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Original article

Effect of Tannin Addition as a Bio-Scavenger on Formaldehyde Content in Particleboard

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ABSTRACT

Tannin is a bitter-tasting organic substance found in plant tissue and finds plausible applications as an adhesive in wood based panel production because it is easy to handle and obtained from renewable sources. The objective of this study was to investigate the influence of tannin addition to particleboard and its affect on formaldehyde content in the particleboard. Tannin had an impact in terms of reducing the amount of formaldehyde content in the particleboard. When particleboards with and without tannin were compared , the addition of 1.2% tannin (based on oven dry weight of Urea formaldehyde (UF) resin) decreased the formaldehyde content by 21.35% and the various properties of tannin had negligible effect on the formaldehyde content. The addition of tannin improved the physical properties, although the effect on mechanical properties was ambiguous. Therefore, the addition of tannin can reduce the formaldehyde content and it can act as a formaldehyde scavenger.

Keywords: Bio-scavenger; Formaldehyde content; Particleboard; Tannin.

INTRODUCTION

Formaldehyde, a colorless flammable chemical, was discovered in 1855 by a Russian scientist Alexander Michailowitsch Butlerow. By the 1860s, formaldehyde had found usage in various industrial applications. Subsequently it has been widely used in adhesive manufacturing for wood based panel production (Salthammer *et al*., 2010). Urea formaldehyde (UF) resin has been widely used as a synthetic resin in the wood-based panel industry due to its ease of handling and application, low cost, and the finished product being colorless (Levendis *et al*., 1992; Pizzi *et al*., 1994; Duan *et al*., 2015). However, UF resin releases some hazardous formaldehyde, which can potentially cause cancer and has been linked with the 'sick building' syndrome (Kavvouras *et al.,* 1998;

Park *et al*., 2013; Duan *et al*., 2015). The emission of hazardous formaldehyde is affected by various factors such as raw materials, types of resin, air humidity, temperature, and other production conditions (Roffael, 1993). In the past, the US Environmental Protection Agency has raised concerns about the emission of volatile organic compounds (VOCs) into the environment. Therefore, in recent decades, several approaches have been developed to solve the problems associated with excessive formaldehyde emission from wood panels (Dunky, 1998; Moubarik *et al*., 2013).

Several techniques for the reduction of formaldehyde in wood-based panel industry have been developed, such as treatment of panels with ammonia gas or ammonia salts, addition of formaldehyde scavengers, and surface treatment with paints, lacquers, veneer, or papers. Among these formaldehyde emission (FE) reduction techniques, surface treatment is the least effective (Eom *et al*., 2006; Duan *et al*., 2015), while the most efficient method is the chemical addition of formaldehyde scavengers (Boran *et al.*, 2012).

Bertaud *et al.* (2012) reported that formaldehyde scavenging is the most effective technique in reducing FE. These scavengers can be divided into two types; synthetic scavengers and bio-scavengers. Bio-scavengers are an environmentally friendly way to reduce formaldehyde emission from wood-based panels and include substances such as tannin, lignin, starch, and soy protein. A bio-scavenger has many advantages such as it can be found abundantly, has a comparatively lower cost, and is eco- friendly (Imam *et al*., 1999). Starch adhesive was developed through fortification with other polymers to be used as an adhesive in plywood (Moubarika *et al.,* 2009). Tannin is suitable as a green adhesive and bio-scavenger due to its availability, low price, and the large quantities of high-quality tannin fractions (Ping *et al*., 2011).

Tannins can be mixed in plywood adhesives and can act as a bio-scavenger in particleboards, plywood, OSB (oriented strand board), MDF (medium density fiber board), glulam, LVL (laminated veneer lumber), and PSL (parallel strand lumber) production (Li and Maplesden, 1998). Condensed tannins, which are a type of tannin, have been used with formaldehyde to develop adhesives to be used in interior and exterior particleboard and plywood applications (Pizzi, 2003, 2006; Pichelin *et al*., 2006; Lei *et al*., 2008; Tondi and Pizzi, 2009). The levels of FE from MDF, which is produced by the UF resin, can be altered by mixing tannin extracted from white oak bark. In addition, the mechanical properties of MDF decrease with an increase in the concentration of tannin in UF resin (Boran *et al*., 2012). However, there has been very little research focusing on using tannin as an easy-to-use bio-scavenger. Therefore, the aim of the current research is to evaluate the influence of tannin and the affect of its properties on formaldehyde content in particleboards.

