UNIVERSITÀ DEGLI STUDI DELLA BASILICATA
SAFE - Scuola di Scienze Agrarie, Forestali, Alimentari ed Ambientali

SAFE - Scuola di Scienze Agrarie, Forestali, Alimentari ed Ambientali

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Title of Thesis

Characterization of particleboards produced with Orange (*Citrus sinensis* L.) and Turkey oak (*Quercus cerris* L.) wood species using Modified Starch as adhesive

Scientific-Disciplinary Sector

“AGR/06 Wood Technology”

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Cycle XXXIV

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Preface

The Ph.D. program was supported by the grant of Region Basilicata-Italy to develop a partnership between the University of Basilicata and the industrial sector, through the "Industrial 4.0" strategy. The research program, supervised by my Tutor Prof. Todaro, had as main objective to highlight the importance of agroforestry biomass, aiming to avoid wastage in the use of raw materials. In particular, the present research focused on developing PBs produced with wood biomass from branches of Orange tree (*Citrus sinensis* L.) crops and Turkey oak (*Quercus cerris* L.) as substitute raw materials for commonly used particles in PBs manufacture. Orange tree crops predominate the southern part of Italy, while Turkey oak forests are widespread in the Mediterranean area. The management of both wood species produces a large amount of residual biomass without an economic value, and often burned in the field or used to generate heat, energy, or in the best case used as compost for the cultivations. To avoid this loss of material, it was decided to assess the suitability of Orange tree pruning residues and Turkey oak wood residues as an alternative raw material for PBs intended for general use as well as in interior design. Furthermore, since the majority of PBs are manufactured with Urea-Formaldehyde (UF) as binder, known as toxic substance for human health due to its formaldehyde release in indoor conditions, it was promoted a more sustainable adhesive, namely modified corn starch (MS), to develop an industrial product with a lower environmental impact. Therefore, to evaluate the performance of the new PBs with MS adhesive, the comparison was made with UF adhesive panels, having the same structure and Orange and Turkey Oak wood particles ratio. Mechanical properties (modulus of elasticity, modulus of rupture and internal bond strength) and physical (thickness swelling and water absorption) were analyzed. Most of the experimental research was conducted at the National Research Council of Sesto Fiorentino (Italy), since the period characterized by the pandemic emergency has led to problems in continuing the tests for one year at the Department of Wood Processing and Wood Products Design in Brasov (Romania). However, during the foreign experience, it has been possible to acquire the panel manufacturing methodology thanks to the precious availability of my Co-tutor Prof.ssa Ing. Octavia Zeleniuc.

Six months internship stage in smart working mode was carried out at the lucanian company Colacicco S. a. s. to share and discuss the results of this experimental work in order to identify the best PB for practical applications. Specifically, results from the experimental panels tests were compared with the European standard requirements that classifies panels according to their final use. The company tutor Luca Colacicco, over the years, evolved and it is well known both in the upholstered furniture sector and in interior design.

Introduction and Aims

The European Union (EU) land surface is 2,213,323.59 ha, of this, about 462,855.94 hectares are occupied by agriculture crops and 1,015,482.48 ha by forests, with produced biomass of approximately 956 Mt of dry matter for agriculture and 18,600 Mt of dry weight for forests. It is worth mentioning also the constantly increase of the forest area in EU, according to the data reported on the State of Europe's Forests (SoEF 2020). It currently amounts to about 227 million ha, without considering the Russian Federation, representing almost 35% of the total European land area. At almost 92%, Central-West Europe has the highest share of forests available for wood supply while South-East Europe has the lowest share at about 53%. The area of forest available for wood supply is increasing between 1990-2020 and 2010-2020, with the exception of North-Europe where the area of forest available for wood supply has declined. In Italy the total forest area is 36.4% of the national surface and it increased by 6.6% in the period 2005-2019 (EUROSTAT 2019). In the period 2015-2020 Italian forests have continued to expand, occupying 11,4 million ha, almost 40% of the national surface (FAO 2020). Despite the increased forest area, and consequently the availability of the wood raw material, the 80% of the wood used by Italian manufacturers is imported from other countries (Marchetti et al. 2018). Therefore, one of the primary objectives of forest policy in Italy is to increase the degree of self-supply in this sector (Romagnoli et al. 2019). In this contest, less used wood species that are found in the Mediterranean region can be considered for industrial purposes.

In the Italian Mediterranean regions, excluding the islands, fruit tree crops occupy an area of approximately 1,059,048 ha, where Orange tree (*Citrus sinensis* L.) crops represent the 3% (ISTAT 2018). In Basilicata region the area destined for agricultural production is equal to 833,847 ha, and of these 50,281 ha are destined for tree crops and 12% is represented by orange groves (ISTAT 2018).

Regarding tree crops management, their canopy requires at least one pruning per year. Furthermore, some of these crops, have a life cycle that lasts about 15/16 years, and then they are explanted. Pruning and harvesting of orchards generate significant amounts of wood, which are often burned or sometimes shredded and left into the soil to increase the organic matter (Bruno et al. 2021).

Turkey oak (*Quercus cerris* L.) coppices is the most widespread forest type throughout all the South-Eastern Mediterranean countries, covering 675,53 ha, which is equal to 18.4% of the forest area under the coppice system in Italy (INFC 2005). In Basilicata region, according to the available data reported by the National Inventory of Forests and Forest Carbon Tanks, Turkey oak forests cover a surface of 83,625 ha, accounting for 23% of the entire regional forest area (356,426 ha) (INFC 2005). The traditional cultivation goals are firewood, charcoal, fodder for cattle breeding and, since the 1800s, the production of railway sleepers (Piussi 2015). Actually, Turkey oak residues sourced from forests management are used mainly for energy purposes since its wood is difficult to treat and, for this reason, less appreciated compared with other deciduous oaks for furniture, floors and veneer (Uzielli 1989; Todaro et al. 2013).
In general, burning orange and oak wood residues generates various problems, the first is the production of CO_2 into the atmosphere (Darley et al. 1966), the second is due to the legislation that has set limits to this practice, so it is not always possible to burn these residues. Due to these problems related to the management of wood residues, possible alternative uses of these materials are being sought in different industrial sectors. Wood residues or biomass coming from agroforestry activities could be considered potential raw materials for new products with high added value, such as particleboards, the basic wood products mainly used in interior design and particularly in furniture industry, considered under the general name of Wood-based panels. Wood-based panels, *e.g.* particleboards, oriented strand board, and medium-density fiberboard are among the most prominent product categories in Northern America and Europe, while veneer and plywood (inclusive of blockboard) represent the main wood-based panel product in the Asia-Pacific region, mostly in China. Among the main particleboard producers are China, Russian Federation, Germany and United States which alone account for 69% of the entire world production (FAOSTAT 2020).
In Latin America and the Caribbean region, each major wood-based panel product accounts for about an equal share of the total production. The total European production of oriented strand board increased from 200,000 m^3 (3%) in 2018 to 6.9 million m^3 due to the strong production capacity in Luxembourg (EPF 2019). Wood-based panel production in the EU grew by 1.7% in 2018, reaching 59.3 million m^3.

Growth in the global production of particleboards was the fastest among all reconstituted panels categories, recorded at 25% from 2014 to 2018. Particleboards in 2018 covered over half of the total wood-based panels production in Europe, increasing by 0.3% to 37.8 million m^3 in 2018. According to the last data, Germany and France are the leading producers of particleboards in Western Europe (FAOSTAT 2020). Figure 1 shows the share of the wood-based panel production in 2020 referred to Europe, United Kingdom and European Free Trade Association Countries.

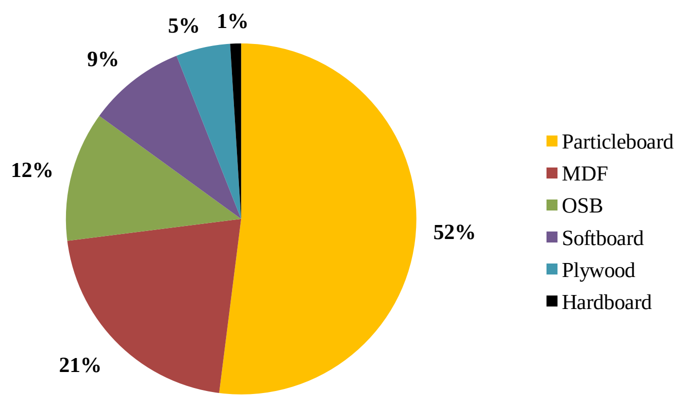

Figure 1. Share of wood-based panel production in Europe, United Kingdom and in the European Free Trade Association Countries (© European Panel Federation 2020).

In the light of the increasing wood-based panel demand and steady rise in raw material prices, particleboards industry needs low price raw material. Several studies have focused on various wood residues and other lignocellulosic material to manufacture particleboards (Barboutis and Philippou 2007; Amini et al. 2013; Klímek et al. 2016). The research project aimed to promote the underutilized wood species such as *Citrus sinensis* L. and *Quercus cerris* L. which have not been the subject of any study to date, as raw material for the prominent particleboard sector. Furthermore, the potential of Modified Corn Starch as bio-adhesive was evaluated, since the majority of particleboards are manufactured with Urea-Formaldehyde resin, which is toxic for human health. This would mean to offer also the opportunity to open avenues to bio-based adhesives for PB producers.

The idea to develop particleboards in terms of attractivity, eco-friendliness, lower cost, is perfectly suited to the concept of "circular economy" according to which it is necessary to achieve "zero waste" and exit from the current economic perspective take, make and dispose" (Harale 2021). The enhanced technological possibilities to utilize wood waste considered as a low quality raw material, and other types of residues in the production of WBP help towards the transition to a circular bioeconomy. The cascading use of wood resources, defined as "*the efficient utilisation of resources by using residues and recycled materials for material use to extend total biomass availability within a given system*" is one of the most important principle to follow (Neykov 2020).

The exploitation of wood residues coming from the major wood species present in Basilicata Region to produce PBs will lead to:

- reduce practices with high environmental impact such as wood biomass incineration;
- increase the opportunity to use wood residues for other purposes besides energy use;
- use of alternative wood materials, such as local resources, to traditional ones in PBs manufacture.

In a historical moment in which environmental issues have assumed a key role in the economic and social planning of all States, the doctoral work aims at the development of industrial prototype produced with a lower environmental impact adhesive and with local wood resources that might gain the attention of the furniture industry and especially to upholstered furniture sector.

In the Figure 2 the specific objectives of the research are presented.

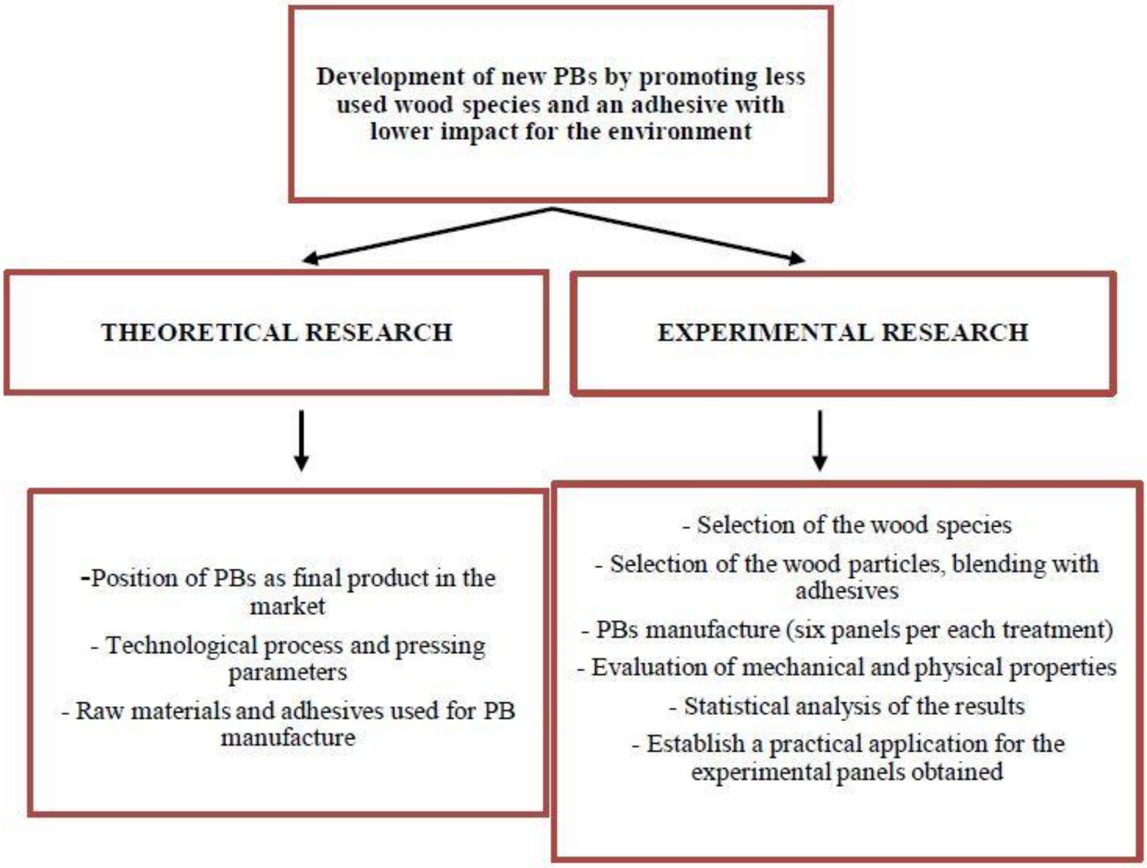

Figure 2. Objectives of the research.

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Abstract

The continuing rise in raw material prices imposes the search for new raw materials with low economic competition. Generally, wood species utilized in particleboards (PBs) production are either softwoods like red fir, spruce and pine, or hardwoods like beech, poplar and birch. In recent years, several investigations into the potential advantages of using annual plants, fast-growing species and agricultural residues in the PBs production and on their performances as a raw material for PBs have been conducted. The present research focused on developing PBs produced with wood branches of Orange tree (*Citrus sinensis* L.) from crops and Turkey oak (*Quercus cerris* L.) wood residues from forest stand. Orange tree crops predominate the southern part of Italy, and Turkey oak forests are widespread along the Apennine mountain, especially in Basilicata Region. The management of both wood species produces a large amount of biomass often burned in the field or sometimes shredded and left into the soil to increase the organic matter, or used for energy purposes. Generally, PBs are manufactured with Urea-Formaldehyde (UF) as a binder, known as a toxic substance for human health. In this research Corn Starch modified with glutardialdehyde (MS) was promoted as a low environmental impact adhesive. PBs produced with orange and oak wood particles and using two types of adhesives, UF and MS, were compared and evaluated in terms of mechanical properties, such as modulus of elasticity (MOE), modulus of rupture (MOR), and internal bond strength (IB), and physical properties, such as thickness swelling (TS) and water absorption (WA). The obtained data were statistically analyzed using analysis of variance (ANOVA) and Duncan's mean separation tests. All panels produced with UF adhesive showed good mechanical performances. Panels produced with the mixture of UF and MS showed acceptable MOE and IB values, complying with the minimum requirements of the standard EN 312:2004 for P2 panels type, namely non-structural panels including furniture for use in dry areas. TS values met also the requirement for P3 panels type intended for use in humid conditions, according to the standard EN 312:2004, but only in the case of the panels produced with UF adhesive. The use of MS has negatively affected the physical properties (TS and WA) and resistance to bending (MOR). However, the produced

panels showed good performance for indoor applications, where dimensional stability is not a strict requirement.

