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Comparison of formaldehyde emission from building finishing materials at various temperatures in under heating system; ONDOL

Abstract The objective of this research was to investigate the effect of various temperatures, room, 37 and 50°C, on formaldehyde emission from floor materials, such as laminate and plywood floorings, and furniture materials, such as MDF and particleboard veneered with decorative paper foil, by desiccator's method. The temperature conditions were set up by, measuring the temperature in a Korean under heating system. To maintain an indoor air temperature of 20°C, the temperature of the flooring surface was about 37°C and the temperature of the cement mortar was 50°C. The initial formaldehyde emission of the laminate flooring and plywood flooring was 1.44 and 0.63 mg/l, and for MDF and particleboard it was 4.73 and 4.95 mg/l, respectively. Floor materials were under E1 grade while furniture materials were under E_2 grade in terms of formaldehyde emission. Because of the under heating system, the flooring materials were exposed to 37 and 50°C, while the furniture materials mostly existed at room temperature. At 37 and 50°C, the formaldehyde emission level of the flooring materials was already under 0.3 ppm (F☆☆☆☆ level by JIS A 1460, application possibility without area limit) after 10 days and the emission had decreased further (0.03–0.10 mg/l) after 28 days. These levels are not injurious to the human body and will not cause sick house syndrome (SHS). The problem, however, is the furniture materials such as MDF and particleboard. As these materials are not exposed to high temperature (50°C in this experiment) in living condition, it was still E_2 grade of formaldehyde emission level at room temperature remained even after 28 days. Although there will be variations with the volume of furniture materials and the indoor conditions, furniture materials are the principal cause of indoor air quality pollution in Korean with the under heating system.

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Key words: Flooring materials; Furniture materials; Under-heating system; Formaldehyde emission; Desiccator; Indoor air quality.

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Practical Implications

Koreans spend most of their time sitting on ONDOL (heated) floors, with their buttocks always in contact with the floor surface. The flooring materials are exposed to high temperatures (37–50°C) why the effect of bake-out is rapid. The emission of formaldehyde from furniture materials are more important for the IAQ because usually MDF and particleboard of E_2 grade are being used as furniture materials in Korea.

Introduction

Formaldehyde (HCOH) is a suspected human carcinogen that is known to be released from pressed-wood products used in home construction, including products made with urea-formaldehyde (UF) resins [e.g. particleboard, hardwood plywood, medium density fiberboard (MDF), and paneling] and those made with phenol-formaldehyde (PF) resin (e.g. softwood plywood, oriented strand board) (Kelly et al., 1999; Otson and Fellin 1992). The toxicity of wood based panels bonded with UF resin, due to the emission of formaldehyde and the associated possible health hazard, could act as an obstacle to their acceptance by the public, given the prevailing climate of environmental awareness and concern. As a result, the European and Northern American governments have already or are about to impose regulations limiting the emission of formaldehyde from building materials and from the materials used for the manufacture of furniture and fittings (Kavvouras et al., 1998). Many consumer products containing formaldehyde-based resins release formaldehyde vapor, leading to consumer dissatisfaction and health-related complaints. These emissions have resulted in various symptoms, the most common of which is irritation of the eyes and the upper respiratory tract. Formaldehyde has also been found to produce nasal carcinomas in mice and rats after exposure to 14.1 and 5.6 mg/l of formaldehyde, respectively, over a long period of time. These findings have led to an intensified interest in the indoor environment. Consumer products, specifically construction materials, are a major source of formaldehyde in the indoor environment (Pickrell et al., 1986).

The Korean government started controlling Indoor Air Quality since 2004. The law from the Ministry of Environment regulates the use of pollutant emission building materials. The use of materials that emits formaldehyde more than 1.25 mg/m^2 h (JIS A 1901, Small chamber method) is prohibited. This is E_2 grade (> 5.0 mg/l) when changed to the desiccator's method (JIS A 1460). Most suppliers and people are concerned about how to reduce pollutants from building materials and how to control indoor air quality.

