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論　文（Original article）

Emission of volatile organic compounds during drying of veneer: Hinoki (*Chamaecyparis obtusa* Endl.)

Atsuko ISHIKAWA [1)*], Tatsuro OHIRA [2)], Kohta MIYAMOTO [3)], and Akio INOUE [3)]

Abstract

Estimation of volatile organic compounds (VOCs) emitted from the wood-based material industry has become an urgent matter following the 2004 amendment of the Air Pollution Control Law. Meanwhile, complaints about the odors from wood products facilities have been made by neighboring residents. At the same time, aldehydes emission from wood products has raised concerns over its effect on human health. We have reported VOCs emissions from wood drying process of sugi, larch and meranti. In this study, emissions of VOCs and aldehydes during hinoki (*Chamaecyparis obtusa* Endl.) veneer drying were estimated. Hinoki veneer was dried at 180ºC, and the discharged air was analyzed by high-performance liquid chromatography (HPLC) and gas chromatography / mass spectroscopy (GC/MS). The emissions of VOCs and aldehydes increased with increasing drying time. The discharge amount of hinoki was higher than that of sugi, larch, and meranti. The dominant compounds in the discharged air were terpenes. Most of the discharged formaldehyde was removed by means of a drain trap up to 25 minutes. Moreover, discharged amounts of VOCs and aldehydes from the industrial drying process were estimated using the experimental data obtained in this study. These results can be of help to understand emissions and to establish effective emission control of VOCs and aldehydes from hinoki drying process.

Key words : Hinoki (*Chamaecyparis obtusa* Endl.), Veneer, Drying, Volatile organic compounds (VOCs), Aldehydes

1. Introduction

Estimation of the amount of volatile organic compounds (VOCs) emitted from the wood-based material industry has become an urgent matter following the 2004 amendment of the Air Pollution Control Law to control the emission of VOCs from factories (Ministry of the Environment, 2004). Wood includes VOCs, such as terpenes. Meanwhile, complaints about the odors from wood products facilities have been made by neighboring residents. At the same time, aldehydes released from wood and wood products have raised concerns over its effects on human health, and the Ministry of Health, Labour and Welfare proposed a set of guidelines for formaldehyde and acetaldehyde (Ministry of Health, Labour and Welfare, 2002; Tohmura et al., 2005). Under these circumstances, it is important to clarify the components, particularly VOCs and aldehydes, of discharged air from wood products facilities in Japan.

Among wood processing, drying process is considered to emit VOCs, such as terpenes. In Japan, only a few researches have been done about the VOCs released during drying. A number of foreign research projects have been performed to measure the amount of VOCs released during the wood drying process; however, each emitted compound is not always analyzed (Ishikawa, 2009; Milota, 2000). Even in the few cases where each compound is measured, the measurement systems differ due to the variation in equipment and conditions of the drying processes (Bengtsson & Sanati, 2004; Cronn et al., 1983; Fritz et al., 2004; Granström, 2003; McDonald et al., 2002; Otwell et al., 2000).

We have investigated VOCs emissions from wood drying process of sugi, larch and meranti (Ishikawa et al., 2009; Ishiakwa, 2009). In this study, emissions of VOCs and aldehydes during hinoki (*Chamaecyparis obtusa* Endl.) veneer drying were estimated. Hinoki is now being used for plywood. Hinoki green veneer was dried at 180ºC, and the discharged air was analyzed by high-performance liquid chromatography (HPLC) and gas chromatography / mass spectroscopy (GC/MS). Using this data, the amount of VOCs and aldehydes discharged during drying of 1 m^3 of veneer was calculated in order to estimate emissions from industrial wood-drying facilities. These results can be

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1) Department of Wood Processing, Forestry and Forest Products Research Institute (FFPRI)
2) Department of Biomass Chemistry, Forestry and Forest Products Research Institute (FFPRI)
3) Department of Wood-Based Materials, Forestry and Forest Products Research Institute (FFPRI)
* Department of Wood Processing, Forestry and Forest Products Research Institute (FFPRI), 1 Matsunosato, Tsukuba, Ibaraki 305-8687, Japan; e-mail: aishi@ffpri.affrc.go.jp

of help to understand emissions and to establish effective emission control of VOCs and aldehydes from hinoki drying process.