Keywords: Particleboard; Orange wood; Turkey oak wood; Modified Corn Starch; Urea-Formaldehyde; Mechanical properties; Physical properties

List of abbreviations

ANOVA= Analysis of Variance

MC= Moisture Content

MDF = Medium Density Fiberboard

MS= Modified Starch

MOE= Modulus of Elasticity

MOR= Modulus of Rupture

IB= Internal Bond

OSB= Oriented Strand Board

O UF= Orange particleboards with Urea-Formaldehyde

TO UF= Turkey oak particleboards with Urea-Formaldehyde

O+TO UF= mixed Orange and Turkey oak particleboards with Urea-Formaldehyde

O (UF+MS)= Orange particleboards with Urea-Formaldehyde and Modified Starch

TO (UF+MS)= Turkey oak particleboards with Urea-Formaldehyde and Modified Starch

O+TO (UF+MS)= mixed Orange and Turkey oak particleboards with Urea-Formaldehyde and Modified Starch

PB= Particleboard

PBs= Particleboards

TS= Thickness swelling

WA= Water Absorption

WBP= Wood-Based Panels

READING GUIDE

A brief guide on what is reported in each chapter is summarized to facilitate the reading of this thesis.

CHAPTER 1 provides basic data on wood-based panels, including their market and industrial applications.

CHAPTER 2 provides an exhaustive description of the typical manufacturing process of particleboards.

CHAPTER 3 reports researches on different types of lignocellulosic biomass as raw materials to produce particleboards and adhesives used.

CHAPTER 4 reports botanical and other characteristic of the wood species, such as *Quercus cerris* L. and *Citrus sinensis* L., the raw materials selected for the experiments.

CHAPTER 5 presents materials and methods to manufacture the experimental panels as well as the mechanical and physical tests to which the panels were subjected.

CHAPTER 6 provides the results and discussions of the experimental work.

CHAPTER 7 shows the conclusions and remarks.

CHAPTER 1 OVERVIEW

1.1 Wood-based panels

Wood-based panel (WBP) is a general term to indicate a wide range of engineered composite type panels produced from byproducts such as chips, fibres or veneers of the wood processing adhered together with termosetting resins (Papadopoulou 2009). PBs, medium density fiberboards (MDF), and oriented strand board (OSB) are the most commonly used products of the wood panel industry (Irle and Barbu 2010; Gonçalves et al. 2018). While some panel types are relatively new on the market, others have been developed and successfully introduced more than hundred years ago. The first wood based panel dates back to 1887, where sawdust bonded with a blood-albumin adhesive was pressed to obtain a flat panel (Hubbard 1887). Over time, the WBP development was driven by tuning and further evolution of woodworking machines, synthetic adhesives, and some technological innovations. Technological development together with new market requirements and the steadily changing raw material situation led to continuous improvements of WBP and their manufacturing processes (Irle and Barbu 2010). The reasons for their rapid affirmation are reflected in the need to overcome the typical dimensional irregularities of the roundwood retractable from the arboreal stems, obtain products with specific geometric characteristics, and create semi-finished products with large surfaces and high dimensional stability. The main advantage is that WBP can reduce wood defects and their influence, limit the moisture gradients and anisotropic behavior typical of many wooden materials (Zanuttini 2001). In addition, their importance is related to their function of representing an alternative and sustainable use of lignocellulosic residues. Depending on the type of the starting wooden material including solid wood, veneers, particles or fibres which comes from hardwoods and softwoods as well as from other lignocellulosic materials, WBP aim to cover specific needs of the modern industry, as illustrated in Figure 3.

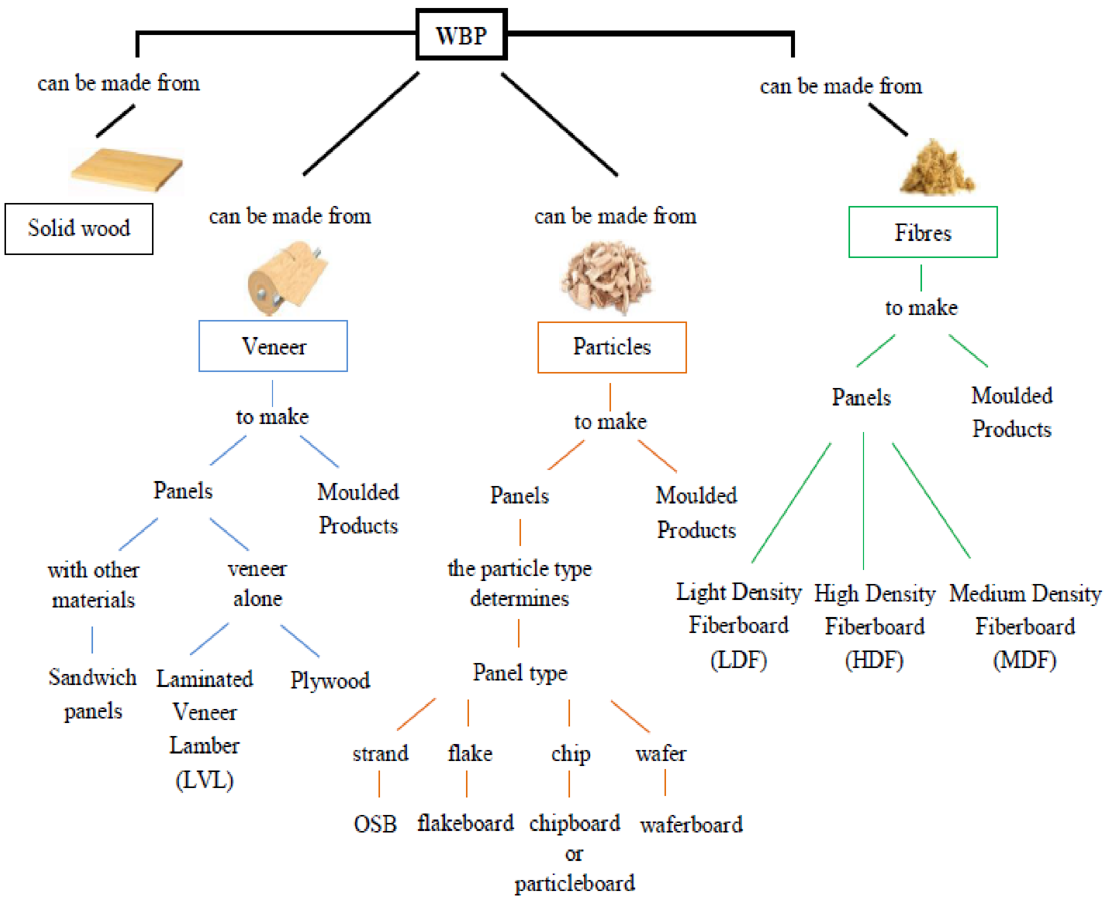

Figure 3. A map summarizing a share of the wide range of WBP (figure adapted and partially modified from Irle and Barbu 2010).

Specifically, biomass material for panels production is represented by unprocessed forest products, industrial residues from wood processing or agricultural wastes in the shape of fibres, shaving, chips or other type of particles (Maloney 1993).

1.2 The Wood-Based Panel market

1.2.1 Europe

The total WBP production decreased by 0.9% in Europe to 74.0 million m^3 in 2019. Plywood experienced a 2.4% drop. MDF and PB which comprise the group of non-structural panels, contracted by 2.2%, and softboard posted the largest production decrease among the subcategories, dropping by 8.6%. The production of hardboard weakened by 2.9% (EPF 2020). OSB and plywood, defined as structural panels, in 2019 declined by 1.4% in terms of production (Figure 4).

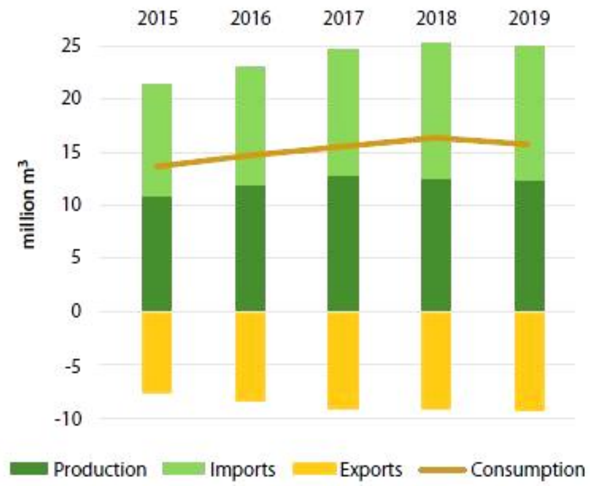

Figure 4. Europe: structural panels production, trade and consumption in the period 2015-2019. *Note*: exports are shown as negative numbers. (Source: UNECE/FAO database, 2020).

Demand for structural panels dropped by 3.4% in 2019 and for non-structural panels kept constant (-0.1%). Exports decreased only slightly in volume (by 0.5%) but significantly in value (by 5.2%). Higher exports of structural panels (up by 1.9%) offset lower exports of non-structural panels (down by 1.4%). Imports were stable in 2019 for non-structural panels (up by 0.2%) but contracted significantly (by 1.6%) for structural panels (Figure 5). The total consumption of WBP decreased by 0.8%.

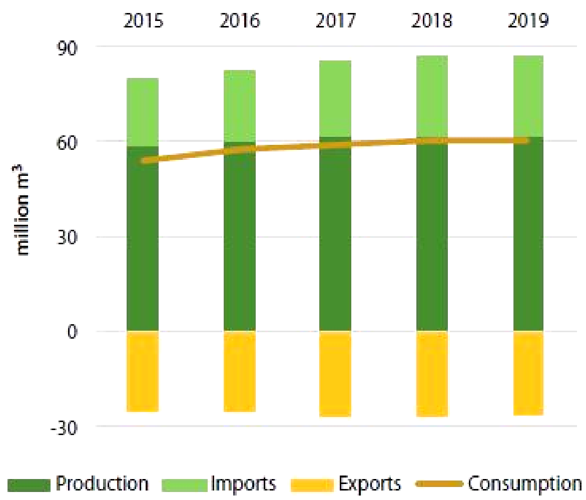

Figure 5. Europe: non-structural panels production, trade and consumption in the period 2015-2019. *Note*: exports are shown as negative numbers. (Source: UNECE/FAO database 2020).

1.2.2 Eastern Europe, Caucasus and Central Asia

Total WBP production increased by 1.1% in 2019 in Eastern Europe, Caucasus and Central Asia (EECCA), and by 1.3% in Russian Federation, reaching 26.2 million m^3 and 17.6 million m^3, respectively. Apparent consumption increased in EECCA in 2019 for both structural panels up by 0.5%, to 3.9 million m^3 and non-structural panels up by 0.6%, to 17.4 million m^3 (Figures 6, 7). The apparent consumption of WBP fell by 1.7% in the Russian Federation, to 12.4 million m^3.

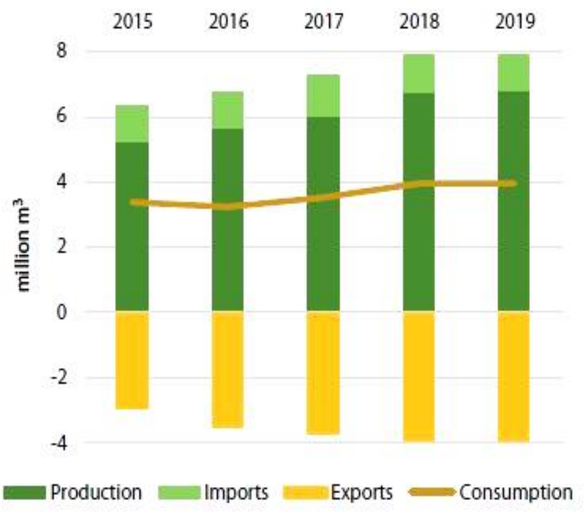

Figure 6. EECCA: Structural panels production, trade and consumption in the period 2015-2019. *Note*: Exports are shown as negative numbers. (Source: FAOSTAT 2020).

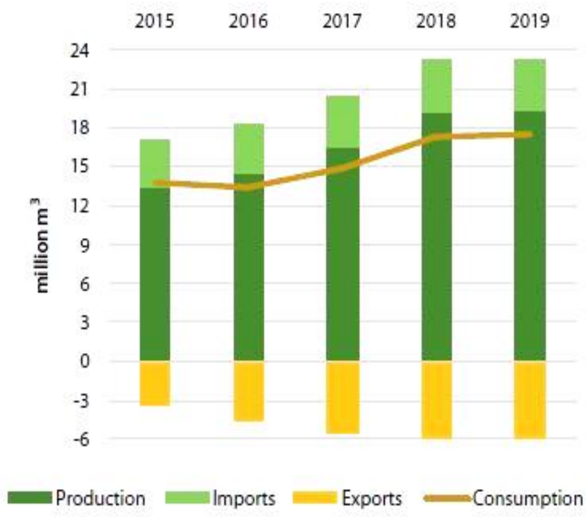

Figure 7. EECCA: Non-Structural panels production, trade and consumption in the period 2015-2019. *Note*: Exports are shown as negative numbers. (Source: FAOSTAT 2020).

The trade volume of WBP grew over three years (2016-2018) in EECCA due to the trade developments in the Russian Federation, but in 2019 it remained constant. Imports by the Russian Federation is decreased in 2019 for all WBP (plywood by 14%, MDF by 4%, PB by 14% and OSB by 36%) due to the weakening of the Russian rouble against the US dollar (by 3.1%).

1.2.3 North America

Total WBP production was 45.9 million m^3 in North America, decreasing by 1.3% (Figure 8). The production capacity in North America increased by 1% in 2019 (CPA 2020). Figure 8 shows as the consumption of structural WBP decreased gradually by 4.7% in North America during the period 2018-2019, and the demand for OSB and plywood decreased by 4.1% and 5.6%, respectively. Trends in the consumption of structural WBP in the four principal end-use applications are as it follows: +0.04% in the residential construction sector, +0.6% in the restructuring sector, -1.8% in the non-residential sector (APA 2020).

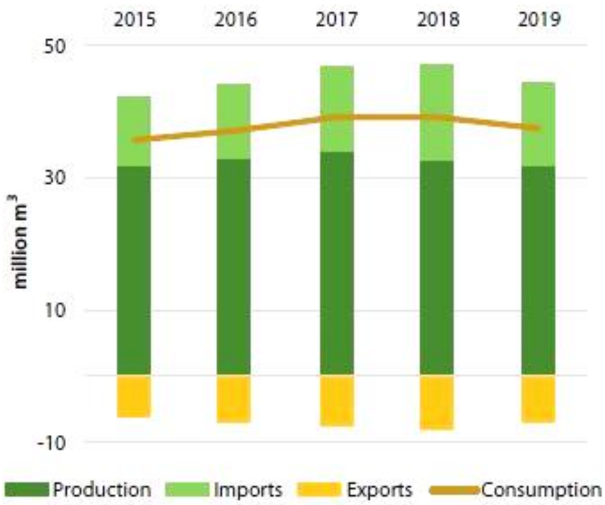

Figure 8. North America: Structural panels production, trade and consumption in the period 2015-2019. *Note*: Exports are shown as negative numbers. (Source: UNECE/FAO database 2020).

PB and MDF consumption, known as non-structural panels, increased by 0.9%. However, MDF consumption registered a decrease of 4.0% balanced by an increase of 11.9% in PB consumption (Figure 9).

