Formaldehyde and TVOC emission behavior of laminate flooring by structure of laminate flooring and heating condition

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A B S T R A C T
Formaldehyde was measured with a desiccator, a 20 L chamber and the FLEC method. The formaldehyde emission rate from laminate was the highest at 32 °C using the desiccator, which then decreased with time. The formaldehyde emission using the 20 L small chamber and FLEC showed a similar tendency. There was a strong correlation between the formaldehyde and total volatile organic compounds (TVOCs) with both types of floorings using the two different methods. The formaldehyde emission rate and TVOC results were higher when tested using the FLEC method than with the 20 L small chamber method. The emission rate was affected by the joint edge length in laminate flooring. Toluene, ethylbenzene and xylene were the main VOCs emitted from laminate flooring, and there were more unidentified VOCs emitted than identified VOCs. The samples heated with a floor heating system emitted more formaldehyde than those heated using an air circulation system due to the temperature difference between the bottom panel and flooring. The TVOC emission level of the samples was higher when an air circulation system was used than when a floor heating system was used due to the high ventilation rate.

1. Introduction

Currently, buildings contain many synthetic building materials and furnishings, such as the walls, carpets, and air conditioning systems. The use of these newer materials and furnishings has resulted in a larger number of new indoor pollutants in greater concentrations [1–3]. Waxes, paints, polishes, cleansers, air fresheners, fabric protectors and composite materials are all sources of various organic and inorganic chemicals [4].

Due to the large quantity of materials used in buildings, and their constant exposure to indoor air, there is a growing concern regarding the effects of these indoor pollutants on the health and comfort of the building occupants. More than two hundred volatile organic compounds (VOCs) have been identified in the indoor environment [5–7]. Over the past decade, these VOCs have been associated with adverse health effects, such as sensory irritation, odor and a more complex set of symptoms referred to as sick building syndrome (SBS). Researchers have also found that neurotoxic effects can result from low level of exposure to air pollutants [8]. These reactions can include watery eyes, runny nose, a high frequency of airway infections, asthma-like symptoms in non-asthmatics, and odor or taste complaints [9,10].

There are many gases, materials, particles, etc. in a building that can cause indoor air-quality-related health problems and sick building syndrome. Building materials can release a range of pollutants, particularly volatile organic compounds (VOCs), which can make the indoor air quality worse than outdoor air. Recent studies of VOC emissions in four newly built, unoccupied test houses also reported building materials to be the main source of indoor air pollution [11–14].

Laminate wood flooring consists of HDF (high-density fiberboard) as the core material, while engineered flooring consists of plywood with a thin fancy veneer bonded onto the face of the plywood using urea–formaldehyde and melamine–formaldehyde (MF) resins as hot-press adhesives. Most wood laminate floors are simply a photographic representation of wood grains. Laminate wood flooring consists of four main components bonded together. A wear resistant, decorative surface made from a resin-based MF resin and aluminum oxide is bonded to a moisture resistant, wood composition based core. A balancing backing is bonded to the underside of the core. On the top there is a clear cap sheet of aluminum oxide, which provides protection and stain resistance [15–17].

This floor heating system is quite popular in Korea. Koreans spend a considerable proportion of their time sitting directly on the floor heating system and have traditionally slept on thin cotton mattresses warmed directly from the floor surface [18].

In this study, a desiccator method was used to measure the levels of formaldehyde emission from laminate flooring at various
temperatures set by the heater system to evaluate under heating system effect. A 20 L small chamber and the FLEC were used to evaluate both the formaldehyde and total volatile organic compounds (TVOCs) emission rates when the joint edge length was 0 cm, 14 cm and 28 cm. FLEC was also used to determine the levels of formaldehyde and TVOC emission from laminate flooring at a variety of surface temperatures by adjusting the floor heating system and air circulation system.

