Evaluation of the relation between sick-building syndrome and aldehydes emitted from lime plaster containing vegetable oil

(植物油を添加した漆喰から放散するアルデヒド類と シックハウス症候群との関係性の評価)

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Contents

Abstract

Lime plaster applied to walls and ceilings often contains vegetable oil for improving water resistance, and is considered to be harmless owing to its natural origin. The purpose of this study was to assess aldehyde emissions from plaster containing vegetable oil in order to predict the concentrations of aldehydes emitted into the indoor air. A passive emission chamber method was used to observe the emissions of aldehydes from vegetable oil when mixed with sodium hydroxide solution. The findings show that the alkalinity of calcium hydroxide in the plaster would accelerate the formation of aldehydes significantly. Furthermore, aldehyde emissions from the plaster to which vegetable oil was added were tested using a dynamic emission chamber (the small chamber method). A plaster containing soybean oil strongly emitted hexaldehyde throughout the test period. Plasters containing linseed oil or perilla oil strongly emitted propionaldehyde and acetaldehyde. The calculated indoor air concentrations of the aldehydes emitted from the lime plasters containing vegetable oil exceeded the air quality guidelines of the Japanese Ministry of Health, Labour, and Welfare, the Japanese Ministry of the Environment, and the German Federal Environmental Protection Agency. Moreover, these aldehydes are irritants and have unpleasant odours. Therefore, adding vegetable oil to plaster should be avoided to prevent sick-building syndrome.

1. Introduction

In recent years, sick-building syndrome caused by the increasing air tightness of modern houses and chemicals emitted from building materials has become an issue (Mori and Todaka, 2011; Saijo et al., 2004). The symptoms of sick-building syndrome include eye irritation, throat pain, rhinorrhea, and headaches (Mori and Todaka, 2011; Saijo et al., 2004). Thus, to protect building occupants from such health impairments, building materials of natural origin are currently more favoured than the materials that may emit chemicals.

Lime plaster (hereafter, plaster) is considered to be a natural material. The principal component of plaster is slaked lime (calcium hydroxide), which is from limestone, a natural mineral found in abundance globally. Hence, plaster has been used as a building material worldwide since ancient times (Yang et al., 2010). The advantages of plaster include humidity tolerance and heat insulation, while low water resistance is one of its disadvantages (Green Home Building). Thus, vegetable oils are often added to plaster to improve its water resistance. Various vegetable oils, such as rapeseed oil and linseed oil, can be used (Free Patents Online). However, aldehydes are typically generated by autoxidation of vegetable oils (Lazzari and Chiantore, 1999; Toda et al., 1996; Juita et al., 2012; Lee et al., 2010; Knudsen et al., 2007; Kumarathasan et al., 1992). The majority of the detected reaction products, such as propionaldehyde originate from the oxidation of linolenic compounds, which dominate in the composition of linseed oil (Juita et al., 2012). Moreover, aldehydes are emitted from paint and floor finish products that contain natural products such as vegetable oils. Such products have been found to cause persistent odours and hence affect the perceived indoor air quality more than similar synthetic products (Knudsen et al., 2007; Kim et al., 2011). Therefore, autoxidation of vegetable oils contained in plaster may produce aldehydes, which can cause unpleasant odours and even sick-building syndrome.

Elevated levels of indoor aldehydes have been associated with the occurrence of sickbuilding syndrome (Takigawa et al., 2012). High indoor air concentrations of acetaldehyde can induce asthma (Hulin et al., 2010). Furthermore, the odours of aldehydes, such as those emitted from building materials after office renovation (peng et al., 2009) or from insulating materials in residences (Van der Wal et al., 1989), can be unpleasant.

In the present study, aldehydes generated from vegetable oils and from vegetable oil-containing plaster were analysed. Then, the aldehyde concentrations in a model room with vegetable oil-containing plaster were estimated.

