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Sick House/Building Syndrome in JAPAN –Current Status and Practical Research on Indoor Air Quality–

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Abstract : Many Japanese today are suffering from health disorders related to their living environment, such as allergies and hypersensitivity to chemical substances. The Sick House/Building Syndrome has been a serious problem since 1996, due to low level exposure to hazardous chemicals such as formaldehyde and volatile organic compounds (VOCs) emitted in airtight houses. This paper aims to show current status of the syndrome in Japan and practical researches to promote prevention of, and improvement to indoor air pollution due to chemicals.

Keywords : indoor air quality, sick house/building syndrome

Introduction

Many Japanese today are suffering from health disorders related to their living environment, such as allergies and hypersensitivity to chemical substances. In particular, indoor air pollution by volatile organic compounds (VOCs) has been a serious problem since 1996 or so and became a matter of great social concern. The cases are popularly known as the Sick House Syndrome among Japanese people rather than the familiar Sick Building Syndrome, because majority of the patients consists of housewives and children living in newly-built apartment or detached houses. Besides, recent serious indoor air pollution problems in school facilities produced the new word, the Sick School Syndrome.

Traditional Japanese houses had been made of natural wooden materials and well ventilated, adapting to warm and humid climate. Nowadays, modern houses are being built with higher air-tightness (air change rate without mechanical ventilation is smaller than 0.2/h) to realize greater heating and cooling efficiency and hence save energy. In order to supply large number of such types of houses in a stable manner and low cost, highly processed building materials using chemical substances must be used such as plywood, particle board, MDF, paper walls and so on. In these highly airtight houses, indoor air was polluted by hazardous chemicals emitted from the materilas and then caused idiopathic environmental intolerance of residents, also known as multiple chemical sensitivities (MCS).

Although there are potentially numeral risks caused by environmental chemical substances, for example dioxins, endocrine disrupters, heavy metals and pesticides for agricultural uses, countermeasures regarding the syndrome have a higher priority to be carried out because of the fact the victims are consciously or in many cases unconsciously facing fear from the invisible gaseous chemicals. This paper aims to describe present status of the syndrome in Japan and practical researches to promote prevention from, and improvement to indoor air pollution by chemicals.

Case study in Tokyo (Kawashima *et al.*, 2003)

A woman, who had moved and lived alone in a newly-built apartment house at Tokyo in April 1999, began to feel unpleasant odor in the house. She gradually became annoyed from health disorders such as eye mucus, throat pain, bronchus irritation, headache, unpleasant feeling with muscles, diarrhea, fatigue and failure of memory. As she was working at a public library, she made a literature search by herself and suspected they were typical symptoms of the Sick House Syndrome. Then, she tried to grasp present status of air quality using a conventional gas monitor and found indoor air concentrations of formaldehyde at the living room ranged from 0.14 to 0.23 ppm. These values were much

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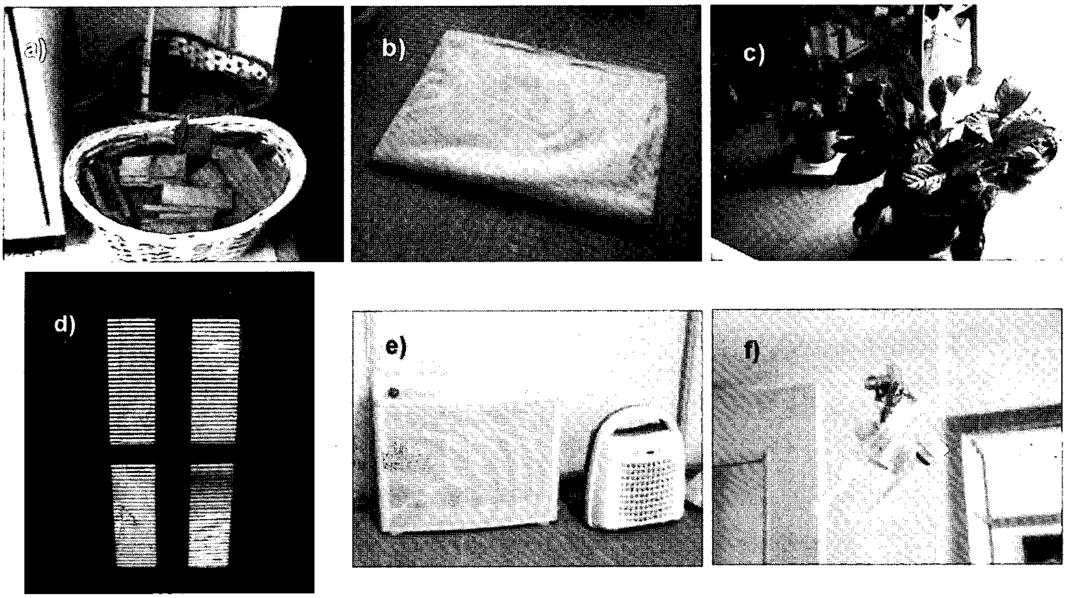

