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Alternative low cost based core systems for vacuum insulation panels

Flávia A. Almeida[a,b], Jorge Corker[a,b,*], Nuno Ferreira[c], Miguel A. Neto[d], Mizi Fan[e], Hermann Beyrichen[f], Roland Caps[f]

[a]*Laboratory of Wear, Testing and Materials (Led&Mat), Instituto Pedro Nunes. Rua Pedro Nunes, Coimbra 3030-199, Portugal*
[b]*SEG-CEMMPRE – Centre for Mechanical Engineering, Materials and Processes; University of Coimbra, Rua Luís Reis Santos, 3030-788 Coimbra, Portugal*
[c]*I3N, Department of Physics, University of Aveiro, 3810-193 Aveiro, Portugal*
[d]*CICECO, Materials and Ceramic Eng. Dept., University of Aveiro, Campus de Santiago, P-3810-193 Aveiro, Portugal*
[e]*Department of Civil Engineering, Brunel University, UB8 3PH, UK*
[f]*va-Q-tec AG, Karl-Ferdinand-Braun-Str., 7, Würzburg D-97080, Germany*

Abstract

Vacuum Insulation Panels (VIP) are presently regarded as one of the most promising state-of-the-art building insulation solutions. Based on their thermal conductivities of about 4 mW/(m K), with a thickness below 40 mm, they have a great potential for near zero-energy buildings (nZEB) and for applications where high insulation standards and living space savings are crucial. However, VIP are still unaffordable for the majority of homeowners and contractors (up to 100 €/m^2), mostly due to the cost of the conventional fumed silica used as core material to secure the long service life requirements of building applications. This study presents the early developments of alternative cores engineered for VIP targeting the building market. The adopted strategy is to replace fumed silica with cheaper natural inorganic/organic lightweight materials or, alternatively, by creating multimaterial nanostructured composite matrices. The different compositions were analysed according to their physical, chemical and morphological characteristics and their respective thermal conductivity ranks. Promising lambda values as low as 5.3 mW/(m K) have been achieved for gas pressures below 10 mbar (1 kPa). It is expected that these novel core systems will be capable of suppressing the different heat transfer mechanisms at more reasonable costs than the current VIP fumed silica ones.

Keywords: Vacuum insulation panels; core materials; thermal conductivity; near zero-energy buildings.

1. Introduction

Currently about 40% of the European energy consumption is for buildings and building related activities, mostly related to inefficient building insulation systems. Capable of reaching outstanding thermal conductivities around 4 to 5 mW/(m K) (5 to 10 times better than any conventional insulation available), the state-of-the-art Vacuum Insulation Panels (VIP) are especially attractive solutions when trying to achieve the standards and requirements of the latest European Energy Directives and H2020 objectives towards near zero-energy buildings [1]. Such low thermal conductivity ranks are mainly achieved by suppressing the gas conduction heat transfer mode inside an open porous core material, which is evacuated and sealed within an air and vapour tight envelope barrier. One of the most used VIP core material is glass fibre, especially amongst Asian manufactures. Although capable of reaching lambda values, at pristine conditions, as low as 1.5 mW/(m K), glass fibre cores must be evacuated well below 0.1 mbar (10 Pa) to reach such value [2]. However, having relative coarse pore sizes, glass

* Corresponding author.
E-mail address: jcorker@ipn.pt (Jorge Corker)