MATERIALS AND METHODS

Materials

Industrial wood particles (*Havea basilensis*) (fine and coarse particles for face and core layer) derived from plants are used to produce particleboards in Thailand. The wood particles have a moisture content 3-5%.

Commercial UF-E2 resin (65.02% of solid content, pH of 8.5, 1.58 poise of viscosity) with a resin content of 8% and 10% (oven-dry weight of wood basis) was used in the face layer and core layer, respectively. Wax emulsion of 3% (oven-dry weight of wood basis) was used in both the face and core layers. A 1% ammonium chloride solution (oven-dry weight of resin basis) was used as a hardener. Tannin extracts, derived from mangrove bark, were added as a bio-scavenger by spraying as solution.

Methods

The experimental design and pressing conditions of the particleboard samples are shown in Table 1. The dimensions and density of particleboard was selected as 35 cm × 35 cm × 1 cm (length × width × thickness) and 650 g/cm^3, respectively. The percentage of tannin mixed with resin was varied from 0 to 1.2%, to produce eight different combinations (coded A–H). Codes A–D were prepared so as to be the control condition with tannin extracted from mangrove barks, under reflux conditions for 3 hours in a ratio of 100:500:0:0 (bark sample: mixture of acetone and water: sodium sulfite: sodium carbonate). Codes E–H were prepared under optimal condition with tannin extracted from mangrove barks under reflux conditions for 3 hours in a ratio of 100:500:4:1 (bark sample: mixture of acetone and water: sodium sulfite: sodium carbonate), as shown in Table 1. Five replicates were constructed using the procedure, resulting in a total of 40 test samples.

Table 1 Experimental design for tannin as a formaldehyde scavenger.

Panel Code	Tannin additive	
	Loading (% OD weight of resin)	Tannin source
A	0 .0	**Control conditions** (extracted from mangrove barks
B	0.4	under reflux conditions for 3 hours using a 100:500:0:0 (by wt.)
C	0.8	ratio of bark sample: acetone- water (50:50 v/v):
D	1.2	no sodium sulfite : no sodium carbonate.)
E	0.0	**Optimal conditions** (extracted from mangrove barks under reflux
F	0.4	conditions for 3 hours using a 100:500:4:1 (by wt.) ratio of bark sample:
G	0.8	acetone-water (50:50 v/v):
H	1.2	sodium sulfite: sodium carbonate.)

A spray gun was used to apply the UF resin and wax to the wood particles in a rotary drum mixer. Different amounts of tannin extract were sprayed onto separate wood particle samples. Then, each sample mixture was weighed and placed in a forming box to produce the particle mat. The mat was hot pressed at a pressure of 250 MPa and temperature of 160°C for 3 min. After that, the 40 panels (samples) were conditioned at 20°C and 65% relative humidity (RH) until they reached a constant weight. Quantities such as the modulus of rupture (MOR), modulus of elasticity (MOE), internal bond strength (IB), thickness swelling (TS), and water absorption (WA) were determined in accordance with the TIS standard 876 (TIS Standard, 2004). Formaldehyde content was determined in accordance with the EN Standard 120 (EN Standard, 1992). Tannin characterization was determined in accordance with the Neimsuwan's method (Neimsuwan *et al.*, 2017)

RESULTS AND DISCUSSION

Characterization of tannins

Two sets of conditions were used in this experiment. The first set, called the control conditions, involved tannin extracted from mangrove bark under reflux conditions for 3 hours using a 100:500:0:0 (by weight) ratio of bark sample:acetone-water (50:50 v/v):sodium sulfite:sodium carbonate. The second set, called the optimal conditions, involved tannin extracted from mangrove bark under reflux conditions for 3 hours using a 100:500:4:1 (by weight) ratio of bark sample:acetone-water (50:50 v/v):sodium sulfite:sodium carbonate. The obtained tannin characteristics are shown in Table 2. The Stiasny number (SN) of the tannin extracted under the control and optimal conditions was 76.76% and 87.69%, respectively. The gel time was 1.11 mins and 14.35 mins while the pH level was 4 and 6, for the control and optimal conditions, respectively.