In renovated or completely new buildings, levels of indoor air pollutants, especially of volatile organic compounds (VOCs), resulting from emissions from constructions and building materials are often several orders of magnitude higher than the VOC levels in buildings with normal use (Brown, 1999; Rothweiler et al., 1992; Tuomaninen et al., 2001; Wolkoff et al., 1991). A procedure known as 'bake-out' has been used to reduce VOC emissions from newly installed materials, products, and furnishings (Etkin, 1996; Girman, 1989; Girman et al., 1987). The principle of this procedure is to drive VOCs out of the construction materials into the indoor air by raising the temperature in the building up to a level of 32–40°C, while increasing the outdoor air exchange so that emissions are removed from the building. The procedure generally takes several days to 2 weeks and is performed before occupancy. According to the results of a few pilot studies, a decrease of 60–94% in total VOC levels was achieved with bake-out (Follin, 1997), although contradictory results have also been reported (Bayer, 1990; Bayer, 1991; Offerman et al., 1993). There are some limitations in the practical use of bake-out, for example material damage caused by excessive temperature and humidity changes, possible sorption effects, risks for odor and irritation complaints of occupants after bake-out and overall costs of the procedure (Levin, 1989). Though the elevation of indoor air temperature is an essential part of bake-out by increasing the diffusion coefficient of VOCs, there are signs that a significant decrease in indoor air concentrations of VOCs may be achieved merely by improving ventilation using a continuous supply of fresh air instead of re-circulated air after construction, installation of new materials, painting and carpeting before occupancy (Batterman and Peng, 1995; Valicenti and Wenger, 1997).

The ONDOL floor heating system has been used conventionally in Korea since 400 b.c. (Park et al., 1995) Before the 1950s, firewood was used as fuel, with a gate for forest fuel input. After the 1950s, the ONDOL system has been modified many times for various reasons, such as to protect forestry resources and to prevent death from CO gas poisoning from anthracite, etc. (Song, 1996). For a short while, briquettes were used, but after the 1970s, Korean economic power has increased annually. Residential buildings were developed as high-rise apartments. The ONDOL system was modernized, with a gas boiler installed instead of using forest and briquette fuel. Hot water from a boiler is supplied to the floor coil, which is an X-L pipe underneath the floor surface. The thermal storage mass consists of the cement mortar in place of the stone slab. The principle of the ONDOL floor heating system has remained essentially the same, even as its form has changed. At the floor surface, heat is radiated to warm the air temperature, consequently, keeping the human body warm. Koreans enjoy using this ONDOL floor heating system. Koreans used to spend most of their time sitting on ONDOL floors, with their buttocks always in contact with the floor surface. The finishing materials used should be thermo-physiologically comfortable (Song, 2005). Various researchers have endeavored to study the physiological response of the human body to the surface of various flooring finishing materials. The recommended floor surface temperature is in the range of 19–26°C. Nonetheless, floor-heating systems can set at maximum floor surface temperature as high as 29°C (Banhidi, 1991; Fanger, 1970; Olesen, 1977).

Although previous research yielded significant benefits, it proved to be insufficient because it did not consider the Korean peoples unique habit of sitting on the floor. Consequently, a number of Korean researchers explored various flooring finishing materials and determined the ideal floor surface temperature while a person is in a sitting position of 22.0–38.8°C (Kong et al., 1988; Kang et al., 2003; Kong and Sohn, 1988; Lee and Rhee, 1992; Yoon et al., 1991).

Building, finishing and furnishing materials may emit a large number of VOCs into the indoor air. However, some laminate flooring may emit small quantities of formaldehyde and VOCs, at 23°C. It is therefore possible that laminate flooring may contaminate indoor air in the presence of floor heating. Under such conditions, the maximum temperature of the floor is 29.4°C, and the temperature at the interface between the concrete screed and the flooring may reach 50–60°C, accelerating the chemical reactions, which produce the emissions (Gustaffson and Jonsson, 1993; Wiglusz et al., 2002a,b).

This study analyses the effect of various indoor temperatures in under heating system on the emission of formaldehyde from building finishing material such as laminate flooring, plywood flooring, MDF and Particleboard. Furthermore, we examine the environmental safety of flooring materials compared with MDF and particleboard in terms of formaldehyde emission in Korean under heating system.

