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Formaldehyde emission and VOCs from LVLs produced with three grades of urea-formaldehyde resin modified with nanocellulose

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ABSTRACT

Three grades of liquid urea-formaldehyde (UF) resins with different formaldehyde emission (HCHO) levels such as super E0 (SE0), E0 and E1 were modified by adding different amounts of microfibrillated cellulose (MFC, 5 wt%). The laminated veneer lumbers (LVLs) were produced from the unmodified and modified SE0, E0, and E1 grade resins. The total volatile organic compounds (TVOC) and HCHO of the LVLs were determined at 25 °C, 35 °C, and 45 °C for 30 min using a thermal extractor. The highest VOC emitted from the LVLs was found to be toluene, followed by xylene, benzene, and ethyl-benzene, respectively. Styrene, however, was not detected at all in any of the systems. The incorporation of the MFC into the SE0 up to 30 wt% significantly decreased the formaldehyde emission from of the UF resin while this not observed for E0 and E1 grade resins. The TVOC from the LVLs considerably decreased with increasing MFC content at 25 °C and 35 °C. The use of MFC in the UF resin can be environmentally friendly solution for reducing the TVOC from the wood-based panels, in particular for indoor furniture at 25 °C and 35 °C.

1. Introduction

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. Building materials can release wide range of pollutants. One of the hazardous pollutants of the indoor air are volatile organic compounds (VOCs) emitted from wood-based panels, which can cause indoor air related health problems. More than two hundred volatile organic compounds (VOCs) have been identified in the indoor environment [1,2]. Formaldehyde and VOCs emitted from wood-based panels has become one of the major causes of degrading indoor air quality, which can negatively affect human comfort, health and productivity. In Korea, the Ministry of Environment provides total VOC (TVOC) guidelines for building materials. Even natural VOCs from wood are considered to be harmful and are included in the TVOC calculation [3].

Wood-based panels are important materials for the construction of homes, buildings and floor furniture making. In particular, laminated wood materials produced with synthetic resins such as urea-formaldehyde or melamine formaldehyde are used as laminated wood flooring. Underfloor heating system is quite popular in many countries, such as Korea and in many areas of Europe, and its popularity is increasing at a rapid rate. People 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 [4]. Indoor environment quality has a significant impact on modern life around the globe. Americans, on average, spend approximately 90% of their time indoors. For this reason, the reduction of VOCs and formaldehyde emission emitted from the laminated flooring material are very important to protect

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human health, in particular for kids and babies having a weak immune system. According to the "Guidelines for Air Quality" published by the World Health Organization (WHO), the main health effects of indoor air pollution include irritation, respiratory infection, and Sick Building Syndrome (SBS), among others. Hence, to guarantee the health of occupants in buildings, it is important to maintain high indoor air quality [5].

UF resin is the major resin of the wood industry because of its high bonding strength and low cost. One of the most important factors in this area is the mole ratio of formaldehyde to urea in UF resin [6]. With increasing concern about environmental pollution and from the formaldehyde of the UF resin, researchers has focused on environmentally friendly polymers for reducing the amount of free-formaldehyde. The formaldehyde content of UF resins has been gradually lowered over the years because it is one of the most effective ways of reducing formaldehyde emission. The emission of formaldehyde from wood-based panels decreases as the molar ratio of formaldehyde falls [7], but unfortunately, the other physical and mechanical properties are influenced negatively at the same time.

Inorganic nano materials such as nano SiO, nano Al_2O_3, and nano ZnO have been studied to reduce the formaldehyde emission from the wood-based panels [8]. The effect of nanocrystalline cellulose (NCC) on the formaldehyde emission from the plywood was studied, which resulted in a significant reduction in the formaldehyde emission (53.2% of control) from the plywood [9]. However, the formaldehyde emission and TVOCs emitted from wood-based panels produced with the different molar ratio UF resins modified with the microfibrillated cellulose (MFC) was not investigated based on the extensive literature search. The MFC has recently received considerable attention for reinforcements of resins because of high surface area, high stiffness and strength [10]. These properties make the MFC an evident potential for environmental friendly reinforcement for wood resins and composites [10–13]. The MFC consists of a network of interconnected cellulose microfibrils with diameter in range of 10–40 nm and length of several micrometers. The aspect ratio of MFC is normally about 100–150 [10]. Previous studies showed that the incorporation of MFC into the UF resin improved the bond performance of the UF resin [10–12].