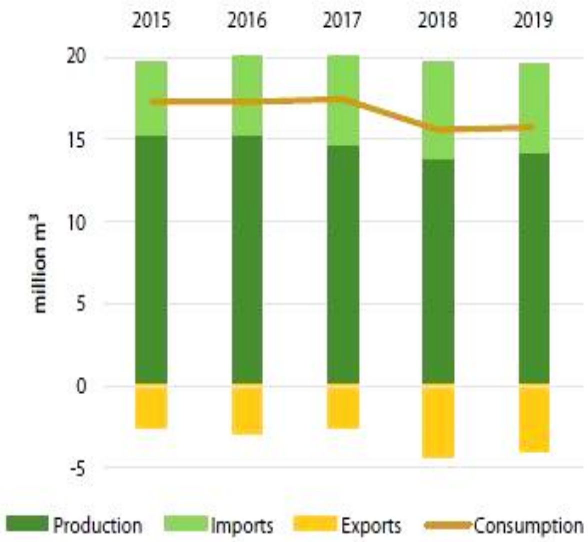

Figure 9. North America: Non-structural panels production, trade and consumption in the period 2015-2019. *Note*: Exports are shown as negative numbers. (Source: UNECE/FAO database 2020).

In North America imports of WBP sharply dropped by 21.0% in 2019, to $5.8 billion. The value of US imports decreased, except for PB which increased by 10.2%. In general, the value of Canada WBP imports, decreased by 8.1% in 2019, even though imports of PB and OSB increased. WBP exports from North America decreased by 19.3% in 2019. The exports from North America inclusive of the trade between Canada and US, decreased by 32.3% for OSB, 14.8 for plywood and 1.1% for MDF, while PB registered an increase over 2018. As shown in the figure 8, the consumption of WBP dropped down after 2017, due to an overall weakening in demand in North America continued until 2019, leading to a decrease of the capacity utilization rates.

1.3 Industrial application

Due to their high performances and low cost, WBP have revolutionized the woodworking industry, leading to the creation of wood-based products in accordance with market demands, trends in furniture design developing and with the modern national furniture industry (Rossi and Spallazzo 2021). WBP is the basic material used in different industrial applications (Figures 10, 11, 12 and 13).

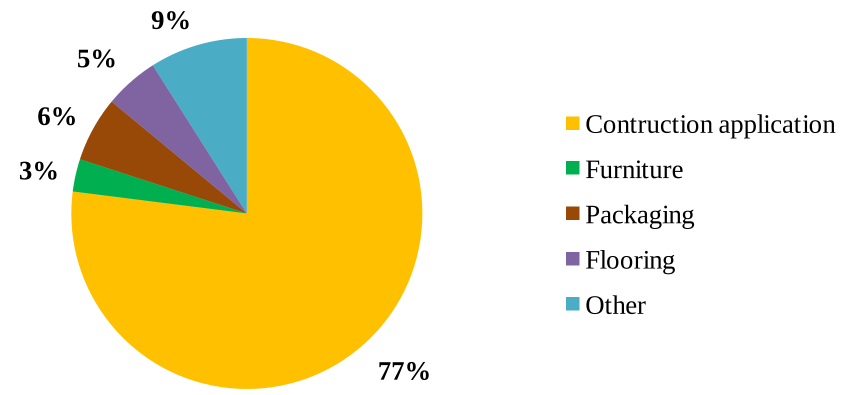

Figure 10. OSB End-Uses, 2020 (© European Panel Federation).

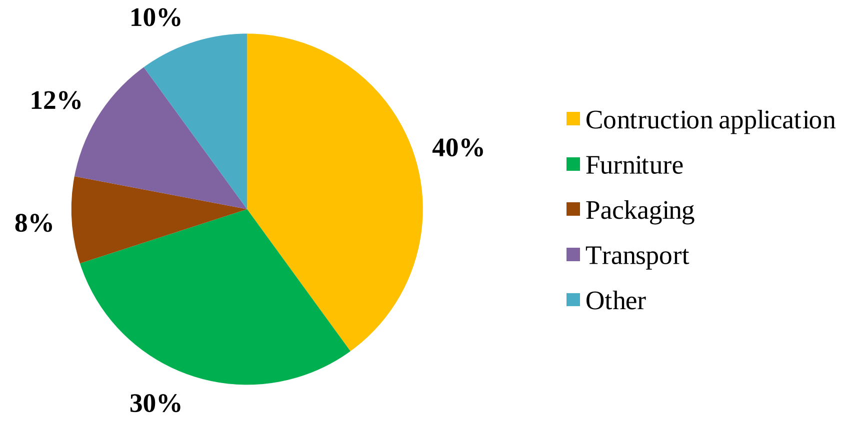

Figure 11. Plywood End-Uses, 2020 (© European Panel Federation).

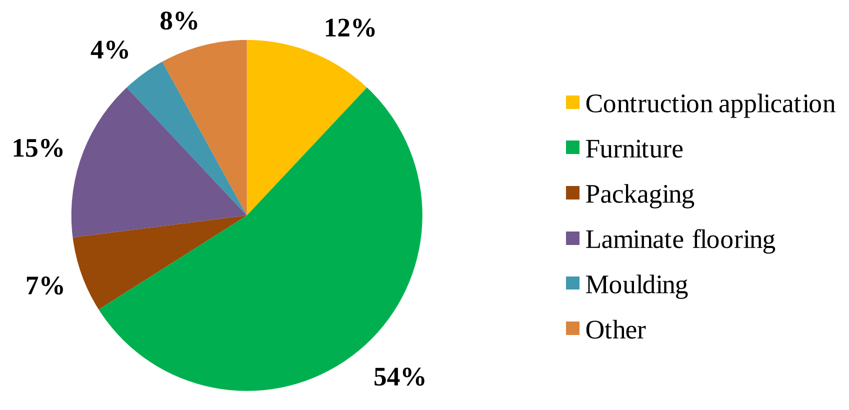

Figure 12. MDF End-Uses, 2020 (© European Panel Federation).

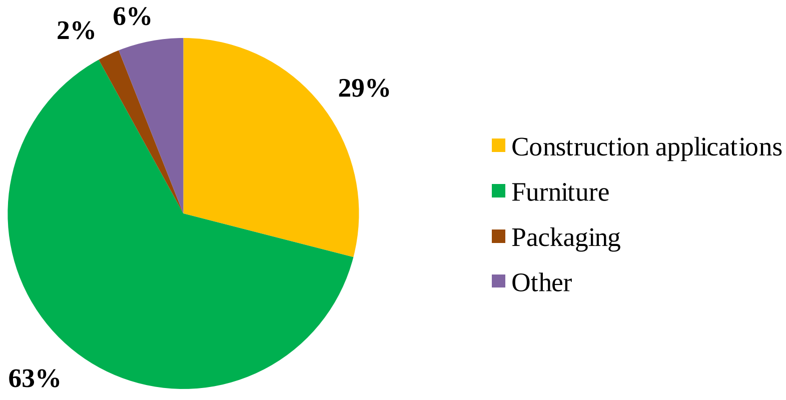

Figure 13. PB End-Uses, 2020 (© European Panel Federation).

OSB is mainly a panel for construction application, including flooring, flat roof decking and wall sheathing (Figure 10). Plywood is mostly applied in the fields of construction and furniture (Figure 11). Standard MDF, or specifically its variant High Density Fiberboard, is used for 54% in furniture industry (Figure 12). As shown in the figure 13, PBs are used for under-floors as insulating material, for wall, in packaging but most of the applications refer to the furniture industry for kitchen, bathroom, dining-room, bedroom, and hall. It is clear that, overall, WBP production play an important role in the construction and furniture industries worldwide.

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CHAPTER 2 PARTICLEBOARD TECHNOLOGY

Technology for PB manufacture has developed so that almost every type of wood and non-wood lignocellulosic materials can be use as raw material in its manufacture.

2.1 Definition of Particleboard

According to the Standard EN 309 (2007), a PB is defined as a “wood-based panel manufactured under pressure and heat from particles of wood including wood flakes, chips, shavings, sawdust and similar and/or other lignocellulosic material in particle form (flax shives, hemp shives, bagasse fragments, straw and similar) with the addition of an adhesive”. Most of the European countries use the term “particle” instead of “chip”, therefore, to avoid confusion, the term particleboard with its abbreviation PB, will be used within the text. The wood species from which the particles are made is a crucial factor in PB production technology. Panels made from softwood species having low density, such as pine or poplar, demonstrate better mechanical properties than panels made from hardwood species with high density, such as birch or beech (Starecki et al. 1994). Most of the PBs are manufactured using a layering system, where the core layer contains large particles and the outer layers finer particles, allowing improved surface finish (de Barros Filho et al. 2011). The type of particle determines the type of panel. For example, a chipboard is made with chips, according to the English terminology, a flakeboard with flakes, OSB with strands, and so on (Irle and Barbu 2010). The particles are bonded together by adding a synthetic adhesive, and then pressed into a large board shape inside a heated press under high pressures and temperatures values, then cooled, cut into various sizes and prepared for delivery. PB can be coated or laminated to improve its aesthetic value and resistance, and can be easily treated with fire retardants or with a certain types of preservative chemicals (Popescu 2017). The preparation of the adhesive mixture is an important step and it is a part of the typical sequence of technological operations to manufacture a PB as depicted in figure 14.

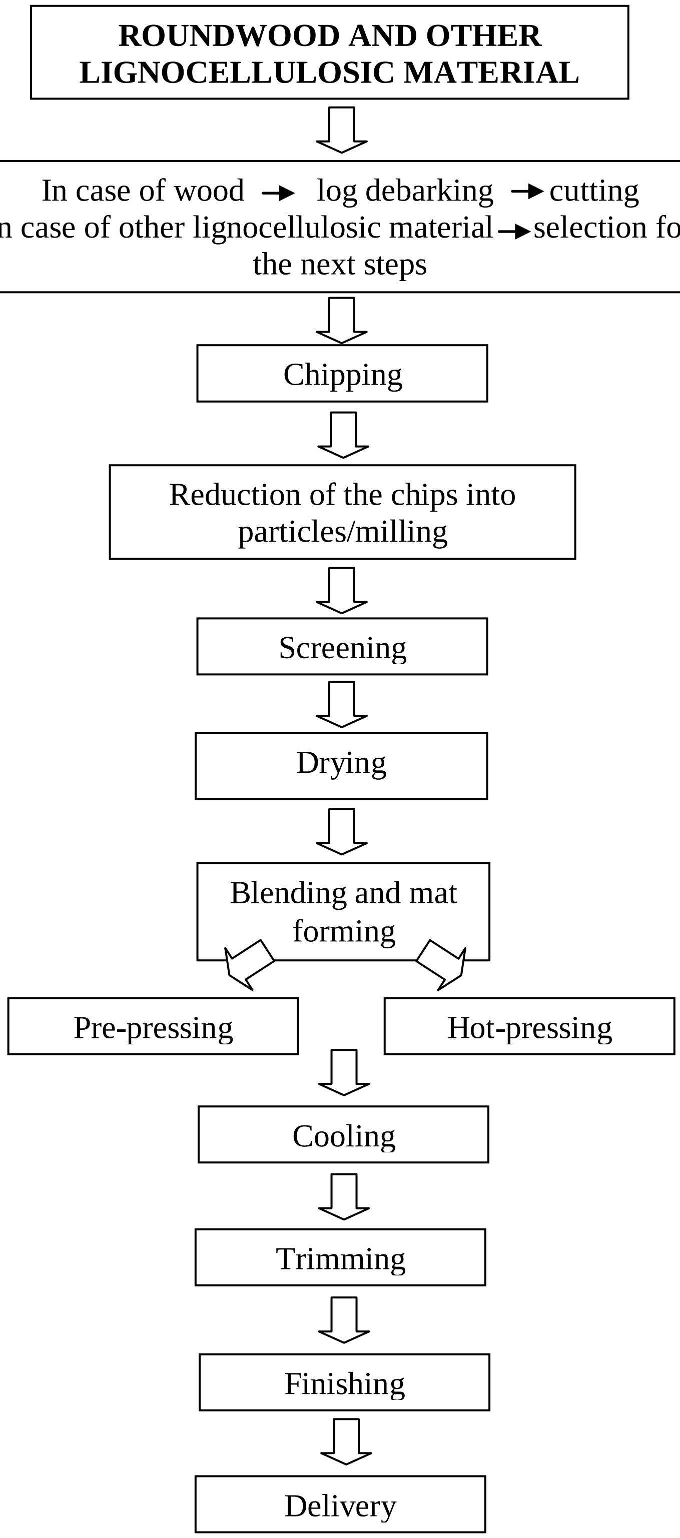


Figure 14. Diagram of the PB manufacturing process.

The pressing parameters, and especially the temperature involved in the production process, have a marked influence on the subsequent properties of the final panel.

2.1.1 Roundwood and other lignocellulosic materials

Generally, the raw material used for PB production comes from roundwood reduced into particles. The demand for solid wood and solid wood residues is in continue increase, thus PBs manufacturers are looking for other resources from category of low grade residues, such as, sawdust, planer shavings, and unused wood species as well as seasonal crops, such as straw from cereals, flax, bagasse and many others (Papadopoulou and Chrissafis 2017).

2.1.2 Debarking/Raw material preparation

Ideally, the bark should not be included in PB as it reduces the strength properties of the panel and increases resin demand. Logs are therefore debarked (Figure 15). When recycled wood is involved as raw material in PB production, this step is also mandatory to remove impurities, sand, dust and other abrasive contaminants (Irle and Barbu 2010).

Figure 15. Logs debarking. Source: https://juwal.eu/en/debarking-machine/

2.1.3 Chipping Process

Depending on the raw material size, first are used primary breakdown machines (Figure 16). The raw material is transformed into almost square chips, 30 to 60 mm in size and 3 to 9 mm in thickness. Average dimensions of wood chips reach 30–50 mm in length, 25–40 mm in width, and 4–6 mm in thickness (Mirski et al. 2019).

Many other sizes of chips could be used, from a chipper canter of a sawmill, or resulted from wood processing, depending on the source of the raw material. The chips are screened to remove under- (<2 mm) and over-sized (>50 mm) particles which are involved in a further chipping phase (Irle and Barbu 2010).

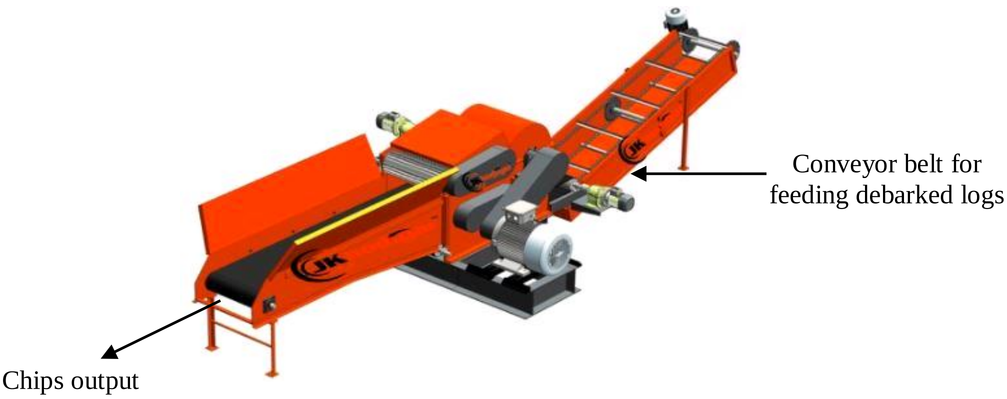


Figure 16. Commercial drum chipper. Source: https://www.indiamart.com/

2.1.4 Reduction of the chips into particles/milling

This additional reduction is accomplished further in different ways, for example, using an hummermill to grind and crush or using a knife system to cut and slice (Figure 17). The chips are transformed into proper particles, 10 to 20 mm long, 3 to 10 mm wide, and 0.4 to 0.7 mm thick. Disc refiners may also be used to produce very fine particles via attrition (Nishimura 2015).