Experimental

Materials

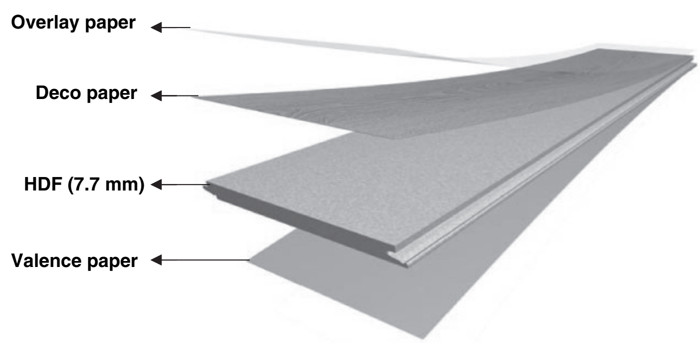
For building finishing floor materials, we chose laminate flooring and plywood flooring. Currently, those are extensively used in new apartment interiors and in the remodeling market in Korea. Laminate flooring is composed of waterproof, high-density fiberboard as core material, overlay paper, deco paper and valance paper. Each paper is impregnated with melamine-papers pressed at about 200°C in the order shown in Figure 1. Finally, the product is processed side and edge by the tenoner. In the case of plywood flooring, fancy veneer of 0.5 mm thickness; a wood such as birch, oak, beach, cherry, or maple is glued on plywood of 7.2 mm thickness and pressed at about 160°C. Ultra-violet (UV) curable coating is coated on this fancy veneer. The structures of both flooring products are shown in Figure 1. For comparison with flooring, we used MDF and particleboard veneered with decorative paper foil as furniture material and other building finishing materials. Both were 18 mm thickness. The moisture contents of these materials were showed in Table 1.

Table 1 Moisture contents of materials

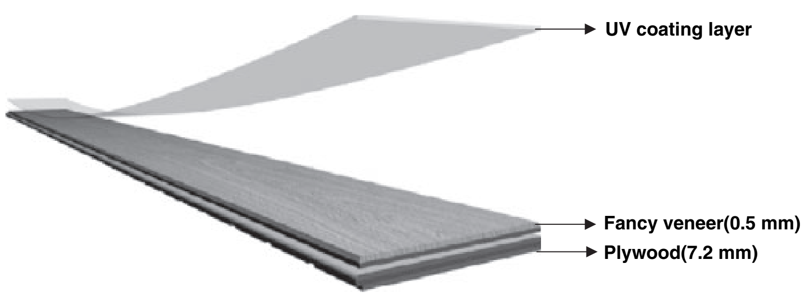
Materials	Laminate flooring	Plywood flooring	MDF	Particleboard
Moisture Contents (%)	7.2 ± 0.7	6.5 ± 0.5	7.6 ± 0.8	7.9 ± 0.9

Floor construction and indoor temperature

The floor construction for this experiment is shown in Figure 2. First, we installed two kinds of floorings on 9.5×9.5 m of square floor. Each flooring was installed on half of the floor. For the under heating system, a 15 mm diameter copper pipe was installed with 50 mm narrow pitch in a 70 mm thick cement mortar. In the case of laminate flooring, the flooring method was used with PVC vinyl and 0.2 mm polyethylene foam. The laminate flooring was not glued with cement mortar. Hot water from a boiler was supplied to the floor coil, which was an X-L pipe underneath the floor surface.



