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SHORT NOTES

THE TEA LEAVES AS A FILLER FOR UF RESIN PLYWOOD PRODUCTION

JOANNA WALKIEWICZ, JAKUB KAWALERCZYK, RADOSŁAW MIRSKI,
ZUZANNA SZUBERT
POZNAŃ UNIVERSITY OF LIFE SCIENCES
POLAND

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ABSTRACT

The addition of green tea leaves fractions < 0.315 mm as a filler to urea-formaldehyde resin to reduction of emission of formaldehyde from three-layer birch plywood was investigated. Moreover, other properties such as bonding quality and water resistance of plywood were investigated. It was found that green tea had an ability to absorb formaldehyde. This phenomenon was caused by phenolic compounds contained in green tea leaves. The addition of green tea in the amount of 20% and 25% resulted in a decrease in formaldehyde emission from plywood comparing to the reference sample. The bonding quality test showed that the addition of tea leaves as a filler did not affect the strength properties of the tested bond lines.

KEYWORDS: Plywood, formaldehyde, filler, green tea.

INTRODUCTION

In the last 20 years, the production of wood-based materials has increased by 180 in 2000 to over 361 million m^3 in 2020. For the production of most materials, the amino resins are widely applied. These resins contain both free and bound formaldehyde. Free formaldehyde permeates directly to the environment and its emission usually quickly fades away, however, the bound one is released steadily as a result of the degradation of the bond lines, and it intensifies under the influence of change in air humidity and temperature. Products manufactured with wood-based materials can therefore be a major source of formaldehyde released into the indoor environments (Risholm-Sundman et al. 2007, Zhang et al. 2018).

The disadvantageous feature of cured urea-formaldehyde (UF) adhesives is brittleness and tendency to crack because of the internal stresses, especially of the shrinkage type, as well as

thermal and humidity. The effect of these phenomena is the intensified emission of formaldehyde from the adhesives in UF-bonded wood-based materials (Jóźwiak 2004).

Formaldehyde is emitted from wood-based materials to the surroundings. Therefore, from the point of view of hygiene and care for human health, reducing its emissions is an important issue. There are many literature data regarding the reduction of formaldehyde emission (Tab. 1).

Tab. 1: The examples of methods applied to reduce formaldehyde emissions.

Method	Additional results	Reference
Veneer moisture, composition of the adhesives and parameters of the pressing process	-	Jóźwiak 2007
Amine-based compounds addition	-	Maurer et al. 2008
Ethyl acetoacetate addition	-	Dziurka et al. 2014
Nano-SiO_2 addition	physical and mechanical properties improved	Salari et al. 2013
Microfibrillated cellulose addition in LVL	reduced VOC emission	Ayrilmis et al. 2016
Heat treatment (or bake-out)	reduced TVOC (Total volatile organic compounds) emissions of particleboards	Jiang et al. 2017
Pumpkin flour addition	improved mechanical properties	Kawalerczyk et al. 2019a
Hemp flour addition	improved mechanical properties	Kawalerczyk et al. 2020b
Oak bark addition	improvement of bonding quality	Mirski et al. 2020

An interesting way to reduce formaldehyde emissions may be addition of the tea leaves as a filler. The green tea leaves contain mainly polyphenols such as catechins epigallocatechin, epicatechin, galocatechin, as well as flavonoids: quercetin, kaempferol, myricetin, phenolic acids: gallic acid, p-coumaric acid. The leaves are also a source of purine alkaloids (Kania and Baraniak 2011). Moreover, it was previously found that tea catechins are reactive with formaldehyde (Takagaki et al. 2000). Hojo et al. (2000) used green tea catechins as a filler for plywood manufacturing. It led to the reduction of formaldehyde emission. The results suggest that the utilization of catechin as a formaldehyde scavenger is a proper way.

Therefore, the aim of the conducted study was to investigate whether the addition of green tea as a filler to urea-formaldehyde resin will reduce emission of formaldehyde from wood based materials, such as three-layer plywood.

MATERIAL AND METHODS

Materials

For the production of plywood, the birch veneers with dimensions of 320 x 320 mm and a thickness of 1.5 mm were used. Moreover, the commercially available urea-formaldehyde resin with the solid content of 69.6%, gel time at 100°C of 68 s was applied. Rye flour was used as the reference filler in the amount of 20% (based on the solid content of the resin). Green tea leaves, ground to obtain the fractions < 0.315 mm were used at the loads of: 15%, 20%, 25% (based on the solid content of the resin). Tab. 2 showed adhesives formulation.

Tab. 2: Adhesives formulation.

Sample code	Type of filler	Quantity (g/100 g dry weight of resin)	
		Filler	Hardener
REF	Rye flour	20	2
H15	Tea leaves	15	2
H20	Tea leaves	20	2
H25	Tea leaves	25	2

Methods

The resin was applied to the veneers in the amount of 170 $g \cdot m^{-2}$ and the open time was 5 min. Three-layer plywood were pressed using the following parameters: unit pressure of 1.4 MPa, temperature 120°C for 4 min.

