EFFECTS OF TEMPERATURE AND HUMIDITY ON FORMALDEHYDE EMISSION FROM COMPOSITE FURNITURE COMPONENTS

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Key words
Formaldehyde emission, Furniture component, Impregnation, Varnish, Adhesive

Abstract
The aim of this study was to investigate the effects of various temperature and humidity environments on formaldehyde emission from composite furniture components. To accomplish this, experimental samples were collected from various composite furniture components processed with wood-based panels, wood materials, various glues, varnish, and impregnation substances. The experiments were conducted in three different environments, namely, a winter and humid environment (10 °C, humidity 95%), a spring and warm environment (20 °C, humidity 65%), and a humid-summer and dry environment (40 °C, humidity 35%), complying with TS EN 717-1 principles. The results of the experiments indicated that the highest formaldehyde emission values were found in Scots pine wood, raw particle board, wood materials with urea formaldehyde adhesive, un-impregnated, varnished, and uncoated-edge materials located in an environment where the temperature was 40 °C and the humidity was 35%. The lowest formaldehyde emission values were found in oriental beech, beech plywood, wood material with phenol formaldehyde adhesive, impregnated, and unvarnished materials located in an environment where the temperature was 10 °C and the humidity was 95%. Formaldehyde emission values obtained in this research can be taken into consideration during manufacturing of composite furniture components.

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1. INTRODUCTION

Wood-based panels (e.g., particleboard, fiberboard, plywood, blockboard, melamine coated chipboard, MDFlam, OSB, and LVL) Laminated Medium Density Fiberboard, Oriented Strand Board, Laminated Veneer Lumber created in factory settings through various manufacturing techniques using trees as raw material have significant consumption of wood-based materials rates thanks to their various sizes, stability, high strength, low cost, broad anisotropic and hygroscopic borders, and economy. Wood type, resin amount and type, and additives (hardeners and adhesives) used in manufacture and use of these panels lead to the rise of an enormous problem, namely formaldehyde emission (Çolakoğlu 1993).

Because of a decrease in internal-external air circulation caused by exterior thermal sheathing systems, which appeared as a required economic measure because of increasing global energy consumption, a remarkable amount of formaldehyde emission has begun to accumulate indoors, posing negative effects for human health (Baumann et al. 2000).

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A study suggesting that formaldehyde emission depends on temperature, relative humidity, degree of weather change, and endogenous factors (i.e., wood type, binder, level binder type, and manufacturing conditions) has reported that formaldehyde emission significantly decreases as the wood panels age (Roffael 2006).

Another study explored the effects of furniture and floor materials such as particle board, plywood, laminate, and MDF on formaldehyde emission in indoor air at two different room temperatures, i.e., 37 and 50 °C. The results of the study indicate that the use of MDF and particle board does not create health risks for humans under certain conditions, but these materials must not be used at high temperatures (50 °C, in that study) (Kim and Kim 2005). Another study that monitored the change in formaldehyde emission at different temperatures (23, 29, and 50 °C) found a significant increase in emission rate at temperatures above 29 °C (Wiglusz et al. 2002).

The present study attempted to determine changes in formaldehyde emission from composite wood furniture components. Wood-based panels (particle board, fibreboard, plywood, melamine coated chipboard, MDFlam), wood materials (oriental beech, Scots pine), various glues (phenol formaldehyde, urea formaldehyde), varnish (cellulosic), and an impregnation substance (Imersol-Aqua) were studied under three different humidity and temperature conditions. Recommendations for decreasing formaldehyde emission are also presented.

2. MATERIAL AND METHODS

Materials

Wood
Oriental beech (Fagus orientalis Lipsky) and Scots pine (Pinus sylvestris Lipsky), which are widely used as materials in the forest products industry of Turkey, along with wood-based panels (particle board, fibreboard, plywood, melamine coated chipboard, MDFlam) which are widely used in the furniture industry, were chosen for this study. Wood materials used in the trials were supplied from businesses in Ankara based on completely random methods. The wood materials chosen were sound, smooth, fibrous, without internal decay, knotless, with normal growth level, without reaction wood, and unharmed by fungi and insects (TS 2470 1976).