Fig. 1. User-friendly techniques employed by the patient, a) bamboo charcoal, b) pillow filled with charcoal flakes, c) plants which absorb VOCs, d) front door with air pathway, e) air cleaners, d) passive samplers for air quality monitoring.

higher than the indoor air quality guideline level (0.08 ppm). In March 2000, she was referred to a hospital and diagnosed as a possible patient of the multiple chemical sensitivities. Most patients are forced to either move to other houses (aged and stand in suburban area) or replace building materials suspected to emit the hazardous chemicals at great expense. However, those manners were not practical for her. She then decided to reduce indoor concentrations of chemicals by introducing air cleaning methods as shown in Fig. 1. The front doors with parallel open slits increased air exchange rate in the house. Air cleaners, bamboo charcoals and plants must have reduced indoor concentration of formaldehyde and VOCs. Significant field measurements using passive samplers conducted in 2002 proved effect of those efforts on the improvement of indoor air quality; concentrations of formaldehyde decreased to approximately 0.03 ppm.

In this case, the patient fortunately got over the symptom by identifying and removal of causing substances employing user-friendly techniques such as air cleaners and simple measurement devices. However, many patients do still suffer from the symptom. A questionnaire survey was conducted against 4,000 adults (older than 20) via interviews using the Environmental Exposure and Sensitivity Inventory (Miller, 1999) and showed 0.74% of the respondents had a potential risk for chemical sensitivities (Uchiyama and Murayama, 2003). This result, however, seems to be underestimated due to a bias of specific questionnaire to western people (it should be revised for Japanese people) and lack of data on children, probably more sensitive to environmental impurities.

Political Countermeasures

Indoor air quality guidelines have been set for concentrations of chemical substances in indoor air by Ministry of Health, Labour and Welfare, Japan (MOHLW). Chemical substances subjected to the guideline were added stepwise as shown in Table 1. Committee of the Ministry has also prepared manuals on the standard sampling and analytical procedures for each pollutant in a newly-built and occupied house, employing both active and passive sampling methods. These guidelines and manuals were aggressively referred by stakeholders on this matter, particularly by house-builders to consider the indoor air quality of houses. The indoor environment was then recognized as one of the housing performance. In accordance with the Housing Quality Assurance Act (2002), measurement of concentrations of chemical substances (formaldehyde, toluene, xylene, ethylbenzene and styrene) was included in fields of the Housing Performance Indication System, which enables a housing purchaser to compare housing from the view point of performance. Even though house-builders are not obliged to the system and measurements are carried out only when the client requests it, a number of builders have been voluntary adopting the indoor air quality monitoring following the manuals to enhance reliability on the houses they built.

Ministry of Land, Infrastructure and Transport, Japan (MLIT) have conducted a nation-wide survey on indoor air concentrations of chemical substances for approximately 4,600 housing units in 2000 (Murakami, 2003). The results showed priority should be given to taking measures to reduce formaldehyde concentration, because 27.3% of the units showed a higher value than the guideline with 0.071 ppm in average. Furthermore, toluene should receive priority after formaldehyde, because 12.3% of the units showed a higher value than the

Table 1. Chemical substances given priority for political countermeasures regarding the Sick House Syndrome in Japan

