligned and highly ordered, and between them there is a strong hydrogen bonding network. These two features confer cellulose, and wood implicitly, high stiffness. Thus, adding wood filler to a polymer matrix can increase the rigidity of WPCs by creating a skeleton inside the polymer matrix.

Table 4. Mechanical Parameters Evaluated for WPCs Comprising Organic Anhydride Modified Wood and HDPE/LDPE as Thermoplastic Matrices

Composite Sample	Density (g/cm ³)	Charpy Notch Impact Strength (kJ/m ²)	Young Modulus (GPa)	Tensile Strength (MPa)	Elongation at Break (%)
HDPE	0.95	no break	1.0	32	150
MW5 / HDPE	1.05	3.14	1.03	15.09	2.02
MW10 / HDPE	1.15	3.31	1.05	16.71	2.25
MW15 / HDPE	1.21	4.10	1.75	18.19	2.68
MW20 / HDPE	1.26	4.52	1.86	18.78	3.98
MW25 / HDPE	1.31	5.16	1.97	20.09	3.68
LDPE	0.92	no break	0.29	10	400
MW5 / LDPE	0.91	10.22	0.41	7.01	3.44
MW10 / LDPE	0.92	10.33	0.43	7.16	3.32
MW15 / LDPE	0.93	9.63	0.55	7.22	2.54
MW20 / LDPE	0.94	8.43	0.59	8.22	2.11
MW25 / LDPE	0.95	7.03	0.70	9.12	1.76

On the other hand, lignin, which is another wood component and has amorphous character, does not significantly add to the mechanical properties of WPCs, but it contributes to the binding of the cellulose fibrils, allowing an efficient stress transfer from the polymer matrix to the wood filler. Hence, the wood filler increased the stiffness (modulus of elasticity) of HDPE in the corresponding composites without increasing their density excessively as presented in Table 5 (re-written from Zănoagă and Tanasă 2011).

Table 5. Some Mechanical Properties of WPCs Comprising Softwood and HDPE as Polymer Matrix

Composite Code	Density (g/cm ³)	Tensile Strength (MPa)	Elongation at Break (%)	Flexural Strength (MPa)
HDPE 100	0.930	21.4	8.1	22.4
HDPE 60 / Wood 40	0.940	11.8	3.9	17.9
HDPE 50 / Wood 50	0.938	9.5	2.6	14.4

Wood particles can act as nucleation seeds for the crystal growth in polyalkenes, resulting in a trans-crystalline layer that can influence the mechanical behavior of WPCs (Quillin *et al.* 1993). The wood filler stiffens these polymer matrices, but it also makes them brittle, with reduction in strength properties.

In favorable cases, it can be possible to maintain tensile and flexural strengths. However, it is common that these properties are reduced without the addition of a compatibilizing agent (Lu *et al.* 2000; Clemons and Caulfield 2005). Even the use of a coupling agent cannot grant improvements in mechanical strength, because they are still limited by the low aspect ratio of the wood particles as a result of the processes used to produce the fibers from wood (Stokke and Gardner 2003), which can be either chemical (yielding carbohydrate-rich surfaces) or mechanical (yielding lignin-rich surfaces).

Processing methods used for WPCs production have a significant effect on the mechanical properties (Adhikary *et al.* 2008; Migneault *et al.* 2009; dos Santos *et al.* 2015). Thus, an injection molding process can result in better physical and mechanical properties and lower water absorption and swelling than an extrusion process (Migneault *et al.* 2009). Applying the extrusion processing method, the resulting WPCs present a decreased density

and strength due to the CO₂ released from intensive degradation of the lignin polymer component from wood filler. Differences between the processing methods can be explained by the structural variations (*e.g.* fiber alignment) and the surface quality (Migneault *et al.* 2009). Thus, surface defects (*e.g.* voids, fiber ends) result when WPCs are produced by an extrusion process, and consequently these composites present an increased water sorption and swelling behavior with adverse effects on mechanical properties. An injection molding process results in WPCs with smooth, polymer-rich surfaces, and aligned fibers, all of which lead to a better stress transfer between wood filler and polymer matrix, with further positive impact on the mechanical performance.