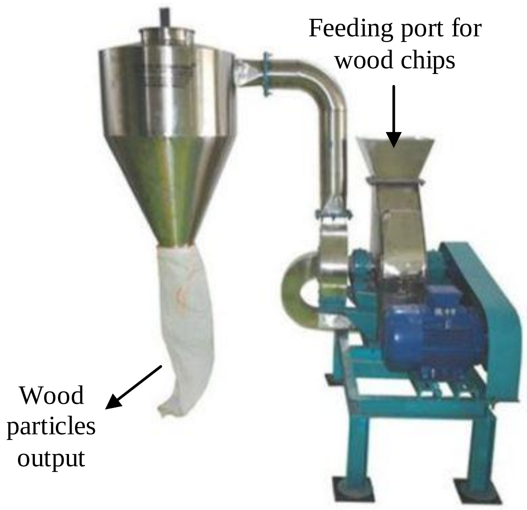


Figure 17. Commercial hummermill to grind and crush the chips into particles. Source: https://www.indiamart.com/

2.1.5 Screening Process

Milled particles include a range of particles sizes from small to large which are classified by size using a mechanical sieves (Figure 18) and air sifter which ensures that the degree of separation is reliably maintained. PB usually has a three layer structure where the surface layer contains small particles to result in a smooth surface for bonding on films and painting, and the core layer is comprised of large particles (Irle and Barbu 2010) .

Figure 18. Classifier. Source: http://acientech.com/

2.1.6 Drying Process

The particles are dried in a rotary drum dryer (Figure 19) which is the main method for drying because the problem of raising dust and explosions are avoided. This phase allows to reduce the moisture content of the particles to between 2% and 8% depending on the adhesive system to be used to make PB. The moisture content must remain within this percentage range because residual moisture is converted to stem during the hot-pressing stage, if too much steam is generated, then, when the press opens, the panel is likely to be delaminated by the sudden release of steam pressure (Irle and Barbu 2010).

Figure 19. Rotary drum dryer. Source: https://www.spraydryersandcoolers.com/Rotary-Dryer.aspx

2.1.7 Blending Particles with resin

The adhesive solution is blended, according to proven recipes, with water, release agents and other additives, *e.g.* hardeners, fire retardants, preservatives, wax and then applied on the dried particles (Figure 20). In particular, the amount of adhesive mix is calculated on the solid adhesive substance to oven dry wood basis. The hardener solution is added to catalyse the resin curing reaction and it is referred as a percentage of solid hardener substance to solid resin basis. The most widely used hardener is ammonium chloride (NH_4Cl).

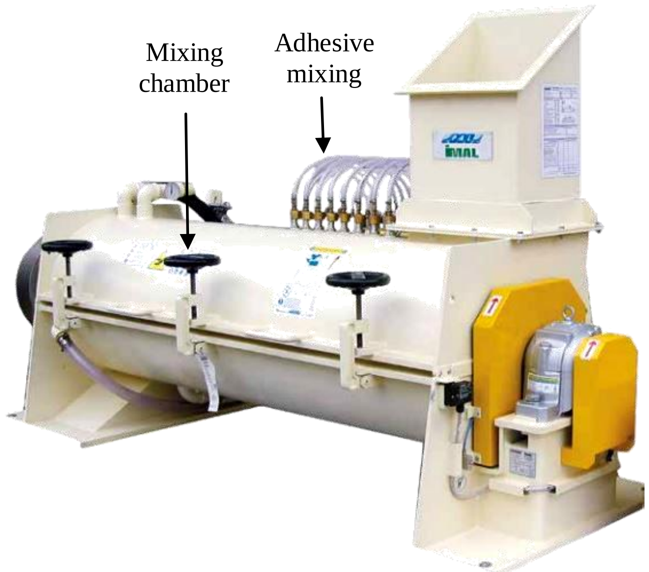


Figure 20. Typical commercial blender. Source: https://www.imalpal.com/

The mixing between adhesive and particles occurs into a rotate drum (Irle and Barbu 2010). Melamine formaldehyde, phenol formaldehyde, UF resins are among the most commonly used adhesives for PBs production as their chemical formulas displayed in Figure 21. The type of resin is one of the major parameters determining the physical and mechanical properties of the final panel (Nishimura 2015).

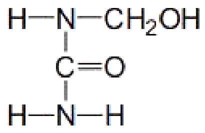
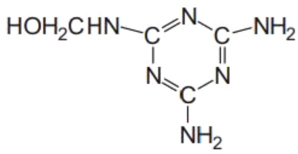
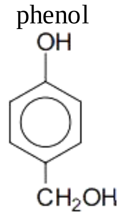
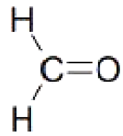
Urea	**Melamine**	**Phenol**	**Formaldehyde**
Mono metylol urea 	Mono methylol melamine 	Mono methylol phenol 	

Figure 21. Common adhesives used in PBs production. Figure adapted from Irle and Barbu 2010.

Despite the fact that UF is the most used adhesive, it has certain disadvantages: it is not moisture resistance, thus panels made with UF resin are not suitable for exterior applications where they could be exposed to moisture. In addition, this resin releases formaldehyde in the air during the production process of the panel causing damage to the manufacturers health due to its toxicity. The UF panels are intended for indoor applications, thus strict regulations should be applied by producers in order to minimize the effect on human health by formaldehyde release indoor. There are regulations which limit the maximum concentration of formaldehyde emission in the air, leading to a reduction in the number of internal uses for panels bonded with this resin (Irle and Barbu 2010). The addition of melamine to urea formaldehyde resin can improve the moisture resistance of UF. Therefore, the melamine urea formaldehyde resin is a clear and it produces panels with very enhanced mechanical properties however it has limitation due to its high cost. Phenol formaldehyde resin is also water resistant being excellent product to be used for outdoor type panels but it is more expensive than that of UF, and it needs high temperature and longer time to cure (Irle and Barbu 2010).

2.1.8 Mat forming Process

The mat or pre-pressed panel should be laid down uniformly across its length and width as depicted in Figure 22. Several forming heads can be used in series, for particles separation and distribution, based on air currents, spinning rollers or a bed of separated rolls. These systems produce a gradation of particle sizes from face to core, resulting in a multi-layered structure. Single and five layer panels may also be formed (Irle and Barbu 2010; Nishimura 2015).

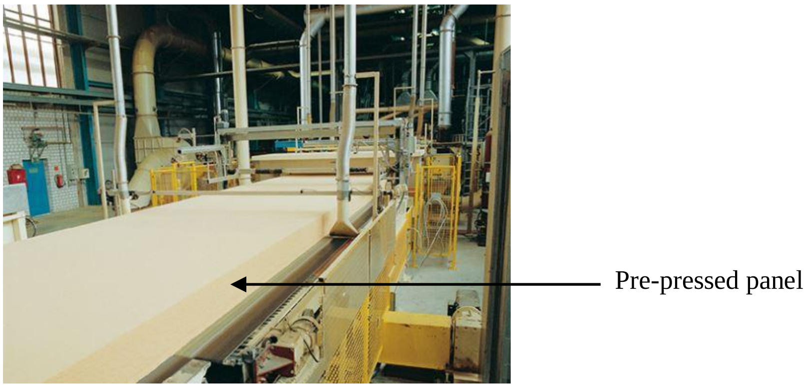


Figure 22. Mat forming stage. Source: https://www.forbo.com/movement/en-jp/industries-applications/raw-materials/wood/wood-board-manufacture

2.1.9 Pre-pressing

Some manufacturers cold press the pre-board to reduce its height and to increase stability preventing the collapse of pre-board during hot pressing (Nishimura 2015) (Figure 23).

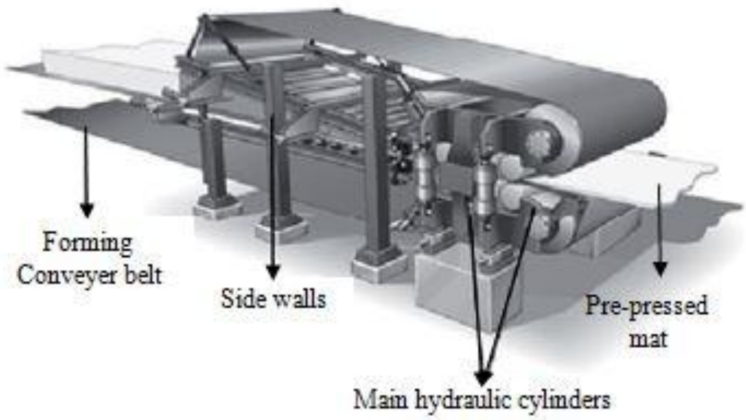


Figure 23. Pre-pressing stage. Source: https://mefexport.com/metso-panelboard (Metso Panelboard).

2.1.10 Hot-pressing

The hot-pressing takes place usually in single or multi-opening presses or continuous presses where panels are pressed between heated plates under specific values of temperature and pressure (Figure 24). There are typically three stages of pressing. The first stage is to press until the required thickness is reached. In the second stage the press is maintained at maximum pressure. During the third stage the pressure is gradually released which eases internal stresses within the panel.

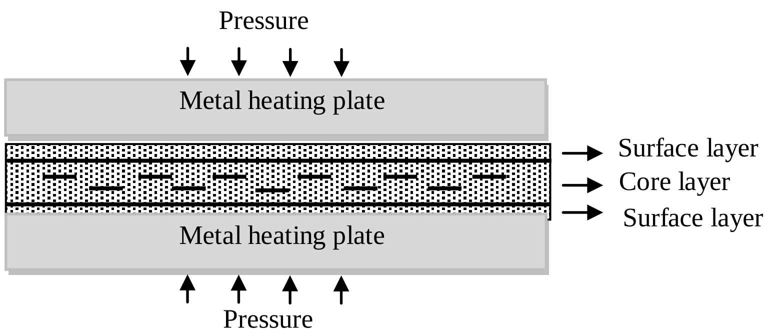


Figure 24. Schematic representation of a three layers panel placed between two metal heating plates during the hot-pressing process.

The temperature used during the pressing stage depends on the type of adhesive. Generally, temperature values between 150 and 160 °C are applied for UF resin, while temperature values between 180 and 200°C are suitable for phenol formaldehyde resin (Irle and Barbu 2010; Nishimura 2015). The specific pressure could range from 2 to 4 MPa depending on final panel density (Irle and Barbu 2010).

2.1.11 The Cooling Process

Manufactured PB is then cooled as shown in Figure 25 after it has been taken out of the press. Then, panels are trimmed to provide straight edges, but also to remove edge effects. Sanding process is the next step following trimming of the panels before they are left to cool again and cure allowing the release of volatile compounds and stabilizing its moisture content. Finally they are packaged and shipped for delivery (Irle and Barbu 2010; Nishimura 2015).

Figure 25. Cooling system. Source: https://sickusablog.com/

2.1.12 Classification of the Finished Panels

The final PB is classified in Europe, based on use, by the EN 312 Standard (2004) which specifies the minimum requirements for uncoated PBs bonded with resin according to the final mechanical and physical characteristics measured. The Table 1 summarize the different uses, classified according to seven grades, from P1 to P7.

Table 1. Classification of PB according to UNI EN 312 (2004).

CLASSIFICATION	PANEL APPLICATION
P1	General panel for use in dry areas
P2	Non-structural panel (including furniture) for use in dry areas
P3	Non-structural panel for use in dry or humid areas
P4	Load-bearing panel for use in dry areas
P5	Load-bearing panel for use in dry or humid areas
P6	Heavy-duty load-bearing panel for use in dry areas
P7	Heavy-duty panel for use in humid areas

The common tests and corresponding standards to evaluate mechanical and physical properties are defined in Table 2.

Table 2. Common test methods to evaluate PBs properties.

PROPERTIES	EN STANDARDS
Modulus of elasticity	EN 310
Modulus of Rupture	EN 310
Tensile strength perpendicular to the plane	EN 319
Swelling in thickness after immersion in water	EN 317
Moisture Content	EN 322
Density	EN 323

All these properties will be explained specifically in the chapters concerning the experimental part of the work.

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CHAPTER 3 LIGNOCELLULOSIC RAW MATERIALS AND ADHESIVES FOR PARTICLEBOARD PRODUCTION

The main sources of raw material for PBs are regenerated forest land or plantation species, however, the constant growth in demand for PBs combined with an increasing consumption of wood led to an increase of wood raw material prices (Dukarska 2013; Górna and Adamowicz 2020). Raw material costs represent a significant proportion of total panel manufacturing costs, amounting for 40–60% (Solt et al. 2019). To overcome this issue, alternative and less expensive lignocellulosic materials considered as a waste, or other unused wood species, could be therefore an alternative raw material sources for the panel industry aiming to find a place in the common market (Grigoriou and Ntalos 2001). In addition, adhesives have been always an important topic of discussion for the panel industry. For PBs manufacture it is desired to use effective adhesives, at a competitive price, with the lowest possible formaldehyde emissions. Due to formaldehyde release, it has been shown that formaldehyde-based adhesives are not environmentally friendly products, therefore solutions should be found to modify or replace them.

3.1 Research for the Appropriate Lignocellulosic Raw Material

Three types of raw material sources are considered: residues from agricultural crops, from fruit trees and finally wood provided by less used wood species. Among the most studied materials, are agricultural residues such as stalks from sunflower (*Helianthus annus* L.) (Bektas 2005; Binici et al. 2013), wheat straw (Han et al. 1998; Mo et al. 2003), castor (*Ricinus communis* L.) (Grigoriou et al. 2001), eggplant (*Solanum melongena* L.) (Guntekin and Karakus 2008), vine (*Vitis vinifera* L.) (Ntalos and Grigoriou 2002), cotton (*Gossypium hirsutum* L.) (Guler and Ozen 2004), stalks from kenaf (*Hibiscus cannabinus* L.) (Kalaicioglu and Nemli 2006), husks from rice (Ciannamea et al. 2010), almond shell (Guerue et al. 2006), sugar cane bagasse-bamboo (Lee et al. 2006), flax, hemp, jute, sisal (Papadopoulou and Chrissafis 2017) and many others. Although agricultural materials are prominent substitutes for traditional type raw materials in the manufacture of panels, there are still obstacles to their wide use.

The main reason is that they are available only seasonally and big storage facilities are required, and also because of wax and silica components (*e.g.* wheat straw) that interfere with adhesion properties (Liu et al. 2004). In addition, their cultivation is limited to certain geographic regions and each of them requires different technology for its successful use in the production of PBs. Some studies have dealt with material provided by fruit trees, such as orchard pruning residues in PBs production. Three-layered composite materials were successfully produced also from plum and apple tree branches at a target density level of 700 kg/m^3 and using UF resin. The composite materials were pressed at 2.5 MPa with temperature of 200 °C. In general, these composites produced with waste biomass from orchard pruning showed physical and mechanical properties values above the minimum requirements by European standards (Kowaluk et al. 2019). In terms of less used species, Tayo et al. (2020) assessed the suitability of Black locust wood from short-rotation plantation in the manufacture of PBs. The target density level was 650 kg/m^3, and the panels were pressed at 20 MPa with temperature of 200°C. The research showed that Black locust as hardwood species could be one of the future alternative for the wood-based panel industries due to the good physical and mechanical properties obtained. Experimental PBs made with cherry and apple pruning have been produced using UF resin with a pressure of 3 MPa and a temperature of 165 °C. Based on the results, it has been shown that it is possible to produce PBs from a mixture of apple and cherry pruning particles without falling below the property value required by the standards (Sahin and Arslan 2013). There is a little knowledge about orange tree pruning, for example, Reixach et al. (2015) tried to use it in form of fibers as reinforcement in polypropylene-based composites in order to investigate their thermal properties. Another research aimed to evaluate the possibility of using orange wood from agricultural conversions to make high-value products and it was concluded that it could be an excellent material for flooring (Berti et al. 2017).

