

Permeation of tracer gases through building materials—consequences for ventilation measurements with tracer gas techniques

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ABSTRACT

Tracer gas sorption in and permeation through building materials influence tracer gas ventilation measurements. Therefore, it is important to have knowledge about these processes.

The permeation of three commonly used tracers (SF₆, N₂O and the PFT C₆F₆) through untreated gypsum board has been experimentally investigated. The result shows that all three tracers diffuse readily through this material (diffusion coefficients in the order of 1×10^{-6} m²/s). Caution should therefore be exercised when using tracer gas measurements in rooms with walls of gypsum board or other porous materials.

INDEX TERMS

Tracer gas; Ventilation measurement; Permeation

INTRODUCTION

Air change rates in buildings are often determined from tracer gas measurements. The most commonly used tracer gases are sulfur hexafluoride (SF₆), nitrous oxide (N₂O) and perfluorinated hydrocarbons (PFT). In order to be useful for ventilation measurement the only sink for the tracer gases should be their dilution with ventilation air. Otherwise, the measurement will yield erroneous results. Tracer gases should therefore be inert, neither sorbing onto and nor diffusing through building materials. It is generally assumed that the commonly used tracer gases fulfil these requirements. To the knowledge of the authors no systematic scientific investigation of possible sinks for such tracer gases has been published. However, Dorer *et al.* (2000) and Hill *et al.* (2000) recently reported that erroneous ventilation measurement results were obtained using some PFTs. Their explanation was that the PFTs used exhibit strong sorption to indoor materials.

The aim of the present work is to contribute with a systematic study of possible interaction between various tracer gases for ventilation measurement and indoor materials. In this paper, we reported on the permeation of three tracer gases—SF₆, N₂O and the PFT C₆F₆ through untreated 13 mm gypsum board.

METHODS

Test Chamber

The measurements were performed using a 0.85 m³ chamber ($L = 2.36$ m, $H = 0.6$ m, $W = 0.6$ m) (see Figure 1). The chamber is designed to minimize possible sink effects. The upper surface is a glass plate, while the other sides are made of MDF-board coated on the inside with 2 mm aluminium plate. A solvent-free epoxy resin is used to seal walls to each other. A 1.73 m section of one long side is removable, in order to facilitate its exchange with a gypsum board (exposed area = 0.878 m²). The presence of rubber, Teflon and silicone gaskets is avoided.

The test chamber is ventilated by shunting a part of a main airflow using an adjustable ball valve. A calibrated measurement flange measures the main airflow rate which is kept at approximately 100 m³/h. The main air flow is exhausted outdoors. The air pressure in the main plenum is approximately 350 Pa. The airflow rate to the chamber inlet is measured with a rotameter calibrated by a soap bubble meter. Air mixing is arranged so that the boundary conditions at the inside of the chamber walls are constant and well defined. Air flows along the surfaces at a constant velocity despite different air change rates. This is accomplished by an

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internal air return flow through a 2 m long steel tube with a fan mounted in one end. In order to avoid turbulence, the air passes through two metal screens of different mesh size, before it enters the zone with the material to be tested.

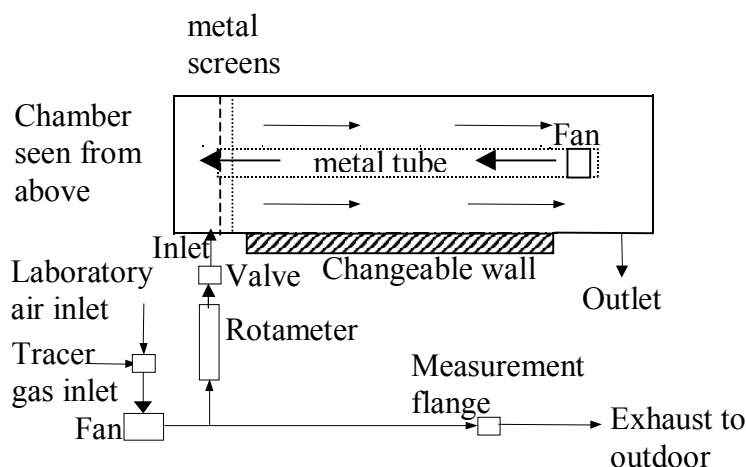


Figure 1 The aluminium/glass chamber. The chamber is equipped with a removable wall and an internal air flow controller which provides well-defined boundary conditions at the inside and mixing of chamber air.

The internal return airflow rate is approximately 70 m³/h, which is at least 30 times the supply airflow rate. The air velocity inside the chamber was measured using thermo-anemometry at several locations in-between the gypsum wall and chamber centre at two heights (0.3 and 0.15 m). The mean velocity was 0.16 m/s. The over-pressure in the test chamber was also measured.