The objective of this research was to investigate the formaldehyde emission and TVOC emitted from the laminated veneer lumber (LVL) produced with the different grade UF resins (super E0, E0, and E1 grades) modified with different amounts of the MFC using a thermal extractor. Previous studies reported that thermal extractor method of VOCs and formaldehyde emission analysis was a good alternative to the traditional chamber method for determining the VOCs emission levels from wood-based flooring materials such as laminated wood or plywood [14–16].

2. Materials and methods

2.1. Materials

Three different formaldehyde emission grades of commercial liquid UF resins, such as (E1, E0, and super E0 (SE0)) used in the production of LVLs were supplied from a resin manufacturer (Hansol Home Deco Company Ltd.) in Seoul, Republic of Korea. The specifications of three different types of the UF resins are given in Table 1.

Three commercial rotary veneers (3.6 mm thickness) of debarked pine logs (*pinus densiflora*) were used in the production of LVLs with three-layers. The original size of first class veneers were about 2100 mm × 1700 mm. The veneers having 7% moisture content did not have defects such as knots, checks, splits, and surface irregularities. The veneers were cut into 250 mm × 250 mm square parts and then conditioned in a climate room at 65% relative humidity and 20 °C.

Pine wood flour (40 mesh, *pinus densiflora*) was used in the production of the MFC. The wood flour was suspended in distilled water and then the concentration of suspension was adjusted to 1 wt%. The suspension was then passed through the wet disk mill (*Supermasscolloider* MKCA6-2, Japan). The rotational speed of disks was set to 1800 rpm. The reduced clearance between rotational disks was 150 μm from fiducial zero point. Operation cycles used for MFC-production were 15 passes. The average size of resulting MFC was found to be 79 nm (average of 100 MFC samples). The concentration of the MFC suspension was adjusted to 5 wt%. The MFC suspension and its SEM image are presented in Fig. 1.

2.2. Preparation of UF resins modified with MFC

The super E0, E0, and E1 grade UF resins were modified with different amounts of 5 wt% MFC suspension (Table 2). The UF resins were mixed with different amounts of the MFC suspension using a magnetic stirrer (1000 rpm) for 3 h at room temperature to achieve a proper distribution of MFC in the UF resin. One percent of ammonium chloride as hardener was added to the resin mixture based on solid content of the UF resin. The hardener was added into the resin mixture during the last 5 min of mixing process.

2.3. Production of LVL panels

After preparing pine wood (*pinus densiflora*) veneers with dimensions of 250 mm × 250 mm × 3.6 mm, 3-layer, the LVL panels were produced under laboratory conditions. Each type of modified UF resin was uniformly spread on single bonding surface of veneers using a plastic brush at the rate of 180 g/m^2. After the gluing process, the individual veneers were assembled with the same grain directions for all veneers. The LVL mats were hot pressed under 1.5 N/mm^2 of pressure at a temperature of 150 °C for 13 min in a laboratory hot press. Two LVL panels (10 mm thick) were produced for each type of MFC modified UF resin. The densities of LVL panels ranged from 0.58 to 0.59 g/cm^3. The experimental design is given in Table 2.