Water Sorption Properties

The assessment of the moisture-dependent effects, such as the fungal attack and decay of mechanical properties, is strongly related to the moisture uptake and transport properties of the WPCs (Steckel *et al.* 2007). In WPCs with PE or PP matrices, the wood particles are at least partially coated by the polymer because their hydrophobic nature creates an efficient moisture barrier, thus ensuring a certain degree of protection against moisture sorption. Water diffusion and sorption occur in WPCs in a highly complex and very rigorous manner.

The demand for polyolefin-based composites, such as PP/wood flour composites, is continuously growing. In this context, the availability of alternative wood sources could represent an opportunity to develop additional fields of application and consequently to relax constraints on the market supply. A recent study (Krause *et al.* 2017) has evidenced a significant reduction in water sorption of polypropylene composites comprising wood derived from alternative sources (*e.g.* from short-rotation coppice species, such as poplar, or from large unutilized hardwood reserves, such as beech). Nevertheless, a concept of full-tree utilization should be applied when investigating wood sources from short-rotation coppice (*e.g.* poplar, willow) because they present a considerable amount of bark content.

A significant limitation of the water uptake in the case of WPCs was reached when the wood component was thermally treated prior to their blending with the polymer (Kaboorani and Englund 2011). The pre-heated wood showed an increased dimensional stability. There are two factors that can be considered in this respect: (a) the loss of intrinsic water during the thermal treatment, and (b) an extended interpenetrating network that may have occurred between the constitutive polymers of wood as a result of the cross-linking reactions between formaldehyde (formed during the thermal decomposition of organic acids from wood) and phenolic units present in the lignin.

Table 6. Composites Samples from Wood Filler and HDPE as Thermoplastic Matrix with Addition of MAPP as Coupling Agent

Composite Code	Wood (%)	HDPE (%)	MAPP (%)	Wood Heating Temperature (°C)
C1	50	50	0	-
C2	50	48	2	-
C3	50	50	0	175
C4	50	48	2	175
C5	50	50	0	200
C6	50	48	2	200
C7	50	50	0	220
C8	50	48	2	220

In a previous work (Zănoagă and Tanasă 2012), composite samples were produced using wood filler, previously dried for removing moisture and further subjected to heat treatment (for 45 min at different temperatures—175, 200, and 220 °C—in an air circulated oven), and HDPE, adding MAPP as coupling agent in some formulations, as presented in Table 6.

Water sorption data for composite samples after immersion in water for 10, 20, and 30 weeks showed that the addition of the coupling agent and thermal pre-treatment significantly affected this phenomenon (Figs. 5 and 6). Wood pre-heating at adequate temperatures reduced the water sorption in composites samples, as shown by the water sorption decrease of 18% by heating wood at 200 °C and 220 °C, respectively. No significant water sorption changes were noticed for composite samples comprised of wood heated at 175 °C as they were absorbing water in a similar manner as the samples comprised of untreated wood.