2. Materials and methods

Hinoki (*Chamaecyparis obtusa* Endl.) green veneer measuring 220 × 220 × 3 (thick) mm was used.

2.1 Measurement of VOCs and aldehydes

Discharged air during drying was measured as described in the former report (Ishikawa et al., 2009). Two pieces of the specimens were heated in an oven (inner size: 30×30×34 cm, DKN302, Yamato Scientific Co. Ltd., Tokyo) at 180ºC until the moisture content was reduced to below 10%. The drying temperature and final moisture content were determined based on the conditions of industrial veneer drying. Experimental conditions and moisture content of the specimens before and after drying are listed in Table 1. During the drying process, the discharged air was passed through a heat-resistant tube and a drain trap, and was collected in a sampling bag (10 L, GL Science Co. Ltd., Tokyo) fixed in a sampling case equipped with Pump A (MP-Sigma 30, Shibata Scientific Technology, Tokyo). Hereafter, the trapped water and the gas collected in a sampling bag are described as “discharged moisture” and “discharged gas”, respectively. The discharged moisture and gas were sampled from the beginning to the end of the drying. After drying, the discharged gas was sampled to a 2, 4-dinitrophenylhydrazine (DNPH) cartridge (LpDNPH S10L, Supelco Inc., Tokyo) and a Tenax-TA tube (25090-U, Supelco Inc., Tokyo) by Pump B, and the discharged moisture was DNPH derivatized. The collected compounds were analyzed by GC/MS (GC type 6890, MSD 5973, Hewlett-Packard Japan, Ltd., Tokyo) and HPLC (LC-10 ADvp, Shimadzu Co. Ltd., Kyoto). Details of the analysis are described in the former report (Ishikawa et al., 2009). Three tests were performed for one condition.

2.2 Discharge amount during drying of 1 m^3 of wood

In order to estimate emissions from industrial wood-drying facilities, the amount of VOCs and aldehydes discharged during drying of 1 m^3 of veneer was calculated based on the method described in the former report (Ishikawa et al., 2009). The discharged amount from 1 m^3 of wood was calculated using the following equation:

Discharged amount from 1 m^3 of wood (g) = Air concentration (g/m^3) × Air exchange rate (m^3/h) × Drying time (h) / Total volume of specimens (m^3) …… (1)

where air concentration is the value obtained for VOCs and aldehydes in the experiments described in 2.1, and total volume of specimens is the sum of specimen volumes placed in the oven. Formaldehyde was detected both in discharged gas and discharged moisture; therefore, the discharged amount was calculated using Eq. (2):

Discharged amount of formaldehyde from 1 m^3 of wood (g) = {Air concentration (g/L) + Concentration in discharged moisture collected during sampling of 1 L of discharged gas (g/L)} × 1000 × Air exchange rate (m^3/h) × Drying time (h) / Total volume of specimens (m^3) …… (2)

2.3 Ether extraction

The amount of ether extractives included in specimens before and after drying was measured in order to estimate the amount of compounds included in the specimens before and after drying. First, half the hinoki specimens were dried at 180ºC for 39 min as well as under the conditions of the experiment described in 2.1. Undried and dried samples were cut into pieces, and ground in a mill to pass through a 2.0-mm screen. Ground samples in 5-g units were Soxhlet-extracted with diethyl ether at 40ºC for 5 h. The solvents were evaporated in vacuo at 40ºC. Other 1-g ground samples were oven-dried at 105ºC until their weight stabilized, after which the moisture content was calculated. From these results, the rate of ether extractives relative to the oven-dried weight of the wood was calculated. Compounds in the extractives were then analyzed using GC/MS. The analysis conditions were the same as described

Table 1. Experimental conditions and moisture content of specimens before and after drying.

