Outgassing characteristics of a polycarbonate core material for vacuum insulation panels

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ABSTRACT

Performance deterioration with time is one of the most important issues in a vacuum insulation panel (VIP), which is mostly due to the inner gas pressure rise. Outgassing from the interior of core materials is the major gas source when the core material is a polymer. Outgassing characteristics of a polycarbonate as the VIP's core material are examined theoretically and experimentally. To measure the outgassing rate, specific outgassing tests are carried out using a pressure rise method. Diffusive outgassing mechanism is discussed based on the Fick's law. As the result, the total amount of dissolved gas and the diffusion coefficients of various gases in the polycarbonate are obtained by using the measured outgassing rate. Temperature dependence of the diffusion coefficient of nitrogen is also examined. It is shown that the outgassing rate of polymer core materials can be significantly reduced to a negligible level by a baking pre-treatment in vacuum and/or by a metal coating on the polymer surface.

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1. Introduction

Vacuum insulation panels (VIPs) as a high-performance thermal insulator have been widely developed to reduce not only energy consumption but also CO₂ emissions. The VIP has various applications such as buildings, refrigerators and cryogenic devices. Although insulation performance of VIPs usually degrades with time, the initial thermal resistance of VIPs for refrigerator application is about 5 times as high as that of conventional insulation materials at the same thickness. A VIP consists of a core material to withstand the external atmospheric pressure, an almost gas-tight envelope to maintain the inside vacuum and a getter or desiccant to adsorb various gas flowing into the panel. The core material has to be porous to easily evacuate and to have a minimum solid conduction effect. For this reason, bulk type materials in the form of powders, foams and fibers are frequently used as the core material [1]. In addition to these bulk type core materials, an artificial structure like multi-layered staggered beams (Fig. 1) can be applied [2]. It was shown that the initial thermal resistance of the staggered beam VIP is similar to that of the VIP with glass fiber arranged perpendicular to the heat flow direction.

The effective thermal conductivity k_{eff} (center-of-panel k value, excluding the edge conduction effect) is the most important measure of initial and long-term performance of a VIP. It can be determined by summation of the solid conductivity k_s, the gaseous conductivity k_g and the radiative conductivity k_r as

$$k_{eff} = k_s + k_g + k_r. \tag{1}$$

Among these, k_g is responsible for the long-term deterioration. It increases with pressure and pore size of the core. Because the gas pressure in VIPs increases with time due to leaks and inner outgassing, the performance deterioration is an inevitable consequence to some degree. Especially in case of the VIP with the multi-layered staggered beam core, slight pressure increase can increase k_g significantly due to the relatively large pore size [3]. Pressure rise should be suppressed, thus.

The increase of pressure in VIPs, initially at a high vacuum, is attributed to gas permeation through the envelope surface and sealing edge, and outgassing from the core material. Gas permeation through heat-sealed flanges is the most important factor responsible for the pressure increase when inorganic materials such as glass fiber or silica powder are used for the VIP's core. However, for the case of polymer core materials, outgassing from them has been suspected of the major gas source. The author's group has been working on such structures, including the aforementioned multi-layered staggered beam core made of polycarbonate. For this background, outgassing from the core material is investigated in this research.

There have been a few studies on the outgassing contribution in VIPs. For instance, outgassing in the VIP consisting of glass or ceramic spacer and stainless steel foil envelope is experimentally

Nomenclature

A	surface area, m^2
C	gas concentration, Pa L/cm^3
C_0	initial gas concentration, Pa L/cm^3
d	thickness, m
D	diffusion coefficient, cm^2/s
D_0	diffusivity at 'infinite' temperature, cm^2/s
E_d	activation energy of desorption, J/mole
E_v	latent heat of vaporization, J/mole
k	thermal conductivity, W/m·K
N	number of molecules
P	pressure, Pa
q	specific outgassing rate, Pa L/s·cm^2
Q	gas flow, Pa L/s
R_u	universal gas constant, J/mol·K

t	time, s
T	temperature, K
V	volume, m^3

Greek symbols

α	decay exponent
τ_0	nominal period of vibration

Subscripts

a	apparatus
c	coating
eff	effective
out	outgassing
p	polymer
s	sample

measured by Nemanic and Setina [4]. Outgassing of rigid open-celled polyurethane foam is investigated by Yang et al. [5]. However, a comprehensive interpretation of the diffusive outgassing mechanism for the VIP with the polymer structured core, as the staggered beam VIP is such an example with high outgassing potential, has not been reported yet.

