



Mass transfer of volatile organic compounds from painting material in a standard field and laboratory emission cell

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Received 21 April 2002; received in revised form 8 November 2002

Abstract

The field and laboratory emission cell (FLEC) is becoming a standard method of characterizing pollutant emissions from building materials. It is significant to use the emission profiles from FLEC to scale the emissions of building materials in real buildings. The dynamics of mass transfer in such an FLEC are the key to perform this task. In this study, the mass transfer mechanisms of the total volatile organic compounds from a wet painting in an FLEC are experimentally and numerically investigated. A three-dimensional mass transfer model, which takes into account the convective mass transfer between the material and the air, the diffusion in the paint film and in the substrate, is developed. The emissions from a water-based emulsion paint are quantified to assess the model. The concentration fields in the film and substrate are calculated to demonstrate the processes of internal volatile organic compounds diffusion. © 2003 Elsevier Science Ltd. All rights reserved.

Keywords: Indoor air quality; Field and laboratory emission cell (FLEC); Mass transfer; VOCs

1. Introduction

Volatile organic compounds (VOCs) constitute an important class of indoor air contaminants. Evidence from a variety of non-industrial building investigations and systematic studies have found that 60% of indoor VOCs come from building materials and furnishings [1]. Various VOCs have been associated with certain symptoms of sick building syndrome, multiple chemical sensitivity, and other health effects. In addition, excessive exposure to VOCs in the work environment can lower people's productivity and cause material and equipment damage.

The impact of increased consciousness about indoor environment has created a demand for low-emitting (healthy) building materials. This has prompted several activities for standardized methods to characterize and

quantify the emissions from building materials and consumer products as determination of VOCs in finished products. Further there is a need for a method for easy source identification of VOCs from potential emitting building materials on site and a quantification of them. From these reasons, the field and laboratory emission cell (FLEC) has been proposed and has becoming an European standard for emission testing [2]. This is a kind of micro emission cells which have high sensitivity due to the large loading ratio (surface area/volume). Nowadays, a large percentage of emission tests for various materials were performed with FLEC [3–5].

Mass transfer in a standard FLEC has been investigated by several authors. Uhde et al. [6] were the first who measured the velocity distributions in a FLEC. They found that a surface area of very low air velocities exists within a radius of 20 mm. Later, Murakami et al. [7] conducted a three-dimension analysis of diffusion and emissions of VOCs in a FLEC cavity. They found that in the case of the internal diffusion material, the local VOC emission rate becomes uniform, while in the case of evaporation type materials, emission rates vary greatly

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Nomenclature

C_a	concentration of TVOCs in air flow (kg/m^3)	R	ideal gas constant ($\text{kJ kg}^{-1} \text{K}^{-1}$)
C_m	concentration of TVOCs in material film (kg/m^3)	r	radial coordinates (m)
C_{ma}	TVOCs concentration at the material/air interface (kg/m^3)	r_0	radius of paint film (m)
C_s	concentration of TVOCs in substrate (kg/m^3)	Re	Reynolds number
D_0	constant (m^2/s)	Sc	Schmidt number
D_a	diffusivity of TVOCs in air (m^2/s)	T	temperature (K)
d_e	hydrodynamic diameter of the flow channel (m)	u_a	air velocities in angle direction (m/s)
D_m	diffusivity of TVOCs in material film (m^2/s)	u_m	mean air velocity at the slit (m/s)
D_s	diffusivity of TVOCs in the substrate (m^2/s)	v_a	air velocities in radial direction (m/s)
E_d	activation energy (kJ kmole^{-1})	z	axial coordinates (m)
h_m	local convective mass transfer coefficients between the material surface and air (m/s)	<i>Greek symbols</i>	
K_{ma}	material–air partition coefficient for paint	ν	kinematic viscosity of air (m^2/s)
K_{mas}	partition coefficient for substrate	α	liquid expansion factor
K_{sa}	partition coefficient between substrate and air	τ	time (s)
		θ	angle
		δ_a	height of air slit (m)
		δ_m	thickness of paint film (m)
		<i>Superscript</i>	
		*	dimensionless