Table 2 Tannin characterization obtained from the extract of mangrove bark.

Conditions	Stiasny Number (%)	pH	Gel Time (min)
Control conditions	76.76	4	1.11
Optimal conditions	87.69	6	14.35

Effect of tannin addition on the mechanical properties of particleboard

The highest MOR value of 4.07 MPa and 6.28 MPa was obtained for panels with 0.8% added tannin, under control and optimal conditions, respectively (Tables 3–4 and Figure 1). The second highest MOR value was 3.92 MPa and 5.94 MPa, obtained from panels with 1.2% added tannin, under control and optimal conditions, respectively. However, the MOE results also showed a similar trend as MOR. It can be concluded that adding tannin, extracted from optical condition, can improve the MOR and MOE levels for particleboard, because tannin can also act as a matrix in particleboard. The highest IB value obtained was 0.037 MPa, which was from a panel with no tannin addition. However, when using tannin from optimal conditions, the highest IB value was 0.167 MPa, for a panel with 0.4% tannin addition. Although tannin could play a significant role as a binding agent in particleboard, the effect on particleboard properties was unclear due to different tannin properties and loading on its production. Tannin could be carefully applied during particleboard production, and its application mechanism and properties should be studied more deeply. Eom *et al.* (2006) found that adding tannin to particleboard only had a marginal effect on the IB level, because the key influence was probably the quantity and distribution of the resin along with the wood particle geometry and the mechanism of interaction between the tannin, UF resin, and wood. This interaction is depicted in Figure 2.

Table 3 Properties of particleboard after the addition of tannin extracted using control conditions.

Panel Code	Value	Density	Bending MOR	Bending MOE	IB	TS	WA	Formaldehyde content
		g/cm³	MPa	MPa	MPa	%	%	mg/100 g Board OD Weight
A	Ave.	0.55	3.13^{A}	548.29^{A}	0.037^{A}	16.76^{A}	20.58^{A}	17.19^{A}
	Std.	0.07	0.76	165.42	0.001	4.26	3.92	0.61
	n	15	15	15	15	15	15	3
B	Ave.	0.55	2.85^{A}	513.57^{A}	0.026^{A}	12.88^{A}	19.71^{A}	15.49^{AB}
	Std.	0.04	1.05	149.03	0.011	4.13	5.16	1.29
	n	15	15	15	15	15	15	3
C	Ave.	0.57	4.07^{B}	667.81^{A}	0.025^{A}	12.49^{A}	28.28^{A}	15.90^{AB}
	Std.	0.04	1.33	178.63	0.009	3.10	8.04	0.06
	n	15	15	15	15	15	15	3
D	Ave.	0.59	3.92^{B}	617.85^{A}	0.027^{A}	13.75^{A}	27.07^{A}	13.39^{B}
	Std.	0.05	1.31	193.85	0.008	9.70	14.76	0.66
	n	15	15	15	15	15	15	3

Note: * Values with different letters (A-B) indicate the statistical significance ($p \leq 0.05$).
Remarks: MOR = Modulus of rupture; MOE = Modulus of elasticity; IB = Internal bond strength; TS = Thickness swell; WA = Water absorption.

Table 4 Properties of particleboard after the addition of tannin extracted using optimal conditions.

Panel Code	Value	Density	Bending MOR	Bending MOE	IB	TS	WA	Formaldehyde content
		g/cm³	MPa	MPa	MPa	%	%	mg/100 g Board OD Weight
E	Ave.	0.55	2.67^{A}	411.13^{A}	0.122^{A}	17.41^{A}	20.40^{AB}	17.19^{A}
	Std.	0.05	0.90	90.19	0.063	6.94	4.20	0.61
	n	15	15	15	15	15	15	3
F	Ave.	0.63	5.64^{B}	883.46^{B}	0.167^{A}	11.96^{B}	17.31^{AC}	17.19^{A}
	Std.	0.06	1.31	241.46	0.048	4.32	2.42	0.61
	n	15	15	15	15	15	15	3
G	Ave.	0.64	6.28^{B}	891.81^{B}	0.131^{A}	11.79^{B}	19.91^{AC}	15.35^{AB}
	Std.	0.04	1.93	372.49	0.064	2.16	2.29	0.08
	n	15	15	15	15	15	15	3
H	Ave.	0.64	5.94^{B}	909.00^{B}	0.143^{A}	13.67^{AB}	23.41^{B}	13.52^{B}
	Std.	0.05	1.74	273.38	0.044	2.25	5.19	0.47
	n	15	15	15	15	15	15	3

Note: * Values with different letters (A-B) denote the statistical significance ($p \leq 0.05$).
Remarks: MOR = Modulus of rupture; MOE = Modulus of elasticity; IB = Internal bond strength; TS = Thickness swell; WA = Water absorption.

