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Isıl İşlemin Yongalevhalardaki Formaldehit Emisyonuna Etkisi

Effect of Heat Treatment on Formaldehyde Emission in Particleboard

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Öz

Emprenye işlemine alternatif olarak düşünülen, insan ve çevre sağlığına zararlı kimyasallar ilave edilmeksizin uygulanan ısıl işlemin odun esaslı levha ürünleri için birçok avantajı ve dezavantajı bulunmaktadır. Bu ürünlerinin üretiminde çeşitli avantajları nedeniyle önemli ölçüde kullanılan formaldehit esaslı reçineler üretim esnasında ve sonrasında insan sağlığı ve çevre için zararlı olan formaldehit ayrışmasına neden olmaktadır. Formaldehit emisyonunun azaltılması için birçok çalışma uygulanmaktadır. Isıl işlemin farklı amaçlarla uygulanmasından dolayı bu çalışmalarda rastlanılmamıştır. Bu çalışmada, ısıl işlemin kayın (*Fagus orientalis* Lipsky.) odunundan üretilen 3 tabakalı yongalevhaların yüzeylerinden açığa çıkan formaldehit emisyonu oranlarına etkisi araştırılmıştır. Bu amaç için, farklı ısıl işlem sıcaklığı ve süresine göre üç grup oluşturulmuştur. Isıl işlem uygulandıktan sonra, gruplardaki levhaların, alt, orta ve üst tabakaları birbirlerinden ayrılmış ve her bir tabakanın formaldehit emisyonu oranı ısıl işlem uygulanmayan kontrol gruplarıyla karşılaştırılarak değerlendirilmiştir. Sonuç olarak, ısıl işlem ile yongalevhanın formaldehit emisyonunun üst, orta ve alt tabakalarda değişim gösterdiği tespit edilmiştir. 150°C ısı ile muamele edilmiş üst tabakalardaki formaldehit molekülleri sıcak taraftan soğuk tarafa doğru akmıştır. Isı ile muamele edilmiş tabakalardaki formaldehit emisyonu, işlem zamanı arttıkça azalmıştır.

Anahtar Kelimeler: Formaldehit emisyonu, Yongalevha, Isıl işlem, Moleküler iletim

Abstract

Heat treatment was applied without addition of chemicals harmful to human and environmental health, which are considered as an alternative to the impregnation process has many advantages and disadvantages for the wood based panel products. Formaldehyde has been linked to human health and environmental problems during and after manufacturing. Many studies are applied to reduce the formaldehyde emission. Because of the different applications of heat treatment, it has not been found in these studies. In this study, the effects of heat treatment on the formaldehyde emission rate released on the surfaces of beech (*Fagus orientalis* Lipsky.) three-layer particleboard were investigated. For this purpose, three groups were formed according to different time and temperature of heat treatment. After heat treatment, the lower, upper and middle layers of panels in the groups were separated then the formaldehyde emission rates from these layers were determined and compared with the control groups were not applied heat treatment. Consequently, the formaldehyde emission rates from the particleboard varied by layer (the lower, upper and middle) with heat treatment. The formaldehyde molecules in the upper layers treated at 150°C flowed from the hot side to the cold side. The formaldehyde emission rates in the heat-treated layers decreased with the time of treatment.

Keywords: Formaldehyde emission, Particleboard, Heat treatment, Moleculer conductivity

