Article

Particleboards from Recycled Thermally Modified Wood

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Abstract: In recent years, the production and consumption of thermally modified wood (TMW) has been increasing. Offcuts and other waste generated during TMWs processing into products, as well as already disposed products based on TMWs can be an input recycled raw material for production of particleboards (PBs). In a laboratory, 16 mm thick 3-layer PBs bonded with urea-formaldehyde (UF) resin were produced at 5.8 MPa, 240 °C and 8 s pressing factor. In PBs, the particles from fresh spruce wood and mixed particles from offcuts of pine, beech, and ash TMWs were combined in weight ratios of 100:0, 80:20, 50:50 and 0:100. Thickness swelling (TS) and water absorption (WA) of PBs decreased with increased portion of TMW particles, i.e., TS after 24 h maximally about 72.3% and WA after 24 h maximally about 64%. However, mechanical properties of PBs worsened proportionally with a higher content of recycled TMW—apparently, the modulus of rupture (MOR) up to 55.5% and internal bond (IB) up to 46.2%, while negative effect of TMW particles on the modulus of elasticity (MOE) was milder. Decay resistance of PBs to the brown-rot fungus Serpula lacrymans (Schumacher ex Fries) S.F. Gray increased if they contained TMW particles, maximally about 45%, while the mould resistance of PBs containing TMW particles improved only in the first days of test. In summary, the recycled TMW particles can improve the decay and water resistance of PBs exposed to higher humidity environment. However, worsening of their mechanical properties could appear, as well.

Keywords: particleboards; thermally modified wood; recycled wood; physical and mechanical properties; decay and mould resistance; FTIR

1. Introduction

For the production of particleboards (PBs), nowadays, the trend is to use non-wood lignocellulosic raw materials and especially recycled wood, except of the various traditional forms of fresh natural soft- and hardwood species, as are slabs, edge trimmings, chips, or sawdust. Recycled wood is the subject of much research, especially in terms of the different subtle composition [1]—old wood, e.g., from building constructions, furniture and packaging, and also residual wood from industry. Thermally modified wood (TMW) can be one such residual lignocellulosic material from industrial production.

Wood modification (chemical, thermal, biological) represents an assortment of innovative processes adopted to improve the physical, mechanical, or aesthetic properties of sawn timber, veneer or wood particles used in the production of wood composites. The modified wood should be nontoxic under service conditions. Furthermore, there should be no release of any toxic substances during service, or at the end of life following disposal or recycling the modified wood [2,3]. Products from modified wood do not cause environmental hazards greater than those associated with the disposal of unmodified wood [3]. It means that recycled modified wood is a suitable input raw material for new wooden products, including PBs.

Thermal modification of wood is preferentially aimed at increasing its biological resistance and dimensional stability during climate changes, whereby this wood is characterised with brown-darker colours in its all volume [2,4–7]. TMWs have higher resistance to brown-rot fungi compared to the white-rot fungi, e.g., Sivrikaya et al. [8] showed that...
mass loss of TM spruce wood due to its 8-weeks attack by the brown-rot fungus *Coniophora puteana* was only 1.4%. However, TMW is more brittle having increased susceptibility to cracking, and its strength properties decrease usually from 10% to 20% [2,7,9].

Several reviews on the topic of thermal modification of wood from different perspectives were performed [2,7,10–14], including its thermal pretreatments with the aim to improve properties of produced wooden composites [15].

Thermal modification of wood dates back as far as 1915. Currently, the TMW is industrially produced, and its use is increasing. The global production volume of modified wood in the world is estimated to be 1,608,000 m$^3$ per year, which is dominated by thermal modification to be 1,110,000 m$^3$ per year. Today in Europe are developed commercial processes, e.g., in France (Retification), Finland (ThermoWood), Germany (OHT, Menz Holz), Austria (Thermoholz), Suisse (Intemporis), the Netherlands (Plato Wood), and Estonia (Thermory). It allows the growth of TMW production in Europe, especially when used in exterior cladding, decking and joinery applications—so now it exceeds 695,000 m$^3$ [3,12,16–18]. In Slovakia, there are few small producers, the highest TECHNO-PAL (Banská Bystrica) with annual production of TMW less than 500 m$^3$ per year. Import of TMW from other European countries to Slovakia is app. 1000 m$^3$ per year. Along with increased trend of TMW production in the world, there is and will also be more and more waste of TMW. In the future, post-consumer of TMW as a raw material for further processing and not necessarily for energy purposes, is expected to emerge [4,19]—e.g., for PBs, for which world production is now by FAOSTAT approximately 102 million m$^3$ [20].