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fibres experience similar problems as open cell polymeric foams, both unable to suppress gas conduction at moderate vacuum levels, making it more suitable for packaging and appliance applications due to their short service life (15 years maximum) [2].
On the other hand, nanostructured fumed silica is the most used VIP core material for building applications [3]. Due to its nano-sized pore range (30 to 100 nm) and with the help of opacifiers, one should anticipate lambda values between 4 to 6 mW/(m K) under a pressure of 20-100 mbar (2-10 kPa) and, even at non-evacuated conditions, values around 18-20 mW/(m K) can still be expected [2]. Although commercially available, the price of fumed silica VIP is still very high (60-90 €/m^2), mostly due to the cost of the core material. With the majority of conventional insulation offered below 10 €/m^2 and when Europe still faces the aftermath of a deep economic crisis, the only way for VIP to step out from a high-priced niche market is by having its price cut considerably. Under this scope, many research works have been testing diverse core solutions along with fumed silica, such as perlite [4-6], diatomite [4], pumice [7] and fibre-powder composites involving glass and organic cellulose fibres [4,6-8]. All materials presented some drawbacks, mainly related with their high density and/or their inability to minimize the gas conduction influence due to their large pore size. The aim of the proposed work is to select and preliminarily evaluate alternative core materials towards the generation of novel cost-effective VIP building solutions. Based on natural and sustainable materials, mainly in the form of industrial residues, multi-level hybrid structured cores (Fig. 1) were envisaged, by partially or fully replacing the fumed silica content, allowing to cut considerably the raw materials production costs.

2. Materials and Methods

2.1. Core materials and definition of blends

Commercial fumed silica (FS - Aerosil©) was used for comparison and mixing purposes, together with two low price residue materials which have been selected to create the alternative hybrid VIP cores:

- An organic wood-based powder (WM);
- A natural lightweight siliceous mineral powder (LM), with two distinct average particles sizes, referenced as LM-1 and LM-2.

The market price of FS is 4 €/kg while for the two by-products, it is lower in about 10 to 200%; 0.02 €/kg for WM and 0.30 €/kg for LM.

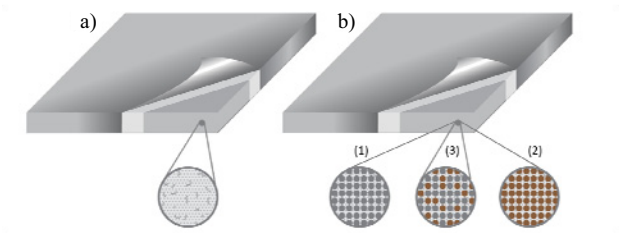

Fig. 1. a) Conventional VIP cores for building applications, made with expensive fumed silica powders, b) Alternative low cost VIP cores solutions, made with cheap lightweight minerals (1), organic by-products from renewable resources (2), or by doping it with fumed silica and creating novel structured hybrid core solutions (3).

VIP of pure compositions of each material were also produced and characterized. At the same time, a set of preliminary blends were selected to represent the novel core systems, without any getters, desiccants or opacifiers:

- 90%LM-1 + 10% FS
- 80% LM-2 + 20% FS
- 80% WM + 20% FS
- 50% LM-2 + 50% FS
- 50% WM + 50% FS

2.2. Raw materials characterization

Raw materials phase identification was determined by X-ray diffraction, using a Co tube Philips-MPD diffractometer. The chemical nature of the mineral samples was evaluated by X-ray fluorescence spectroscopy (PANanalytical Axios^{mAX} WDXRF model) and the organic material characterized by Fourier transform infrared spectroscopy (FTIR), using a Perkin Elmer Spectrum 100. The morphology of the candidate core particles and of the hybrid mixture structures was observed by Scanning Electron Microscopy (SEM) in a JEOL JSM5600LV and a Hitachi SU-70. Complementary characterization included: particle size distribution by laser diffraction (Malvern Mastersizer 3000), pore size distribution by Hg porosimetry (Micromeritics AutoPore IV), real density by He-pycnometry (Micromeritics AccuPyc) and specific surface area and pore size evaluation by N_2 adsorption (Micromeritics ASAP 2000). Initial moisture and drying behaviour analysis was conducted by termogravimetry (TG) with a Setaram Setsys Evolution from 25ºC to 170°C at 3°C/min. Finally, moisture adsorption evaluation was made at 25ºC with a DVS-Advantage equipment, following sorption-desorption stage profiles between 0-95%.

