



## INVESTIGATION OF FORMALDEHYDE EMISSION FROM LIGHT-WEIGHTED MULTI-STRUCTURE WOOD PRODUCTS WITH POLYSTYRENE GRANULE

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### ABSTRACT

Lightweight composite wood products are ones whose demand is increasingly grow year by year. Accordingly it is predicted that it has the highest rate of growth in above mentioned industry during upcoming decades. This issue is becoming more important due to shortage of wood resource, competition to provide this renewable materials, and finally economical components in production of lightweight products. Polystyrene is less expensive and very light product (with density of 30 kg/m<sup>3</sup>). Use of this material in wood industry is continuously growing to produce competitive products. Synthetic resin containing formaldehyde is used in the production of wood composite products. Formaldehyde that is toxic composition and high carcinogenicity is always regarded as human and environmental concerns in the application of these products. This study aims to investigate the rate of formaldehyde release by polystyrene granules based on the main processing components of producing three-layer particleboard.

In this test, 8 treatments with three replications were defined from the ratio of polystyrene to wood (two levels); the percentage of adhesive (two levels); the mixing of the surface layer and middle layer (two levels). The density of produced woods is chosen 0.5 gr/cm<sup>3</sup>. The press components were considered constant for comparability (temperature: 160° C, pressure: 30 kg / cm<sup>2</sup>, time: 6 min). After production, boards are calibrated according to the standard of ISO 12642-4 in condition of acclimatization and are tested to determine rate of formaldehyde release in drying method. The most optimum treatments are selected based on ANOVA analysis.

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### Introduction

One of the main concerns in wood industries is the supply of raw wood material [1, 2, 3]. Currently, producers of wood multi-structures and the industries that extract their required energy from wood are closely competing around the world [3, 4, 5, 6]. As such, it is important to consider measures to address, at least, part of the existing concerns via various solutions such as lower consumption of wood in producing wood products. On this basis, production of light-weighted multi-structure