Pelaez-Samaniego et al. [15] summarized the use of TMW for production of wood composites. The properties of PBs manufactured with content of TMW are by more authors [19,21–28] as follows: better dimensional stability; reduced water absorption; improved MOE; similar MOR (although some authors showed reduction of MOR); reduced internal bond. Oriented strand boards (OSB) with content of TMW obtained similar changes in properties [29–37]: e.g., better dimensional stability; better resistance to decay; at which wood species and thermal pretreatment conditions differently affected (reduced or improved) their mechanical properties.

The aim of this work was to study the effect of the thermally modified particles (from industrial offcuts of TMWs) on the selected physical, mechanical and biological properties of PBs prepared in a laboratory from TMW particles and particles of freshly cut spruce logs.

### 2. Materials and Methods

#### 2.1. Materials

##### 2.1.1. Wood Particles

The spruce wood particles were made from fresh spruce logs (FSL). Used logs were firstly chipped in the company Kronospan s.r.o. Zvolen, Slovakia, and then the chips milled on particles in the grinding SU1 impact cross mill (TMS, Pardubice, Czech Republic) in the laboratories of the Technical University in Zvolen (TÚZVO), Slovakia (Figure 1). Prepared spruce particles were used for the production of reference particleboards (PBs), and also for the production of PBs containing different proportions of TMW particles.

The TMW particles were prepared from waste offcuts of three TMWs—pine, beech and ash—of constant dimension of 25 mm $\times$ 150 mm $\times$ 800 mm. Offcuts of TMWs were obtained from the timber plant TECHNO-PAL, Banská Bystrica, Slovakia. The industrial production of TMWs themselves was performed by a classic technology, i.e., thermal treatment in air at a temperature of 195 °C for a period of 3 h. The spruce wood and TMWs had the following compression strength parallel with grains (averages determined from 15 samples 20 mm $\times$ 20 mm $\times$ 30 mm at moisture content of 4.7–6.2%): spruce 42.3 MPa; pine-TMW 52.4 MPa; beech-TMW 85.3 MPa; ash-TMW 78.6 MPa—i.e., Average-TMWs 72.1 MPa.

The mixture of TMW particles used for the experiment contained one-third of pine, beech and ash TMWs. In the laboratories of TÚZVO, the TMW offcuts were firstly chipped using the 230H drum mower (Klöckner KG, Hirtscheid—Erbach, Westerwald, Germany),
and subsequently milled on particles using the grinding SU1 impact cross mill (TMS, Pardubice, Czech Republic) (Figure 1).

![Milling of wood chips using the SU1 impact cross mill (TMS, Pardubice, Czech Republic): (a) view of the equipment with grinding segments defining the particle size; (b) grate for central particles; (c) grate for surface particles; (d) a detailed view of the grinding device.](image)

**Figure 1.** Milling of wood chips using the SU1 impact cross mill (TMS, Pardubice, Czech Republic): (a) view of the equipment with grinding segments defining the particle size; (b) grate for central particles; (c) grate for surface particles; (d) a detailed view of the grinding device.

The spruce wood and TMW particles (Figure 2) for the core layer of PBs had dimensions from 0.25 mm to 4.0 mm, and for the surface layers from 0.125 mm to 1.0 mm. The particles for the core layer were dried to a moisture content of 2% and for the surface layers to a moisture content of 4%.

![Particles for particleboards (PBs): (a) The spruce (I) and thermally modified wood (TMW) particles of (II) pine; (III) beech; (IV) ash used in surface layers (SL) and core layer (CL); (b) Size characteristics of the spruce wood and mixture of TMW particles—percentages of individual fraction for surface layers (SL) and core layer (CL).](image)

**Figure 2.** Particles for particleboards (PBs): (a) The spruce (I) and thermally modified wood (TMW) particles of (II) pine; (III) beech; (IV) ash used in surface layers (SL) and core layer (CL); (b) Size characteristics of the spruce wood and mixture of TMW particles—percentages of individual fraction for surface layers (SL) and core layer (CL).

