Article

Bonding of Selected Hardwoods with PVAc Adhesive

Ján Iždinský ^{1,*} , Ladislav Reinprecht ¹ , Ján Sedliačik ², Jozef Kúdela ³ and Viera Kučerová ⁴

¹ Department of Wood Technology, Faculty of Wood Science and Technology, Technical University in Zvolen, T. G. Masaryka 24, 960 01 Zvolen, Slovakia; reinprecht@tuzvo.sk

² Department of Furniture and Wood Products, Faculty of Wood Science and Technology, Technical University in Zvolen, T. G. Masaryka 24, 960 01 Zvolen, Slovakia; sedliacik@tuzvo.sk

³ Department of Wood Science, Faculty of Wood Science and Technology, Technical University in Zvolen, T. G. Masaryka 24, 960 01 Zvolen, Slovakia; kudela@tuzvo.sk

⁴ Department of Chemistry and Chemical Technology, Faculty of Wood Science and Technology, Technical University in Zvolen, T. G. Masaryka 24, 960 01 Zvolen, Slovakia; viera.kucerova@tuzvo.sk

* Correspondence: jan.izdinsky@tuzvo.sk; Tel.: +421-455-206-385

Abstract: The bonding of wood with assembly adhesives is crucial for manufacturing wood composites, such as solid wood panels, glulam, furniture parts, and sport and musical instruments. This work investigates 13 hardwoods—bangkirai, beech, black locust, bubinga, ipé, iroko, maçaranduba, meranti, oak, palisander, sapelli, wengé and zebrano—and analyzes the impact of their selected structural and physical characteristics (e.g., the density, cold water extract, pH value, roughness, and wettability) on the adhesion strength with the polyvinyl acetate (PVAc) adhesive Multibond SK8. The adhesion strength of the bonded hardwoods, determined by the standard EN 205, ranged in the dry state from 9.5 MPa to 17.2 MPa, from 0.6 MPa to 2.6 MPa in the wet state, and from 8.5 MPa to 19.2 MPa in the reconditioned state. The adhesion strength in the dry state of the bonded hardwoods was not influenced by their cold water extracts, pH values, or roughness parallel with the grain. On the contrary, the adhesion strength was significantly with positive tendency influenced by their higher densities, lower roughness parameters perpendicular to the grain, and lower water contact angles.

Keywords: hardwoods; extractives; pH value; roughness; wettability; PVAc adhesive; adhesion strength

Citation: Iždinský, J.; Reinprecht, L.; Sedliačik, J.; Kúdela, J.; Kučerová, V. Bonding of Selected Hardwoods with PVAc Adhesive. *Appl. Sci.* **2021**, *11*, 67. <https://dx.doi.org/10.3390/app11010067>

Received: 25 November 2020

Accepted: 18 December 2020

Published: 23 December 2020

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1. Introduction

The strength and stability of glued joints are the priority properties of all construction and decorative composites based on metals, wood, glass, plastics, and also other traditional and modern materials. This also applies to glued solid wood products for industrial, building, and transport structures, furniture, musical instruments, sports equipment, and other uses. With regards to the glued wood products, not only is the initial strength of glued joints important, but also the stability of the joints during indoor and mainly outdoor exposures, causing one-off or cyclical changes in wood moisture and temperature.

The most essential parameters influencing the overall bonding quality of wood products include the following: (a) the wood's species, density, chemical and anatomical structure, physical and strength characteristics, surface machining determining the surface roughness, grain orientation, moisture content, and pre-treatment with biocides or other additives, (b) the adhesive's chemical structure, weight solid, viscosity, surface tension, and mechanism of hardening, and (c) the bonding technology's pressure, time, and temperature [1–13].

The low density, high permeability, and high surface roughness of the individual wood species are basic factors that play an important role in terms of better adhesive penetration depth, usually in connection with a positive impact on the bonding quality. However, Aicher et al. [8] found that the adhesion strength of bonded wood is not always most prominently connected with its density. When compared, experiments of several

researchers who tested bonded hardwoods with density (ρ) in a range of 300–1000 kg/m³ showed that the greater values of the adhesion strength were not in all cases found in specimens prepared from denser species (adhesion = $4.095 + 0.014 \rho$ /MPa; R^2 = only 0.25), but almost always in those prepared from species characterized by a higher shear strength (adhesion = $0.628 + 0.912 \tau$ /MPa; R^2 = 0.88). Shida and Hiziroglu [14] examined these tendencies and found out that the adhesion strength of bonded woods was greater in the denser karamatsu species than in the less dense sugi species. Similar tendencies for nine European wood species observed Konnerth [15]. On the contrary, Alamsyah et al. [16] demonstrated a higher adhesion strength in bonded specimens made from the less dense *Paraserianthes falcataria* tropical wood than from the denser *Acacia mangium*.