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wood products by modifying production processes and raw materials or changing production technologies has gained a great of attention [6, 7]. The idea of producing light-weighted composite products dates back to 2000, although the production of lightweight wood products was also experienced in furniture manufacturing industries in the 1950 due to lack of wood resources because of the World War II [3]. Today, lightweight composite products refer to the products with any density below  $0.5 \text{ gr/cm}^3$  [3, 8, 9]. This classification was first presented by Tuman (2008) and later on accepted by other researchers [5, 6]. Since 2008, production of multi-structure wood products in Europe has decreased by 13% while the demand for such products has increased by 4%; predictions indicate the fastest rate of growth in global production of multi-structure wood products in 2020 [4]. Light-weighted multi-structure wood products can largely contribute to reduced consumption of wood material by reducing the product weight by incorporating such materials as foams [10, 11, 12]. Of other properties of this type of multi-structure wood products, one may refer to lower finished cost of some of these products and easier application of them [4, 13]. On this basis, products with foam interlayers, sandwich panels, light-weighted composite panels, and tubular products can be classified as lightweight multi-structure wood products [14, 15]. Aminoplast resins are the main binding agents use in the process of producing compact wood sheets. Among aminoplast resins, urea-formaldehyde resin is most primarily and frequently used [8]. Even though this resin is associated with limitations when exposed to moisture, but polymerization at low temperature, lower cost, shorter polymerization time, and colorless nature are among its advantages [16, 8, 17]. Formaldehyde emission is referred to as the main disadvantage associated with the application of this resin [18]. Urea-to-formaldehyde ratio, resin consumption rate in wood product manufacturing process, necessity or unnessesity of using a catalyst, pressing parameters during the production process, and panel operation conditions are among the factors affecting formaldehyde emission out of the panel [9]. According to standard code on Control of Substances Hazardous to Health (COSHH), formaldehyde is among the most important pollutants of indoor air. Inside human body, formaldehyde tends converts to formic acid which then permeates into the blood and increases its acidity [19]. Formaldehyde is not detectable via smelling at concentrations below 0.1 ppm. At concentrations between 0.1 and 0.5 ppm, the presence of formaldehyde can be detected via smelling by sensitive individuals who exhibit minor eye, nose, and throat disorders. When ambient concentration of formaldehyde reaches 0.5 – 1 ppm, it causes eye, nose, and throat irritations in most people, and exposure to formaldehyde at concentrations above 1 ppm may end up with severe disorders [20]. On this basis, US Occupational Safety and Health Administration (OSHA) reduced 8-hour occupational limit of exposure to this material to 0.75 ppm and its short-term exposure limit to 2 ppm. Furthermore, US National Institute for Occupational Safety and Health (NIOSH) has declared 0.1 ppm as carcinogenesis threshold of formaldehyde for human. In the study by Tang et al. (2009), it was reported that Chinese workers at factories who were exposed to high levels of formaldehyde had significantly lower numbers of white and red globules as well as blood platelets than a control group who were not exposed to that chemical [17]. Woogan et al. (2009) undertook studies on the impact of formaldehyde on human blood parameters and achieved significant new findings on the associated risks with formaldehyde to the human health. Their report indicated possible disorders in blood cells under the effect of formaldehyde [21]. In the United States, NIOSH has set allowable threshold of exposure to formaldehyde to 0.7 ppm, while the corresponding limit in Sweden and German is set to 1 ppm [22]. Other countries have similar rules in this respect. Finally, International Agency for Research on Cancer (IARC) has classified formaldehyde under potential carcinogens in humans. Recently, remarkable progress has been achieved in the scope of the technology of producing light-weighted multi-structure products using foaming agents. One of the widely applied foaming agents in various industries in polystyrene. Chemical formula of polystyrene is  $n(\text{C}_6\text{H}_5\text{CH}:\text{CH}_2)$ ; it is a member of the family of thermoplastics and classified as a chemical foam [9]. This substance is made up of several phenyl benzene cycles connected to one another. Application of polystyrene for reducing the weight of wood multi-structure wood products throughout special processes has been considered. The most important processes in this respect involve the production of products with polystyrene interlayers with wooden surface layers. In the meantime, these products are associated with many problems in practice; among other, one may refer to the need for special tools and connections, sever strength weakness of the interlayer, and the need for deployment of expensive technologies. As another method, foamed polystyrene granules mixed with wood particles are incorporated into the interlayer in multi-structure products. Numerous research works have investigated various properties of layered products light-weighted with polystyrene foam, honeycomb layers, and sandwich panels (2, 3, 10, 20, 25, 26, 32, 34). One of the most recent research in this respect is that performed by Shalbafan, Veling and Tuman (2013) who investigated the effect of continuous press at 130 and 160°C on physical and mechanical properties of layered products light-weighted with polystyrene foam. Their results indicated lower internal cohesion and bending strength of the products subjected to high-temperature pressing, even though the temperature change imposed no effect on the products' screw withdrawal strength [9]. Also in Iran, application of polystyrene in multi-structure product manufacturing has been considered. In a research, Enayati and Mersad (2002) investigated the use of the waste resulted from polystyrene-made cups in manufacturing particle board of  $0.6 \text{ gr/cm}^3$  in density with the aim of reinforcing the binding connection and improving physical and mechanical properties of the particle board. Their results showed improved bending strength, internal cohesion, and water absorption properties of the board by adding polystyrene waste chips to the product. The best treatment was the one containing binder polystyrene at 10% and 15%, respectively [23]. In another research, Mir, Farokhpayam, and Nazerian (2016) studied the effect of urea-formaldehyde to melamine-formaldehyde binder ratio (UF/MF) on properties of light-weighted particleboard with expansive polystyrene. Results of the research showed that, with

increasing the percentage of polystyrene the granule, tensile strength and water absorption capacity of the product decreased, while its bending strength increased. In all treatments, increased UF/MF ratio enhanced the resultant mechanical characteristics. They attributed the improved water absorption capacity of the product to the presence of nonpolar texture of polystyrene, and related improved mechanical properties to higher compaction of the internal layer due to lower volumetric density of polystyrene [8]. Considering the importance of formaldehyde emission, the present research investigates formaldehyde emission from multi-structure wood products light-weighted with polystyrene granules.

**Materials and Methods**

**Materials**

For the purpose of the present research, three main raw materials were used, including UF resin, wood particles, and polystyrene granule. The UF resin was procured from Samed Adhesives Company. The required wood particles were obtained from industrial production process of North Adhesive Industry Co. for the two layers (surface layer and interlayer) of the multi-structure product. Polystyrene granules were procured from Banyar Polymer Petrochemical Co. and converted to foamed granules of 2-3 mm in diameter through Styrofoam industrial production process. Moreover, some of the required additives for the tests (e.g. paraffin emulsion and ammonium chloride) were procured from local retailers at Tehran. In order to perform formaldehyde emission test, S2100 UV-visible spectrophotometer (UNICO, USA) with an accuracy of 2 nm and wavelengths of 200-1000 nm was used (at Barzanj Azmoon Laboratory).