2. Experimental

2.1. Materials

2.1.1. Laminate flooring

Laminate flooring is used in new apartment interiors and renovated houses in Korea, and is composed of waterproof, high-density fiberboard (HDF) as the core material, an overlay paper, decou paper and balance paper. Each paper was impregnated with melamine–formaldehyde and TVOC emission from laminate flooring at a variety of

2.1.2. Heating systems (floor heating system and air circulation system)

The floor heating system for this experiment was constructed from MDF for the bottom panel, a heater sheet and an engineered floor. The heater sheet employed the same principal as hot water pipes to heat floors. The heater sheet is a commercial product that allows the surface temperature to be varied from 10\(^\circ\)C to 80\(^\circ\)C. An oven-dry was used to assimilate the fan air circulation system.

2.2. Edge treatment of samples

The edges of all experimental samples were sealed with Parafilm [19] to properly analyze the formaldehyde, VOC and TVOC emissions.

2.3. Formaldehyde emission measured by the desiccator method with edge sealing treatment at various temperatures

The formaldehyde emissions from the laminate flooring were determined by using a desiccator [20] according to the standard Japanese method. The desiccator test was used to determine the quantity of formaldehyde that building boards emit and was carried out using a 10 L glass desiccator. The quantity of formaldehyde emitted was obtained by measuring the concentration of formaldehyde absorbed in either distilled or deionized water when the test pieces of a specified surface area were placed in a desiccator filled with a specified amount of distilled or deionized water and allowed to emit for 24 h.

The surface of the samples met the criteria of 1800 mm\(^2\) as defined by the JIS and KS. The temperature of the drying oven was set to 20 ± 1\(^\circ\)C, 26 ± 1\(^\circ\)C or 32 ± 1\(^\circ\)C. The laminate floorings were conditioned and sampled in a dry oven at 20 ± 1\(^\circ\)C, 26 ± 1\(^\circ\)C, and 32 ± 1\(^\circ\)C for 1, 3, 5, 7, 9, 11, 13, 15, 17, 19, 21, 23, 25, 27, and 29 days.

2.4. 20 L small chamber test for formaldehyde and TVOC emission

The use of a 20 L chamber has been standardized in Korea, and was used in this study [21,22]. The air control system consisted of an air supply unit, a humidifier and pumps. The 20 L chamber was placed in a temperature-controlled climate chamber. Purified air was used for ventilation. A stainless steel seal box was used to prevent the cut edge effect, so chemical emission was only allowed to come from one side of the test piece. When two of these seal boxes were used, the total surface area and loading was 0.044 m\(^2\) and 2.2 m\(^2\)/m\(^3\), respectively [23].

Both the chamber and seal boxes were washed with water and baked in an oven at 260\(^\circ\)C prior to use to eliminate the pollutants from the chamber itself. A small 20 L chamber was supplied with purified and humidified air at a given ventilation rate. Both the internal temperature and relative humidity within the chamber were kept constant. Table 1 lists the test conditions of 20 L chamber and the FLEC. All the test pieces were sealed in seal boxes and placed into the chamber. The air inside the chamber was sampled after 24 h. Table 2 shows the sampling conditions. The air temperature and RH inside the test chamber, which was ventilated at 0.5/h, were kept constant throughout the experiment at 25 ± 1\(^\circ\)C and 50 ± 5% RH, respectively. The aldehydes were analyzed by HPLC and TDS/GC–MS was used to measure the VOCs. In this paper, TVOC was defined as the integrated peak area of the peaks between C6 and C16 to concentrations using the toluene response factor. A peak area under 10 was defined as the limit of detection; 1 \(\mu\)g/m\(^3\). Fig. 2 shows the samples with 0 cm, 14 cm, and 28 cm joint edges of laminate flooring. The sample gas was taken using Tenax-TA and the 2,4-DNPH cartridge after the sample specimens were installed for 1, 3, 5, and 7 days in a 20 L small chamber.

### Table 1

Test conditions in the 20 L small chamber and the FLEC.