Water-based adhesives, to which water dispersed adhesives also belong (e.g., polyvinyl acetate (PVAc) adhesives [17]), may reach an adhesion optimum when the water has totally penetrated into the wood substrate [18]. PVAc adhesives are commonly used in the wood industry for general assembly applications, film overlay and high-pressure lamination, edge gluing, wood veneer, and edge bonding [17]. They are safe, non-toxic, non-combustible, easy cleanable, without pollution, cure at room temperature, colorless, transparent and tough after curing, and give a high adhesion strength to bonded wood elements.

Özçifçi and Yapici [19] determined that there was a greater adhesion strength for beech and Scotch pine woods bonded with PVAc adhesive along the tangential direction than the radial one. Burdurlu et al. [20] obtained similar results for Calabrian pine wood bonded with PVAc and polyurethane (PUR) adhesives and recommended performing the bonding process on the tangential surfaces with higher pressures. However, in spite of these results, it is well known that the penetration depth of liquid adhesives within the wood structure is influenced not only by the wood's anatomical direction, density, moisture, and final permeability, but also by the physical and chemical characteristics of the adhesive and the technological bonding conditions, such as pressure, temperature, and time. For example, Sernek et al. [1] found better penetration of the water-based urea-formaldehyde (UF) adhesive into beech wood in the tangential direction at pressure application, while no significant difference between penetrations in the tangential and radial directions occurred when pressure was not applied.

The roughness of wood surfaces depends, first of all, on the wood anatomy and the mode of its machining [19,21–23]. Recognition and quantification of the surface roughness are important from the viewpoint of wood bonding and surface treatment, as the wood surface morphology significantly influences the wood wetting with film-forming materials and the adhesion of these materials to the wood substrate. The circular rotary saw usually causes a higher surface roughness of woods, in comparison with their planning or sanding [22,23]. Shida and Hiziroglu [14] inspected four Japanese wood species—sugi, hinoki, hiba, and karamatsu—and determined that their adhesion strengths with the PVAc adhesive achieved greater values if their surfaces were pre-finished with 80-grit sandpaper, compared to those surfaces pre-finished with finer 120- and 240-grit sizes. Burdurlu et al. [20] found out that a greater roughness of Calabrian pine wood surfaces was caused by their machining in this order, from most to least influential: sawing with a circular rip saw, sanding, and planning. The shear strengths of specimens bonded with the PVAc adhesive were better for sanded or sawed wood surfaces compared with planned ones. Hiziroglu et al. [5,24] also documented the increased roughness of wood surfaces resulting from using sandpapers with lower grit sizes and the better wood surface adhesion with the PVAc adhesive. However, the experiments of Özçifçi and Yapici [19] showed that higher adhesion with various adhesive types had smoother planed wood surfaces than those prepared by band or circular sawing.

The high polarity and good wettability of wood surfaces is given mainly by the presence of hydroxyl, carbonyl, and carboxyl groups in the lignin-polysaccharide matrix of the cell walls. This results in the formation of strong physical bonds with various polar adhesives. Wood surfaces with higher polarities are more wettable with water-based

adhesives [16,25]. The consequence is a higher penetration of the adhesives through the lumens of cell elements on the wood surface. However, the penetration rate can partly be limited by the formation of Van der Waals interactions, dipolar interactions, and hydrogen bonds of polar adhesives with the lignin-polysaccharide matrix of wood cell walls [26].

The wettability of wood, stability of the adhesive systems, and quality of the final adhesion can negatively or positively be influenced by wood extractives and also by preservatives or other excipients added to the wood [27–30]. Polar and nonpolar extractives play a major role in wood bonding processes, as they can contribute to or determine the relevant bonding properties of wood, such as acidity (pH value), wettability (contact angle, surface free energy), or even permeability (clogging of lumens by crystals). Extractives of a high acidity accelerate the curing of acid curing urea-formaldehyde (UF) and melamine-urea-formaldehyde (MUF) resins, decelerate bonding with alkaline hardening phenol-formaldehyde (PF) resins, or degrade PUR adhesives [9,11,31]. Starch and monomeric sugars, which belong to the primary polar water-soluble extractives present in all wood species, have a negative effect on the bonding of wood with cement, MUF, and PF adhesives [11,31]. Secondary extractives specifically occur in various hardwood and softwood species. These extractives, which are typically situated in the heart zones of some European and several tropical hardwood species, contain either various polar polyphenols with a hydrophilic character, such as flavonoids, tannins, sterols, flobafenes, rubrenolide, rubrynnolide, and quinones or coumarins, as well as various nonpolar or semi-polar waxes, fats, and oils with a hydrophobic characteristic [32–35]. Studies of tropical woods bonded together with a PVAc adhesive showed that the extractive content of the wood species had an adverse effect on the bonding quality [36]. In addition, extractives have pronounced inhibitory or supportive effects on the wood sorption capacity, which is reflected in the swelling and shrinkage coefficients associated with moisture changes. The consequence is a stress state at the wood–adhesive–wood interface, impairing the adhesion of the glued joint [37]. Generally, the impact of wood extractives manifests itself more significantly when the bond line is exposed to multiple negative factors. For example, the loss of the adhesion strength in regard to polar extractives is more apparent in the wet state of bonded woods [11].