Tracer Gas Injection

Tracer gas is injected before the centrifugal fan, which feeds the main airflow. SF₆ is injected using an Innova (Brüel & Kjær) 1303 sampling and dosing instrument. PFT is injected using a pack of 10 passive tracer gas sources, each with an emission rate of 2.2×10^{-5} g/h at 20°C.

Tracer Gas Sampling and Analysis

Air samples are taken via 4 mm polyethylene tubing. There are five tracer gas sampling points. One in the chamber inlet, before the adjustable valve, two points in the middle of the chamber, one point close to the exhaust from the chamber and one point in the room air close to the main flow inlet.

SF₆ and N₂O concentrations at the five measurement points are measured continuously with an Innova (Brüel & Kjær) 1302 multi-gas analyser. PFT concentrations are measured intermittently by parallel pumped sampling from the inlet air and the three chamber sampling points onto four charcoal adsorbent tubes. The sorbent tubes are analysed for tracer using liquid extraction and gas chromatography with an electron capture detector (Stymne and Eliasson, 1991).

Experimental Design

Three different tracer gases were tested for their permeation through 13 mm untreated gypsum board: SF₆, N₂O and C₆F₆.

The following experiments were performed:

1. Test of chamber tightness with inert wall
 - 1a. SF₆ decay with inert wall at 1.07 ACH
 - 1b. SF₆ decay with inert wall at no flow (inlet and outlet closed)
2. Test of chamber tightness with gypsum board with an external diffusion barrier
 - 2a. SF₆ steady state at 1.07 ACH
 - 2b. SF₆ decay at no air flow (inlet and outlet closed)
3. Test of SF₆ permeation through gypsum board
 - 3a–3d. SF₆ step-up, steady state and decay at 0.48, 0.70, 1.07 and 1.94 ACH
4. Test of N₂O permeation through gypsum board

- 4a. N₂O step-up, steady state and decay at 1.07 ACH
 5. Test of C₆F₆ permeation through gypsum board
 5a–5b. Steady state after 24 and 48 h at 1.07 ACH
 5c. Steady state after 24 hours at 0.48 ACH
 5d. Steady state with an external diffusion barrier

CALCULATIONS

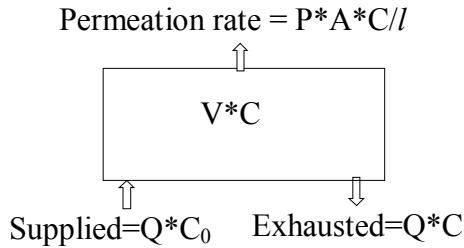


Figure 2 Tracer mass and mass transports in the chamber.

P denotes the permeation coefficient in m²/s.

A is the exposed area of the wall

l is the thickness of the wall

V is the chamber volume

C is the tracer concentration in the chamber

C_0 is the inlet tracer concentration

Q is the flow rate of air delivered to the chamber

Equation 1 shows the mass balance equation in the chamber, assuming steady state diffusion through the wall, zero concentration outside the chamber and no sorption of tracer compound. C_0 denotes the inlet concentration and C_∞ the steady state concentration. V is the chamber volume, P is the permeation coefficient in the board, A is the board area and l is the board thickness.

$$V \frac{dC}{dt} = QC_0 - QC - \frac{PA}{l} C \quad (1)$$

which yields

$$C_\infty = \frac{Q}{Q + \frac{PA}{l}} C_0 \quad \text{and} \quad P = \frac{(C_0 - C_\infty) l}{C_\infty} \frac{Q}{A} \quad (2)$$

$$\frac{PA}{l} = Q \left(\frac{C_0}{C_\infty} - 1 \right)$$

The integrated forms are for step up:

$$(C_\infty - C) = (C_\infty - C_{t=0}) e^{-\frac{\left(Q + \frac{PA}{l}\right)}{V} t} \quad (3)$$

and for step down:

$$C = C_{(t=0)} e^{-\frac{\left(Q + \frac{PA}{l}\right)}{V} t} \quad (4)$$

The permeation coefficient can be calculated from the diffusion constant D in the material and the mass transfer coefficients on the inside (k_i) and the outside (k_e).

$$\frac{l}{P} = \frac{l}{D} + \frac{1}{k_i} + \frac{1}{k_e} \quad (5)$$

Estimations of the mass transfer coefficients (k) are done with the following well-established correlation from boundary layer theory:

$$Sh = \frac{k \cdot L}{D_a} \quad (6)$$

where $Sh = 0.646 \cdot Re_L^{1/2} Sc^{1/3}$ for laminar flow over a flat plate ($Re_L < 5 \times 10^5$) and

$$Re_L = \frac{U_\infty \cdot L}{\nu}, \quad Sc = \frac{\nu}{D_a}$$

L is the length of the plate, D_a is the diffusion coefficient of tracer in air, U_∞ is the bulk air velocity and ν is the kinematic viscosity for air. Calculations for the mass transfer coefficient are based on 20°C temperature with a characteristic length (L) of 1.73 m.

