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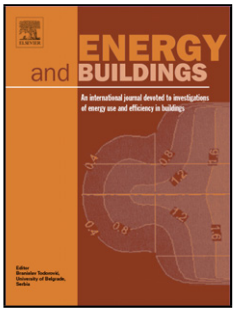

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Cellulosic-crystals as a fumed-silica substitute in vacuum insulated panel technology used in building construction and retrofit applications

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Abstract

This article investigates impact of substituting fumed silica with a cellulosic-crystal innovation in a commercial Vacuum Insulated Panel (VIP) core. High building performance demands have attracted VIP technology investment to increase production capacity and reduce cost. In building retrofit VIPs resolve practical problems on space saving that conventional insulations are unsuitable for. Three challenges exists in fumed silica: *cost*, *low sustainability properties*, and *manufacture technical maturity*. Cellulosic nano-crystal (CNC) technology is in its infancy and was identified as a possible alternative due to a similar physical nano-structure, and biodegradability. The study aim was to determine a *performance starting point* and establish how this compares with the current benchmarks. Laboratory cellulosic-crystal samples were produced and supplied for incorporation into commercial VIP manufacture. A selection of cellulosic-panels with core densities ranging 127 – 170 kg/m^3 were produced. Thermal conductivities were tested at a pressure of 1 Pa (0.01 mBar), with the results compared against a selection of fumed silica-VIPs with core densities ranging 144 – 180 kg/m^3. Conductivity tests were then done on a cellulosic-VIP with 140 kg/m^3 density, under variable pressures ranging 1 – 100,000 Pa (0.01 – 1000 mBar). This investigated panel lifespan performance, with comparisons made to a fumed silica-VIP of similar core density. Manufactured cellulosic-samples were found unsuitable as a commercial substitute, with performance below current standards. Areas for cellulosic nano-material technology development were identified that show large scope for improvement. Pursuit could create a new generation of insulation materials that resolve problems associated with current commercial versions. This is most applicable in building retrofit where large ranges of domestic and commercial cases are marginalised from their construction markets due to impracticalities and high upgrade costs. This being a problem in multiple economies globally.

Key Words: Vacuum Insulated Panels, Building Insulation, Retrofit, Nano-technology

List of variables

λ_r	*Radiation conductivity component in insulation (W/m.K);*
λ_{ss}	*Solid state thermal conductivity in insulation (W/m.K);*
λ_g	*Gas conductivity component in the pores of the insulation (W/m.K);*
λ_{coup}	*Coupling effect impact between gas molecules and matrix (W/m.K);*
λ_{tot}	*Total of all single effects and effective thermal conductivity in VIP application (W/m.K);*
λ_{gas}^0	*Gas thermal conductivity at ambient pressure (W/m.K);*
p_{gas}	*Pressure inside the VIP (Pa);*
$p_{1/2}$	*A material parameter mainly dependent on the pore size of the VIP matrix (Pa);*

1.0 Introduction

Vacuum insulated panel (VIP) technology permits the specification of low thermal U-value elements in buildings. Research and development has been extensive, with reviews covering these including: Simmler *et al.* [1], Fricke *et al.* [2], Baetons *et al.* [3], Alam *et al.* [4], Fuchs *et al.* [5], Johansson [6], Kalnæs and Jelle [7], and Brunner *et al.* [8]. VIP applications are numerous, with example studies including: building retrofit [9], district heating [10], thermal storage [11], and solar energy [12]. The technology is comprised of a fumed silica based core coated in a sealed evacuated aluminium based film. Rigidity of the centre is maintained using small quantities of micro-fibres for shape retention, combined with infrared opacifier for radiation transfer suppression (figures 1 and 2) [13]. Coating films are typically made of polymers, including: Polyethylene Terephthalate [PET], Polyethylene [PE], and Polypropylene [PP], these alternated with metal layers (Al, Steel, AlOx, SiOx) that prevent gas penetration into the core [13-15]. A cross-section of a VIP film envelope is shown in Figure 3 with typical layer thicknesses [1, 13].

VIP thermal conductivities are five to ten times lower than commercial insulations, with the centre-of-panel reaching 4 mW/m.K [3, 4, 14, 16]. Albeit excellent performance, the technology is expensive with panel prices multiples of five to ten that of standard insulations [17, 18]. Perspective is given considering space saving provided in a standard solid brick construction aiming for a 0.28 W/m^2.K U-value; where a reduction from 100mm (mineral fibre or EPS) to 40mm for VIP can be achieved [3]. In light of cost, the technology is mainly used in niche applications where no alternatives are possible, or where space saving is at a premium [5, 10]. Should a cost-effective solution be developed, it would have massive application in the retrofit field where large groups of buildings cannot be upgraded, due to the impracticality of current commercial insulation systems.

Being the constituent that incorporates the bulk of the VIP embodied energy, recent research advances have focussed on VIP core innovations, including studies by Nemanic *et al.* [19], Nemanič and Žumer [20], Chen *et al.* [21], Li *et al.* [22-24], Chang *et al.* [25], and Choi *et al.* [26]. Albeit comprehensive, the focus of these articles leaned towards the physical impacts of fumed silica based core material innovations. The cellulosic innovation was investigated as a fumed silica alternative for VIP cores, this done on the grounds of mitigating the barriers to market mentioned. The content reflects the order of analysis in evaluating the given hypothesis, with sections provided on review of current research, methodology, and results and discussion. The purpose was to establish a starting point for an ongoing body of research, in light of the infancy of nano-cellulose technology. The stakeholders aimed to use outcomes to plan future development paths to enable the substitution concept both technically and economically feasible. The collaborating industrial partner provides multiple insulation system solutions internationally, with its primary business the building industry.

2.0 Review

2.1 Commercial and super insulation heat transfer

Insulation thermal conductivity is the sum of internal solid-state conductivity, convection, and radiative (infrared) transmissions (Figure 4) [12, 13, 27]. The dominant component is gaseous heat transfer with standard commercial product values ranging 0.035 – 0.040 W/m.K [28]. Insulation performance is derived through suppression of micro-void natural convection in the material physical structure [13]. Higher-spec polymer products use blowing agents to replace air during manufacture, enabling conductivities ranging 0.018 - 0.028 W/m.K. [28, 29]. Commercial *super-insulation* performance is attributed to a physical structure size reduction from micro- to nano-scale; examples including aerogels and vacuum insulated panels (VIPs). A resultant suppression of convection emerges through the *mean free-path* of trapped gas molecules equalling that of the mean-diameter of the voids in the physical structure, leading to conductivities ranging 0.005 – 0.015 W/m.K [28, 30]. A phenomenon known as the Knudsen effect [13]**.**

Aerogels have the smallest physical structure of all known insulations at atmospheric pressure, with thermal conductivities ranging 0.014 - 0.016 W/m.K. The lower mentioned conductivities of VIPs provided via the Knudsen effect are caused through a combination of the fumed silica nano-structure, and gas pressure reduction to negligible levels. This impact was covered extensively by Hans *et al.* (Figure 5) [13]. Pore-structures are spherical and linked in three-dimensional networks with numerous dead-ends [30], leading to an emergent inefficiency of conduction, and a suppression of convection to an idealised case. The interaction physics were covered extensively in the works of Kwon *et al.* [30], Kim and Song [31], and Li *et al.* [23].