Five Mediterranean evergreen hardwood species (*Quercus coccifera*, *Quercus ilex*, *Arbutus unedo*, *Phillyrea latifolia*, *Erica arborea*) were investigated as raw materials showing that all properties were affected by wood species and panel density. All single-layer PBs made with five evergreen hardwoods had lower mechanical properties but better thickness swelling/water absorption than panels made from common industrial furnish (Barboutis and Philippou 2007).

Although the use of many types of raw materials were investigated for experimental particleboard manufacture, currently there is no or very limited information on the properties of particleboard made from Orange and Turkey oak wood. For this reason the research intent is to produce PBs using wood branches of Orange tree and Turkey oak wood residues, due to the fact that it is an accessible raw material in the Mediterranean area and is less used in industrial applications.

3.2 Sustainable bio-based adhesives for particleboards manufacture

The majority of PBs are manufactured using UF as adhesive. The advantages are related to its fast curing time, good performance in the panel, low cost and much production experience with this resin system (Irle and Barbu 2010). The main disadvantage of using UF resin is its low resistance to water. Lower resistance to water limits the use of WBPs bonded with UF resin to interior application (Park et al. 2006). According to recent sources (Kutnar and Burnard 2014; Grunwald 2017), it was estimated that the adhesive use in the European PB industry was split between UF (90–92%), Melamine-Urea Formaldehyde (6–7%) and Polymeric Methyl Diphenyl Diisocyanate (1–2%) adhesives. It can be observed that formaldehyde substance play an important role in the adhesive production. Asia-Pacific region held a 56% share of the world's total formaldehyde capacity, followed by China which was a competitor leader in terms of its formaldehyde capacity accounting for over 51% of the total capacity. Europe and North America, with shares of 22% and 15.83%, respectively, where moderate growth was observed (Kutnar and Burnard 2014). Formaldehyde-based wood adhesives are derived from petrochemicals, which are non-renewable (Li et al. 2004). In addition, they are considered as dangerous substance leading to serious problems for human health caused by formaldehyde emission (Salem et al. 2011). The mechanism in formaldehyde emission from UF bonded PB is related to unreached free formaldehyde from the binder and hydrolysis of partially and completely cured adhesive (Que et al. 2007). The International Agency for Research on Cancer (IARC,

2004) conducted an evaluation of formaldehyde and concluded that there is sufficient evidence that formaldehyde causes nasopharyngeal cancer in humans.

Environmental consciousness related to the need to use sustainable raw materials and legislations for the same is becoming the main driving factor towards the production of eco-friendly wood composites comprising new bio-based adhesives instead of the traditional formaldehyde-based synthetic adhesives. Wood composite industries tried to control and reduce formaldehyde emission from wood composite panels. Manufacturers are also interested in using non-formaldehyde based adhesive in their product line to avoid such problem (Amini et al. 2013). Studies on soy, lignin, and tannin-based adhesives have been conducted, focusing on the biopolymers' importance (Hemmilä et al. 2017). Some renewable biopolymers, with the relative resource from which they are derived and their industrial uses, are displayed in the Table 3.

Table 3. Important renewable biopolymers. Source: Hemmila et al. (2017).

Resources	Biopolymer Types	Industrial Uses
Trees, herbaceous species, plant waste	Cellulose	Textile, woodworking, composites
Trees, herbaceous species, vegetational biomass, and agro-industrial residues	Lignin	Adhesives and coatings
Corn, potato, cassava, wheat	Starch	Adhesives, food, plastic, rubber and pharmaceutical products
Soya, vegetables, fruits and animals	Protein	Plastic, adhesives and composites
Soya, horticultural crops	Oils and Waxes	Adhesives, resins, coatings

Starch is the second-most abundant polymer after cellulose. Starch is carbohydrate materials that consist of amylase and amylopectin which could be differentiated by its chemical structure.

The linear α-(1 → 4) linked glucan is called amylase while an α-(1 → 4) linked glucan with 4.2–5.9% α-(1 → 6) branch linkages is amylopectin (Robyt 2008). Starch originates from stalks, roots, and seeds of staple crops such as rice, corn, wheat, tapioca, potato and many more and widely available throughout the world commonly used in food industries (Figure 26).

This polysaccharide has shown great potential as a binder for wood adhesives. As an example of a relatively cheap and renewable product from abundant plant materials, it is easy to process and has been extensively used in the form of binders, glues, and pastes, but its bonding capacity is not strong enough to glue wood, nor is its water resistance (Xu et al. 2014).

Figure 26. Common types of starch

It was found that the adhesive prepared by corn starch oxidation significantly improved the water resistance. Besides oxidation, various modifications of starch were evaluated, including esterification, etherification, and crosslinking (Verwimp et al. 2004). The possibility of developing a new wood adhesive by adding a cross-linker named Hexa-methoxy-methyl-melamine to a mixture of UF and corn starch has been already assessed in the past. The results showed that it has excellent mechanical properties comparable to many of the commercially available UF plywood adhesives used for interior applications (Hemmilä et al. 2017). Amini et al. (2013) conducted a study on experimental PB made from rubberwood (*Hevea brasiliensis* Willd.) using modified corn starch as a binder. The corn starch was modified with glutardialdehyde, which is a cross-linker used in wood composite together with glyoxal and dimethylol-dihydroxy-ethylene-urea (Gadhave et al. 2017), to improve the polymerization of this bio-based adhesive.

Based on the findings of this work, MS may have potential as a binder in PB manufacture with acceptable properties. Adhesives from renewable resources appear to have great potential in the future since they would replace petroleum-based products, but other aspects such as water resistance that is durability must be solved.
Although some recent studies have been conducted on the use of MS in the production of composite materials (Sulaiman et al. 2013; Amini et al. 2015; Akinyemi et al. 2019; Kariuki et al. 2019; Owodunni et al. 2020), the use of this adhesive to manufacture experimental PBs produced with pruning residues of Orange and Turkey oak wood residues has not been investigated.

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CHAPTER 4 TREE SPECIES OBJECTIVE OF STUDY

In Italy the area occupied by orange trees is about 81,583 ha (ISTAT 2020), representing together with other important orchards (i.e. apricot and olive trees) the 54% of total permanent crops. According to Camia et al. (2018), the quantity of biomass coming from citrus fruit is about 2.27 Mt. Pari et al. (2018), have estimated that in Italy around 6 million tons (over dry basis) of pruning biomass is available from the main orchards each year, including uprooted biomass. Most of the created biomass is burned in fields or used to produce heat for farm needs. Nowadays, orange wood is also occasionally used in mosaics, tool handles, and marquetry because it is hard to process and then transform it into a value-added product. There are no reliable information on the produced biomass from Turkey oak stand. Despite the lack of this information, there is sufficient evidence that it is a fairly present species. Turkey oak is one of the forest species which occupies an important area in Italy especially on Apennines mountain range system (INFC 2005), so that introduction in more profitable markets could represent an important opportunity to spark mountain economy in the Apennine area (Todaro et al. 2012). In Basilicata Region, (Southern Italy),Turkey oak forests cover a surface of 83,625 ha, accounting for 23% of the entire regional forest area (356,426 ha) (INFC 2005). Less dimensional stability, elevated internal tensions, strong swelling and shrinkage, low durability are limiting factors of this wood species (Giordano 1981), and for this reason it is less appreciated and mainly used for energy purposes (i.e. firewood) or charcoal, fodder for cattle breeding and, since the 1800s, for the production of railway sleepers (Piussi 2015). The botanical characteristics of the tree wood species are described in the following two paragraphs.

4.1 Orange tree (*Citrus sinensis* L.)

Citrus sinensis L., commonly known as Orange tree (Figure 27), is a fruit tree belonging to the Rutaceae Family, whose fruit is orange. It is a plant that varies in size from a small shrub to a tree 4-5 m high with a stem mean diameter of 14.9 cm. The life cycle of these orchards ranges from 16 years for intensive cultivations to more than 40 years for extensive cultivations, but when trees became unproductive the farmers pull them out. The pruning and explants produce a large quantity of biomass. The pruning biomass of orange trees is estimated to be about 1.8 t ha^{-1} $year^{-1}$ (Burg et al. 2017). It has a round crown and branches with delicate and flexible thorns. Its wood is light yellow passing to grayish, with a fine texture and mostly linear fibers (Giordano 1981). The leaves are oval in shape rounded at the base and sharp at the apex, the margin is whole to slightly notched; the petiole has a small wing on both sides. The flowers, isolated or gathered in small racemes at the axial of the leaves, have five greenish sepals and five pure white petals; they give off a pleasant but not strong scent. The fruit, which varies in shape from ovoid to rounded slightly depressed at the poles, is orange or yellow in color; the surface is usually smooth or sometimes slightly wrinkled, the juice is abundant and sweet. The orange tree cultivation is largely presents in America and Asia, country of origin of these species. In Europe the area harvested is 272,287 ha with a production of about 6,451,581 t. The countries in Europe with the largest number of hectares and production are Spain and Italy respectively 139,626 ha and 81,015 ha and 3,639,853 t and 1,522,213 t (FAOSTAT 2018).

Figure 27. Orange Tree. Source: https://pixabay.com/it/photos/natura-albero-arancia

Woody material from orchards has been poorly investigated. Studies about orchards are mainly reflected to the evaluation of the fruit juices, parts of the fruits (peel, seeds) and leaves, but rarely on the evaluation of the physical and chemical properties of the wood (Bruno et al. 2020). A recent study about orange wood investigated its technical characteristics for use as wood flooring, stating that it is a good material to manufacture a high-quality product (Berti et al. 2018). Berti et al. 2018 assessed the main physical properties of orange wood and it was found that density 827 kg/m^3, basic density 666 kg/m^3, volumetric shrinkage 15.56%, longitudinal shrinkage 0.56%, radial shrinkage 5.20%, tangential shrinkage 10.52% and brinnell hardness about to 4.8 kg/mm^3. Giordano (1981) reported a wood density value at 12% of moisture content ranging from 720 to 780 kg/m^3. In general, its wood has high density and shrinkages.

4.2 Turkey oak (*Quercus cerris* L.)

Quercus cerris L., commonly known as Turkey oak (Figure 28), belongs to the Fagaceae Family. It is a large fast-growing deciduous tree species growing to 40m tall with a trunk up to 1.5-2m diameter (Savill 2019), with a well-developed root system (Di Iorio 2007). This wood species can live for around 120-150 years.

The bark is mauve-grey and deeply furrowed with reddish-brown or orange bark fissures. The leaves are dark green above and grey-felted underneath (De Rigo et al. 2016). Size and shape of the leaves are different, but are normally 9-12cm long and 3-5cm wide, with 7-9 pairs of triangular lobes. The leaves turn yellow to gold in late autumn and drop off or persist on the crown until the next spring. The twigs are long and pubescent, grey or olive-green, with lenticels. The buds, which are concentrated on the tip of the twigs, are egg-shaped and hairy and, typically, they are surrounded by long twisted whiskers (Mitchell 1974). The flowers are wind-pollinated ad appear in April and May The fruit is a large acorn stalkless, 2-3.5 cm long and 2cm broad (Johnson 2006).

Figure 28. Turkey oak. Source: www.giardinaggio.it

Giordano (1981) reported that the wood density value at 12% of moisture content ranges from 600 to 1.05 kg/m^3. This forest species has a range distributed from southern Europe to Asia Minor and it is a dominant species in the mixed forests of the Mediterranean basin (Figure 29).

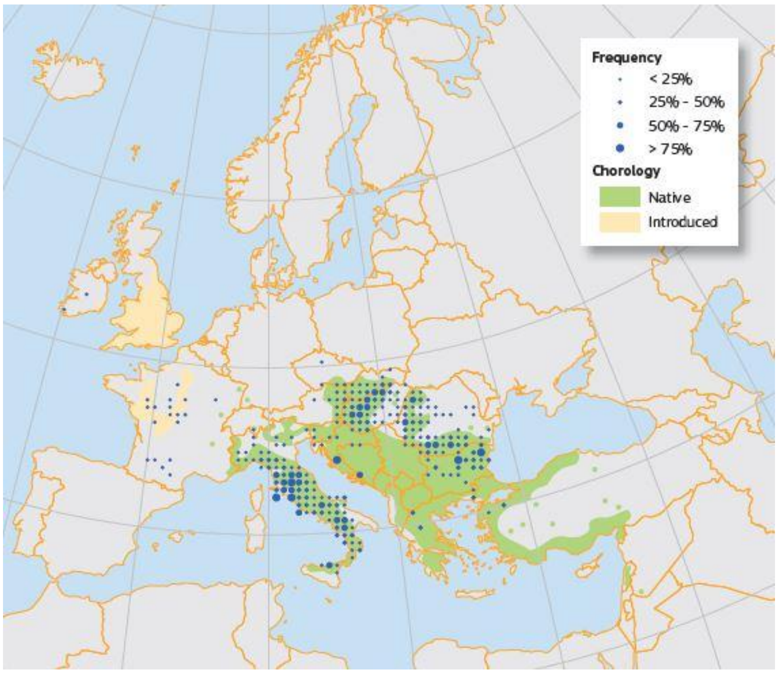

Figure 29. *Quercus Cerris* L. distribution with the frequency occurrences according to the field observations as reported by National Forest Inventories (Caudullo et al. 2017).

It is particularly present in the Balkan and Italian Peninsulas. The western limit of its natural range is France and its northern limit is in Germany, continuing eastward through Austria, Switzerland, eastern Czech Republic, Slovakia and Hungary. In Italy it grows from sea level up to the Appennines and frequently is combined together with Hungarian oak (*Quercus frainetto* L.) (Bozzano and Turok 2003). Turkey oak has been introduced in some other European countries including the UK and France, as well as it is also planted in North America, Ukraine, Argentina and New Zealand (De Rigo et al. 2016). From a qualitative point of view and technological performances (*e.g.* low dimensional stability, prone to crack, different technological properties between heartwood and sapwood, etc.), its wood is poorly appreciated for industrial application but widely used for energetic purposes (Bernetti et al. 1998). The lack of technical references on the technological properties is certainly connected with the fact that most of the attention has been focused on its defects: less dimensional stability, elevated internal tensions, and low durability. Bajraktari et al. (2018) evaluated the chemical composition of Turkey oak wood (Table 4).

Table 4. Chemical composition (% of the total dry mass) and monosaccharide composition (% of total monosaccharides) of the heartwood of Turkey oak wood (Bajraktari et al. 2018)

Characteristics	Values (%)
Ash	0.88 (0.21)
Extractives	1.15 (0.4)
Dichloromethane	1.02 (0.21)
Ethanol	2.12 (0.41)
Water	3.29 (1.06)
Total extractives	**6.43 (1.33)**
Klason lignin	23.23 (1.36)
Soluble lignin	3.05 (0.30)
Total lignin	**26.27 (1.32)**
Monosaccharides	0.66 (0.12)
Ramnose	0.74 (0.08)
Arabinose	1.41 (0.12)
Galactose	1.98 (0.63)
Glucose	62.23 (2.41)
Xilose	30.41 (1.71)
Manose	2.07 (1.55)
Galacturonic acid	1.93 (0.14)
Acetic acid	0.23 (0.01)

Values in Parentheses are Standard deviation.

The chemical composition of the polysaccharides shows that glucose is the major sugar, corresponding to about 60.4% of the total monosaccharides present. The second most important sugar observed was xylose with a value of 31.7% which means that hemicelluloses in Turkey oak heartwood are predominantly xylans with low contents of arabinose and acetyl groups. The monomeric composition of polysaccharides is similar to that found for other oak woods in terms of predominance of glucose followed by xylose. Turkey oak wood shows adequate hardness and density for interior uses like flooring for domestic and commercial applications, with moderate use.