(a) Laminate flooring



(b) Plywood flooring

Fig. 1 Structure of laminate flooring and plywood flooring

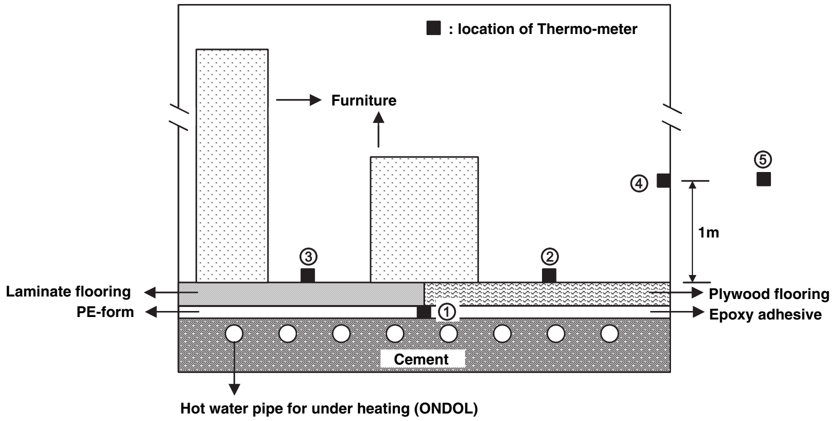


Fig. 2 Floor construction and temperature measurement

Plywood flooring was glued on the surface of the cement mortar with water based, epoxy adhesives.

Second, thermocouples were fixed, (1) on the surface of the cement mortar, (2) on the surface of the laminate flooring, (3) on the surface of the plywood flooring, (4) on the wall 1.2 m above the floor, and (5) outside, to measure the temperatures by the data logger (Advantest R7326B). The data logger is compact and light weight portable loggers suitable for on-site measurement in automobile, machine industries and plants for simplified logging of electrical component data; temperature/voltage/pulse. Other building materials, MDF and particleboard, existed in the space Even though each flooring material was only exposed to hot cement mortar containing the hot water pipes, we checked the temperature on the wall 1.2 m above the floor. We measured the temperature for 8 h from 10:00 p.m. to 6:00 a.m. in early spring season. We controlled the under heating temperature to maintain the indoor temperature at 20°C.

Determination of formaldehyde emission

The Japanese standard method with a desiccator's (JIS A 1460) was used to determine the formaldehyde emissions from the laminate flooring, plywood flooring, MDF and particleboard veneered with decorative paper foil. The test for determination of quantity of formaldehyde emitted from building boards by, desiccator's method is carried out by, using the glass desiccator's. The emitted quantity of formaldehyde is obtained from the concentration of formaldehyde absorbed in distilled water or deionized water when the test pieces of specified surface area placed in the desiccator's filled with the specified amount of distilled water or deionized water and 24 h has elapsed. The principle for determination of concentration of formaldehyde absorbed distilled water or deionized water is based on the Hantzsch reaction in which the formaldehyde reacts with ammonium ions and acetylacetone to yield diacetyldihydrolutidine (DDL) (Japanese Industrial Standard, 2001).

The 24-hour desiccator's method uses a common glass desiccator's with a volume of 10 l. Eight test specimens, with dimensions of 5×15 cm were positioned in the desiccator. The emission test lasted 24 h in the covered desiccator's at a temperature of 20°C. The emitted formaldehyde was absorbed in a water-filled Petri dish and was analyzed by means of the chromotropic acid method. From the results of temperature measurement, we set the temperature of the dry oven, to 20°C, 37°C and 50°C. First, the materials, laminate flooring, plywood flooring, MDF and particleboard, were conditioned in the dry oven at 20°C, 37°C and 50°C for 1, 2, 3, 5, 7, 10, 15, 20 and 28 days to control the same condition with under heating system; ONDOL (Figure 3). Second, we measured formaldehyde emission from these materials by desiccator's method (JIS A 1460). This process is shown in Figure 3.

Results and discussion

Indoor and floor surface temperature

During the experiment, the outdoor temperatures of the laboratory were in the range of 7.5–8.5°C for 8 h, as shown in Figure 4. We set the under heating temperature for a suitable indoor temperature (measuring height was 1.2 m) of about 20°C, at which the temperature of the cement mortar surface was about 50°C. The temperatures of the floor surfaces laminate flooring and plywood flooring all showed variations, although the temperature of the cement mortar surface was consistent same. The mean floor surface temperature ranged from 35.0 to 37.0°C for the laminate flooring and from 36.0 to 38.0°C for the plywood flooring. The floor surface temperature depends on the thermal characteristics of the covering materials. The higher the thermal resistance, the lesser the temperature