The objective of this paper is thus to understand gas transport mechanisms in a vacuum for the VIP's core material and to evaluate it experimentally. Polycarbonate is used as the test specimen in this study, although the reported results are widely applicable to other materials too. A new technique measuring diffusion coefficients depending on gas species and temperature of the bulk type material is proposed. Based on the experimental results, a few methods for reduction of the outgassing contribution are proposed.

2. Gas emission from the core material

When a material is exposed to vacuum, the gas release (or, 'outgassing') occurs due to vaporization of the material, gas desorption from the surface of the material and/or diffusion from inside the bulk material (Fig. 2).

Vaporization occurs in all materials. The vapor pressure is defined as the partial pressure of the vapor over the surface in an equilibrium state, and it can be expressed by the following Clausius–Clapeyron equation [6]:
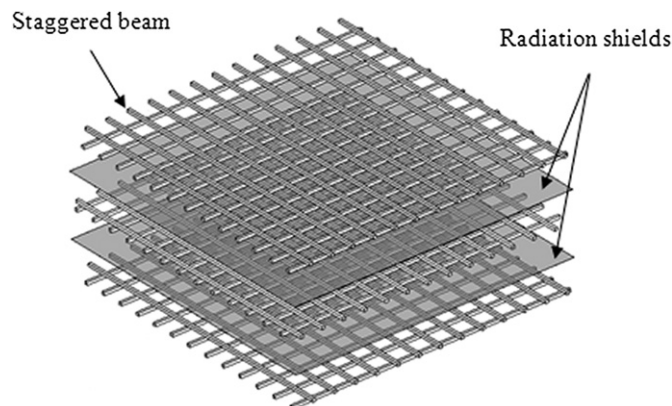
$$\ln P_s = A - \frac{B}{T}, \tag{2}$$

Fig. 1. Multi-layered staggered beam core [2].

where P_s is the saturation pressure, A and B are the constant. Based on the above equation, vapor pressure of various polymer materials is investigated by Jensen [7]. Vapor pressures of all the considered plastic materials in room temperature are in the range of 10^{-7} to 10^{-5} Pa. For example, polyethylene shows vapor pressure of 4.0×10^{-6} Pa. Due to lack of data, the vapor pressure of a polycarbonate is roughly estimated to be in the range of 10^{-7} to 10^{-5} Pa based on Jensen's result. Because the vacuum level in VIPs is generally about 10^{-1} to 10 Pa, the vaporization effect can be safely ignored.

Gases or vapors adsorbed on the surface while in atmosphere are slowly released by desorption phenomena. Gas molecules can be trapped or bound on the surface by physisorption and chemisorption. Physisorption occurs when molecules are bound to the surface by van der Waals force with energy less than 10 kcal/mol and chemisorbed molecules are trapped at energies greater than 10 kcal/mol [8]. Weakly physisorbed gases such as nitrogen, oxygen and rare gases can be removed at the surface in a short pumping time, while chemisorbed gases such as most organic species, water and carbon dioxide can be removed by applying an external heat. Desorption mechanism can be interpreted theoretically using a thermodynamic or a kinetic approach. Desorption rate when the surface is covered with multiple adsorbed layers is given by the zero order desorption model as [9].

$$-\frac{dN(t)}{dt} = a\exp\left(\frac{-E_v}{R_u T}\right), \tag{3}$$

where N is the total number of adsorbed molecules, E_v is the latent heat of vaporization, R_u is the universal gas constant and a is a constant. This equation can be applied to the case of the surface saturated with multiple layers of water vapor. However, the rate of desorption for the surface coverage of a monolayer or less can be expressed by the following first order desorption model [10]:
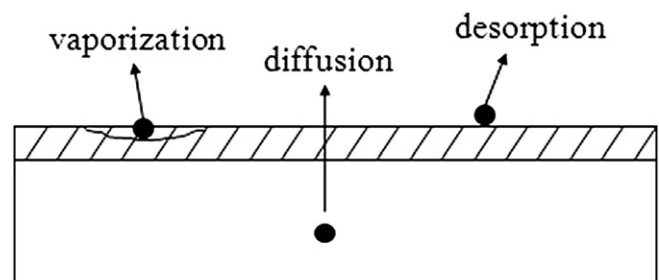

Fig. 2. Outgassing sources from a material in a vacuum.