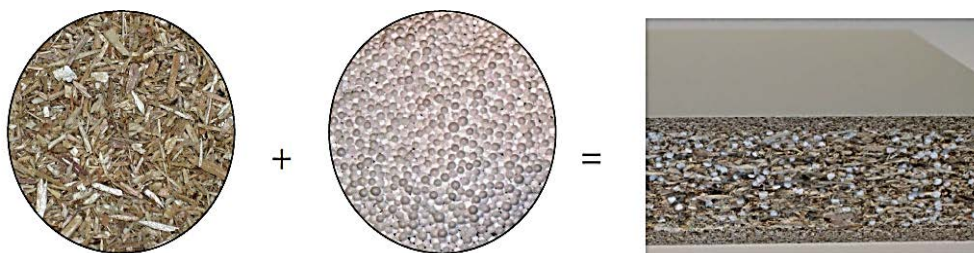
**Methods**

The following treatments were considered for this test:

**Table 1.** treatments of test

Layers ratio (%)		Percentage of resin (%)		Polystyrene/wood particle (%)		Code
40/60	30/70	12	10	5	1	
	*		*		*	1
*			*		*	2
	*	*			*	3
*		*			*	4
	*		*	*		5
*			*	*		6
	*	*		*		7
*		*		*		8

The mentioned three components were selected based on their effectiveness on formaldehyde emission. Layer ratio was considered based on the weight balance between surface layer (30 and 40%) and interlayer (60 and 70%). In order to come with comparable results, the same pressing parameters were considered for all treatments. Treatment level was determined based on pretests. Three repetitions were considered for each treatment, so that a total of 36 boards were produced (inclusive of control samples). Optimal treatments were selected based analysis of variance (ANOVA) at a significance level of 99%. Assumptions of the test (constant parameters were as follows: sample density: 0.5 gr/cm<sup>3</sup>, type of polystyrene granules: expand polystyrene (EPS), moisture contents of surface layer and interlayer: 14% and 8%, respectively (total moisture content: about 10%), dimensions: 2.5 × 50 × 50 cm, polystyrene granule dimensions: 2-3 mm, dimensions of surface layer particles according to mesh numbers 40 and 60 (0.2 – 0.4 mm), thickness of interlayer particles: 0.6 – 0.8 mm (with an elongation factor of 15 – 20), and pressing parameters (temperature: 160°C, pressure: 30 kg/m<sup>2</sup>, pressing time: 6 min). On this basis, weight balance of surface layer and interlayer was performed and appropriate amount of polystyrene granule was added based on the wood particle composition by weight. Once finished with weight determination, particles were binded using an experimental binder at the specified ratio for each treatment. After binding, the particles were mounted in a formed wood mold and pressed (Figure 1).



**Figure 1.** light weight wood based panel product with polystyrene granules (nolte company)

Following the pressing cycle, the produced boards were climatized for 24 hours (at 20°C and relative humidity of 65%). Subsequently, the boards were cut according to the test standard.

For the sake of formaldehyde emission, desiccator method was used according to ISO 12460-4 (2008). In this method, the amount of released formaldehyde in a given volume of water in 24 hours is measured. For the sake of this test, 150 g ammonium acetate (CH<sub>3</sub>COONH<sub>4</sub>) was dissolved in 800 mL of water in a 1000 mL-volumetric flask. Then, 3 mL of glacial acetic acid (C<sub>2</sub>H<sub>4</sub>O<sub>2</sub>) and 2 mL of acetyl acetone (pentane-2,4-dione, C<sub>5</sub>H<sub>8</sub>O<sub>2</sub>) were added to the solution, and finally, the flask volume was fully filled with distilled water (12, 13). The obtained solution was then used for test. Absorbed formaldehyde in water was used for determining the emission from the board using the photo-spectrometer at a wavelength of 412 nm with cells of 50 mm in length. Dimensions of test samples was 10 × 15 cm with independent thickness for sample. Three repetitions were considered for each treatment (Table 2).