2.4. Determination of TVOC and formaldehyde emission of LVLs

A thermal extractor (TE, Gerstel) was used to measure the TVOC and formaldehyde emission from the LVL specimens at different temperatures (Fig. 2). A schematic diagram of the mode of operation is presented in Fig. 2. The TE consists of an adjustable oven (temperature range: room temperature ~ 350 °C) heating a glass tube (length 178 mm, diameter 13.6 mm) with the sample inside. The sample size was limited both by the diameter of the tube and by the heatable length of the oven to a maximum 70 mm. Pure nitrogen was flowed through the glass tube. In normal use, the entire gas flow passes over the adsorbent material. A sample of LVL (10.5 mm × 50.6 mm × 7.4 mm, W × L × T) was placed in a glass extraction tube. The VOCs and formaldehyde were purged under a pure nitrogen gas stream at a constant flow on a Tenax TA tube and 2,4-DNPH cartridge. The thermal extraction process was applied at 25 °C, 35 °C, and 45 °C for 30 min. The test conditions are given in Table 3. The sampling volume was 1 L. Air sampling was performed at a flow rate of 39 ml/min for approximately 30 min. Due to the low volume of the glass tube and high nitrogen gas flow, the air exchange rate was 90 h^{-1}. The list of the test conditions is presented in Table 3.

The tests were carried out at three temperatures, 25 ± 1 °C, 35 ± 1 °C and 45 ± 1 °C, with the humidity uncontrolled but it was

Table 1
Specifications of three different types of the UF resins.

UF resin grade	Non-volatile solid content (wt %)	Jel time (s, 100 °C)	Viscosity (cPs)	pH	Specific gravity	F/U ratio
E1	60.5	124	184	8.11	1.250	1.2
E0	63.3	119	160	7.87	1.255	0.9
SE0	66.3	725	104	7.7	1.252	0.7

F/U: formaldehyde/urea.

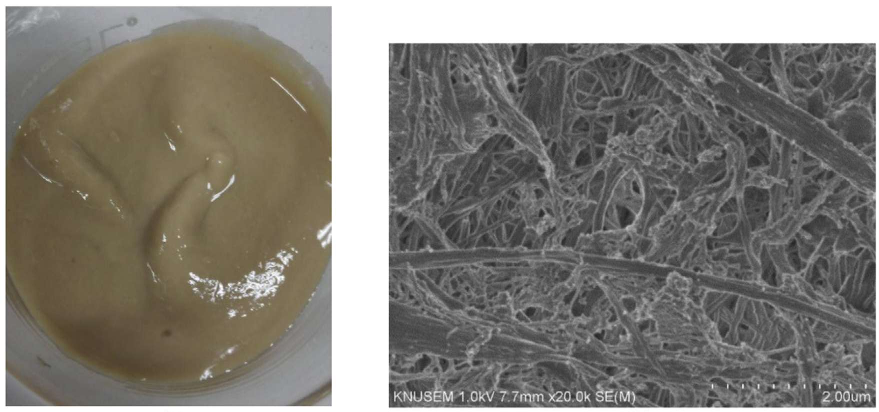

Fig. 1. (a) photograph showing microfibrillated cellulose (5% microfibrillated cellulose (MFC) and 95% water) and (b) SEM image showing the microstructure of the cellulose fibril.

Table 2
Experimental design and properties of UF resins modified with MFC.

LVL code		Quantity* (Applied on a single surface of veneer) (25 cm × 25 cm)			
		Liquid UF resin (g)	MFC suspension (g)	MFC suspension rate (weight %)	Solid hardener (g)
A	SE0 class UF	11.25	0.00	0	1.125
B		10.00	1.25	10	1.0
C		8.75	2.50	20	0.875
D		7.50	3.75	30	0.75
E		6.25	5.00	40	0.625
F	E0 class UF	11.25	0.00	0	1.125
G		10.00	1.25	10	1.0
H		8.75	2.50	20	0.875
I		7.50	3.75	30	0.75
J		6.25	5.00	40	0.625
K	E1 class UF	11.25	0.00	0	1.125
L		10.00	1.25	10	1.0
M		8.75	2.50	20	0.875
N		7.50	3.75	30	0.75
O		6.25	5.00	40	0.625

*The quantity of each type of resin mixture was calculated based on the 180 g/m^2 of resin spreading. The concentration of the MFC suspension was adjusted to 5 wt%.