Drying temperature (ºC)	Drying time (min)[1)]	Flow rate (pump A) (L/min)	Collected air volume (L)[2)]	Flow rate (pump B) (L/min)	Sampled air volume (L)		Moisture content (%)[3)]	
					DNPH	TENAX	Before drying	After drying
180	11 (0-11)	0.45	4.95	0.10	2.50	2.00	104	0
	11 (14-25)		4.95					
	11 (28-39)		4.95					

[1)] Discharged air was collected three times during drying: stage 1 (0-11 min), stage 2 (14-25 min), stage 3 (28-39 min).

[2)] Collected air volume = Drying time × Flow rate (pump A)

[3)] n=3

in the former report (Ishikawa et al., 2009).

3. Results and discussion

Fig. 1 shows the moisture content of wood and the discharged amount of formaldehyde, acetaldehyde and VOCs during drying. All of the discharged amounts increased with increasing drying time. It has been reported that the discharged amount of VOCs and formaldehyde increases with decreasing moisture content (Granström, 2003; Otwell et al., 2000). These results indicate that the discharged amount of VOCs and aldehydes could be decreased by preventing extra drying time.

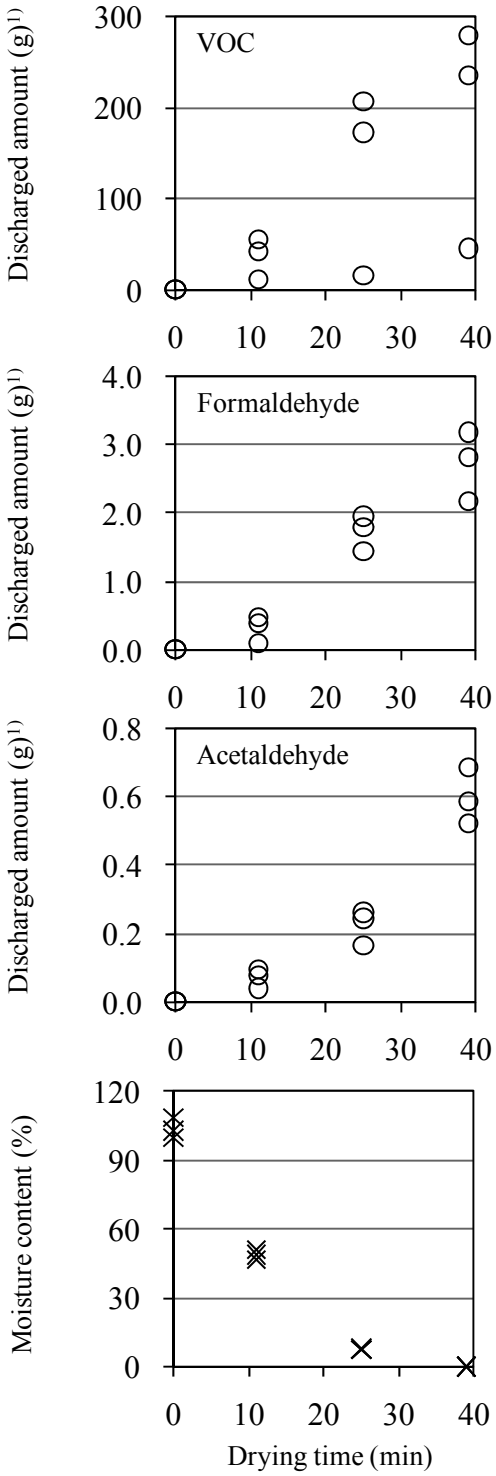

Fig. 1. Relationships between drying time, moisture content, and emissions from 1m^3 of veneer.

1) Cumulative amount.