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CHAPTER 5 MECHANICAL AND PYSICAL PROPERTIES OF EXPERIMENTAL PANELS

5.1 Materials and Methods

5.1.1 Biomass material

The woody pruning residues of Orange comes from trees with an age of 12 years located in Bernalda (Matera), southern Italy (Basilicata Region) (Lat.: 40°25'16.0''N; Long.: 16°45'24.0''E), at 127 meters above sea level. Turkey oak wood material was supplied by Meridiana Legnami Company located in Tito Scalo (Potenza). Both types of biomass material were supplied in the form of debarked chips, with dimensions of about 5 cm long and of various thicknesses. Chips were stored in the laboratory for air-drying to a moisture content (MC) of about 12% for 60 days and then reduced into particles using a Multi-functional ensilage crusher with a produced efficiency ≥ 800 Kg/h. The MC of the wood chips was determined in accordance with the Standard EN 322:1993. The resulting particles were then screened with horizontal vibrating sieves (Standard Test Method for Particle-sizes distribution [ASTM] E-11 specification) with opening sizes of 4 mm to remove the oversized and 0.8 mm to remove the dust.

The fraction of particles which passed through the sieve with opening sizes of 4 mm and remained on the sieve with opening sizes of 0.8 mm was chosen for the production of homogenous single-layer PBs. The particles had the length ranging from 3 to 10 mm, width from 0.5 to 1.5 mm, and thickness from 0.3 to 1 mm. Length, thickness and width of randomly selected particles (about 30 particles) were measured by a micrometer. Slenderness ratio (SR) (length/thickness) and flatness ratio (FR) (width/thickness), were also determined (Table 5).

Table 5. Average particles dimensions. Slenderness ratio (SR), Flatness ratio (FR) of wood particles.

Dimensional characteristics	**Average value**
Length, mm	7.05 (2.54)
Width, mm	1.12 (0.31)
Thickness, mm	0.56 (0.24)
Slenderness ratio (SR)	12.58 (7.79)
Flatness ratio (FR)	6.26 (4.38)

Values in Parentheses are Standard deviation.

The values of FR are similar to those found in the literature (Dukarska et al. 2022). The SR values are slightly lower than the values found in the literature, which means that the obtained particles had larger surface area and thus greater resin coverage which led to improved mechanical strength (Barnes 2001; Miyamoto et al. 2002). The particles were stored in a chamber to maintain a constant 10 % MC at 22 % relative humidity (RH) and 21 ± 1°C until they were used. Figure 30 shows the first stages for the biomass material preparation.

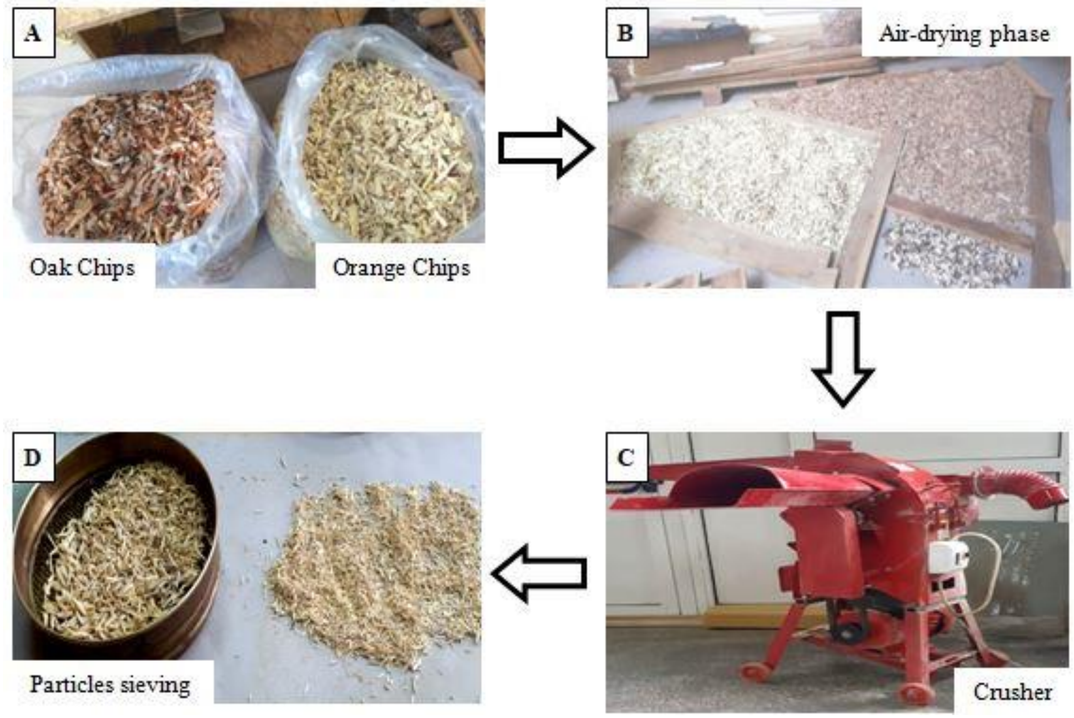

Figure 30. First stages before PBs manufacture. (A) Raw material obtained in the form of chips; (B) air-drying phase; (C) crusher to reduce the chips into particles; (D) particles sieving.

5.1.2 Particleboard Manufacture

The produced panels were divided into three categories based on the wood species used: PBs with 100% Orange wood particles, PBs with 100% Turkey oak wood particles, and PBs with 80%-20% Orange-Turkey oak wood particles. Each of these categories is divided according to two adhesives used: UF at amount of 10% and UF mixed with MS at amount of 2% and 15%, respectively (Table 6). A total of thirty six panels, six for each trial were produced for the experiments.

Table 6. Panel types, proportion of raw material and adhesive amount.

	RAW MATERIAL		ADHESIVE	
PANEL TYPE	ORANGE (%)	TURKEY OAK (%)	UF (%)	MS (%)
O UF	100	_	10	_
TO UF	_	100	10	_
O+TO UF	80	20	10	_
O (UF+MS)	100	_	2	15
TO (UF+MS)	_	100	2	15
O+TO (UF+MS)	80	20	2	15

The percentages of adhesive and the production parameters were decided on the basis of previous studies carried out in this area (Amini et al. 2013; Chotikhun and Hiziroglu 2017). A target density level of 780 Kg/m^3 was assumed.

The nominal dimension of panels was 300mm×400mm×10mm (width × length × thickness). The production parameters used for panels made with UF resin and for panels made with UF+MS resins are listed in the tables 7 and 8.

Table 7. Production parameters of panels made with UF adhesive.

Parameter	Value
Press temperature (°C)	160
Pressing time (min)	5
Pressure (Bar)	60
Dimensions (mm)	300×400
Thickness (mm)	10

Table 8. Production parameters of panels made with UF+MS resins.

Parameter	Value
Press temperature (°C)	160
Pressing time (min)	20
Pressure (Bar)	60
Dimensions (mm)	300×400
Thickness (mm)	10

The pressure value was decided on the basis of the natural density of the wood species used. Furthermore, such a high pressure (60 Bar) was deemed necessary to ensure a good interaction between wood particles and adhesive. Particles were oven dried at 70±3 °C to achieve MC of less than 5%. Based on the oven dry particles weight, 10% UF resin in liquid form (65% solid content) was applied for the panels produced with this commercial resin. The properties of UF adhesive are given in Table 9.

Table 9. Properties of UF resin.

Properties	UF
Solid (%)	65%
Density (g/cm^3)	1.273
pH	7.6
Viscosity (cps)	64
Free formaldehyde (%)	0.15
Gel point (100 °C)	55

As a hardener, ammonium chloride (NH_4Cl, 2% based on the resin weight) in powder form was previously added to the UF solution. Regarding the panels produced with only 2% of UF combined with 15% of modified bio-based adhesive, the procedure was as follows: corn starch was obtained in powder form and the glutaraldehyde solution was purchased from Merck chemical Company. It is a colorless oily liquid organic compound with the formula CH_2 $(CH_2CHO)_2$ widely used as disinfector agent for medical equipment. It has a specific density ranging from 1.06 to 1.12 g/cm^3 at a temperature of 20°C (Lenga 1988). Corn starch in powder form modified with glutaraldehyde in liquid form in a ratio of 1:2 (w/w) was used as binder at amount of 15% based oven dry particle weight.

Initially, corn starch powder was dissolved in distilled water at a temperature of 30°C with stirring, followed by addition of glutaraldehyde solution 25% (Amini et al. 2013; Sulaiman et al. 2013). No wax or other hydrophobic substances were used for the panels. The adhesive and wood particles were blended mechanically for 4 minutes using a hand mixer. The resinated particles were placed in a wooden frame suitable for mat formation and pre-pressed manually with a panel 15 mm thick. The MC of the mat was around 10-12% after adding UF adhesive and around 13-16% by adding MS adhesive. The frame was previously placed on a 3 mm thick steel sheet, covered with heat-resistant paper. After pre-pressing the mat, the frame and the panel were removed and new heat-resistant paper was used for the top of the mat (Balea Paul et al. 2021). The obtained mat was hot pressed using an automatic laboratory press (P 600 LAB 100 TON | Nicem SpA) according to the production parameters listed in tables 7 and 8. The final steps for PBs manufacture are illustrated in Figure 31.

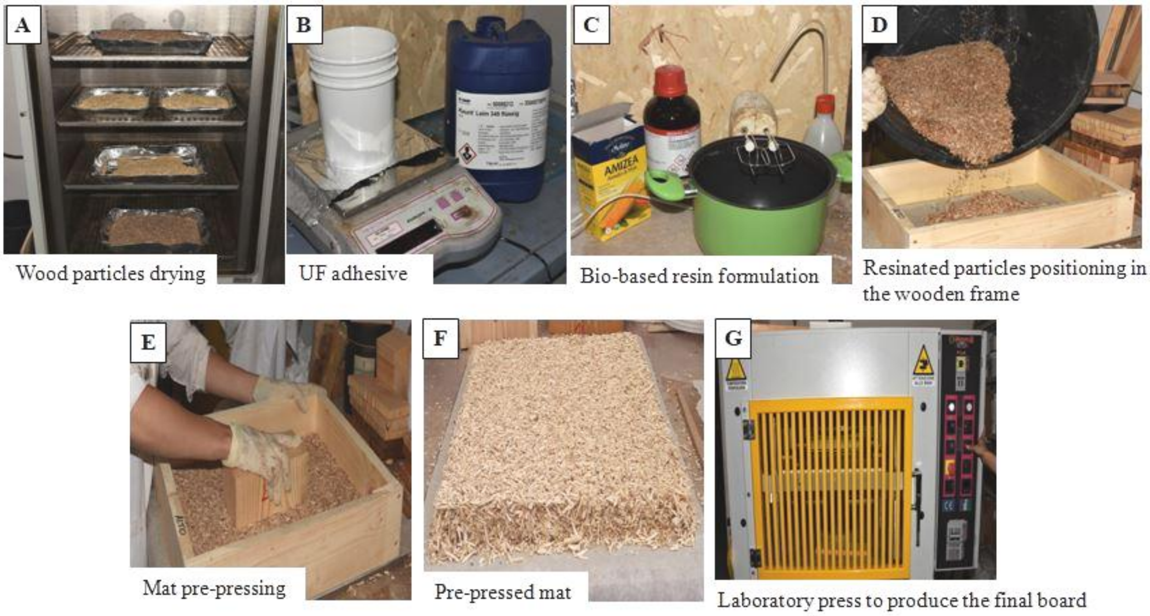

Figure 31. Final steps for PBs manufacture. (A) Wood particles drying in a laboratory oven; (B) UF adhesive; (C) bio-based resin formulation; (D) positioning of the resinated particles in the wooden frame; (E) mat pre-pressing; (F) pre-pressed mat; (G) laboratory press to produce the experimental panels.

After being removed from the press, the produced panels (Figure 32) were trimmed to a final size of 298×398 mm to avoid edge effects and conditioned at a temperature of 20°C and RH of 65% for two weeks.

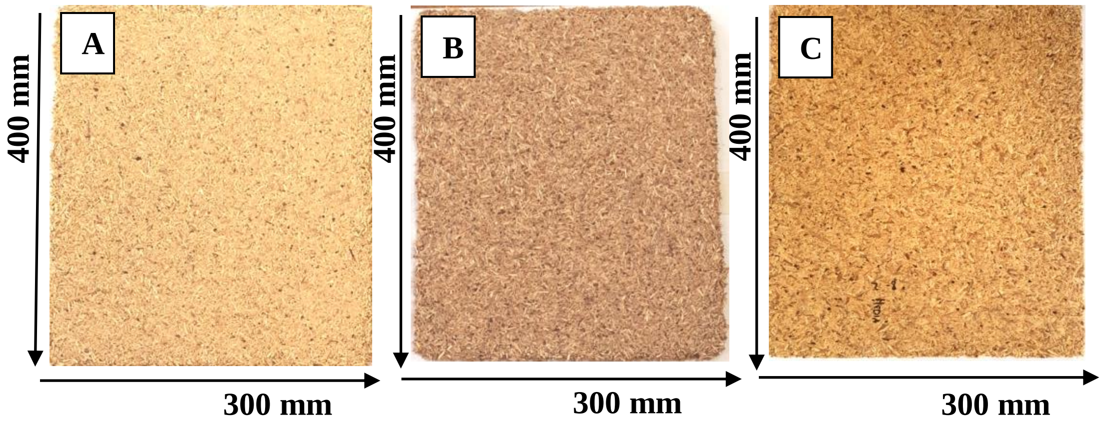

Figure 32. Representation of the panels produced according to the wood species used. (A) Panel with 100% Orange wood particles (O); (B) panel with 100% Turkey oak wood particles (TO); (C) panel with 80%-20% Orange-Turkey oak wood particles (O+TO).

The conditioned panels were cut into various sizes for the evaluation of mechanical and physical properties. The specimens from each panel were then conditioned at a temperature of 20°C and RH of 65% before testing. MC (EN 322:1993) and density (EN 323:1993) of the specimens were determined in parallel to the mechanical and physical properties evaluation.

5.1.3 Evaluation of Properties of the Panels

Mechanical properties, including modulus of elasticity (MOE), modulus of rupture (MOR) (EN 310:1993), and internal bond strength (IB) (EN 319:1993), as well as the physical properties, including thickness swelling (TS) and water absorption (WA) (EN 317:1993) of the samples were evaluated, the size and the number of specimens were determined according to the mentioned standards. Results were compared with limits imposed by standard UNI EN 312:2004 for P2 type panel, intended for non-structural panel, including furniture for use in dry conditions, as well as with limits imposed by the same standard for P3 type panel, intended for non-structural panel for use in humid conditions. Before testing the panels, MC and density were checked as stated in EN 322:1993 and 323:1993 standard.

5.1.3.1 Determination of the Mechanical Properties of the Panels

Both bending and IB tests were carried out on using Universal testing machine model 5567 (INSTRON). Rectangular-shaped specimens (250 × 50 mm) were used for three-point flex measurement of MOE and MOR as shown in Figure 33.

Figure 33. Bending strength test.

This bending test consists in applying a load in the middle of the test piece supported on two fulcrums. Results were expressed according to the Equations (1) and (2), as follows:

$$MOE = \frac{l_1^3(F_2 - F_1)}{4bt^3(a_2 - a_1)} \left[N/_{mm^2} \right] \quad (1)$$

where l_1 is the distance between the centres of the supports, in millimetres; b is the width of the test piece, in millimetres; t is the thickness of the test piece; F_2-F_1 is the increment of load in the straight line portion of the load-deflection curve, in Newton; a_2-a_1 is the increment of deflection at the mid-length of the test piece.

$$MOR = \frac{3F_{max}\, l_1}{2bt^2} \left[N/_{mm^2} \right] \quad (2)$$

where F_{max} is the maximum load, in Newton; l_1 is the distance between the centres of the supports, in millimetres; b is the width of the test piece, in millimetres; t is the thickness of the test piece, in millimetres.