<table>
<thead>
<tr>
<th>Sample size</th>
<th>0.0432 m(^2)</th>
<th>0.0177 m(^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air flow rate</td>
<td>(0.147 m × 0.147 m × 2)</td>
<td>167 mL/min (VOCs),</td>
</tr>
<tr>
<td>Ventilation rate</td>
<td>0.5/h</td>
<td>471.43/h</td>
</tr>
<tr>
<td>Sample loading factor</td>
<td>2.16 m(^2)/m(^3)</td>
<td>506 m(^2)/m(^3)</td>
</tr>
<tr>
<td>Temperature</td>
<td>25 ± 1(^\circ)C</td>
<td>25 ± 1(^\circ)C</td>
</tr>
<tr>
<td>Humidity</td>
<td>50 ± 5%</td>
<td>50 ± 5%</td>
</tr>
</tbody>
</table>

### Table 2

Sampling conditions in the 20 L small chamber method.

<table>
<thead>
<tr>
<th>Formaldehyde</th>
<th>VOCs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sampler</td>
<td>2,4-DNPH Cartridge (Supelco, USA)</td>
</tr>
<tr>
<td>Air flow rate</td>
<td>167 mL/min</td>
</tr>
<tr>
<td>Total volume</td>
<td>10 L</td>
</tr>
</tbody>
</table>
2.5. Field and laboratory emission cell (FLEC)

The loading factor, which is defined as the test material area to the emission cell volume, was a maximum of 506 m²/m³. An emission-free silicon rubber foam was used to seal the interface between the FLEC and test material surface [24]. All the tubes and couplings were made from high quality stainless steel. Air (or nitrogen) was introduced through two diagonally positioned inlets into a circular shaped channel (depth 7 mm × 7 mm) at the perimeter of the cell, so that the air was distributed over the test material surface through a circular air slit (1 mm). The air then exited from the cell from above at the top-center of the FLEC. Such an arrangement provided a constant and efficient air velocity over the entire surface, with the exception of a smaller part near the center, because the cylindrical cross-section area is constant from the perimeter. The FLEC was supplied with clean and humidified air (or nitrogen) from an air supply control unit as outlined. The FLEC was supplied with purified and humidified air at a set ventilation rate. The emission sample was collected under the FLEC lid at an air flow rate of 275 mL/min after a 30 min equilibration time. For formaldehyde, 5.0 L of gas was collected in a 2,4-DNPH cartridge for 30 min under a gas flow rate of 167 mL/min, while 1.6 L of gas was collected in a Tenax-TA tube for 30 min under a gas flow rate of 53 mL/min.

In this experiment, FLEC was used to evaluate both the formaldehyde and the TVOC emission rates when the joint edge length was 0 cm, 14 cm, or 28 cm. FLEC was also used to determine both the formaldehyde and TVOC emission levels of the laminate flooring at a surface temperature of 20 °C, 26 °C, or 32 °C by adjusting the floor heating and air circulation systems (Fig. 3). The formaldehyde and VOCs emitted were collected 1, 3, 5, and 7 days after installing the sample. The same HPLC and TDS/GC–MS instruments employed for the 20 L chamber were used to analyze the formaldehyde and VOCs levels in this experiment.

3. Results and discussion

3.1. Formaldehyde emission behavior using a desiccator

Formaldehyde is emitted from wood-based products, such as particleboard (PB), medium density board (MDF), and plywood that has been bonded with a formaldehyde-based resin. This is particularly true in case of urea–formaldehyde (UF) resin, which has the highest formaldehyde emission rate because a proportion of the UF resin is incompletely cured, resulting in free formaldehyde and formaldehyde emission due to hydrolysis of the cured UF resin. Fig. 4 shows the results of formaldehyde emission at 20 °C, 26 °C, and 32 °C. Each sample was tested three times. The results show that the level of formaldehyde emitted from the laminate flooring increased with increasing temperature. The sample held at 32 °C for 3 days had more than twice the formaldehyde emission than the sample held at 20 °C over the same period of time. A sample held at 32 °C for 9 days showed more than 1.5 mg/L formaldehyde, and is classified as E2 grade [20]. After a period of 9 days, the formaldehyde emission was <1.5 mg/L (E1), and was reduced by 0.85 mg/L. The sample kept at 26 °C emitted formaldehyde at a concentration of 0.97 mg/L after the first day, which decreased slightly to 0.75 mg/L after 29 days; this sample was regarded as E1. The sample at 20 °C emitted a constant level of formaldehyde after installing the sample; this sample was classified as E1. There are two types of laminate flooring commonly available. One type is produced by bonding a decorative plastic laminate onto the substrate using a wet bonding agent. The other consists of several layers of specially
saturated paper that is thermofused directly onto the substrate. The substrates are high-density fiberboard (HDF). Some laminate flooring emits small quantities of formaldehyde and volatile organic compounds (VOCs). The surface of the laminate flooring that is made from low-pressure melamine (LPM) prevented the emission of formaldehyde from the surface of the raw materials. This type of laminate flooring can affect the level of chemical contamination of indoor air when floor heating is used[25].

