



Control of formaldehyde and TVOC emission from wood-based flooring composites at various manufacturing processes by surface finishing

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ABSTRACT

This paper assesses the reproducibility of testing formaldehyde and TVOC emission behavior from wood flooring composites bonded by urea–formaldehyde resin at various manufacturing steps for surface finishing materials. The surface adhesion step of laminate flooring for this research was divided into two steps; HDF only and HDF with LPMs. In the case of engineered flooring, the manufacturing steps were divided into three steps; plywood only, fancy veneer bonded on plywood and UV coated on fancy veneer with plywood. Formaldehyde and VOCs emission decreased at the process of final surface finishing materials; LPMs were applied on the surface of HDF for laminate flooring. Although emissions increased when fancy veneer was bonded onto plywood in the case of engineered flooring, emission was dramatically reduced up to similar level with plywood only when final surface finishing; UV-curable coating was applied on fancy veneer. This study suggests that formaldehyde and VOCs emission from floorings can be controlled at manufacturing steps for surface finishing.

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1. Introduction

In renovated or completely new buildings, levels of indoor air pollutant emissions from construction and building materials, especially of VOCs, are often several orders of magnitude higher than the VOC levels in buildings under normal use [1–4]. The use of small-scale environmental chambers with volumes ranging from a few liters to a few cubic meters has been increasing [5]. The weakness of these traditional chamber techniques is that they cannot be used to investigate emissions from existing real building structures. The emission of VOCs from a material in a real building structure is affected not only by the material but also by the environmental conditions and other surrounding materials [6]. Secondary emissions can develop under the influence of humidity, ozone, UV-light, etc. [7,8]. Hydrolysis reactions in the floor structure (PVC/adhesive/casein containing leveling agents) can produce 2-ethylhexanol, butanol and ammonia [9]. Thus, the emission measured on site can differ considerably from the emission measured from a single material under laboratory conditions [10]. The impact of the increased consciousness about indoor environment has created a demand for low-emitting (healthy) building materials, and hence also for standardized methods to characterize and quantify the VOC emissions from building materials and consumer products. Furthermore, methods for easy source identification of potential VOC emission from building materials on site and for their quan-

tification are required. Therefore, the field and laboratory emission cell (FLEC) has been proposed and has become a European standard for emission testing (prENV 13419-2, 1998). This is a kind of micro-emission cell featuring a high sensitivity due to the large loading ratio (surface area/volume). Nowadays, a large percentage of emission tests for various materials are performed with FLEC [11–13]. As a pre-test method for TVOC emission from building material, the VOC analyzer was successfully applied. There was a good correlation between TVOC emission concentration by the VOC analyzer and TVOC EF by the standardized 20 L small chamber method in the case of paints [14,15].

Recently, there have been many concerns about human health and the environment. PVC flooring, laminated paper lacquered with bean oil, which was used in most Korean houses has been replaced by wood flooring materials, especially in new apartments [16]. There are three kind of flooring composites; engineered flooring, laminate flooring and parquet flooring used in Korea. Among these, parquet is made from thick veneer which is more expensive than the others for normal living rooms. Therefore, engineered flooring and laminate flooring are commonly used in new apartment interiors and in the remodeling market in Korea. Laminate flooring is a composite floor with either a chipboard or HDF core that is bonded to a film of wood effect veneer and covered with a laminated surface; overlay paper, deco paper and valance paper. Each paper is impregnated with melamine-papers pressed at about 200 °C. A clear cap sheet made of an aluminum oxide saturated film (overlay paper) and a balancing backing are also put on the top of deco paper and on the back of the panel, respectively. The main purpose of the alu-

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Table 1
Composition of melamine–formaldehyde resin for impregnation.

Composition	Amount added (g)
Melamine–formaldehyde resin	100
Hardening agent	0.82
Plasticizer	2
Release agent	0.4
Water	3

minimum oxide film is to protect the surface against any stain [17]. Finally, the product is processed on the sides and edge with the tenoner. In the case of engineered flooring, fancy veneer of 0.5 mm thickness; woods such as birch, oak, beach, cherry, or maple are glued on plywood of 7.2 mm thickness and pressed at about 160 °C. UV-curable coating is placed on this fancy veneer [18].