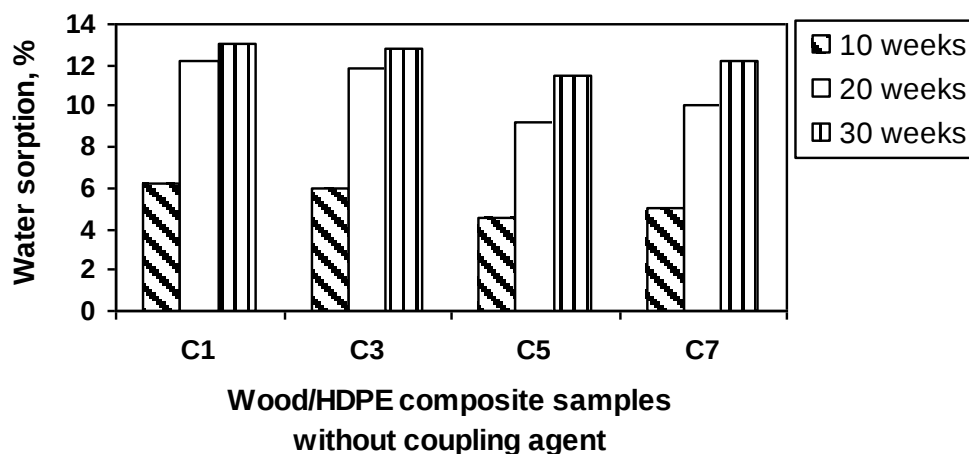


Fig. 5. Water sorption evolution for wood (non-heated and heated) - HDPE composites with no addition of coupling agent (re-drawn from Zănoagă and Tanasă 2012)

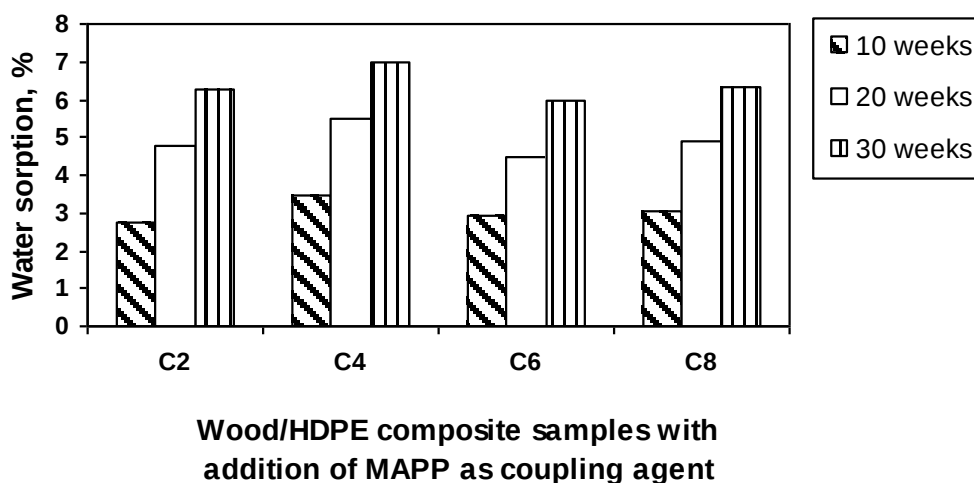


Fig. 6. Water sorption evolution for wood (non-heated and heated) - HDPE composites with addition of MAPP as coupling agent (re-drawn from Zănoagă and Tanasă 2012)

Degradation of hemicelluloses (a wood component with significant hydrophilic character) during the thermal pre-treatment can explain the reduction in water sorption values for the resulting WPCs. Composites made with wood pre-heated at 220 °C presented higher water sorption values than samples with wood pre-heated at 200 °C. This behavior can be explained by the char formation on the wood filler surface. As can be observed from Figs. 5 and 6, higher water sorption values were recorded for composites with slightly charred wood due to a more intense capillarity phenomenon favored by the structure of char, which is more porous than the corresponding wood. Adding 2% MAPP as a coupling agent to the composites formulation decreased the water sorption to about 57%. This effect of the coupling agent is more evident when comparing with reference composite samples comprised of non-heated wood.

Reduced water sorption was noticed for composite samples constituted by wood previously heated up to 220 °C, the significant effect being mainly observed for those with wood treated at 200 °C, regardless of whether or not a coupling agent was used in the composite formulation.

Moisture uptake and transport properties of WPCs, mainly those comprising PP as polymer matrix, are strongly influenced by different factors, such as: wood filler content, wood filler particle size, and use of a compatibilizer and/or other additives. Different studies showed that increasing the wood content in composite formulations determines an increased moisture uptake. The particle size is also important: the wood filler having large particle size resulting in composites having higher moisture content than wood particles of smaller size used in the same amount (Stark 2001; Bledzki and Faruk 2004). Generally, the addition of coupling agent reduced the moisture uptake and transport coefficients (Steckel *et al.* 2007).