The modelling of VIPs involves reduction of thermal conductivity via manipulation of individual heat transfer modes, and quantifying impact takes into account all of these as they are coupled; i.e. changing the infrared absorbency consequently impacts solid conductivity. It has been shown more efficient to measure total thermal conductivity directly, than to predict the effect of changing one component. This was shown by Lorentzati *et al.* [16], Alam *et al.* [17], Chen *et al.* [21], and Di *et al.* [32]. In standard modelling thermal conductivity is separated into the following components, these dependent on underlying physical processes [27]:

$$\lambda_{tot} = \lambda_r + \lambda_{ss} + \lambda_g + \lambda_{coup} \quad [1]$$

The effects of radiation, solid-state conductivity, and coupling for low pressures is combined into the value λ_{vac}:

$$\lambda_{vac} = \lambda_r + \lambda_{ss} + \lambda_{coup} \quad [2]$$

Literature shows thermal conductivity (λ) does not follow a linear dependence on gas pressure and is instead formulated as given in *equation (3)* [27]. This assertion was confirmed experimentally in studies by Isaia *et al.* [14], Alam *et al.* [17], Kwon *et al.* [30], and Di *et al.* [32]:

$$\lambda_{tot} = \lambda_{evac} + \left(\frac{\lambda_{gas}^{0}}{\left[1 + \frac{p_{\frac{1}{2}}}{P_{gas}} \right]} \right) \qquad [3]$$

It is observed $p_{1/2}$ should be maximal to supress gas conductivity for any given pressure, or p_{gas} should be maintained at low value over the VIP product lifetime post manufacture. The standard fumed silica filling material has typical values of 60000 Pa for $p_{1/2}$ in its lifetime building application. Hans *et al* [13] covered impacts of pore diameter to conductivity change with pressure variation ($\delta\lambda/\delta P$ - Figure 5). Fricke *et al* [2, 33] reported the differential curve for thermal conductivity and pressure for various core materials, providing the methodology to calculate the effective average pore diameter (ϕ) using experimental data. This was calculated using the variation with gas constant ($\lambda_{gas}^{0}/2$), with air as the gas removed (Equation 4 and Figure 6); these works have been cited in multiple studies since their publication [8, 13, 32, 34].

$$\Phi(\mu\mathrm{m}) \approx 230(mbar.\mu\mathrm{m}) \big/ p_{\frac{1}{2}}(mbar) \qquad [4]$$

Multi-layer envelopes for maintaining VIP vacuum have finite air tightness leading to degradation in pressure on aging, with a typical annual 1 mbar a year loss [13]. Figure 6 shows fumed silica retains low conductivity and outperforms alternatives, even at pressure increases of a factor of a thousand. Additionally, when atmospheric pressure is attained on end of functional life, a fumed silica based VIP conductivity is approximately half that of alternatives. Hence it's use in commercial products, and it being the benchmark for new innovation comparison.

2.2 Manufactured fumed silica and reviewed VIP core studies

Energy intensive oxidation of silicon tetrachloride is used to manufacture fumed silica [35, 36], and its application advantages are widely known. Physically, it is comprised of SiO_2 molecules arranged in tetrahedral structures, these forming amorphous particles typically 12nm in diameter [37]. Prior VIP inclusion, these particles form agglomerates typically 4 to 200 µm in diameter. The material unfortunately has key limiting factors. Firstly, commercial products are manufactured as a by-product of silicon purification, and the limited global supplies are accounted for by a small organisation group. Secondly regarding sustainability, it is not biodegradable and no uses were reviewed for a recycled version of the material.

A fumed silica market review was constrained by lack of independently produced information, and sourced figures on capacity were limited to producer publications. It is estimated the annual global production capacity is approximately 1 million tonnes per year, with the majority of output in the east of Asia.

VIP core studies are limited in literature and generally focus on either substitution of the fumed silica supporting fibre structure, or methods to remove the material altogether. Li *et al* completed two studies, the first assessing the impact of fumed silica hybrid materials [23], the second investigating inclusion of rice husk ash [24]. In both articles, samples combined with varying fumed silica ratios were tested in a range of vacuum pressure set points to assess thermo-physical property impact. Results showed core structure pore diameters could be engineered to finer or courser values, manipulating change in conductivity with pressure (δλ/δP). This provided advancement in VIP optimisation, although the limitation of fumed silica content was not addressed. Chang *et al.* [25] studied diatomaceous earth and glass bubbles core composites comparing these with fumed silica. Although these performed at pressures below 10^4Pa, pore sizes were significantly larger and caused rapid performance loss. Chen *et al* [21] tested the VIP thermal conductivity impact of super stratified glass-fibre used as the core material. Under pressures ranging 0.03 - 10 Pa samples exhibited a commercially promising thermal conductivity range of 1.25- 13.03 mW/m.K. However, cores analysed were composed of fibres with diameters ranging 1 - 10μm, with performance loss significant in the 1-10 Pa region. Choi *et al* [21] researched fumed silica omission through VIP coating material alternatives, and inclusion of internal supporting pillars for shape maintenance. Using evacuated stratified glass fibre inter-separated with radiation shields as the core, and stainless steel coating, the measured central panel thermal conductivities were below 2 mW/m.K. This was lower than previously reported, albeit with no reference performance loss with pressure (δλ/δP), manufacture costs, or edge effect impacts.

Review highlights the achievable impact limits in VIP performance improvement through focus on the core fibre structure supporting fumed silica, or on the containment film retaining the imposed vacuum. The remaining innovation direction is in identification and implementation of a feasible fumed silica substitutes. The challenge is finding materials that: (a) have similar nano-structures for providing high thermal resistance and low loss of performance with internal panel pressure, and (b) can be produced by low-energy means. Monocrystalline cellulose nano-rods (or cellulose nano-crystals (CNCs)) were identified for this purpose.

