Vapour diffusion calculations through graphite-enhanced polystyrene

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Abstract. Buildings account for almost 40% of total energy use, in the European Union. Energy losses in this sector should be reduced. To reach this, thermal insulation of buildings is a key solution. Polystyrene insulations are ordinary, and their thermal properties are well known. However, the most promising type is the graphite enhanced one with relatively less information. These graphite doped material products have much better thermal insulation properties than the conventional (white) ones. This paper will present measurements of sorption isotherms, combined with kinetic vapour experiments performed in a climatic chamber. Based on the results, the identification of the sorption isotherm will be shown. Moreover, the other scope of the paper is to apply a theoretical model used for diffusion after immersion to calculate the vapour diffusion in the samples treated in a climatic chamber. Furthermore, grain size analysis will be executed from microscope images, too.

INTRODUCTION

Thermal insulation is a very important action in both the building and vehicle industries. The ever-strictness of thermal engineering requirements and the increasingly energy-conscious needs of investors are creating products with more favourable thermal insulation parameters. The goal is to achieve the required thermal level with the possible thinnest layer thickness. Thicker thermal insulation also requires more attention to construction, and may also impose architectural constraints on the designer, e.g. the shape of the building may change, the proportions may be distorted, and the doors and windows will be shaded. In the case of renovations in addition to new buildings, it is necessary to adapt to the existing structural conditions, so the lack of space affecting the thickness of the thermal insulation is exaggerated. There are also financial benefits from using better thermal insulation: greater built-in costs, and lower logistics costs (transport, material handling, storage) [1-4].

Wetting properties of thermal insulation materials are important characteristics from the stability of thermal performance point of view. Sorped water in gas/vapour, liquid or solid form can cause unexpected changes in both thermal properties and the physical parameters of the sample. A key graph for understanding the wetting characteristics of the sample is the sorption isotherm curve (relative humidity vs equilibrium moisture content at a certain temperature) [5]. The two frequently applied sorption isotherm graphs are the Langmuir and the Brunauer – Emmet -Teller (BET) type ones (see Fig. 1). These two sorption characteristics are the most important ones, however other shapes might happen but these two uniform ones. As the presentation of them from 0 % relative humidity till about 40 % both shows a square root type function, this means that firstly during the water up-taking the adsorption process at the surface takes place (monolayer part), however after this with increasing relative humidity value the BET isotherm shows a continuously increase while the Langmuir ones showing a constant equilibrium moisture content independently from the relative humidity. The curves are plotted at constant temperature by measuring the equilibrium moisture content of specimens dried to constant weight in the air at a given temperature and relative humidity. The horizontal axis of the sorption isotherm shows the relative humidity of the air in contact with the specimen and the vertical axis the moisture content of the material (expressed as a percentage by weight or volume), so the sorption isotherm curve expresses the relationship between the relative humidity and the moisture content of the material. The curves usually have an inflexion point where the slope of the curve suddenly increases. This point marks the beginning of capillary condensation when moisture no longer covers only the surface of the pores but begins to fill the entire cross-section of the capillary passages. This is the so-called capillary condensation, which in the case of materials used in the construction industry usually develops at a relative humidity of $\varphi = 75\%$. Capillary action can occur with any building material where there are defects in the material, such as pores, larger cavities or free surfaces. Pores can account for up to 80% of the volume of materials that can be counted in masonry, where moisture can penetrate into building materials [6, 7]. The connection between the vapour and liquid transport can be the capillary condensation, where the phase change from vapour to liquid state happens. Open pores fill with water when exposed to moisture, the easier it is to do this, the larger the pore. Water can seep into the rest of the building material due to the force of gravity. Under the influence of external pressure, water moves in the capillary pores according to specific laws, even against the force of gravity. Moreover, the vapour diffusion can take place also by the pressure difference [8-12].

