



Studies on the barrier performance and adhesion strength of novel barrier films for vacuum insulation panels

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ABSTRACT

Novel cost efficient high barrier envelopes are developed for vacuum insulation panels. In order to fulfil the required maximum oxygen permeability (at 23 °C and 50% relative humidity (RH)) of $1 \times 10^{-14} \text{ m}^3(\text{STP})\text{m}^{-2} \text{ day}^{-1} \text{ Pa}^{-1}$ and water vapour transmission rate (at 23 °C and 85% → 0% RH) of $1 \times 10^{-6} \text{ kg m}^{-2} \text{ day}^{-1}$, hybrid polymeric (ORMOCER[®]) and aluminium barrier layers are combined. Using roll-to-roll processes, these materials are deposited from the liquid or gas phase on top of a poly(ethylene terephthalate) substrate film. The low adhesion strength observed between aluminium and ORMOCER[®] layers is significantly increased by the deposition of an additional aluminium oxide layer between the two materials. The lamination of a polyethylene or polypropylene sealing film does not weaken the adhesion strength. The resulting barrier structure has reached an oxygen permeability of lower than $5 \times 10^{-14} \text{ m}^3(\text{STP})\text{m}^{-2} \text{ day}^{-1} \text{ Pa}^{-1}$ (at 37 °C and 30% RH). Electrochemical impedance spectroscopy gives evidence for a possible penetration of the ORMOCER[®] lacquer into the aluminium oxide layer (synergistic effect), which explains the improved adhesion and barrier performance due to the aluminium oxide layer.

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1. Introduction

Vacuum insulation panels (VIPs) can meet the demand for durable, efficient, space saving and sustainable thermal insulation materials to be applied at buildings and refrigerators. The VIP core consists of a highly porous material having pore diameter in the micro- to nanometer scale and is encapsulated by a barrier envelope [1,2]. Typical VIP core materials used are open cell polyurethane, fumed and precipitated silica, glass fibres and aerogel based materials.

After evacuating the core material, the following mechanisms of heat transfer arise in a VIP: heat conduction by the residual gas (oxygen and nitrogen) or water vapour within the pores, heat conduction by the solid skeleton of the core and radiation [1,3,4]. The first contribution is the most significant one and it is either due to gas or water vapour permeation from the environment into the

VIP or due to outgassing from the core material or from the envelope. However, especially for more hygroscopic core materials such as fumed silica, a significant part of the water vapour within the pores is adsorbed at the inner surface of the core material resulting in heat conduction by this adsorbed phase or in heat transfer by evaporation, diffusion and condensation of water vapour [1,4].

In order to maintain the thermal insulation performance of VIPs during their intended lifetime, the barrier envelope has to significantly reduce the permeation of oxygen, nitrogen and water vapour into the VIP [1,4,5]. However, permeation takes place not only through the barrier envelope but also through the material of the sealing seam and through micro-defects possibly contained in it [5,6].

As an example, fumed silica based VIP having dimensions of $0.5 \text{ m} \times 0.5 \text{ m} \times 0.03 \text{ m}$ is considered. After 50 years a maximum thermal transmittance of $0.2 \text{ W m}^{-2} \text{ K}^{-1}$ is allowed [7], which corresponds to a thermal conductivity of $6.0 \times 10^{-3} \text{ W m}^{-1} \text{ K}^{-1}$ for such a VIP. In this case, the required barrier performance of a VIP envelope to gases is calculated for a temperature of 23 °C and 50% relative humidity (RH) in the environment. In order to simplify the situation, it is assumed that 50% RH will be reached in the VIP core material within the considered time period of 50 years.

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Neglecting coupling effects, the thermal conductivity λ of a VIP at dry conditions is the sum of the thermal conductivity of the evacuated VIP and of the gaseous conductivity λ_{gas} . The latter one as a function of the total gas pressure p within the pores of the core material is given by [8]:

$$\lambda_{\text{gas}} = \frac{\lambda_{\text{gas},0}}{1 + p_{1/2}/p}. \quad (1)$$

Here, $\lambda_{\text{gas},0}$ denotes the thermal conductivity of the free still gas and $p_{1/2}$ the gas pressure where the gaseous conductivity is equal to $\lambda_{\text{gas},0}/2$.