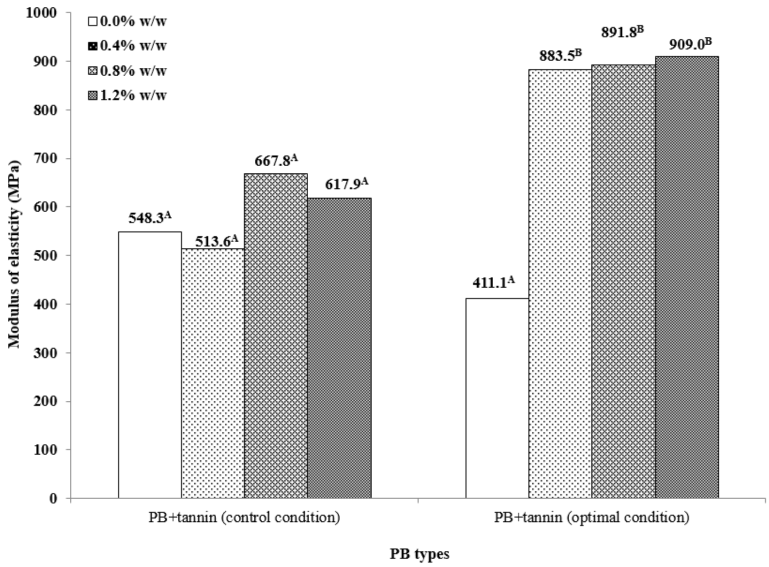

Figure 1 Modulus of rupture (MOR)valuesforparticleboard (PB) after the addition of different levels of tannin.

Note : * Values with different letters (A-B) indicatethe statistical significant (p ≤ 0.05).

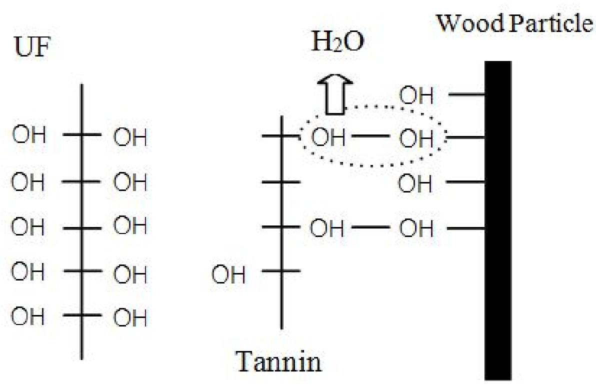

Figure 2 The mechanism of tannin, UF resin, and wood particle interaction (Kim *et al.,* 2006).

Effect of tannin addition on the physical properties of particleboard

Quantification of the TS levels indicated that the lowest value was 12.49% and 11.79%, for a panel with 0.8% tannin addition, extracted under control and optimal conditions, respectively (Tables 3−4 and Figure 5). The lowest WA value was 19.71% and 17.31% for a panel with 0.4% tannin addition, again extracted under control and optimal conditions, respectively (Figure 6). From the results it can be seen that the tannin extracted using optimal conditions, effects the level of TS and WA, although the effect on WA was ambiguous. It is possible that the TS of particleboard was affected by three sources: 1) swelling of the wood particles from the stress that occurs during hot press; 2) swelling due to water absorption; and 3) swelling due to stress in the board. WA might be influenced, to a large extent, by the voids in the particleboard structure. Tannin could improve the physical properties of particleboard by reducing the stress in the panel resulting from the collapse of gaps.