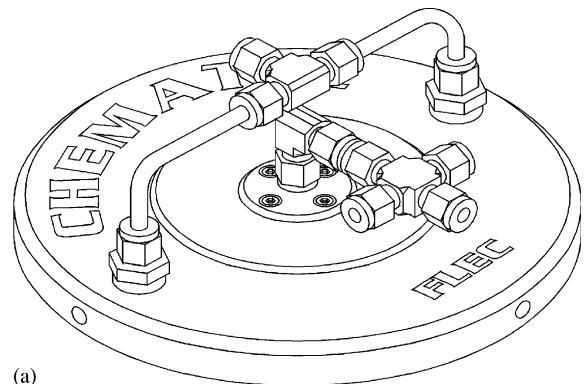
along the surface. Further, Sjöberg [8] calculated the resistance to transmission of *n*-butanol and 2-ethylhexanol through two floorings, PVC and Linoleum, with a modified bottom part for the FLEC. Recently, Zhang and Niu [9] conducted a complete CFD modeling of the fluid flow and surface mass transfer in the FLEC. The effects of fluid inlets and outlets were considered. Experiments with water emission confirmed their predictions of convective surface mass transfer coefficients in the FLEC. These investigations provided the fundamentals of VOCs emissions in such a system.

This study is a step forward from previous researches. Convection with air stream, internal diffusion in material, and diffusion in substrate, are taken into account simultaneously. In real buildings, influences of mass diffusion in the material film, as well as in the substrate may become substantial.

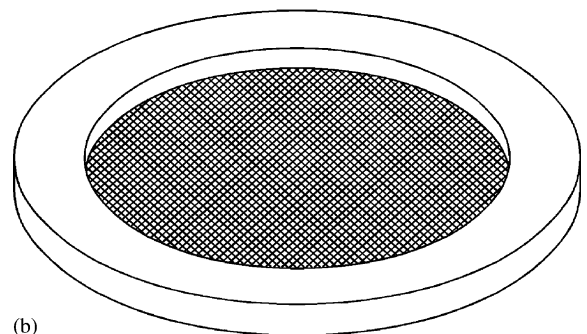
2. Mathematical model

2.1. Introduction of the FLEC

The FLEC has become a standard equipment for emission testing. It is composed of two parts: cap (Fig. 1(a)) and lower chamber (Fig. 1(b)). The flow geometry of the FLEC is shown in Fig. 2. When testing, the planar specimen of the emission material is placed in the lower chamber and becomes an integral part of the emission cell. The upper surface of the specimen (the emission



(a)



(b)

Fig. 1. Pictures of the FLEC, showing the cap (a) and lower chamber (b).

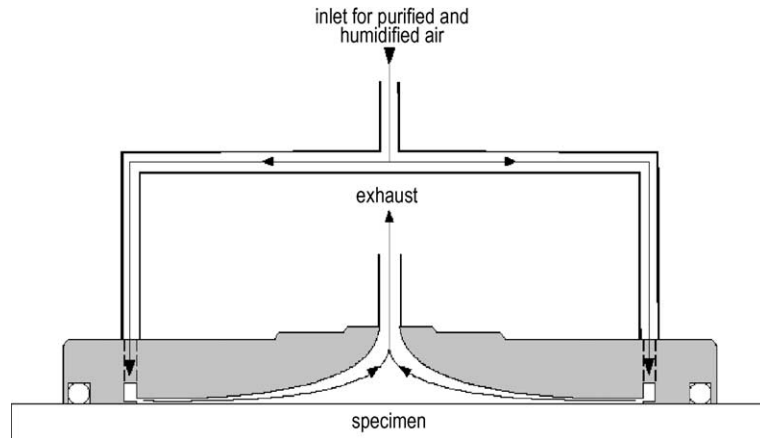


Fig. 2. Schematic diagram showing the flow geometry of the FLEC.

surface) and the inner surface of the FLEC cap form a cone-shaped cavity. The air is supplied through the air slits in the cap. It is introduced through two diametrically positioned inlets (symmetrically placed) into a circular-shaped channel at the perimeter, from where the air is distributed over the emission surface through the circular air slit. The air flows radially inward, until it exits the FLEC outlet in the center.

2.2. Model setup

The model will focus on the emissions of wet paintings in a FLEC, since wet painting material represents a major source of indoor air pollution. Consumers regularly apply paints. It has been found that 2–9% of the households used paints during the week that VOCs were monitored in their homes [10]. These paints usually contain more than 25% of the solvent that will be released into air during the drying period.