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1. Introduction

The importance of wood composite products has increased due to the increasing demand for wood products and the reduction in the quantity and quality of the raw material. The production of particleboards in 2015 were 21 million m^3 for Chine, 16 million m^3 for U.S.A., 7 million m^3 for Germany and 4 million m^3 for Turkey (FAO, 2016). The increased production of these materials have in turn increased use of adhesives in the wood products industry (Wålinder, 2000; Aydin et al., 2010). Formaldehyde-based resins are significantly used in the wood-based panel industry because of various advantages and excellent performance (Colak, 2002; Marutzky, 1994). Urea formaldehyde resins are the most widely used adhesives in the wood-based panels such as particleboards, medium density fiberboards and plywood due to provide high reactivity, low cost and excellent adhesion to wood (Costa et al., 2013). However, formaldehyde released from the panels manufactured with formaldehyde-based adhesives such as urea formaldehyde resins poses significant environmental health risks and this release can take years (Colak, 2002; Marutzky, 1994). Exposing to formaldehyde for short and long terms causes problems in human health such as eye, nose, and throat irritation and cancer (Salem et al., 2011). It is stated that the formaldehyde emission decreased with the increase in pressing temperature of particleboards (Brinkman, 1978; Roffael, 1982). The content of formaldehyde emission depends on various factors. These factors are mole ratio of F/U, resin level, catalyst level and composition, moisture content and distribution prior to pressing, board post-treatment, and duration of storage before use (Petersen, 1973; Meyer et al., 1980; Roffael, 1975; Sundin and Hanetho, 1978; Wang and Gardner, 1999; Park et al., 2006; Que et al., 2007). Myers (1984) stated that the variables such as resin synthesis details, cure catalyst, press time and temperature, moisture content, board conditioning or aging can strongly influence formaldehyde emission and the mechanical properties of particleboard. Formaldehyde emissions are also significantly affected by pressing conditions (Jiang et al., 2002). Wolcott et al. (1996) studied the effects of pressing variables on formaldehyde and methanol emissions from UF-bonded particleboard with a mixture of Douglas-fir and southern pine furnish. As a result of their study, formaldehyde emissions increased with increasing pressing time, moisture content, platen temperature, resin level, and formaldehyde-to-urea (F/U) mole ratio. Moreover, the additional high-frequency heating during pressing significantly decreased the formaldehyde emissions in the particleboards (Roffael, 1982).

Petinarakis and Kavvouras (2006) studied that effects of the production variables on formaldehyde emission from particleboards. They found that the release of formaldehyde from particleboards of low formaldehyde emission type, glued with urea-formaldehyde glues, can be further significantly decreased though the optimization and monitoring of the production process variables such as the pressing temperature, the panel density and the moisture content of the wood-glue mixture before the hot pressing.

Many studies are applied to reduce the formaldehyde emission in literature. However, due to the different applications of heat treatment, it has not been found in these studies. In this study, the effects of heat treatment on the formaldehyde emission rate released on the surfaces of beech (*Fagus orientalis* Lipsky.) three-layer particleboard were investigated.

2. Materials and Methods

Eastern beech (*Fagus orientalis* Lipsky) wood particles were used in the manufacture of particleboards. They were chipped using a hacker chipper before the chips were reduced into smaller particles using a knife ring flaker. First, the wood particles were screened using a horizontal screen shaker. The chips that pass through a 3 mm mesh screen and leave on a 1.5 mm mesh screen are classified in the middle layer and the chips that pass through a 1.5 mm mesh screen and leave on a 0.5 mm mesh screen are classified in the outer layer for use. After these processes, particles were dried using a lab-customized hot air-dryer at 110°C to 3% moisture content. It was used urea formaldehyde resin with a solid content of 55%. Based on oven-dry particle weight, 8% and 10% resin were applied using an atomizing spray gun for the core and face layers, respectively.. The ratio of the face thickness to the total thickness of a panel known as the shelling ratio was 0.40 for all samples. 25% solution of ammonium chloride (NH_4Cl) as a hardener was added at 1% in oven-dry-weight basis to resin.

Boards were manufactured at a pressure of 23-25 kg cm^2 at 150°C for 9 min. The dimensions and target density of particleboards were 43 cm × 43 cm × 1.8 cm, and 0.70 g cm^3, respectively. After pressing, panels were conditioned at a temperature of 20°C and 65% relative humidity for three weeks. Then, three groups were formed according to different time and temperature of heat treatment. Details related to the control group was not applied heat treatment and three groups were given in Table 1. The particleboard panels were heat-treated with conduction mechanism.

Table 1. Form of the groups according to time and temperature of heat treatment

Groups	Time of Heat Treatment (min)	Temperature (^{o}C)	
		Lower Layer	Upper Layer
Control	-	-	-
A	10	20	150
B	20	20	150
C	10	150	150

The hydraulic press was used for heat treatment (Figure 1). Then, the lower, upper and middle layers were separated for each panel group, and the formaldehyde emission and moisture content values of these layers were separately determined. Then, the samples were conditioned prior to testing at 20°C and 65% relative humidity for two weeks.