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2. Materials and methods

2.1. Vegetable oils

Food-grade vegetable oils were used in this study to make plasters. They included rapeseed oil (Nisshin Oillio Group Co., Ltd., Tokyo, Japan), soybean oil (Showa Sangyo Co., Ltd., Tokyo, Japan), linseed oil (Nippon Flour Mills Co., Ltd., Tokyo, Japan), and perilla oil (Asahi Group, Tokyo, Japan). Oils were used immediately after opening the container prior to the best-before date.

2.2. Aldehydes generated from vegetable oils

Emissions of aldehydes from vegetable oils alone or mixed with water or sodium hydroxide solution were determined using the passive emissions chamber method at room temperature (JSA, 2001). Three samples $-$ A, B, and C $-$ were tested, as described as follows. Sample A was 0.5 mL of vegetable oil. Sample B contained 0.5 mL of vegetable oil and mixed with 20 mL of ultra-pure water. Sample C contained 0.5 mL of vegetable oil and mixed with 20 mL of 2.5 mol/dm³ (100 g/dm³) sodium hydroxide solution (Wako Pure Chemical Industries, Ltd., Osaka, Japan). The samples were placed in sealed 120-mL glass bottles. For samples B and C, the bottles were shaken by hand for 1 min to mix the two phases. A DSD-DNPH cartridge (Sigma-Aldrich Co., St. Louis, MO, USA) was then hung in the bottle (Figure 1). The emitted aldehyde vapours were collected for 24 h.

2.3. Aldehydes generated from plasters containing vegetable oils

Emissions of aldehydes from plasters containing vegetable oils were determined using a dynamic emissions chamber method; the Japanese Industrial Standard (JIS) A 1901 Small Chamber Method (JSA, 2009) (based on ISO 16000-9 (ISO, 2006)).

The plaster and oil were mixed at a ratio corresponding to the average commercial products in Japan. The principal components of plaster -50 g slaked lime (calcium hydroxide; Wako Pure Chemical Industries, Ltd.), 45 mL ultra-pure water, and 0.5 mL vegetable oil ― were placed in a beaker and mixed with a spatula. The plaster sample was spread with a spatula over an 18×18 cm² area on a cleaned glass plate of 20×20 cm^2 in size. The thickness of the plaster layer was approximately 1 mm (Figure 2). A plaster sample containing 50 g slaked lime and 45 mL ultra-pure water was used as a control.

Prior to the experiment, the chamber was washed with methanol (Wako Pure Chemical Industries, Ltd.) and dried to remove any contaminants from the chamber itself. The 2.0 \times 10⁻² m³ chamber was supplied with purified and humidified air at a ventilation rate of 0.5 ± 0.05 times/h, which is equivalent to an air flow rate of $1.0 \times$

 10^{-2} m³/h. The temperature and relative humidity inside the chamber were held constant at 28 ± 1 °C and 50 ± 5 %, respectively (JSA, 2009).

The aldehydes in the air inside the chamber were collected on an LpDNPH S10L (Sigma-Aldrich Co.) cartridge for 30 min with a sampling volume of approximately 5.0 \times 10⁻³ m³. The air samples were collected at elapsed times of 1 day, 2 weeks, and 4 weeks according to JIS A 1901 (JSA, 2009). An extra sample was taken after 13 weeks.

2.4. Conditions of analysis of high-performance liquid chromatography

2,4-Dinitrophenylhydrazine (DNPH) derivatives collected on DSD-DNPH and LpDNPH S10L cartridges were eluted using 5.0 mL acetonitrile (High-performance liquid chromatography (hereafter, HPLC) grade; Wako Pure Chemical Industries, Ltd.). Aldehydes were analysed using a Shimadzu Nexera HPLC (Shimadzu Corp., Kyoto, Japan) with a Zorbax Bonus-RP column $(4.6 \text{ mm } i.d., 250 \text{ mm } length, 5 \text{ \mu m } particle)$ size; Agilent Technologies Inc., Santa Clara, CA, USA). The flow rate was 1.0 mL/min with the spectrophotometric sensor set at a wavelength of 360 nm. The column oven temperature was 40°C. The eluting solvent was 60% acetonitrile and 40% water and the injection volume was $20 \mu L$ (JSA, 2009).