Another important factor that affects the moisture uptake is represented by the processing method applied to manufacture WPCs. When composites are produced through an injection-molding method, the water content is reduced (Clemons and Ibach 2004), while WPCs resulting from extrusion absorb more moisture (Stark *et al.* 2004). Extruded composite samples present more exposed wood particles as a consequence of the removal of the polymer-rich surface layer, and, subsequently, these samples have an enhanced affinity for water. Composite samples obtained through injection-molding are subjected to higher processing pressures and acquire a higher density than samples prepared by extrusion. Moisture uptake, water-vapor transmission rate, and diffusion coefficients (Steckel *et al.* 2007) may be effectively determined by performing diffusion experiments (the values of the diffusion coefficients thus obtained are closer to the actual ones) and sorption tests (useful for predicting the moisture uptake regardless of the mechanisms).

Controlling moisture in WPCs is essential for improving their outdoor performance under weathering conditions. It was evidenced that co-extrusion of WPCs with a clear cap layer (Stark and Matuana 2007) greatly reduced the moisture sorption, but weathering cracked the cap layer with occurrence of delamination and subsequently changes in mechanical properties through providing a pathway for water sorption by WPCs. Some protection of WPCs against change in color after weathering was observed by adding the clear cap layer over WPCs (Stark and Matuana 2009), further protection being achieved through the addition of either a compatibilizer (for preventing delamination of cap layer), a nano-sized TiO₂ (acting as photo-blocker), or a combination of light stabilizers (UVA and HALS).

Mechanical properties of WPCs are greatly influenced by moisture sorption. Thus, the stiffness of WPCs is adversely affected by the increased water absorption level

(Tamrakar and Lopez-Anido 2011). The WPCs with high wood filler contents lost flexural properties suddenly due to the reduced strength of interfacial adhesion between wood filler and polymer matrix (Homkhiew *et al.* 2016). Flexural modulus significantly decreased rather than the strength because wood filler swelling through water sorption causes its plasticization and makes it ductile. Processing methods also can play a significant role in the mechanical performance and moisture resistance. Thus, the WPCs *via* injection molding method present lower water absorption and swelling and subsequently better mechanical properties than those *via* the extrusion method (Migneault *et al.* 2009).

CONSIDERATIONS ON THE LIFE CYCLE ASSESSMENT (LCA) OF WPCs

The environmental impact of WPCs is a constant concern of both scientists and manufacturers involved in the research and development of these materials. As the nature and type of the thermoplastic matrix varies, the environmental effects vary as well. Polymers from fossil resources are difficult to integrate into an eco-friendly cradle-to-grave cycle (Rajendran *et al.* 2012), but when recycled polymers are used for WPCs, the expectations increase (Vidal *et al.* 2009; Oneil *et al.* 2013). The use of renewable-based and biodegradable polymers, such as polylactic acid (PLA), associated with an optimized consumption of energy, may yield materials with a reduced environmental impact (Qiang *et al.* 2014).

When comparing solid timber decking products (lumber treated with alkaline copper quaternary ACQ) *versus* WPCs made of 50% recycled wood and a mixed matrix based on equal amounts of virgin and recycled HDPE, it was concluded that the environmental impact of the solid wood was significantly lower than WPCs, considering two assessments, cradle-to-gate and cradle-to-grave (Bolin and Smith 2011). Thus, according to published data, the use of fossil fuels was 14 times lower for lumber, as well as the acid rain, while the water consumption and greenhouse gas emission was 3 times lower; smog and ecological toxicity were reduced by half. As for the eutrophication, values were equal for timber and WPCs. But, when WPCs were produced using biomass or renewable energy sources, the difference in fossil fuel consumption was significantly reduced.