Formaldehyde was detected both in the discharged moisture and the discharged gas. The mass of formaldehyde collected in discharged moisture and discharged gas during sampling of 1 L of discharged gas are shown in Fig. 2. The data shows that most of the discharged formaldehyde was collected in the drain trap up to 25 min, but was not collected after 28 min. These results might indicates that formaldehyde can be trapped in the water until saturation, but after that passes through the trap and be discharged in the air. Taking the former report (Ishikawa et al., 2009) into consideration, it could be concluded that formaldehyde can be efficiently removed at wood processing plants by condensing and collecting discharged moisture with sufficient amount of water or using a water scrubber. Organic acids, such as acetic acid, might be included in the drain as the result of thermal decomposition of wood. However, these compounds were not analyzed in this study because the main purpose was to estimate the amount of aldehydes.

Tabel 2 shows the major compounds emitted during drying. All the compounds listed in Table 2 are terpenes, therefore terpenes are the main compounds released during hinoki drying. Generally, compounds, such as furfural, are included in wood extractives as a result of thermal decomposition. However these compounds were

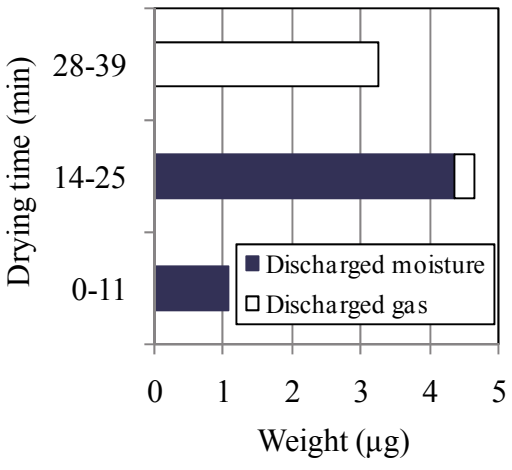

Fig. 2. Comparison of formaldehyde amount in discharged moisture and discharged gas during sampling 1L of discharged gas.

Table 2. Major compounds released during drying.

Compounds	Air concentration (μg/m^3)		
	0-11 min	14-25 min	28-39 min
α-Pinene	45008	45484	30476
Limonene	15507	25834	13455
γ-Cadinene	4331	52809	10950
α-Muurolene	3818	25823	7451
γ-Muurolene	2411	18108	4657
Neoalloocimene	2604	5026	3568
α-Fenchene	7890	4855	2871
β-Selinene	879	8395	1866
α-Copaene	750	3256	990
β-Elemene	667	6827	726
Dauca-5,8-diene	296	1202	230
α-Cadinol		16633	16282
Ferruginol		1738	4292
Hinokione		449	2479
Muurola-3,5-diene<cis->		1070	157
Cadina-1(6),4-diene<cis->			16587

Table 4. Results of GC/MS analysis of ether extractives.

(%)

Compounds	Heartwood		Sapwood	
	Undried	Dried	Undried	Dried
γ-Cadinene	45.0	43.5	49.1	31.0
Dauca-5,8-diene	11.6	7.8	9.1	6.5
α-Copaene	9.6	8.8	10.4	5.7
Muurola-3,5-diene<cis->	7.4	9.2	9.3	15.3
γ-Muurolene	4.3	6.2	5.1	10.2
α-Muurolene	3.5	7.5	6.5	10.3
Cadina-1(6),4-diene<cis->	0.6	1.0	0.4	0.4
Hinokione	0.7	0.6	-	-
Ferruginol	0.6	0.7	-	-
β-Elemene	0.2	0.5	-	-
Total	82.9	84.8	89.9	79.3

-: undetected

not significantly released from wood during the drying process. The differences in wood extraction and wood drying, for example sample size and treating conditions, might have an influence on the result. Tables 3 and 4 list the results of ether extraction. Both compounds included in the discharged gas (Table 2) and extractives (Table 4) are reported to be included in untreated hinoki wood (Forestry and Forest Products Research Institute 2004; Kinjo and Yaga, 1986; Kinjo et al., 1988; Ohashi et al., 1987; Ohtani et al., 1997; Terauchi et al., 1993). Tepenes such as α-cadinol, are reported to have termiticidal and antifungal activity (Kinjo and Yaga, 1986; Kinjo et al., 1988). Some compounds detected from the drying process were found in ether extractives, while the others were not. The differences in the kinds of compounds might be due to their susceptibility to ether extraction and drying at 180ºC. Only a trace of α-pinene, which is well known to exist in hinoki wood, could be found in the extractives. The reason is unclear but α-pinene existed in the hinoki sample might have been removed during sample preparation. The percentage of Muurola-3,5-diene<cis>, γ-Muurolene, and α-Muurolene in extractives of dried sample was higher than that of undried wood. This might be due to the differences in volatility of compounds included in the wood sample, chemical conversion of the other compounds and so forth. Further study is needed to clarify the reason.

