

Potential Use of Waste Marble Powder as Adhesive Filler in the Manufacture of Laminated Veneer Lumber

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The waste powder produced during the manufacture of marble, which is presently mostly discarded in landfills, has the potential for higher-valued usage. Recycling marble waste powder will contribute to the protection of nature as well as economic gain. The potential use of waste marble powder as filler in the adhesive for manufacturing of laminated veneer lumber (LVL) was investigated in this study. With this objective, LVL panels were manufactured by adding marble factory waste powder in different ratios to polyvinyl acetate (PVAc) and urea-formaldehyde (UF) adhesives. The waste powders of beige marbles and travertine marbles were mixed by weight with adhesives in the percentage levels of 0%, 10%, 20%, 30%, 40%, and 50%. Physical and mechanical tests were performed on the specimens obtained from the LVL panels. According to the results obtained from the present study, as compared to the control specimens, higher mechanical performance was obtained with the waste beige marble powder at high contents and with the waste travertine marble powder at low contents with the PVA adhesive. A poorer mechanical performance was found in the travertine powder with the UF adhesive. The dimensional stability of LVLs containing travertine powder was better than that of the LVLs containing beige marble powder.

Key words: Adhesive; Filler; Laminated veneer lumber (LVL); Waste marble dust

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INTRODUCTION

Currently, great effort is being put into finding ways to reuse and recycle materials resulting from wastes. The reutilization of wastes prevents damage to nature by decreasing the use of limited natural resources, increasing productivity in manufacturing, and minimizing the environmental problems that form as a result of storing wastes (Bilensoy 2010).

Marble, used frequently in construction applications, is utilized by an industry that produces a significant amount of solid waste during the cutting process. Recycling the marble powder produced during cutting is an important issue because of the current development of environmental awareness and costs for filler (Bilensoy 2010). It is estimated that approximately 2.2 million tons of marble blocks are processed in Turkey annually, of which 660,000 tons of marble powder is thrown out without being recycled. Utilization of the waste marble powder in various industrial sectors waste marble powder in industry can provide significant economic gains.

Approximately 2.7 million tons of waste mud has been produced annually by the industries that process natural stone in Turkey, and these wastes are dumped on vacant lots or valleys. When studies on the economic use of marble wastes were examined, it was observed that they reported on the construction sector in general, such as using marble powder wastes in filler processes, the manufacture of light construction blocks, improving floors with marble powder, and as an additive substance in the manufacture of cement (Khristova and Aniskevich 1994; Ali *et al.* 2000; Garcia *et al.* 2003; Zorluer and Usta 2003; Agarwal and Gulati 2006; Bilgin 2010; Tanyıldızı and Coşkun 2011; Erdem and Öztürk 2012). Excluding the use of wastes from marble processing facilities as construction materials, it was observed that they are used as powder wastes reduced to different dimensions as a raw material decoration in architecture, a filler material, and as an additive substance for agricultural purposes (Gündüz and Şentürk 1996; Demirel and Yazıcıoğlu 2010).

For LVL manufacturing, fillers are added to the adhesive mix to improve its workability. The fillers used in glue mixtures typically act to fill holes and irregularities in veneer and to retain adhesive on the glueline (Avery *et al.* 1989). Specifically, these finely ground organic and/or inorganic materials promote bonding by holding the adhesive on the veneer surface where it is needed. Aside from performance, desirable filler features include low cost, consistent quality, and sufficient supplies (Eberhardt and Reed 2006). Examples of inorganic fillers include clays such as attapulgite and bentonite (Avery *et al.* 1989). To our knowledge, there are no reports on the utilization of waste marble powder of marble factories as adhesive inorganic filler in the wood-based panel industry. The objective of this study was the manufacture of laminated veneer lumber (LVL) by adding two kinds of marble in different ratios within adhesive and studying their physical and mechanical properties. The aim of the present work was to investigate the effect of marble powder content on the bond performance of polyvinyl acetate (PVAc) and urea-formaldehyde (UF) adhesives. The dimensional stability and mechanical properties of the LVL panels manufactured with the PVA and UF adhesives containing marble powder were determined.