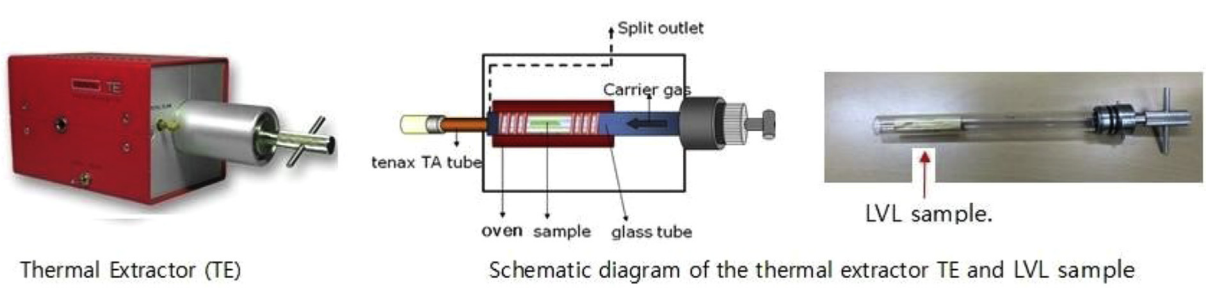

Fig. 2. Thermal extractor and schematic diagram of the thermal extractor (TE) and LVL sample.

about 55 ± 10% relative humidity. The TVOC and formaldehyde emission were analyzed by thermal desorption-gas

Table 3
Test conditions in the thermal extractor test method.

Test condition	Thermal extractor method
Surface area of test specimen (m^2)	0.002076
Chamber volume (L)	0.26
Loading factor (sample area/chamber volume)	79.85
Air change rate (h^{-1})	90
Air supply (mL/min)	39
Sampling	after 30 min
Temperature (°C)	25, 35, 45 ± 1
Humidity (% relative humidity)	Room condition (50 ± 10)
VOCs sampling flow rate (ml/min)	39
VOCs sampling volume (L)	1
Inlet air	High purity N_2
Cleaning process	Cleaning with pure water then placed in an TE for 30 min at 260 °C
VOCs analysis method	TDS-GC/MS
Formaldehyde analysis method	HPLC

chromatograph/mass spectrometer (TD-GC/MSD) and high performance liquid chromatography (HPLC), respectively. The formaldehyde emission and TVOC from the LVLs were measured as μg/m^3 (micrograms per cubic meter) (Concentration (ug/m^3) = 0.0409 × concentration (ppb: per billion) × molecular weight).

3. Results and discussion

3.1. Formaldehyde emission emitted from LVLs

The formaldehyde emission values of the LVLs depending on the F/U ratio of UF resin and MFC content at 25 °C, 35 °C, and 45 °C using the thermal extractor are given in Table 4. The formaldehyde emission from the LVLs produced with the SE0 grade UF resin considerably decreased with increasing MFC content at 25 °C while this was not observed for the E0 and E1 grade resins. The results revealed that the MFC did not work for the E0 and E1 resins at 35 and 45 °C. As expected, the highest formaldehyde emission rate was found in the LVL specimens produced with E1 grade resin, followed by E0 and SE0 grade resins, respectively. The formaldehyde emission rates from the LVLs significantly increased with increasing environment temperature. At the environmental temperatures of 35 °C and 45 °C, the rates of formaldehyde emission was more than two and five times higher than that at 25 °C, respectively (Table 4). Similar results were observed in previous studies [17–19]. Resin free formaldehyde is frequently reported to be cause a positive correlation generally exists between it and formaldehyde emission from boards. At low F/U ratio resin, the free formaldehyde content and board emission rate are lower [20].