Adhesives
The urea-formaldehyde (UF) solution used in this study has a density of 1.23 ± 0.02 g.cm⁻³, an approximate pH value of 7.5 to 8, and a viscosity of 2500 to 4000 mPa.s at 20 ± 2 °C, with a gel time of 35 to 40 s at 100 °C. The maximum free formaldehyde ratio of UF is 1%, and the solution has a resin of 50 gr + ammonium chloride (NH4Cl) of 10%. It is recommended that UF adhesive be applied to a surface at approximately 190 g.m⁻². The phenol-formaldehyde (PF) solution used in this study has a density of 1.18 ± 0.03 g.cm⁻³, a viscosity of 13,000 ± 2000 mPa.s, and a pH value of 7.5 to 8.5 at 20 ± 2 °C, with a gel time of 10 to 20 s at 100 °C and a solidification time of 2 to 4 min at 110 °C. It is recommended that PF adhesive be applied to a surface at approximately 180 g.m⁻². The UF and PF adhesives were directly applied to one of the surfaces of the respective wood materials, and the bonding process was conducted at 20 ± 2 °C and 65 ± 5% relative humidity. Polisan, a manufacturing firm based in Izmir, Turkey, supplied the PF and UF adhesives (Polisan 1999).
Impregnation material
Imersol Aqua (IA), used as an impregnation material, was supplied by Hemel-Hickson Timber Products Ltd. (Istanbul). The material is a non-corrosive, non-flammable, odourless, fluent, water based material with a pH of 7 and a density of 1.03 g.cm$^{-3}$. It is provided as a ready-made solution, and contains 0.5% (w/w) tebuconazole, 0.5% (w/w) propiconazole, 1% (w/w) 3-Iodo-2-propynl-butyl carbonate, and 0.5% (w/w) cypermethrin. All drilling, cutting, turning and milling operations should be completed and the relative humidity should be in equilibrium with the test environment before the IA is applied to the wood material. For the impregnation process, dipping duration was a minimum of 6 min in an impregnation pool that contained 15 litres of impregnation material for each cubic metre of wood. The impregnated wood must dry for at least 24 h (Hickson’s Timber Impregnation 2000; Keskin 2009).

Methods
Determination of density
The densities of wood materials were determined according to TS 2472 (1976). To determine the air-dry density, test samples with dimensions of 20 mm × 30 mm × 30 mm were kept under conditions of 20 ± 2 °C and 65 ± 5% relative humidity until they reached a constant weight. The weights were measured with an analytical scale with a sensitivity of ± 0.01 g. Afterwards, the dimensions were measured using a digital caliper with a sensitivity of ± 0.01 mm. The air-dried densities ($\delta_{12}$) of the samples were calculated using the following equation:

$$\delta_{12} = \frac{W_{12}}{V_{12}} \text{ g.cm}^{-3}$$

where $W_{12}$ is the air-dry weight (g) and $V_{12}$ is the volume (cm$^3$) at air-dry conditions.

Preparation of experimental samples
The wood materials used in this study were chosen from first class timber and cut from annual rings from regions vertical to the surface according to TS 2470 (1976). The pieces were then kept in an air-conditioned cabinet at a temperature of 20 ± 2 °C and relative humidity of 65 ± 5% until their weight became constant. The experiment was conducted after the samples were dried to 8% to 12% of their original moisture content. A total of 312 experimental samples with dimensions of 100 mm × 500 mm ×18mm were prepared, and 234 wood-based panels were prepared with dimensions of 500 mm × 500 mm ×18mm. Of the 100 mm × 500 mm materials, 156 were impregnated for 24 h with a dipping method following ASTM D 1413-76 (2005). Panels were produced from the impregnated and un-impregnated samples using the two types of adhesives. The produced panels were classified as varnished and unvarnished. Wood-based panels were classified as raw particle board, fibreboard, MDFlam, melamine-coated chipboard, poplar plywood, and beech plywood.(Güller 2001) Additionally, MDFlam and melamine-coated chipboard were prepared as coated-edge and uncoated-edge. Figure 1 shows the some test samples.
**Determination of formaldehyde emission**

Average formaldehyde emission from wood and wood-based panels was determined in this study as per the principles of TS EN 717-1 (2006). Formaldehyde emission was measured using a gas detector produced by RAE Systems in USA. The appearance of gas detector is given in Figure 2. The detector uses PID technology. A photoionization detector (PID) uses an ultraviolet (UV) light source to break down chemicals to positive and negative ions. The gas becomes electrically charged. In the PID, these charged particles produce a current that is then amplified and displayed on the meter as “ppm” or “ppb” (Rae Systems Application Note). The experiments were carried out in three different environments with the following humidity and temperature parameters: (1) a winter and humid environment (10 °C, humidity 95%), (2) a spring and warm environment (20 °C, humidity 65%), and (3) a humid-summer and dry environment (40 °C, humidity 35%), complying with TS EN 717-1 (2006). The view of test cabin is given in Figure 3.