3.2. Emission behavior of formaldehyde and aldehydes from adhesive, flooring and flooring with adhesives using a 20 L small chamber and FLEC

Fig. 5 shows the rate of formaldehyde emission in the 20 L small chamber. The samples were tested for 7 days in a 20 L small chamber, and the formaldehyde emissions in the 2,4-DNPH cartridge were sampled after 1, 3, 5, and 7 days. The results showed that the rate of formaldehyde emission decreased with increasing joint edge length. Therefore the rate of formaldehyde emission decreases and may become stable 7 days after installing the sample. According to the joint edge length, the rate of formaldehyde emission in the 20 L small chamber was in the following order: 28 cm > 14 cm > 0 cm.

Laminate flooring made from formaldehyde-based resins, such as UF and MUF, which is common in industry, emit high amount of formaldehyde. In addition, formaldehyde was detected at an early retention time of 4.98 min because it is an aldehyde with a simple molecular structure: HCHO. Formaldehyde and acetaldehyde, 2-propenal/acetone, propionaldehyde, benzaldehyde, and tolualdehyde were detected as aldehyde compounds. Kim et al. [26] reported that wood-based composites, such as PB, MDF, engineered flooring and laminate flooring, emitted similar kinds of aldehydes. The order of emission of the aldehydes from the laminate with a 28 cm joint edge length according to their peak area was 2-propenal > formaldehyde > acetaldehyde > hexaldehyde > valeraldehyde. Valeraldehyde emission decreased to zero after 5 days. In contrast, the rate of hexaldehyde emission increased after 3 days. Samples with a joint edge of 28 cm emitted a higher concentration of aldehydes than those samples with a 14 cm or a 0 cm joint edge length, which is similar in behavior to that of formaldehyde emission. Fig. 6 shows the rate of aldehyde emission determined using the 20L small chamber for a period of 7 days. Although the results from the different joint edge lengths showed complex variations, the sample with a 28 cm joint edge length had the highest emission rate, while the sample with the 0 cm joint edge had the lowest, which is similar to the results of formaldehyde emission. The total aldehyde and formaldehyde emission rates decreased for every sample.

FLEC has the advantage of a reduced testing time compared to 20 L chamber method, and only requires a few hours for testing compared to the 7 days required when using the 20 L chamber method. Nevertheless, FLEC was used to measure the formaldehyde emission rate in a similar manner to that measured using the 20 L chamber method, namely samples were tested after 1, 3, 5, and 7 days. Fig. 7 presents the FLEC results of the formaldehyde emissions. The results show that the formaldehyde emission rate decreases with increasing installation time. The emission results from the FLEC method showed a 135% difference compared to the formaldehyde emission rates of the 20 L chamber. The samples with a 28 cm joint edge emitted 25 µg/m² h formaldehyde, which was more than the emission of samples with either a 14 cm or a 0 cm joint edge after a period of 1 day. The amount of formaldehyde released was proportional to the joint edge length of the laminate flooring.

The formaldehyde emission rates using the FLEC method were higher than those obtained using the 20 L chamber method. This is because the formaldehyde emission factor increases with increas-
ing ventilation rate, i.e., the product emitted more formaldehyde as the ventilation rate was increased [27]. FLEC has a higher ventilation rate than the 20L chamber, and hence a higher loading factor.