EXPERIMENTAL DETAILS

Materials

Two kinds of marble powder, travertine and beige, were used in this study. The wastes were obtained from the collected liquid precipitation ponds at a factory manufacturing travertine marble in Denizli, Turkey, and a factory manufacturing beige marble in Burdur, Turkey. The technical properties of two kinds of marble powder are presented in Table 1. The waste travertine and beige marble powders retained on a 0.25-mm ring sieve were used as filler in the adhesive for the LVL manufacturing (Fig. 1). The waste marble powder is collected on a 0.25-mm ring sieve during the marble production process in the factory. The marble powders were then oven-dried to a moisture content of 0 to 1% using a laboratory oven at 100 °C for 48 h, and then stored in a sealed container.

Wood material

Wood material from Oriental beech (*Fagus orientalis* Lipsky) was chosen for the manufacturing of LVLs because of its widespread use in the furniture industry in Turkey.

The LVL panels were manufactured by bonding nine layers of beech veneers with a thickness of 2.2 mm.

Table 1. Technical Properties of the Marbles

Property	Unit	Beige Marble ¹	Travertine Marble ²
Hardness	Mohs	3	3 to 3.5
Density	g/cm ³	2.72	2.66
Particle size	µm	<250	<250
Water absorption	By weight (%)	0.08	1.62
Porosity	%	0.40	3.94
Compressive strength	N/mm ²	156.0	54.4
Degree of pores	%	0.30	3.94
SiO ₂	%	2.81	1.91
F ₂ O ₃	%	0.10	79.10
MgO	%	1.40	1.15
CaO	%	52.90	2.43

¹ Test report of Demirkayalar Marble Company, Denizli, Turkey, 2009.

² Test report of Erdem Marble Company, Denizli, Turkey, 2011.



Fig. 1. Waste marble powder

Adhesives

The PVAc and UF adhesives were obtained from an adhesive company in Turkey. The specifications of the adhesives are presented in Table 2.

Table 2. Specifications of the Polyvinyl Acetate and Urea-Formaldehyde Adhesives

Property	Unit	Polyvinylacetate (PVA) ¹	Urea-formaldehyde resin (UF) ²
Solid content	% (after 2 h, 120 °C)	48	50-70
Viscosity	mPas (at 20 °C)	15000	7000
pH	-	2.5-3.5	7
Density	g/cm ³	1.05	0.5 ³
Period of hardening	min (at 60 °C)	20-30 (20°C)	9-11 (60°C)

¹ Catalogue of Wurth Inc., D3/D4 189211014 Adhesive, USA.

² Catalogue of Durante & Vivian Company, Duroxill 850 UF adhesive, Italy.

³ Bulk density.

Methods

Manufacturing of the LVL panels

After preparing the veneers with dimensions of 200 mm (width) \times 1000 mm (length) \times 2.2 mm (thickness), the LVL manufacturing commenced. First, the different mixtures of adhesive and waste marble powder were prepared (Table 3). The mixtures were obtained by mixing the adhesives and the waste marble powder in different containers in ratios of 10%, 20%, 30%, 40%, and 50% by weight. The liquid UF adhesive solution containing the waste marble powder as filler was applied using a brush on single bonding surfaces of veneers at approximately 180 g/m². After the gluing process, nine veneers were placed with their grain directions parallel to the grain direction of the neighbour veneers. The hot-press pressure, temperature, and time for the UF adhesive bonded LVLs were 1.2 N/mm², 105 °C, and 10 min, respectively. As for the PVA adhesive, the nine glued veneers were assembled together, one on the top of the other, and pressed in a hot-press for 12 h under a pressure of 1.2 N/mm² at a temperature of 20 °C. The resulting LVL panels (20 mm thick) were trimmed to 180 mm \times 900 mm and conditioned in a conditioning room at a temperature of 23 ± 2 °C and a relative humidity (RH) of 65 ± 2%. Six LVL panels were produced for each type of adhesive formulation (Table 3). The densities of the LVL panels varied from 0.66 to 0.77 g/cm³.