Before they can be used as furnishing materials, wood-based panels have to be treated to match the specific requirements of their final use. Therefore, finishing treatment methods which produce an overlay or coating, such as paints, prints, varnishes, veneers, laminates, impregnated papers, finishing foils, etc., are used to reduce the absorption of water and humidity, and eliminate the release of harmful gases [19]. Interior fitment and furniture manufacturers are using more surfacing materials for decorating fiberboard. This material is manufactured as uniform, flat panels that provide excellent surfaces for the application of coating materials [20,21]. These surface materials, such as decorative vinyl film and melamine impregnated paper, can lower the formaldehyde emission concentration from wood-based panels [22]. Nemli examined the effects of the coating materials process parameters on the technological properties of particleboard and stated that the surface coating decreased the formaldehyde emission [23].

In this study, FLEC was used to measure formaldehyde and VOCs emission levels from engineered flooring and laminate flooring throughout the manufacturing steps and to compare the VOCs results with those obtained from the VOC analyzer. Especially, it was used to investigate the effect of various manufacturing steps of surface finishing treatment to TVOC/formaldehyde emission behaviors of each flooring.

2. Experimental

2.1. Materials

Among the various floorings, we chose laminate flooring and engineered flooring. Currently, these are extensively used in new apartment interiors and in the remodeling market in Korea. Laminate flooring is composed of waterproof, high-density fiberboard (HDF) as the core material, with overlay paper, deco paper and valance paper. The wood fiber, for HDF, distributed from Dongwha Enterprise was made from Korean pine (*Pinus densiflora*) with a moisture content of 4%. Urea–melamine–formaldehyde (UMF) resin was used to manufacture HDF. The molar ratios of the resin was 0.95 (F/M + U). Before the HDF was manufactured, 3 parts (to resin) of 25% ammonium chloride as a hardener and 13 parts of 44% wax solution for water-proofing were added. The wood fiber distributed from Dongwha Enterprise was made from Korean pine (*P. densiflora*) with a moisture content of 4%. The MDF was manufactured using the above adhesives, in order to have a specific gravity of 0.8. The mixture was then hot pressed, to form composite boards, at a peak pressure of 40 kgf/cm² and a temperature of 180 °C. The main pressing time was 4 min and the pressure was then released in two steps of 1 min each. Overlay paper, deco paper and valance paper were impregnated with melamine–papers pressed at about 200 °C and called low pressure melamine (LPM) as shown in Fig. 1a. The resin for impregnation is shown in Table 1. Finally, the edges of the

product are machined to produce a tongue and groove profile. In the case of engineered flooring, 0.5 mm-thick fancy veneer of maple is glued to a 7.2 mm-thick plywood sheet and pressed at about 160 °C. A UV-curable coating is coated on this fancy veneer as shown in Fig. 1b. The UV-curable urethane acrylate system consisted of three main components; urethane diacrylate oligomers, reactive diluent monomer and photoinitiator. The manufacturing step of laminate flooring for this research was divided into two steps; HDF only and HDF with LPMs. In the case of engineered flooring, the manufacturing steps were divided into three steps; plywood only, fancy veneer bonded on plywood and UV coated on fancy veneer with plywood.

2.2. Methods

2.2.1. Field and laboratory emission cell (FLEC)

The FLEC was used as a micro-emission cell in this experiment [24]. When the circular stainless steel cell is put onto the surface of the planar test material, the material surface becomes an integral part of the cell itself. The material surface area exposed to airflow inside the FLEC is 177 cm² and the internal volume of the cell is 35 mL. The FLEC was supplied with purified and humidified air at a given ventilation rate. The temperature (20 °C) and relative humidity (50 ± 5%) inside the chamber were kept constant. The emission sample was collected after 5 min of equilibration time and 5 min of cleaning time under the FLEC lid at an airflow of 250 mL/min. For formaldehyde, 4.5 L of gas were collected in a 2,4-DNPH cartridge

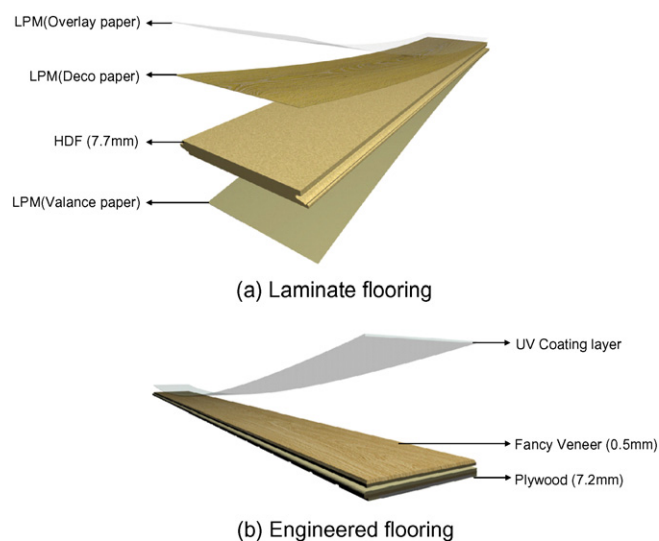


Fig. 1. Structure of laminate flooring and engineered flooring.