Particles with a dimension of ≤0.25 mm were subjected to FTIR spectra analysis performed on a Nicolet iS10 spectrometer equipped with Smart iTR ATR accessory using diamond crystal. For all particle types, 4 spectral measurements were performed in the range from 4000 cm⁻¹ to 650 cm⁻¹ with a resolution of 4 cm⁻¹. Measured spectra were baseline corrected and analyzed in absorbance mode by OMNIC 8.0 software (Table 1).
### Table 1. Intensity of FTIR spectra (normalized at 898 cm\(^{-1}\)) for particles from the fresh spruce logs (FSL) and thermally modified woods (TMWs)—pine, beech and ash.

| FTIR (cm\(^{-1}\)) | Spruce-FSL | Pine-TMW | Beech-TMW | Ash-TMW | Average-TMWs |
|---------------------|------------|-----------|------------|---------|--------------|
| 1274                | 1.64       | 0.33      | 0.25       | 0.31    | 0.30         |
| 1334                | 0.15       | 0.16      | 0.13       | 0.19    | 0.16         |
| 1372                | 1.11       | 1.10      | 1.35       | 1.31    | 1.25         |
| 1430                | 1.06       | 0.97      | 1.31       | 1.54    | 1.27         |
| 1510                | 2.42       | 2.28      | 1.92       | 3.00    | 2.40         |
| 1600                | 0.75       | 1.09      | 1.91       | 2.31    | 1.77         |
| 1653                | 0.17       | 0.06      | 0.08       | 0.05    | 0.06         |
| 1730                | 1.09       | 1.30      | 2.91       | 2.46    | 2.22         |
| 2900                | 1.60       | 1.61      | 1.46       | 1.64    | 1.57         |

\(TCI = 1372/2900\) 0.69 0.68 0.92 0.80 0.80

\(LOI = 1430/898\) 1.06 0.97 1.31 1.54 1.27

#### 2.1.2. Resin and Additives

The UF resin KRONORES CB 4005 D was added into the surface layers of PBs in amount of 11%, and the UF resin KRONORES CB 1637 D was added into the core layer of PBs in amount of 7%. Molar ratio of UF resins was 1.2 (U:F). Other physical-chemical characteristics of used UF resins were mentioned in the work Iždinský et al. [38]. Required curing of UF resins was achieved by the hardener ammonium nitrate, used as 57 wt.% water solution. Hardener was applied in amount of 2% or 4% to the dry mass of UF resins used for the surface or core particles. Paraffin, used as 35 wt.% water emulsion, was applied on the surface and core particles in amount of 0.6% and 0.7%, respectively.

#### 2.2. Particleboard Preparation

The 3-layer PBs with the dimension of 400 mm × 300 mm × 16 mm and with the density of 650 kg m\(^{-3}\) ± 10 kg m\(^{-3}\) were produced under laboratory conditions. UF resin with the hardener was applied on the conditioned wood particles in laboratory rotary mixing device (TU Zvolen, Slovakia). Wood particles mixed with the adhesive had moisture content from 9.1% to 10.4% (for the surface layers) and from 6.3% to 7.1% (for the core layer). Surface/core particles ratio was 35:65. The particle mats were layered manually in wooden forms. Each particle mat was cold pre-pressed at a laboratory temperature, at a pressure of 1 MPa, and then it was pressed in the CBJ 100-11 laboratory press (TOS, Rakovník, Czech Republic) in accordance with the pressing diagram (Figure 3), i.e., at a maximum temperature of 240 °C, a maximum pressing pressure of 5.75 MPa, and a pressing factor of 8 s/mm. In total, 24 PBs were manufactured, i.e., 6 from each type (Table 2).

### Table 2. Individual types of manufactured particleboards (PBs).

| Variant of PB | Thermally Modified Wood Amount in PB, w/w (%) | Number of Produced Boards | Board Type |
|---------------|-----------------------------------------------|---------------------------|------------|
| PB-C:         | 100% particles of spruce wood                  | 0                         | C          |
| PB-TMW:       | 20%, 50% or 100% particles from mixture of thermally modified woods (TMWs), combined with 80%, 50% or 0% particles of spruce wood | 20, 50, 100 | TMW-20, TMW-50, TMW-100 |
2.3. Physical and Mechanical Properties of PBs

Physical and mechanical properties of PBs were determined by the European (EN) and Slovak (STN) standards: the density by EN 323 [39]; the moisture content by EN 322 [40]; the thickness swelling (TS) and the water absorption (WA) after 2 and 24 h by EN 317 [41] and STN 490164 [42]; the modulus of rupture (MOR) in bending and the modulus of elasticity (MOE) in bending by EN 310 [43]; and the internal bond (IB)—the tensile strength perpendicular to the plane of PB—by EN 319 [44]. Samples were prepared from the 4-weeks air-conditioned PBs (Figure 4). Their mechanical properties were determined with the universal machine TiraTest 2200 (VEB TIW, Rauenstein, Germany).

