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ARAŞTIRMA MAKALESİ RESEARCH PAPER

# The Effects of Some Synthetic Adhesives on the Formaldehyde Emission of the Particleboard

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**Abstract:** The formaldehyde emission is the one of the major problems of the particleboard industry that harms to human health and environment. The purpose of this study was to determine the effect of the adhesive type on the formaldehyde emission of three-layered particleboard. The urea formaldehyde (UF), phenol formaldehyde (PF) and melamine-urea formaldehyde (MUF) adhesives were used as adhesives. Black poplar *(Populus nigra)* was used as wood raw material. The panels produced with PF and MUF adhesives had the required level of formaldehyde emission value specified in EN 312-2 (1996) and these panels could be classified as E-1 grade particleboard. The particleboard samples produced with UF adhesive had more than the formaldehyde emission value required for E-1 class particleboards.

**Keywords:** Environment, formaldehyde emission, particleboard, thermosetting adhesives.

# Bazı Sentetik Tutkalların Yongalevhanın Formaldehit Emisyonu Üzerine Etkileri

**Öz:** Formaldehit emisyonu, yongalevha endüstrisinin insan sağlığına ve çevreye zarar veren en büyük sorunlarından biridir. Bu çalışmanın amacı, tutkal türünün üç tabakalı yongalevhanın formaldehit emisyonu üzerindeki etkisini belirlemektir. Tutkal türü olarak üre formaldehit (ÜF), fenol formaldehit (FF) ve melamin-üre formaldehit (MÜF) kullanılmıştır. Odun hammaddesi olarak Karakavak *(Populus nigra)* kullanılmıştır. FF ve MÜF yapıştırıcılarla üretilen paneller, EN 312-2 (1996) 'de belirtilen formaldehit emisyon değerine sahip olduğundan E-1 sınıf yongalevha olarak sınıflandırılabilir. ÜF tutkalı ile üretilen yongalevha örneklerinin formaldehit emisyon değeri ise, E-1 sınıf yongalevhalar için belirtilen değerden daha fazla bulunmuştur.

**Anahtar sözcükler:** Çevre, formaldehit emisyonu, termosetting tutkallar, yongalevha.

## INTRODUCTION

Particleboard, since it has a homogeneous structure, easy processing properties and it is used in building and especially in furniture production, it is a wood composite material preferred by manufacturers (Baharoğlu et al., 2012; Yang et al., 2007). Another preferred reason of particleboard is its cheap cost due to the use of cheap raw materials such as low cost wood and other lignocellulosic materials in its production. Due to the above-mentioned features, the demand for particleboard is increasing every year throughout the world (Ciannamea et al., 2017).

The most widely used adhesives in particleboard production; urea formaldehyde (UF), phenol formaldehyde (PF) and melamine-urea formaldehyde (MUF). These are classified as the thermosetting adhesives. The PF glue has a large using are such as: wood composites, bonding of insulations materials, electrical and automotive parts, indoor and outdoor applications. Besides, PF adhesives have good bonding properties, thermal stability and high resistance to moisture. UF glue has low resistance to moisture as well as being cheap and is therefore suitable for internal use (Sheng et al., 2008; Bardak et al., 2011). MUF adhesives commonly use in the wood composite panel industry because of their low cost short curing cycle and higher resistance to moisture according to UF adhesive. MUF adhesive is suitable for both indoor and outdoor applications (Jiang & Lu, 2017). The above-mentioned glues have a disadvantage other than these properties. It is also the formaldehyde emission. The formaldehyde emission affects negatively environment and human health (Que et al., 2013). During the production of particleboard and during the use of the final products produced from these particleboards, formaldehyde is released. Workers are affected by formaldehyde emissions during the production of particleboard in the factory. In addition, users are also affected by the formaldehyde emission of products produced from the particleboard used in indoors, such as furniture. The US Environmental Protection Agency declared formaldehyde as a priority pollutant (Moubarik et al., 2012). Formaldehyde was recognized as ‘‘carcinogenic to humans (Group 1)’’ in 2004 by the International Agency for Research on Cancer (IARC). Formaldehyde can cause some diseases such as irritation on the eyes and nose, asthma, headache, throat irritation and cancer (Baharoğlu et al., 2012; Costa et al., 2013). Besides, the formaldehyde emission was recognized as a reason of the sick house syndrome in 1983 by World Health Organization (Ghani et al., 2018).