It was also demonstrated that the use of recycled WPCs yielded reduced environmental impacts as compared with virgin WPCs or PVC (Oneil *et al.* 2013), except for eutrophication and smog impact. On the other hand, the use of redwood lumber produced a negative effect on global warming unless it is based on resources from sustainable managed forests.

Sometimes, studies on the same biopolymer, material, or class of WPCs have provided conflicting results and opposite conclusions, as the working hypotheses, selected methods, and considered processes, or even basic aims vary (Yates and Barlow 2013). Thus, a life cycle assessment (LCA) study may be used to highlight the environmental benefits of a particular lumber relative to plastic decking (Eastin 2011; Bergman *et al.* 2014).

Nevertheless, recent comparative studies have emphasized that WPCs made of virgin materials may have low environmental impact if the wood content is significant. Even more, using wood and plastic waste will result in an even lower environmental impact, as the amount of wood in WPCs can be increased and plastics recycling industry is an already established provider of good quality materials (Miller *et al.* 2015; Wager and

Hischier 2015; Sommerhuber *et al.* 2015, 2016, 2017; Wenker *et al.* 2016). Under these circumstances, an assessment of the presence of the potentially hazardous organic compounds is required, and further LCA studies are needed in order to investigate this aspect considering the effective resources use.

CONCLUSIONS AND FUTURE PERSPECTIVES

Tailoring new wood-polymer composites within the comprehensive concept of environmental friendly and sustainable development is attracting increasing interest. This is consistent with present concerns over the rational exploitation of the renewable and non-renewable resources, materials recovery and re-(up- or down-)cycling, a more consumer-oriented manufacture, and a strongly limited environmental impact.

In terms of sustainable production, it is important to use recycled materials (the cascading principle) not only for wood, but also for the polymer matrices (Sokka *et al.* 2015). Since the interfacial interactions are of main relevance for the composites level of performance, various approaches have been designed and successfully tested: the modification of wood prior to its use in composites, by physical, chemical, and radiative methods; the reactive functionalization of the polymer matrix; the use of polymer blends as matrices; the use of compatibilizing and/or coupling agents; and the addition of various other additives, such as clays, ashes, *etc.* Even combinations of different, wisely selected methods have been employed to overcome the inherent incompatibility of wood and polymers entailed by the polarity disparity of components. The processing method and parameters contribute to the composites level of performance as well, in strong correlation with the specifications for different applications and consumer demands. Processing challenges may negatively affect the final products (fiber damage during processing, weak bonding, poor wettability, various degradation processes at the wood/polymer interface, high moisture absorption, exfoliation, and cracking), if not properly addressed. The fine tuning of the formulation-processing-properties relationship is assisted by the wide variety of investigation methods that focus on structural and functional characterization (surface and bulk, as well as aesthetic, properties). Data obtained from different tests can be interpreted in an integrative perspective, which enables the recommendation of a specific product for a particular application or type of consumer.

Still, the most sensitive issue remains the life cycle assessment (LCA) of WPCs, especially the cradle-to-grave LCA, as it aims not only at the service life, but also at the final disposal. Following the cascading use principles (Sikkema *et al.* 2013; Geldermann *et al.* 2016; Thonemann and Schumann 2017) and considering their environmental soundness (which is still under debate since it strongly depends on the selected impact categories and methodological options), it is easy to notice that repeated exploitation of plastics and wood is not realistically possible over a certain limit without a severe reduction of properties, which strongly limits the range of applications and unbalance the costs-effectiveness. Wood and plastic waste recycled from industry or landfill are already employed in composite formulations, but the final disposal of WPCs is still under research (Sommerhuber *et al.* 2017).

A current challenge is the increasing market share of WPCs. A forecast released in 2013 estimated that, in the interval 2010 to 2015, the specialized market in Europe would increase by over 65%, in North America by a third, while in Japan it would grow twofold; the average annual rate was estimated to range between 8% for North America and up to

52% for India (Eder and Carus 2013). Since WPCs containing >50% wood could be classified as wood waste, the most part of the post-consumer WPCs is often wrongly sorted and collected, and finally incinerated, which is the least preferred option for their end-of-life (EoL) with regard to environmental issues (Miller *et al.* 2015).