2.3 Cellulosic nano-technology as a potential fumed silica substitute

Cellulose is naturally abundant, with extractable nano-crystalline domains in its structure. Cellulosic nano-crystals (CNCs) provide a direct route to sustainable nano-technologies in building insulations. Procurement is done through selectively degrading the cellulose structure amorphous portions, this typically through *acid-catalysed hydrolysis*. CNC cross-section dimensions are typically 6-20 nm, with 100-400 nm lengths [47]. Owing to their structure, they are very strong and thus have attracted wide interest for applications including reinforcement in: polymer composites [48-51], bio-composites [50-52], and electrode materials [53-55]. They are biodegradable, and are safe to handle with naked skin as far as current knowledge extends. These advantages led to their investigation as a substitute for VIP core fumed silica in this study.

There are currently limitations in CNC manufacture. Global output capacity is provided by facilities in North America and Europe, manufacturing close to 1 ton per day [56]. Typical factory yields range multiples of singles to tens of kilograms per week, with all providers using acid-catalysed hydrolysis [56]. Large-scale commercial production involves inorganic acid at high concentration [50, 57], with output reaction media diluted for CNC separation using membrane filtration [58-61]. Being difficult, separation requires considerable energy, time, cost, and occupies a large footprint area. Laboratory scale acid-CNC separation is faster, being via high-speed centrifugation (>10000 rpm) [62-65]. However, practicalities in scaling up are hindered by a high power requirement. Overcoming the difficulty of the acidic reaction mixture CNC separation bottleneck would improve competitiveness for future technology exploitation.

The limitations discussed render the technology unfeasible for current VIP products, with respect to production-turnaround demands. However, recent articles on high-efficiency separation techniques indicate these may soon be overcome [66], with innovations including the high energy ball milling [67], and the subcritical water hydrothermal method [68]. These processes provide potential to increase future market penetration on the basis of lower economic and environmental costs. This justified the investigation.

3.0 Methodology

The comparison method was centred on University manufactured cellulosic-particles delivered to the industrial partner, for inclusion as the fumed-silica substitute in their VIP manufacture process. Thermal conductivity at high vacuum pressure of 1 Pa (0.01 mBar), and change in thermal performance with loss of pressure ($\delta\lambda/\delta P$) were assessed and compared with data produced on conventionally manufactured fumed-silica VIP panels. This section covers all relating process detail and methodology.

3.1 Cellulosic-crystal preparation

Owing to a high aspect ratio, commercial CNCs tend to become arranged in highly porous structures post drying from aqueous dispersions [69], with atmospheric drying resulting in rigid nano-porous films [70]. Early discussions identified this issue on account of its potential in increasing product density and thermal conductivity. Dispersion freeze drying was therefore used, on it producing larger agglomerates collectable as a highly-porous low density powder. It was anticipated VIPs produced with this would have good insulation performance, with the powdered form providing assembly convenience.

Process cellulose used was cotton wool. 64 Wt% sulphuric acid was prepared by dilution of concentrated sulphuric acid (95 Wt%). The dialysis membranes were *Spectra/Por® 4 dialysis tubings* of 12000 -14000 *Daltons* molecular weight cut off. The extraction method of cellulosic-particles from the cotton wool was covered by Revol *et al*.[71] Acid hydrolysis of cotton wool was done on batches of 600 ml 64 Wt% sulphuric acid, preheated to 45^{o}C before addition of 75 grams cotton wool. The reaction mixture was left to process at 45˚C for 35 minutes, followed by dilution with deionised water and centrifugation twice, 10 minutes each at 10000 rpm to remove the acid from the dispersion. Cellulosic-particles settled during this process, with the supernatant discharged. On completion, the dispersion was dialysed against tap water for 48 hours to remove the remaining free acid, and particles were dispersed using ultra-sonication. Subsequent filtration over a No. 2 frit filter ensured that no micro-sized aggregates remained in the final dispersion. The resultant content was approximately 1 Wt%, this frozen through submerging in liquid nitrogen, with freeze drying following to yield the dry powder. Cellulosic-particle solids were placed over a 2mm sieve so the final evenly sized powder was formed for VIP assembly.

3.2 Preparation of the vacuum insulated panels with nano-cellulose used as the core material

On delivery to the industrial partner, the cellulosic powder was dried prior its incorporation into the standard VIP preparation method. This was done through a temperature controlled laboratory oven (*salvis LAB*) having a stability of +/-0.3 °C, and constant forced air movement [22]. Various drying temperatures where tested to ascertain the physical impacts to the particles when subject to heat. Visible degeneration was observed at 110°C, therefore a process temperatures ranging 60-80°C were selected as arbitrary safety points, with mass reduction monitored. Losses stabilized after 90 minutes for the higher 70-80°C settings, with overall 4.5% reductions recorded.

Prior drying the core was loaded to form a right angular shape using a press. A typical weight for the dried material was 500g per panel, with the core was wrapped in a polyester filter bag and sealed post formation. The product was dried in the oven prior placement in a pre-manufactured VIP envelope and placed in the vacuum chamber. The core forming was delivered prior drying to reduce the required time between oven and vacuum chamber. The typical powder temperature prior vacuum chamber closing was above 70°C. This ensured no moisture could settle in the powder between oven and chamber, as its presence would compromise the final panel thermal conductivity. Vacuum time was 20 minutes with a final chamber pressure of 0.01 mbar, prior final sealing of the VIP. Figure 7 shows the process steps.

Prior gas removal a load is applied to the panel via a special press to form its shape (Figure 8), with a compromise made between thermal performance and structural capacity. In general a higher core density is advantageous in being rigid and easily handled, while a low density core is fragile. The latter leads to uneven panel edges that cause breaks and thermal bridges when panels are placed in parallel, as is normal in installation. However, a lower core density has a higher thermal performance and therefore a balance is aimed for. In the cellulosic-VIP panel manufacture this balance was assessed. Multiple density variations were produced by gradually increasing the press load to create cores of various densities for thermal testing. The value for each core was determined by carefully weighing the panels after drying, and measuring the final VIP dimensions post evacuation. The process showed a higher degree of uncertainty for low density cores, which tended to have non-straight edges due to the phenomena explained. On completion, the cellulosic-core density of 140 kg/m^3 showed sufficient stability to be handled and processed.

In assessing VIP core performance two factors were considered. Firstly, general thermal conductivity dependence on density was analysed with this measured using a *Lasercomp Fox 600* (Figure 9) *[16, 72].* Secondly, the impact gas conductivity (*dLamba [gas]* with the inertial parameter *p1/2 – equations [3] and [4]*) was assessed to appraise the life time performance of the CNC core material.

3.4 Panel thermal conductivity tests with variable densities

Several core samples composed of pure cellulosic-powder were produced and tested, these having different core densities. The same drying and evacuation process was used in all cases. While the core filling weight could be measured accurately, the dimensions of a final vacuum panel were difficult to predict. This would normally not be a concern for rigid right-angled insulation products. The test series indicated a margin of error for the VIP core density of <2%, with the same applying to the thermal conductivity tests. The repeatability of the used FOX 600 device was +/- 0.2 mW/m.K, and the final error expectation was <= +/-0.3 mW/m.K (including the thickness measurement uncertainty). A set of six densities were manufactured at: 127 kg/m^3, 134 kg/m^3, 140 kg/m^3, 151 kg/m^3, 160 kg/m^3, and 170 kg/m^3.