Much progress has been made in developing mass transfer models for the evaporation-controlled process. The earliest semi-physical models to predict emission rates from materials were proposed by Black and Bayer [11]. In the development of these models, it was assumed that the internal resistance of materials to mass diffusion was negligibly small. This assumption restricts the application of the model, where the transport of vapor through a board-air interphase region is a rate limiting process. Dunn [12] proposed a model for the prediction of emissions from a thin film. The main assumption was that the emission is merely a surface phenomenon. The model considers sink effects, however, the assumption of thin film restricts its application to thick materials where diffusion is a controlling factor. Later, Tichenor et al. [13] developed an evaporative mass transfer model for total volatile organic compounds (TVOCs) emission from interior architectural coatings. This model is called the vapor pressure and boundary layer (VB) model, and

it considers the emission as a pure evaporation process and neglects internal diffusion. Spark et al. [14] discussed the application of gas-phase mass transfer models for indoor pollutant sources. Guo [15] further developed a mass transfer model for predicting the emission rates of individual VOCs from petroleum-based indoor coatings (VBX model). It has been found that such a model does not work for the entire emission process, especially when the material becomes relatively dry. Yang et al. [16,17] proposed a model that considers VOC mass transfer in the air and material–air interface, diffusion in the material film, and also diffusion in the substrate. The model presented here is similar to that endorsed by Yang et al. However, modifications are made to take into account the special flow geometry of the cavity and the corresponding convective surface mass transfer coefficients. Furthermore, VOCs profiles and transfers in the substrate could be investigated. Also with this model, the concentration fields in the paint and in the substrate could be calculated to explore the mechanisms of VOCs emissions.

To ease the model setup, several assumptions are made as following:

- (1) Fick's law applies to mass transfer in both material film and substrate.
- (2) The material is homogeneous and the VOCs diffusion coefficients in the material and the material–air partition coefficients are constant. TVOCs is used to represent the lumped parameter of various VOCs quantified.
- (3) The mass transfer rate between the air and material is very small. Hence, the heat generation/release associated with the sorption is negligible.
- (4) Steady state stream flow over the surface.
- (5) The amount of coating material applied is small so that the material is quickly absorbed by the porous substrate after the application, and a wet layer with

a uniform initial VOCs concentration is formed inside the substrate.

- (6) During the emission process, VOCs in the material film and the VOCs vapor pressure are in thermodynamic equilibrium at the material–air interface. Similarly, VOCs in the substrate and film are in equilibrium at the material–substrate interface.

Transfers of VOCs in material are very complicated. During the emission process, several mechanisms of external and internal VOC mass transfer occur. These mechanisms include evaporation at the material surface, movement of free or bound VOCs, VOCs vapor diffusion in the material film, and VOCs diffusion from the material film to the substrate. In fact, a microscopic view of the entire diffusion processes will generate a phenomenon that is too complicated to be solved. To simplify the situation, the continuum model, which treats the material as a homogeneous medium and characterized by an effective diffusion coefficient, is always used. The benefits of this approach is that Fick's law can be used to predict the mass transfer in porous media.

2.2.1. Air flow

Air flow equation above the material is assumed to be two-dimensional and is governed by a mass conservation as

$$\frac{\partial C_a}{\partial \tau} + u_a \frac{\partial C_a}{r \partial \theta} + v_a \frac{\partial C_a}{\partial r} = D_a \frac{1}{r} \frac{\partial}{\partial \theta} \left(\frac{\partial C_a}{r \partial \theta} \right) + D_a \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_a}{\partial r} \right) + \frac{8r}{d_e^2} h_m (C_{ma} - C_a) \quad (1)$$

where C_a is the concentration of TVOCs in air flow (kg/m^3), τ is time (s); u_a and v_a are air velocities in angle and radial directions (m/s), respectively; θ , r , z are angle, radial, and axial coordinates, respectively; D_a is the diffusivity of TVOCs in air (m^2/s); d_e is the hydrodynamic diameter of the flow channel (m); h_m is the local convective mass transfer coefficients between the material surface and air (m/s); C_{ma} is the TVOCs concentration (kg/m^3) at the material/air interface.