The viscosity was determined in accordance with the EN ISO 2555 standard with the use of rotary viscometer Brookfield DV-II + Pro. The determination of the formaldehyde absorption capacity from aqueous solution was performed according to the methodology described in details by Bekhta et al. (2021). Determination of formaldehyde emission was carried out using the "flask method" in accordance with EN 717-3 (1996) (2 repetitions). The bonding quality was tested in accordance with the EN 314-1 (2004) standard (10 repetitions). The results of the bonding quality were subjected to statistical analysis - ANOVA using the Statistica 13.0 program (StatSoft Inc., Tulsa, USA). The delamination study was conducted according to the methodology described in details by Taghiyari et al. (2020).

RESULTS AND DISCUSSION

The effect of the addition of ground tea leaves on the viscosity of the resin is shown in Tab. 3. It was observed that the addition of tea increased the viscosity of the resin. It is especially important since too high viscosity leads to the difficulties in application of the resin evenly enough to ensure the full cover of the veneer surface. A similar phenomenon was observed by Réh et al. (2021), who stated that the viscosity of the glue mixture and the ease of its application are very important issues in the plywood production process. Moreover, in the studies presented by Kawalerczyk et al. (2019b) it was observed that improperly selected parameters of the adhesive mixture may have a negative effect on the adhesive joints. Nevertheless, when compared to the reference sample where the rye flour addition was 20%, the viscosity for the mixture containing 20% of hemp flour was lower. This phenomenon could be caused by the size of the filler particles or their chemical composition (Mirski et al. 2020).

Tab. 3: Viscosity of the resin with various filler addition.

Resin	**Pure**	**REF**	**H15**	**H20**	**H25**
Viscosity (MPa x s)	984	1450	1960	3272	11500

Fig. 1 shows the formaldehyde content in the water solutions with tea powder and reference filler-rye flour. In both cases, a decrease in formaldehyde content was observed. It is worth

adding that a considerably positive effect of tea leaves addition was observed. The tea leaves showed a greater ability to absorb formaldehyde than rye flour. Compared to rye flour since these values were lower after 3 h of incubation at 65°C, for samples with 1, 3 and 5% (by weight) of traditional filler. A similar phenomenon of formaldehyde absorption was observed by Bekhta et al. (2021) in research regarding bark powder use as a filler.

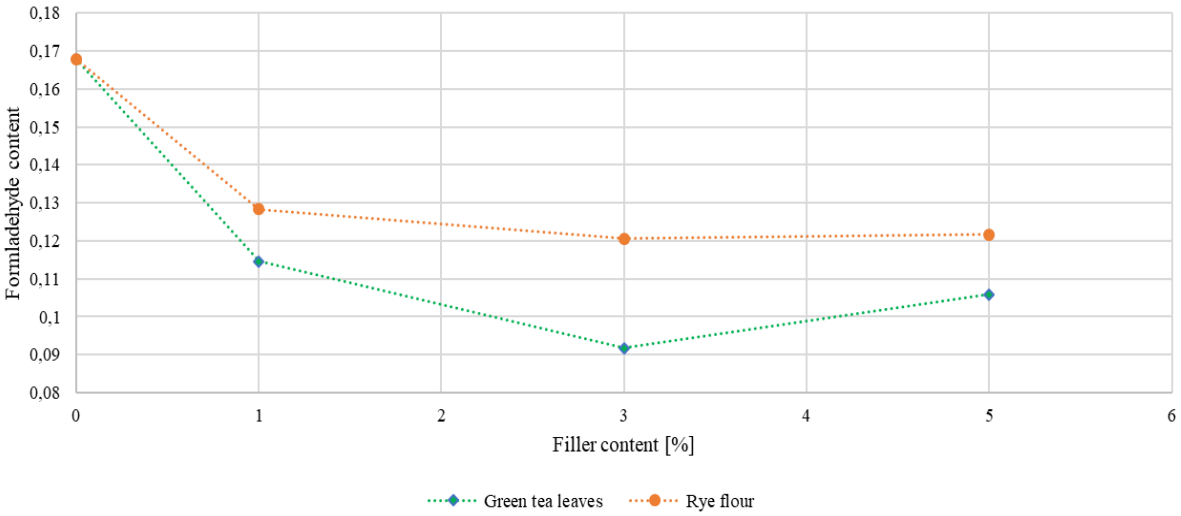

Fig. 1: Formaldehyde content in aqueous solution with 1%, 3% and 5% addition of filler.

In the case of green tea, the presence of chemicals such as e.g. catechins had probably a positive effect on the ability to absorb formaldehyde (Takagaki et al. (2008). These compounds are reactive towards formaldehyde and thus, they show the ability to bond its molecules. Bonding of HCHO by compounds contained in green tea contributes to the reduction of its content in the tested aqueous solutions. The formaldehyde absorption capacity of tea was shown also in the results of emission from the produced three-layer plywood (Fig. 2).