Cellulose is a naturally efficient absorber of IR radiation; however to compliment this one sample was mixed with a SiC powder IR opacifier, this being a standard procedure in fumed silica based VIP manufacture. Gradual increases of SiC content were done, with ratios tested of: 0%, 5%, 20%, 40%, 50%, 60%, and 70%. For practicality reasons only the cellulosic-VIP core of 140 kg/m^3 density was measured in this procedure, on account of the quantity of powder supplied being small due to limitations in the research project time.

3.5 Panel gas conductivity tests at varying pressure

In determining impact to thermal conductivity with change in panel pressure ($\delta\lambda/\delta P$), several samples where produced. Measurements for each were tested at variations in pressure. Only pure cellulosic- powder was used in these tests, with a density ranging between 125-130 kg/m^3. Gas pressure dependence was measured, this used to calculate the material parameter $p_{1/2}$ (equation [4]). Internal VIP gas pressure was calculated using the *film lift method* [13].

In manufacturing panels with defined internal gas pressures the vacuum methodology was altered. Firstly, the chamber was evacuated to 10Pa for 20 minutes, this ensuring sample moisture content was the same. Vacuum pumps were then stopped and the chamber was filled with dry air until the desired pressure was reached. Levels were held for 10 minutes to ensure equilibrium between core and chamber was obtained. The cellulosic-VIP was then sealed and the vacuum chamber opened. At least one hour post completion internal gas pressure was measured. To check any leaks or defects, gas pressure was tested 24 hours after production. The thermal conductivity and dimensions where then measured in the Fox 600 device, followed by further gas pressure tests. Samples with 3 gas pressure tests having >0.4 mbar variations where then used.

4.0 Results and discussion

This section details all outcomes of the industry partner test procedures completed.

4.1 Panel thermal conductivity tests with variable densities

The mid-panel thermal conductivity measurements for the set of six cellulosic-VIP core densities manufactured are shown (Figure 10). For reference, margins of error are visible on both axes. The starting 127 kg/m^3 core density recorded a thermal conductivity of 11.7 mW/m.K (+/- 0.3). Measured values then increased linearly with the density, giving a line of best fit parameter α = 0.5095. Results were then compared with standard fumed silica VIP products, with variable core densities (Figure 11). The fumed silica cores tested contained SiC powder as an opacifier; therefore provide the final thermal performance benchmark the research was aimed towards.

4.2 Impact of opacifier addition to cellulosic-VIP panel thermal conductivity

Impacts to the thermal conductivity of 140 kg/m^3 cellulosic-sample cores through addition of SiC opacifier density are shown (Figure 12). An optimum performance weight between 50-60% was found, this mix permitting suppression of IR transmission and a 2.2 unit decrease in thermal conductivity to 10.3 (+/-0.4) mW/m.K.

4.3 Panel thermal conductivity at variable gas pressure

Cellulosic-VIP thermal conductivity results were recorded at various gas pressures for the 140 kg/m^3 sample, and compared against other know cores (Figure 13). The applied fitting curve uses the parameter lambda (vac) = 12.5 mW/m.K, and $p_{1/2}$ = 1300 Pa. Results show a cellulosic-VIP performance loss at a significant lower pressure than fumed silica; however, a higher performance retention than the glass fibre or PU core materials. Degradation was observed at a pressure of ~1 mbar (100 Pa), compared to the fumed silica-core at ~50 mbar (5000 Pa), and the PU and glass fibre cores at ~0.1 mBar (10 Pa). Considering VIP pressure loss is consistent in application, the results indicate the cellulosic-VIP would lose application performance after 5-6 months. Using the Fricke *et* *al* [33] methodology, the effective pore size in the cellulosic-particle core was calculated in the 17.7 µm range. This a factor of forty larger than the fumed silica based core 0.4µm pore size, using the same calculation.

4.4 Discussion

Test results highlight cellulosic-particle technology improvements are required for them to be a viable fumed silica replacement in building based VIPs. Comparisons are made with reference to: production time, physical characteristics, and thermal properties. Albeit a developed industry, access to information pertaining to fumed silica production was limited, this likely due to commercial interests.. Assumptions made were used in knowledge of this fact, and it is recognised independently produced figures would be required to validate the research conclusions herein.

In cellulosic-particle manufacture, the process done showed omission of the *acidic reaction mixture* would remove a production bottleneck, on account of it requiring 48 hours during dialysis. This would enable the capacity supply demanded by VIP producers, on provision of it having physical characteristics similar to fumed silica. Considering energy, innovations that replace the *centrifuge* and *freeze drying* stages with less intensive methods would reduce cost; however, it is appreciated this is challenging based on the physical reasons these steps are included for. Investment in the cellulosic-particle manufacture innovations is highly advised, as the potential benefits are clear for the application investigated and the wider global insulation industry for buildings. Going forward, cost reduction opportunities are likely via implementation of manufacture innovations reviewed; these including: the *high energy ball milling method [67]* and the *subcritical water hydrothermal method [68]*.

Further opportunities exists on account of this study's cellulose being sourced from pharmaceutical grade cotton wool. Being abundant, alternative sourcing options exist and supply of waste products could be used to offset costs. One possibility could be suppliers paying cellulosic-particle manufacturers to take unwanted waste cellulose, as oppose to paying landfill tax; examples including: waste wood from construction, paper from publishing, waste clothing, etc. This route may uncover further challenges however in terms of process pollutant removal, and uniformity CNC cross-sections and lengths.

Cellulosic-VIP thermal performance ranged 11.6 – 13.5 mW/m.K in cores with densities similar to fumed silica-VIPs (*Figure 11*). Albeit this was a factor of three larger than commercial versions, results indicate further improvements are available. The first included use of standard SiC opacifier, with thermal conductivity reduced 2.2 W/m.K (*Figure 12*). The second would be based on manipulation of cellulosic-particle cross-section and length. Limitations of this study did not permit their measurement in manufacture, and resultant impacts are unclear at this time. However it is recommended methods be used to determine them in future, as reviewed in the works of Elazzouzi-Hafraoui, S., *et al*. [47]. Theoretically, identification of smaller ranges in the physical parameters mentioned, would enable manipulation of cellulosic-particle insulation performance through engineering of the core pore size. Justification of this is based on the theoretical framework reviewed (S*ection 2.1*), where performance is dominated by the material pore size parameter ($p_{1/2}$ - *Equation [3]).* Going forward, innovation in manufacture techniques to facilitate production with smaller cross-section and length ranges is required. Methods exist for use as a starting point [67, 68].