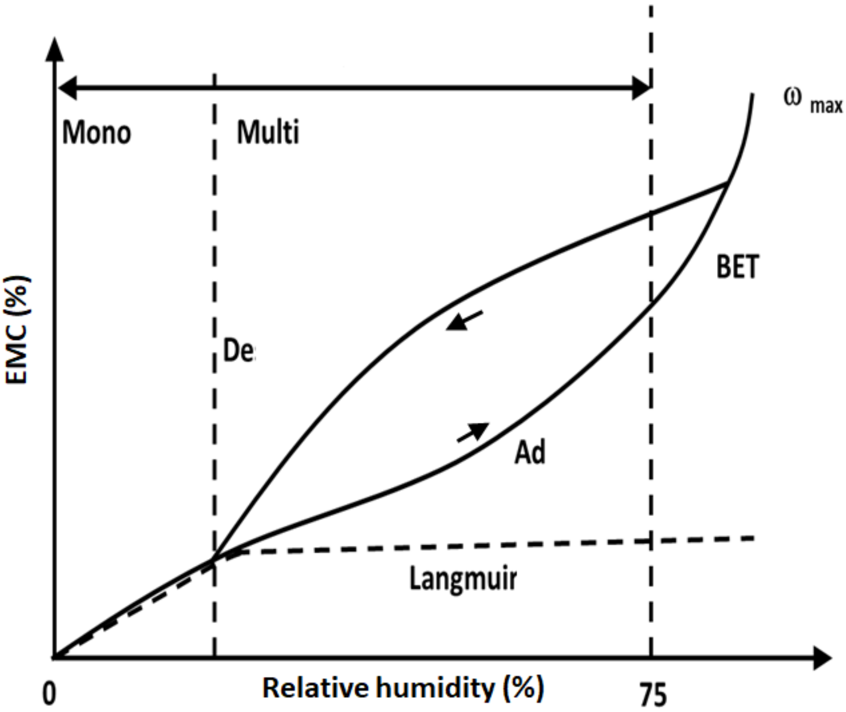

FIGURE 1. Typical sorption isotherm curves

MATERIALS AND METHOD

During the examinations, we investigated graphite-enhanced expanded polystyrene as promising insulation for common use. The addition of graphite grains improves the thermal reflectivity of the product, resulting in nearly 15% or even 35% better thermal insulation properties than conventional white EPS and fibrous insulation materials. Its other material properties are the same as those of a normal white EPS product (eg flammability, dimensional stability, strength characteristics) except the moisture uptaking. In a previous paper, it is presented that under the same humid circumstances graphite EPS takes up more water than conventional white ones. It is caused by its graphite/carbon content which is said to be a good getter material having a relatively high specific surface to absorb moisture [13]. This also gives a reason for executing laboratory examinations on this material.

Due to the dark colour of the product, it must be protected from direct sunlight and UV radiation, and excessive heating and cooling during storage, transport and use.

The density of the EPS samples strongly depends on the length of the graphite EPS beads and it has a strong connection with the macroporosity (the gap among the beads) which drives the moisture adsorption, too. The tested material during the measurements is graphite enhanced expanded polystyrene having 16-17 kg/m^3 density. To see the distribution of the length of the beads, an analysis with a microscope was executed and the results are presented in Figure 2. One can see the Gaussian distribution of the length of the beads, with about a 12 mm mean value.