Indoor Air Quality Guidelines			Housing Performance Indicat. System	Standard for School Environ. Sanitation	Amended Building Standard Law	Major usage for housing
MOHLW			MLIT	MEXT	MLIT	
Date	Chemicals (cas no.)	Concnetration*	Subjectto measurement of indoor air concentration		Sublect to regulations	
1997. 6	formaldehyde (50-00-0)	100 μg/m³ (0.08 ppm)	○	○	○	synthetic resin used for plywood, particle board etc. and adhesives for wall paper
2000. 6	toluene (108-88-3)	260 μg/m³ (0.07 ppm)	○	○	-	paints, interior furnishing mat.
	xylene (1330-20-7)	870 μg/m³ (0.20 ppm)	○	○	-	paints, interior furnishing mat.
	p-dichlorobenzene (106-46-7)	240 μg/m³ (0.04 ppm)	-	○	-	insecticide
2000. 1	ethylbenzene (100-41-4)	3800 μg/m³ (0.88 ppm)	○	○	-	paints, interior furnishing mat.
	styrene (100-42-5)	220 μg/m³ (0.05 ppm)	○	○	-	insulator
	chlorpyriphos (2921-88-2)	1 μg/m³ (0.07 ppb) 0.1 μg/m³ (0.007 ppb) for infant	-	-	○	anti-ant agent
	di-n-butylphthalate (84-74-2)	220 μg/m³ (0.02 ppm)	-	-	-	plasticizer for paints
	TVOC	400 μg/m³**	-	-	-	
2001. 7	tetradecane (629-59-4)	330 μg/m³ (0.04 ppm)	-	-	-	paints, etc
	di-2-ethylhexylphthalate (84-66-2)	120 μg/m³ (7.6 ppb)	-	-	-	plasticizer for wall paper
	diazinone (33-41-5)	0.29 μg/m³ (0.02 ppb)	-	-	-	insecticides
	nonanal (124-19-6)	41 μg/m³ (7.0 ppb)**	-	-	-	paints, etc
2002.1	acetaldehyde (75-07-0)	48 μg/m³ (0.03 ppm)	-	-	-	synthetic resin for plywood etc.
	fenobucarb (3766-81-2)	33 μg/m³ (3.8 ppb)	-	-	-	Insecticide

MOHLW: Ministry of Health, Labour and Welfare, MLIT: Ministry of Land, Infrastructure and Transport.
MEX: Ministry of Education, Culture, Sports, Science and Technology.
*( ): coverted at 25 degC.
**darft value due to lack of enough information on hazardous properties, etc.

guideline (0.07 ppm) with 0.038 ppm in average.

The amended Building Standard Law, which stipulates minimum standards for building lots, structure and facilities, was enforced in July 2003, considering those results and strong toxicity on nerve system of anti-ant agent. This is a first regulation on a housing regarding the Sick House Syndrome, covering formaldehyde and chlorpyrifos as chemical substances subject to the regulation. The use of building materials containing chlorpyrifos is prohibited. Technical standards concerning formaldehyde are briefly as follows.

- Area size of formaldehyde-emitting materials used for interior furnishing is restricted according to the type of habitable room and the frequency of ventilation.
- Installation of ventilation equipment, which enables 24h ventilation, is mandatory in all buildings, considering the emissions from furniture.

The formaldehyde-emitting building materials are classified under four categories according to their formaldehyde emission rates; Type 1, whose emission rate of formaldehyde is greater than 0.12 $mg/m^2/h$, is prohibited for the use of interior furnishing materials. Type 2 (emission rate: 0.02-0.12 $mg/m^2/h$) and Type 3 (0.005-0.02 $mg/m^2/h$) are allowed to use in a limited area. No restriction was set for the materials whose emission rate is up to 0.005 $mg/m^2/h$. The emission rates are strictly determined by an established small chamber method (JIS A 1901, 2003).

It must be noted that the law allows following types of habitable rooms are exempted from the restriction: rooms equipped with centrally controlled air conditioner and those approved by the Minister as rooms in which it is possible to maintain the formaldehyde concentration at no more than 0.1 mg/m^3(0.08 ppm) throughout the year. In the latter case, the use of air cleaning materials which remove formaldehyde gases by adsorption and decomposition are allowed to use.

