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Control of emission rates of chemical compounds emitted by controlling their mass transfer coefficients on the surface of the tested building material

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The air change rate in the chamber, the loading factor of the materials, and the mass transfer coefficient are very important factors in the measurement of chemical compounds, because they have a decisive effect on emission rates of chemical compounds emitted from materials. Small 20-liter chambers, such as the advanced pollution and air quality chamber, are generally used in Korea and Japan for measuring the amount of released chemicals. In this study, chemical compounds released from building materials and adhesives were measured using a chamber proposed by the authors to control the mass transfer coefficient on the surface of the tested building material and we examined the distribution of chemical compounds concentrations in the chamber by means of computational fluid dynamics to confirm test reliability. The chamber was controlled and maintained at 28 °C, a relative humidity of 50%, a mass transfer coefficient of 14 m/h with an air change rate of 0.50 h⁻¹, and formaldehyde and total volatile organic compounds were emitted from the flooring material and adhesive. As the mass transfer coefficient on the surface of the tested building material increased, the emission rates of chemical compounds measured using the proposed chamber increased. The mass transfer coefficient on the surface of the tested building material significantly influenced the emission rates of the chemical compounds released from the building material and adhesive.

Keywords: adhesive; flooring materials; emission test; mass transfer coefficient

1. Introduction

For measuring emission rates of chemical compounds released from building materials, many countries, including Korea, have instituted laws or specifications for measurement methods applicable to their own country. For measuring emission rates of chemical compounds released from building materials, small chambers are generally used, considering the advantages of the ease in carrying out experiments, low expenses, etc. Meanwhile, small 20-liter chambers are also generally used for measuring emission rates of released chemical compounds in Korea and Japan [1,2].

The water vapor mass transfer coefficient can be related to the convective heat transfer coefficient through Lewis, relation. The mass transfer coefficients of water vapor and volatile organic compounds (VOCs) are generally almost identical. Indoor convective heat transfer

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coefficients are generally $3\sim 6 \text{ W m}^2/\text{K}$. This value of indoor convective heat transfer coefficient is converted to a water vapor mass transfer coefficient of $9\sim 18 \text{ m/h}$ [3,4]. The aforementioned small chambers maintain a uniform mass transfer coefficient for water vapor diffusion on the surface of tested building materials at approximately $2\sim 3 \text{ m/h}$, which is considerably below the mass transfer coefficient of $9\sim 18 \text{ m/h}$ recommended in the Japanese industrial standards (JIS) A 1901. This means that mass transfer coefficient for chemical compounds in the small chamber differs from that inside an actual architectural space [5,6].

In order to settle the aforementioned problems, in this study we developed an airflow control unit for a small chamber for uniformly controlling airflow on the surface of a tested building material in a small chamber. The mass transfer coefficients on the surface of the tested building materials are presented, corresponding to those on an actual building, in order to exactly evaluate the concentration of released chemical compounds using the small chambers. We measured the airflow velocity on the surface of the building materials in the small chamber with the developed airflow control unit and the fan speed for airflow control. The distribution of water vapor concentration in the small chamber by means of computational fluid dynamics (CFD) was examined and the mass transfer coefficient on the surface of the building material was estimated to confirm test reliability [7]. Pollutants released from the adhesive and the wallpaper attached with the adhesive were measured under controlled a mass transfer coefficient on the surface of the tested building material, to examine the effect of mass transfer coefficient on the rate of pollutant release.

2. Configuration of the airflow control unit and the small chamber

In order to avoid adsorption and decomposition of chemical compounds, the airflow control unit was made of SUS 304 stainless steel and a fan made of Teflon was used as shown in Figure 1.

The fan for generating uniform airflow in the airflow control unit was installed on the bottom of the chamber in order to minimize the influence of air flow generated by the air supply and exhaust of the chamber as shown in Figure 2. Mechanical ventilation was performed in the chamber and was controlled to maintain a uniform wind velocity on the surface of the tested