2.2.2. Material film

The transient VOCs diffusion process in the material film can be expressed in a form analogous to Fick's law as

$$\frac{\partial C_m}{\partial \tau} = \frac{1}{r} \frac{\partial}{\partial \theta} \left(D_m \frac{\partial C_m}{r \partial \theta} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left(D_m r \frac{\partial C_m}{\partial r} \right) + \frac{\partial}{\partial z} \left(D_m \frac{\partial C_m}{\partial z} \right) \quad (2)$$

where C_m is the TVOCs concentration in paint material (kg/m^3), D_m is the diffusivity of TVOCs in material film

(m^2/s). It is a function of the pore structure, the material type, compound properties, temperature, and the VOCs concentration in the material film. For a given wet material–substrate–VOC system, the D_m is considered a thermally activated process and can be expressed by

$$D_m = D_{m,0} \left(\frac{C_m}{C_{m,0}} \right)^3 \quad (3)$$

$$D_{m,0} = D_0 \exp \left(- \frac{E_d}{RT} \right) \quad (4)$$

where D_0 is a constant (m^2/s), E_d is the activation energy (kJ kmole^{-1}), R is ideal gas constant ($\text{kJ kg}^{-1} \text{K}^{-1}$), T is temperature (K). Chen and Lin [18] suggested that $E_d = 30.5 \text{ kJ kmole}^{-1}$ for VOCs. The temperature for indoor applications only vary within a small range. Hence, it may be possible to estimate the E_d using the above results for reference data.

2.2.3. Substrate

By using the “continuum model” and Fick's law, the transient VOC diffusion process in the substrate may be described by the following equation

$$\frac{\partial C_s}{\partial \tau} = \frac{1}{r} \frac{\partial}{\partial \theta} \left(D_s \frac{\partial C_s}{r \partial \theta} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left(D_s r \frac{\partial C_s}{\partial r} \right) + \frac{\partial}{\partial z} \left(D_s \frac{\partial C_s}{\partial z} \right) \quad (5)$$

where C_s is the concentration of VOCs in substrate (kg/m^3); D_s is the diffusivity of VOCs in the substrate (m^2/s). The dependence of D_s on temperature is assumed to follow the Arrhenius equation as

$$D_s = D_{s,0} \exp \left(- \frac{E_d}{RT} \right) \quad (6)$$

where $D_{s,0}$ is a constant (m^2/s).

2.2.4. Material–air interface

At the material–air interface, the VOC changes phase from the material (liquid or solid) side to the air side. Henry's law states that at equilibrium, the ratio of the concentration of a volatile solute in a solvent, C_m to the solute's concentration in the vapor phase above the solvent–solute mixture, C_{ma} is a constant. Although Henry's law is valid only for low-solute concentrations, it can be applied to a wide range of gases and VOCs of environmental interest. Henry's law can be expressed as

$$C_m = K_{ma} C_{ma} \quad (7)$$

where K_{ma} is a constant, which frequently is called the dimensionless material–air partition coefficient. Assuming the K_{ma} at the reference temperature, T_{ref} , is $K_{ma} \times (T_{ref})$, the K_{ma} at a different temperature, T , will be given by the equation

$$K_{\text{ma}}(T) = K_{\text{ma}}(T_{\text{ref}}) \frac{T}{T_{\text{ref}}} 10^{0.2185A[(1/T)-(1/T_{\text{ref}})]} \quad (8)$$

where A is the property of the compounds [16].

2.2.5. Material–substrate interface

The TVOCs concentration at the material–substrate interface is given by the equation

$$C_s = K_{\text{sm}} C_m \quad (9)$$

where

$$K_{\text{sm}} = \frac{K_{\text{sa}}}{K_{\text{ma}}} \quad (10)$$

and K_{sa} is the partition coefficient between substrate and air.

2.2.6. Boundary conditions

The boundary conditions for VOCs are:

$$C_{\text{a,inlet}} = 0 \text{ at the air inlet;} \quad (11)$$

$$-D_m \left. \frac{\partial C_m}{\partial z} \right|_{z=\delta_s+\delta_m} = h_m(C_m - C_a) \quad (12)$$

at the film and air interface

$$-D_s \left. \frac{\partial C_s}{\partial z} \right|_{z=\delta_s} = -D_m \left. \frac{\partial C_m}{\partial z} \right|_{z=\delta_s} \quad (13)$$

at the substrate and material film interface

and

$$-\left. \frac{\partial C_s}{\partial z} \right|_{z=0} = 0 \text{ at the other side of substrate} \quad (14)$$

$$-\left. \frac{\partial C_s}{\partial n} \right|_{z=0} = 0 \text{ at other surfaces for the film and substrate} \quad (15)$$

where h_m is the local convective mass transfer coefficient between the air stream and emission surface (m/s).