Table 3. Composition of the LVL Panels

Adhesive	Kind of waste marble	Amount of waste marble powder in adhesive (weight %)	LVL type
PVA (P)	Travertine (T)	10	PT10
		20	PT20
		30	PT30
		40	PT40
		50	PT50
	Beige (B)	10	PB10
		20	PB20
		30	PB30
		40	PB40
		50	PB50
	Control		PK
UF (U)	Travertine (T)	10	UT10
		20	UT20
		30	UT30
		40	UT40
		50	UT50
	Beige (B)	10	UB10
		20	UB20
		30	UB30
		40	UB40
		50	UB50
	Control		UK

Physical properties of LVL panels

LVL specimens with dimensions of 20 mm \times 30 mm \times 20 mm were prepared according to ISO 3131 (1975) to determine the air-dried and oven-dried density of the LVL panels. The width and thickness variations of the LVL specimens with dimensions of 30 mm \times 30 mm \times 20 mm were determined in conformance with ISO 4859 (1982)

standard. According to ISO 4859, the width and thickness variations of LVL panels, between two equilibrium moisture contents, are calculated as a percentage of the initial specimen width and thickness. First, the width and thickness of the specimens conditioned at 20 ± 2 °C and $65 \pm 5\%$ RH were measured with a digital micrometer having an accuracy of ± 0.001 mm as the specimens reached the equilibrium moisture content of $12 \pm 2\%$. When the same specimens reached the equilibrium moisture content of $21 \pm 2\%$ in the conditioning room at 20 ± 2 °C and $90 \pm 5\%$ RH, they were measured again.

Mechanical properties of LVL panels

LVL specimens with dimensions of $360 \text{ mm} \times 20 \text{ mm} \times 20 \text{ mm}$ (thickness) were prepared to determine the bending properties. Three-point bending strength or modulus of rupture (MOR) and modulus of elasticity (MOE) in bending were determined in accordance with ISO 3133 (1975) and ISO 3349 (1975), respectively. The MOR and MOE specimens were prepared for the parallel direction to the grain of the surface layers of the LVL panels. The specimens were tested in a universal testing machine (UTEST 7012, Ankara, Turkey) with a 5-kN load cell at a crosshead speed of 2 mm/min.

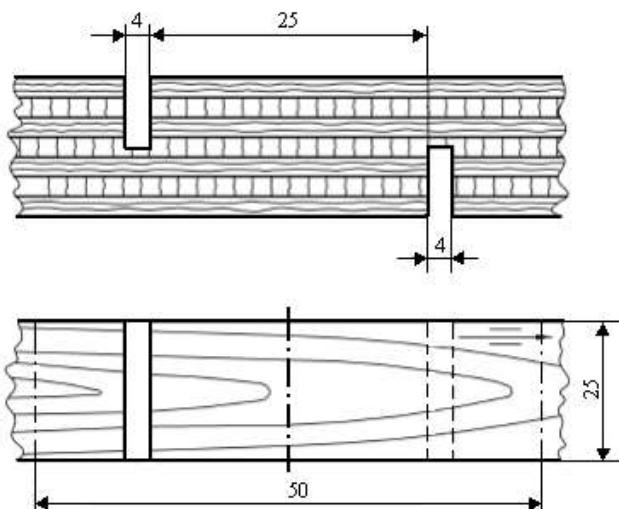


Fig. 2. Test piece schematic for tensile-shear strength parallel to the fibers (from EN 314-1 2004)

Tensile-shear strength parallel direction to the grain of the surface layers of specimens with dimensions of $50 \text{ mm} \times 25 \text{ mm} \times 20 \text{ mm}$ (thickness) was determined according to EN 314-1 (2004) (Fig. 2). Before the mechanical tests, all the specimens were conditioned in a conditioning room at a temperature of 23 ± 2 °C and a RH of $65 \pm 2\%$.

Statistical analysis

Statistical data analysis was performed using SPSS Software Program (SAS statistical package for Windows, SAS Institute Inc, NC, USA) for analysis of variance (ANOVA). Mean differences among the LVL types were analyzed using Duncan's multiple range test at a significance level of 0.05.

RESULTS AND DISCUSSION

Physical Properties

Physical properties of the LVL panels manufactured with the PVA or UF adhesives containing the waste marble powder are presented in Table 4. The significant differences ($p < 0.05$) found by Duncan test analysis for each test among the LVL types are indicated in Table 4.