In the case of wood bonding with adhesives, it is necessary to consider wood's rheological properties that might induce additional retardation of the process of wood surface wetting with the gluing substance on its own. The impact of rheological performance can be, to a considerable extent, mitigated with the aid of mechanical and physical forces applied during adhesive application, during the pressing of the wood at bonding, as well as at exposure of the bonded wood to climatic changes.

The issue of the bonding of European and tropical hardwoods for construction and furniture purposes has been addressed by several researchers, evaluating the factors influencing the shear strength and delamination of glued joints in particular [7,8,38–40]. In summary, they proved that several European and tropical hardwood species meet the requirements for the adhesion strength of glued joints set by the relevant standards and are potentially suitable for glued furniture and construction products.

The aim of this work was to analyze the impact of the selected structural and physical characteristics of hardwoods (e.g., the density, cold water extract, pH value, roughness, and wettability) on their adhesion strength with a water-based PVAc adhesive in dry, wet, and reconditioned states.

2. Materials and Methods

2.1. Hardwoods

2.1.1. Wood Species

The heart zones or central zones of 10 tropical and 3 European wood species were used for the experiment (Figure 1). Their names are usually defined by the standard EN 13556 [41]: bangkirai (*Shorea obtusa* Wall.; Sh. Spp.), European beech (*Fagus sylvatica* L.), black locust (*Robinia pseudoacacia* L.), bubinga (*Guibourtia demeusii* (Harms) J. Léon.), ipé

(*Tabebuia serratifolia* (Vahl) Nicholson), iroko (*Milicia excelsa* (Welw.) C. C. Berg), maçaranduba (*Manilkara bidentata* A. Chev.), dark red meranti (*Shorea curtisii* Dyer ex. King), European oak (*Quercus robur* L.), Santos palisander (*Machaerium scleroxylon* Tul.), sapelli (*Entandrophragma cylindricum* Sprague), wengé (*Millettia laurentii* De Wild.), and zebrano (*Microberlinia brazzavillensis* A. Chev.). The experimental wood material was obtained from the trading company JAF Holz, Ltd. (Špačince, Slovak Republic) in the form of naturally dried boards, having a moisture content of $13\% \pm 2.5\%$. Test samples with dimensions of $80 \text{ mm} \times 20 \text{ mm} \times 5 \text{ mm}$ (longitudinal $\times$ radial $\times$ tangential) were prepared with a circular rip saw (Freud Pro LP30M 026P) having these parameters: a diameter of 255 mm, a cutting thickness of 2.8 mm, a sawblade body thickness of 1.8 mm, a number of teeth of 40, and a maximum rotation speed of 7800 rpm. The wood samples were of a high quality (i.e., without bio-damages, knots, or other inhomogeneities), and before other technological operations, they were conditioned at a temperature of $20 \text{ }^{\circ}\text{C} \pm 2 \text{ }^{\circ}\text{C}$ and a relative air humidity of $50\% \pm 5\%$, achieving an equilibrium moisture content of $8\% \pm 2\%$.

Figure 1. Hardwoods used in the experiment, numbered (from 1 to 13) according to the increasing values of the adhesion strength in the dry state demonstrated in Section 3.2: (1) bangkirai, (10) beech, (4) black locust, (13) bubinga, (7) ipé, (2) iroko, (11) maçaranduba, (3) meranti, (8) oak, (12) palisander, (9) sapelli, (5) wengé, and (6) zebrano.

2.1.2. Characteristics of Hardwoods: Density, Cold Water Extract, pH Value, Roughness, and Wettability

The density ρ of hardwoods was determined in accordance with the standard EN 323 [42].

The cold water extract from hardwoods was obtained in accordance with the standard ASTM D1110 [43], followed by measurement of the pH values of these extracts with a pH meter 7110 (WTW, Wellheim, Germany).

The wettability of the hardwood surfaces was associated with determining the contact angle with a redistilled water drop with a volume of 0.0018 mL up to its complete soaking into the wood substrate, using a goniometer Krüss DSA30 Standard (Krüss, Hamburg, Germany). The course of the water drop profile evolving parallel to the wood grain, from first contact up to the complete soaking, was recorded with a camera, its scanning frequency set in accordance with the wetting interval. The initial contact angle θ_0 was evaluated at the beginning of the wetting process, meaning at the moment of first contact between the water drop and the wood substrate. The drop's contact angle, from the moment of reversion from the acceding contact angle into the receding one, was considered the equilibrium contact angle θ_e . From the values of the contact angles θ_0 and θ_e , the abstract contact angle θ_w , corresponding to an ideal smooth surface, was calculated by the method of Liptáková and Kúdela [44].

Three parameters of wood roughness, the R_a (arithmetic mean deviation), R_z (arithmetic mean of the heights and depressions of the profile at the basic length), and R_{Sm} (mean dis-

tance between the valleys), were inspected on the radial surfaces of samples parallel with and perpendicular to the grain, using the Surfcom 130A surface roughness measuring instrument (Carl Zeiss, Jena, Germany) in accordance with the standard EN ISO 4287 [45]. A total measured length for one replicate was 12.5 mm, and a basic length for one analysis was 2.5 mm.

