International Communications in Heat and Mass Transfer 87 (2017) 105–111

Contents lists available at ScienceDirect

International Communications in Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ichmt

The combined effects of temperature and electric fields on formaldehyde emission from building materials: Experiment and molecular dynamics simulation

Bo Xu[a], Zhenqian Chen[a,b,*]

[a] Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education, Southeast University, Nanjing 210096, PR China

[b] Jiangsu Provincial Key Laboratory of Solar Energy Science and Technology, School of Energy and Environment, Southeast University, Nanjing 210096, PR China

ARTICLE INFO

Keywords:
Temperature
Electric field
Formaldehyde
Building materials
Three key parameters
Molecular dynamics

ABSTRACT

An experimental and molecular dynamics simulation study concerning the emission of formaldehyde from building materials in different temperature and electric fields was conducted. Formaldehyde concentration was tested at the temperature of 298 K, 303 K, 308 K, 313 K and electric field of 0 kV, 12 kV, 18 kV, 24 kV. A new calculation method for solving three key parameters of formaldehyde emission was proposed and verified by experimental results. It was concluded that building materials emitted more formaldehyde at higher temperature and electric field. With the increase of temperature and electric field, experimental results indicated that both initial concentration (C_0) and diffusion coefficient (D_m) increased while partition coefficient (K_m) decreased. In addition, the temperature made larger contribution than electric field not only on formaldehyde emission and three key parameters by experimental results, but also on adsorption potential in the molecular dynamics simulation. What's more, the adsorption potential decreased while diffusion coefficient increased with greater temperature and electric field in the molecular dynamics simulation system, corresponding well with experimental results. The results of this study confirmed the contribution of electric field and temperature for the reduction of formaldehyde in the man-made boards.

1. Introduction

Building materials such as man-made boards are the most dominating formaldehyde emission sources indoors [1]. It has been shown that C_0, K_m and D_m are three key parameters affecting the emissions characteristics of indoor materials [2]. The key parameters are affected by environmental conditions, such as temperature and high-voltage electric field. The effect of temperature on emission parameters has been studied by many scholars. Huang [3], Deng [4] and Zhang [5] proposed a theoretical correlation between C_0-T, D_m-T and K_m-T respectively for formaldehyde emissions. However, the effect of electric field on formaldehyde emissions from building materials has been rarely studied.

Many methods were used by researchers to study emission parameters including three key parameters and formaldehyde concentration. Yan [6] used multi-gas/solid ratio method and Xiong [7] proposed a multi-emission/flash regression method to obtain three parameters with complicated experiment procedure. Wang [8] established a multiple equilibrium regression method (MERM) which needed much more time in the whole experiment. Considering that, a new method was proposed to calculate three key parameters with short experimental time and procedure.

Most of scholars were focused on macro-scale study of formaldehyde diffusion within building materials and had little research on microscopic mechanism. The structure of medium density fiberboard (MDF) was very complicated and it couldn't be constructed completely in micro-scale. Cellulose accounts for more than 70% of MDF and was the best choice. In that case, the research of formaldehyde diffusion within cellulose in micro-scale represents for formaldehyde diffusion within MDF in macro-scale. The molecular dynamics [9] has been used to simulate the diffusion movement among molecules successfully.

In this study, experiment on formaldehyde emissions at different temperature and electric field was performed and molecular dynamics simulation of formaldehyde diffusion within cellulose was carried out to verify and explain the experimental results. A new calculation method was proposed to calculate three key parameters.

* Corresponding author at: Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education, Southeast University, Nanjing 210096, PR China.
E-mail address: zqchen@seu.edu.cn (Z. Chen).

http://dx.doi.org/10.1016/j.icheatmasstransfer.2017.06.006

Nomenclature

C_0	initial concentration of formaldehyde in building material (mg/m^3)
$C_a(t)$	calculation results of formaldehyde concentration in chamber (mg/m^3)
$C_e(t)$	experimental results of formaldehyde concentration in chamber (mg/m^3)
C_{equ}	equilibrium concentration of formaldehyde in test chamber (mg/m^3)
D_m	mass diffusion coefficient for formaldehyde in building material (m^2/s)
h_m	convective mass transfer coefficient (m/s)
E_{ads}	adsorption potential (kcal/mol)
$E_{cellulose}$	energy of cellulose Iβ (kcal/mol)
$E_{formaldehyde}$	energy of formaldehyde molecules (kcal/mol)
$E_{formaldehyde + cellulose}$	total energy of formaldehyde diffusion on cellulose Iβ (kcal/mol)
K_m	partition coefficient between building material and air (dimensionless)
T	temperature (K)
τ	time (s)