A standard TO11/IP-6A aldehyde/ketone-DNPH mix (Sigma-Aldrich Co.) was used

for quantification of formaldehyde, acetaldehyde, propionaldehyde, butyraldehyde, valeraldehyde and hexaldehyde.

2.5. Calculation of the emissions factor

The area-specific emissions factor EF_a (μ g/m²/h) was calculated using equation (1) as described in JIS A 1901 (JSA, 2009)

$$
EF_a = C_t \times n/L \tag{1}
$$

where C_t is the concentration of the aldehydes in the small chamber at elapsed time t $(\mu g/m^3)$, *L* is the sample load factor (m^2/m^3) and *n* is the air change rate (times/h).

2.6. Calculation of the indoor air concentration

The incremental increase in the air concentration ΔC was calculated as described in JIS A 6921 (JSA, 2003). The surface area was assumed to be 24 $m²$ for the walls and 7 $m²$ for the ceiling, and the volume of the room was assumed to be 17.4 $m³$ based on the Danish Standard, INF 90:1994 (DS, 1994). The incremental increase in the air concentration ΔC (μ g/m³) when using building materials over surface area A_R was calculated using equation (2) as follows

$$
\Delta C = (E F_a \times A_R) / (n_R \times V_R) \tag{2}
$$

where EF_a is the area-specific emissions factor (μ g/m²/h), A_R is the surface area of the walls and ceiling $(24 + 7 \text{ m}^2)$, n_R is the air exchange rate in the room model $(0.5$ times/h), and V_R is the volume of the room model (17.4 m³). This equation was developed on the basis of a single-compartment mass balance for the emitted aldehydes in the well-mixed room air. Room ventilation was assumed to be the only removal mechanism for the emitted aldehydes, and the outdoor concentrations were set to zero. The emission rates were assumed to be constant at a given time.

3. Results

3.1. Aldehydes generated from vegetable oils

Emissions of aldehydes from the vegetable oils are shown in Table 1. For samples A and B, no aldehydes were detected from the oils except for perilla oil. In contrast, for sample C (to which sodium hydroxide was added), aldehydes were detected from all of the vegetable oils tested. Acetaldehyde and propionaldehyde were detected from all types of oil. Hexaldehyde was detected from soybean oil, linseed oil, and perilla oil. The amounts of aldehydes, particularly propionaldehyde and acetaldehyde, generated from linseed and perilla oils were, in general, higher than those from rapeseed oil and soybean oil.

3.2. Aldehydes emitted from plasters containing vegetable oils

Emissions of aldehydes from plaster containing vegetable oils are shown in Table 2. No aldehydes were emitted from the control plaster, to which no vegetable oil was added (Table 2). Emission factors for all the aldehydes other than formaldehyde were highest one day after spreading and then decreased over time. Total aldehyde emissions after 13 weeks from plasters with rapeseed oil, soybean oil, linseed oil, and perilla oil decreased to 3.7%, 1.7%, 2.4%, and 5.5% of their initial values, respectively.

The plaster with soybean oil strongly emitted hexaldehyde throughout the study period. On the other hand, the plasters with linseed oil or perilla oil strongly emitted propionaldehyde and acetaldehyde. For the plaster containing rapeseed oil, propionaldehyde had the highest emission factor (262 μ g/m²/h) one day after the test started. Emissions of acetaldehyde, propionaldehyde, and hexaldehyde continued throughout the test period.

For the plaster containing soybean oil, hexaldehyde had the highest emissions factor of 2080 μ g/m²/h one day after spreading. Although the emission factor for hexaldehyde decreased over time, emissions continued at a high level throughout the testing period. Other aldehydes, including acetaldehyde, propionaldehyde, and valeraldehyde, were also quantifiable throughout the test.

For the plaster containing linseed oil, propionaldehyde had the highest emission factor of 1680 μ g/m²/h one day after spreading. Although emissions of propionaldehyde decreased over time, they continued at high levels throughout the test period. Significant emissions of propionaldehyde and acetaldehyde continued for as much as 13 weeks. The emission patterns for the plaster containing perilla oil were similar to those for the plaster containing linseed oil.