Classification of PBs was performed in accordance with the European standard EN 312 [45], taking into account the requirements of the PB (type P2) with the thickness ranging between 13 mm and 20 mm.

![Diagram](image)

**Figure 3.** Standard three stage pressing diagram in the manufacturing of particleboards.

![Diagram](image)

**Figure 4.** Scheme of samples preparation from the particleboard (PB) (a), i.e., 1—MOR and MOE by 3-point bending test [43], 2—TS and WA after 2 and 24 h [41,42], 3—IB [44], density [39], and moisture content [40], 4—decay resistance to the fungus *Serpula lacrymans* [46], 5—mould resistance [47], 6—spare samples; Display of PB-samples 50 mm × 50 mm × 16 mm with different amount of recycled TMW w/w (%) (b), i.e., (A) 0%, (B) 20%, (C) 50%, and (D) 100%. 

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*Forests* 2021, 12, x FOR PEER REVIEW 6 of 16
2.4. Decay Resistance of PBs

The decay resistance of PBs to the interiors very dangerous brown-rot fungus \textit{Serpula lacrymans} (Schumacher ex Fries) S.F. Gray / \textit{S. lacrymans} (Wulfen) J. Schröt. — by IndexFungorum/, strain BAM 87 (Bundesanstalt für Materialforshung und -prüfung, Berlin) was tested on samples 50 mm \times 50 mm \times 16 mm. Before the mycological test, performed by ENV 12038 [46]: (a) the edges of samples (50 mm \times 16 mm) were sealed in the amount of 200 ± 10 g \cdot m^{-2} with epoxy resin CHS-Epoxy 1200 mixed with the hardener P11 in weight ratio 11:1 (Stachema, Melnik, Czech Republic); (b) the samples were oven dried and sterilized at rising temperature from 60 °C to 103 ± 2 °C for 10 h, cooled in desiccators, and weighed \(m_0\); (c) all surfaces of samples were repeatedly sterilized twice for 30 min with a UV light radiator; (d) the samples were soaked in distilled water for 240 min to achieve the moisture content of 25% to 30%; (e) the samples were placed into 1 L Kolle-flasks on a top of the stainless steel grids with the fungal mycelium inoculant grown on the agar-malt soil (HiMedia Laboratories Pvt. Ltd., Mumbai, India)— into each flask one sample of PB containing recycled TMW and one sample of reference PB.

Kolle-flasks with samples were incubated for 16 weeks at a temperature of 22 ± 2 °C and a relative humidity of 75% to 80%. After the mycological test, the samples of PBs—after depriving the fungal mycelia from their surfaces—were underwent to a gradual drying process to the constant weight \(m_0/Fungally\_Attacked\), i.e., 100 h at 20 °C, 1 h at 60 °C, 1 h at 80 °C, 8 h at 103 ± 2 °C, and final cooling in desiccators. Their mass losses \(\Delta m\) in percentage were calculated by Equation (1), and their moisture contents \(w\) in percentages by Equation (2).

\[
\Delta m = \frac{m_0 - m_{0/Fungally\_Attacked}}{m_0} \times 100\% \quad (1)
\]

\[
w = \frac{m_{w/Fungally\_Attacked} - m_0/Fungally\_Attacked}{m_0/Fungally\_Attacked} \times 100\% \quad (2)
\]

2.5. Mould Resistance of PBs

The mould resistance of PBs against the mixture of microscopic fungi (moulds) often active in interiors (\textit{Aspergillus versicolor} BAM 8, \textit{Aspergillus niger} BAM 122, \textit{Penicillium purpurogenum} BAM 24, \textit{Stachybotrys chartarum} BAM 32 and \textit{Rhodotorula mucilaginosa} BAM 571) was tested according to a partly modified EN 15457 [47]—it means the samples from PBs had other size, i.e., 50 mm \times 50 mm \times 16 mm, and their sterilization was performed by other method, i.e., twice for 30 min by UV light radiator. Air-conditioned samples were placed into Petri dishes with a diameter of 180 mm (two pieces in one dish) on the Czapek-Dox agar soil (HiMedia Laboratories Pvt. Ltd., Mumbai, India), and then were inoculated with water suspension of mould spores. Incubation lasted 28 days at a temperature of 24 ± 2 °C and relative humidity of 90–95%.