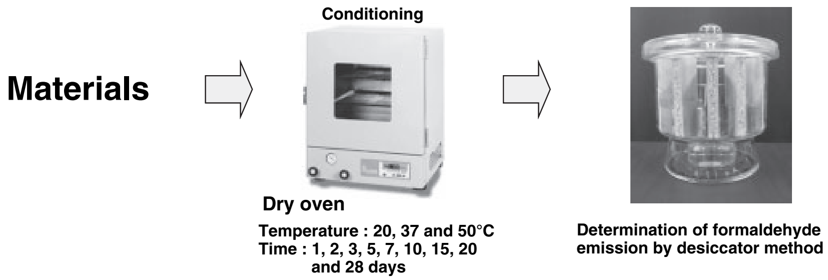


Fig. 3 Temperature changes of measuring points in the under heating system

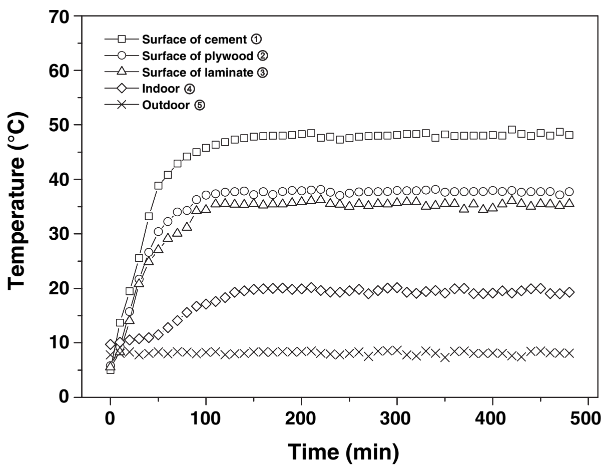


Fig. 4 Temperature conditioning of materials and desiccator method

fluctuation. Wood covering is known to resist the heat flow from the hot water to the floor surface because of its thermal resistance. According to the research of Cho (1996), however, finishing materials with high thermal resistance retained heat longer.

Because of the different installation systems of laminate and plywood floorings, the temperature of the laminate flooring surface was lower than that of the plywood flooring. The floating and non-glued installation method of laminate flooring affected the thermal conductivity. The empty gaps between the laminate, PE foam and cement mortar reduced the thermal conductivity from the under heated cement mortar to the laminate flooring surface. The surface temperature shows the ability of heat flux generated from the covering material. It is required to maintain the heating load. Residential buildings in Korea have high-quality insulation. In recent years, wall insulation and double skin structure on the exterior envelope have become popular. Thus, the heating load has decreased, and the Korean under heating (ONDOL) system has increased the use of wood covering for its floors. The choice of floor finishing material should be determined by two factors: the ability to manage heat flux based on the heating load and the comfort it provides to the human body (Song, 1996). The most important result from this experiment was that the floor, furniture and building finishing materials all faced different temperatures in the same space with the ONDOL system. We can therefore presume that formaldehyde emission from each installed building material will be different.

Formaldehyde emission in under heating system

The initial formaldehyde emission results of each flooring material, and from MDF and particleboard with decorative paper foil, obtained by desiccator's method are shown in Figure 5. Each material was tested three times. The emission of laminate flooring was 1.44 mg/l and of plywood flooring was 0.63 mg/l. Both these floor materials were under E1 grade in formaldehyde emission. Generally, laminate flooring is manufactured as E1 grade in Europe. The greatest effect on formaldehyde emission in laminate flooring is exerted by, High Density Fiberboard (HDF), the core of laminate flooring. It was made by UF resin or urea-melamine-formaldehyde condensed resin. This grade of laminate flooring can be used for residences. Because the plywood that was used as core in plywood flooring