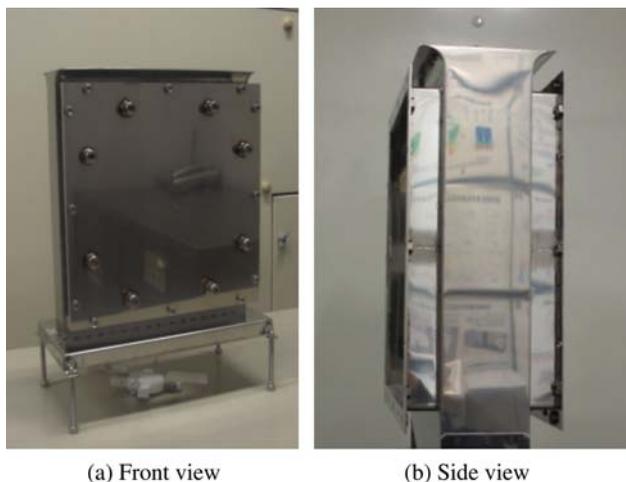


Figure 1. Airflow control unit.

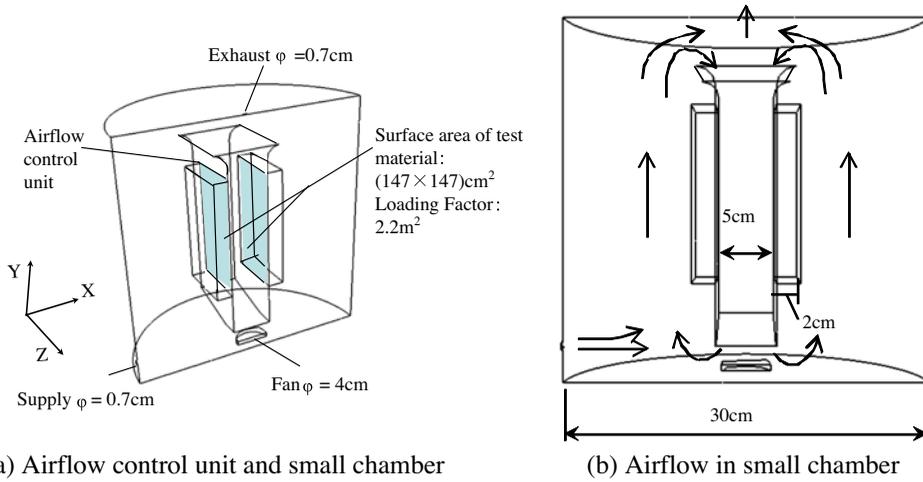


Figure 2. Simplified views of airflow control unit and small chamber.

building material. To avoid pollutants being generated by the fan in the chamber, the fan was rotated by means of a magnetic force. The area for installing the tested building material in the airflow control unit was 0.043 m^2 , which corresponds to product loading factor of $2.2 \text{ m}^2/\text{m}^3$.

3. Method

3.1. Measurement of mass transfer coefficient

Several sheets of filter paper wetted with distilled water were attached on to the surface of the building material as shown in Figure 2. Subsequently, dry air at a constant temperature was supplied to the chamber to measure the mass transfer coefficient when the temperature and the (RH) reached the steady state. The temperatures at the air supply inlet and the outlet of the chamber, and the RH and the temperature on the surface of the filter paper were continuously measured. In this study, the mass transfer coefficient was measured in the airflow control unit from the amount of evaporating vapor from the filter paper per hour. A T-type thermocouple was attached in three places at the center of the filter paper to measure its surface temperature. The air change rate in the chamber was set to 0.50 h^{-1} , and the temperature was maintained at 28°C . The measured temperature and RH were used to calculate absolute humidity and the mass transfer coefficient [8].

The example to analyze the airflow field on the basis of average wind velocity (U_{center}) in the center of the airflow control unit and the wind velocity of the fan (U_{fan}) is shown in Table 1.

Table 1. Cases of numerical analysis (Temperature: 28°C).

| Case | $U_{\text{center}} (U_{\text{fan}})$ (m/s) | Reynolds number ($U_0 L_0 / \nu$) | Air change rate (h^{-1}) |
|------|--|-------------------------------------|-------------------------------------|
| 1 | 8.27×10^{-4} (0.01) | 1 | 0.50 |
| 2 | 3.86×10^{-3} (0.05) | 10 | |
| 3 | 9.31×10^{-2} (1) | 160 | |
| 4 | 2.07×10^{-1} (2) | 350 | |
| 5 | 3.57×10^{-1} (4) | 600 | |

The temperature and the air change rate in the small chamber were set to 28 °C and 0.50 h⁻¹, respectively. With respect to the average wind velocity U_{center} in the airflow control unit, the airflow field was analyzed for five cases from 8.27×10^{-4} m/s ($U_{\text{fan}} = 0.01$ m/s) to 3.57×10^{-1} m/s ($U_{\text{fan}} = 4$ m/s). For each case, the Reynolds number ($= U_0 L_0 / \nu$) was 1, 10, 160, 350, and 600. U_0 and L_0 are the average wind velocity in the airflow control unit and half of the internal height of the airflow control unit, respectively. ν means coefficient of kinematic viscosity.