2.2.7. Initial conditions

The TVOCs initial conditions are:

$$C_{\text{a},0} = 0 \text{ in the air,} \quad (16)$$

$$C_{\text{m},0} = C_{1,0}/\alpha \quad (17)$$

where α is the liquid expansion factor which reflects the absorptivity of the substrate. Physically, α means that once a wet material is applied to an absorptive substrate, the volume of the liquid film absorbed by the substrate will expand by a factor of α to the initial volume, and the initial VOC concentration in the film will decrease by a factor of α .

$$C_{\text{s},0} = 0 \text{ in the substrate} \quad (18)$$

2.2.8. Local mass transfer coefficient in air side

An expression has been summarized to predict the local convective mass transfer coefficient h_m on the material surface. This expression is obtained by curve fit of experimental data from water emissions in previous studies [9]. Based on the results, the local Sherwood number is given by the equation

$$Sh_L = 0.3359 Re Sc \left(\frac{r_0 - r}{2\delta} \right)^{-0.834} \quad (19)$$

It is defined as

$$Sh_L = \frac{2h_m \delta_a}{D_a} \quad (20)$$

where δ_a is the height of air slit (m); D_a is the diffusivity of VOCs in air (m^2/s); Re is the Reynolds number and Sc is the Schmidt number. They are defined as

$$Re = \frac{2u_m \delta_a}{\nu} \quad (21)$$

$$Sc = \frac{\nu}{D_a} \quad (22)$$

where u_m is the mean air velocity at the slit (m/s), and ν is the kinematic viscosity of air (m^2/s).

3. Experimental studies

A commercially available water-based emulsion paint was used for the model validation. The air flow through FLEC was adjusted to 509 ml/min with air flow through the sample tube at 111 ml/min, providing an air exchange rate of 873 per hour and $50 \pm 3\%$ relative humidity. The temperature was measured by the built-in temperature sensors in the air pump at 23 ± 0.5 °C. The whole test rig is shown in Fig. 3.

The air samples were taken by using sorbent tubes connected as in the above Figure. The sorbent tubes used in this experiment are 7 in. long with a 12 cm sorbent bed and 1/4 in. O.D. stainless steel. The sorbent selected is Tenax® TA.

The paint test sample was prepared using the FLEC test plate for paint (Part no.: FL-0161). The test plate is designed for applications of test materials onto a defined surface area with a defined paint thickness and with the possibility to check the weight. The application thickness is defined by the thickness of a foil laminated on an aluminium plate and the application surface area is defined by the area of a circular hole in the foil. An application thickness of 0.08 mm can be obtained by applying the test material to the hole on the plate and distribute it using the FLEC Application Roller. The defined application area was obtained by removing the foil from the plate. The pictures of the test plate and application roller are shown in Fig. 4.

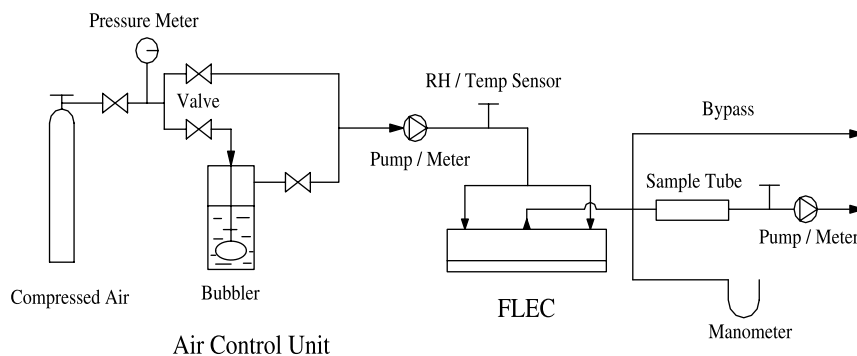


Fig. 3. Set up of the emission test with a FLEC.