The thermal performance change with vacuum pressure (δλ/δP) results provide further questions and potential opportunities (*Figure 13*). The study's 5-6 month outcome is currently not viable for commercial exploitation, comparing to fumed silica-VIPs performance loss post ~20 years.

Models show this was caused by average cellulosic-core pore size, this forty times larger than the fumed silica version. The result casts doubt on whether the cellulosic-particles were manufactured to the nano-scale; this stance based on two observations: (1) the 17.7μm pore size indicating micro-scale lengths; and, (2) the 38 mW/m.K panel thermal conductivity at atmospheric pressure indicating micro-scale cross-sections. Contrary to this, cellulosic-core performance loss occurred at a higher pressure (1 mBar) than the polyurethane and glass fibre versions (0.1 mBar). This indicates smaller scale voids in light of the theoretical framework set out in *equation (3)*. However, it is unclear if particles were manufactured to nano-scale, and further investigation on this is required going forward.

With unknown parameters mentioned it is unclear what level of reduction is possible to pore size on completion. However, research to investigate reduction levels achievable in pursuit of improvement is recommended. *Equation (4)* dictates reduction of cellulosic core pore size would lead to lower thermal conductivity at atmospheric pressure [27]. There are likely to be physical limits on achievable performance, albeit routes to manufacture development are available. With cellulosic nano-crystal technology in its infancy, developments may produce samples with cross-sections <6nm and lengths <100nm. If done economically, using a process with desired manufacture output capacity (*i.e. 500 – 1000 tonnes weekly*), it is feasible the technology could be used as the biodegradable substitute commercially pursued for VIP cores.

On basis of these achieving required *manufacture capacity*, *cost*, *thermal conductivity at pressure*, *maintenance of thermal performance with pressure loss (δλ/δP*), and *biodegradability* standards, a new generation of nano-scale insulations could be developed. This enabling the sustainable retrofit of building stocks in multiple countries, currently too expensive or impractical to complete, based on the short comings of current commercial (micro-scale) insulation technologies. This would have repercussions globally in many industries, as numerous additional applications may be identified.

Conclusions

This article studied the impact of replacing fumed silica in commercial vacuum insulated panels (VIPs) with a biodegradable cellulose-crystal innovation. Cellulosic nano-crystal manufacture technology is in its infancy, and the work aim was to determine a *performance starting point* and establish a comparison with current fumed silica VIP core benchmark: production capacity, and thermal performance parameters.

Albeit reviewed information was limited on fumed silica industry output, current commercial cellulosic-crystal production capacity estimates are under-developed when comparisons were made. The latter being a minimum 1000 times lower than the former. Analysis of laboratory samples identified three key *bottleneck* processes contributing to this issue, these being the presence of the *acid reaction* process, *centrifuge*, and *freeze drying* phases. Together, these phases accounted for the bulk of production time. Manufacture innovations were reviewed that remove the requirement of these processes including the *high energy ball milling method* [67] and the *subcritical water hydrothermal method* [68]. However it was not in the scope of this study to assess these, albeit clear opportunities exist to exploit them for the application analysed, and it is advised research be pursued.

Analyses of thermal performance was done for cellulosic-VIPs with variable core densities at pressure of 1 Pa (0.01 mBar). These were compared with standard fumed silica-VIPs containing opacifier of the same dimensions and similar densities. Thermal conductivity measurements of the cellulosic-VIPs manufactured ranged 11.7 – 13.6 mW/m.K for core-densities between 127 – 170 kg/m^3, whereas the fumed silica-core VIPs ranged 3.9 – 4.2 mW/m.K for core-densities between 145 – 180 kg/m^3. In terms of thermal performance with loss of vacuum pressure ($\delta\lambda/\delta P$), similar densities of cellulosic -core and fumed silica-core were compared directly, with these being ~140 kg/m^3. Results showed the cellulosic -core degraded at a pressure of 1 mBar (100 Pa), compared to the fumed silica-core degradation at 50 mbar (5000 Pa). Additionally when fully degraded to atmospheric pressure, the conductivity of the cellulosic panel (~38 W/m.K) was twice that of the fumed silica version (~20 W/m.K). The research indicated the cellulosic-VIP had an average core pore-size of ~17.7 µm and would lose performance after 5-6 months; this compared to the commercial fumed silica-core VIP pore-size of 0.4 µm that would lose performance after ~20 years. Evidently the fumed silica core outperformed the cellulosic version by a factor of three in thermal performance under vacuum and a factor of two at atmospheric pressure. In terms of performance retention with loss of pressure ($\delta\lambda/\delta P$) fumed silica-VIP outperformed the cellulosic version by a factor of fifty.

Study limitations did not permit the measurement of manufactured cellulosic-particle cross-section and length averages, and results indicate these are relatively large using the production techniques and calculation methodologies. The physical results casted doubt on whether the particles were nano-scale considering the theoretical framework reviewed. However, contradictions were observed in the aging performance retention of the cellulosic-VIP panels compared to other micro-scale equivalents. Evidently future research would incorporate this analysis.

In conclusion improvements are clearly desired and possible, if done through analysis and engineering of cellulosic-particle size. This outcome may arise through manufacturing innovations reviewed [67, 68]. However, it is recognised this would be a difficult considering the physical production time challenges this paper has shown. Future research should be done in the technology's application in building insulation materials. On assumption of this being successful, new areas and industry in insulation manufacture may arise. It is envisioned production of nano-scale biodegradable insulations could replace the micro-scale non-biodegradable versions currently used globally. This would have significant application in the retrofit sector where large groups of buildings cannot be upgraded, due to the impracticality and costs in technology implementation.