**Table 2.** Test results of formaldehyde emission and Duncan's grouping

Absorption Average	Absorption number			Formaldehyde concentration (ppm or mg/ml)	Code
	Repeat 1	Repeat 1	Repeat 1		
0.26	0.26	0.26	0.25	0.989 <sup>a</sup>	1
0.26	0.26	0.27	0.26	0.992 <sup>a</sup>	2
0.27	0.27	0.28	0.26	0.966 <sup>ab</sup>	3
0.27	0.26	0.27	0.28	0.959 <sup>ab</sup>	4
0.18	0.18	0.17	0.19	0.666 <sup>c</sup>	5
0.18	0.18	0.18	0.18	0.657 <sup>c</sup>	6
0.16	0.16	0.16	0.17	0.701 <sup>b</sup>	7
0.16	0.16	0.17	0.16	0.718 <sup>b</sup>	8
0.32	0.32	0.32	0.33	1.101 <sup>d</sup>	treatment

Measurements were carried out based on Hunch reaction wherein blue formaldehyde reacts with ammonium ions and acetyl acetone to produce di-acetyl-di-hydrolutidine (DDL). At the wavelength of 412 nm, DDL exhibited peak absorption [24, 25]. Calibration curve is obtained from standard formaldehyde solution with its concentration determined from iodometric titration. Formaldehyde emission was calculated as concentration in mgr/liter from Equation (1):

$$\rho(\text{HCHO}) = (V_0 - V) \times 15 \times c(\text{Na}_2\text{S}_2\text{O}_3) \times 1000/20 \quad \text{Equation (1)}$$

$c(\text{Na}_2\text{S}_2\text{O}_3)$ : Sodium thiosulfate concentration (mol/L)

$V_0$ : Volume of thiosulfate titration solution consumed for titrating the test sample (mL)

$V$ : Volume of thiosulfate titration solution consumed for titrating the control (mL)

Once finished with determining the formaldehyde, concentration of the formaldehyde emitted from each sample was calculated via Equation (2):

$$G = f(\alpha_d - \alpha_b) \times 1800/A \quad \text{Equation (2)}$$

$\alpha_a$ : Background formaldehyde solution absorption

$\alpha_d$ : Test samples formaldehyde solution absorption

$A$ : Combined area of test samples (cm<sup>2</sup>)

$f$ : Formaldehyde calibration curve slope (mg/L)

Emitted formaldehyde from the samples was measured and reported with an accuracy of 0.001 mg/mL. Absorption number of each sample was read for three times using the spectrophotometer device.

### Results and Discussion

Results of the research showed that, with increasing the content of polystyrene granule in the composition of the manufactured product, free formaldehyde emission decreases, so that with increasing the fraction of polystyrene granule from 1 to 5%, concentration of formaldehyde exhibited a decrease from 0.989 ppm to 0.657 ppm. This is while the constant concentration of 1.101 ppm was recorded for control (Table 2 and Figure 2).