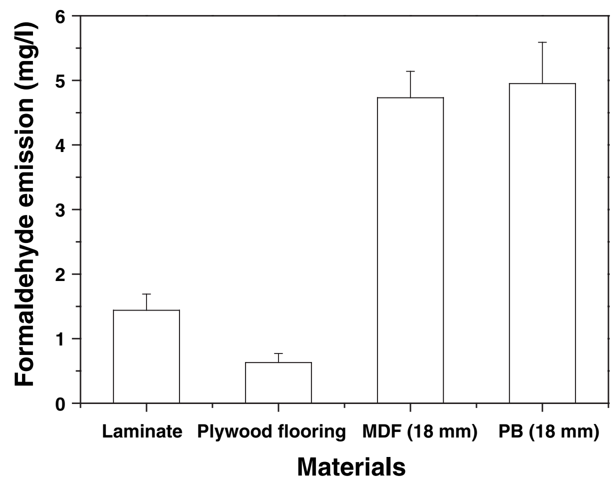


Fig. 5 Formaldehyde emission from each building and furniture material by desiccator's method

was glued with PF resin, its formaldehyde emission was lower than that of laminate flooring. E1 grade of wooden flooring materials has been circulated in Korea. To maintain under E1 grade, the manufacturers use MUF (melamine-urea-formaldehyde) condensed resin for HDF and PF resin for plywood as adhesives. On the other hand, the MDF and particleboard furniture materials in the experiment were veneered with decorative paper foil, with a formaldehyde emission of E_2 grade. The emission of MDF was 4.73 mg/l and that of particleboard was 4.95 mg/l. The highest initial emission level was observed with the particleboard that was veneered with decorative paper foil. In Korea, MDF and particleboard of grade more than E_2 are circulated and used for furniture manufacture. From Wigluszs result (Cho, 1996), formaldehyde emission was reduced when wood-based panels were veneered with decorative paper foil. However, the sample we used for this study still emitted a lot of free formaldehyde even though it was veneered with decorative paper foil. This indicates that these panels would be more than E_2 grade. The authors considered that furniture materials give a more serious effect to the human body in indoor air quality problem than flooring materials.

From the results of measuring the temperature in the under heating system, we analyzed the formaldehyde emission of flooring and furniture materials at the dry oven temperatures of 20, 37, and 50°C.

Comparison of formaldehyde emission from each material at 20, 37, and 50°C by desiccator's method for 4 weeks is shown in Figure 6. It is well known that formaldehyde emission is decreased with time. Especially, in the case of flooring materials at 37 and 50°C, a very low formaldehyde emission was observed at 28 days. At room temperature, furniture materials of E_2 grade were still about 4 mg/l at 28 days. Furthermore, in the case of flooring materials at 28 days, free formaldehyde was emitted from the flooring materials at the same level as the initial emission level. This confirms the necessity of bake-out (Bayer, 1990; Bayer, 1991; Follin, 1997; Offerman et al., 1993).

Because of the under heating system, the flooring materials are exposed to 37 and 50°C, while the furniture materials mostly exist at room temperature. At 37 and 50°C, the formaldehyde emission level of the flooring materials was already under 0.3 mg/l (F☆☆☆☆ level by JIS A 1460, application possibility without area limit) after 10 days and the emission had decreased further (0.03–0.10 mg/l) after 28 days. These levels are it is not injurious to the human body and will not cause sick house syndrome (SHS).

The problem, however, is the furniture materials such as MDF and particleboard. Because these materials