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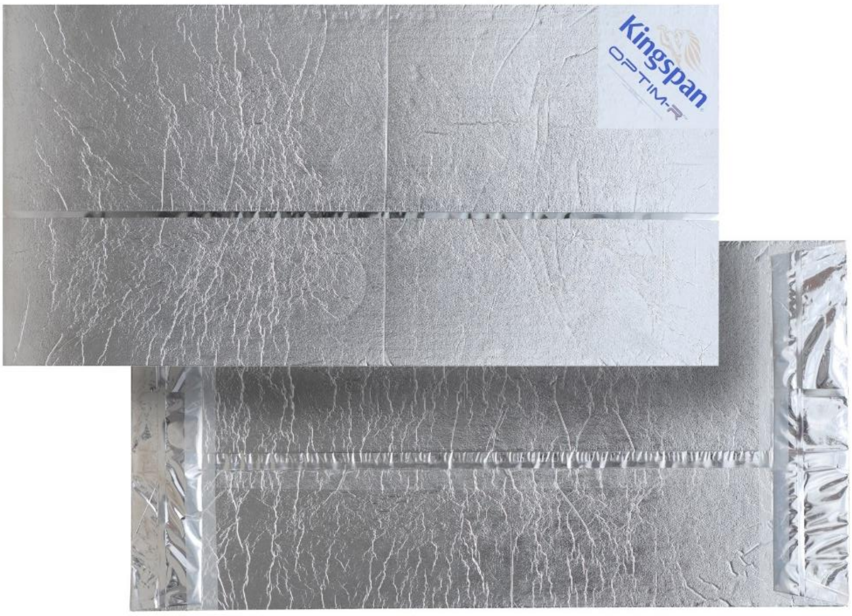


Figure 1 – A standard commercial vacuum insulated panel

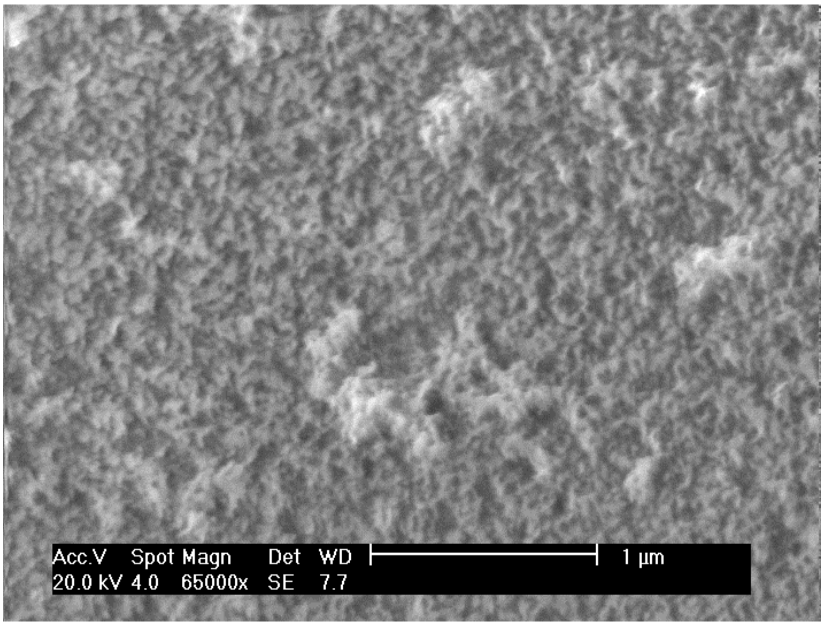
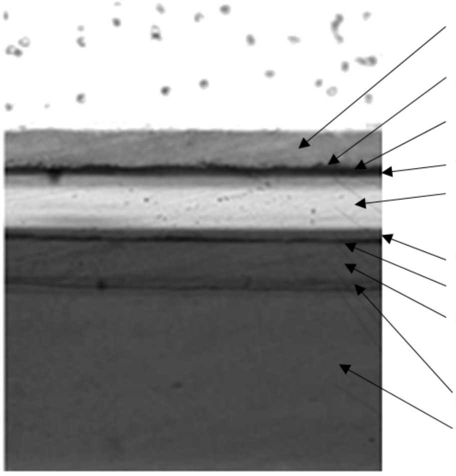


Figure 2 – Macro and microscopic images of the fumed silica based VIP core material



Layer	Material	Function	Thickness
1	PET	Protecting layer (substrate for 2)	12 µm
2	ALU	Aluminium barrier layer	30 nm
3	PUR	Polyurethane glue layer	2 µm
4	ALU	Aluminium barrier layer	30 nm
5	PP	Polypropylene (substrate for 4)	18 µm
6	PUR	Polyurethane glue layer	2 µm
7	ALU	Aluminium barrier layer	30 nm
8	PET	Polyethylene Terephthalate layer (substrate for 7)	12 µm
9	PUR	Polyurethane glue layer	2 µm
10	PE-LD	Polyethylene sealant layer	60 µm

Figure 3 - Details of film composition, [1, 13]

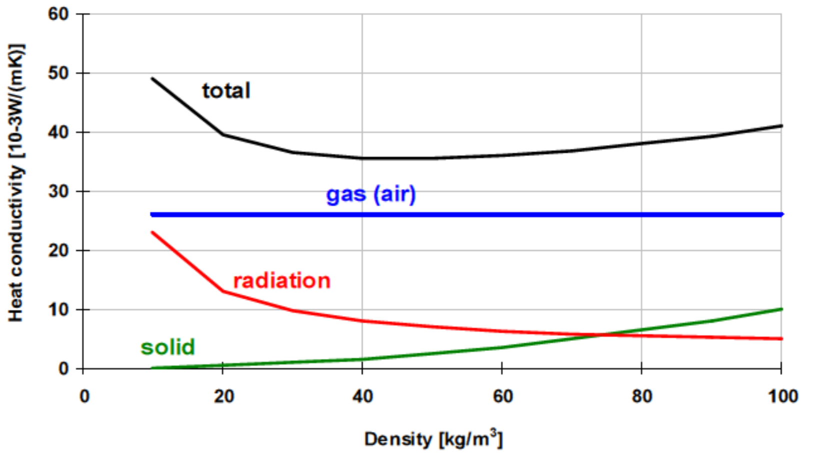


Figure 4 – Graph showing the distribution of solid, liquid, and radiation In relation to heat transfer in conventional insulations with changing density [13]

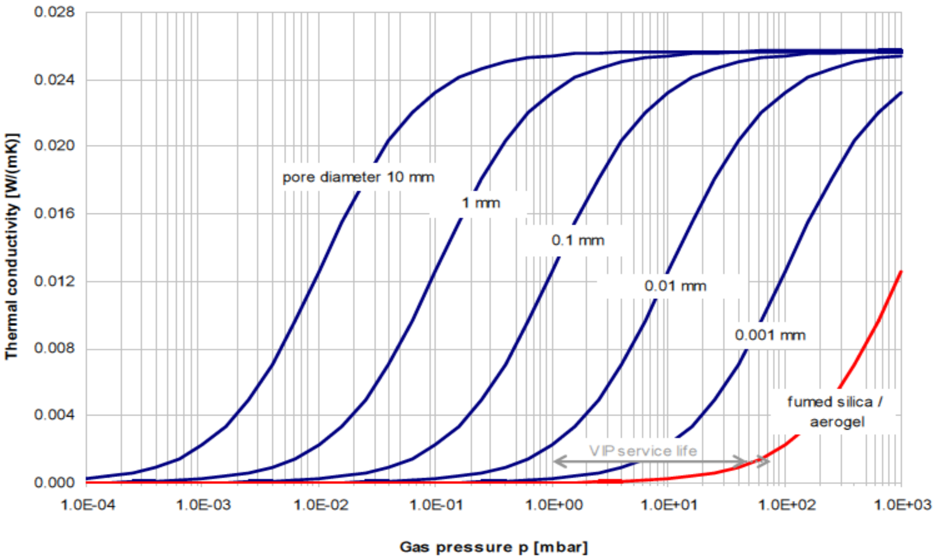


Figure 5 – Impact to gas pressure retention of core pore-size [13]