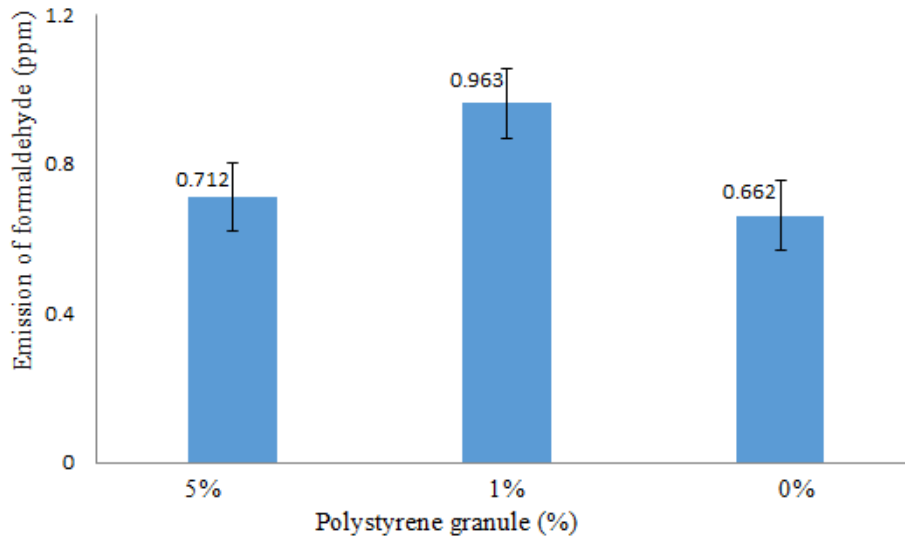


Figure 2. The release of formaldehyde from treated and control sample

Among the treatment parameters, polystyrene granule fraction was significant at 99% level. Surface layer-to-interlayer mixing ratio exhibited no significant effect on formaldehyde emission. In all of the considered cases, formaldehyde emission from treated samples was lower than that from control. Table 2 also distinguishes five classes based on Duncan classification method. According to Duncan mean classification, polystyrene granule consumption ratio, polystyrene granule-to-binder ratio, layers ratio, binder-layers ratio, and treatment sample were categorized into groups. On the basis of Duncan classification, formaldehyde emission decreases with increasing polystyrene granule ratio. Investigating Figure 3, it seems that formaldehyde emission is a function of polystyrene granule ratio; so that, in general, emission from the samples containing higher polystyrene granule (5%) is lower at both binding levels. At 1% granule level, it was observed that formaldehyde emission increased from 0.958 ppm to 0.975 ppm as binder consumption increased from 10% to 12%, while the corresponding figures at 5% granule level were 0.666 ppm and 0.712 ppm, respectively.

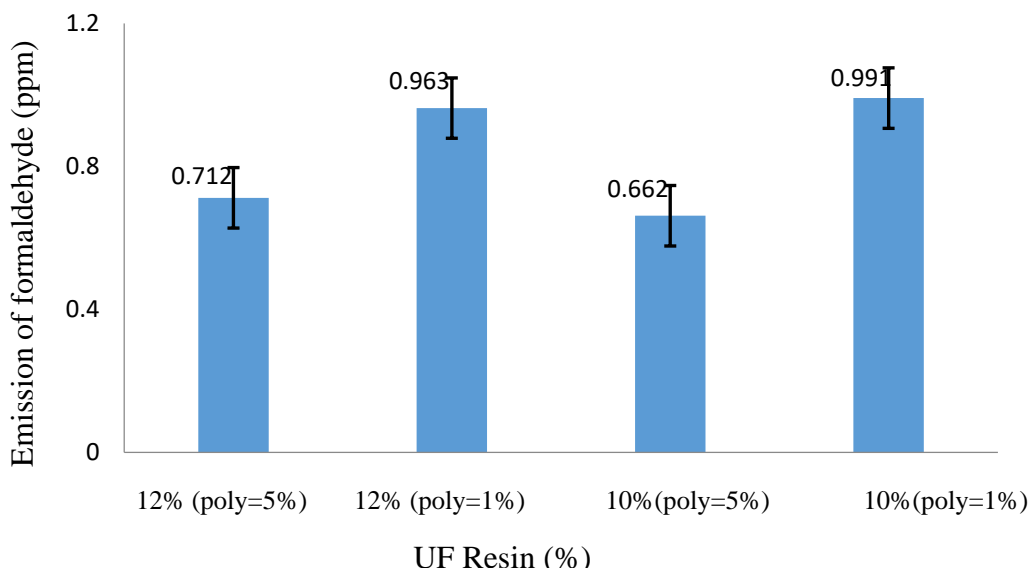
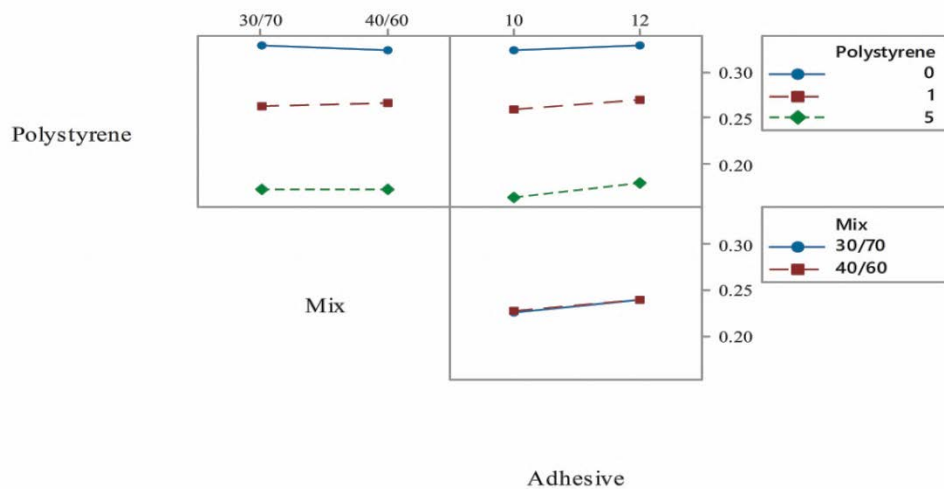