2.2. Adhesive

A bonding of hardwoods was performed with the water resistant, one-component crosslinking polyvinyl acetate (PVAc) adhesive Multibond SK8 (Franklin International, Columbus, Ohio USA). This adhesive is characterized by the following basic technical parameters: weight solids 48.7–52.3%, pH 2.4–3.5, viscosity approximately 4000 mPa.s, and specific gravity 1.1 g cm⁻³.

2.3. Wood Bonding

For the adhesion strength, each individual specimen was prepared by bonding two samples (80 mm × 20 mm × 5 mm) of the same wood species. The PVAc adhesive was applied on the contacting surfaces of both wood pieces in an amount of 120 g ± 10 g per square meter. The bonding process was performed in the press for 60 min at a pressure of 1.2 MPa and a temperature of 20 °C ± 2 °C. The conditioning of the bonded hardwood specimens lasted 7 days at a temperature of 20 °C ± 2 °C and a relative air humidity of 50% ± 5%.

2.4. Adhesion Strength: Tensile Shear Strength

The adhesion strength test—the tensile shear strength of lap joints—of bonded hardwoods was performed by the standard EN 205 [46] in the dry state, wet state, and reconditioned state and evaluated in accordance with the criteria of the standard EN 204 [47] for water-resistant adhesives belonging to the D3 class (Figure 2).

Figure 2. Adhesion test for bonded hardwood specimens according to the standard EN 205 [46]. Dimensions are in mm.

2.5. Statistical Analyses

The statistical software STATISTICA 12 was used to analyze the gathered data. Descriptive statistics deals with the basic statistical characteristics of studied properties: arithmetic mean and standard deviation. Differences of the adhesion strength in the dry state of the bonded hardwoods were analyzed by the Duncan test. The simple linear correlation analyses together with the coefficient of determination R^2 and the significance level parameter p were used as method of inductive statistics to evaluate the measured data.

3. Results and Discussion

3.1. Density, Cold Water Extract, pH Value, Roughness, and Wettability of Hardwoods

The selected structural and physical characteristics of 13 hardwoods, theoretically important from the point of view of their bonding with adhesives, are present in Tables 1–3.

The density ρ of 13 hardwood species, determined at a moisture content of $8\% \pm 2\%$, ranged from 636 kg/m^3 for meranti to 1105 kg/m^3 for maçaranduba (Table 1). The cold water extract of the hardwoods, obtained from their sawdust by the standard ASTM D1110 [43], ranged from lower values of 0.9–1.6% for zebrano, wengé, meranti, and sapelli to higher values of 4.2–4.6% for palisander, oak, and black locust (Table 1). The pH of individual hardwood species ranged from a neutral acidic value of 5.8 for palisander and beech to more acidic values in the scope of 3.4–3.9 for meranti, bangkirai, oak, and ipé (Table 1).

The type and amount of extractives in wood varies from species to species, and for the same wood species this can also be influenced by the geographical origin, climate conditions, tree age, and part of the tree from which a sample originates [48]. Several studies have been carried out on the extractives of tropical woods. For example, Wanschura et al. [49] described the benefits of extractives present in tropical woods for their surface treatments. Kilic and Niemz [50], in the structures of 12 tropical wood species, found very low amounts of lipophilics ($0.05\text{--}0.38 \text{ mg/g}$); the constituent consisted mainly of fatty acids, while the hydrophilics were composed of phenolic acids, flavonoids, sterols, stilbenes, and a lignan. Jankowska et al. [30] determined that there were large differences in quantity of the hot water soluble extractives in the European and tropical hardwood species, which have been researched by us as well (e.g., 2.92% in light red meranti, 4.24% in beech, 4.33% in wengé, 6.07% in sapelli, 6.47% in iroko, 11.21% in oak, and 12.63% in ipé). The values of the cold water extracts of the same species determined in our experiment were evidently lower, from 1.53% in wengé to 4.33% in oak (Table 1). This difference was probably caused by the fact that hot water dissolves not only polar extractives, which also easily dissolve in cold water (e.g., tannins, gums, sugars, and coloring matter), but also starches. The value of the cold water extract for meranti wood was comparable with that found by Yamamoto and Hong [51]; however, those for the wengé and zebrano woods were smaller compared with the values obtained by [50].

Similar pH values for some of the same tropical hardwood species (Table 1) were reported by Yamamoto and Hong [51], Torelli and Čufar [52], and Ikenyiri et al. [53].

Table 1. Densities, cold water extracts and pH values of the hardwoods.