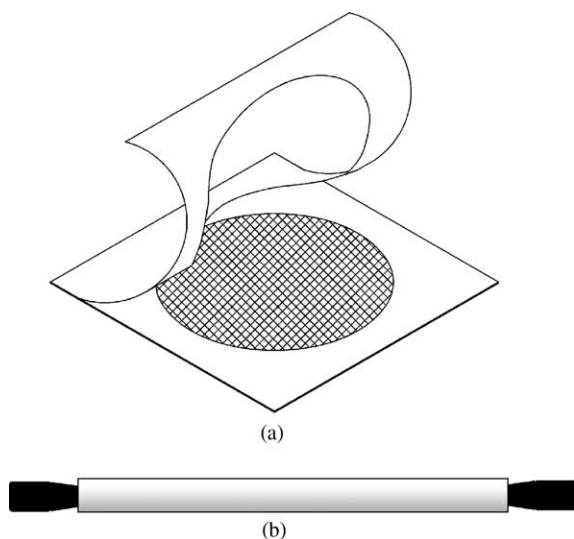


Fig. 4. Pictures showing the test plate (a) and the application roller (b).

The sampling period of each sorbent tube was set in the range from 10–30 min with a shorter period at the start and a longer period after few hours. The sampling time was controlled by the FLEC pump operation timer. The sampled tubes were recapped, recorded and replaced in the storage container immediately after sampling. The samples were refrigerated at 4 °C during storage. The sampled tubes were then transferred to the laboratory for analysis within 30 days of sample collection. The sampled tubes underwent thermal desorption followed by gas-chromatography/mass-spectroscopy according to the descriptions given in the US Compendium Method TO-17 [19].

Totally, there were 23 individual VOCs quantified. The most dominant VOCs are *n*-hexane, toluene, *o*-xylene, propylbenzene, 1-ethyl-3-methylbenzene, 1,3,5-trimethylbenzene, and 1,2,4-trimethylbenzene. All

individual VOCs are summed up to find the emission data of TVOCs.

4. Results and discussion

The emission process is modeled in following way: a CFD code FLUENT is used to calculate the steady state distributions of air velocity in the chamber. The flow is then incorporated into the VOC mass transfer Eqs. (1), (2) and (5) for simulating emissions. The convergence criteria used at each time step ensured that the total normalized residuals were less than 1% for flow and 3% for concentrations. A detailed description of the fluid flow in a FLEC can be found in Ref. [9]. Some parameters used in the test or simulation are shown in Table 1. They are obtained from headspace tests. For the experimental case, the effects of substrate are negligible, since aluminum plate is used as the substrate. It was also assumed that the TVOCs mixture has lumped properties.

The calculated TVOCs concentrations at FLEC outlet versus time are plotted in Fig. 5. The discretized values are the experimental data. From this figure, it is shown that the model predicts the emissions well. The

Table 1
Parameters in the emission test

Property	Symbol (unit)	Value
Initial VOC concentration in paint	C_{m0} (kg/m ³)	132
Partition coefficient for paint	k_{ma}	41560
Chamber volume	m ³	0.000035
Expansion factor	α	1.0
Constant for diffusivity in paint	D_0 (m ² /s)	2.13×10^{-6}
Diameter of emission surface	mm	150
Product density	kg/l	1.399
Wet film thickness	μ m	80
Temperature	°C	23 ± 0.5
Relative humidity		$50 \pm 3\%$

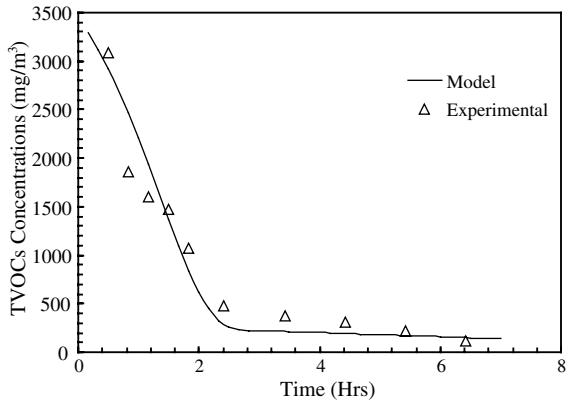


Fig. 5. Concentrations of TVOCs at the outlet of FLEC.

TVOCs decay fast at the beginning, and then emit steadily after 2 h.

Fig. 6 shows the percentages of mass of TVOCs emitted, and left in the paint material with time. It can be found that at the beginning, VOCs emitted very quickly. Actually, about 65.2% of the VOCs were emitted within the first 4 h, while 34.7% of the VOCs were left in the material. After that, the emission amount increased slowly. At 24 h, 85.8% of the VOCs are emitted, while only 13.9 remain in the material. Emissions from this small amount, however, is expected to last for a long period of time. After 10 days, about 99% of the VOCs are released.