For fumed silica panels filled with nitrogen the following relation between λ_{gas} , pressure p and temperature T is obtained by fitting Eq. (1) to measured conductivities for pressures up to 5000 Pa [8]:

$$\lambda_{\text{gas}} = \frac{25.5 \cdot \sqrt{T/293.15\text{K}}}{1 + (T/293.15\text{K}) \cdot (5.93 \cdot 10^4 \text{ Pa}/p)} \cdot 10^{-3} \text{ W/mK}. \quad (2)$$

It will be shown that the gas pressure in the considered example does not exceed a value of 5000 Pa. Since the values of the mass and diameter of O_2 molecules differ from the correspondent values for N_2 molecules by less than 15%, Eq. (2) will approximately also be used for oxygen.

When a silica panel absorbs water vapour and the water vapour partial pressure and the temperature are sufficiently low, the thermal conductivity in the absence of gases is given by [8]:

$$\lambda = \left(0.624 + 0.0133 \cdot T/\text{K} + 1.14 \cdot \frac{p_{\text{H}_2\text{O}}(T)}{p_{\text{sat}}(T)} + 1.08 \cdot \frac{25.5}{6 \cdot 10^4 \text{ Pa}} \cdot p_{\text{H}_2\text{O}}(T) \right) \cdot 10^{-3} \text{ W/mK}, \quad (3)$$

where partial pressure and saturated pressure of water vapour are denoted by $p_{\text{H}_2\text{O}}$ and p_{sat} , respectively. In the case of the maximum possible water vapour pressure of 1400 Pa within the pores, corresponding to 23 °C and 50% RH, Eq. (3) gives a thermal conductivity of $5.8 \times 10^{-3} \text{ W m}^{-1} \text{ K}^{-1}$.

Since the considered gas and water vapour pressures are very low, it is assumed that their contributions presented in Eqs. (2) and (3) can be added up to the total thermal conductivity of the VIP. Consequently, the contribution of the permeating dry gases (oxygen and nitrogen) to the thermal conductivity should not be more than $0.2 \times 10^{-3} \text{ W m}^{-1} \text{ K}^{-1}$ after 50 years. According to Eq. (2) this corresponds to a total pressure of about 500 Pa for dry gases. Therefore, a maximum increase of gas (oxygen and nitrogen) pressure of 10 Pa year^{-1} is allowed within this VIP in order to be able to maintain a thermal conductivity of at most $6.0 \times 10^{-3} \text{ W m}^{-1} \text{ K}^{-1}$ for 50 years.

In order to estimate how the gas permeation through sealing seams contributes to the gas pressure increase, low density polyethylene (LDPE), a typical material used for sealing layers, is considered. A sealing layer thickness of 50 μm resulting in a seal thickness of 100 μm , and the typical seal width of 1 cm are taken for the calculations. The oxygen and nitrogen permeabilities of LDPE were used as given in [9]. A gas (oxygen and nitrogen) pressure increase of 7 Pa year^{-1} is found to be due to permeation through the sealing seams.

Based on that, a maximum pressure increase of about 3 Pa year^{-1} should be only due to gas permeation through the VIP barrier envelope in order to fulfil the requirements. This corresponds to an oxygen permeability requirement of lower than $2.5 \times 10^{-14} \text{ m}^3(\text{STP}) \text{ m}^{-2} \text{ day}^{-1} \text{ Pa}^{-1}$ at 23 °C and 50% RH for the VIP barrier envelope.

It has to be pointed out that this requirement is a worst case estimation for a water vapour pressure within the VIP being equal to the value in the environment from the beginning. In the case of less severe conditions than 23 °C, 50% RH, where the water vapour

pressure is lower than 1400 Pa, the requirements for oxygen barrier performance could be less. However, in building applications it is recommended to use VIP barrier envelopes with the calculated performance.