Removal of Formaldehyde and VOCs

Numerous studies relating to removal of formaldehyde and VOCs by air-cleaning materials

Table 2. Methods and materials used for removal of formaldehyde and VOCs

Principle	Methods/Materials	Mechanism
Physisorption	Activated charcoal	Adsorption by charcoals having micro porous structures
	Porous ceramics	Adsorption by natural and synthesized ceramics (zeolite, sepiolite, silica etc)
	Natural fibers	Adsorption by fibers and unwoven clothes such as cellulose, wool and so on
Chemisorption	Inorganic compounds	Fixation of HCHO by chemical reactions with inorganic compounds (metal complex, ammonium sulfate)
	Organic compounds	Fixation of HCHO by chemical reaction with organic compounds having a NH_2 group, normally impregnated on the solid sorbent
	Extracts	Fixation of HCHO by chemical reaction with natural products such as chitin, chitosan, green tea catechins
Decomposition	Photocatalyst	Oxidative decomposition of VOCs by photocatalyst (typically TiO_2) under UV/Visible light
	Ozonolysis	Oxidative decomposition of VOCs by ozone (attention; ozone is harmful)
	Catalytic decomposition	Oxidative decomposition of VOCs by metal/metal oxides at room to elevated temperature
	Modified charcoal	Decomposition of HCHO by $KMnO_4$ supported on charcoal
	Plasma discharge	Decomposition of VOCs by radials produced during plasma discharge
	Minus ions	Definition and effect of "minus ions" are controversial (suspicious)
	Activated oxygen	Oxidative decomposition of VOCs by activated oxygen oroduced by a catalyst
Digestion	Bacterial	Bacterial digestion of VOCs
Blocking	Resins	Coating of resinson the building materials

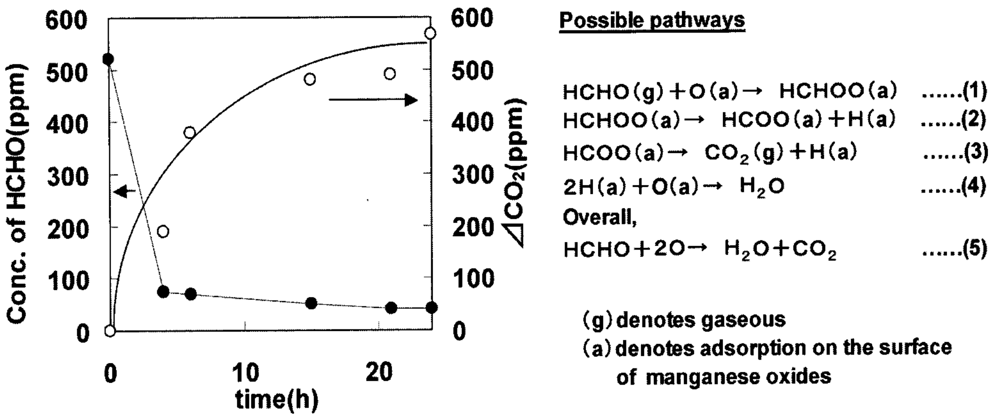

Fig. 2. Time courses of concentrations of HCHO (●) and produced CO_2 (○) through the reaction between fine MnO_2 and HCHO at 21°C.

have been conducted in Japan. There are also many kinds of commercially available products, such as air cleaners, ventilation system with purifying filters, passive type air cleaning materials, paints, spray and so on, based on various principles as shown in Table 2.

Author has developed air-cleaning materials consisting of activated carbon particles and manganese oxides, by which formaldehyde is decomposed into carbon dioxide even at room temperature (Sekine and Nishimura, 2001a). According to the results of screening of reactivity between metal oxides and formaldehyde vapor, MnO_2 showed highest removal efficiency with a significant production of carbon dioxide in vapor phase (Fig. 2) and without emissions of harmful byproducts such as formic acid and carbon monoxide (Sekine, 2002a).