3.3. Estimated indoor air concentrations of aldehydes emitted from plaster with vegetable oil in a model room

Predicted indoor concentrations of total aldehydes in a model room the day after plaster application were 1970, 11,200, 8890 and 10,100 μ g/m³ for plasters with rapeseed oil, soybean oil, linseed oil and perilla oil, respectively (Table 3). Indoor air concentrations of aldehydes and total aldehydes for the plaster with rapeseed oil were the lowest among the tested oils. The incremental room air concentration of aldehydes 13 weeks after plaster application was predicted to be from 72.4 (rapeseed oil) to 550 $g/m³$ (perilla oil).

4. Discussion

In the present study, aldehyde emissions from plasters containing vegetable oil were assessed and the resulting concentrations of aldehydes in room air were calculated. The results show that the aldehydes emitted from plaster varied depending on the type of vegetable oil the plaster contained. The aldehydes emitted from the plasters were assumed to be related to the types of the unsaturated fatty acids contained in the vegetable oils.

Vegetable oils contain the unsaturated fatty acids ― oleic acid, linoleic acid and α -linolenic acid — in different percentages (Japan Oil Chemists' Society, 2001). Rapeseed oil, soybean oil and linseed/perilla oil primarily contain oleic acids, linoleic acids and α -linolenic acids, respectively (Japan Oil Chemists' Society, 2001). Furthermore, other unsaturated fatty acids are also present in various quantities (Japan Oil Chemists' Society, 2001). In lipid oxidation, a free radical is formed by removing a labile hydrogen from a carbon atom adjacent to a double bond (Vollhardt and Schore, 2011). Therefore, among the three unsaturated fatty acids, α -linolenic acid with three carbon–carbon double bonds is most unstable, and linoleic acid with two double bonds is more unstable than oleic acid with one double bond. Hexaldehyde could be generated from linoleic acid by cleavage at the sixth carbon from the end (Figure 3), whereas

propionaldehyde could be generated from α -linolenic acid by cleavage at the third carbon from the end (Figure 3). Additional generation of propionaldehyde could presumably occur by cleavage at the sixth and ninth carbons. Because the vegetable oil samples used in this experiment were relatively fresh, autoxidation did not begin immediately and there was very little generation of aldehydes at the beginning of the tests.

The aldehydes were likely generated from the unsaturated fatty acids contained in the vegetable oils by hydrolysis owing to the high alkalinity. When sodium hydroxide is added to vegetable oil (triglycerides), triglycerides will break down to di- and mono-glycerides through hydrolysis and will ultimately degrade to free fatty acids (salts). The unsaturated free fatty acids are subject to oxidation to hydroperoxides, which decompose to give by-products consisting mainly of aldehydes (Figure 3). Propionaldehyde was strongly emitted from both linseed oil and perilla oil under alkaline conditions, likely owing to the similar compositions of unsaturated fatty acids in these vegetable oils. Moreover, acetaldehyde was emitted from all of the vegetable oils contained in the plasters, possibly owing to a radical oxidation reaction in which the double bond of an unsaturated fatty acid is cleaved and an adjoining carbon–carbon double bond stabilizes the radical, yielding acetaldehyde (Diaz et al., 2008; Tanaka et al., 2009; Diaz et al., 2005; Juita et al., 2011).

The Japanese Ministry of Health, Labour, and Welfare has a guideline for acetaldehyde concentration of 48 μ g/m³ to prevent the sick-building syndrome (Japan Ministry of Health, Labor, and Welfare). The guideline represents the concentration of a substance which, if reached or exceeded, requires immediate action, as this concentration could pose a health hazard, particularly for sensitive people who stay indoors over long periods of time. The calculated concentrations of acetaldehyde emitted from the plasters containing soybean, linseed and perilla oil four weeks after the plaster application were higher than this indoor air guideline. A guideline of 100 μ g/m³ has been established by the German Federal Environmental Protection Agency for aldehydes for C_4-C_{11} saturated acyclic aliphatics (The German Federal Environmental Protection Agency). The concentration of hexaldehyde emitted from the plaster containing soybean oil 13 weeks after plaster application exceeded this guideline.

The indoor air concentrations in new residential housing in which symptoms of sickbuilding syndrome appeared were reported by Takeda (Takeda et al., 2009) and Takigawa et al. (Takigawa et al., 2010, 2012). The lowest reported values are 20.8 $g/m³$ for acetaldehyde, 7.7 $\mu g/m³$ for propionaldehyde, 2.2 $\mu g/m³$ for butyraldehyde, 3.7 μ g/m³ for valeraldehyde and 9.5 μ g/m³ for hexaldehyde. The indoor air

concentrations shown in Table 3 were higher than these reported values for all the plasters throughout the test period, up to 13 weeks after plaster application.

Furthermore, propionaldehyde is regulated as a malodorous compound at the site boundary of an establishment such as a factory by the Japanese Ministry of the Environment (Japan Ministry of the Environment). The calculated concentrations of propionaldehyde emitted from plaster containing linseed oil and perilla oil exceeded the guideline (119 μ g/m³) even 13 weeks after plaster application.

In this study, reagents and ultra-pure water were used to eliminate the influence of impurities. However, in actual use, plaster may be mixed with seaweed, sand, and tap water, among other things. Iron and copper, in particular, accelerate oxidization of fatty acids (Juita et al., 2011). Therefore, aldehyde concentrations may vary owing to impurities in actual indoor air. Moreover, in this study, indoor air concentrations were predicted using a room space model (JSA, 2003). The predicted concentrations were dependent on the surface area to volume ratio of the room (A/V) and the air exchange rate. The indoor chemical reactions that have received most attention to date are those involving interactions of ozone with terpenoids in the gas phase and with the surfaces of common materials, furnishings and the occupants themselves (Morrison, 2008; Weschler, 2004, 2011).

5. Conclusions

This study demonstrated that aldehydes are emitted from plasters containing vegetable oils. Aldehydes are believed to be emitted from the plaster by autoxidation of the unsaturated free fatty acids generated by hydrolysis of vegetable oils due to the high alkalinity of the plaster. The findings show that the alkalinity of calcium hydroxide in the plaster would accelerate the formation of aldehydes significantly. Predicted concentrations of aldehydes in indoor air exceeded the guidelines of the Japanese Ministry of Health, Labour, and Welfare, the Japanese Ministry of the Environment and the German Federal Environmental Protection Agency. These results suggest that using plaster containing vegetable oil may pose a health hazard, particularly for sensitive individuals. Thus, addition of vegetable oil to plasters used as indoor building materials should be avoided.

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Tables and Figures

Table 1. Aldehydes generated from vegetable oils.

^aNot detected: \leq 0.20 μg.

Table 2. Emission factors of aldehydes emitted from plaster containing vegetable oil.

^aNot detected: $\leq 5.0 \text{ µg/m}^2/h$.

Table 3. Indoor air concentrations of aldehydes predicted from the emission factor of the test specimen of the plaster with vegetable oil.

^aNot detected: <17.8 μ g/m³.

Figure 1. Example of the experiment emissions of aldehydes from vegetable oils alone

or mixed with water or sodium hydroxide solution.

Figure 2. Example of the experiment emissions of aldehydes from plasters containing

vegetable oils.

Figure 3. Generation of specific aldehydes from vegetable oils mixed into lime plaster. Linoleic acid from soybean oil is oxidized to form the main precursor hydroperoxides, which break down to secondary byproducts such as hexaldehyde (major product), propionaldehyde and acetaldehyde (Diaz et al., 2008; Tanaka et al., 2009; Diaz et al., 2005). α-Linolenic acid from linseed or perilla oil produces different aldehydes, including propionaldehyde (the major product) and acetaldehyde, through the same process (Diaz et al., 2008; Tanaka et al., 2009; Diaz et al., 2005).

Linseed or perilla oil Primarily contain α-linolenic acid

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