Fig. 7 shows the mean concentrations of TVOCs in the air, at the paint surface, and in the paint film. It is clear from this figure that the three concentrations in the air and in the paint decrease drastically in the first 4 h, and they decrease steadily afterwards. The concentrations in the paint were higher than those at the surface, meaning TVOCs emitted from the paint to the air con-

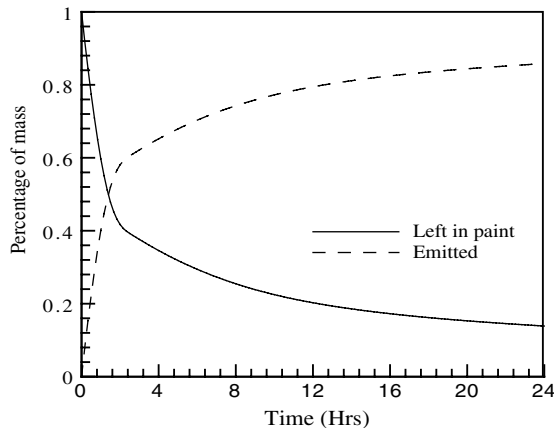


Fig. 6. Percentages of TVOCs mass emitted, and left in the material film in a FLEC.

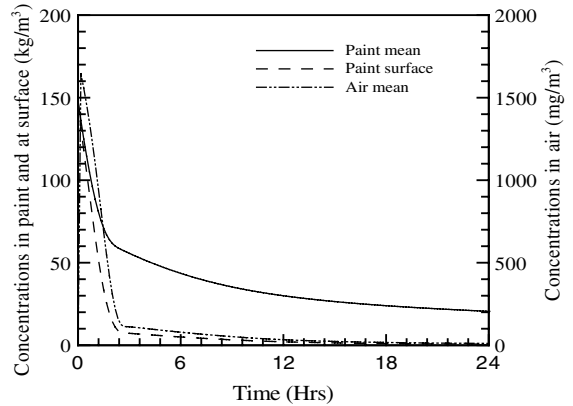


Fig. 7. Evolutions of air-based TVOCs concentrations in air flow and paint film.

tinuously. The differences between the concentrations of paint and air, which are the driving forces for emission, first increase and then decrease with time. This implies that the emission rates first increase, reach a climax, and then decrease gradually. There are great differences of concentrations between TVOCs in the solid and at the surface, implying that internal resistance is the predominant resistance in the FLEC. The resistance between the surface and air, i.e., convective transfer resistance, is relatively small.

To know the mechanisms of emissions clearly, the fields of TVOCs in the paint are plotted in Fig. 8. Two profiles at 3 and 15 h are drawn, respectively. The values in the figure are concentrations of TVOCs in the material. The dimensionless coordinates are defined as height

$$z^* = \frac{z}{\delta_m} \tag{23}$$

where δ_m is the thickness of paint film; radius

$$r^* = \frac{r_0 - r}{r_0} \tag{24}$$

where r_0 is the radius of paint film.

From Fig. 8, it can be seen that at the beginning, only a small portion of the surface, which locates at the perimeter of the chamber, emits VOCs to the air stream. In Fig. 8(a), the TVOCs gradients have the highest values at $r^* = 0$, and decrease with r^* . In the radius of $0 \leq r^* \leq 0.4$, the VOCs diffuse mainly in axial direction and releases large quantities of VOCs. We call this part of the surface as the active area. Accordingly, we call the part of $0.4 < r^* \leq 1.0$ as inactive area. In this area, VOCs mainly diffuse radially in an outward direction and little VOCs are released to the air stream directly. This is because air stream becomes saturated with emission surface shortly after it enters the chamber. This phenomenon has been proved with our previous experiments with water emission [9].