Table 2
Test condition of FLEC.

Test condition	FLEC
Sample area	0.0177 m ²
Volume	0.035 L
Loading factor (area of sample/volume)	504.64 m ² /m ³
Air change rate	428.57 h ⁻¹
Air supply	250 mL/min
Equilibration time	Sampling after 15–30 min
Temperature/humidity	23 ± 2.0 °C/50 ± 5%
Compounds, sampling flow and total sampling	VOC: 50 mL/min, 1.5 L formaldehyde: 150 mL/min, 4.5 L
Inlet air	High purity air
Background concentration	VOC: 2 µg/m ³ , TVOC: 20 µg/m ³
Cleaning process	Vacuum oven or cleaning by methylene then high purity air for 1 day
Analysis method	VOC: GC/MS, formaldehyde: HPLC

Table 3
Analysis conditions of HPLC for formaldehyde.

Variables	Formaldehyde analysis condition
HPLC	Agilent HP1100
Detector	UV-vis 365(Bw.30), ref. 590(Bw.10)
Column	Supelco C18. 4.6 mm × 250 mm
Mobile phases	Acetonitrile:water = 45:55
Analysis time	25 min
Injection volume	20 μ L
Column temperature	40 °C
Mobile phase flow rate	1.0 mL/min

Table 4
Analysis conditions of GC/MS for VOCs.

Variables	VOCs analysis condition
TDS	PerkinElmer ATD400
GC/MS	HP6890/Agilent5973
Column	RTX-1 (105 m × 0.32 mm × 3 μ m)
Carrier gas and flow	He (99.99%)
Temperature program	40 °C (5 min) → 70 °C (5 min) → 150 °C (5 min) → 200 °C (5 min) → 220 °C (5 min) → 240 °C (5 min)
MS condition	
Mode	EI (electron ion)
Electron energy	70 eV
Detection mode	TIC (scan), m/z : 35/350

for 30 min under a gas flow rate of 150 mL/min while 1.5 L of gas was collected in a Tenax-TA tube for 30 min under a gas flow rate of 50 mL/min. The condition of the FLEC for the correction gas is listed in Table 2. The replicates of each sample were three times.

Formaldehyde and VOCs were analyzed by HPLC and TDS/GC-MS, respectively, as listed in Tables 3 and 4. In this paper, TVOC was defined as the conversion of all peak areas between C_6 and C_{16} to concentrations using the toluene response factor. A peak area under 10 was defined as the limit of detection. The sample gas was taken with Tenax-TA and 2,4-DNPH cartridges. The calculation of the emission factor (EF) is explained in ASTM D5116. Two technical terms of EF and ER are commonly used to describe the rate of emissions from indoor materials, and are related as follows:

$$ER = A(EF)$$

where ER is the emission rate (mg/h); A is the source area (m^2); EF is the emission factor ($mg/m^2 h$).

2.2.2. VOC analyzer

The VOC analyzer is a portable device to measure the four main aromatic hydrocarbon gases: toluene, ethylbenzene, xylene and styrene. To prepare the samples for the VOC analyzer, the each flooring samples were conditioned at 20 °C and 50 ± 5% in a thermo-hygrostat for 15 days, then cut into four pieces measuring 50 mm × 50 mm and placed in a 3 L polyester plastic bag. The polyester plastic bag was sealed with teflon tape, purged three times with N_2 gas, and then filled with N_2 gas by pulling up the plunger. For the blank control, an empty bag with N_2 gas was prepared. The gases for the VOC analyzer were collected from the 3 L polyester plastic bag in a gas tight (0.5 cc) manner after 4 days, placed into the VOC analyzer and analyzed. This process is shown in Fig. 2. This 4-day period was determined by the authors' previous study to find the optimum method for VOC emission test by the VOC analyzer [25]. The measurement procedure comprised three steps. First, the product was inserted into the 3 L polyester plastic bag. Then, the plunger was slowly pulled, pushed in again, and pulled out for the second time before the syringe was removed from the plastic bag. If the top of the syringe was wet, it was wiped dry with a tissue. A dedicated needle was attached and 0.5 cc (1/2 calibration) of the sampled gas was ejected by pushing the plunger. The remaining gas was injected into the inlet on the main unit of the VOC analyzer, after which the measurement was automatically started [26].