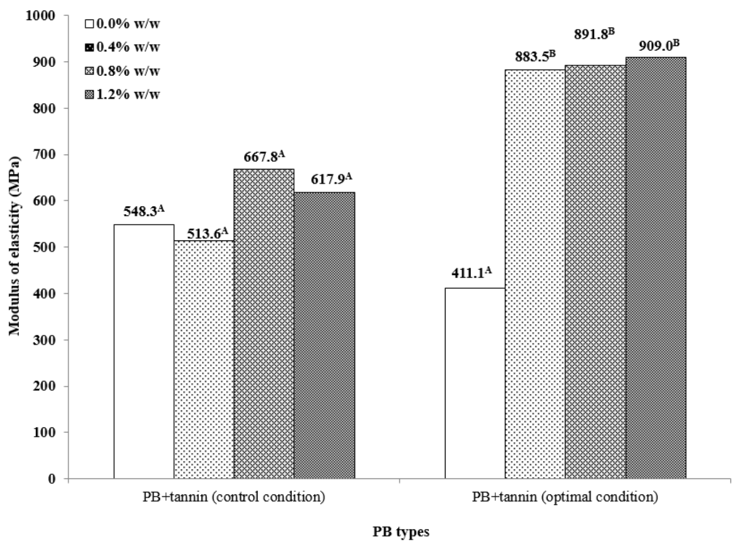

Figure 3 Modulus of Elasticity (MOE) valuesfor particleboard (PB) after the addition of different levels of tannin.

Note : * Values with different letters (A-B) are statistically significant ($p \leq 0.05$).

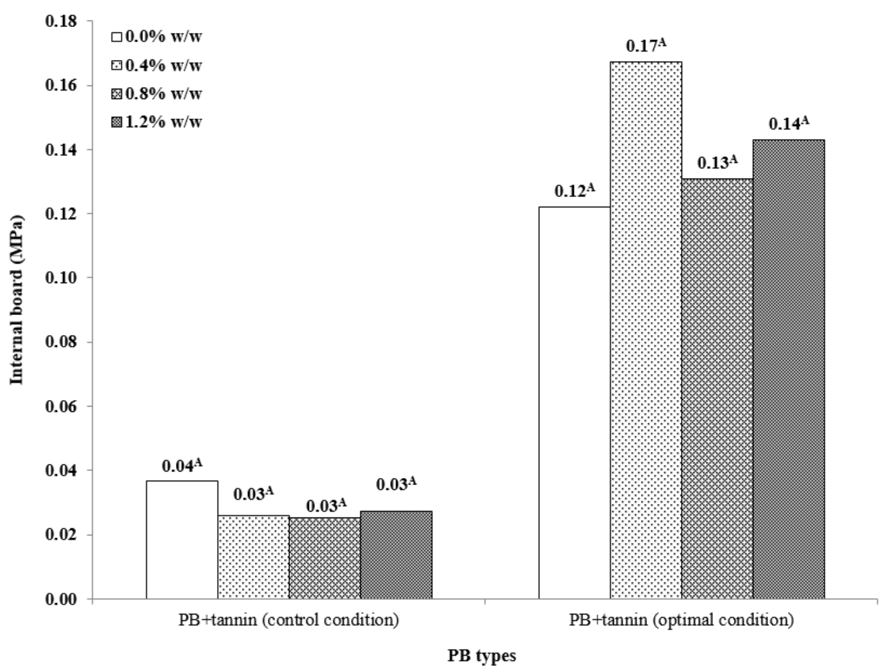

Figure 4 Internal board valuefor particleboard (PB) after the addition of different levels of tannin.

Note : * Values with different letters are statistically significant ($p \leq 0.05$).