The incorporation of the MFC into the SE0 up to 30 wt% decreased the formaldehyde emission from the UF resin. As the amount of MFC increased to 30 wt%, the formaldehyde emission from LVL (code: D) produced with the SE0 grade UF resin decreased from 17.3 to 3.4 μg/m^3 at 25 °C. However, the formaldehyde emission from the LVLs produced with the SE0 modified with the MFC increased from 13.9 to 34.9 μg/m^3 at the 35 °C. At the 45 °C, as the amount of the MFC in the SE0 resin increased to 20 wt%, the formaldehyde emission decreased very slightly (73.8–71.2 μg/m^3), then it increased up to 89.9 μg/m^3 as the MFC content was raised to 40 wt% in the SE0 resin. A similar result was found in a previous study [21]. Liu and Zhu [21] reported that compared with control panels, formaldehyde emissions from wood-based panels coated with 0.5% nanoparticles reinforced melamine-impregnated papers decreased slightly. They reported that nanoparticles could absorb free formaldehyde from resin and the barrier properties of the nanoparticles reduced the formaldehyde emissions of panels because of their shielding effect. However, in our study, this effect was only observed for the SE0 grade resin in our study at 25 °C. The increases in the loading level of the MFC may have caused aggregation in the MFC filled UF resins, negatively affecting formaldehyde emission values and the decline of formaldehyde is nonlinear [21].

As for the E0 grade resin, the formaldehyde emission from the LVL (code: E0-C) tested at 25 °C decreased from 15.2 to 13.5 μg/m^3 when 20 wt% MFC was incorporated into the resin. Further

Table 4
Formaldehyde emission values of LVLs at three different temperatures.

LVL code	25 °C	35 °C	45 °C
	Formaldehyde (μg/m^3)	Formaldehyde (μg/m^3)	Formaldehyde (μg/m^3)
SE0-A	17.3	13.9	73.8
SE0-B	19.3	14.9	72.5
SE0-C	9.7	24.2	71.2
SE0-D	3.4	22.1	75.5
SE0-E	5.7	34.9	89.9
E0-A	15.2	44.3	82.6
E0-B	17.2	45.1	88.3
E0-C	13.6	54.2	87.9
E0-D	25.9	43.7	118.8
E0-E	21.9	52.7	146.6
E1-A	26.5	102.1	167.3
E1-B	29.9	107.1	195.3
E1-C	52.4	115.1	179.5
E1-D	46.5	122.0	178.4
E1-E	71.8	113.2	221.8

increment in the MFC content did not decreased the emission (21.9 μg/m^3) from the LVL. At the environmental temperature of 35 °C, the emission from the LVLs produced with the E0 resin increased 44.3–52.7 μg/m^3 as the 40 wt% MFC was incorporated into the resin. A similar trend to the results obtained at 35 °C was found in the LVL tested at 45 °C.

The formaldehyde emission from the LVL produced with E0 resin increased from 82.6 to 146.6 μg/m^3 at 45 °C as the 40 wt% MFC was incorporated into the resin. The incorporation of the MFC into the E1 grade resin did not decrease the formaldehyde emission. The emission from the LVL tested at 25 °C increased from 26.5 to 71.8 μg/m^3 as the amount of the MFC increased from 0 to 40 wt% in the E1 grade resin. Similar results were found in the specimens tested at 35 °C and 45 °C. According to the World Health Organization (WHO) [22], the formaldehyde concentration level from building materials is measured post construction (but pre-occupancy) and should be less than or equal to 100 μg/m^3 [15]. In addition, the E1 emission measured for formaldehyde must be less than 124 μg/m^3 (as required by EN 636:2003). The formaldehyde emission values of the LVLs produced with the modified E1 grade resins at 25 °C and 35 °C met the maximum value (124 μg/m^3) of EN 636 standard [23]. In addition, the LVL specimens modified with the MFC and control specimens at 25 °C complied with the maximum value (100 μg/m^3) specified by the WHO.