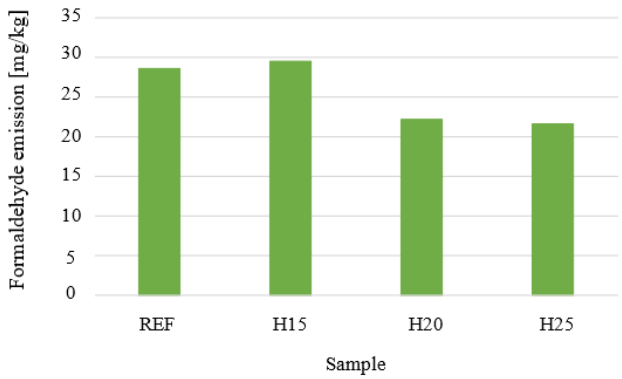

Fig. 2: Results of formaldehyde emission depending on amount of filler.

A significant decrease in formaldehyde emission from plywood was observed in case of 20% and 25% load of green tea. For plywood with an additive of 15%, the emission of formaldehyde was at a similar level as the reference sample. This phenomenon could be caused by uneven

application of resin. Another explanation for this could be that the amount of added filler was too low. Nevertheless, the obtained results confirm the research hypothesis that the green tea may contribute to the absorption of formaldehyde and reduction of its emission.

Tab. 4 shows the results of the delamination tests. No delamination was observed after each of the three cycles. This indicates that replacing rye flour with an unconventional filler did not adversely affect the water resistance of plywood panels.

Tab. 4: Results of the delamination test.

Sample	Number of delaminated samples		
	Cycle I	Cycle II	Cycle III
REF	0	0	0
H15	0	0	0
H20	0	0	0
H25	0	0	0

Fig. 3 shows the results of the bonding quality test. Statistical analysis showed no statistically significant changes. However, there is no reason to reject the null hypothesis about the equality of mean values. In other words, the introduction of a filler as a filler (differing in its content), did not change the bonding quality of the resultant plywood.

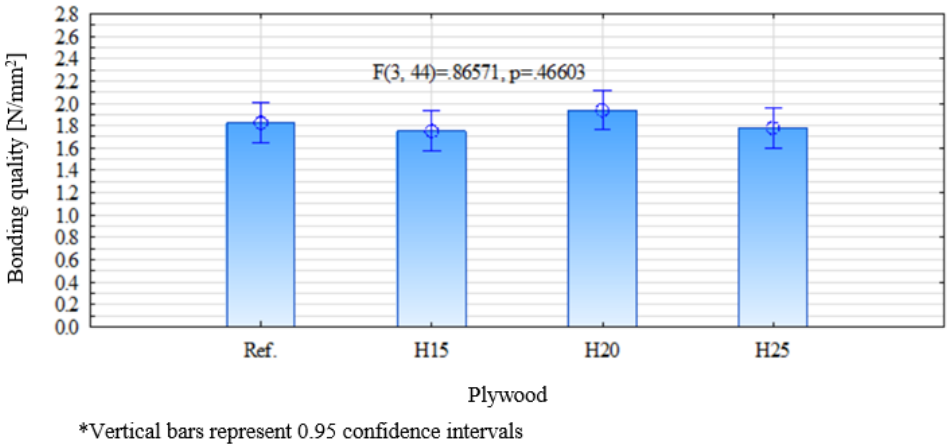

Fig. 3: Results of the bonding quality test *depending on the variant.*

Regardless of the variant, the gluing quality of all plywood has been achieved value greater than 1 $N \cdot mm^{-2}$. This means that the produced plywood panels meet the requirements of the PN-EN 314-2 standard.

CONCLUSIONS

(1) Addition of the ground green tea leaves as a filler to the UF adhesive, caused the increase in the viscosity of the mixture. *(2)* It was found that green tea showed a greater ability to absorb formaldehyde than the commonly applied rye flour. This phenomenon was caused by bonding of formaldehyde by chemical/phenolic compounds contained in green tea. *(3)* The 20 and 25% addition of green tea resulted in a significant decrease in formaldehyde emission from plywood

comparing to the reference sample. *(4)* The delamination tests performed showed no delamination of the plywood glued with green tea-filled adhesive which indicates no adverse effect on the water resistance of plywood panels. *(5)* The evaluation of the bonding quality did not show statistically significant differences. The addition of tea leaves as a filler did not affect the strength properties of the tested bond lines.

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JOANNA WALKIEWICZ*, JAKUB KAWALERCZYK, RADOSŁAW MIRSKI,
ZUZANNA SZUBERT
POZNAŃ UNIVERSITY OF LIFE SCIENCES
FACULTY OF FORESTRY AND WOOD TECHNOLOGY
DEPARTMENT OF MECHANICAL WOOD TECHNOLOGY
WOJSKA POLSKIEGO 28, 60-637 POZNAŃ
POLAND
*Corresponding author: joanna.walkiewicz@up.poznan.pl