Wood Species	Scientific Name	Density (kg/m^3)		Cold Water Extract (%)	pH
		EN 350 [54]	Obtain		
Bangkirai	<i>Shorea obtusa</i>	700-930-1150	834	2.70	3.82
Beech	<i>Fagus sylvatica</i>	690-710-750	705	2.13	5.79
Black locust	<i>Robinia</i>	720-740-800	726	4.56	4.65
	<i>pseudoacacia</i>				
Bubinga	<i>Guibourtia demeusii</i>	700-830-910	887	3.15	4.12
Ipé	<i>Tabebuia serratifolia</i>	900-1050-1150 ¹	957	2.08	3.94
Iroko	<i>Milicia excels</i>	630-650-670	641	3.51	5.60
Maçaranduba	<i>Manilkara bidentate</i>	1000-1100-1150 ¹	1105	3.66	4.60
	<i>Shorea curtisii</i>				
Meranti	<i>Shorea curtisii</i>	600-680-730	636	1.55	3.38
Oak	<i>Quercus robur</i>	670-710-760	779	4.33	3.83
Palisander	<i>Machaerium scleroxylon</i>	700-900-1000 ¹	818	4.21	5.84
	<i>Entandrophragma cylindricum</i>				
Sapelli	<i>Entandrophragma cylindricum</i>	640-650-700	693	1.62	5.23
Wengé	<i>Millettia laurentii</i>	780-830-900	881	1.53	4.32
Zebrano	<i>Microberlinia brazzavillensis</i>	700-770-850 ¹	777	0.91	5.62

Notes: Mean values of density are from six measurements, and the cold water extract and pH value are from three measurements. ¹ By Wagenführ [55].

The roughness parameters Ra and Rz determined for 13 hardwoods exhibited, on average, 48.2% and 79.1% higher values, respectively, measured perpendicularly to the grains (on average: $Ra = 10.4 \mu\text{m}$, $Rz = 85.6 \mu\text{m}$) than those determined parallel with the grains (on average: $Ra = 7.0 \mu\text{m}$, $Rz = 47.8 \mu\text{m}$). Generally, these represented differences from 8% to 114%, according to the relevant wood species (Table 2). However, the parameter R_{Sm} was approximately comparable in both measured directions of the wood (Table 2).

Table 2. Roughness parameters Ra , Rz , and R_{Sm} of the hardwood surfaces.

Wood Species	Roughness Parallel with Grain (μm)			Roughness Perpendicular to Grain (μm)		
	Ra	Rz	R_{Sm}	Ra	Rz	R_{Sm}
Bangkirai	10.4 (3.0)	68.4 (18.5)	698.4 (193.6)	17.1 (3.6)	131.9 (19.7)	560.5 (89.5)
Beech	6.0 (1.8)	41.2 (12.6)	673.9 (142.9)	8.2 (1.9)	63.7 (12.8)	408.5 (56.7)
Black locust	5.8 (2.7)	40.5 (19.3)	572.4 (121.8)	9.6 (2.7)	86.7 (20.7)	565.5 (149.7)
Bubinga	6.5 (3.6)	44.9 (22.8)	591.4 (150.1)	7.0 (3.1)	69.3 (22.2)	688.5 (233.6)
Ipé	7.9 (4.6)	51.5 (25.8)	737.1 (197.5)	9.3 (3.8)	77.2 (20.5)	572.4 (105.9)
Iroko	7.1 (1.8)	51.6 (13.7)	580.5 (134.8)	10.0 (3.2)	82.5 (24.7)	661.1 (256.5)
Maçaranduba	3.6 (1.5)	26.7 (8.7)	555.1 (104.7)	4.2 (0.9)	46.8 (13.5)	643.9 (213.3)
Meranti	9.9 (5.1)	60.8 (27.3)	655.0 (179.8)	17.6 (4.6)	126.6 (21.3)	760.2 (197.7)
Oak	9.4 (6.4)	59.5 (35.2)	574.5 (195.2)	16.8 (5.1)	123.4 (33.7)	534.5 (119.5)
Palisander	4.7 (1.3)	34.3 (10.1)	679.2 (239.1)	5.9 (2.1)	57.6 (14.7)	555.7 (138.8)
Sapelli	7.3 (2.8)	52.8 (19.4)	565.1 (109.0)	10.2 (2.3)	87.6 (11.1)	533.8 (93.0)
Wengé	5.4 (3.3)	39.2 (20.0)	545.6 (145.7)	9.8 (2.7)	78.5 (17.8)	712.1 (174.5)
Zebrano	7.1 (3.2)	50.3 (22.4)	588.5 (159.5)	9.5 (3.3)	81.6 (21.7)	622.5 (195.1)

Note: Mean values are determined from 90 values (15 different measuring spots on 6 replicates). Standard deviations are in parentheses.

The surfaces of the densest maçaranduba wood were characterized by the lowest roughness parameters Ra and Rz . On the contrary, the highest roughness parameters were exhibited by the least dense wood, the meranti wood, but at the same time by the bangkirai and oak woods also, which had a medium density (Tables 1 and 2). In these circumstances, an unexpectedly higher surface roughness of the bangkirai and oak, belonging to the ring-porous wood species [55], can be explained by their naturally more porous morphological structure.

The water's potential for surface wetting of the tested hardwood species was assessed based on the water contact angles θ_0 , θ_e , and θ_w (Table 3). Iroko had the highest initial and abstract contact angles ($\theta_0 = 112.5^\circ$, $\theta_w = 78.5^\circ$), and black locust had the lowest equilibrium and abstract contact angles ($\theta_e = 19.7^\circ$, $\theta_w = 25.4^\circ$), while bubinga had the lowest initial contact angle ($\theta_0 = 54.8^\circ$) (Table 3).