Figure 3. the effect of resin ratio on emissions of formaldehyde

The increase in formaldehyde emission at polystyrene granule ratio of 1% was mostly affected by binder consumption. Investigating interactive effects of test parameters, it seems that, the increase in polystyrene granule ratio is the most important factor affecting formaldehyde emission, so that changes in layers ratio and binder consumption are functions of changes in polystyrene granule ratio (Figure 4).



**Figure 4.** The interaction of the components of the emission of formaldehyde treatment

Furthermore, with increasing the consumption of polystyrene granule from 1 to 5%, formaldehyde consumption was observed to decrease by 30%, while the decrease in formaldehyde emission in the boards containing granule at 1% was only 11%, as compared to control samples. Formaldehyde emission from the manufactured board depends on two parameters, namely remaining free formaldehyde content of the board and UF binder hydrolysis during the operation phase [26]. At a given binder consumption level, an increase in density of the product or the use of lighter raw materials affects the product compaction, thereby limiting the availability of air and moisture to particle surfaces, contributing to reduced formaldehyde emission rate [27]. Due to its foaming effect (up to 40 times as its initial volume) and low density ( $0.03 \text{ g/cm}^3$ ), incorporation of polystyrene granule enhances the compaction of final product [28, 29]. Net result of using less wood material in a lightweight multi-structure product (as compared to other products with intermediate density) was reduced formaldehyde emission from the multi-structure product [30, 31]. Higher compaction of treated samples which ended up with better inter-particle connections has been referred to as another root cause of such behavior [9, 32]. Firmer adhesion of binder due to the obtained higher compaction enhances the connections while reducing the level of free formaldehyde in the products [33]. In a research, it has been reported that, upon FTIR spectrometry analysis, increased compaction of the board results in enhanced extended cross-connections at wavenumber  $3440 \text{ cm}^{-1}$  due to hydroxyl groups of the resin, thereby reducing the hydrolysis. In the present research, the absorption peak at  $3350 \text{ cm}^{-1}$  was attributed to NH connections, the one at  $1663 \text{ cm}^{-1}$  was attributed to  $-C=C-$  bonds, the one at  $1547 \text{ cm}^{-1}$  was due to CO, and the one at  $1253 \text{ cm}^{-1}$  was attributed to the bonding group CN in the resin [19]. Faster hydrolysis of the resin within a shorter period of time results in increased formaldehyde emission during the consumption [21]. Increased compaction of the board reduces air and water availability for the bonds, thereby affecting formaldehyde emission at a given binding level [34]. The results of study show that with increase of polystyrene granules in the composition of produced materials, rate of free formaldehyde release is reduced. Accordingly when the amount of polystyrene granules is increased from 1 to 5, the concentration amount of formaldehyde is reduced from 0.989 ppm to 0.657 ppm, while recorded concentration is 1.101 ppm for control sample. When assessing interaction of test components, it seems that increase of polystyrene is considered as main factor effective on the rate of formaldehyde release. Furthermore, with the increase of polystyrene granules from 1 to 5, release of formaldehyde is reduced by 30%, while reduction in formaldehyde release between boards with 1% granule in compared to control sample is just 11%.

### Conclusion

Results of the present research showed that, formaldehyde emission from light-weighted multi-structure wood products with polystyrene is also directly related to resin consumption. On the other hand, with increasing polystyrene granule-to-wood ratio in the constructed test samples, formaldehyde emission decreased. Indeed, in different treatments, formaldehyde emission acted as a function of the content of polystyrene. Increased compaction of the product due to low density of polystyrene at a given weight of raw material was identified as the most important cause of the mentioned observations. Mixing ratio of surface layer with interlayers exhibited no significant effect on free formaldehyde emission. Increase in product's compressibility due to low density of polystyrene in a specified weight of raw material is considered as the most important factor of this component. The mixing ratio of the surface layer to the middle layer did not show significant effect on the formaldehyde release

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