Moreover, by using a VIP envelope having also good barrier properties to water vapour, e.g. a water vapour transmission rate of lower than $1 \times 10^{-6} \text{ kg m}^{-2} \text{ day}^{-1}$ at 23 °C and 85% → 0% RH, the increase of thermal conductivity with time can be reduced. This results in a further enhanced thermal insulation performance of the VIP.

Novel energy and cost efficient VIPs with thermal transmittances of lower than $0.2 \text{ W m}^{-2} \text{ K}^{-1}$ and lifetimes of more than 50 years are developed within the European project *NanoInsulate – Development of Nanotechnology-based High-performance Opaque & Transparent Insulation Systems for Energy-efficient Buildings* [7]. Opaque VIPs with nanofoam based cores and translucent VIPs with cores based on silica aerogel polymer composites [10] were developed within this project.

For the opaque VIPs to be developed within the *Nanoinsulate* project, barrier envelopes with oxygen permeabilities and water vapour transmission rates of lower than $1 \times 10^{-14} \text{ m}^3(\text{STP}) \text{ m}^{-2} \text{ day}^{-1} \text{ Pa}^{-1}$ at 23 °C and 50% RH and $1 \times 10^{-6} \text{ kg m}^{-2} \text{ day}^{-1}$ at 23 °C and 85% → 0% RH, respectively, are aimed [7].

In the case of translucent VIPs with aerogel cores the barrier requirements are less strict due to the smaller size of aerogel pores and the resulting higher value of $p_{1/2}$ in Eq. (1) [1]. For aerogel based materials the value of $3.3 \times 10^5 \text{ Pa}$ [11] for $p_{1/2}$ is by a factor of about 5.5 higher than the value $6 \times 10^4 \text{ Pa}$ for fumed silica (Eqs. (1) and (2)). This means that for aerogel based materials, λ_{gas} as a function of the gas pressure increases by the factor 5.5 slower than for fumed silica in the low gas pressure region. Consequently, the requirements for oxygen barrier performance can be reduced in the case of aerogel based VIPs. In order to give exact values, also the thermal conductivity of the evacuated aerogel material, its water vapour sorption isotherm and the effect of absorbed water on its thermal conductivity have to be taken into account.

Aluminium foils would fulfil the required barrier performance for fumed silica based VIPs but they cause a thermal bridge effect when they are used for encapsulation [1,2,5]. In contrast, the barrier properties of standard polymeric films are not sufficient for VIPs by far [1,5,12]. Consequently, their gas and water vapour permeabilities have to be reduced via deposition of barrier layers on top of them [5,12]. Barrier envelopes currently used for VIP encapsulation are produced by vacuum deposition of aluminium layers on top of poly(ethylene terephthalate) (PET) films and lamination of up to three such films together with a polyethylene (PE) sealing film [1,5]. Due to their much lower thickness, the thermal bridge effect for vacuum deposited aluminium layers is significantly lower than for aluminium foils [13]. Laminates containing up to three metallized films can reach gas permeabilities down to $2\text{--}5 \times 10^{-15} \text{ m}^3(\text{STP}) \text{ m}^{-2} \text{ day}^{-1} \text{ Pa}^{-1}$ at 23 °C and 50% RH and water vapour transmission rates of $1\text{--}5 \times 10^{-6} \text{ kg m}^{-2} \text{ day}^{-1}$ at 23 °C and 85% → 0% RH [1] and therefore can fulfil the barrier requirements for the building applications.

In this paper, a novel concept for the development of opaque high barrier films is described. The barrier and mechanical performance of these films and their suitability for the VIP application in buildings are discussed.

2. Concept of novel barrier films for VIPs

One aim of the *Nanoinsulate* project is the development of novel barrier films suitable for VIP encapsulation, which are more cost efficient than current materials. Cost efficient production might be

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