**Data analyses**

Multiple analyses of variance were performed using MSTAT-C statistics software version 1.42 developed by Michigan State University in USA to determine the effects of wood type, adhesive type, type of impregnated material, and humidity and temperature conditions, as well as the interaction among these variables, on formaldehyde emission into the environment by these samples. Where the results of variance analysis produced at 0.05 confidence level (α<0.05) significant correlations, Duncan’s test was performed. In this way, success ratings among tested factors were identified according to least significant difference (LSD) critical value by dividing them into homogeneous groups.
3. RESULTS AND DISCUSSION

Average formaldehyde emission values by material type are provided in Table 1.

Table 1. Average Formaldehyde Emission Values by Material Type (ppm)

<table>
<thead>
<tr>
<th>Types of materials</th>
<th>Formaldehyde Emission (ppm)</th>
<th>HG*</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Wood panels</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oriental Beech (Ob)</td>
<td>0.5596</td>
<td>A</td>
</tr>
<tr>
<td>Scotch Pine (Sp)</td>
<td>0.4646</td>
<td>B</td>
</tr>
<tr>
<td><strong>Wood-based panels</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Raw particle board (Pb)</td>
<td>0.8451</td>
<td>A</td>
</tr>
<tr>
<td>Raw fibreboard (Fb)</td>
<td>0.7841</td>
<td>AB</td>
</tr>
<tr>
<td>Raw MDFlam (MDF)</td>
<td>0.7828</td>
<td>AB</td>
</tr>
<tr>
<td>Raw melamine coated chipboard (Cb)</td>
<td>0.6346</td>
<td>ABC</td>
</tr>
<tr>
<td>Poplar plywood (PP)</td>
<td>0.5438</td>
<td>BC</td>
</tr>
<tr>
<td>Oriental Beech plywood (BP)</td>
<td>0.3974</td>
<td>C</td>
</tr>
<tr>
<td><strong>Adhesives</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Urea-Formaldehyde (UF)</td>
<td>0.5560</td>
<td>A</td>
</tr>
<tr>
<td>Phenol-Formaldehyde (FF)</td>
<td>0.4681</td>
<td>B</td>
</tr>
<tr>
<td><strong>Impregnation Processes of types</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Unimpregnated (UI)</td>
<td>0.3324</td>
<td>A</td>
</tr>
<tr>
<td>Impregnated (I)</td>
<td>0.2802</td>
<td>B</td>
</tr>
<tr>
<td><strong>Varnishing Processes of types</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Varnished (V)</td>
<td>0.5434</td>
<td>A</td>
</tr>
<tr>
<td>Unvarnished (UV)</td>
<td>0.4807</td>
<td>B</td>
</tr>
<tr>
<td><strong>Environments of types I</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>O3 (Temperature 40 °C-Humidity 35%)</td>
<td>0.6767</td>
<td>A</td>
</tr>
<tr>
<td>O2 (Temperature 20 °C-Humidity 65%)</td>
<td>0.4868</td>
<td>B</td>
</tr>
<tr>
<td>O1 (Temperature 10 °C-Humidity 95%)</td>
<td>0.3926</td>
<td>C</td>
</tr>
<tr>
<td><strong>Environments of types II</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>O3 (Temperature 40 °C-Humidity 35%)</td>
<td>0.9182</td>
<td>A</td>
</tr>
<tr>
<td>O2 (Temperature 20 °C-Humidity 65%)</td>
<td>0.6367</td>
<td>B</td>
</tr>
<tr>
<td>O1 (Temperature 10 °C-Humidity 95%)</td>
<td>0.4391</td>
<td>C</td>
</tr>
<tr>
<td><strong>Processes of edge coating</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Uncoated-edge (UC)</td>
<td>0.6181</td>
<td>A</td>
</tr>
<tr>
<td>Coated-edge (C)</td>
<td>0.5336</td>
<td>B</td>
</tr>
</tbody>
</table>

Formaldehyde emission was found to differ depending on wood type, wood-based panel type, adhesive type, impregnation, varnishing, ambient temperature, and edge coating. The highest formaldehyde emission values were found, by material type, with Scots pine, raw particle board, wood material having urea formaldehyde adhesive, un-impregnated, varnished, and uncoated edges that were located in an environment where the temperature was 40 °C and the humidity was 35%. The lowest formaldehyde emission values were found, by material type, with oriental beech, beech plywood, wood materials with phenol formaldehyde adhesive, impregnated, and unvarnished that were located in an environment where the temperature was 10 °C and the humidity was 95%. Multiple variance analysis results for the panels produced in this study are provided in Table 3.