The discharged amounts from 1 m^3 of wood are listed in Table 5 with the results of the other species (Ishikawa et al., 2009). The discharge amount of VOC from hinoki was higher than that from sugi, larch, and meranti. One reason for the high discharged amount for hinoki is considered to be the longer drying time.

Assuming that the difference between the amount of ether extractives of undried and dried wood is the

Table 3. Ether extractives of undried and dried hinoki wood.

Specimen	Ether extractives (%)[1]						Oven-dried weight of 1m^3 wood (g)[3]	Ether extractives (g/m^3 wood)[4]
	Undried	Average	Dried[2]	Average	Undried - Dried	Average		Undried - Dried
Heartwood	0.97	0.89	0.42	0.35	0.55	0.54	370,000	1,998
	0.98		0.21		0.76			
	0.73		0.43		0.31			
Sapwood	0.32	0.43	0.21	0.28	0.11	0.15	340,000	513
	0.65		0.42		0.23			
	0.33		0.21		0.11			

[1] Relative to oven-dried weight

[2] Drying conditions: 180ºC, 39 min.

[3] Calculated from the oven-dry specific gravity of hinoki (Fushitani et al., 1985).

[4] Ether extractives (g/m^3 wood) = Ether extractives (undried - dried) (%) / 100 × Oven-dried weight of 1 m^3 wood (g)

Table 5. Emissions of VOC and aldehydes during drying.

Specimen	Temperature (°C)	Time (min)	Emissions from 1m^3 green wood (g)			Ovendried weight of 1m^3 wood (g)	Discharged VOC (%)[3)]
			VOC	Acetaldehyde	Formaldehyde		
Hinoki	180	39	186.7[1)]	0.6	2.7	355000[2)]	0.0526
Sugi[4)]	180	27	34.8	0.5	2.9	325000	0.0107
Larch[4)]	180	12	6.1	0.1	0.4	500000	0.0012
Meranti[4)]	180	15	1.4	0.1	0.5	410000	0.0003

[1)] Average of data at 39 minutes in Fig. 1.

[2)] Calculated from the oven-dry specific gravity of hinoki (Fushitani et al., 1985).

[3)] Discharged VOC (%) = Emissions from 1m^3 green wood (g) / Ovendried weight of 1m^3 wood (g) × 100

[4)] Ishikawa et al., 2009

maximum amount of VOCs discharged during drying, the maximum amount of VOCs discharged during drying 1 m^3 of wood can be calculated to be 513−1998 g as shown in the rightmost column in Table 3. During actual industrial drying, only a part of the maximum amount would be emitted. As shown in Table 5, the amount of discharged VOCs during drying of 1 m^3 of hinoki was estimated to be 186.7 g. The amount of discharged VOCs during drying of other species (pine, fir, hemlock, etc.) at 49−160ºC has been reported to be 20−4000 g/m^3 of wood (Ingram et al., 2000; McDonald et al., 2002; McDonald et al., 2004; Milota, 2003; Milota, 2006; Milota & Mosher, 2006; Shmulsky, 2000a; Shmulsky, 2000b; Shmulsky, 2000c). Our result is in these published values. The discharged amount of formaldehyde and acetaldehyde during drying of radiata pine at 100ºC dry-bulb temperature (DBT) with 70ºC wet-bulb temperature (WBT) has been reported to be 1.1 and 8.7 g/m^3 of wood, respectively (McDonald et al., 2002). Our results were almost the same level (Table 5).