2.3. VIP trials production, gas pressure assessment and thermal conductivity evaluation

Pure powders and their different blends were prepared by mechanical mixture, before being dried at 90ºC for 24h. Afterwards, materials were poured inside plastic bags and placed in special moulds to be uniaxially compacted at 1.0 MPa, creating regular shaped VIP boards (≈1.5 cm thick) with densities ranging 0.18-0.20 g/cm^3. The boards were placed inside U-shaped commercial multi-laminated metalized PET foils and evacuated inside a vacuum sealing machine. To study the influence of gas pressure on thermal conductivity, all samples were sealed at different pressures, checked either visually by the lift-off method or by using internal va-Q-check sensor [9]. Thermal conductivity evaluation was made using an in-house built equipment dedicated for VIP testing and following ASTM C518 standard as guideline, where two precision sensors were placed in each side of the testing panel to measure heat fluxes across the sample thickness at 30ºC/45ºC, in steady state conditions.

3. Results and Discussion

3.1. Raw materials characterization

XRD patterns (not shown) attested the amorphous nature of all materials under study, while presenting characteristic broadened SiO_2 bands for the inorganic materials and a pronounced amorphous halo for WM powders due to its bio-polyester content, as confirmed by FTIR analysis (not shown). On the other hand, WDXRF attested the siliceous nature of the LM materials and have revealed an additional alumina predominance, with SiO_2 (74.86 wt.%) and Al_2O_3 (14.04 wt.%) being the most abundant oxides registered. FTIR evaluation of the WM samples also showed that no significant structural changes occurred to the WM powders after being dried at 180ºC for 30 min. This is important since any moisture and volatile substances must be removed from the core to reach high vacuum levels during the evacuation process. TG evaluation confirmed that most of the weight loss registered by all samples occurred up to 120ºC, thus mainly related to natural moisture in about 5 wt.% for WM, 4 wt.% for FS, 0.4 wt.% for LM-1 and 0.9 wt.% for LM-2. Complementary, moisture vapour sorption results (Fig. 2) have shown that all materials exhibit very distinct behaviours. While LM presented by far the smallest total water uptake (0.17%, inset graph of Fig. 2), both WM (7.68%) and FS (23.11%) showed much higher moisture sorption ranks, indicating bulk absorption features. The higher sorption capability of FS is known to be due to its huge specific surface area given by its nanometric particles sizes [10]. Fig. 2 shows a very small hysteresis between the sorption and desorption cycles of FS, suggesting a reversible sorption mechanism, while the sudden exponential increase of its DVS profile above 60% is well known and mainly related to capillary condensation in the FS nanometric pores [10]. Contrarily, the isotherm plots of LM and WM present open hysteresis gaps, indicating that desorption from pores is very slow and often not completed. This behaviour shows that it will be highly advisable to use getters/desiccants with both alternative materials, especially for the WM compositions, in order to counteract and delay the moisture ageing effects due to permeation through the envelope foils [3].

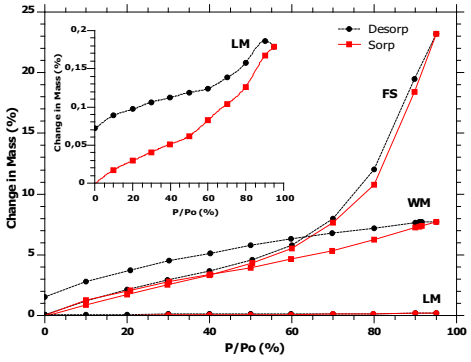

Fig. 2. Dynamic Vapour Sorption plots of the raw materials.

Particle size distributions of all materials are given in Fig. 3, with respective SEM micrographs in Fig. 4. Particle size and shape are especially important for the solid skeleton heat conduction contribution, which is mainly determined by the effectiveness of energy transfer between grains [11]. In Fig. 3 it is possible to observe that, not only WM presented the largest particle size distribution of all materials (D_{50} 150 µm), but both LM powders were still well inside the micrometre range.