Table 4. The Physical Properties of the LVL Panels

LVL type	Physical properties*			
	D ₀ ** (g/cm ³)	D ₁₂ *** (g/cm ³)	Swelling (%)	
			Width (α _w)	Thickness (α _t)
PT10	0.68	0.71	7.45 (1.32) ^d	6.63 (0.64) ^{abc}
PT20	0.70	0.72	5.89 (1.56) ^{abcd}	7.63 (1.26) ^{bc}
PT30	0.67	0.69	5.72 (0.61) ^{abc}	7.01 (0.61) ^{bc}
PT40	0.70	0.72	7.46 (1.31) ^d	5.34 (0.75) ^a
PT50	0.70	0.72	5.93 (1.70) ^{abcd}	6.81 (1.10) ^{abc}
PB10	0.72	0.74	6.40 (1.23) ^{abcd}	7.20 (1.84) ^{bc}
PB20	0.71	0.73	7.06 (1.87) ^{bcd}	7.00 (0.94) ^{bc}
PB30	0.70	0.72	6.08 (1.49) ^{abcd}	6.64 (1.03) ^{abc}
PB40	0.73	0.75	5.84 (1.00) ^{abcd}	7.21 (0.85) ^{bc}
PB50	0.70	0.74	6.55 (0.81) ^{abcd}	8.24 (1.34) ^{cd}
PK	0.71	0.74	5.46 (1.70) ^{ab}	9.71 (1.93) ^d
UT10	0.68	0.70	6.52 (1.12) ^{abcd}	6.39 (0.53) ^{ab}
UT20	0.67	0.70	4.96 (0.65) ^a	7.00 (1.74) ^{bc}
UT30	0.70	0.72	4.98 (0.62) ^a	7.06 (1.04) ^{bc}
UT40	0.72	0.75	5.80 (1.75) ^{abcd}	6.89 (1.82) ^{abc}
UT50	0.70	0.72	6.40 (1.20) ^{abcd}	6.27 (1.71) ^{ab}
UB10	0.66	0.67	6.19 (0.73) ^{abcd}	6.57 (1.02) ^{abc}
UB20	0.77	0.73	5.99 (1.53) ^{abcd}	7.08 (0.78) ^{bc}
UB30	0.69	0.70	7.22 (0.50) ^{cd}	7.61 (1.30) ^{bc}
UB40	0.66	0.69	6.28 (0.71) ^{abcd}	8.27 (1.22) ^{cd}
UB50	0.70	0.72	6.20 (0.72) ^{abcd}	7.98 (1.29) ^{bc}
UK	0.68	0.74	4.99 (0.60) ^a	7.55 (0.59) ^{bc}

*Groups with the same letters in each column indicate that there is no statistical difference ($p < 0.05$) between the LVL types according Duncan's multiple range test. The values in parentheses are standard deviations; ** Oven-dry density (0% moisture content);

*** Air-dry density (12% moisture content)

According to the results of the swelling tests, the changes in the thickness of the specimens decreased with the incorporation of the marble powder into the PVA or UF adhesive. The swelling values of the LVLs containing the travertine powder were lower than those of the LVLs containing the beige marble powder (Table 4). The lowest thickness swelling values (α_t) among the PVA adhesive/marble powder treatment group were found in the LVL types PT40 (5.34%) and PB30 (6.64%). As for the UF adhesive/marble powder treatment group, the lowest thickness swelling values were found in the LVL types UT50 (6.27%) and UB10 (6.57%). The width swelling values of the specimens (α_w) were higher than those of the control LVLs.

Among the PVA/marble powder treatment group, the lowest width swelling values within the specimens (α_w) were found in the LVL types PB40 (5.84%) and PT30 (5.72%). As for the UF adhesive/marble powder treatment group, the lowest width

swelling values within the specimens (α_w) were found in the LVL types UT20 (4.96%) and UB20 (5.99%). The width swelling values (α_w) of the control LVL manufactured with the PVA adhesive or the UF adhesive were found to be 5.46% and 4.99%, respectively. Based on the swelling values, it can be said that the optimum marble powder ratio for the PVA adhesive was 30 wt%, while it was 10 wt% for the UF adhesive.