The manufacturers of wood composite materials, glue manufacturers and researchers have studied to reduce the harmfulness of formaldehyde emission to the environment and human health.

Pizzi et al. produced a new type of adhesive from the tannin they obtained from pine bark and spruce. They analyzed the values of formaldehyde emission by producing a single-layer particleboard with this adhesive. At the end of this study, the value of the formaldehyde emission in the particleboard produced with these adhesives was lower than that of the particleboard produced with conventional glues (Pizzi et al., 1994). In another study, the produced particleboards were heated at 50 and 60ºC for 6 and 12 hours. After this process, the formaldehyde emission values of the heat treated particleboard samples were found to be lower than untreated particleboard samples (Jiang et al., 2017). Some researchers added nanoparticles (nanosilica, nanoalumina, and nanozinc oxide) to urea formaldehyde and melamine urea formaldehyde resin. So, these adhesives were reinforced with those nanoparticles. After this process, particleboard samples were produced and detected the value of formaldehyde emission. According to detected formaldehyde values, adding nanoparticles reduced the formaldehyde emission (Candan & Akbulut, 2013). Pirayesh et al. stated that, the using of almond and walnut shell in the mix of particleboard mat caused reducing formaldehyde emission. They reported that, the extractives in the shell of almond and walnut could cause decreasing the formaldehyde emission of particleboard panels (Pirayesh et al., 2013). The other way to decrease formaldehyde emission is the addition of some materials named “scavengers” to the wood particles or the resin such as sodium metabisulfite, ammonium bisulfite and urea (Costa et al., 2013). Park et al. investigated the effects of formaldehyde to urea mole ratio on the some properties of particleboard. They synthesized urea adhesives at different mole ratios. According to their test results, decreasing of the mole ratio of formaldehyde to urea caused decreasing in the free formaldehyde amount (Park et al., 2006). Researchers produced the particleboards with corn flour/NaOH and mimosa tannin/hexamine adhesives. The formaldehyde emission levels were found lower than those of the boards made with urea formaldehyde adhesive (Moubarik et al., 2013). The purpose of this study was to determine the effect of the adhesive type on the formaldehyde emission of three-layered particleboard.

## MATERIAL and METHODS

Black poplar *(Populus nigra)* was used as wood raw material in this study. The trees obtained from Trabzon in Turkey. The trees was harvested the city of Trabzon in the eastern Black Sea region of Turkey. Firstly, bark of the trees was peeled. After debarking, the trees were chipped using a ring type flaker before the chips were reduced into smaller particles using a hammer mill. The particles were dried to 3% moisture content in a dryer. The dried particles were classified into two sizes using a 3.0–1.5–0.5 mm openings vibrating screen for the core and face layers. In the next process, resin was applied with a pneumatic spray gun. Three types of adhesives were used in the production of the particleboard panels. The urea formaldehyde (UF), phenol formaldehyde (PF) and melamine-urea formaldehyde (MUF) adhesives were used as adhesives. MUF resin was at Formaldehyde/(Melmamine+Urea) molar ratio of 1.08 and solid content of 44.7%. The viscosity was 300–550 mPa.s and pH was 7.67. PF resin was at Formaldehyde/Phenol molar ratio of 1.3 and solid content of 63%. The viscosity was 60 cps and pH was 9.3. UF resin was at Formaldehyde/Urea molar ratio of 1.2 and solid content of 65%. The viscosity was 142 cps and pH was 7.95. Based on the oven-dried chip weight, a 11-wt.% resin was used for the surface layer particles and a 9-wt.% resin was used for the middle layer particles. For the resin hardener, ammonium sulfate (concentration: 25%) was used as a resin hardener during the blending process by about 1% based on the solid amount of adhesive. The shelling ratio of the particleboard mat was 40%. The mats were hot pressed at a temperature of 150 °C for 6 min at 25 kg/cm$^2$ with a panel thickness of 12 mm. Any hydrophobic materials and other additive materials were used during panel production. Two particleboard panels were produced for each of the groups. The produced particleboard panels had the dimensions 550 mm × 550 mm × 12 mm and were conditioned at 20 °C and 65% humidity for three weeks. The experimental design is presented in Table 1.