RESULTS

The test of the chamber tightness with the inert wall (experiment 1a) shows a strictly exponential decay, with a calculated ACH, which is within 5% of the one given by the rotameter flow. The second experiment (1b) is also strictly exponential decay, with a calculated ACH of 0.04 h^{-1} , which corresponds to a leakage flow of 35 l/h.

The test of the chamber tightness with the gypsum board wall, through which permeation is hindered by an aluminium foil attached to the external side of the board, (experiment 2a), shows that all three concentrations in the chamber are close to the inlet concentration. Experiment (2b) is also a strictly exponential decay, with a calculated ACH of 0.08 h^{-1} , which corresponds to a leakage flow of 65 l/h.

The step-up, steady state and step-down processes for experiments 3a–3d are shown in Figure 3 for SF_6 for the four different residence times 2.1, 1.43, 0.93 and 0.52 h. The fifth diagram shows the steady state and decay when permeation is hindered by an aluminium foil attached to the external side of the gypsum board (experiment 2a and 2b). It is clearly demonstrated from these diagrams that SF_6 has a sink, which strongly depends on the residence time in the chamber.

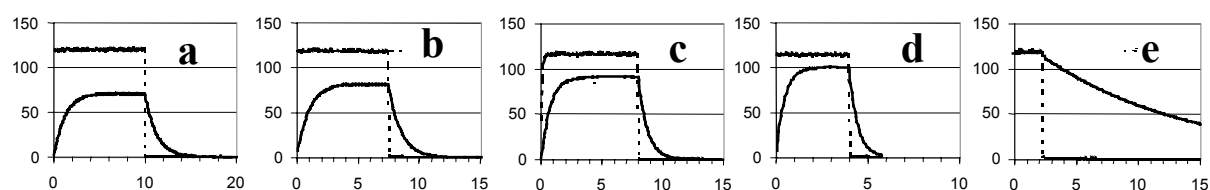


Figure 3 Concentration (mg/m^3) as a function of time (h) at step-up, steady state and decay of SF_6 tracer for different air change rates ($0.48, 0.70, 1.07$ and 1.94 h^{-1}). The fifth diagram shows the steady state at an ACH of 1.07 h^{-1} with a permeation barrier and the decay without flow for this case. Upper curves show the inlet concentration.

Nitrous oxide has only been examined in one experiment (4a) at a residence time of 0.93 h. The step-up, steady state and decay is shown in Figure 4. This diagram demonstrates that N_2O also has a sink in the chamber, which is even larger than that of SF_6 (compare with diagram c in Figure 3).

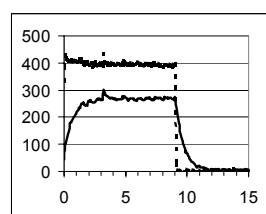


Figure 4 Concentration (mg/m^3) as a function of time (h) at step-up, steady state and decay of N_2O tracer for an ACH of 1.07 h^{-1} .

The steady state concentrations of the perfluorocarbon C_6F_6 in the chamber (experiments 5a–5c) are shown in the bar graph in Figure 5. Data are displayed after 24 and 48 h at 1.07 ACH and after 24 h at 0.48 ACH . All concentrations are relative to the ones analysed in the inlet. These experiments show that C_6F_6 also has a sink comparable to that of SF_6 .

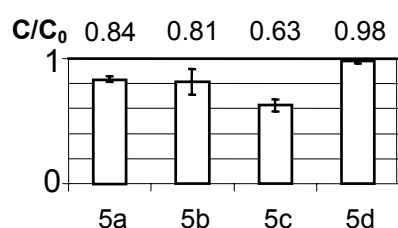


Figure 5 The ratio between the steady state concentrations of C_6F_6 in chamber and the inlet concentration.

- (5a) at 1.07 ACH after 24 h
 (5b) at 1.07 ACH after 48 h
 (5c) at 0.48 ACH after 24 h
 (5d) at 1.07 ACH with an external diffusion barrier

The permeation and diffusion coefficients of SF₆, N₂O and C₆F₆ in the gypsum board are calculated both from the steady state concentrations and from the decays using Eqns (2) and (4), respectively. The results are shown in Table 1. The mass transfer coefficient on the inside (k_i) is estimated to be 5×10^{-4} m/s and on the outside (k_e) to be 3×10^{-4} m/s, using Eqn (6) and a diffusion constant of tracer in air of 1×10^{-5} m²/s.