Mechanical Properties

The mechanical properties of the LVL panels manufactured with the PVA or UF adhesives containing the waste marble powder in different ratios are presented in Table 5. The MOR of LVL types UB10, UB50, UB30, and UT10, which were manufactured with UF adhesive, were higher than that of the control LVL (92.2 N/mm²). The highest MOR value (102.5 N/mm²) was found in the LVL type UB10, followed by the LVL types UB50 (95.8 N/mm²), UB30 (95.3 N/mm²), and UT10 (94.1 N/mm²), respectively. According to these results, the mixture of UF adhesive (U) and the waste beige marble powder (B) provided the best formulation. It was determined that the addition of 10 wt% marble powder into the adhesive provided the best MOR result. The incorporation of the marble powder into the adhesive increased the MOE values of the LVLs more than the MOR values. In general, an increment was observed in the MOE values of the LVLs as the amount of the marble powder increased to 50 wt% in the PVA or UF adhesive.

The tensile-shear strength of the LVL specimens decreased as the amount of marble powder in the adhesive increased. This was generally attributable to the hydrophobic (apolar) surface of the marble powder, which was incompatible with the hydrophilic characteristics of the UF adhesive and the wood surface. In particular, it was estimated that a high ratio of inorganic filler decreased the chemical bonding of the adhesive with the hydroxyl groups of the wood. The present results were consistent with the results of Lee *et al.* (2009), who determined that inorganic substances weakened the bond performance of UF adhesives. As for the PVA adhesive, the tensile-shear strength of the LVL types PB40, PT10, PB20, PT40, and PT50 were higher than that of the control LVL. The PVA adhesive displayed a better performance for tensile-shear strength compared to the UF adhesive. As for the UF adhesive, the tensile-shear strength of the LVL types UB20 and UB30 was higher than that of the control LVL.

When the UF adhesive was mixed with marble powder, it exhibited a higher bending strength compared to the PVA adhesive. As the ratio of marble powder within the adhesive increased up to 10%, it decreased the absorption of the adhesive by the veneer, which caused the adhesive to remain at the glue line. As the ratio of marble powder within the adhesive increased to 30 wt%, it improved the spreadability of the adhesive. This situation showed that a significant portion of the adhesive containing marble powder is not absorbed by the veneer and remains at the glueline. The MOE of LVL types PT50, PT40, and PT10 were better than that of the control LVL. According to these results, in complete contrast to the bending strength analysis, the mixture of PVA (P) adhesive and waste travertine marble powder (T) provided the highest MOE for the LVL. Based on the MOE property, the optimum amount of the marble powder in the adhesive was found to be 50 wt%. The fact that there was a complete contrast in the results between the bending strength and modulus of elasticity values is a situation that should be considered in future work.

When the results were closely examined, it was observed that the LVL types PT50, PT40, and PT10 showed a good performance for both the MOE and tensile-shear

strength while the LVL types UT20 and UT40 showed a poor performance for the MOR and tensile-shear strength. Based on the MOR results, a good compatibility was found between the waste beige marble powder and the UF adhesive, while this was not observed for the waste travertine marble powder. In contrast to this, a good result was found in the MOE values of the LVLs bonded with the mixture of PVA adhesive and waste travertine marble powder. Despite this, the travertine marble powder with the PVA adhesive gave a good result for the MOE. As compared to the control group, a high tensile-shear strength was determined for the PVA adhesive containing the waste beige marble powder or the waste travertine marble powder (Table 5).

As shown in Table 1, the porosity of the travertine marble is considerably higher than that of the beige marble. The higher porosity of the travertine marble could be a reason for the lower tensile-shear strength and MOR. This is because the adhesive can penetrate into the porous structure of the inorganic and decreases the amount of the adhesive in the glue line. Adhesives can effectively transfer and distribute stresses, thereby increasing the strength and stiffness of the composite (Vick 1999). As compared to the control LVLs, the improvement in the MOE values of LVLs can be explained by the hardness of the waste marble powder. The higher MOE of the LVLs containing waste travertine powder was mainly attributed to its higher hardness. The hardness of a material tends to increase with an increase in the elastic modulus (Lan and Venkatesh 2014).