2. Experiment

2.1. Experiment apparatus and conditions

2.1.1. Experiment apparatus

Experimental principle, apparatus, samples of MDF and h_m were described in ref. [10]. One plate electrode was connected to a high-voltage power supply which provided a constant voltage of 0–50 kV and the other plate electrode was connected to the ground. The static electric field was generated between two plate electrodes. A water bath was applied to keep or change the air temperature in chamber. The concentrations of formaldehyde in chamber were continuously recorded by HTV-m formaldehyde analyser. A calibrator with formaldehyde concentration and temperature corresponding one by one was used to calibrate formaldehyde analyser. Calibrator was placed in the experimental table for more than 1 h and the environmental temperature was recorded. When analyser extracted formaldehyde from calibrator, adjusted the formaldehyde concentration of analyser to the formaldehyde concentration corresponded by environmental temperature. Repeat this process for several times until the difference between formaldehyde concentration of analyser extracted formaldehyde from calibrator and formaldehyde concentration corresponded by temperature was less than 5%.

2.1.2. Experiment conditions

In the experiment, the temperatures were 298 K, 303 K, 308 K, 313 K and electric voltage were 0, 12 kV, 18 kV, 24 kV. As the relative humidity of air tested in the chamber was 40 ± 5%, the chamber could be considered as a constant humidity environment. It is because that relative humidity from 20% to 50% has no significant impact on initial formaldehyde concentration and emission rate at the experimental temperatures when formaldehyde and VOCs emit from building material [11]. The experiment began when formaldehyde concentrations in chamber were below 0.01 mg/m^3. Then keep the temperature 298 K in the chamber and put the MDF sample into the chamber. Turn on the electric power to produce the electric field and keep the electric intensity constant in the process of a complete experiment. The concentrations of formaldehyde were recorded every half an hour. The formaldehyde diffusion reached equilibrium when relative changes in five consecutive formaldehyde concentrations were less than 5%. When the formaldehyde diffusion reached equilibrium, keep the electric field constant and change the temperature to 303 K until a new equilibrium was reached. Then the temperature was changed to 308 K and 313 K in turn. This completed the experimental procedure. Electric intensity was changed to begin a new experiment with a new MDF sample.

2.2. Error analysis

2.2.1. Formaldehyde concentration measurement

The detection range of HTV-m formaldehyde analyser was 0–10.00 ppm (1 ppm = 1.34 mg/m^3 for formaldehyde) with precision of 2%. The minimum formaldehyde concentration was measured as 0.93 mg/m^3. Therefore, the maximum relative error was as followed:

$$\varepsilon_{C,max} = \Delta C / C_{min} = (10 \times 2\% \times 1.34)/0.93 = 28.8\% \tag{1}$$

The maximum relative error happened at the beginning of experiment and the relative error decreased over time. When it reached equilibrium, the relative error was less than 20%. What's more, the maximum formaldehyde concentration reached two-thirds of the maximum range of formaldehyde analyser. In this situation, the maximum relative error was acceptable and the choice of formaldehyde analyse was reasonable.

2.2.2. Temperature measurement

The hot-wire anemometer (ST-732) was used to measure air temperature with the precision of ± 0.8 °C.

2.3. New calculation method

2.3.1. Description

A new method was proposed to calculate three key parameters only with real-time concentration curve of formaldehyde concentration in chamber. What's more, the experiment took a short time with a simple procedure. The development of computer made the new method possible for it had a quite high requirement for computer. The equations of formaldehyde emission from building materials in the airtight chamber are described in ref. [12] with Eqs. (1)–(10). There was an exact equation between C_0 and K_m. Therefore, only two variables (D_m and K_m) were in the process of solving equations. According to previous research experience, the range of K_m and D_m were 500–15,000 [10] and 10^{-12} m^2/s–10^{-10} m^2/s [8]. Hence, K_m increased from 500 to 15,000 with a step of 50 and D_m improved from 10^{-12} m^2/s to 10^{-10} m^2/s with a step of 10^{-12} m^2/s during calculation. Then do the iteration and obtain the real-time curve ($C_a(t)$) of formaldehyde concentrations in chamber. The least square method (LSM) was applied compared $C_a(t)$ with experimental results of $C_e(t)$. At last, the curve of $C_a(t)$ which was closest to $C_e(t)$ was gained and the corresponding C_0, K_m and D_m will be output. The specific process was shown in Fig. 1.

2.3.2. Validation

Fig. 2(a) described the average value of formaldehyde concentration in real-time and relative errors obtained by more than three groups of experiment. Nearly all relative errors were less than 5% except the early stage of emission (10%) caused by large fluctuation of concentration in initial emission process. The low relative errors verified the reliability of experimental results. The increase of formaldehyde concentration in chamber means more formaldehyde emission from boards. Formaldehyde emission increased with greater temperature, corresponding well with ref. [5]. The calculation result of C_a was also shown in Fig. 2(a) using the above calculation method. The calculation results corresponded well with experimental results verifying the reliability and accuracy of new method. In addition, the relationship between K_m and T was followed [5]: