MATERIAL EMISSION AND IAQ
– RECENT EXPERIENCES IN JAPAN–

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ABSTRACT
In Japan problems caused by aldehydes and VOCs have been recognized to be serious especially in new constructed houses. During 2000 a big field study on over 4500 houses was conducted in Japan by the Ministry of Land, Infrastructure, and Transport (formally Ministry of Construction). For indoor air concentration of formaldehyde, 27% of houses were over the guideline of 100μg/m^3. About 12.3% houses were over the toluene guideline of 260μg/m^3. From 2000 the Ministry of Health, Labor and Welfare has started to establish guidelines on VOCs. Finally new building standard law on sick house issues in Japan was promulgated on 2002 and it was enforced since July 1, 2003. To prevent "sick house syndrome", suitable ventilation and reduction of emission rates from building products are strongly required. In Japan a small-scale chamber method was standardized as JIS A 1901 in 2003. In this paper background of recent issues is described. Examples of emission rates measurement with 20 L chamber were also shown for flooring materials and EPS (expanded polystyrene form).

Keywords: Regulation, formaldehyde, chamber, VOCs, emission rate, building materials

INTRODUCTION
In Japan problems caused by aldehydes and VOCs have been recognized to be serious especially in new constructed houses. The Ministry of Health, Labor and Welfare, Japan published a first guideline on August 1997, where concentration of formaldehyde in the air should be less than 100μg/m^3. In 1998 the Government Committee on Healthy House (under the Ministry of Construction, the Ministry of Health, Labor and Welfare, the Ministry of International, Trade and Industry, the Forestry Agency, formal names in 1998) published a guidebook for architects and builders. They also published a manual for house users and owners. In this guidebook six chemical substances were pointed out. Namely, they are formaldehyde, toluene, xylene, wood preservatives, termite chemicals, and plasticizers. Japanese Home Builders Association published their guidelines on 1999 and 2001. During 2000 a big field study on over 4500 houses was conducted in Japan by the Ministry of Land, Infrastructure, and Transport (formally Ministry of Construction). It shows that the average concentration of formaldehyde is 89μg/m^3. For indoor air concentration of formaldehyde, 27% of houses were over the guideline of 100μg/m^3. About 12.3% houses were over the toluene guideline of 260μg/m^3.

From 2000 the Ministry of Health, Labor and Welfare has started to establish guidelines on VOCs [1]. Table 1 shows the guideline values on the different kinds of chemicals published by January 2002. This guideline has strong impacts on Japanese housing manufactures and industries because more than 1.2 million new houses have been constructed per year (2003). Also a scientific project on indoor chemical pollution (IAPOC) has been promoted by the Architectural Institute of Japan. Furthermore, standardization actions on the emission test and chemical analysis have been conducted under the Ministry of Economy, Trade and Industry,

where the small chamber method is applied. Finally new building standard law on sick house issues in Japan was promulgated on 2002 and it was enforced since July 1, 2003. [2]
To prevent “sick house syndrome”, suitable ventilation and reduction of emission rates from building products are strongly required. Several small-scale chambers for measuring aldehydes and VOCs emission rates have been proposed in the world (Tichenor [3], Wolkoff et al. [4], Gunnarsen et al. [5], Brown [6]). These small test chambers have often been used to determine the chemical emission rates from building materials (Colombo et al, [7], Jensen et al. [8], Lundgren et al, [9], Knudsen, et al. [10], Risholm-Sundman [11], Uchiyama et al. [12]). In Japan a small-scale chamber method was standardized as JIS A 1901 in 2003. In this paper examples of emission rates measurement were also shown for flooring materials and EPS (expanded polystyrene form).

Table 1. Guideline values for VOCs by the Ministry of Health, Labor and Welfare (Jan. 2002)

Substance	Guideline
formaldehyde	100μg/m^3
acetaldehyde	48μg/m^3
toluene	260μg/m^3
xylene	870μg/m^3
p-dichlorobenzene	240μg/m^3
ethylbenzene	3800μg/m^3
styrene	220μg/m^3
chlorpyrifos	1(0.1)* μg/m^3
DBP	220μg/m^3
tetradecane	330μg/m^3
DEHP	120μg/m^3
diazinon	0.29μg/m^3
fenobucarb	33μg/m^3
TVOC (tentative)	400μg/m^3

*for children

THE AMENDED BUILDING STANDARD LAW ON SICK HOUSE ISSUES IN JAPAN

Overview

The Amended Building Standard Law was promulgated on July 12, 2002 and it has been enforced from July 1, 2003.

Two chemicals of chlorpyrifos and formaldehyde were regulated. The use of building materials containing chlorpyrifos in buildings with occupied space is prohibited. The area size of formaldehyde emission building materials which can be used as interior finishing materials is restricted according to the type of room and the ventilation rate. Even if the emission from building materials is negligible, formaldehyde may be also emitted from furniture and other products. For this reason, the mechanical ventilation system is, in principle, mandatory to install in almost all types of buildings.

The materials used in ceiling cavities, attics, cavities underneath floors, wall, storerooms and other similar locations must be low formaldehyde emission rates, or ventilation system should be installed.

Building materials - formaldehyde

The building materials, which have become subject to controls, are listed exclusively in Notification No.1113 of the Ministry of Land, Infrastructure and Transport, 2003. Built-in furniture, kitchen cabinets, etc. using the Notification building materials are also subject to controls.[13] When using such building materials, it is required to classify these materials into the class (grade) of formaldehyde emission rate. Materials other than Notification building materials can be used without restriction as interior finishing materials such as stone, metal, natural wood and so on. In Table 2 the classes of formaldehyde emission rates are shown.

Table 2. Classes of formaldehyde emission rates

Formaldehyde Emission rate (*1)	Building materials stipulated in Notification		Restrictions on interior finishing materials
	Name	Relevant Standard	
Over 120 μg/m^{2}h	Type 1 formaldehyde-emitting building materials	Equivalent to old E_2, Fc_2 standards under JIS, JAS systems, unclassified	Use prohibited
Over 20 μg/m^{2}h But not more than 120 μg/m^{2}h	Type 2 formaldehyde-emitting building materials	F** under JIS, JAS systems	Limited area of use
Over 5 μg/m^{2}h But not more than 20μg/m^{2}h	Type3 formaldehyde-emitting building materials	F*** under JIS, JAS systems	
Up to 5 μg/m^{2}h		F**** under JIS, JAS systems	No restrictions

(*1) test condition: 28°C, 50%RH, 100μg/m^3 (formaldehyde concentration=guideline value)

Designers, superintendents of construction work and housing suppliers are required to submit the records of acceptance inspection which give a concrete and detailed description of the class (grade), category, quantity, labels or documents confirmation (JIS, JAS, copy of Ministerial approval certificate). Therefore, it has been highly recommended for the manufactures by the Ministry to obtain JIS, JAS, and Ministerial Approval of the Notification building materials. Until April 2004, over 1400 materials are certified as Ministerial Approval ones.

HOW TO DETERMINE THE GRADE OF BUILDING MATERIALS BY THE MINISTRY APPROVAL

Small chamber method (JIS A1901)

To determine the emission rate of formaldehyde, a sample is tested in accordance with the provisions of JIS A 1901 [14]. JIS A 1901 is a standard on “Determination of the emission of volatile organic compounds and aldehydes for building products - Small chamber method”. Twenty-litter chamber is widely used in Japan as shown in Appendix 2 of JIS A 1901. The test conditions, in principle, comply with the conditions stipulated in Section 6 of JIS A 1901. The following conditions should be applied.

Test temperature	28°C ± 1°C
Relative humidity	50% ± 5%
Ventilation rate	0.5 1/h ± 0.05 1/h
Loading factor	2.2 m^2/m^3 as a standard

However, the loading factor can be adjusted whenever necessary to prevent the formaldehyde concentration in the chamber from exceeding 100μg/m^3 during the testing

Sampling of chamber air is carried out one day, three days and seven days after the start of the tests. The grade of material is based on the value on the seventh day. At the moment, JIS A1901, the small chamber method is used for only formaldehyde by the regulation purpose. However, a lot of manufactures and housing suppliers voluntary measure the emission rates of VOCs by this method. ENV-717-1 [15] method and ENV-13419-1 [16] are compatible with JIS A1901.

Desiccator method (JIS A1460 and JAS)

Specimens of plywood, timber flooring, structural panels, MDF, particleboard, wallpaper, starch wallpaper adhesives, starch adhesives and paints for fixtures containing formaldehyde-water solutions can be tested by using the glass desiccator method (JIS A1460) [17] and evaluation criteria defined in individual Japan Industrial Standards (JIS) or Japan Agricultural Standards (JAS) [18] as shown in Table 2. Materials for which the Japan Agricultural Standards stipulate testing and evaluation criteria based on the acrylic desiccator method, such as glued laminated timber and laminated veneer lumber, can be checked using the acrylic desiccator method. We pointed out there is a disclipancy between acrylic and glass methods. JIS A 1460 is a standard on "Building boards, Determination of formaldehyde emission - Desiccator method". The determination of concentration of formaldehyde absorbed in distilled water or deionized water is analized based on the Hantzsch reaction in which the formaldehyde reacts with ammonium ions and acetylacetone to yield diacetyldihydrolutidine (DDL). Figure 1 shows the scheme of glass desiccator method.

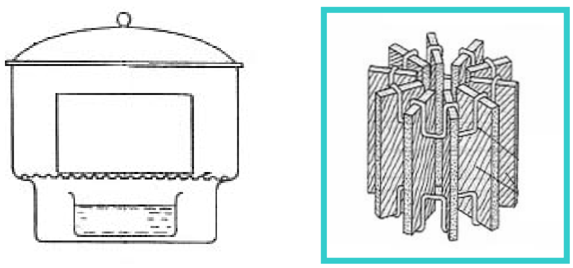

Fig. 1. Scheme of desiccator method

CHARACTERISTICS OF SMALL-SCALE CHAMBER ADPAC

Small-scale chamber, ADPAC [19], was developed in Japan by us with its performance in compliance with JIS A1901, ASTM [20, 21], ECA reports [22, 23, 24, 25], and ENV 13419-1. At the moment over 800 chambers of ADPAC are already used in Japan. Figure 2 shows the main chamber made of stainless steel (SUS304) and the air control unit. Air control system has an air supplying unit, a humidifier, and pumps. Purified air was used for ventilation. Figure 3 shows the schematic diagram of ADPAC system. Seal box was used to prevent cut ure 3 shows the schematic diagram of ADPAC system. Seal box was used to prevent cut edge effect, which allowed chemical emission only from one side surface of the test piece. When two seal boxes are used, total surface area is $0.044m^2$ and the loading is $2.2m^2/m^3$.

Fig. 2. 20L chamber (ADPAC)

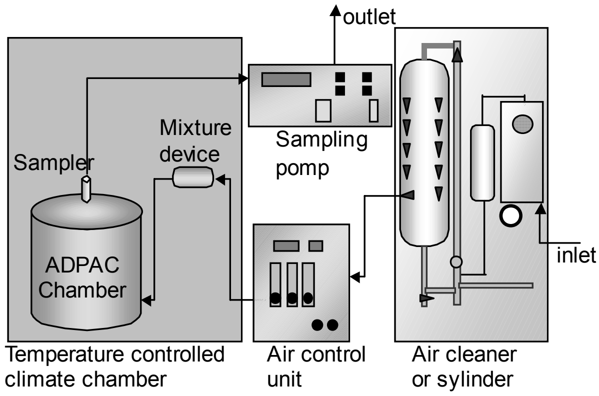

Fig. 3. Schematic diagram of ADPAC system

Methods

Before setting up the chamber and seal boxes, they were washed with water, and baked out using oven at 260°C to eliminate any pollutants from the chamber itself. ADPAC System upplied purified and humidified air with a given ventilation rate. Temperature and relative humidity inside the chamber were kept constant. The test conditions in this paper were shown in Table 3. Test pieces sealed with seal boxes were set in the chamber, and the air inside the chamber was sampled after 15 hours. Sampling conditions were shown in Table 4. Throughout the measurements, air temperature and relative humidity inside the test chamber were kept constant at 25±1°C and 50±4%, and ventilated at $0.5\ h^{-1}$. This test was carried out before JIS A1901 was established, so ambient air temperature was kept at 25°C in stead of 28°C. Aldehydes were analyzed by HPLC, and TDS/ GC/MS was used for VOCs as shown in Tables 5-6. Analysis conditions for VOCs were slightly different among materials, so that for EPS was tabled here. In this paper, TVOC is defined that the all areas of the peaks after n-hexane were converted to concentrations using the toluene response factor. The peak area under 10 was defined as the limit of detection.

Table 3. Test conditions

Chamber Volume	20 L
Loading Factor	$2.2\ m^2/m^3$
Temperature	25 ± 1 °C
Relative Humidity	50 ± 4 %
Ventilation Rate	$0.5\ h^{-1}$

Table 4. Sampling conditions

	aldehydes	VOCs
Sampler	DNPH-Silica cartridge	for Flooring: Tenax TA (60/80mesh) for EPS: Carbopack B (60/80mesh) + Carboxen 1000
Air Flow Rate	167 mL/min	167 mL/min
Total Volume	10 L	3.2 L

Table 5. Analysis conditions for aldehydes

HPLC	Waters 2640 Separations Module alliance, Waters 2487 Dual λ Absorbance Detector
Column	Nova-Pak® C_{18}, $\varphi 3.9 \times 150$ mm
Mobile Phase	65%water /30%acetonitrile /5%THF
Column Oven Temperature	40°C
Sample Injection Volume	20 μL
Detector	UV, operating at 360 nm

Table 6. Analysis conditions for VOCs

TDS	Perkin Elmer ATD400
GC/MS	HP6890 /5973B
Column	HP-VOC, 0.32mmφ×60m×1.8μm
Oven temperature	35ºC(2min) →15°C/sec(4min) → 2.5°C/sec(2min) → 5°C/sec → 250°C(1min)
Mode	SCAN

MEASUREMENT OF FLOORING MATERIALS

In Japan majority of wooden flooring material in houses is made of plywood. This may be the main source of formaldehyde in houses. Three kinds of flooring materials were tested during 28 days. Table 7 shows the characteristics of flooring materials. Test pieces were cut in the factory before sending to the laboratory by 165mm x 165mm. Test pieces were carefully sealed by the aluminum sheet and they were packed in the PET bag. Just before measurements were started, seal was opened. Cut edges of test pieces were sealed by the box.

Table 7. Characteristics of flooring

	Base	Grade of JAS*	Surface coating
A	Plywood	F_{C0}	Urethane type
B	Plywood	F_{C0}	UV cure coating
C	Plywood	F_{C0}	UV cure coating

* JAS defines formaldehyde concentration under 0.5mg/L in water by desiccator method as F_{C0}

Figure 4 shows the emission rates of formaldehyde for flooring materials. At the first day emission rates for flooring A and B were almost same as 23μg/m^{2}h. That for flooring B was 17μg/m^{2}h. After 28 days emission rate of formaldehyde for the flooring material A was 15μg/m^{2}h and those for flooring B and C were around 10μg/m^{2}h. These emission rates are relatively high, because all of them are classified into F*** category by the Japanese Agricultural Standard. Figure 5 shows the decay of TVOC for three flooring materials. Here TVOC is calculated by toluene equivalent. At the first day TVOC for the flooring material A was 570μg/m^{2}h and that for the flooring B was 490μg/m^{2}h and that for the flooring C was 300μg/m^{2}h.

Ninety four percent of TVOC from the flooring material B was toluene. On the other hand, for the flooring B, ethyl benzene was 20% and xylene was 15%. Flooring A also included aromatic hydrocarbon. For the flooring C, 20% of them was 1-methoxy butane. Little emission of toluene and ethyl benzene was observed. Even when the emission rates of TVOC were same level, characteristics of VOCs were quite different by materials. For three flooring materials, even they are made of wood, α-pinene and limonene were under detected limit. We observed α-copaene, β-caryophyllene, β-elememe.

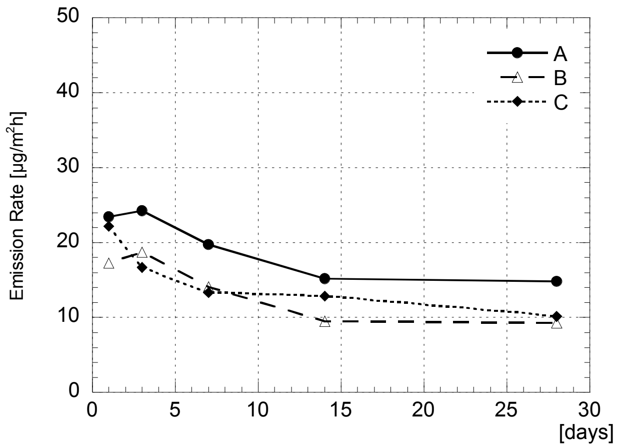

Fig. 4. Decay process of formaldehyde –Flooring

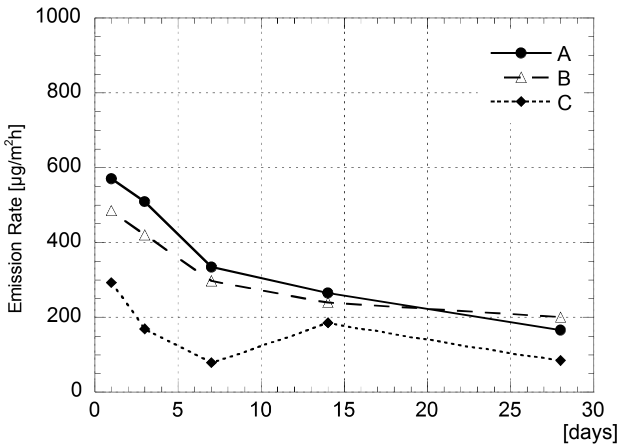

Fig. 5. Decay process of TVOC –Flooring

MEASUREMENT OF THERMAL INSULATION BOARD EPS

Preparation of test pieces

Four kinds of expanded polystyrene form (EPS) were measured during 14 days. EPS A was bought at a DIY shop. EPS B was one of the present market products in Japan. EPS C was the improvement product of B. EPS D was made in USA. Just before the measurements were started, test pieces were cut from the board. Cut edges of test pieces were not sealed.

Results

Figure 6 shows the decay process of TVOC. The emission rate for EPS A was 586μg/m^{2}h after one day and 206μg/m^{2}h after 14 days. That for EPS B was around 40μg/m^{2}h until 7 days. However, after 10 days the emission from EPS B was increased. That of EPS A was 210μg/m^{2}h after 14 days. The reason was not clear. Presumably it may be estimated that the chemical substances inside EPS was diffused to the surface and increased the emission rates. We observed same phenomena for EPS at several different measurements. It was found that the emission of VOCs from EPS A might be continued for long period. At the first day emission rate for EPS C was 42μg/m^{2}h, it was still continued at 10~20μg/m^{2}h level from 5 days after.

Figure 7 shows the decay process of styrene. Styrene was major substances of TVOC. These decay curves were coincide with those of TVOC. As described in the introduction section, the government shows guideline value of styrene (see in Table 1). EPS is made of styrene monomer. Some amount of styrene monomer is still remained in it. Emission rates of styrene for EPS A and B were higher than American EPS D. That for EPS D was observed less than 10μg/m^{2}h during that period. Improvement for Japanese EPS is highly recommended. Aromatic hydrocarbons such as styrene, toluene, xylene and ethyl benzene were highly emitted, and halocarbons such as chloromethane and dichloromethane were also emitted. Particularly toluene and styrene emission from EPS A was observed. Emission rate of toluene for EPS A was 140μg/m^{2}h at the first day and that of styrene was 125μg/m^{2}h. Although it was decreased after 14 days that of toluene was 87μg/m^{2}h and that of styrene was 70μg/m^{2}h. The emission of styrene from EPS C was low, but emission of chloromethane after one day was observed at 39μg/m^{2}h. Aldehydes were not detected from all EPS.

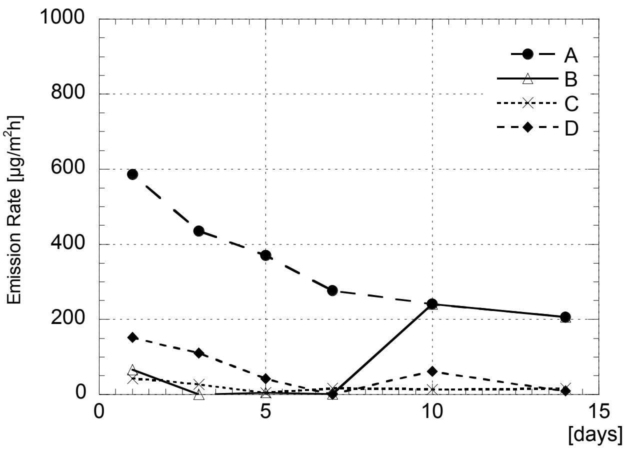

Fig. 6. Decay process of TVOC –EPS

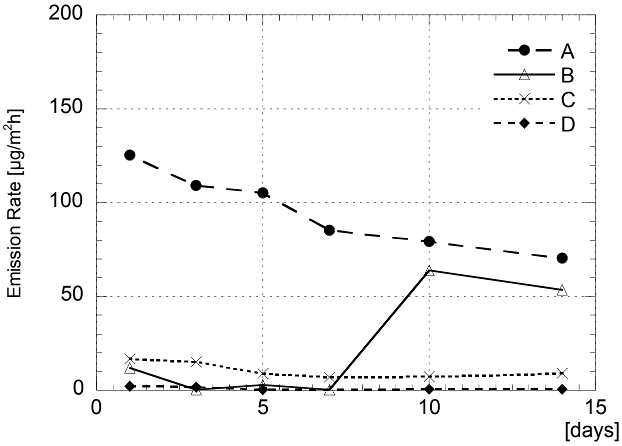

Fig. 7. Decay process of styrene –EPS

CONCLUSIONS

Recent experiences in Japan regarding to the sick house issues are mentioned. The amended building law and guidelines are also described. A small-scale chamber method was developed and standardized as JIS A1901 to measure the emission rates of chemical substances from building materials. By using 20L small chamber, flooring materials, and expanded polystyrene forms were measured in case studies.

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