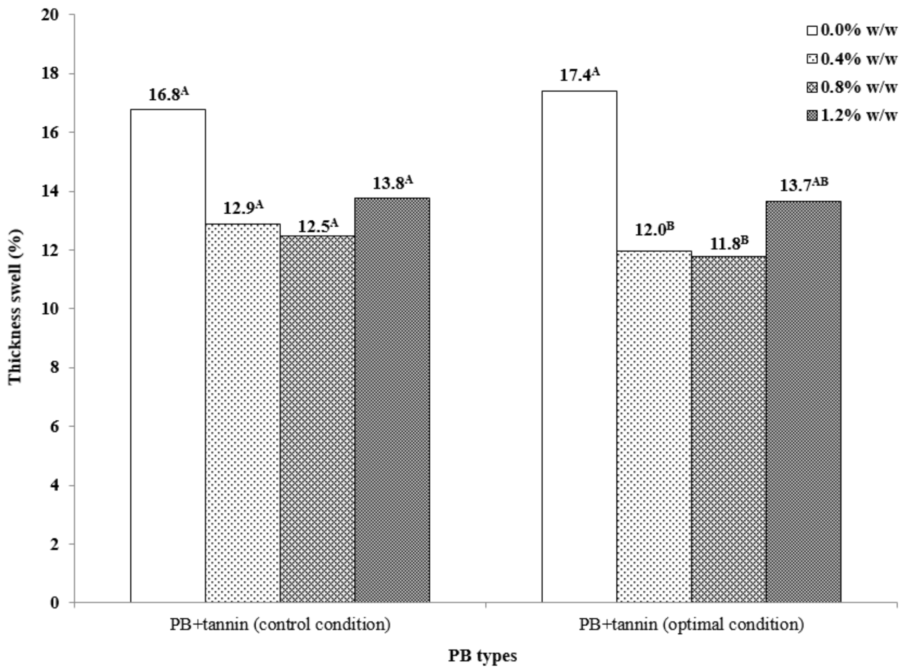

Figure 5 Thickness swelling (TS)of particleboard (PB) adding different levels of tannin.
Note : * Values with different letters (A – B) are statistically significant ($p \leq 0.05$).

Effect of tannin addition on the reduction of formaldehyde content of particleboard

The lowest formaldehyde content was 13.39 mg/100 g board OD weight and 13.52 mg/100 g board OD weight, for a panel with 1.2% tannin addition, extracted under control and optimal conditions, respectively. From the results, it can be deduced that tannin can reduce the formaldehyde content in particleboard. The higher the amount of added tannin, the lower the formaldehyde content in the particleboard. However, the tannin properties might not influence the formaldehyde content, but tannin loading does effect the overall particleboard properties, especially, adhesion in the particleboard structure. From Figure 7 and Table 4, adding tannin extracted using control and optimal conditions can decrease the formaldehyde content, measured according to the European Standards, when compared with the E2 levels (8-30 mg/100 g board OD weight). However, Eom *et al*. (2006) and Kim *et al*. (2006) found that adding tannin to particleboard can reduce the formaldehyde emission by 60%, 14% and 10% when using the desiccator, small chamber, and flask methods, respectively. The tremendous variability among different approaches is apparent in Table 5 (Sundman *et al*., 2007).

Table 5 Comparison of formaldehyde emission from different approaches.

Method Name	Standard	Sample Loading Factor	Class	German Regulations	Source
Chamber	EN 717-1	1 m^2/m^3	E1	≤ 0.1 ppm	Roffael, 2006
Gas analysis	EN 717-2	0.4 x 0.05 m	E1	≤ 3.5 mg/h*m^2	
Flask method	EN 717-3	0.025 x 0.025 m, 20 g	E1	no official limit or 4 mg/kg dry board	Sundman *et al.*, 2007
Desiccator	JIS A 1460	0.18 m^2	F**	≤ 1.5 mg/l (average) 2.1 mg/l (maximum)	Roffael, 2006; Eom *et al.*, 2006
Perforator	EN 120	0.025 x 0.025 m,110 g	E1 E2	≤ 8 mg/100 g ≥ 8–30 mg/100g	Roffael, 2006; Eom *et al.*, 2006
CARB (Phase II)	-	-	-	0.09 ppm for PB	Salem *et al.,* 2012