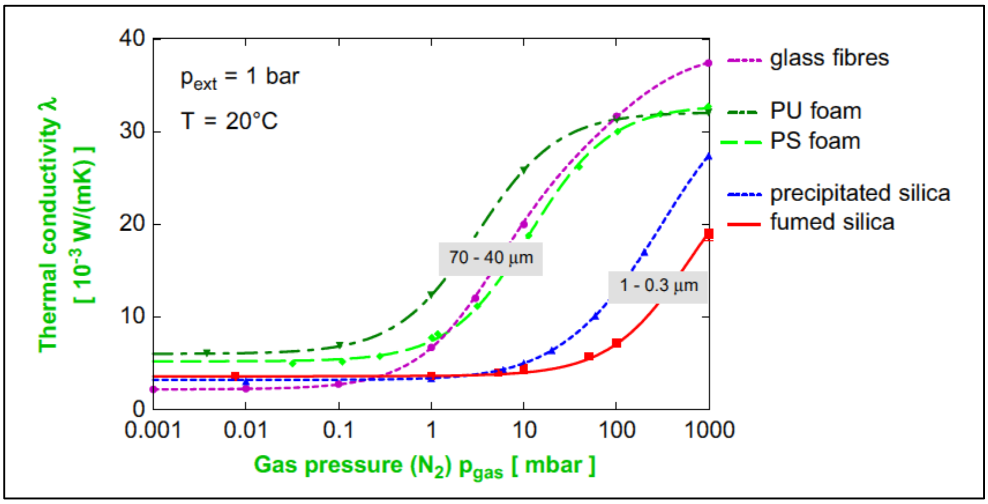


Figure 6 - Thermal conductivity of fibres, powders, and foams as a function of gas (air) pressure [2, 8, 32, 34].

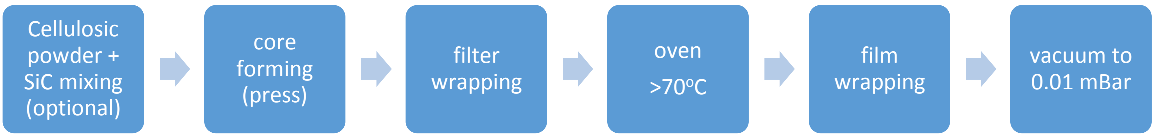


Figure 7 – Process steps for the cellulosic-VIP manufacture

Figure 8 – Press used to form VIP core shape via load application

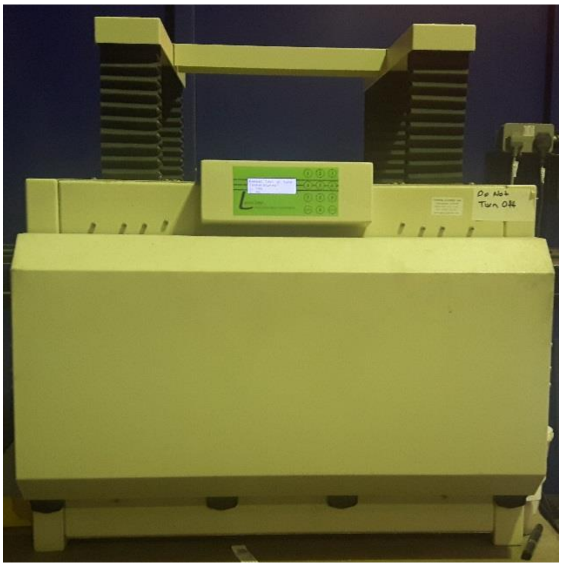


Figure 9 – Fox 600 device repeatability +/- 0.2 mW/m.K, and final error expectation <= +/-0.3 mW/m.K (including the thickness measurement uncertainty)

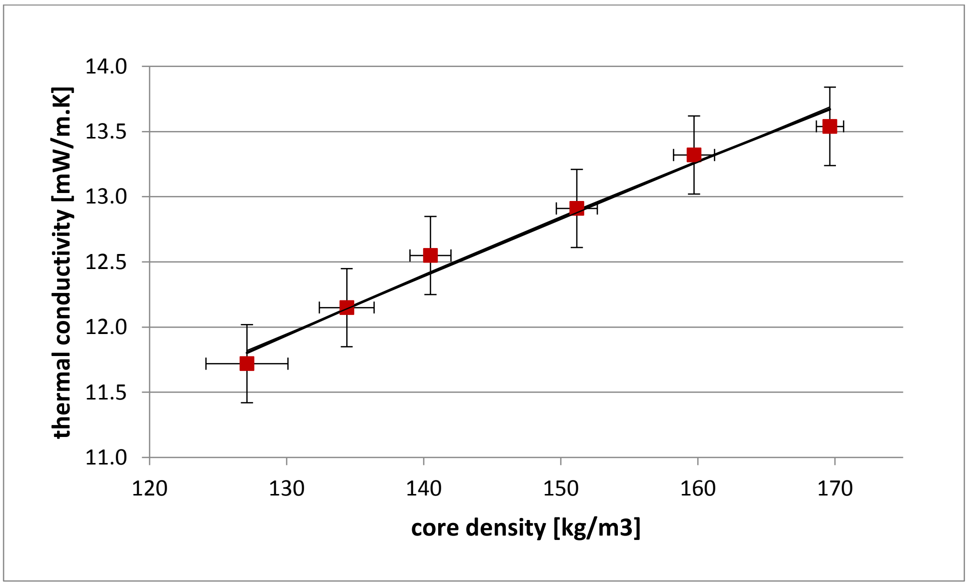


Figure 10 – Thermal conductivity measurements of VIP panels with cellulosic-core material of variable density