A three-dimensional analysis of the flow field was performed based on a low Reynolds number type k - ϵ model [9]. Numerical analysis was carried out only for half of the space as shown in Figure 2(a). The boundary conditions for the CFD analysis are shown in Table 2.

Following the analysis of the airflow field, the boundary condition for emission was set at the surface where the building material was placed and then the diffusion was analyzed [10]. The surface of the building material was modeled with a single-component liquid. The model of the building material consisted of water (distilled water) and water evaporation from the liquid surface to the air, i.e., the mass transfer coefficient on the surface of the building material was measured. The diffusion was analyzed assuming an isothermal state (28 °C). The transportation of water (vapor) is expressed and analyzed by Eq (1). The water (vapor) diffusion coefficient D_a in the air calculated from Eq (2) to (4) is 2.30×10^{-5} m²/s [11–13]:

$$\frac{\partial \bar{C}_1}{\partial t} + \frac{\partial \bar{U}_j \bar{C}_1}{\partial x_j} = \frac{\partial}{\partial x_j} \left(\left(D_a + \frac{v_t}{\sigma_t} \right) \frac{\partial \bar{C}_1}{\partial x_j} \right) \quad (1)$$

$$\log_{10} P_w = \frac{A - B}{(C + T)} - 3 \quad (2)$$

$$C_0 = \rho_a \frac{M_1}{M_2} \frac{P_w}{P - P_w} \quad (3)$$

$$D_a = \frac{6.7 \times 10^{-8} \times T^{1.83}}{P} \times \left[\left(\frac{T_{c1}}{P_{c1}} \right)^{\frac{1}{3}} + \left(\frac{T_{c2}}{P_{c2}} \right)^{\frac{1}{3}} \right]^{-3} \sqrt{\frac{1}{M_1} + \frac{1}{M_2}} \quad (4)$$

where,

- C_1 : vapor phase concentration ($\mu\text{g}/\text{m}^3$)
- D_a : molecular diffusion coefficient (m^2/s)

Table 2. Conditions of numerical analysis.

| Turbulent flow model | Low Reynolds number, k - ϵ type model |
|----------------------|--|
| Number of meshes | 270,690 |
| Scheme | Space difference: Second-order upwind |
| Inlet | $U_{x,\text{in}} = 7.2 \times 10^{-2}$ m/s, $U_{y,\text{in}} = 0$, $U_{z,\text{in}} = 0$ $k_{\text{in}} = 3/2$ $(U_{\text{in}} \times 0.05)^2$, $\epsilon_{\text{in}} = C_{\mu} \cdot k_{\text{in}}^{3/2} / L_{\text{in}}$ $L_{\text{in}} = 1/7 L_0$, $L_0 = 7.0 \times 10^{-4}$ m |
| Fan | $U_{\text{fan}} = 0.01, 0.05, 1, 2, 4$ (m/s) |
| Wall | No-slip |

- U_j : wind velocity (m/s)
 ν_i : eddy viscosity (m^2/s)
 σ_t : turbulent Schmidt's number (-)
 P_w : water vapor pressure (Pa)
 A, B, C : empirical constants (A : 7.74, B : 1554.16, C : 219)
 T : temperature ($^{\circ}\text{C}$)
 C_0 : saturation concentration (g/m^3)
 ρ_a : air density (g/m^3)
 M_1, M_2 : molecular weight
 P : atmospheric pressure in the chamber (Pa)
 T_{c1}, T_{c2} : critical temperatures ($^{\circ}\text{C}$)
 P_{c1}, P_{c2} : critical pressures (Pa)