3. Results and discussion

Generally laminate flooring is manufactured to satisfy the E₁ grade in Europe. The greatest influence on formaldehyde emission in laminate flooring is exerted by HDF, which is the core of laminate flooring. This grade of laminate flooring can be used for residences. E₁ grade wooden flooring materials have been circulating in Korean flooring market [27]. The formaldehyde emission results obtained by FLEC for each flooring samples are shown in Fig. 3. When LPMs were applied on each side of the HDF core, formaldehyde emission was reduced compare to HDF only. Although LPM is made by impregnation into melamine-formaldehyde resin, the effect of formaldehyde reduction appeared when it was used as a cover-

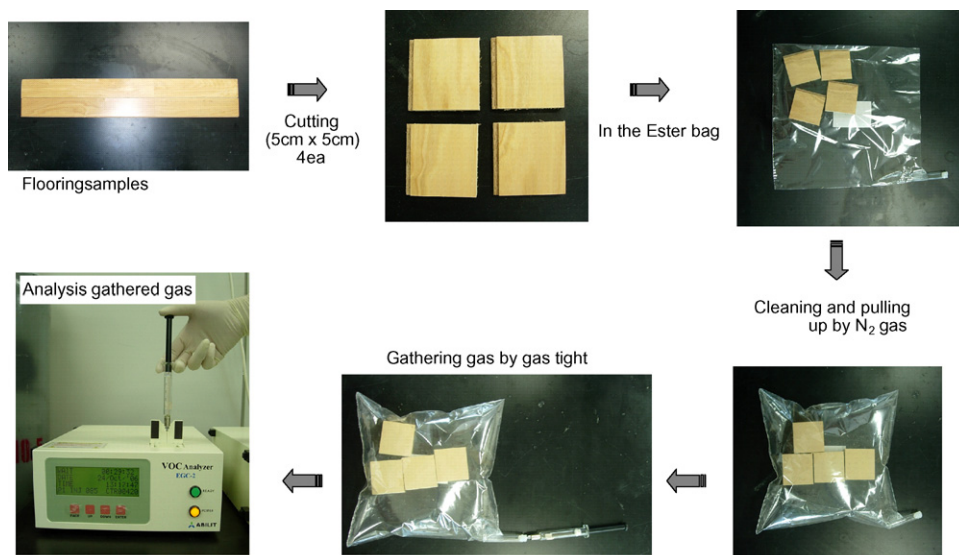


Fig. 2. Test method for volatile organic compound (VOC) emissions from engineered flooring by the VOC analyzer [26].

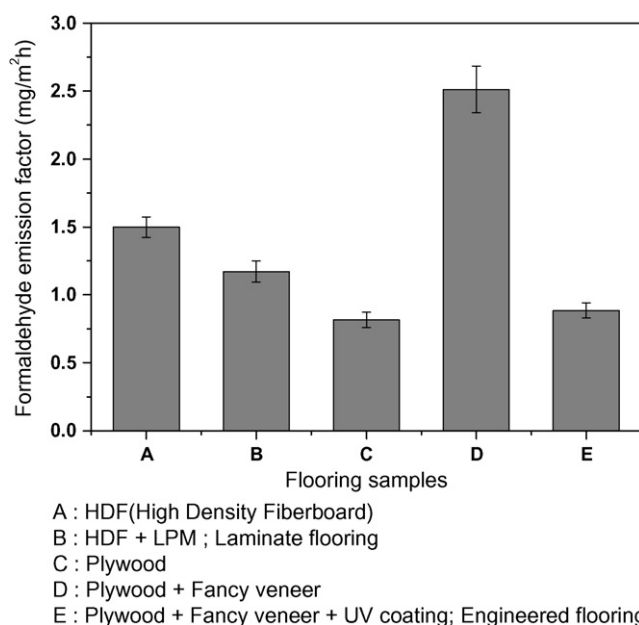


Fig. 3. Formaldehyde emission behaviors of flooring materials at various manufacturing steps for surface finishing materials by FLEC.