Mould growth activity (MGA) on the top surfaces of PBs was assessed on the 7th, 14th, 21st day (and also on the 28th day—in addition to EN 15457 [47]) after inoculation by a scale from 0 to 4, using the following criteria: 0 = no mould on the surface; 1 = mould up to 10%; 2 = mould up to 30%; 3 = mould up to 50%; 4 = mould more than 50% on the surface.

2.6. Statistical Analyses

The statistical software STATISTICA 12 (StatSoft, Inc., Tulsa, OK, USA) was used to analyze the gathered data. Descriptive statistics deals with basic statistical characteristics of studied properties—arithmetic mean and standard deviation. The simple linear correlation analysis, determining also the coefficient of determination, was used as method of inductive statistic to evaluate the measured data.
3. Results and Discussion

3.1. Physical and Mechanical Properties of PBs

The basic physical and mechanical properties of PBs manufactured in the laboratories of TUZVO are presented in Table 3. The effect of recycled TMWs on the properties of PBs was analyzed by the linear correlations and the coefficients of determination $r^2$ in Figures 5–9.

Table 3. Physical and mechanical properties of the reference/control particleboard (PB-C) and the particleboards (PBs) containing particles from a mixture of recycled thermally modified wood offcuts (PB-TMW).

| Property of PB          | Thermally Modified Wood (TMW) in PB w/w (%) |
|-------------------------|---------------------------------------------|
|                         | 0   | 20  | 50  | 100 |
| Density (kg·m$^{-3}$)   | 654 (15.7) | 659 (23.6) | 657 (21.7) | 653 (23.6) |
| Thickness swelling (TS) after 2 h (%) | 6.01 (0.53) | 3.78 (0.47) | 3.00 (0.27) | 1.97 (0.45) |
| Thickness swelling (TS) after 24 h (%) | 23.73 (1.38) | 15.06 (0.61) | 10.73 (1.12) | 6.58 (0.59) |
| Water absorption (WA) after 2 h (%) | 27.40 (2.04) | 16.34 (0.91) | 12.16 (0.81) | 6.38 (0.42) |
| Internal bond (IB) (MPa) | 68.18 (2.31) | 45.28 (2.04) | 37.32 (1.51) | 24.55 (2.24) |
| Modulus of rupture (MOR) (MPa) | 14.6 (1.56) | 13.1 (1.45) | 10.3 (0.96) | 6.5 (0.74) |
| Modulus of elasticity (MOE) (MPa) | 2611 (285) | 2649 (390) | 2189 (443) | 2225 (253) |

Notes: Mean values of density are from 42 samples, TS from 12 samples, WA from 12 samples, IB from 24 samples, MOR and MOE from 18 samples. Standard deviations are in the parentheses.

Figure 5. Density of PBs containing different amount of thermally modified wood recyclates.

The average density of PBs ranged in a very narrow interval between 653 kg·m$^{-3}$ and 659 kg·m$^{-3}$, i.e., without any apparent influence of the different mutual weight portion of particles from the FSL and recycled TMWs (Table 3). In the linear correlation, it confirmed the $r^2$ value lower than 0.002 (Figure 5). Following it means that the tested moisture, mechanical and biological properties of PBs could not be affected by the density factor.

However, in theory, the searched properties of PBs could be affected by the structure and properties of used wood species. In this study the following analyses were performed: (1) FTIR spectroscopy of wood particles for identification some molecular characteristics of the lignin-polysaccharide matrix in cells walls (Table 1); (2) Compression strength of wood parallel with grains (Section 2.1.1). An attempt to explain the differences in selected properties of PBs by the FTIR and the strength of the types of wood used is presented at the end of the discussion.
Forests 2021, 12, x FOR PEER REVIEW...process of PBs production, when the PBs from thermally modified wood prepared at pressing factor 8 s/mm had the IB 0.43.

Figure 6. Thickness swelling (TS) after 2 h (a) and 24 h (b) of PBs containing different amount of thermally modified wood recyclates.

Figure 7. Water absorption (WA) after 2 h (a) and 24 h (b) of PBs containing different amount of thermally modified wood recyclates.