According to Liptáková et al. [56], the values of the contact angles θ_0 and θ_e depend on the wood surface roughness and chemistry. These authors supposed that the calculated contact angle value θ_w , valid for an ideal smooth surface, depends exclusively on the wood chemistry. Consequently, the different values of the contact angle θ_w in Table 3 indicate species-related differences in the wood surface chemistry. This concerns not only the different types and contents of extractives, but also the chemical composition of the lignin and polysaccharides in tested hardwoods.

The wettability of wood is a substantial parameter which gives basic information on the interaction between the solid wood surface and liquids, such as adhesives and paints (e.g., how easily and efficiently the liquids spread over a solid surface) [57,58]. The smaller water contact angles, which usually result from the rougher and more polar characteristics of wood surfaces, indicate deeper penetration of the water-based adhesive into the wood structure [16,59].

When comparing the roughness parameters and the wettability parameters of 13 hardwoods (Tables 2 and 3), it is evident that there was not always a more apparent connection between these two groups of parameters. For example, the maçaranduba and meranti

woods, characterized by totally different values for the density (Table 1) and the parameters of roughness (Table 2), had comparable values for the water contact angles (Table 3).

Table 3. Wettability of hardwood surfaces, characterized by the contact angles.

Wood Species	Contact Angle (°)		
	θ_0	θ_e	θ_w
Bangkirai	87.6 (20.7)	46.1 (14.2)	58.6 (22.3)
Beech	71.8 (9.3)	27.6 (11.6)	32.4 (12.9)
Black locust	80.7 (14.8)	19.7 (10.7)	25.4 (13.8)
Bubinga	54.8 (7.5)	30.5 (17.6)	32.2 (18.7)
Ipé	101.3 (7.5)	46.2 (16.4)	61.8 (21.0)
Iroko	112.5 (10.3)	54.0 (10.7)	78.5 (15.7)
Maçaranduba	94.0 (5.7)	23.5 (10.4)	31.5 (14.5)
Meranti	92.0 (15.1)	32.5 (23.7)	41.1 (25.0)
Oak	80.5 (12.2)	33.6 (14.6)	41.0 (16.7)
Palisander	88.4 (12.7)	29.9 (9.3)	40.0 (14.3)
Sapelli	111.7 (14.0)	32.0 (16.5)	52.2 (26.5)
Wengé	86.8 (8.4)	59.1 (7.6)	68.3 (7.9)
Zebrano	71.4 (9.9)	45.0 (13.0)	50.3 (14.2)

Notes: Mean values are from 30 measurements (5 different measuring spots on 6 replicates). Standard deviations are in parentheses.

3.2. Adhesion of Hardwoods with PVAc Adhesive

The values of the adhesion strength (the tensile shear strength of the lap joints) of hardwood specimens bonded with the PVAc Multibond SK8 adhesive are shown in Table 4 and Figure 3.

Table 4. The adhesion strength (tensile shear strength of lap joints) of hardwood specimens bonded with the PVAc Multibond SK8 adhesive, determined in the dry state, wet state, and reconditioned state by the standard EN 204 [47].

Wood Species	Adhesion-Tensile Shear Strength of Lap Joints (MPa)		
	Dry	Wet	Reconditioned
Bangkirai	9.53 (1.68) -	2.29 (0.54)	11.24 (1.58)
Beech	15.65 (2.73) a	2.64 (0.20)	16.19 (2.56)
Black locust	11.69 (2.65) c	1.58 (0.24)	12.62 (2.04)
Bubinga	17.20 (1.09) a	2.00 (0.17)	19.24 (1.62)
Ipé	13.99 (0.51) a	1.27 (0.08)	12.10 (1.41)
Iroko	10.69 (2.14) d	2.01 (0.10)	10.26 (1.30)
Maçaranduba	15.76 (2.23) a	0.56 (0.19)	8.48 (0.78)
Meranti	11.40 (1.54) d	2.34 (0.24)	10.85 (1.73)
Oak	14.27 (1.47) a	2.33 (0.24)	15.57 (1.97)
Palisander	15.86 (1.18) a	1.26 (0.18)	13.67 (2.86)
Sapelli	15.13 (1.85) a	1.98 (0.29)	14.90 (2.36)
Wengé	11.90 (2.02) c	1.57 (0.22)	13.53 (0.51)
Zebrano	13.85 (2.52) a	1.12 (0.30)	9.80 (2.12)

Note: Mean values are from six values. Standard deviations are in parentheses. The Duncan test, using the indexes (a, b, c, and d), identified the significance level of the higher adhesion strength of bonded wood species in relation to the reference bonded bangkirai wood having the lowest adhesion strength in the dry state (e.g., a: very significantly higher, >99.9%; b: significantly higher, >99%; c: less significantly higher, >95%; and d: insignificantly higher, <95%).