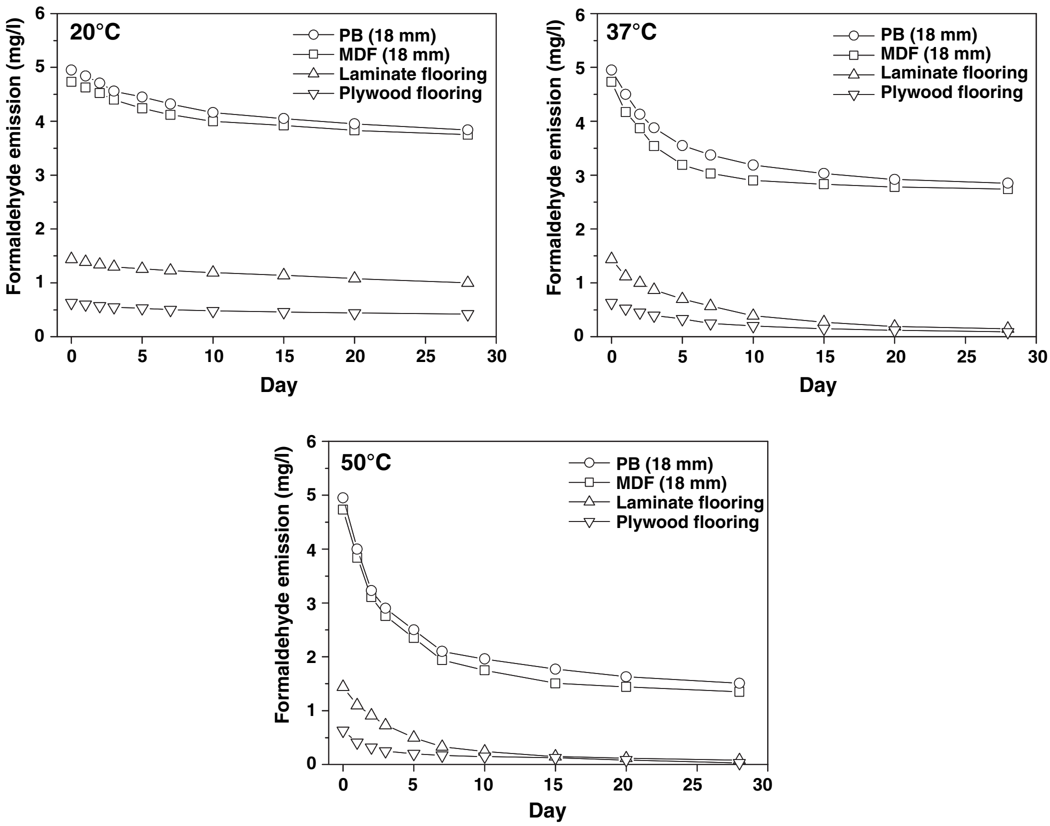


Fig. 6 Change of formaldehyde emission over time from each material at 20, 37, and 50°C