Table 1 Permeation and estimates of diffusion coefficients from steady state and decay

	SF ₆	SF ₆	SF ₆	SF ₆	[†] SF ₆ re-run	[†] SF ₆ re-run	N ₂ O	PFT 24 h	PFT 48 h	PFT 24 h	PFT barrier
Over-pressure in chamber [Pa]	0.5	1	2	5	<0.5	<0.5	2	2	2	0.5	2
Air change rate [h ⁻¹]	0.48	0.70	1.07	1.94	1.07	1.94	1.07	1.07	1.07	0.48	1.07
Residence time [h]	2.10	1.43	0.93	0.52	0.93	0.52	0.93	0.93	0.93	2.10	0.93
Air flow rate [m ³ /h]	0.41	0.60	0.91	1.65	0.91	1.65	0.91	0.91	0.91	0.41	0.91
PA/l [m ³ /h] steady state	0.28	0.28	0.27	0.22	0.25	0.23	0.37	0.17	0.21	0.24	0.02
PA/l [m ³ /h] decay	0.30	0.28	0.28	0.33	0.26	0.19					
P [10 ⁻⁶ m ² /s] steady state	1.2	1.1	1.1	0.9	1.0	1.0	1.5	0.7	0.9	1.0	0.08
P [10 ⁻⁶ m ² /s] decay	1.2	1.2	1.1	1.4	1.1	0.8					
D^* [10 ⁻⁶ m ² /s] steady state	2.2	2.1	2.0	1.4	1.8	1.6	4.0	1.0	1.4	1.6	0.08
D^* [10 ⁻⁶ m ² /s] decay	2.5	2.3	2.2	3.1	2.0	1.2					

*The calculation of the diffusion coefficients is based on the experimentally determined permeation coefficients P and estimated values of mass transfer coefficients on the chamber inside (5×10^{-4} m/s) and the outside (3×10^{-4} m/s).

[†]The re-runs of SF₆ are performed with reduced chamber over-pressure.

DISCUSSION

The estimated permeation and diffusion coefficients are of the same order and reasonably independent of the residence time of tracers SF₆ and C₆F₆. N₂O seems to diffuse faster than the other two tracers, but as this conclusion is only based on a single measurement on N₂O, it is only preliminary. The permeation coefficients seem to be more accurately determined from steady state measurements ($1.0 \times 10^{-6} \pm 0.1 \times 10^{-6}$ for SF₆) than from decay ($1.1 \times 10^{-6} \pm 0.2 \times 10^{-6}$).

The results show that the untreated gypsum board exhibits a strong and continuous sink for the investigated tracer compounds. When sealing the external side of the board, the sink effect ceases. The sink is therefore probably primarily due to permeation of the tracer gases through the gypsum board. The rate of permeation is governed by the diffusion coefficient of tracer in the board and on the mass transfer coefficients on the inner and outer sides of the board. The permeation rates are so high for the investigated compounds, so that the mass transfer coefficients play an important role for the permeation rate. Due to lack of precise data, the diffusion coefficients shown in Table 1 are estimated from calculated reasonable values of the mass transfer coefficients and the same for all tracers.

A pressure difference over a porous material will induce a convective flow through the material. In a similar gypsum board this pressure induced air velocity was found to be $v = 2.5 \times 10^{-7} \Delta p$, where v is the air velocity in m/s through the material and Δp is the pressure difference in Pa. The pressure induced airflow at 5 Pa will therefore be less than 4 l/h through the investigated gypsum board. This convective flow is negligible compared to the diffusion induced permeation and will not influence the calculation of permeation coefficients.

Gypsum board is the most widely used building material for cladding of indoor walls in modern buildings. The permeation rates of commonly used tracer gases are so high through this material that this sink must be considered when performing ventilation measurements using tracer gas. Usually, the gypsum board is surface treated with wallpaper or paint. In colder climates, external walls are also usually lined with a plastic foil between the insulation and the gypsum board. Such factors will drastically diminish the permeation rate. In order to investigate the effect of surface treatment and liners, we plan to continue this systematic investigation of tracer permeation.

There may also be sorption effects in building materials. In contrast to permeation, sorption is a transient effect. If sorption is small compared to permeation it might be difficult to observe. It would best be observed as a 'tail' in the decay curve, when permeation is hindered through the material. If the permeation effect is not accounted for, observed sinks of tracer gas may be misinterpreted as only due to sorption effects.

CONCLUSIONS

Untreated gypsum board shows a large permeability towards tracer compounds commonly used for ventilation measurements. Caution should therefore be used when using tracer gas measurements in rooms with walls of gypsum board or other porous materials. Further studies of tracer permeability through surface treated building materials are needed.

ACKNOWLEDGEMENTS

The authors thank workshop staff, Ragnvald Pelttari and Larry Smids for skilled construction work and Anita Eliasson at Pentiaq for performing PFT analysis.

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