Figure 3. The growth tendency of the adhesion strength from bangkirai (No. 1) to bubinga (No. 13) determined in the dry state of bonded hardwoods (a) unequally changed for the individual wood species only in the wet state (b), based on the evidently decreased parameter R^2 and reduced significance p (from 99.9% to 99%) of the linear correlation $\text{Adhesion} = a + b \times \text{Number of Hardwood}$, while this tendency returned in the reconditioned state (c).

The Multibond SK8 adhesive was as a good glue type for the bonding of several hardwoods exposed in dry and water-soaked conditions. This type of PVAc adhesive, for all 13 bonded hardwoods, usually secured the minimum adhesion strengths required by the standard EN 204 [47] (i.e., on average, (a) in the dry state of 13.6 MPa, from 9.5 MPa for bangkirai to 17.2 MPa for bubinga (required minimum = 10 MPa), (b) in the wet state of

1.8 MPa, from 0.6 MPa for *maçaranduba* to 2.6 MPa for beech (required minimum = 2 MPa), and (c) in the reconditioned state of 13.0 MPa, from 8.5 MPa for *maçaranduba* to 19.2 MPa for *bubinga* (required minimum = 8 MPa)) (Table 4, Figure 3).

In the summary evaluation, using a linear correlation $\text{Adhesion} = a + b \times \text{Number of Hardwood}$ (for the numbering of hardwoods, see Figure 1), the adhesion strength of 13 bonded hardwoods determined in the dry state (Figure 3a) unequally decreased in the wet state due to 4 days of soaking in water (Figure 3b). This was based on the coefficient of determination R^2 declining from 0.6 to 0.06, and on the change of the significance level parameter p from 0.000 to 0.021. However, due to 7 days of reconditioning of the wet samples in a dry environment, the adhesion strength recovered quite equally to the initial values found in the dry state, which was in the linear correlation confirmed by the increasing of R^2 to 0.21 and with p equal to 0.000 (Figure 3c).

Partially worsened results were achieved in the wet state of the bonded hardwoods, when the adhesion strength decreased in more cases under the criteria value of 2 MPa (i.e., to 0.6 MPa–2.6 MPa) (Table 4, Figure 3), which could be explained, among other factors, with a different penetration of the PVAc adhesive into the individual hardwood species. For example, the lowest adhesion of 0.6 MPa was determined for the *maçaranduba* wood, the densest species (Table 1) having the lowest roughness (Table 3). On the contrary, the second-highest adhesion of 2.3 MPa was determined for the *meranti* wood, the least dense species (Table 1) having the highest roughness (Table 3). The highest adhesion of 2.6 MPa was determined for the beech wood (i.e., the species characterized by very good permeability [60,61]). Generally, different microstructures of the wood–adhesive–wood interfaces, when a probable better penetration of adhesives into less dense and more porous woods, as well as into more permeable woods, could be connected with a higher and more water-stable mechanical adhesion of adhesives with wood surfaces.

Results achieved in this work for the bonded beech wood were in accordance with the work of He and Chiozza [62]. For this wood species, bonded it at 23 °C with the PVAc glue Vinavil 2259 L, they determined by the standard EN 205 [46] the adhesion strength in the dry state 15.8 MPa and in the wet state 2.7 MPa.

3.3. Connections between Bonding and Selected Characteristics of Hardwoods

The adhesion strengths of 13 bonded hardwoods, valued in the dry state, were positively influenced—on the 99% or 95% significance level ($p < 0.01$; $p < 0.05$)—by their higher densities ρ , lower roughness parameters R_a and R_z perpendicular to the grain, and lower water contact angles θ_0 , θ_e , and θ_w , as was documented by the linear correlations ($\text{Adhesion} = a + b \times \text{Property of wood}$) (Table 5 and Figure 4a,d–f).

On the contrary, the adhesion strengths of the bonded hardwoods were not significantly influenced by the cold water extract, pH value, or roughness parameters R_a , R_z , and R_{Sm} parallel with the grain ($p > 0.5$) (Table 5 and Figure 4b,c).

Generally, the joint quality of bonded timbers depends on several surface characteristics of the wood, including its wetting capacity. For example, beech and *bubinga* woods were characterized with good wettability values (Table 3), and this property resulted in a positive impact on their adhesion strength with a PVAc adhesive in the dry state (Table 4 and Figures 3a and 4e,f), as well as in the wet state and reconditioned state (Table 4). However, the wettability may not always be a presuming factor. For example, a good bonding quality in the dry state was also determined for *ipé*, *iroko*, *sapelli*, and *wengé* woods, whose surfaces had evidently higher water contact angles.

Table 5. Linear correlation analyses between the hardwood characteristics or properties and the adhesion strength, determined for the wood–PVAc adhesive–wood interface in the dry state.