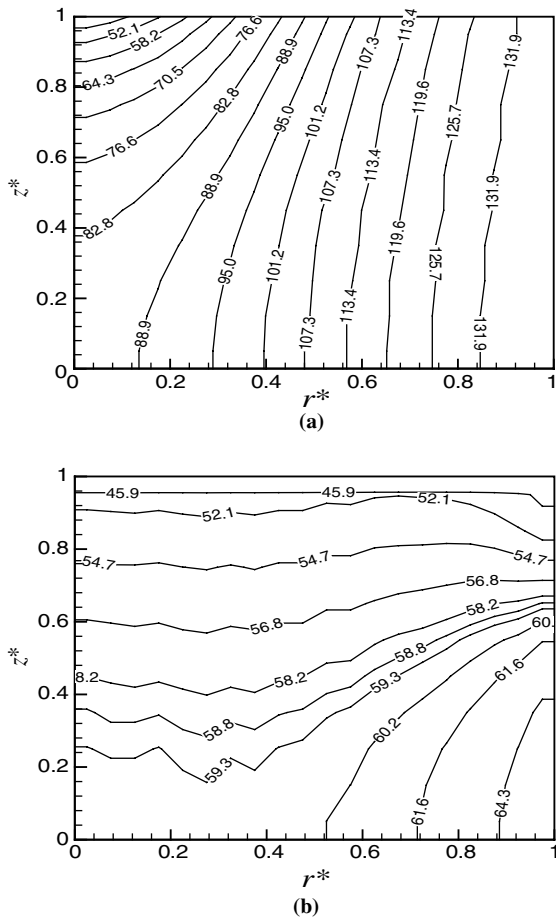


Fig. 8. Profiles of concentrations of TVOCs (kg/m^3) in paint: (a) after 3 h and (b) after 15 h.

The active area extends toward the center of the chamber with time lapsing. After a long period, the emissions along the chamber surface become relatively uniform, see Fig. 8(b). In this period, the internal diffusion dominates the emission process. This period lasts very long.

The adsorptivity of the substrate has a great impact on the emissions of the paint. Fig. 9 shows the effects of substrate partition coefficient on the percentages of mass emitted at 24 h. The thickness of the substrate is selected as 10 mm, the diffusivity is fixed at $1.0 \times 10^{-14} \text{ m}^2/\text{s}$, which is a typical value for TVOCs in oak board [17].

From Fig. 9, it is concluded that the greater the partition coefficient for substrate, the more VOCs are diffused into substrate from paint film, and the less VOCs are emitted to the air flow. The substrate acts as a sink for VOCs: it first absorbs VOCs from painting, and then emits them to air as a secondary source. The sink effect prolongs the emissions of VOCs from wet paintings. Such sink effects have negative effects on indoor air quality.

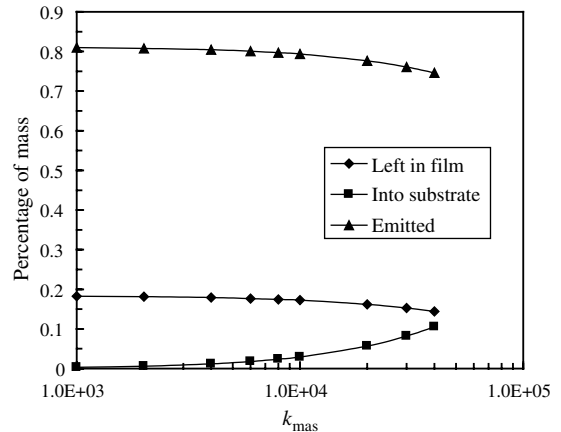


Fig. 9. Effects of partition coefficient of substrate on percentages of mass, emitted, left in material, and diffused into substrate, at 24 h.

5. Conclusions

This study presents a detailed modeling of emissions of VOCs from wet painting in a standard FLEC. The study found that since air stream becomes saturated with paint surface shortly after it enters the FLEC chamber, only the active area emitted VOCs to the air during the beginning hours. During this period, evaporation dominates the emission process. The active area extends to the whole surface with the time passing. Then, the internal diffusion will dominate the emission process.

A case study with a water-based emulsion paint shows that about 65.2% of the VOCs were emitted within the first 4 h. The emissions then slow down and persist for a long period. After 10 days, about 99% of the VOCs were released to the air stream. This fast decay is due to the very large convective mass transfer coefficients above the emission surface.

The substrate which has a high adsorptivity acts as a secondary source for VOCs emissions. Such effects may prolong the decay of VOCs, and the emissions from “wet” materials.

Acknowledgements

This research is funded by the Postdoctoral Fellowship of the Hong Kong Polytechnic University and the PolyU 2000/01 large equipment funding.

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