Figure 8. Modulus of rupture (MOR) (a) and modulus of elasticity (MOE) (b) of PBs containing different amount of thermally modified wood recyclates.
The moisture properties of PBs, i.e., the thickness swelling (TS) and water absorption (WA), significantly improved with higher portion of TMW particles in them. For example, TS-2h decreased from 6.01% for PB-C to 1.97% for PB-TMW-100; TS-24h decreased from 23.73% for PB-C to 6.58% for PB-TMW-100; WA-2h decreased from 27.40% for PB-C to 6.38% for PB-TMW-100; and WA-24h decreased from 68.18% for PB-C to 24.55% for PB-TMW-100 (Table 3). The effect of the recycled TMW particles on the moisture properties of PBs was in all cases considered as significant, as values of the coefficient of determination $r^2$ for the linear correlations “TS or WA = $a + b \times w/w$” were high and ranged between 0.77–0.85, at which $p$ was always 0.000 (Figures 6 and 7). Achieved results related to a better moisture resistance of PBs containing the TMW particles can be explained by their most evident hydrophobic characters compared to particles from fresh spruce wood [12].

Similar results related to moisture properties of solid TMW and PBs based on various TMW types were obtained in more research works, e.g., in Majka et al. [48], Cai et al. [49], Altgen et al. [50]. Ohlmeyer and Lukowsky [24] found out for one layer PBs produced from non-treated and thermally-treated pine wood particles, that the TS-24h was due to presence of TMW reduced from 12.9% to 6.7% and the WA-24h from 59% to 46%. By Borysiuk et al. [4], for PBs based on TMW particles the TS-2h reduced by 20% and the TS-24h by 5%, while the WA-2h reduced by 30% and the WA-24h by 10%. A similar effect of TMW in OSBs on reduction of their TS obtained Paul et al. [29].

Boonstra et al. [25] found out that the WA-24h of one-layer PBs produced from spruce and pine particles pre-heated in two-stages at temperatures below 200 °C was improved only partly. However, some process conditions seem to result in a significant reduction of the WA values, especially at using a higher thermolysis temperature and a longer time of curing process, probably due to a lower sorption capacity of bonded water in the cell walls of wood. By Melo et al. [28], the additional thermal treatments of PBs significantly decreased their WA-24h, the best at using a temperature of 180 °C for six minutes. As for the mechanical properties, the thermal treatments did not significantly influence the PBs strength. Zheng et al. [26] produced PBs from in hot-water pre-treated wood particles, but on the contrary, such PBs had a significantly higher TS and WA, i.e., the TS-24h and WA-24h increased by about 21% and 31%, respectively. Therefore, a hot water pre-treatment of wood particles is not recommended for making of PBs.

Presence of the TMW particles in PBs had a negative effect on their mechanical properties—significantly and very apparently on the modulus of rupture (MOR) and internal bond (IB) with $r^2$ equal to 0.87 and 0.86 for the linear correlations “MOR or IB = $a + b \times w/w$” (Table 3 and Figures 8a and 9). The TMW particles had also a significantly negative
effect on the modulus of elasticity (MOE) of PBs, however, with a lower manifestation when \( r^2 \) of the linear correlation “MOE = a + b \times \frac{w}{w'}” was only 0.19 (Table 3 and Figure 8b).

Compared to the requirements by the standard EN 312 [45], the PBs based on recycled TMWs achieved in all cases the IB 0.35 MPa needed for the type P2.

Generally, results related to lower mechanical properties of PBs containing thermally modified wood are in accordance with work of Ohlmeyer and Lukowsky [24], which determined that the IB equal 0.81 MPa of PBs from non-treated pine wood particles was significantly higher compared to IB equal 0.57 MPa of PBs from thermally treated pine wood particles. Important was also technological process of PBs production, when the PBs from thermally modified wood prepared at pressing factor 8 s/mm had the IB 0.43 MPa, while at 12 mm/s their IB was higher 0.70 MPa. The MOR of PBs prepared from TMW particles was significantly reduced to 14.5 MPa, from 18 MPa of the reference PB.

Borysiuk et al. [4] found out that PBs from TMW particles had a significant decrease of the MOR (about 28%) and the IB (about 22%), while the MOE increased (about 16%). The decrease in the strength properties of MOR and IB was probably related to limitation of surface wettability of the thermal modified particles at the gluing process using UF resins [51]. These authors also state that the increase in the MOE of PBs containing thermally treated wood particles is associated with embrittlement and higher rigidity of these particles, which decrease their deformability in accordance with the knowledge of Hill [2] and also other researchers. In this case, the plasticized lignin stiffens the cell walls in the thermally modified wood particles [6], which resulted in an increase in the stiffness of the entire PB produced from them. A similar effect on the increased MOE of PBs as well as on the shortened lifetime of tools used for machining of PBs could have higher amounts of minerals often presented in recycled woods [52].