Square-shaped specimens (50 × 50 mm) were then used for IB measurement. The faces of the specimens were glued to the auxiliary wooden devices and conditioned for adhesive curing for 12 hours. Specimens were subjected to an uniformly distributed tensile stress, until breaking (Figure 34).

Figure 34. Internal bond test.

The tensile stress perpendicular to the panel faces (IB) is determined as the ratio between the maximum load and the squared specimen surface, and it was calculated automatically by the software of the equipment according to the following Equation (3):

$$IB = \frac{F_{max}}{\mathrm{a} \times \mathrm{b}} \left[\mathrm{N}/\mathrm{mm}^2 \right] \tag{3}$$

where F_{max} is the maximum load, in Newton; a is the length of the test piece, in millimetres; b is the width of the test piece, in millimetres.

5.1.3.2 Physical Properties of the Panels

Square-shaped specimens (50 × 50 mm) (Figure 35) were used for TS and WA measurements.

Figure 35. Some specimens for thickness swelling and water absorption measurements.

For the evaluation of these physical properties, the same specimens were used. Specimens were fully soaked in distilled water at room temperature (20-22°C) for 2 and 24 hours to determine short- and long-term water resistance properties, respectively. The thickness and weight of the sample were measured before and immediately after soaking. Results, calculated according to the Equations (4) and (5), are reported as percentages values before soaking.

$$TS = \frac{t_2 - t_1}{t_1} \times 100\ [\%] \tag{4}$$

where t_1 is the thickness of the test piece before immersion, in millimetres; t_2 is the thickness of the test piece after immersion, in millimetres.

$$WA = \frac{w_2 - w_1}{w_1} \times 100\ [\%] \tag{5}$$

where w_2 is the weight of the test piece after immersion, in grams; w_1 is the weight of the test piece before immersion, in grams. Properties were evaluated for each produced PB, and statistical data analysis was performed using SPSS v.21 software (IBM Corp.).

The statistical significance among the different treatments was determined with analysis of variance (ANOVA) and Duncan's mean separation tests (Duncan 1955). A 5% difference ($p<0.05$) was considered to be statistically significant.

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CHAPTER 6 RESULTS AND DISCUSSION

6.1 Density of the Panels

The MC values of the panels ranged from 8.70% to 8.55%. The target density level for the manufactured panels was 780 kg/m^3 at a panel thickness of 10 mm. The actual density values resulted to be slightly lower than the intended target density as displayed in Table 10 and shown in Figure 36.

Table 10. Mean values of the actual density for each panel type.

PANEL TYPE	N. of samples	Actual density[I] (Kg m^{-3})	X_{min}[II]	X_{max}[III]
O UF	30	684 ^{c} (0.03)	0.625	0.769
TO UF	30	718 ^{b} (0.04)	0.651	0.850
O+TO UF	30	740 ^{ab} (0.05)	0.600	0.816
O (UF+MS)	42	724 ^{b} (0.06)	0.623	0.822
TO (UF+MS)	42	762 ^{a} (0.05)	0.600	0.869
O+TO (UF+MS)	42	767 ^{a} (0.06)	0.600	0.900

Values having the same letter are not significantly different based on Duncan’s test at the 0.05 significant level.
Values in parentheses are standard deviations.
[I]Mean values of the actual density.
[II]Minimum value.
[III]Maximum value.

Usually, there are differences between the obtained densities and the target ones, mainly due to the characteristics of the wood particles, the pressing parameters as well as lower compaction ratio, even in industrial conditions (Zeleniuc et al. 2020). The low values of the actual density may be due to the swelling of the panels that occurred during the conditioning process. On the other hand, weight loss of particles during the blending process could also affect the low density of the panels (Iswanto et al. 2017)

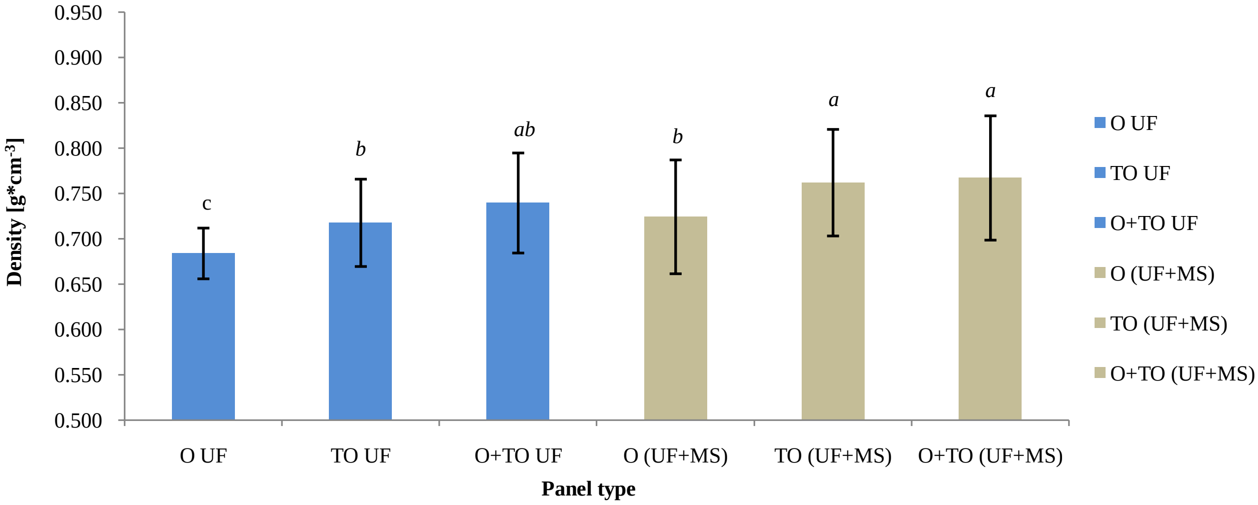

Figure 36. Density values of produced particleboards. O UF-Orange particleboards with Urea-Formaldehyde; TO UF-Turkey oak particleboards with Urea-Formaldehyde; O+TO UF-mixed Orange and Turkey oak particleboards with Urea-Formaldehyde; O (UF+MS)-Orange particleboards with Urea-Formaldehyde and Modified Starch; TO (UF+MS)-Turkey oak particleboards with Urea-Formaldehyde and Modified Starch; O+TO (UF+MS)-mixed Orange and Turkey oak particleboards with Urea-Formaldehyde and Modified Starch. Different letters above bars indicate statistically different mean values ($p<0.05$).

Kelly (1977) reported that higher pressure is required to reach a desired specific gravity for thicker particles and for high density wood species, such as Orange and Turkey oak, whose densities at 12% of MC, are approximately equal to 800 Kg/m^3 and 780 Kg/m^3, respectively (Giordano 1981). The natural variability of such parameter has a significant effect on the density value variability of the panel itself (Kelly 1977).
Kelly (1977) stated that the compaction of the mat is usually performed to an average density higher than the original wood density to obtain a sufficient inter-contact between particles. Taking into consideration the target density, the thickness of panel as well as the higher density of the original wood, the obtained compaction ratio was between 0.83 and 0.95. As a consequence, the average density of the experimental panels was lower than the target density. The lower compaction ratio, the lower density of panel was observed for the panels produced with orange particles. On the other hand, the experimental panels produced with MS adhesive, showed a slight increase in density by about 6% compared to those with UF adhesive, probably caused by the MS solution added to the wood particles at amount of 15%. The increase in their MC value up to 15% resulted in the increase of the compression ratio (Dukarska et al. 2022).

6.2 Mechanical Properties of the Panels

The average values of MOE, MOR and IB of the experimental panels are presented in Table 11.

Table 11. Mean values of the mechanical properties for each panel type.

PANEL TYPE	N. of samples	Bending properties ($N\ mm^{-2}$) MOE	MOR	IB ($N\ mm^{-2}$)*
O UF	30	2028.7 ^{bc} (666.8)	13.8 ^{b} (2.7)	1.79 ^{a} (0.34)
TO UF	30	1790.9 ^{c} (625.7)	16.9 ^{a} (4.5)	1.59 ^{ab} (0.38)
O+TO UF	30	2042.8 ^{bc} (517.4)	15.8 ^{a} (3.4)	1.52 ^{b} (0.45)
O (UF+MS)	42	2380.6 ^{b} (829.4)	6.1 ^{d} (2.6)	0.65 ^{d} (0.10)
TO (UF+MS)	42	3630.1 ^{a} (1051.4)	8.5 ^{c} (2.7)	0.63 ^{d} (0.18)
O+TO (UF+MS)	42	3385.1 ^{a} (1253.2)	9.1 ^{c} (3.5)	0.90 ^{c} (0.24)

*Each Internal Bond (IB) value is an average of 18 measurements.
Values having the same letter are not significantly different based on Duncan's test at the 0.05 significant level.
Values in parentheses are standard deviations.

Statistical analysis found some significant differences ($P<0.05$) between some groups means for MOE, MOR and IB values. MOE is a stiffness parameter, but is also serviceable for the evaluation of the strength parameters, such as MOR, because there is an adequate positive correlation between strength and stiffness (Panshin AJ and Zeeuw 1970). Results of this study exceptionally showed an opposite behaviour in both parameters. Increase in MOE values of UF+MS panels did not mean a substantial increase in MOR values. Due to the prolonged exposure of 20 minutes at high pressure temperature of 160°C, the outer layers of the panels became more stiffer than in the case of pressing UF panels (5 minutes), which led to high MOE but low MOR (Bekhta 2021, personal communication). The presence of the bio-adhesive (UF+MS) resulted in significantly higher values of MOE for panel types TO (UF+MS) and O+TO (UF+MS), which means an increase of about 60% compared to UF panels (Figure 37). High MOE values could be due to the higher density values compared to UF panels (Ashori and Nourbakhsh 2008).

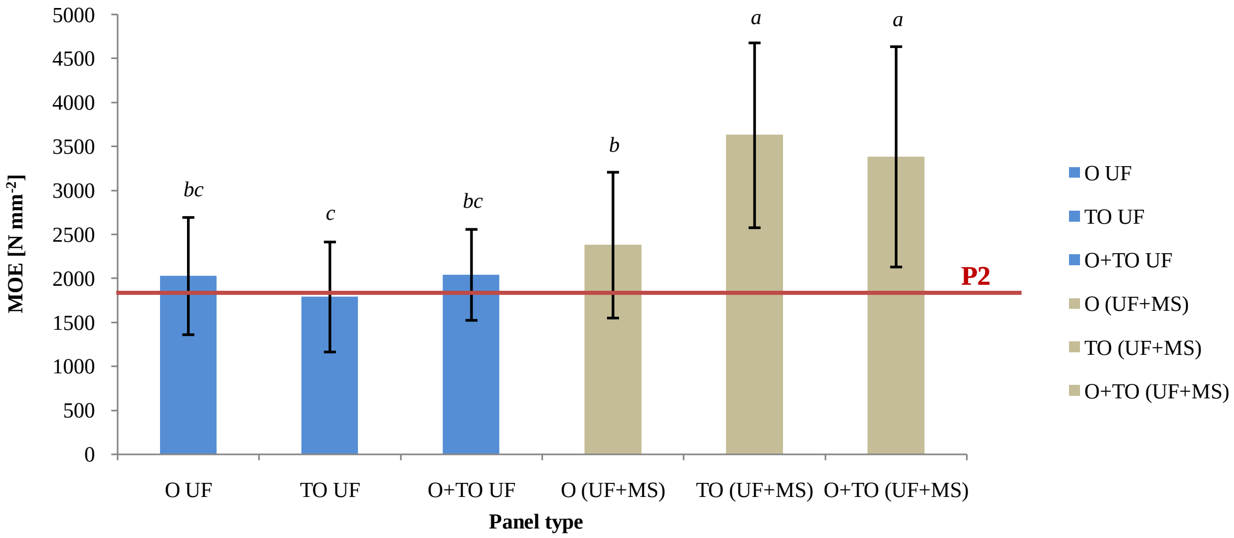

Figure 37. Modulus of elasticity (MOE) values of produced particleboards. O UF-Orange particleboards with Urea-Formaldehyde; TO UF-Turkey oak particleboard with Urea-Formaldehyde; O+TO UF-mixed Orange and Turkey oak particleboards with Urea-Formaldehyde; O (UF+MS)-Orange particleboards with Urea-Formaldehyde and Modified Starch; TO (UF+MS)-Turkey oak particleboards with Urea-Formaldehyde and Modified Starch; O+TO (UF+MS)-mixed Orange and Turkey oak particleboards with Urea-Formaldehyde and Modified Starch. Different letters above bars indicate statistically different mean values ($p<0.05$).

Comparing MOE properties under investigation with other published research, it was found that also Amini et al. (2015) confirmed higher MOE values for panel produced with the mixture of UF and MS compared to those produced with UF alone. Furthermore, Ye et al. (2018) observed that when Dialdehyde Starch dosage exceeded 8%, the rigidity (MOE) of the panels increased, resulting in a decrease of the resistance to deflection (MOR). All the tested panels, except for TO UF panel type, shown to have MOE properties higher than the minimum requirement stated in EN 312:2004, which set up a MOE minimum value of 1800 N/mm^2 for panel type P2, namely non-structural panel including furniture for use in dry areas. In Figure 38 are shown MOR values of the experimental panels. The presence of UF resin resulted in significantly higher MOR values for UF panels compared to the other panels (UF+MS).

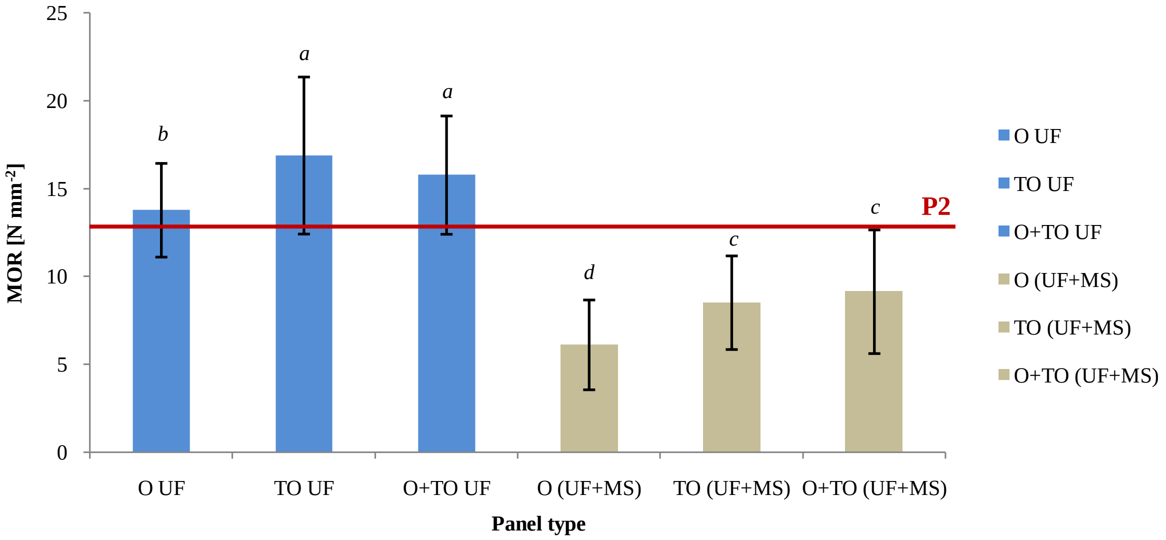

Figure 38. Modulus of rupture (MOR) values of produced particleboards. O UF-Orange particleboards with Urea-Formaldehyde; TO UF-Turkey oak particleboard with Urea-Formaldehyde; O+TO UF-mixed Orange and Turkey oak particleboards with Urea-Formaldehyde; O (UF+MS)-Orange particleboards with Urea-Formaldehyde and Modified Starch; TO (UF+MS)-Turkey oak particleboards with Urea-Formaldehyde and Modified Starch; O+TO (UF+MS)-mixed Orange and Turkey oak particleboards with Urea-Formaldehyde and Modified Starch. Different letters above bars indicate statistically different mean values ($p<0.05$).