3.2. Measurement of chemical compounds

Two sealed boxes made of stainless steel were used to prevent the cut edge effect, which allows only chemical emission from surface of the test piece, and the surface area of the tested building material was $(0.147 \times 0.147) \text{ m}^2$. The adhesive applied to the plywood and the wallpaper applied with adhesive, which are used as building interior materials, were selected in this study. The amount of adhesive applied for the wallpaper used on the plywood was approximately $310 \text{ g}/\text{m}^2$. The wallpaper was made of materials including poly (vinyl chloride) resin, plasticizers, filling agents, pigments, etc., and its weight was approximately $190 \text{ g}/\text{m}^2$. Air was captured at the outlet of the test chamber with a DNPH cartridge (Waters) and a Tenax-TA tube (60/80 mesh, Gerstel Inc., USA), and analyzed for aldehydes and VOCs, using High Performance Liquid Chromatography and Gas Chromatography/Mass Spectrometry Thermal Desorption System. Measurement of the release rate of pollutants was carried out at 28°C , 50% RH and air change rate of 0.50 h^{-1} . The concentration of pollutants was measured at 1, 3, 7, 14, and 28 days after initiating the test. The Reynolds number was controlled at 10 and 160 to examine the effect of mass transfer coefficient on the release rate of pollutants.

4. Results and discussion

4.1. Mass transfer coefficient predicted by CFD analysis

The mass transfer coefficient was measured from the absolute humidities at the air supply inlet and the outlet of the small chamber and the amount of evaporating distilled water from the wet filter paper. Table 3 shows the results of four measurements. The temperature and RH were $28.0 \sim 28.2^{\circ}\text{C}$ and 12%, respectively, at the air supply inlet of the chamber and were $28.3 \sim 28.5^{\circ}\text{C}$ and 95~96%, respectively, at its outlet. The amount of evaporating moisture

Table 3. Experimental results on mass transfer coefficients.

| Measurement No. | Supply | | Exhaust | | Surface Temperature of filter paper ($^{\circ}\text{C}$) | Amount of evaporating vapor (g) | Mass transfer coefficient (m/h) |
|-----------------|------------------------------------|-----------------------|------------------------------------|-----------------------|--|---------------------------------|---------------------------------|
| | Temperature ($^{\circ}\text{C}$) | Relative humidity (%) | Temperature ($^{\circ}\text{C}$) | Relative humidity (%) | | | |
| 1 | 28.2 | 12 | 28.4 | 96 | 28.2 | 0.24 | 11.5 |
| 2 | 28.2 | 12 | 28.5 | 95 | 28.1 | 0.23 | 12.1 |
| 3 | 28.1 | 12 | 28.4 | 96 | 28.1 | 0.23 | 12.8 |
| 4 | 28.0 | 12 | 28.3 | 96 | 28.1 | 0.23 | 11.6 |

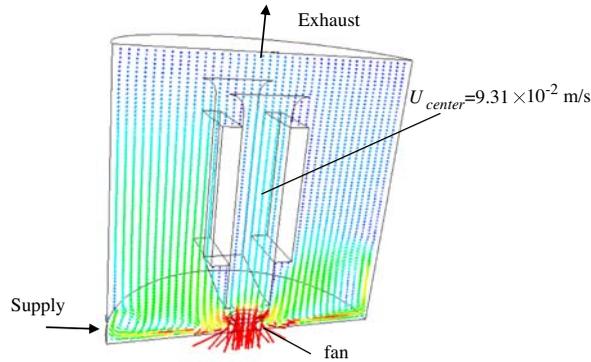


Figure 3. Velocity vector in the chamber (Case 3).

was 0.23~0.24 g which did not differ much in the four measurements. When the air velocity was controlled at 1 m/s, the air velocity on the surface of the building material tested in the airflow control unit was measured as approximately 0.1 m/s. The measured water vapor mass transfer coefficient was 11.5~12.8 m/h. This meets the range of 9~18 m/h for the mass transfer coefficient recommended in JIS A 1901 and corresponds to the mass transfer coefficient in a general building space. The Schmidt number and the Reynolds number were 12.5~12.6 and 134~167, respectively.

Figure 3 shows the average velocity vector (case 3) of airflow in the airflow control unit and the small chamber. The supplied air is mixed with the air in the small chamber immediately before reaching the airflow control unit. It is found that the mixed air flows uniformly from the upper part to the lower part of the airflow control unit.