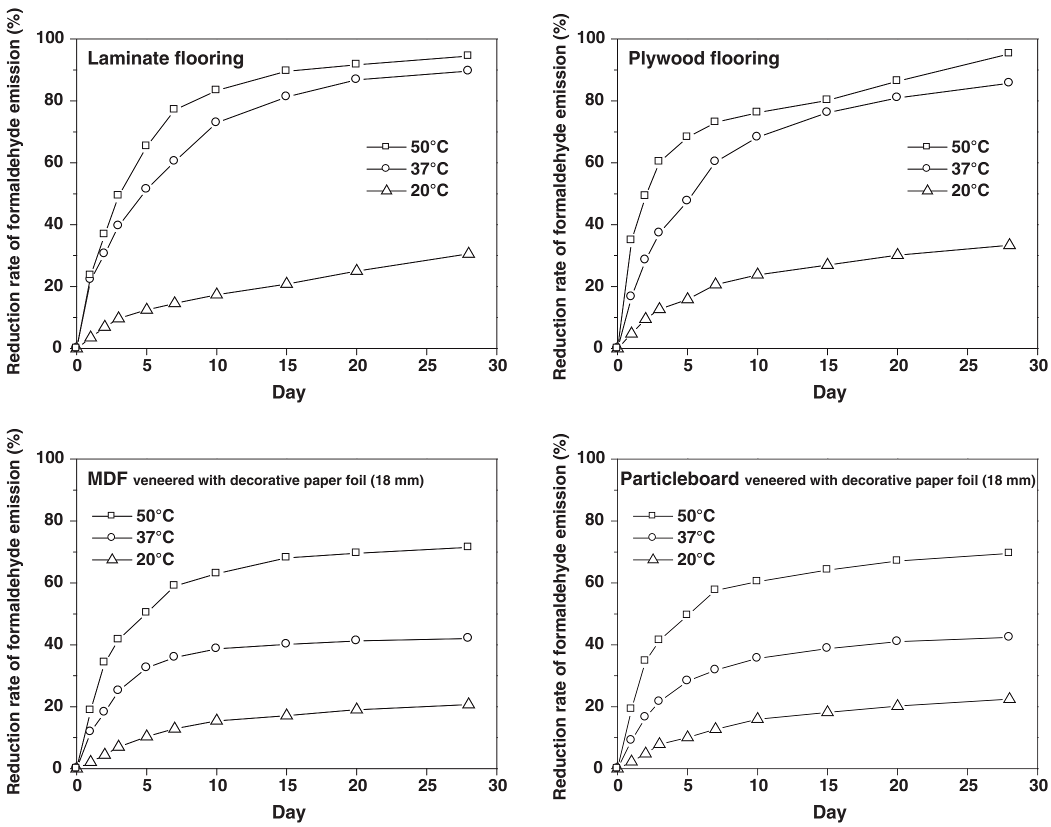


Fig. 7 Reduction rates of formaldehyde emission of flooring materials and furniture materials

are not exposed to high temperature (50°C in this experiment) in living condition, an E_2 grade of formaldehyde emission level at room temperature still remained after 28 days. Although there will be variations with the volume of furniture materials and the indoor conditions, furniture materials are the principal cause of indoor air quality pollution in the Korean under heating system.

Figure 7 shows the reduction rate of formaldehyde emission of each material with time and temperature. The flooring materials, shows a much higher reduction rate of formaldehyde emission at high temperature than the furniture materials does. After 28 days, the reduction rates of formaldehyde emission of the flooring materials were about 90%, compared to 30% for the furniture materials. On the other hand, at room temperature, the reduction rates of formaldehyde emission of the flooring materials were almost the same as, or a little higher than, those of the furniture materials. Because the flooring materials were of 8 mm thickness and their formaldehyde emission level was already E_1 grade (under 1.5 mg/l), their free formaldehyde can escape easier than from thick furniture materials (18 mm) at high temperature. On the other hand, the reduction rates of formaldehyde emission for the furniture materials were directly proportional to increasing temperature.

Figure 8 shows the formaldehyde emission of each material in the heating system laboratory. While the formaldehyde emissions of the furniture materials that faced high temperature were still 4 mg/l, those of the flooring materials were under 0.1 mg/l. These results confirm that greater harm is caused by furniture materials than flooring materials in terms of indoor air quality in Korean under the heating system (ONDOL) after bake-out.

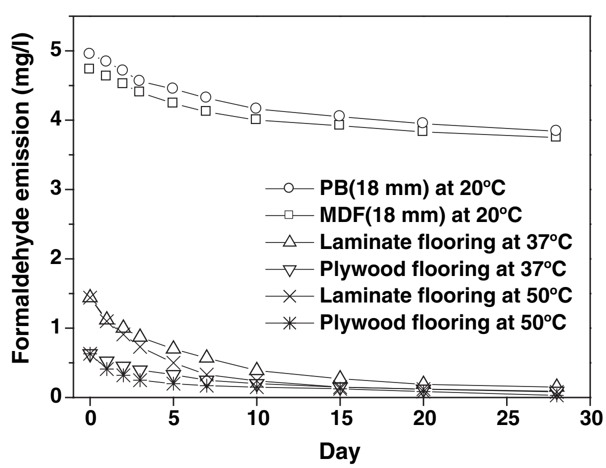


Fig. 8 Formaldehyde emission of each material during under heating

Conclusions

From the results of measuring the temperature in the under heating system, we analyzed the formaldehyde emission of flooring and furniture materials at the dry oven temperatures of 20, 37 and 50°C. Because of the under heating system, the flooring materials are exposed to 37 and 50°C, while the furniture materials mostly exist at room temperature. Flooring materials were exposed to temperatures of 37 and 50°C while furniture materials were only exposed to room temperature during under heating. After bake-out the formaldehyde emissions of flooring materials were much lower than those of furniture materials. Although the results depend on the volume of the installed, building finishing materials, furniture materials such as MDF and particleboard are the principal offenders of indoor air quality pollution in Korea. Furthermore, it is necessary that the temperature factor should be considered for the management of indoor air quality because we live with the under heating system (ONDOL).

Acknowledgements

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