It is evident as the better strength properties of UF resin at amount of 10% compared to the bio-adhesive performance, led to an increase of the resistance to deflection of the panel itself (MOR) (Dunky 2017), as occurred in the first three types of panels.

Higher content of UF adhesive creates a stronger link between the particles and increases the bending resistance of the panels as can be seen for UF panels compared to the MS ones where the UF percentage was lower. The shorter pressing time and higher adhesive content in case of UF panels, contributed to rapid rise of temperature into the core of panel due to heat released from UF polycondensation reaction (Shalbafan and Thoemen 2022). MS panels exhibited the lowest MOR of about 49% compared to UF panels. This is probably due to the higher MC of the mat after adding the adhesive solution of MS. Nemli et al. (2007) observed that high MC of the mat could negatively affect MOR values of the final panel. Istek et al. (2019) also reported that high MC of mat (exceeding 15-16%) increased the surface density and decreased the strength values. On the other hand, as observed by Akinyemi et al. (2019), the MS could only develop to its maximum potential when coarse-sized wood fibres (about 1.7 mm thick) are used to produce PBs.

Lower size of particles (0.85 mm) led to a decrease in MOR values at around 10 N/mm^2, and this is in line with what achieved in this research, where smaller size of particles (below 0.6 mm) were used, and lower MOR values were reached (between 9-6 N/mm^2). A possible explanation was probably the slight interaction occurred between the particle size and the MS adhesive during the compression stage, thus leading also to the lower IB values compared to UF panels. Panels produced with UF resin met and exceeded MOR minimum requirements for panel type P2, as stated in EN 312:2004, which imposes a minimum value of 13 N/mm^2. The average IB values for the experimental panels that demonstrate the good interaction between wood particles and adhesives are presented in the Figure 39.

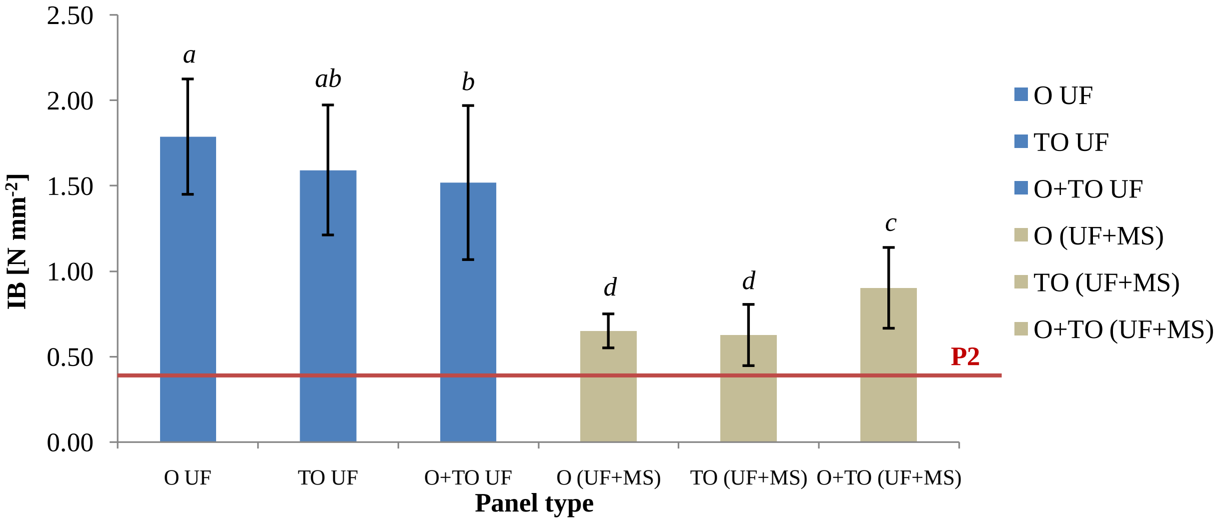

Figure 39. Internal bond (IB) values of produced particleboards. O UF-Orange particleboards with Urea-Formaldehyde; TO UF-Turkey oak particleboard with Urea-Formaldehyde; O+TO UF-mixed Orange and Turkey oak particleboards with Urea-Formaldehyde; O (UF+MS)-Orange particleboards with Urea-Formaldehyde and Modified Starch; TO (UF+MS)-Turkey oak particleboards with Urea-Formaldehyde and Modified Starch; O+TO (UF+MS)-mixed Orange and Turkey oak particleboards with Urea-Formaldehyde and Modified Starch. Different letters above bars indicate statistically different mean values ($p<0.05$).

The highest values ranging between 1.79 and 1.52 N/mm^2 were achieved for UF panels, and the lowest values were achieved for MS panels, ranging between 0.9-0.63 N/mm^2. Similar values of IB were obtained by Amini et al. (2013). Panels made with UF resin alone showed higher IB compared to panels containing the bio-adhesive.

Better interaction between adhesive and wood particles means a more compact panel which led to higher force needed to break the panel perpendicular to its thickness. However, as expected, starch-based adhesives have low reactivity as well as low bonding strength (Hemmilä et al. 2017). The decrease of IB values could be also caused by the increase of the densification duration, when the pressure of 60 Bar was maintained for a longer pressing time for MS panels compared to UF ones. Extension of the first densification step towards the end of the pressing phase negatively influenced the IB, TS, and WA values because of deterioration of the already cured glue bonds in intermediate or inner layers (Shalbafan and Thoemen 2022). Although IB values decreased for MS panels, it is worth mentioning that IB of all panels exceeded the requirements of the EN 312:2004 standard for panel type P2, which imposes a minimum value of 0.40 N/mm^2.

6.3 Physical Properties of the Panels

Concerning the dimensional stability of the panels, all mean values increased as they were subjected to 2 h of immersion into the water, but the difference grew wider after 24 h of immersion (Table 12). UF panels showed significantly lower TS and WA values compared to UF+MS panels.

Table 12. Mean values of the physical properties for each panel type.

PANEL TYPE	Number of samples	TS (%) 2 h	TS (%) 24 h	WA (%) 2 h	WA (%) 24 h
O UF	60	3.78 ^{c} (1.48)	9.98 ^{d} (2.22)	16.68 ^{c} (6.08)	41.86 ^{c} (7.18)
TO UF	60	3.75 ^{c} (1.48)	12.03 ^{d} (2.64)	12.75 ^{d} (5.64)	31.34 ^{d} (6.38)
O+TO UF	60	5.10 ^{c} (1.90)	11.98 ^{d} (2.27)	17.41 ^{c} (6.31)	41.89 ^{c} (6.82)
O (UF+MS)	60	35.46 ^{b} (5.36)	40.57 ^{c} (7.05)	50.65 ^{b} (5.38)	67.43 ^{b} (4.40)
TO (UF+MS)	60	36.22 ^{b} (8.90)	46.59 ^{bc} (10.73)	52.41 ^{b} (5.82)	69.34 ^{b} (5.22)
O+TO (UF+MS)	60	41.84 ^{a} (6.69)	54.97 ^{a} (9.30)	66.58 ^{a} (4.83)	88.00 ^{a} (5.45)

Values having the same letter are not significantly different based on Duncan's test at the 0.05 significant level.
Values in parentheses are standard deviations.

TS value in wood-based panels is determined by swelling of the wood material and springback of the panel after pressing, due to released compression stresses from pressing operation (Shalbafan and Thoemen 2022). Values of TS increased on average from 4% to 37.8% after 2 h of water immersion for UF and UF+MS panels, respectively. After 24 h, UF+MS panels swelled four times more, to about 47.37% compared to UF panels that reached approximately 11.33%. Consequently, the absorbed water was 3.6 greater after 2 h of water immersion and 2 times greater after 24 h of water immersion for UF+MS panels. Similar results were found in the literature (Amini et al. 2013; Akinyemi et al. 2019). Starch adhesives rely on hydrogen bonding forces, which are much weaker then chemical bonds guaranteed by UF resin. They easily form hydrogen bonds with water molecules, leading to poor water resistance (Qiao et al. 2015). In addition, TS and WA values increased with the increasing panel density. UF+MS panels are characterized by higher density values than those of UF panels, as shown in Table 10. This increase probably is due to the release of greater compression stress in high density panels compared with lower density panels (Wong et al. 1998; Xu et al. 2005; Ayrilmis 2007). In addition, the high MC of the mat (between 13 and 16%) could have a negative effect on TS values (Nemli et al. 2007). Moreover, the bonding between the particles decreases, and as a consequence the WA increases if the moisture cannot be sufficiently vaporized, thus preventing the hardening of the adhesive in the core layer of the panel (Nemli et al. 2007). The higher compression ratio of MS panels might cause an increase in TS and WA values (Febrianto et al. 2010). However, in typical commercial panel manufacturer, around 1% wax is used to have better dimensional stability of the products. In this study, no wax was added, as a result the dimensional stability was found to be low. UF panels showed acceptable TS values, in fact they comply with panels maximum property requirement of 14% for 24 h water immersion based on EN 312 type P3 (2004) for non-structural panel for use in humid conditions.

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7. Conclusions and Remarks

This work investigated the suitability of using Orange and Turkey oak wood species in the production of single-layer PBs. In addition two dosages of adhesives were used, namely 10% UF, and 2% UF mixed with 15% MS. All panels produced with UF showed good mechanical performances. Panels produced with the mixture of UF and MS showed good MOE and IB values, complying with the minimum requirements of the standard EN 312:2004 for P2 panels type, namely non-structural panels including furniture for use in dry areas. All panels produced with UF resin met and exceeded MOR minimum requirements for panel type P2. TS values met also the requirement for P3 panels type intended for use in humid condition, according to the standard EN 312:2004, but only in the case of the panels produced with UF adhesive. The use of MS has negatively affected the physical properties (TS and WA) and resistance to bending (MOR). This is probably due to the high MC of the mat after the addition of the MS adhesive and also to the particle size used, which cannot reach the maximum adhesion potential with MS. The increased pressure and long duration of densification during the pressing phase also affected dimensional stability and flexural strength. The increased duration of densification influenced also the dimensional stability and the resistance to bending. However, the produced panels showed good performance for indoor applications, where dimensional stability is not a strict requirement as can be seen in the standard EN 312:2004 for P2 panel type. The results of this research proved that Orange and Turkey oak wood residues are a good alternative as new raw materials for the production of PBs. Developing good properties for bio-based adhesives used in this research was challenging and the obtained results for IB showed that the adhesives based on modified starch could be industrially viable bio-based solutions for PBs manufacture. Furthermore, the idea of making PBs with bio-based adhesive (MS) meant giving further efficiency to the wood raw material, allowing minor wood species to satisfy the future demand for reliable and eco-friendly products. In the future it is necessary to find the optimal factors for a satisfactory panel manufacture, such as the quantity and type of adhesive, as well as all the production parameters involved.

SCIENTIFIC PRODUCTION

Poster and oral presentation to national conference:

❖ XII Congresso Nazionale SISEF, "Underutilised species resources in Italy for particleboards manufacture" - **Valentina Lo Giudice**, Luigi Todaro, Maria Roberta Bruno, Paola Cetera, Octavia Zeleniuc - Palermo, 12-15 November 2019 – Poster

❖ XII Congresso Nazionale SISEF, "Estrattivi da biomassa di Arancio (Citrus Sinensis L.) e Albicocco (Prunus Persicae L.): nuove possibilità di sviluppo sostenibile per le Regioni del Mediterraneo" - Maria Roberta Bruno, Luigi Todaro, **Valentina Lo Giudice**, Paola Cetera - Palermo, 12-15 Novembre 2019 -Poster.

Poster and oral presentation to international conference:

❖ International Conference of Wood Science and Engineering (ICWSE) 2019, "Secondary metabolites of apricot trees (*prunus armeniaca*) pruning: possible sustainable use in the industrial sectors" - Maria Roberta Bruno, Paola Cetera, **Valentina Lo Giudice**, Luigi Todaro, Luigi Milella -Brasov, Romania, 07–09 November 2019 - Oral Presentation.

❖ International Conference of Wood Science and Engineering (ICWSE) 2019, "Underutilized wood species in Italy for particleboards manufacture" - **Valentina Lo Giudice**, Luigi Todaro, Octavia Zeleniuc – Poster

❖ Woodchem 2019, "Biomass from Apricot Tree (*Prunus Armeniaca* L.), Olive Tree (*Olea europaea* L.) and Orange Tree (*Citrus sinensis* L.): new materials for the bioeconomy" - Maria Roberta Bruno, Luigi Todaro, Philippe Gerardin, Stephane Dumarcay, Paola Cetera, **Valentina Lo Giudice**- Nancy, France, 20-22 November 2019.

Online conference:

❖ Society of Wood Science & Technology (SWST) Convention, “Can Woody Biomass from Orchards Still Be Considered a Waste Material?”- Bruno MR, Todaro L, Cetera P, **Lo Giudice V** - Slovenia, 12-15 luglio 2020.

❖ Hardwood Conference Sopron, “Long-term effect on the equilibrium moisture content in thermally modified Quercus cerris L. wood” - Luigi Todaro, Gianluca Ditommaso, **Valentina Lo Giudice**, Roberto Corleto, Milan Gaff- 24-25 June 2021 – Oral Presentation

❖ XIII Congresso Nazionale SISEF, Orvieto (TR), “Characterization of particleboards produced with Orange tree (*Citrus sinensis* L.) and Turkey oak (*Quercus cerris* L.) wood material” – Valentina Lo Giudice, Octavia Zeleniuc, Salim Hiziroglu, Cosimo Marano, Michele Brunetti, Paolo Burato, Giovanni Aminti, Nicola Macchioni, Luigi Todaro. 30 May-2 June 2022.

Paper ISI:

❖ Bruno, M. R., Russo, D., Cetera, P., Faraone, I., **Lo Giudice, V**., Milella, L., Todaro, L., Sinisgalli, C., Fritsch, C., Dumarcay, S., Gérardin, P. (2020). Chemical analysis and antioxidant properties of orange-tree (Citrus sinensis L.) biomass extracts obtained via different extraction techniques. Biofuels, Bioproducts and Biorefining, 14(3), 509-520.

❖ Todaro, L., Liuzzi, S., Pantaleo, A. M., **Lo Giudice, V**., Moretti, N., & Stefanizzi, P. (2021). Thermo-modified native black poplar (Populus nigra L.) wood as an insulation material. iForest-Biogeosciences and Forestry, 14(3), 268.

❖ Mecca, M., Todaro, L., **Lo Giudice, V**., Lovaglio, T., & D’Auria, M. (2021). GC-MS and SPME Techniques Highlighted Contrasting Chemical Behaviour in the Water Extractives of Modified Castanea sativa Mill. and Fagus sylvatica L. Wood. Forests, 12(8), 986.

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❖ **Valentina Lo Giudice**, Paola Cetera, Teresa Lovaglio, Maria Roberta Bruno (2022) "The different levels of valorization of lignocellulosic material obtained from agroforestry activities in a view of circular economy" – Under review

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