Moreover, our results were compared to the discharged amount during industrial drying (Kawarada et al., 2008) (Table 6). The discharged amount of VOCs from hinoki was the same level as that from the industrial drying, and was higher than that from sugi. The reasons for the differences are considered to be the individual varieties of hinoki and sugi, irregularity of air concentration and contamination in the industrial drying duct, and the difference in species (a species of Southeast Asia was dried together with sugi during the industrial drying) and drying conditions.

4. Conclusions

Hinoki veneer was dried under conditions generally used in industrial drying processes (180°C), and discharged VOCs and aldehydes were evaluated. The following results were obtained:

1. VOCs, formaldehyde, and acetaldehyde were emitted during drying. The amounts increased with increasing drying time.
2. The discharge amount of hinoki was higher than that of sugi, larch, and meranti.
3. Main VOCs emitted during drying were terpenes.
4. Most of the discharged formaldehyde up to 25 minutes was removed by means of a drain trap.
5. Discharged amount from industrial drying was estimated using the results obtained in this study.

These results indicate that emissions of VOCs and aldehydes released during hinoki drying can be decreased by controlling drying time. The discharge amount and released compounds of hinoki were different from those of other species. Most of the relased compouns were terpenes which are reported to have several beneficial functions. Therefore, it is desirable to investigate their

Table 6. Comparison of the data in this study with an industrial drying process (Kawarada et al., 2008).

	Our results (Hinoki, 180ºC)	Our results (Sugi, 180ºC)[1)]	Kawarada et al. (Sugi and other species, 180°C)
Ventilation rate (m^3/h)	0.48	0.48	16,100
Dried veneer volume (m^3/h)	4.5 x 10^{-4} [2)]	7.9 x 10^{-4}	5.4
VOC released from 1m^3 veneer (g)	187	35	187

[1)] Ishikawa et al., 2009

[2)] Calculated from the dried veneer volume, 2.90×10^{-4}m^3, during 39 minutes in our experiment.

efficient utilization considering the released compounds of each species. Formaldehyde, on the other hand, is a carcinogen and should be removed. It is also indicated that formaldehyde can be efficiently removed from discharged air by water scrubbing.

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ヒノキの単板乾燥工程で排出される揮発性有機化合物

石川敦子[1)*]・大平辰朗[2)]・宮本康太[3)]・井上明生[3)]

要 旨

2004 年に大気汚染防止法が改正され、木材関連工場からの揮発性有機化合物（VOC）の排出実態を把握することが急務となっている。他方、木材関連工場からの臭気に関する苦情も出ている。また、木材および木質材料から放散するアルデヒド類が健康に及ぼす影響についても関心が持たれている。著者らはこれまでに、スギ、ラーチ、メランチの乾燥工程における VOC の排出挙動について報告してきた。本報告では、ヒノキ (*Chamaecyparis obtusa* Endl.) 単板の乾燥工程における VOC とアルデヒド類の放散について検討した。ヒノキ単板を 180ºC で乾燥し、排気を高速液体クロマトグラフィー (HPLC) とガスクロマトグラフ質量分析（GC/MS）で分析した。VOC とアルデヒド類の排出量は、乾燥時間の経過とともに増加した。ヒノキからの排出量は、スギ、ラーチ、メランチからよりも多かった。排気に含まれる主な化合物はテルペン類であった。乾燥時間 25 分までは、排出されたホルムアルデヒドのほとんどがドレントラップで回収された。さらに、実験値をもとに工場での排出量を推定することができた。これらの結果は、ヒノキの乾燥工程において排出される VOC とアルデヒド類の実態把握および低減化に役立つものである。

キーワード：ヒノキ (*Chamaecyparis obtusa* Endl.)、単板、乾燥、揮発性有機化合物（VOC）、アルデヒド類

1) 森林総合研究所加工技術研究領域
2) 森林総合研究所バイオマス化学領域
3) 森林総合研究所複合材料研究領域
＊森林総合研究所加工技術研究領域　〒 305-8687　茨城県つくば市松の里 1　e-mail: aishi@ffpri.affrc.go.jp