The removal efficiency of a board-like air-cleaning material was investigated in a passive mode using a kinetic approach. First order removal rate constant (/h), k corresponding to air change rate was characterized for the board as a function of the loading factor (ratio of applied board area to space volume, m^2/m^3), L/V, and was found directly proportional to L/V with a slope of 25 at 25°C. When the performance of the board was evaluated in a full-size laboratory with a constant gas generation, the board suppressed the increase of indoor formaldehyde concentration and the time course fitted to a theoretical curve. Then, field tests of the air-cleaning board were conducted in newly constructed multi-family houses in Japan. The board not only reduced

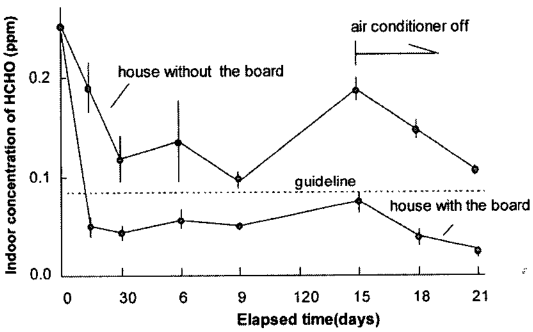

Fig. 3. Variation of indoor concentrations of formaldehyde (HCHO) normalized at 20°C in apartment houses with 9.8 m^2 of air-cleaning board (L/V=0.043 m^2/m^3, k = 1.1/h) and without the board (reference). Air conditioners were operated to prevent dew condensation for the first five months.

indoor formaldehyde concentration from 0.21 ppm to 0.04 ppm for more than 7 months (Fig. 3), but also enhanced the loss of formaldehyde gas from building materials in the houses.

A honey-comb type filter was also prepared and applied to an air cleaner for the reduction of concentrations of formaldehyde in indoor air of school (Sekine, 2002b). When two of the air cleaners, whose space velocity was set at 57,500(/h), were operated in the classroom (for IT education use, volume = 416 m^3, air change rate = 0.95/h), the concentration significantly decreased with time, as was predicted by a mass balance model introducing pre-evaluated equivalent ventilation rate of the device. These results led us the conclusion that certain air cleaning materials and devices, when properly evaluated and applied, prove beneficial to practical use.

Passive Samplers

The use of passive sampling method has greatly increased in recent years to monitor concentrations of indoor air pollutants, because the method has been recognized as an efficient alternative to pumped sampling according to its ubiquitous, cost-effective and user-friendly properties (Brown, 2000). Passive samplers, which employ diffusion process based on Fick's law and hence do not require power supply or other services, are suitable for monitoring personal exposure or indoor concentrations of gaseous components at ppb level. Then, authors have developed new passive samplers consisting of a porous plastic tube uniformly packed with powder-like adsorbent as a trapping media with greater sampling rates. For example, Fig. 4 illustrates construction of the passive sampler for carbonyl compounds employing 2,4-dinitrophenylhydrazine (DNPH) coated silica gel. The porous tube is made of sintered polyethylene particles with 34.5% of porosity and work as a diffuser which limits influence of wind or turbulence on the diffusive movement of gases; sampling rate of such type of samplers does not vary with the wind speed between 0.2 to 4.0 m/s (Sekine, 2001b).

When measuring concentrations of VOCs such as benzene, toluene, xylene, ethylbenzene, styrene, *p*-dichlorobenzene and limonene, carbon molecular sieves were used as collection media for solvent extraction and thermal desorption of analytes (Sekine *et al.*, 2002c, 2004). The VOCs were then determined by GC/FID or GC/MS. Authors has been currently developing a new sampler, which employs *o*-(4-cyano-2-ethoxybenzyl) hydroxylamine phosphate (CNET) as a reactive adsorbent of carbonyl compounds. The CNET is stable in storage and less hazardous than the previous DNPH.

Using such passive samplers, sampling rate, α is a dominant factor for analytical liability. As shown in Eq. (1), collected amount of analyte on adsorbent, W could be converted to air concentration, C using exposure time, t and α, if the adsorbent reduces the concentration of the given analyte at the end of diffusion layer ideally to zero due to sorption or chemical reaction (Brown, 2000).

$$C = W/(\alpha/t) \qquad (1)$$

Therefore, the sampling rate was determined in advance for each air pollutant by a small chamber experiment, deriving them from a slope of linear regressions between air concentrations in the chamber and collected amount of analyte at a certain exposure time. Table 3 shows typical performance of the passive sampler.