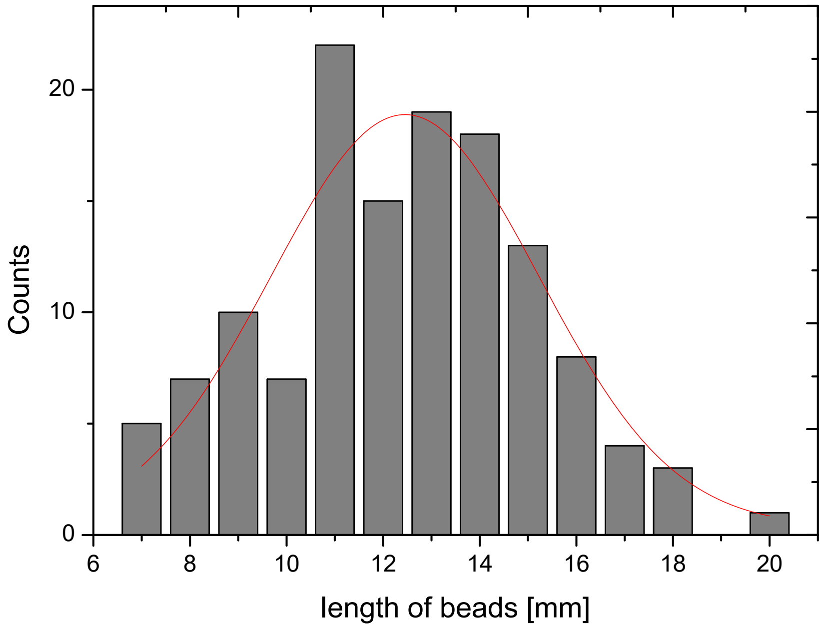

FIGURE 2. The length of beads

The circumstance of the wetting measurements is written in the previous papers of the Author. For the measurement of the moisture/water up-taking phenomenon of materials by the climatic chamber method, using the ISO 12571: 2013 standard, three apparatus must be combined. A dryer or a desiccator (eg: Venticell 111), a climatic chamber (Climacell 111) and a (milligram preciseness) weighing scale (see Figure 3). Four samples with 10 cm x 10 cm x 4 cm geometries were tested. Before wetting the samples, they were cured to constant mass and their mass was measured. After desiccating, probes were placed in a humidity (incubator) chamber under different relative humidity for different times and their wet mass was also measured. In our context, it was executed in a ClimaCell 111 instrument. From these mass values, the moisture content was calculated [5, 10-14]. For graphite-enhanced polystyrene materials, the equilibrium moisture content was measured and presented in the function of the relative humidity (0 to 90%) at 23 ^{o}C. In this paper, these results were used for calculating the vapour diffusivity with two theoretical approximations originating from liquid transport theory [5, 15, 16].

FIGURE 3. The applied equipment

Graphite-containing EPS is a special grey polystyrene thermal insulation board, which, due to its special base material, has a significantly lower thermal conductivity than the white version and already has sound-insulating properties, but takes up more moisture. This graphite polystyrene foam sheet is successfully used in the construction industry, where thermal conductivity is approximately 20% smaller due to the addition of nanoscale graphite powder. It has a thermal conductivity ($\lambda = 0.032$ W / mK) compared to EPS plates without graphite ($\lambda = 0.04$ W / mK).

THEORY

Diffusivity theory

In Ref. 15, 17 and 18 the Authors deduced liquid diffusivity from moisture absorption, but now the theory is applied for vapour diffusion, the theory is as follows.

$$J_m = -D_v \times grad(C_m) = -D_v \times \frac{\partial C_m}{\partial x} \tag{1}$$

where J_m is the rate of a mass transfer per unit area of section (kg/m^{2}s), C_m (kg/m^3) is the volumetric moisture concentration and D_v is a material-specific diffusion coefficient (m^2/s). In the downer equation, ω (kg/kg) is the moisture content and ρ_{dry} is the dry density [5, 15, 17 and 18].

$$J_m = -(\rho_{dry} \times D_v \times (grad(\omega) = -(\rho_{dry} \times D_v) \times \frac{\partial \omega}{\partial x} \tag{2}$$

Here C_m can be substituted with the multiplication of the dry density of the material with the ω (kg/kg) moisture content. A scientific work presented by Ref. [15] gives a simplification to reach an average liquid diffusivity from the water absorption coefficient. After this, we would like to deduce a vapour diffusivity value from the sorped amount of moisture in a climatic chamber. Using Eq. 3 presented in the latest work of the author [11] one can find a method for estimating the vapour sorption coefficient of the samples by interrupted moisture uptake at a fixed humidity and constant temperature. With the downer equation (Eq. 3) from the saturated volumetric moisture content of the material (w_c, m^3/m^3) one can evaluate an average diffusivity coefficient:

$$D_{v2} = \frac{\pi}{4} \times (\frac{A_v}{w_c})^2 \approx D_{v1} = (\frac{A_v}{w_c})^2 \tag{3}$$