Regarding the joint edge length, the emission rate of formaldehyde using the 20L chamber and FLEC showed the same order: 28 cm > 14 cm > 0 cm. The reason why the formaldehyde emission behavior followed a similar pattern for both the 20L chamber and FLEC methods was because laminate flooring with a formaldehyde emission of <1.5 mg/L (E1 grade) prior to the edge sealing treatment showed a significant decrease in emission (50–80%) due to the low-pressure melamine (LPM) used as the top layer, which prevents formaldehyde emission from the HDF core of the laminate flooring. Therefore, the amount of formaldehyde emitted from the edge portion of the laminate flooring was high [28,29].

Fig. 8 presents the rates of aldehyde emission over a 7-day period using the FLEC. The measured emission rate of each aldehyde decreased throughout the 7-day period. The sample with a 28 cm joint edge had the highest emission rate after 7 days, while the sample with a 0 cm joint edge had the lowest. Each sample emitted similar levels of aldehyde. The rates of total aldehyde and formaldehyde emission decreased.

Despite the difference in test principles between the standard method, 20L chamber method, and FLEC method, the FLEC results for formaldehyde emission were similar to those from the 20L chamber. Furthermore, the correlation between the FLEC and 20L chamber method was very high. As shown in Fig. 9, the correlation between the FLEC and 20L chamber can be described by the equation, $Y = 0.476X + 3.068$ with $R^2 = 0.95$.

### 3.3. TVOC and VOCs emission behavior of various joint edge lengths using a 20L chamber and FLEC

In Korea, the Ministry of Environment provides guidelines for VOC emissions from building materials in terms of the TVOC. Even natural VOCs from wood are considered to be harmful and are included in the TVOC calculation. Consequently, it is necessary to consider natural VOCs when reassessing the regulations governing VOC emissions from building materials. More than 80–90% of emissions are unidentified VOCs, which means high emission VOCs need to be regulated and included in the TVOC emission calculations. Furthermore, non-harmful VOCs, such as natural VOCs from wood, α-pine, and β-pine, are considered as harmful VOCs when TVOCs are calculated.

When using the 20L chamber, the highest TVOC emission rate was observed on the first day. The emission rates became constant after 3 days. The sample with a 0 cm joint edge emitted TVOC constantly with an emission rate of approximately 50 μg/m² h (Fig. 10). The sample with a joint edge of 28 cm had the highest TVOC emission, as measured by FLEC (Fig. 11). This sample emitted more than twice the TVOC of the sample with a 14 cm joint edge. The TVOC in these samples decreased rapidly over a 5-day period and stabilized at approximately 180 μg/m² h. The TVOCs measured using the 20L chamber and FLEC method contained more unidentified VOCs than identified VOCs.

Toluene, ethylbenzene and xylene were the main VOCs emitted from the laminate flooring tested. TVOC emission was tested using the 20L small chamber method and FLEC. Although the 20L small chamber method is the standard VOC emission test used in Korea, there is a need for a method that is faster than this chamber method which is why FLEC was used to test the laminate flooring, as shown in Fig. 12. The correlation between the FLEC and 20L chamber can be described by the equation, $Y = 0.179X + 11.151$, with $R^2 = 0.72$. 
3.4. Formaldehyde emission behavior of indoor heating systems (floor heating system and air circulation system) using FLEC