4.2. Experimentally measured mass transfer coefficients

Table 4 shows the predicted average emission rates of water and the average mass transfer coefficients. Figure 4 shows the correlation among Reynolds number, emission rates of water, and the mass transfer coefficient for water vapor.

For case 1 where the Reynolds number is 1, the emission rate of water is 3.29 g/m²h. As the Reynolds number increases to 10 and 160, the emission rate of water increases to 4.07 g/m²h (case 2) and 4.49 g/m²h (case 3). Meanwhile, in cases 4 and 5, although the Reynolds number increased as in Figure 4, the emission rate of water did not increase and was maintained almost at a constant value. The mass transfer coefficients of case 3, where the Reynolds number is 160, and case 4, where the Reynolds number is 350, were 14.43 m/h and 18.07 m/h, respectively. There is no significant difference between the mass transfer coeffi-

Table 4. Predicted average emission rates and average mass transfer coefficients.

| Case | U_{center} (m/s) | Reynolds number ($U_0 L_0 / \nu$) | C_{out}/C_s | C_s (g/m ³) | Average emission rate (g/m ² h) | Average mass transfer coefficient (m/h) |
|------|-----------------------|-------------------------------------|---------------|---------------------------|--|---|
| 1 | 8.27×10^{-4} | 1 | 0.72 | 19.9 | 3.29 | 0.61 |
| 2 | 3.86×10^{-3} | 10 | 0.89 | | 4.07 | 1.89 |
| 3 | 9.31×10^{-2} | 160 | 0.98 | | 4.49 | 14.43 |
| 4 | 2.07×10^{-1} | 350 | 0.99 | | 4.52 | 18.07 |
| 5 | 3.57×10^{-1} | 600 | 0.99 | | 4.53 | 21.55 |

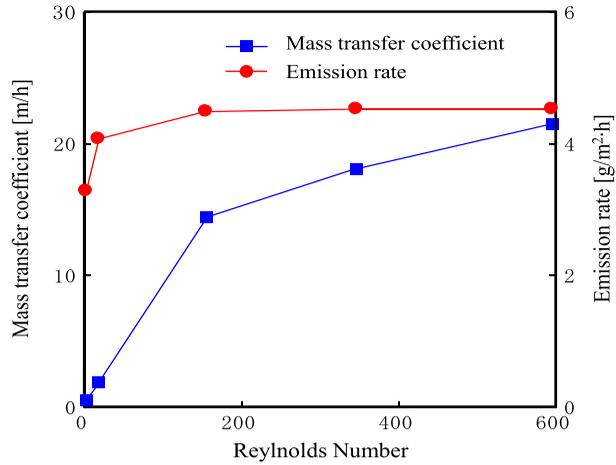


Figure 4. Correlation among Reynolds number, emission rate of water, and mass transfer coefficient for water vapor.