ing material of HDF. In the case of engineered flooring, because the plywood that is used as the core in plywood flooring is glued with phenol–formaldehyde (PF) resin, its formaldehyde emission is lower than that of laminate flooring. However, formaldehyde dramatically increased when fancy veneer was bonded on plywood. Usually, manufacturers use formaldehyde-based resin, such as UF resin and UMF resin, for fancy veneer bonding. This adhesive produced three times more formaldehyde emission compared to plywood only. The final step for engineered flooring is UV-curable coating on fancy veneer. When UV coating was applied on fancy veneer, the formaldehyde emission returned to its lowest point that of plywood only. Basically, UV-curable coating was for surface roughness of soft fancy veneer which is the surface of engineered flooring [28]. It produced reduced formaldehyde emission because it acted as a covering material like LPM in laminate flooring. This sealing effect can be checked from the author's report [29]. When the edges of wood-based composites were sealed by finishing materials, formaldehyde emission was reduced dramatically compared to unsealed edges.

VOC emission behavior of laminate flooring and engineered flooring showed difference with formaldehyde emission. Engineered flooring was higher than laminate flooring even though plywood flooring was glued with PF resin which had the characteristic of low formaldehyde emission. Volatile organic compounds from flooring materials at various manufacturing steps and TVOC calculated between C₆ and C₁₆ are shown in Table 5 and Fig. 4. Besides formaldehyde, wood emits a variety of volatile compounds such as terpenes and some organic acids. The amount of other VOCs released from wood depends highly on the wood species. Hardwoods like beech and oak release mainly high amounts of acetic and formic acid and less terpene compounds, whereas softwoods release much less organic acids but much more terpene compounds [30]. Monoterpene compounds such as α -pinene, β -pinene and 3-carene originating from softwoods and similar products are the most important VOCs. Diterpene compounds, which are less volatile, can be emitted at a relatively high temperature and in comparatively small amounts [31]. Sundin et al. [32] studied the VOCs from wood and wood products and found that about 80% of the VOCs from green wood are monoterpenes and only 1% free aldehydes. In PBs, the authors found a different pattern with monoterpene

Table 5

Volatile organic compounds from flooring materials at various manufacturing steps for surface finishing materials by FLEC, as detected by GC/MS analysis (unit: $\mu\text{g}/\text{m}^2 \text{ h}$).

Flooring samples	A	B	C	D	E
Benzene	5.55	0.94	2.36	6.94	3.83
Toluene	65.66	29.98	115.96	179.11	62.41
Ethylbenzene	6.94	5.05	13.07	16.86	6.85
Styrene	8.55	0.87	5.77	10.22	3.79
<i>o</i> -Xylene	9.98	6.03	7.66	9.77	4.98
<i>m,p</i> -Xylene	18.65	15.23	25.79	29.65	14.78
1,3,5-Trimethylbenzene	0.00	0.00	1.50	2.41	1.15
Identified total	115.31	58.10	172.11	254.95	97.79
Unidentified total	980.12	612.44	1166.21	850.32	662.22
Total concentration	1095.45	670.54	1338.33	1855.37	760.02

A: HDF (high-density fiberboard); B: HDF + LPM; laminate flooring; C: plywood; D: plywood + fancy veneer; E: plywood + fancy veneer + UV coating; engineered flooring.

compounds comprising only 20–22% and aldehydes levels as high as 27–32%. The emission of VOCs from softwoods depends upon a number of factors including the age of the wood and the time after cutting down for the manufacturing process of wood products such as flooring and furniture [33]. However, many natural VOCs such as α -pinene and β -pinene are emitted from wood-based panels. In Korea, the Ministry of Environment provides guidelines for VOC emissions from building materials in terms of 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. Because more than 90% of the emissions are unidentified VOCs, 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 -pinene and β -pinene, are selected as harmful VOCs when TVOCs are calculated [26]. However, the tendency of TVOC emission in each manufacturing step was similar with formaldehyde emission. Although TVOC emission increased when fancy veneer bonded on plywood was used, it decreased with LPM covered for laminate flooring and UV-curable coating for engineered flooring.