Property of Hardwood	N	R ²	T	p	Adhesion = a + b × Property
Density ρ (kg/m ³)	78	0.086	2.68	0.009	$8.36 + 0.007 \times \rho$
Cold water extract (%)	39	0.004	0.39	0.699	$12.94 + 0.16 \times \text{extract}$
pH	39	0.040	1.24	0.224	$10.06 + 0.71 \times \text{pH}$
Roughness parallel with grain (μm)					
<i>Ra</i>	78	0.027	−1.47	0.147	$14.22 - 0.097 \times Ra$
<i>Rz</i>	78	0.015	−1.06	0.292	$14.14 - 0.012 \times Rz$
<i>RSm</i>	78	0.039	1.75	0.085	$12.43 + 0.002 \times RSm$
Roughness perpendicular to grain (μm)					
<i>Ra</i>	78	0.071	−2.41	0.018	$14.56 - 0.087 \times Ra$
<i>Rz</i>	78	0.081	−2.59	0.011	$15.05 - 0.016 \times Rz$
<i>RSm</i>	78	0.006	−0.68	0.498	$14.03 - 0.001 \times RSm$
Contact angle (°)					
θ_0	78	0.11	−3.03	0.003	$17.52 - 0.044 \times \theta_0$
θ_e	78	0.10	−2.94	0.004	$15.62 - 0.051 \times \theta_e$
θ_w	78	0.13	−3.40	0.001	$15.82 - 0.043 \times \theta_w$

Note: N is the number of samples. However, several times more measurements were performed and by linear correlations analyzed for the roughness parameters and the contact angles, as for each individual sample, the roughness parameters were determined on 15 spots and the contact angles on 5 spots (see Tables 2 and 3).

Figure 4. Cont.

Figure 4. Expression of the linear tendency changes of the mean adhesion strengths of 13 bonded hardwoods, determined in the dry state, in relation to the mean properties of these wood species – (a) density, (b) cold-water extract, (c) pH value, (d) roughness parameter R_a perpendicular to the grain, (e) initial contact angle, (f) abstract contact angle. Note: In Table 5, statistical analyses carried out from all individual measurements are present.

4. Conclusions

The adhesion strengths of 13 hardwoods bonded with the PVAc adhesive Multibond SK8 met, in most cases, the requirements of the standard EN 204 [47] (i.e., it ranged from 9.5 MPa to 17.2 MPa (limit 10 MPa) in the dry state, from 0.6 MPa to 2.6 MPa (limit 2 MPa) in the wet state, and from 8.5 MPa to 19.2 MPa (limit 8 MPa) in the reconditioned state).

The water contact angles, expressing the wettability of wood surfaces, were not clearly affected by the other measured structural and physical characteristics of the hardwoods. For example, for the most dense wood, *maçaranduba* wood (1105 kg/m^3), in comparison with the less dense *meranti* wood (636 kg/m^3), was determined to have a 136% greater cold water extract, 36% higher pH value, 63.3% lower roughness parameter R_a parallel with the grain, and 71.6% lower roughness parameter R_a perpendicular to the grain. However, the initial contact angles of these two tropical woods θ_0 were essentially the same, being 94.0° and 92.0° , respectively. This means that the wettability of wood surfaces with water, which usually dominantly affects the adhesion strength between wood surfaces and water-based adhesives or coatings, can simultaneously be dependent on the combination of several other structural parameters of the wood, including its specific molecular structure. This should be analyzed in any following experiments.

The linear correlations indicated that the mutual relations between the adhesion strength values in the dry state of 13 hardwoods bonded with a PVAc adhesive and the cold water extract, pH value, or roughness parameters parallel with the grain were not statistically significant. On the contrary, the adhesion strength values showed negative tendencies on the 95% significance level, influenced by the increased roughness parameters R_a and R_z perpendicular to the grain, and on the 99% significance level, influenced by the increased water contact angles θ_0 , θ_e , and θ_w of the individual hardwoods.

Author Contributions: Conceptualization, J.I., L.R. and J.S.; methodology, J.I., L.R., J.S., J.K. and V.K.; software, J.I. and L.R.; validation, J.I., L.R. and J.K.; formal analysis, J.I., L.R. and J.K.; investigation, J.I., L.R., J.S., J.K. and V.K.; resources, J.I., L.R., J.K. and V.K.; data curation, J.I., L.R. and J.K.; writing—original draft preparation J.I., L.R. and J.K.; writing—review and editing, J.I. and L.R.; visualization, J.I. and L.R.; supervision, J.I. and L.R.; project administration, L.R. and J.K.; funding acquisition, J.I. and L.R.. All authors have read and agreed to the published version of the manuscript.

Funding: This work was supported by the Slovak Research and Development Agency under the contracts no. APVV-17-0583 and no. APVV-16-0177, and the VEGA project 1/0729/18.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data sharing is not applicable to this article.

Acknowledgments: The authors would like to thank the Slovak Research and Development Agency under contracts no. APVV-17-0583 and no. APVV-16-0177, and also to the VEGA project 1/0729/18 for funding and financial support. This publication is also the result of the following project implementation: Progressive research of performance properties of wood-based materials and products (LignoPro), ITMS 313011T720 supported by the Operational Programme Integrated Infrastructure (OPII) funded by the ERDF.

Conflicts of Interest: The authors declare no conflict of interest.

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