The passive samplers were applied for filed measurements at residential houses in Japan, and successfully used for determination of carbonyl compounds and VOCs at ppb level and gave similar results to active samplings. The results show excellent

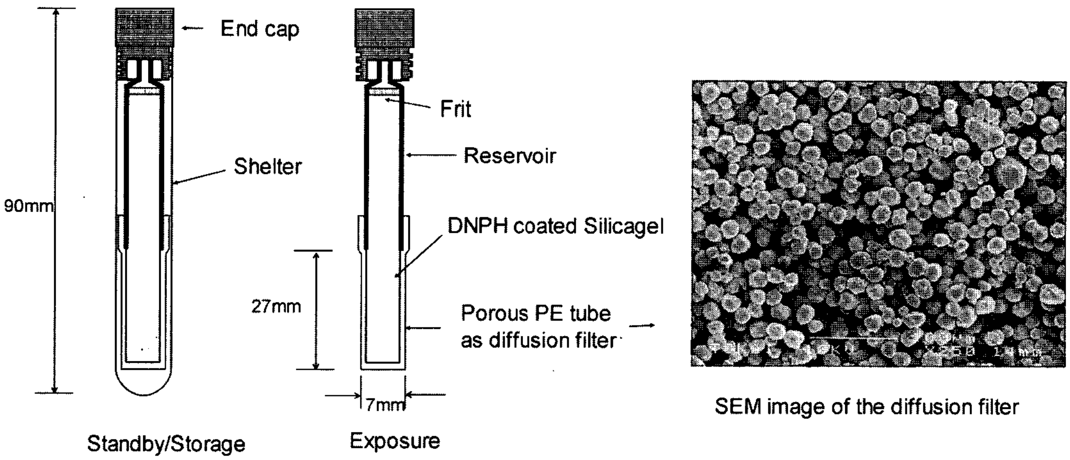

Fig. 4. Construction of passive sampler for carbonyl compounds.

Table 3. Performance of the passive samplers employing porous PE tube as diffuser for determinations of formaldehyde and toluene in indoor air

Analyte sampler	Formaldehyde		Toluene	
	DSD-DNPH	CNET-P	VOC-SD	VOC-TD
Company	Supleco	SCAS*	Supleco	Supleco
Collection media	DNPH/silicagel	CNET/silicagel	Carbon molecular sieves	
Analyte eluted by,	Acetonitrile	Acetonitrile	Carbon disulfide	Thermal desorption
Analyzed by,	UV/HPLC	UV/HPLC	GC/FID, MS	GC/FID, MS
Sampling duration	8-24 h	8-24 h	24 h	2 h
Sampling rate				
□ in m*l*/min	89	74	42	49
$\square_{w}$ in µg/(mg/m^3)/h	5.4	4.4	2.5	2.9
$\square_{v}$ in µg/ppm/h	6.7	5.4	9.7	11
Detection lomit (S/N=3)	0.31 ppb(24 h)**	0.12 ppb(24 h)	7.1 ppb	0.022 ppb

*Sumika Chemical Analysis Service,**3$\square_{b}$

linearity of the technique and suggest that reasonable accuracy can be expected after establishing the sampling rate under given exposure conditions.

Indoor Air Chemistry

In modern living environments, we are exposed numerous types of pollutants in indoor air. However, secondary emission products generated through chemical reactions have not been fully concerned in relation to the Sick House Syndrome. Authors focused on formic acid (HCOOH) suspected to have impact on human health such as chemical sensitivity due to its low irritant level. Our previous study performed in multi-family houses suggested that a room temperature and photochemical oxidants, which ordinarily came from outdoor air by ventilation, had potential influences on the formation of indoor formic acid from formaldehyde. Furthermore, laboratory studies using small and large chambers showed that a heterogeneous reaction between formaldehyde and nitrate radical on the surface of plywood played an important role of the indoor formic acid production. These findings suggest scientists should continue to explore and identify the hazardous substances in indoor environment.

Concluding Remarks

Air pollution is one of the typical worries for human well-being and yet we are unable to see the pollution, until the appearance of severe impacts on human health, such as the Sick House Syndrome. Author believes air quality watching with a strong scientific basis and collaboration of stakeholders plays an important role in enhancing a freedom of individuals from fear of environmental risks and leads to useful scientific inputs in political decision-making for environmental protection.

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