The effects of humidity and temperature conditions for samples produced from wood material (Scots pine and oriental beech) with different adhesives (urea formaldehyde and phenol
The effects of humidity and temperature conditions for laminated wood material, both edge-coated and edge-uncoated, on formaldehyde emission were found to be statistically significant ($\alpha = 0.05$). Duncan’s test was applied to determine for which groups the difference was significant. Duncan’s test results of triple interaction among laminated wood material type, type of treatment, and experiment environment are illustrated in Table 3.

Figure 4. Average formaldehyde emission values with respect to interactions among wood type, adhesive type, varnishing, and experimental environments

The effects of humidity and temperature conditions for laminated wood material, both edge-coated and edge-uncoated, on formaldehyde emission were found to be statistically significant ($\alpha = 0.05$). Duncan’s test was applied to determine for which groups the difference was significant. Duncan’s test results of triple interaction among laminated wood material type, type of treatment, and experiment environment are illustrated in Table 3.

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Table 3. Duncan Test of Triple Interaction among Laminated Wood Material Type, Type of Treatment, and Experiment Environment

<table>
<thead>
<tr>
<th>Type of process x</th>
<th>HG</th>
<th>Type of process x</th>
<th>HG</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cb+UC+O3</td>
<td>0.9515</td>
<td>A</td>
<td>MDF +UC+O2</td>
</tr>
<tr>
<td>Cb+C+O3</td>
<td>0.7515</td>
<td>B</td>
<td>Cb+UC+O1</td>
</tr>
<tr>
<td>MDF+UC+O3</td>
<td>0.6777</td>
<td>BC</td>
<td>MDF +UC+O1</td>
</tr>
<tr>
<td>MDF+C+O3</td>
<td>0.6331</td>
<td>C</td>
<td>MDF +C+O2</td>
</tr>
<tr>
<td>Cb+UC+O2</td>
<td>0.6062</td>
<td>C</td>
<td>Cb+C+O1</td>
</tr>
<tr>
<td>Cb+C+O2</td>
<td>0.5077</td>
<td>D</td>
<td>MDF +C+O1</td>
</tr>
</tbody>
</table>

The highest formaldehyde emission value (0.9515 ppm) was obtained during trials of coated-edge melamine coated chipboard samples in the 3rd environment, whereas the lowest formaldehyde emission value (0.4092 ppm) was obtained during trials of coated-edge MDFlam in the 1st environment. These results are shown in Figure 5.

![Figure 5. Average emission values with respect to interactions among laminated wood material type, type of treatment, and experimental environments](image)

4. CONCLUSIONS

In this section, some recommendations are made comparing the results obtained from the trials with the studies available in relevant literature.

Regarding wood type, Scots pine emitted higher levels of formaldehyde than oriental beech. This can be attributed to the amount of extractives in Scots pine wood (Schafer and Roffael 2000). Likewise, a study that explored the effect of wood type on formaldehyde emission found that poplar plywood emission was higher than that of beech and okoume plywood (Colakoğlu 1993). Moreover, another study comparing particle boards manufactured from oak and pine chips, with all other factors held constant, reported that particle board manufactured from oak had less formaldehyde emission than particle board manufactured from pine and that particle board manufactured from spruce had higher formaldehyde emission than particle board manufactured from beech (Colakoglu 1993). Another study that emphasized the fact that wood type affects formaldehyde emission found that among beech, poplar, spruce, birch,
and pine blockboards, birch had the highest emission rate (1.35 mg.m\(^{-2}\).h), while poplar had the lowest (0.9 mg.m\(^{-2}\).h) (Böhm et al. 2012).

The formaldehyde emission in unimpregnated samples was found to be 0.3324 ppm, while it was found to be 0.2802 ppm in impregnated samples, suggesting that impregnation decreases formaldehyde emission. A previous study reported that samples impregnated with borax exhibited decreased formaldehyde emission and that formaldehyde emission and impregnation have a significant correlation (Colak 2002).