3.2. Biological Resistance of PBs

Today, many wood-based composites, including PBs, are produced with no preservative at all. This is partially due to their dominance application in interiors, rather than in outdoor locations with a higher risk of microbial attack. However, for special products from PBs, e.g., used in bathroom or unventilated cellar, it is possible protect them with nano zinc-oxide and other environmentally acceptable biocides [53], using three primary strategies [54,55]: (1) treatment of the wood components prior to PB manufacture; (2) addition of biocides to the adhesives or matrix of the composite; and (3) biocidal posttreatment of PB after its manufacture. The PBs made from recycled TMWs can be considered as more ecofriendly due to the absence of any additional chemicals, at which such thermally modified wood substance has improved also dimensional stability.

TMW displays that the degree of improved decay resistance is positively related to the heating temperature and its duration [56–60]. Unfortunately, the experiences with fungal durability tests for thermally treated composite panel products are still rather limited. Durability tests of the thermally modified strands used for OSB at 200 °C, 220 °C and 240 °C showed improved resistance to the brown-rot fungus Postia placenta compared with untreated ones [30]. Similarly, thermally modified wood fibers showed that wood plastic composites produced on their basis had improved resistance against fungi [61,62]. The study of Barnes et al. [63] assessed thermally modified plywood, OSB, laminated strand lumber, and laminated veneer lumber as a posttreatment, using a closed pressurized treatment method, with reduction in mass loss when subjected to a laboratory soil block durability test.

Biodegradation of PBs with the brown-rot fungus Serpula lacrymans was more suppressed if the boards contained higher number of particles from the recycled TMWs (Table 4, Figure 10). This result is in accordance with work of [64], by which the resistance of PBs to decaying fungi and termites increased with using of higher number of particles from more durable wood species. The reference PB-C had the greatest mass loss (\( \Delta m \) equal 13.20%), while the PB-TMW-100 containing only particles from TMW offcuts had the smallest mass loss (\( \Delta m \) equal 7.26%)—so the \( \Delta m \) decreased by 45%. The coefficient of determination
r² of the linear correlations “Mass loss = a + b × w/w” was 0.67, on the basis of which a less intense decay of PBs containing TMW particles was confirmed with high significance (Figure 10a). Validity of the performed decay test was confirmed by an enough virulence of the fungal strain which at the Pinus sylvestris sapwood samples caused mass loss of 26.80%, i.e., more than the level 20% required for this reference wood species by standard EN 113 [65].

Table 4. Biological resistance of the reference/control particleboard (PB-C) and the particleboards (PBs) containing particles from a mixture of recycled thermally modified wood offcuts (PB-TMW): (I) decay resistance valued on the basis of mass losses (Δm) caused by S. lacrymans; (II) mould resistance against mixture of microscopic fungi valued on the basis of mould growth activity on surfaces of PBs (MGA from 0 to 4).

| Biological Resistance of PB | Thermally Modified Wood (TMW) in PB w/w (%) |
|----------------------------|---------------------------------------------|
|                            | 0   | 20  | 50  | 100 |
| Decay attack by *S. lacrymans* |     |     |     |     |
| Δm (%)                     | 13.20 (0.48) | 8.42 (0.99) | 8.27 (0.72) | 7.26 (0.18) |
| w (%)                      | 87.3 (7.2) | 70.1 (5.3) | 69.4 (5.2) | 68.7 (2.8) |
| Attack by mixture of moulds MGA (0–4) |     |     |     |     |
| 7th day                    | 1.33 | 1.00 | 1.00 | 0.67 |
| 14th day                   | 2.33 | 2.00 | 2.00 | 2.00 |
| 21st day                   | 3.00 | 3.00 | 3.00 | 3.00 |
| 28th day                   | 4.00 | 4.00 | 4.00 | 4.00 |

Notes: Mean value for mass loss of PB caused by *S. lacrymans* is from 6 samples, and for mould growth activity on the top surface of PB is also from 6 samples. Standard deviations are in the parentheses.

Figure 10. Loss of mass (a) and moisture content (b) of PBs containing different amount of thermally modified wood recyclates after their attack by the brown-rot fungus *S. lacrymans*.

The moisture content of PBs exposed to *S. lacrymans* was lower for those ones containing TMW particles in comparison to the reference PB (Table 4, Figure 10b), in accordance with their higher hydrophobicity determined by the water absorption test (Table 3, Figure 7). The thermal modification of wood contributed to the increase in resistance of produced PBs to water, when after decay action of *S. lacrymans* the reference PB noted 87.29% moisture content while the PBs containing 100% particles from recycled TMW had the smallest moisture content of 68.69% (decrease about 21.3%).