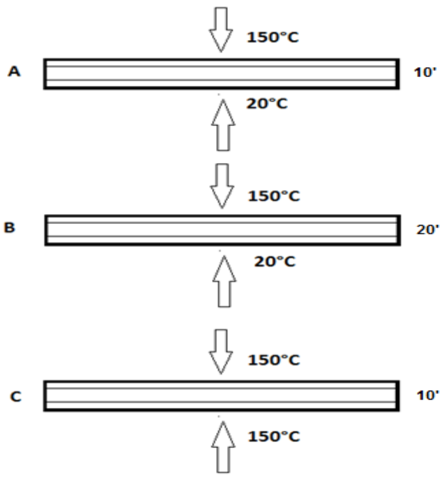

Figure 1. Application of heat treatment on the particleboard panels in the press

Formaldehyde emission contents of particleboard panels were determined according to flask method described in EN 717-3 (1996) standard. This is a simple and inexpensive method for testing formaldehyde release and suitable for testing of uncoated boards (Aydin et al., 2006). This method is suitable only for internal production control of wood-based panels and no official limit values published. For testing, it was taken test pieces of 25 mm x 25 mm x board thickness. For each flask, 15-17 gr weight samples are prepared (Figure 2). The test temperature and time used were 40°C and 3 h, respectively. Formaldehyde emission determined in test samples with two repetitions for each groups. The moisture contents of particleboard panels were determined according to EN 322 (1993) standard.

Multifactor analysis of variance was performed for statistical evaluation of the changes in formaldehyde emission depending on layers in the particleboard panels. After ANOVA, Student–Newman–Keuls test with 95% confidence level was used to compare the mean values of variance sources.

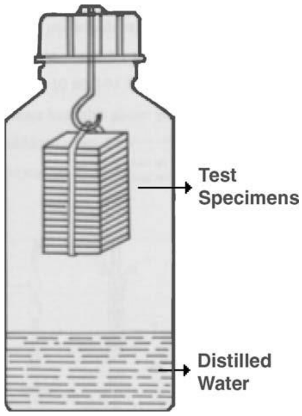

Figure 2. Test apparatus used for the determination of formaldehyde emission content of particleboard panels.

The amount of formaldehyde absorbed in a 10 ml of solution from the containers is determined spectrophotometrically. The flask value (Fv) in milligrams per kilogram of oven-dry board is calculated by the following equation:

$$F_v = \frac{(A_S - A_B) \times f \times 50 \times 10(100+H)}{m} \quad (1)$$

where:

A_S is the absorbance of the analyzed solution from the containers;

A_B is the absorbance of an analysis with distilled water;

f is the slope of the calibration curve, in milligrams per milliliter;

H is the moisture content of the test pieces, in percent;

m is the mass of the test pieces, in grams.

3. Results and Discussion

Formaldehyde emission values of the particleboard panels and homogenous groups are given in Table 2. The mean formaldehyde emission rate of the middle layer for the control group was significantly higher than those of the outer layers and the complete test samples (Table 2). No significant differences were found among the upper and lower layers and complete panel in the control groups.

Table 2. Results of Student–Newman–Keuls test at 95% confidence level for formaldehyde emission values (mg/100g oven dry panel).

Test Groups	Control		A		B		C	
Complete Panel[1]	11.3	a[2]	12.8	ab	14.0	b	10.2	a
Lower Layer	11.6	a	13.2	b	16.8	c	9.7	a
Upper Layer	11.7	a	11.7	a	10.0	a	9.8	a
Middle Layer	14.3	b	14.5	c	14.1	b	12.5	b

[1] Complete Panel; Samples of panel thickness without departing from layers; [2] The means with different letter are statistically different ($p \leq 0.05$).

Assessing groups A and B, which were applied heat treatment from the upper layer, the lowest formaldehyde emission rates were found in the upper layers. While the highest values were found in the middle layer for A, the highest rates occurred in the lower layer for B. This could be explained by the longer heat treatment in group B. Also, the formaldehyde emission rates of the complete group was lower than those of the middle layer for A and the lower layer for B while they were higher than the upper layer which received the heat treatment. The highest formaldehyde emission rates for C were found in the middle layer. The formaldehyde emission rates in the complete panel, lower and upper layers were not significantly different.