RESULTS AND DISCUSSION

Firstly, the paper presents the results of sorption measurements executed on the graphite EPS samples, the equilibrium moisture contents were calculated from the average of the results of the measurements of four samples and we can conclude that their results did not vary significantly. In Figure 4 one can see the BET type II isotherm, for the samples. This process shows higher interaction between the adsorptive and adsorbent than between the adsorptive and adsorbate. Type II is most frequently encountered when adsorption occurs on non-porous powders or no powders with pore diameters larger than micropores. The inflexion point of or knee of the isotherm usually occurs near the completion of the first adsorbed monolayer and with increasing relative pressure, second and higher layers are completed until at saturation the number of adsorbed layers becomes infinite. In Figure 4 one can see a continuous increase in the moisture content to 50 % relative humidity and after a relaxing state, the EMC further increases.

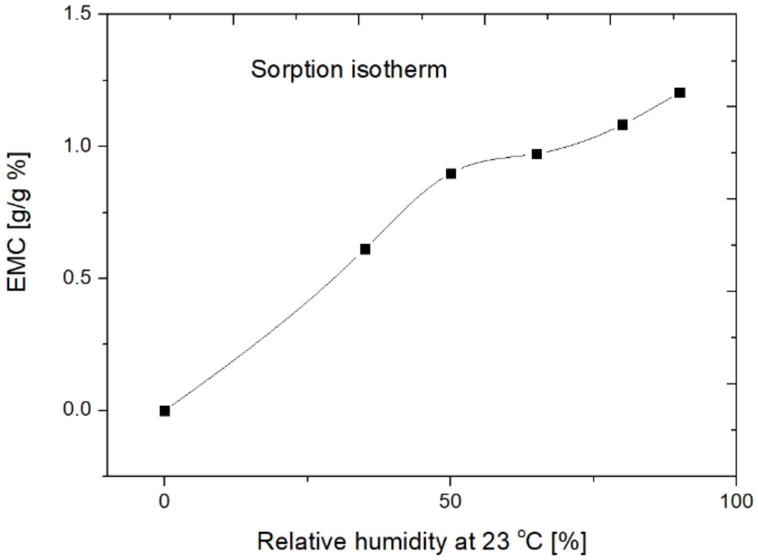

FIGURE 4. The equilibrium moisture content vs. relative humidity

Both the above-mentioned values such as moisture contents and the measured A_v values are highlighted in Table 1. Moreover, in this table, the diffusivities calculated by using Eq. 1-3 are also presented.

Table 1. The measurement and calculation results.

Reletive humidity (%) at 23 °C	**Pressure (Pa)**	**Av [kg/(m²*s¹ᐟ²)]**	**Equilibrium moisture content (kg/kg%)**	**Volumetric moisture content [m³/m³]**	**Dv1 [m²/s]**	**Dv2 (× π/4) [m²/s]**
35	980	0	0	0	0	0
50	1400	0.01	0.614	0.382	1.46E-05	1.15E-05
65	1820	0.014	0.899	0.56	5.98E-05	4.69E-05
80	2240	0.0175	0.973	0.61	1.12E-04	8.80E-05
90	2520	0.018	1.083	0.674	1.47E-04	1.16E-04