SEM analysis of WM particle structure revealed that this residue is formed by irregular shredded fragments containing small cavities (Fig. 4 a)). Between the two LM types, LM-1 presented its median peak centred at higher values, roughly around 40 µm, while LM-2 at about 20 µm. This size difference was corroborated by SEM observations (Figs. 4 b) and c)), also showing that both are formed by tiny irregular shaped plates with walls thickness ranging from 300 nm to 1 µm.

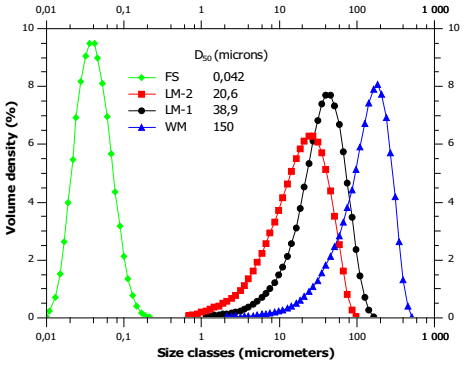

Fig. 3. Particle size distributions of the natural raw materials.

As expected, FS registered the smallest particle size distribution (Fig. 3), being entirely made by aggregates of spherical particles with 40 to 70 nm (Fig. 4 d)). Its nanometric nature is also responsible for the high surface area presented (263.47 m^2/g), two orders of magnitude above those of WM and LM as shown in Table 1, where other relevant features of all materials can be found.

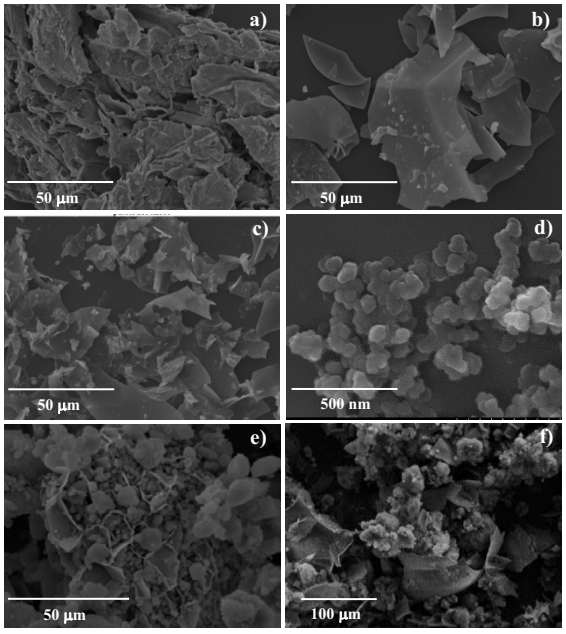

Fig. 4. SEM micrograph of a) WM residues, b) LM-1, c) LM-2, d) FS powders, e) 50% WM + 50% FS, f) 50% LM-2 + 50% FS.

Low bulk density core materials are also desirable to minimize solid conduction per square meter of the porous material [12]. Moreover, spherical nanosized materials with point-like contacts interrupting the heat flow on the nanometric level tend to be less influenced by solid conduction. On the other hand, denser and coarser granular materials having well conducting grains, like LM and WM, are likely to have an inferior insulation behaviour [12]. Nonetheless, following the bulk densities of LM and WM powders (Table 1), it's reasonable to anticipate the production of VIP cores around the desirable range of 150-220 kg/m^3 [3,6]. Table 1 also shows that all powders presented high levels of open porosity, one of the main criteria for VIP cores [13]. In this context, FS have clearly set apart for having the highest porosity level (86.7%) and by registering the lowest median pore diameter (7.20 µm). In fact, contrarily to the LM and WM powders, Hg porosimetry did not properly represents the pore size distribution of FS, since a major part of it was expected to be inside the nanometric range. This was accurately confirmed by N_2 adsorption analysis which registered the FS pore size distribution centred on 25 nm.

Table 1. Loosely powders densities, porosity and surface area characterization.