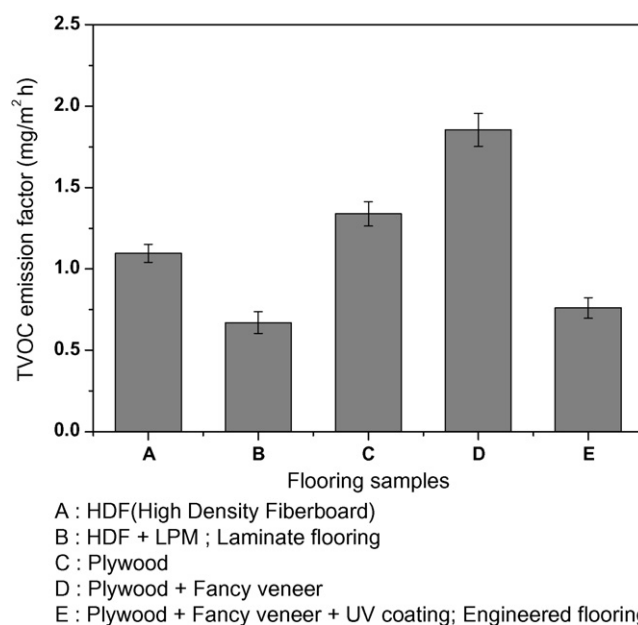


Fig. 4. Total volatile organic compounds emission factors of flooring materials at various manufacturing steps for surface finishing materials by FLEC.

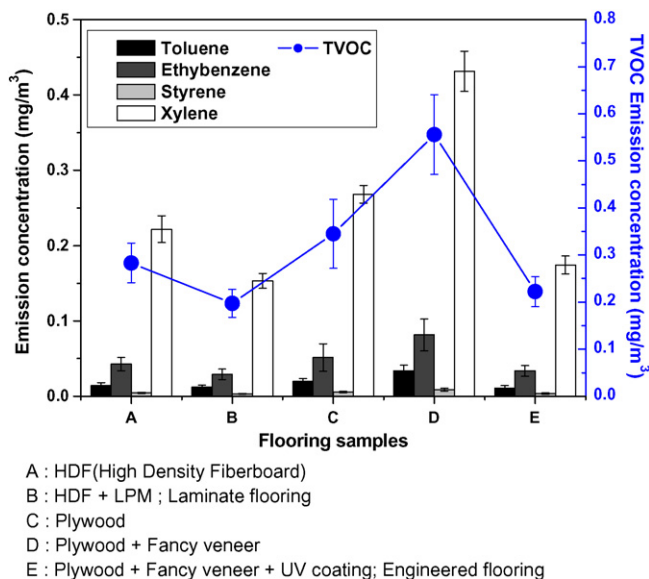


Fig. 5. VOC emission concentrations (toluene, ethylbenzene, xylene and styrene) of flooring materials at various manufacturing steps for surface finishing materials as determined by the VOC analyzer.

Fig. 5 presents concentrations of the four indicated VOCs from each sample, as determined by the VOC analyzer. From all samples, xylene was the highest detected compound. The second highest detected compound was ethylbenzene, followed by toluene and styrene. In the VOC analyzer test, we defined TVOC as the sum of the four detected main aromatic hydrocarbon gases: toluene, ethylbenzene, xylene and styrene. Although it was hard to compare directly the FLEC results because these data were based only on the sum of four VOC compounds, when it was compared to the results of Table 5, the VOC analyzer was found to be a suitable pre-test method for application as a TVOC emission test [26]. The TVOC emission results were also shown to be similar to those of FLEC.

4. Conclusion

Flooring is one of major formaldehyde and TVOC emission wood products. However, formaldehyde and TVOC emission can be reduced during the manufacturing process if the emission levels at each manufacturing steps are found. When LPM and UV coating were applied on HDF for laminate flooring and fancy veneer with plywood for engineered flooring, formaldehyde and TVOC emission decreased because these LPM and UV coatings functioned as a covering material. Although formaldehyde and TVOC emission dramatically increased when fancy veneer was bonded on plywood with formaldehyde-based resin, UV-curable coating returned it to the same emission levels of plywood. FLEC was a useful test method for flooring materials during manufacturing steps because this method considered the surface of samples. Knowing the impact of such an interaction on the emission of volatile organic compounds and change in the distribution of VOCs qualitatively and quantitatively during the different process steps of flooring products could pave the way to understand the release behavior of the VOCs and to decrease their emission Roffael [33]. A deep understanding of the process variables affecting the release of VOCs from wood products such as flooring during various manufacturing steps with different wood species is needed. Therefore, formaldehyde and VOCs emission from floorings can be controlled at manufacturing steps for surface finishing.

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