The best long-range approach would be the selective separation of components and their appropriate disposal: wood can be used for energy purposes (gasification, anaerobic transformation in biogas, preparation of bio-oil by fast pyrolysis, direct combustion) or in compost formulations, while plastics can be further recycled or even submitted to biological degradation in controlled biological media or landfill sites (La Mantia and Curto 1992; Arvanitoyannis 1999; Pandey *et al.* 2003; Shah *et al.* 2008; Yoshida *et al.* 2016). Soil composting is an already established method for plastic waste management – polyolefins, PET, PVC, PS, and aliphatic copolyamides. However, the overall costs associated with such a hi-tech disposal strategy are prohibitive. Moreover, the industrial flow is difficult to organize, and there are still no investors interested in complex collecting-processing-separating-recovering facilities that, in the end, will be supplying the final disposal stations.

Nevertheless, WPCs research and development remains an active field for academic and industrial media as far as the societal demand for such versatile materials is increasing not only in volume, but in quality as well. At the same time, these issues may raise concerns on the finite land resources for wood and biopolymers and the competition with food crops and bioenergy crops. Even more, considering the increasing rate of replacement of fuels and plastics from fossil resources with alternatives from renewable resources, it has been estimated that by 2050 the competition between food consumption, on the one hand, and industrial and energy consumption, on the other hand, will be so tight that agricultural production will hardly cover the demands (Colwill *et al.* 2012). In terms of a technological approach, a low-energy method to fast heating at high temperatures in order to enable a satisfactory dispersion, a high quality product with low moisture content and high load of wood, might prove to be one innovative direction for the development of WPCs. Comparing impulse and cyclone heat treatment to other heating methods, these methods can readily remove free water of materials (both free and bonded) because of the long stage of material processed inside the cyclone unit and to the acceleration-deceleration cycles in the impulse unit (Chen *et al.* 2017). Thus, the mass and heat transfer efficiency can be increased, as well as the fiber quality due to the high temperature treatment (less free hydroxyl groups on the wood surface, lower surface polarity, enhanced surface roughness, and increased cellulose crystallinity).

At the same time, the use of nanometric-scale additives may significantly improve the WPCs performance, *e.g.* nanoparticles of titanium oxide (Stark and Matuana 2009), or cellulose nanofibrils (Hubbe *et al.* 2008; Siró and Plackett 2010), thus widening the range of applications of WPCs.

The use of biopolymers instead of virgin and secondary polymers can reduce up to a certain degree the environmental negative effects, but new approaches are required. Recent studies reported on a novel way to transform oil-based plastic waste into completely biologically compostable materials using concepts and methods from synthetic microbiology/biology and employing the bacteria *Pseudomonas putida* (Wierckx *et al.* 2015). Thus, new WPCs based on bioplastics obtained from classic plastics waste may be developed in the coming years, allowing a complete recycling circle.

Even recycling technologies can benefit from innovative research, as it is part of the development of a sustainable global economy. Molecular recycling, employing novel

methods such as molecular sorting (Forberger and Becker 2016; Woidasky et al. 2016), fluorescence sorting (Langhals *et al.* 2014, 2015), and NIR (Meinlschmidt *et al.* 2014; Li *et al.* 2015), is now under research and seems to be a viable alternate option as compared to bulk sorting.

Nevertheless, nowadays market demands and economic limitations remain of utmost importance. Human society has used wood for millennia, and wood sources will continue to have a significant role if only forest management considerably improves. On the other hand, the plastics industry has to be more environmentally sensitive and employ innovative solutions provided by research advances. The recovery and re-(up-) cycling concerns must get increased attention and funding. All these directions, as well as associated and subsidiary interests, must converge so as to meet the challenges of society not only for present needs, but foremost for the generations to come.

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