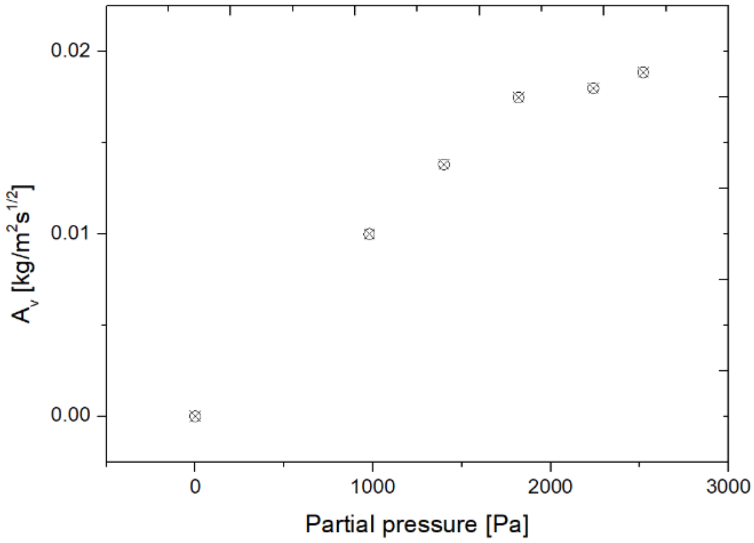

FIGURE 5. The A_v values

Figure 5 represents the increase in the A_v values in function of the partial pressure (relative humidity). The partial pressure was calculated with the saturation pressure (2800 Pa) at 23 ºC. Between 0 and 2000 Pa a continuous strong increase while, after 2000 Pa a slight increase is visible. These values were estimated from the initial water up-taking process, before reaching the sample the equilibrium moisture content at different relative humidities.

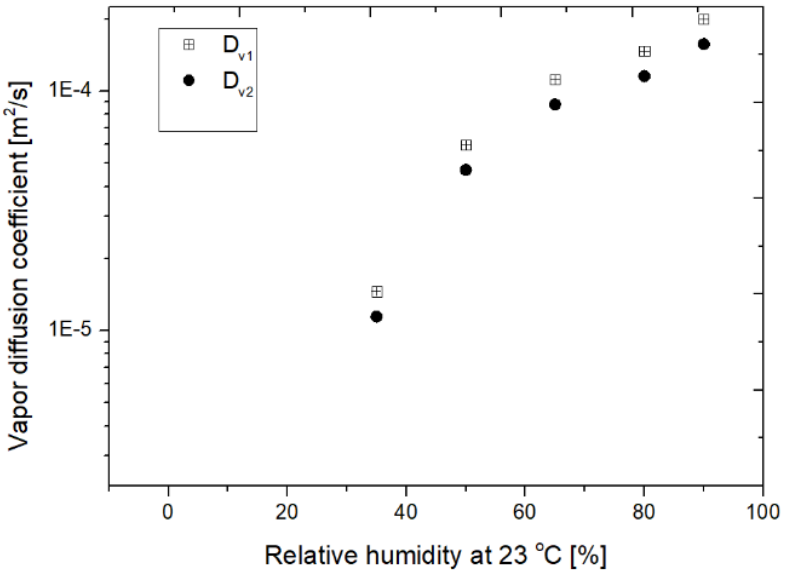

FIGURE 6. The estimated vapour diffusivities

In Fig. 6. one can see the varying D_v values in function of the partial pressure as the results of the calculations. One could observe that the diffusivities are ranging between 10^{-5} and 10^{-4}, moreover, for 90% relative humidity, the diffusivity increases with an order of magnitude compared to the value at 35% rh. As we see, with increasing pressure as the driving force, the diffusivity is increasing. The values are under the values presented in Ref. [19] for white EPS materials since these values represent faster diffusion. Upon this, we can conclude that through graphite-enhanced EPS the diffusion is faster than the white one.

CONCLUSIONS

This paper presents moisture-related investigations executed on graphite-enhanced insulation materials. During the measurements, the properly dried samples were measured in a device designed for sufficiently high moisture content of 30-95% humidity load at 23 ° C for equilibrium. The measurements were performed in the Building Physics Laboratory of the Faculty of Engineering of the University of Debrecen, where I got a fairly accurate picture of the moisture uptake process of graphite-enhanced polystyrene insulation material. In the first step, I determined the A_v specific vapour uptake data, and from this, the vapour diffusion coefficients were estimated through a model applied for liquid transport. We could declare that the vapour diffusivities are ranging between 10^{-5} and 10^{-4} m^2/s which represents faster diffusion than in white EPS, moreover for 90% relative humidity the diffusivity increases by an order of magnitude. Furthermore, we have characterized the sorption isotherm curve of the material, too. The paper dealt with graphite eps insulation materials because it is very popular in the construction industry. This insulating material has many advantages. A first significant advantage is that it is cheaper than other types of insulating material such as aerogel or vacuum insulation panels. The second advantage is low density and weight, thus placing a small load on the outer wall. Moreover, it has 20-30% less thermal conductivity than the conventional white EPS. But, from a humidity point of view, this material should be handled with care.

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