The formaldehyde emission rates from the LVLs produced with SE0, E0 and, E1 grade resins modified with the MFC at 25 °C complied with the standard of WHO. At 35 °C, the formaldehyde emission values from the LVLs produced with the SE0 and E0 grade resins, except for the E1 grade resin, were lower than the maximum standard value of WHO. However, at 45 °C, the LVLs produced with all the modified SE0 and E0 grade resins, except for the E0-D and E0-E grades, met the maximum value specified by the WHO.

Mechanical properties of the LVLs used in this research which was a part of the collaboration between research groups of Istanbul University, Kangwon National University (KNU), and Seoul National University were investigated at the laboratory of department of Forest Biomaterials Engineering of KNU [24]. They reported that limited amounts of the MFC suspension (5 wt%) in the UF resin formulation significantly improved the mechanical performance of UF bonds. According to their results, the tensile shear strength of LVLs improved with increasing content of the MFC suspension up to 30 wt% in the E0 class UF adhesive while this was found to be 20 wt % MFC suspension in the E1 class UF adhesive. Further increment in the content of the MFC suspension decreased the bond performance and ductility of the resin. Although the amount of the UF resin decreased with increasing MFC content in the production of LVLs, the improvement in the tensile strength of the LVLs bonded with the UF resin modified with a certain level of the MFC was explained by the chemical reaction between the methylol groups of the UF adhesive and the free hydroxyl groups of the cellulose in the bond line, and the improved ductility of the resin [24].

3.2. TVOC emitted from LVLs

The TVOC emission from the LVLs produced with SE0, E0, and E1 grade UF resins modified with MFC at 25 °C, 35 °C, and 45 °C are given in Table 5. The VOCs emitted from the LVLs considerably decreased with increasing MFC content in the SE0 and E0 grade resins at 25 °C and 35 °C. However, at the 45 °C, the incorporation of the MFC into the all grades of UF resins did not decrease the VOCs. The highest reduction in the VOCs was observed for the LVLs bonded the with E1 grade UF resin at the environmental temperatures of 25 °C and 35 °C, followed by the SE0 and E0 grade resins. For example as the amount of the MFC increased by 40 wt% in the E1 grade UF resin, the TVOC decreased from 94.9 to 43.4 μg/m^3 at 25 °C. This was found to be 89.5 to 68.3 μg/m^3 for the E0 grade resin and 122.7 to 88.5 μg/m^3 for the SE0 resin at 25 °C. A similar trend was observed for the LVLs tested at 35 °C (Table 5). As for the environmental temperature of 45 °C, the incorporation of the 10 wt % MFC into the super E0, E0, and E1 grade resins decreased the VOCs, but further increment in the MFC content did not decreased the VOCs (Table 5).

Table 5
Total volatile organic compounds (TVOC) emitted from LVLs at three different temperatures.

LVL code	25 °C	35 °C	45 °C
	TVOCs (μg/m^3)	TVOCs (μg/m^3)	TVOCs (μg/m^3)
SE0-A	122.7	301.9	2321.4
SE0-B	36.1	274.1	2037.1
SE0-C	55.2	157.7	2484.2
SE0-D	51.6	188.9	2130.9
SE0-E	88.5	200.4	2322.8
E0-A	89.5	341.0	1750.4
E0-B	65.0	282.1	1633.9
E0-C	79.2	221.9	1765.6
E0-D	88.7	244.9	1852.1
E0-E	68.3	234.9	2048.9
E1-A	94.9	478.1	1055.8
E1-B	96.5	430.1	933.4
E1-C	84.2	388.2	1473.9
E1-D	60.5	154.1	1897.5
E1-E	43.4	126.9	1111.3

In a previous study it was reported that the amount of TVOC emitted from a commercial plywood was 145.1 μg/m^3 [25].