Varnished samples had higher formaldehyde emission (0.5434 ppm) than unvarnished samples (0.4807 ppm), suggesting that varnishing the wood material increases formaldehyde emission. This can be attributed to the fact that root VOC emission agents that form the structure of varnish may have increased formaldehyde emission of plain wood material because cellulosic varnish is composed of nitrocellulose and sulphuric acids, solvents such as acetone, amyl acetate, butyl acetate, ethyl acetate, varnish resins, thinners such as toluene and gasoline, and plasticizers or softeners (phosphoric and phthalic acids) (Hammond 1969).

The highest formaldehyde emission in massive panels was found with urea formaldehyde adhesive (0.6767 ppm) in the 3rd environment, and the lowest was found with phenol formaldehyde adhesive (0.3926 ppm) in the 1st environment. This could be explained by the activation of components within the adhesive through changes in temperature and humidity. As a matter of fact, a study by Colak (2002) indicated the core reason for formaldehyde emission was the 1st stage of reaction in two-stage urea and formaldehyde reaction of urea formaldehyde adhesives, which showed low levels of resistance against hydrolysis caused by water and humidity. It was also demonstrated that the bond in the phenol formaldehyde adhesives, which ensure adhesion through the reaction of phenol with formaldehyde, is stronger than urea formaldehyde adhesives (Colak 2002).

The highest formaldehyde emission was found at Sp+UF+UV+O3 (0.8954 ppm), and the lowest formaldehyde emission at Ob+FF+V+O1 (0.3300 ppm) according to interaction among wood type, adhesive type, varnishing, and experimental environment. Formaldehyde emission and adhesive are directly related to each other (Akbulut 1998). A previous study demonstrated that formaldehyde emission stems from formaldehyde-based resins (Kim et al. 2007).

The highest formaldehyde emission value (0.8451 ppm), by type of laminated wood material, was measured on raw particle board that did not go through any processing. The lowest formaldehyde emission value (0.3974 ppm) was measured from raw beech plywood.

The reason for the high formaldehyde emission from raw particle board may be the fact that the material is not coated and there are plenty of emission exit channels on the surface left after manufacturing. In fact, it was reported that treating the particle board with a surface treatment system that contains active agents that bind formaldehyde decreases emission. It was noted that a decorative vinyl coating agent can be applied to seal all pores on the outer surface, which helps decrease formaldehyde emission. It was also reported that uncoated cross sections have high rates of emission (Kurtoglu and Ucar 1986). Another study indicated that coating with decorative coatings may be used to eliminate free formaldehyde on panel surfaces (Pirayesh et al. 2013).
The poplar plywood emission value (0.5438 ppm) was found to be higher than that of beech plywood (0.3974 ppm). Another study also found the average emission value of poplar plywood (66.24 mg/100 g) to be higher than that of beech plywood (48.28 mg/100 g) (Sensogut et al. 2009). The fact that the measured formaldehyde emission value is higher in poplar plywood than in beech plywood stems from the fact that acetyl groups in the wood have effects on formaldehyde emission and that the increasing rates of acetyl groups leaving the wood during hot-pressing decreases emission (Akbulut, 1998). A previous study found that the formaldehyde emission released from pine plywood is greater than that from beech plywood (Colakoglu et al. 1998).

The reason for the relatively high formaldehyde emission in plywood may be that the surface of plywood is loose or tightly-structured and porous. Plywood is a surface-glued material, and formaldehyde is emitted through the surface openings to the exterior (Kurtoglu and Ucar 1986).

The highest emission value was found at 40 °C and a humidity of 35%. A decrease in the emission value was observed as the temperature decreased. This may be due to the direct relationship between the temperature and disintegration rate of the adhesive in the structure. In a study conducted in Korea regarding widespread use of under floor heating systems and their effects on VOCs emitted from parquet floors, formaldehyde emission was measured via a desiccator in a 20 L room using the Field and Laboratory Emission Cell FLEC method; the formaldehyde emission rate of the laminate was measured to be the highest at 32 °C at the highest desiccator. Desiccator is a method for determination of the formaldehyde emission formaldehyde emission These under floor heating systems spread more formaldehyde emission than air-circulation heating systems, perhaps due to the temperature difference between the ground and flooring (Kim et al. 2011).

The highest formaldehyde emission (0.6181 ppm) was measured from the wood-based panels with uncoated edges. Coating the edges of panels after manufacture along with other treatments have been shown to decrease formaldehyde emission (Athanassiadou 2000).

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