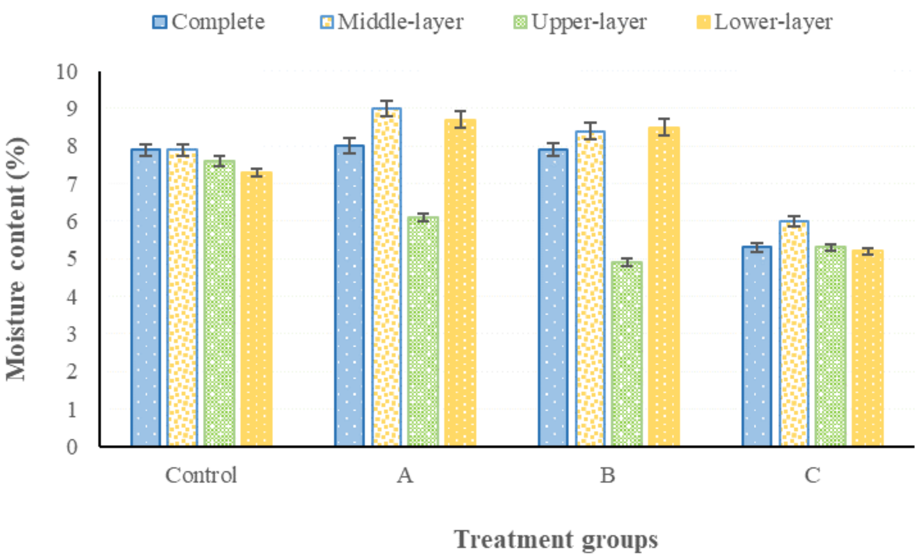

Figure 3. Mean moisture content of panel layers (bars indicate std deviation)

In terms of the moisture content of the control group, the average moisture content rates were found very similar for the all of the layers (Figure 3). The lowest moisture content for A and B were found in the upper layer which was received the heat treatment. The moisture content reduction in the upper layer of B was greater than that of the upper layer of A. The highest moisture content for C which received the heat treatment for both layers in press was found in the middle layer.

It is stated that no significant differences in moisture content of particleboards do not have an effect on results of formaldehyde emission with flask method (Roffael, 1982). Therefore, it should be considered that the differences in moisture content do not have a significant effect on the formaldehyde emission rates in this study. Also, it is possible that the moisture content with a part of formaldehyde gas molecules in the cell cavities releases because of the effect of heat treatment applied during the evaporation moisture content from surface layers.

According to the results, the formaldehyde emission rates in the surfaces that received heat treatment were lower. It is reported in several studies that press temperature had a great effect on formaldehyde emission from particleboards (Brinkman, 1978; Myers, 1984; Petinarakis and Kavvouras, 2006; Tekin and Keskin, 2015), and it significantly decreased formaldehyde emission. Likewise, formaldehyde emission decreases with longer pressing time (Kollman et al., 1975).

The reason for the high formaldehyde emission in the middle layers is not only formaldehyde molecules released from the surfaces by the effect of the moisture evaporating with temperature. It is well-known that heat energy is transferred through the movement of molecules (Philip and De Vries, 1957). With an increase in temperature of a material, the movement of molecules is accelerated, and heat flows to the colder side. The velocity of the gas molecules in a mixture form of air and moisture in wood pores is directly proportional to the absolute temperature. As temperature increases, velocity of molecules increases (Philip and De Vries, 1957). Therefore, as a result of the high formaldehyde emission in the middle layer that is not significantly affected by the applied heat treatment should be considered a part of the gas mixture of formaldehyde and moisture in the wood pores flows into the middle layers during the flow of heat to the cold side.

4. Conclusions

The formaldehyde emission rates in the middle layer (forming 60% of panels) were higher than the upper and lower layers (forming 40% of panels) in the control group. Formaldehyde emission values were not found significantly differences between upper and lower layers. It was found that the formaldehyde emission of the particleboard changed at the lower, upper and middle layers with heat treatment.

The lowest formaldehyde emission values were found in the upper layers for A and B groups. It was determined that molecules of formaldehyde in the both groups flowed to cold side from hot side with heat treatment. The formaldehyde emission in the heat treated layers decreased as the processing time increased.

Among formaldehyde emission values of the upper and lower layers were treated equal heat in C group were not found clearly. While the highest formaldehyde emission values were obtained from the middle layers for A and C groups, they were obtained from the lower layer for B.

In generally, free formaldehyde molecules are more concentrated in the middle layer at non-heat treated panels and two surfaces with equal temperature applied panels. If the heat treatment is applied only from the upper layer, the free formaldehyde is collected in the lower layer and the amount of formaldehyde increases as the heat treatment time increases.

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