Raw materials	Real density (g/cm^3)	Bulk density (g/cm^3)	Median pore diameter (µm)	Open porosity (%)	Surface area (m^2/g)
WM	1.18	0.13	25.0	80.0	1.3
LS-1	1.22	0.17	18.7	72.8	1.3
LS-2	1.17	0.18	9.1	72.2	2.4
FS	1.94	0.075	7.2	86.7	263.5

3.2. Thermal conductivity of the core candidates in function of internal gas pressure

Figs. 5 a) and b) present the thermal conductivity evaluation in function of air pressure for LM and WM hybrid blends, as well as for pure compositions, including pure FS for comparative purposes. The most noticeable aspect shown is the high different thermal behaviour profiles of the pure core candidates compared to FS. This is due to the nanometric pore size range of the latter being on the same order of magnitude to the mean free path of air molecules [13]. The pronounced rising of FS thermal conductivity only starts for pressures around 100 mbar (10 kPa), determining a much smaller gas pressure dependency of this material. Conversely, the larger pore sizes of all candidate powders clearly provide the conditions towards a significant contribution of gaseous conduction even at just a few mbar. For instance, at 1 mbar (0.1 kPa), the WM lambda value (22 mW/(m K)) represents more than 60% of its pristine value, while lower differences were found for LM-1 and especially for LM-2, the latter still able to secure an interesting score (8.6 mW/(m K)). Also, while FS lambda values are always below air thermal conductivity (26 mW/(m K)) over the full pressure

range, all candidates proved to be much more sensitive to envelope's air permeation and have inferior insulation performance in the case of a barrier failure, registering much higher lambda values at 1 atm (0.1 MPa), from 44 to 58 mW/(m K). In order to minimize the cost of VIP core by using FS powders and the strong gas conduction experienced by the candidate materials, hybrid core systems were created with FS, as described in section 2. Test results proved that, especially for LM-2/FS mixtures (Fig. 4 f)), a substantial insulation improvement was attained when comparing with pure LM cores, even surpassing the ones for pure FS for pressures below 0.6 kPa (the case of the 50% LM-2/FS blend, Fig. 5 a)). This particular behaviour was possibly due to the fact of LM being less permeable to infrared radiation than FS, although such statement is to be confirmed in on-going work.

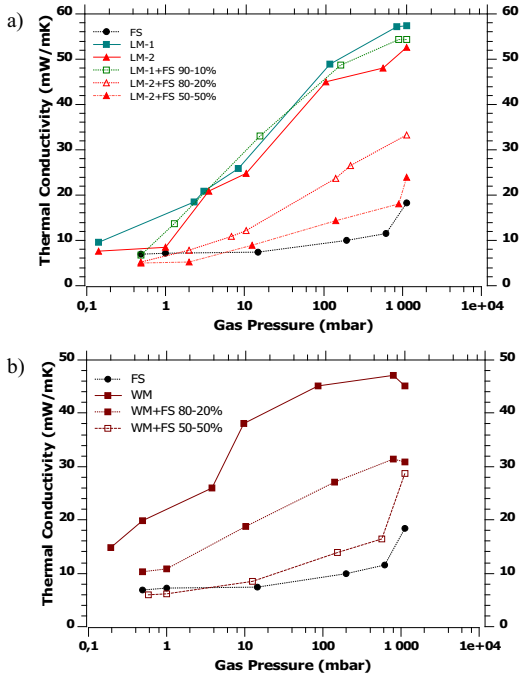

Fig. 5. Thermal conductivity in function of gas pressure of the lightweight mineral (LM) and wood-based material (WM) mixtures with fumed silica (FS).

Fig. 5 b) shows that the thermal conductivity of the WM material at 0.4 mbar (40 Pa) could be lowered in about 50% by adding just 20% of FS, while reaching a remarkable 70% improvement for mixtures having a 50% of FS content. These results suggest that by filling the large pores of WM cores with FS nanoparticle aggregates (Fig. 4 e)), it was possible to largely reduce gas conduction contribution experienced by the pure WM cores along the full gas pressure range. This evidence is especially interesting up to 10 mbar (1 kPa) in the case of a 50% blend, which showed a similar performance as the one registered by the 50% LM-2/FS hybrid core mixture (Fig. 5 a)).