Table 5. The Mechanical Properties of the LVL Panels

LVL type	Mechanical properties*		
	Bending strength (MOR) (N/mm ²)	Modulus of elasticity (MOE) (N/mm ²)	Tensile-shear strength (N/mm ²)
PT10	90.0 (5.76) ^{bcd}	345.8 (10.67) ^{ab}	9.25 (0.94) ^a
PT20	80.3 (13.69) ^{efg}	293.5 (55.90) ^{def}	6.36 (0.61) ^{efg}
PT30	67.17 (11.53) ^h	234.6 (16.63) ^h	5.57 (1.17) ^{gh}
PT40	81.7 (5.90) ^{ef}	346.5 (27.46) ^{ab}	9.08 (0.56) ^a
PT50	89.2 (6.83) ^{bcd}	368.8 (29.09) ^a	8.78 (1.09) ^{ab}
PB10	79.8 (6.96) ^{efg}	246.2 (37.31) ^{gh}	6.32 (1.36) ^{fg}
PB20	76.5 (10.06) ^{fgh}	318.7 (31.57) ^{bcd}	9.14 (1.08) ^a
PB30	70.7 (3.82) ^{gh}	285.9 (11.56) ^{ef}	7.50 (0.66) ^{cd}
PB40	82.4 (10.25) ^{def}	329.6 (20.82) ^{bc}	9.42 (0.93) ^a
PB50	76.3 (5.34) ^{fgh}	338.3 (18.75) ^b	7.03 (0.61) ^{def}
PK	82.4 (11.32) ^{def}	249.2 (39.42) ^{gh}	8.07 (0.86) ^{bc}
UT10	94.1 (5.89) ^{ab}	304.8 (12.07) ^{cde}	5.57 (1.09) ^{gh}
UT20	53.7 (23.31) ^l	307.5 (13.90) ^{cde}	2.58 (0.44) ^k
UT30	82.8 (12.66) ^{def}	304.4 (26.59) ^{cde}	3.76 (1.07) ^{ij}
UT40	49.9 (9.15) ^l	269.3 (17.58) ^{fg}	2.97 (0.41) ^{jk}
UT50	90.1 (4.38) ^{bcd}	326.0 (24.33) ^{bc}	4.01 (0.58) ^l
UB10	102.5 (9.55) ^a	289.7 (26.73) ^{ef}	5.33 (1.13) ^h
UB20	85.5 (8.83) ^{cdef}	306.5 (41.08) ^{cde}	7.08 (0.73) ^{def}
UB30	95.3 (8.45) ^{ab}	340.1 (38.02) ^b	7.21 (1.35) ^{de}
UB40	87.1 (4.45) ^{bcd}	326.1 (2.47) ^{bc}	4.02 (0.50) ^l
UB50	95.8 (8.06) ^{ab}	303.3 (15.61) ^{cde}	4.06 (0.50) ^l
UK	92.2 (9.35) ^{bcd}	303.9 (12.04) ^{cde}	6.02 (0.97) ^{gh}

*Groups with the same letters in each column indicate that there is no statistical difference ($p<0.05$) between the LVL types according to Duncan's multiple range test. The values in parentheses are standard deviations.

CONCLUSIONS

1. As compared to the control specimens, the incorporation of the marble powder into the PVA or UF adhesive decreased the thickness swelling of the LVLs as the RH of the climate room increased from 50 to 90% while the width of the LVLs increased. The dimensional stability of LVLs containing travertine powder was better than that of the LVLs containing beige marble powder.
2. The mechanical properties of the LVLs increased as the amount of the waste beige marble powder increased in the PVA adhesive while the same properties decreased as the amount of waste travertine marble powder in the adhesive increased. Consequently, the addition of travertine marble powder into the PVA adhesive should be made at the low ratio of 10%.
2. The addition of waste travertine marble powder into the UF adhesive displayed a poor mechanical performance. Therefore, if the waste travertine marble powder is to be used in LVL manufacture, then UF adhesive should not be used.
3. The waste beige marble powder displayed a better compatibility with the UF adhesive than the waste travertine marble powder, based on the mechanical properties of the LVL specimens.
4. The utilization of waste marble powder in the UF or PVA adhesive can improve the mechanical properties of the LVL. Moreover, the waste marble powder can decrease the manufacturing cost of LVL because it considerably decreases the cost of adhesive.

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