Figs. 13 and 14 show the emission of formaldehyde from laminate flooring when using an air circulation system and floor heating system, respectively, which were obtained using the FLEC method. The rate of formaldehyde emission from laminate flooring using a heating system was approximately 12, 13, 20 (μg/m² h) for temperatures of 20, 26, and 32 (°C) after 1 day and the formaldehyde emission rate was approximately 8, 9, 12 (μg/m² h) for temperatures of 20, 26, and 32 (°C) after 7 days, respectively. The rate of formaldehyde emission from laminate flooring using a floor heating system was approximately 12, 20, 45 (μg/m² h) at 20, 26, and 32 (°C) after 1 day, respectively. The rate of formaldehyde emission was approximately 8, 7, and 12 (μg/m² h) at 20, 26, and 32 (°C) after 7 days, respectively. After 7 days, the rate of formaldehyde emission became stable with both types of heating systems. Formaldehyde is emitted mainly from high-density fiberboard (HDF), which is the core of laminate flooring. HDF is made from urea–formaldehyde resin or melamine–urea–formaldehyde condensed resin. Laminate flooring is manufactured as E1 grade in both Europe and Korea.
Wood flooring material that is of E1 grade has been circulated in Korea. In order to maintain the E1 grade, the manufacturers employ MUF (melamine–urea–formaldehyde) resin for HDF [15]. The rate of formaldehyde emission increased with increasing temperature, indicating that it is affected by the heating system. The rate of formaldehyde emission from laminate flooring with a floor heating system was higher than that of laminate flooring with an air circulation system because formaldehyde was emitted mainly from the HDF of the laminate flooring. The temperature between the bottom panel and flooring of the floor heating system was 48 °C, which was much higher than the temperature between the bottom panel and flooring when air circulation was used (30 °C). Fig. 15 shows that the temperature of each location was different. Formaldehyde emission decreases with time. High indoor temperatures that result from these heating systems allow the emission of free formaldehyde on the laminate flooring, and the floor heating system was more effective than the air circulation system for facilitating the emission of formaldehyde.

3.5. TVOC emission behavior of indoor heating system systems (floor heating system and air circulation system) using FLEC

Fig. 16 displays the rate of TVOC emission from laminate flooring when using an air circulation system, which were 664, 407 and 402 (μg/m²·h) at 20, 26, and 32 °C after 1 day, respectively. The rate of TVOC emission from laminate flooring using an air circulation system was 188, 199, and 73 (μg/m²·h) at 20, 26, and 32 °C after 7 days, respectively. The samples at 26 °C and 32 °C showed similar emission rates throughout the 7-day period. The sample held at 32 °C decayed rapidly and after 3 days, the emission rate was below that of the samples at 20 °C and 26 °C. The rate of TVOC emission from the laminate flooring when the floor heating system was used was 230, 287, and 451 (μg/m²·h) at 20, 26, and 32 °C when measured on the first day and 154, 203, and 249 (μg/m²·h) at temperatures of 20, 26, and 32 °C after the seventh day, respectively (Fig. 17). The samples at 32 °C using the two different heating systems showed a similar trend in their TVOC emission rates. The VOCs were emitted mainly from the water-based decor paper that expressed the veneer patterns. Although the surface temperature of these samples between heating systems was the same (32 °C), the rate of TVOC emission using air circulation was higher than using a floor heating system because air circulation is similar to the ventilation rate and the floor heating system is similar to the indoor air temperature. The ventilation rate and indoor air temperature for the samples using the air circulation system was higher than the samples using the floor heating system because the floor heating system emits heat from the surface but the air circulation system circulates warm air throughout the entire room. Brown [30] reported that changes in the ventilation rate had an effect on the emission rate. The rate of TVOC emission from laminate flooring increased with increasing indoor air temperature and ventilation rate.

4. Conclusions

The formaldehyde emission results using the 20 L small chamber and FLEC methods showed a similar trend. The rate of formaldehyde emission decreased as a function of time. Furthermore, there...
was good correlation ($R^2 = 0.95$) between the 20 L small chamber method and FLEC method. The emission of total aldehydes was also similar to the formaldehyde emission behavior: as the joint edge length of the sample increased, the emission of formaldehyde also increased. The VOCs emission results measured using the 20 L small chamber and FLEC showed similar trends. There was a strong correlation ($R^2 = 0.72$) between the rates of TVOC emission, as measured by the 20 L small chamber and the FLEC. These results also indicate that the rate of TVOC emission increased with increasing joint edge length. The main VOCs emitted from laminate flooring were toluene, ethylbenzene and xylene. There were more unidentified VOCs emitted than identified VOCs. Laminated flooring that is heated using a floor heating system emitted higher levels of formaldehyde than when an air circulation system was used. The levels of TVOC emission from laminate flooring heated using an air circulation system were higher than when heated by the floor heating system, due to the higher ventilation rate. In conclusion, the rates of both formaldehyde and TVOC emission were affected by the length of the joint edge of laminate flooring, as well as the temperature and ventilation rate.

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