The VOCs values of all the LVLs at three environmental temperatures were significantly below the minimum value (300 μg/m^3) of BREEAM (Building Research Establishment's Environmental Assessment Method, United Kingdom) is the world's first sustainability rating scheme for the built environment [26]. In addition, the European Community has prepared a target guideline value for TVOC of 300 μg/m^3. TVOC emitted from the LVLs produced with SE0, E0, and E1 grade UF resins modified with the MFC were lower than the standard value of BREEEAM and EU. At 35 °C, the VOCs from the LVLs produced with three different grade resins (SE0, E0, and E1) modified with the MFC, except for the control LVLs, and E1-B and E1-C grade resins, complied with the standard. At the environmental temperature of 45 °C, although the TVOC of the LVLs containing the MFC did not comply with limit values of the BREEAM and EU, they met the maximum value of allowable VOC content level (500 μg/m^3) under LEED standard for Indoor Environmental Quality developed by the U.S. Green Building Council [5].

The TVOC emission from the LVLs was significantly affected by the test temperature. The increment in the temperature resulted in a higher TVOC emission rate from the LVLs. In particular, as the temperature increased from 35 to 45 °C, the amount of TVOC significantly increased. TVOC values increased more than three times as the test temperature increased from 25 to 35 °C. In particular, as the test temperature was increased from 35 to 45 °C, the TVOC emitted from the LVLs increased about 6–10 times. The results showed that the VOCs from the LVLs had more active mobility at higher temperatures, which was easier to emit from the composites. Similar results were observed in previous studies [1,3,15]. For example, Kim [3] reported that the VOCs emitted from wood-based panels such as engineered flooring and laminate flooring, increases in temperature cause a remarkable increase in the VOCs emission. To know the temperature on the emission parameters such as the partition coefficient and the diffusion coefficient is an effective approach to know the temperature on the emission characteristics of building materials [17]. Zhang et al. [17] reported that the partition coefficients decreased while the diffusion coefficients increased with increasing temperature. For example, when the temperature approached 50 °C, the partition coefficients became less than 25% of those at 18 °C while the diffusion coefficient for each material at 50 °C was more than 5 times than that at 18 °C [17].

The VOCs contained a lot of unidentified compounds. Thus, in our study the VOCs were divided into 5VOC (benzene, toluene, ethyl benzene, xylene, stylene). The highest VOC emitted from the LVLs was found to be the toluene, followed by the xylene, the benzene, and the ethyl-benzene, respectively. The stylene was not determined in the LVL specimens with and without MFC. The benzene was only found in some LVL groups produced with SE0 and E1 grade resins while the ethyl benzene was only found in LVL groups produced with the E0 and E1 grade resins. However, benzene and toluene were found in all the LVLs produced with the SE0, E0, and E1 grade resins.

The variety and amount of VOC emissions are strongly dependent on the veneer species. Miyamoto et al. [27] reported that the predominant VOCs were derived from the veneer. The reduction in the VOCs emission of LVLs could be due to the fact that the MFC functioned as a scavenger for the VOCs emitted from the veneers (*pinus densiflora*). The results showed that the MFC was found to be the most effective for the reduction of VOCs from LVLs produced with the E1 grade resin among the UF resin grades. This is important for the wood-based panel industry because the panel manufacturers focus on environmentally friendly solution for the reduction of VOCs from furniture made from particleboard and medium density fiberboard.

4. Conclusions

The results of the study showed that the formaldehyde emission from the LVLs produced with the SE0 grade UF resin considerably decreased with increasing MFC content at 25 °C while this was not observed for the E0 and E1 grade resins. The results revealed that the MFC did not work for decreasing of formaldehyde emission of the LVLs produced with the E0 and E1 resins at 35 °C and 45 °C. However, the VOCs emitted from the LVLs considerably decreased with the incorporation of the MFC at environmental temperatures of 25 °C and 35 °C, except for 45 °C. This showed that the MFC was not effective to reduce the VOCs from the LVLs at higher temperatures. The xylene was the highest detected compound in all samples, followed by ethylbenzene and toluene. Styrene, however, was not detected at all in any of the LVLs. The use of MFC in the UF resin can be environmentally friendly solution for reducing the VOCs from the wood-based panel used for indoor furniture.

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