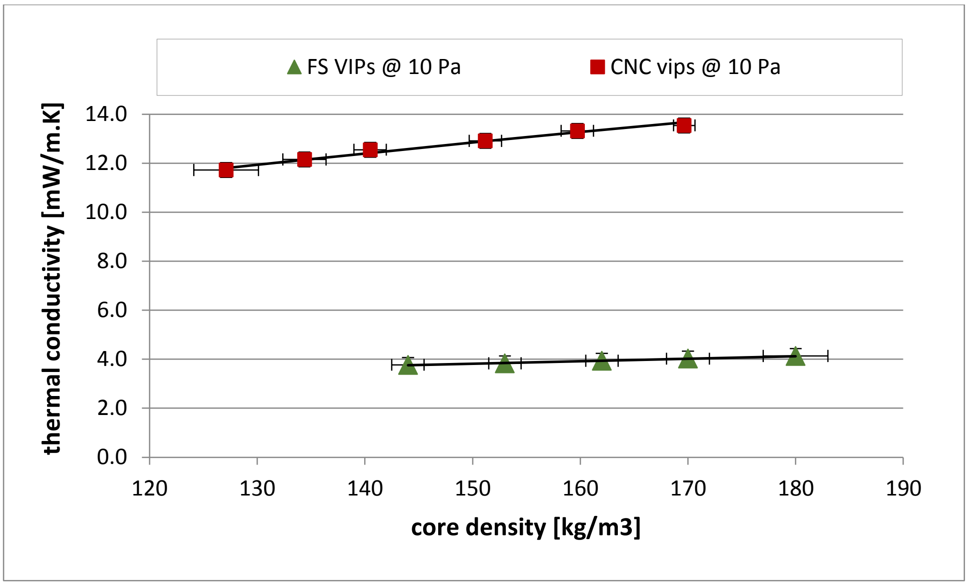


Figure 11 – Comparison of variable density cellulosic and fumed silica VIP cores

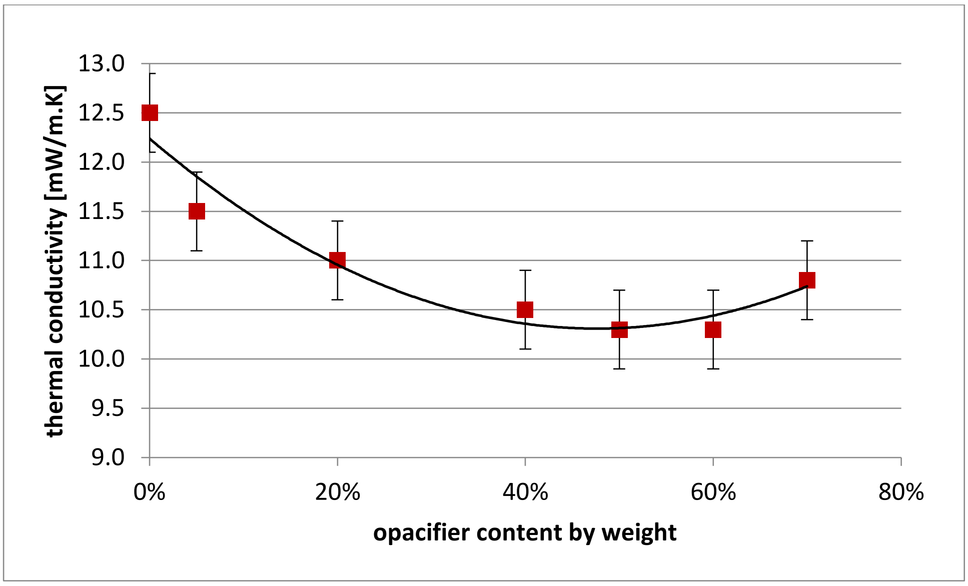


Figure 12 – Thermal conductivity of 140 kg/m³ cellulosic-VIP core panels with inclusion of IR opacifier at various content ratios

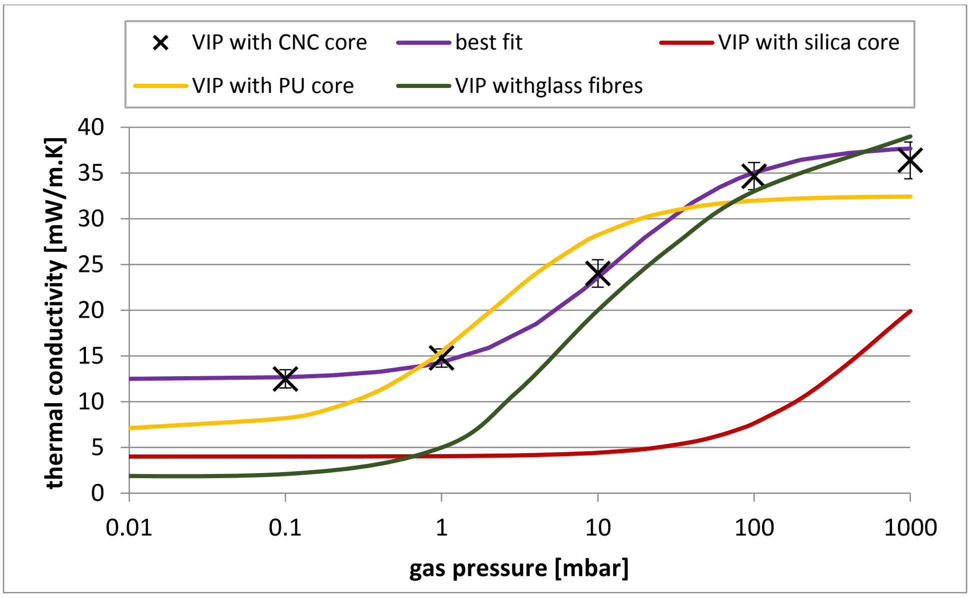


Figure 13 – Comparative thermal conductivity measurements of VIP panels with degradation of vacuum pressure

Table 1 – List of main fumed silica producers globally

Company	**Brand name**	**Production capacity**
Evonik	AEROSIL®	This Japan based company is the largest known producer. Capacity is not specific, although advertised: “*Evonik’s global annual production capacity for precipitated and fumed silica, metal oxides and matting agents now exceeds 550000 tons, with many more projects in the planning…..” [38]*
Wacker	HDK®	Located in China, with a capacity approximately half of Evonik. Advertised: “*the production capacity for siloxane and pyrogenic silica plants to approximately 210000 t/yr*.” [39]
Cabot	Cabosil®	The annual output ranged various values in China factories. Sources including*: 15000 t/yr at the Jiangsi plant [40]; 7000 t/yr at the Tianjin plant [41]; and, 8000 t/yr at the Wuhai plant [42]. Five* more plants are listed worldwide with no output capacity referenced in: USA, Wales, Germany, and India.
OCI	Konasil®	Factories in Korea and China having a combined annual output of *15000 t/yr [43]*
Tokuyama	Reolosil®	A China factory having an annual output of *11200 t/yr [44]*
Xunyu	Xysil®	A China factory having an annual output of *6000 t/yr [45]*
Orisil	Orisil®	A known supplier. Albeit no information production capacity available
Various smaller producers		A 2013 web based report stated: *“By 2015, China’s fumed silica capacity is expected to reach nearly 100000 metric tonnes per annum.”[46]*