Similar increase in the decay resistance of PBs and also other wood-based composites containing TMW was obtained by some other researchers. According to the studies [8,57,66–68], decay resistance of TMW to be related to reduction in maximum moisture capacity of the cell wall due to changed and partly degraded wood polymers, mainly hemicelluloses and due to modification of lignin and creation of new linkages in the polysaccharide-lignin matrix of wood. This phenomenon is similar to our previous study.
when occurred a positive effect of recyclate from faulty PBs on the evidently improved moisture properties and decay resistance to the brown-rot fungus *Coniophora puteana* of newly-prepared PBs [38]. Improved decay resistance of PBs from TMW is in accordance with various types of wood-based composites containing thermally modified components as listed in [30,61,62], as well as wood-based composites exposed to post-heating with reduced mass loss at their following attack by wood decaying fungi [63].

The resistance of PBs to mixture of moulds is summarized in Table 4. The PBs containing 100% TMW recyclates had lower mould rating only on the 7th day of mould test, probably due to reduction the water affinity [8], whereas on the final 28th day after inoculation, 50% and more of their area was covered with the mould hyphae with the sporangium (MGA ranged as 4). The MGA on other PBs containing lower number of particles from recycled TMW and reference PBs had a very similar tendency over time (Table 4).

The differences in the compression strength and FTIR analyses of the used TMWs and spruce wood on properties of prepared PBs can be explained as follows:

- An apparently higher compressive strength parallel with grains of three TMWs (in average 72.1 MPa), compared to less dense spruce wood (42.3 MPa), did not ultimately result in higher MOR of PBs based on TMW particles, but in the opposite effect (Table 3, Figure 8a). It can be attributed to a higher hydrophobicity of TMW particles having a lower polar component of surface free energy [69] with a negative impact on the strength of adhesive joints created between wood elements and polar adhesives [70], including particles in PBs jointed with the polar UF resin.

- FTIR analysis (Table 1) showed that the mixture of three used TMWs, comparing to spruce wood particles, had in accordance with the absorbance peaks an evidently higher portion of lignin determined at 1600 cm\(^{-1}\) (1.77:0.75) and unconjugated \(-\text{C}=\text{O}\) groups determined at 1730 cm\(^{-1}\) (2.22:1.09), but on the contrary, a lower portion of guaiacyl lignin determined at 1274 cm\(^{-1}\) (0.30:1.64) and conjugated \(-\text{C}=\text{C}=\text{O}\) groups determined at 1653 cm\(^{-1}\) (0.06:0.17). Differences in the lignin portion and composition can be attributed: (1) to different wood species (pine/beech/ash TMWs vs. spruce wood) used in the experiment (e.g., generally is known that spruce and other coniferous contain mainly quaiacyl lignin); (2) to creation of new carbonyl and carboxyl groups due to oxidation of wood components in the presence of oxygen at preparation of TMWs; (3) to thermal degradation of hemicelluloses and condensation reactions in TMWs with an indirect increase of lignin and condensed substances similar to natural lignin [71,72].

From the practical point of views, the PBs made with a portion of recycled TMW could be applied for special interior exposures, e.g., kitchens and bathrooms, where sometimes a relatively high humidity of air acts with the aim to protect products made from these boards from moisture and fungal attacks.

4. Conclusions

- The moisture properties of PBs significantly improved with a higher number of hydrophobic TMW particles in their surface and core layers, e.g., after 24 h the thickness swelling (TS) reduced from 23.7% up to 6.58% and the water absorption (WA) from 68.2% up to 24.6%.

- TMW particles had a negative effect on the all mechanical properties of PBs—the most on the modulus of rupture (MOR) in bending with worsening from 14.6 MPa up to 6.5 MPa and the internal bond (IB) with a decrease from 0.78 MPa up to 0.42 MPa, while drop of the modulus of elasticity (MOE) in bending was milder from 2611 MPa maximally to 2189 MPa.

- The biological resistance of PBs to the brown-rot fungus *Serpula lacrymans* significantly increased (maximally about 45%) with a higher portion of more durable TMW particles in their surface and core layers. On the contrary, the presence of TMW particles in PBs had not an evidently positive effect on their mould resistance.
• Generally, adding the recycled TMW particles to produced PBs can be important in terms of increase in their decay and water resistance—required in practice for products exposed to humid environments—but at the same time, worsening of their mechanical properties should be taken into account.

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