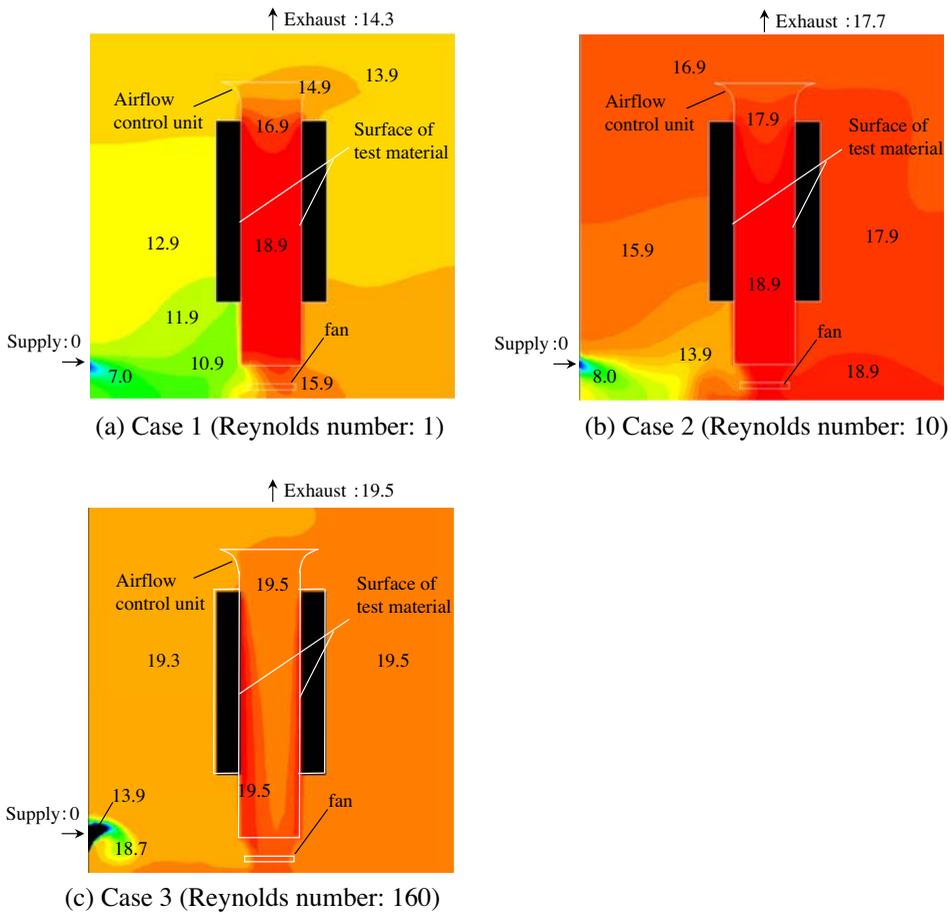


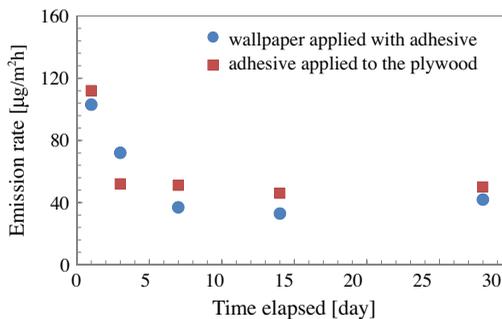
Figure 5. Distribution of water concentration in the chamber (g/m³).

cient of case 3 analyzed under the experimental conditions and that measured in the experiment. For case 5, where the Reynolds number is 600, the mass transfer coefficient was predicted to be 21.55 m/h, which is slightly above the specification of JIS.

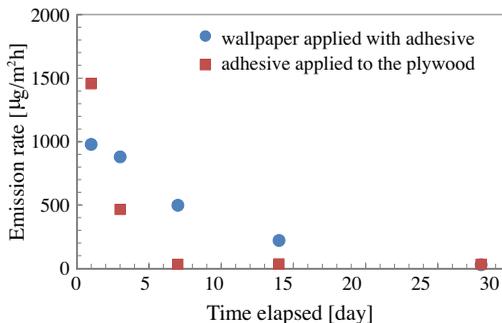
Table 4 and Figure 5 show the water concentration distribution in the chamber. The value of concentration was normalized by C_s , which is the water concentration on the surface of the building material. In case 3, it can be seen that the water concentration in the chamber reached almost saturation and it was almost the same as the measurement result found in the experiment. In cases 1 and 2, the water concentration in the airflow control unit exhibited uniform distribution, but a slight difference from the concentration distribution in the chamber outside of the airflow control unit was observed.

4.3. Chemical compounds emitted from adhesive and wallpaper

Figures 6(a) and (b) show the release rates for formaldehyde and (TVOCs) from adhesive and wallpaper when the Reynolds number was controlled to be 10. The formaldehyde released from the adhesive applied to plywood showed decreasing rates for 7 days after initiating the test as shown in Figure 6(a). The release rate of formaldehyde from the plywood to which wallpaper was attached stayed constant over time, but was somewhat lower compared to the rate for the adhesive applied to the plywood. The release rates of TVOCs significantly differed depending on the presence of wallpaper as shown in Figure 6(b). The rate of TVOCs released from the adhesive stayed constant at approximately $30 \mu\text{g}/\text{m}^2\text{h}$ for 7 days after initiating the test.