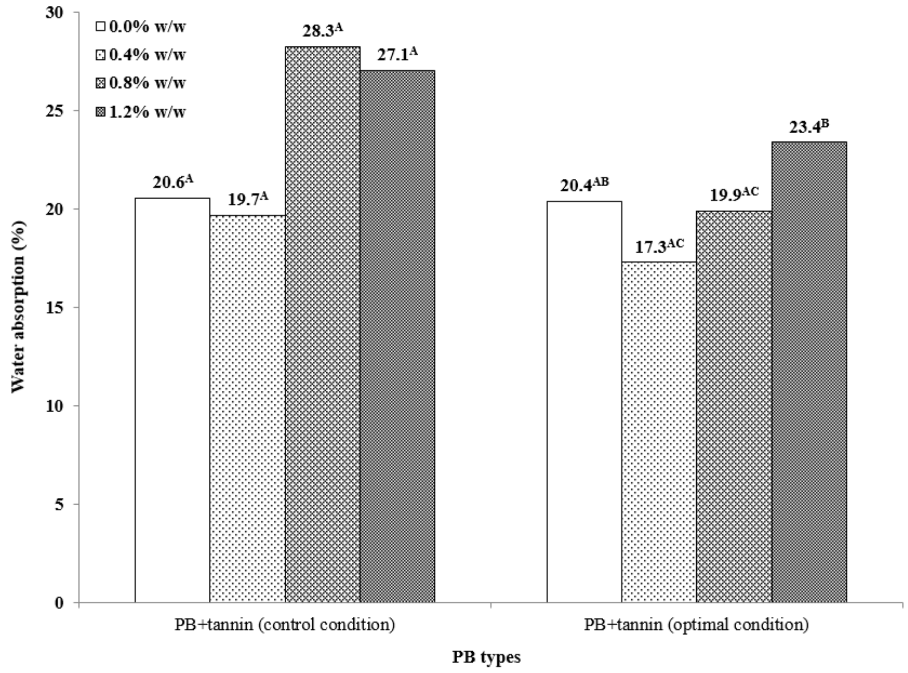

Figure 6 Water absorption value for particleboard (PB) after the addition of different levels of tannin.
Note : * Values with different letters (A – C) are statistically significant (p ≤ 0.05).

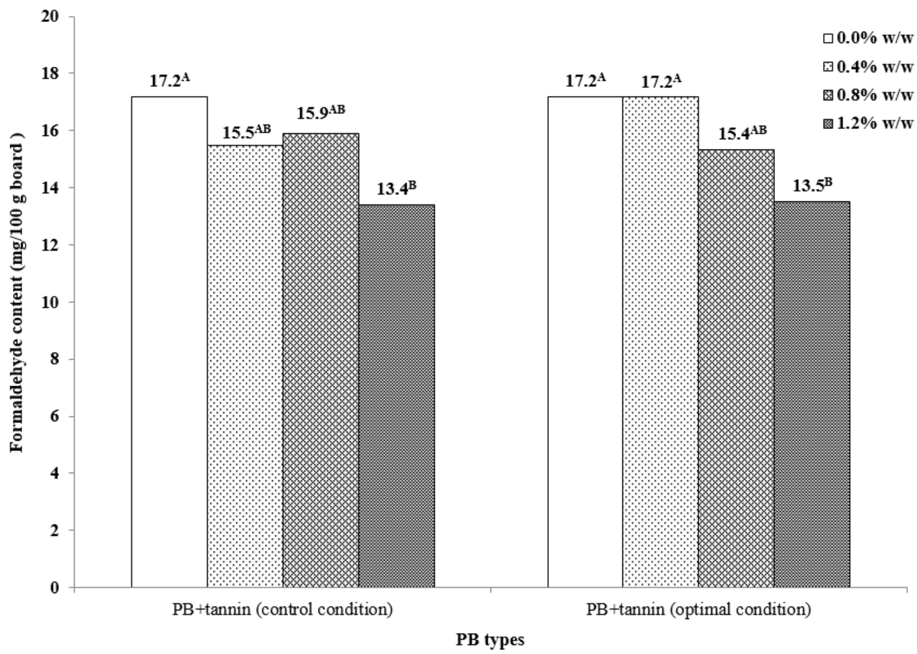

Figure 7 Formaldehyde content in particleboard (PB) after the addition of different levels of tannin.
Note : * Values with different letters (A – B) are statistically significant ($p \leq 0.05$).

CONCLUSION

The loading and properties of tannin, extracted from optimal conditions, had an influence on the MOR, MOE, and on IB, to a lesser extent. However, the investigation of tannin as bio-scavenger in reducing the formaldehyde content in particleboard, was ambiguous. Tannin loading led to a reduction in the amount of formaldehyde content in particleboard, but its properties may have no influence on the reduction of formaldehyde content. The formaldehyde emission reduction in particleboard could be studied as a future study.

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