The formaldehyde emission test was performed on the test samples according to EN 120-1 (1994). The formaldehyde emission was measured via the perforator method. The formaldehyde test was repeated in triplicate for each parameter.

A simple variance analysis with a 95% confidence level was used to determine the significant effects of the adhesive types on the formaldehyde emissions of the particleboard. A Newman Keuls test was conducted to compare the board groups that showed a significant difference.

**Table 1.** Experimental Design for Particleboard Production.

| Particleboard Group | Amount of Adhesive (%) | | | Adhesive Type |
|---|---|---|---|---|
| | Surface Layer | Core Layer | | |
| A | 11 | 9 | 0,650 | Urea Formaldehyde |
| B | 11 | 9 | 0,650 g/cm$^3$ | Melamine-Urea Formaldehyde |
| C | 11 | 9 | 0,650 g/cm$^3$ | Phenol Formaldehyde |

## RESULTS and DISCUSSION

The average formaldehyde emission values and statistical analysis results are given in Table 2. Statistical analysis showed significant differences between the average formaldehyde emission values of the samples at 95% confidence level. The differences between the groups are given in Table 2 as letters.

**Table 2.** The average formaldehyde emission values.

| Type of Particleboard | Target Density of Particleboard (g/cm$^3$) | Formaldehyde Emission (mg/100g) |
|---|---|---|
| A | 0,650 | 10,45 a (0,06) |
| B | 0,650 | 7,13 b (0,08) |
| C | 0,650 | 3,02 c (0,04) |

The values in parentheses are the standard deviation; different letters in the same column indicate statistical differences at the 95% confidence level

According to formaldehyde emission test results of particleboard panels the highest formaldehyde emission value was detected from Group A was made with urea formaldehyde adhesive (10,45 mg/100g). The group C had the lowest formaldehyde emission value produced with phenol formaldehyde adhesive (3,02 mg/100g). The formaldehyde emission value of the group B was 7,13 mg/100g produced with melamine-urea formaldehyde adhesive. According to EN 312-2 (1996) standard, the maximum permissible formaldehyde content for E1-grade formaldehyde emissions is 8 mg of $CH_2O$/100 g for oven-dried particleboard samples. B and C type panels had the required level of formaldehyde emission value specified in EN 312-2 (1996). These panels could be classified as E-1 grade particleboard. The formaldehyde emission value could be low due to the strong adhesive bond formation due to the crosslinking structure of methylene bridges formed during the curing of PF resin used in particleboard samples (El Mansouri et al., 2007). Hydrolysis as an adverse reaction in amino resins such as UF and MUF is the main cause of formaldehyde emissions in these resins. In addition, acid catalysts used to hardening in amino resins cause an increase in formaldehyde emissions. However, the melamine in the MUF adhesive is more stable against hydrolysis (Conner & Service, 2013; Salem et al., 2011). Therefore, formaldehyde emission of particleboard produced with MUF adhesive may be lower than those produced with UF.

## CONCLUSIONS

1. The adhesive type significantly affected the formaldehyde emission value of the particleboard.
2. Because of the lower formaldehyde emission the particleboards produced with MUF and PF adhesives were found to have an E1- grade formaldehyde emission, which is suitable for internal use.
3. UF glued particleboards are not suitable for internal use since they have higher formaldehyde emission values than those specified for E-1 grade particleboards according to EN 312-2 (1996).

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