The cost effectiveness of the studied core solutions was estimated taking into consideration the materials costs (€/kg) and panel's density (kg/m), as specified in section 2, and their initial thermal conductivities (W/m K, Fig. 5). The comparison among the distinct specific costs, in € W/m^4 K, can be visualized in Fig. 6 for all compositions. It is clear that a core cost reduction potential of at least 50% or more can be envisaged by using the proposed by-products studied in this work.

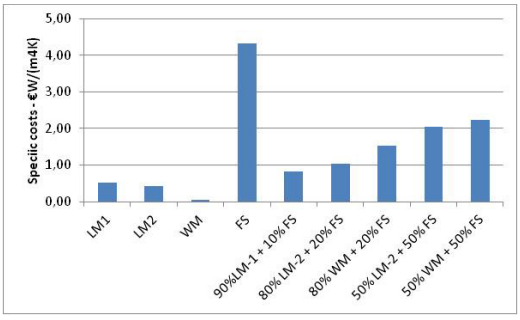

Fig. 6. Core cost estimation of the pure and the blended composites of lightweight mineral (LM), wood-based material (WM) and fumed silica (FS).

Finally, the results presented in this work attest that, by filling the large pore and voids present on LM and WM cores with nanosized FS powders, air conduction contribution can be strongly attenuated together with a potential core cost reduction. Nevertheless, in order to properly address its viability, other issues have to be fully evaluated, such as: the thermal conductivity variation with time, the final density of the panels, the right mass ratio of fumed silica and the need for opacifiers, getters and dryers to secure a long service life. Moreover, the thermal performance of VIP panels degrades with time as moisture and gas permeate through the barrier envelope. In this sense, the selection of the right barrier envelope by means of accelerated experiments is also being investigated, towards meeting the strict requirements of long-term building applications.

4. Conclusions

An initial evaluation of novel low cost hybrid core VIP solutions for the building sector has been carried out through experimental work, based on wood residues and siliceous lightweight powders, along with

controlled fumed silica content. The results showed that cores having a 50-80% content of these alternative materials can present thermal conductivities close to those of pure fumed silica up to gas pressures around 10 mbar (1 kPa) and a preliminary cost effectiveness of more than 50%. The outcomes of this work suggest a good potential upon the usage of these alternative powders to create less costly VIP with good insulating properties.

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References

[1] B.P. Jelle, Energy and Buildings 43 (2011) 2449.
[2] S.E. Kalnaes, B.P. Jelle, Appl. Energy 116 (2014) 355.
[3] H. Simmler, S. Brunner, Energy and Buildings 37 (2005) 1122.
[4] M. Davraz, H.C. Bayrakci, Sci. Eng. Compos. Mater. 21 (2014) 1.
[5] R. Caps, J. Fricke, Int. J. Thermophys. 21 (2000) 445.
[6] M. Alam, H. Singh, S. Brunner, C. Naziris, Energy and Buildings 69 (2014) 442.
[7] P. Mukhopadhyaya, M.K. Kumaran, N. Normandin *et al.*, J. Cold Reg. Eng. 22 (2008) 103.
[8] K. Araki, D. Kamoto, S.-ich Matsuoka, J. Mater. Process. Technol. 209 (2009) 271.
[9] R. Caps, H. Beyrichen, D. Kraus, S. Weismann, Vacuum 82 (2008) 691.
[10] H. Keskinen, S. Romakkaniemi, A. Jaatinen *et al.* Aerosol Sci. Technol. 45 (2011) 1441.
[11] M. Tenpierik, PhD thesis, Technische Universiteit Delft, The Netherlands, 2009.
[12] J. Fricke, Proceedings of 7th International Vacuum Insulation Symposium, Zurich-Duebendorf, Switzerland, September 28-29, 2005, pp. 5-14.
[13] R. Baetens, B.P. Jelle, J.V. Thue *et al.*, Energy and Buildings 42 (2010) 147.