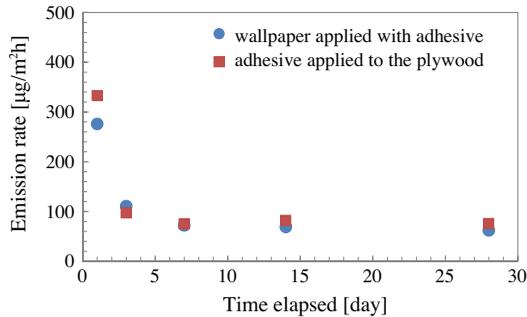


(a) Emission rates for formaldehyde

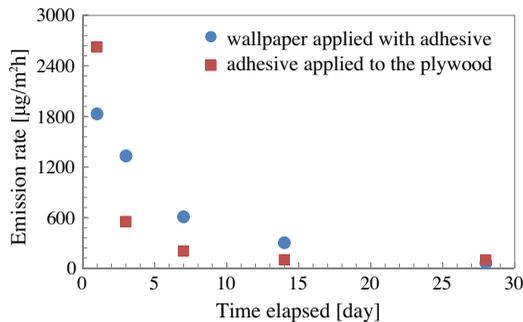


(b) Emission rates for TVOCs

Figure 6. Chemical compounds emitted from the adhesive and the wallpaper applied with adhesive at Reynolds number of 10.



(a) Emission rates for formaldehyde



(b) Emission rates for TVOCs

Figure 7. Chemical compounds emitted from the adhesive and the wallpaper applied with adhesive at Reynolds number of 160.

The release rates of formaldehyde and TVOCs are shown in Figures 7(a) and (b) when the Reynolds number is controlled to be 160 and the mass transfer coefficient is set to approximately 14.43 m/h. Compared to Figure 6, as the Reynolds number increases, the increasing mass transfer coefficient contributes to significantly increasing the release rate of formaldehyde and TVOCs. For the case of the increasing Reynolds number, it is predicted that formaldehyde significantly diffuses into the wallpaper due to the increasing mass transfer coefficient. Accordingly, the release rate of formaldehyde was found to be constant in the presence of wallpaper as shown in Figure 7(a).

On the next day after initiating the test, the release rate of TVOCs from the adhesive exceeded $2,500 \mu\text{g}/\text{m}^2\text{h}$, but showed tendency toward sharply decreasing rate below $600 \mu\text{g}/\text{m}^2\text{h}$ after a lapse of three days. Compared to the case in which the Reynolds number was 10, the rate of TVOCs release was higher as the Reynolds number increased, but the TVOCs emission did not depend significantly on the Reynolds number.

5. Summary and conclusions

In this study, we developed an airflow control unit for small chambers for controlling the mass transfer coefficient at a constant value on the surface of the building material tested in a small chamber. We used the mass transfer coefficient on the surface of the tested building material corresponding to an actual architectural space for which emission rates of the released chemicals were measured.

Pollutants released from the adhesive applied to the plywood and the wallpaper applied with adhesive were measured, using the chamber and the airflow control unit proposed in this study and we examined the effect of mass transfer coefficient on the release rate of pollutants.

When installing the airflow control unit in the small chamber to control the fan speed (U_{fan}) at 1 m/s ($U_{\text{center}}=9.31 \times 10^{-2}$ m/s, Reynolds number 160), the mass transfer coefficient of water vapor was found to be approximately 12 m/h from the experiments, and 14.43 m/h from the numerical analysis. Therefore, it is considered that the values are very similar to the mass transfer coefficient in an actual architectural space, and it is expected that the application of the airflow control unit and the airflow velocity proposed in this study will contribute to achieving an even higher level of accuracy in measuring emission rates of released chemical compounds. When using the airflow control unit in the small chamber, it was seen that the velocity of the released compounds and the mass transfer coefficient highly depended on the airflow velocity by the fan, i.e., the Reynolds number. Increasing the Reynolds number, due to increase of airflow velocity by the fan, resulted in smaller concentration difference of the compounds on the surface of the building material and in the air.

The release rates of formaldehyde and TVOCs from adhesive were significantly affected by the mass transfer coefficient and the release rates increased with increasing Reynolds number. The release rates of pollutants from the adhesive were the highest at 1 – 3 days after initiating the test and decreased significantly after 3 days. It was observed that formaldehyde and TVOCs were released from the plywood to which wallpaper was attached with adhesive continuously over time, but the wallpaper contributed to inhibiting pollutant release.

With the chamber and the airflow control unit proposed in this study, it is possible to accurately control the mass transfer coefficient, and thus to accurately evaluate the rates of pollutants released from all types of building materials in the simulated conditions of an actual indoor space.

Acknowledgements

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