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# The Path to the High Performance Thermal Building Insulation Materials and Solutions of Tomorrow

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## ABSTRACT

In todays society there is an increased focus on various energy aspects. Buildings constitute a large part of the total energy consumption in the world. In this respect it is important to have the optimum heat balance in buildings. That is, in a cold climate one wants to have as well thermally insulated building envelopes as possible. However, even in cold climates there might often be relatively long periods with overheating in the buildings, e.g. due to solar heat gains and excessive heat loads from miscellaneous indoor activities. In warm climates overheating is most often the case, e.g. in office work spaces with large window glass facades and extensive use of electrical equipment. Insulation retrofit is among the most cost-effective measures, even more cost-effective than e.g. solar photovoltaics. The traditional thermal insulation materials of today have typically thermal conductivities between 33 and 40 mW/(mK). State-of-the-art thermal insulation includes vacuum insulation panels (VIPs) with conductivities from between 3 to 4 mW/(mK) in fresh condition to typically 8 mW/(mK) after 25 years ageing due to water vapour and air diffusion into the VIP core material which has an open pore structure. Puncturing the VIP envelope causes an increase in the thermal conductivity to about 20 mW/(mK).

The main emphasis of this work centers around the possibilities of inventing and developing innovative and robust highly thermal insulating materials. That is, within this work the objective is to go beyond VIPs and other current state-of-the-art technologies. New concepts are introduced, i.e. advanced insulation materials (AIMs) as vacuum insulation materials (VIMs), gas insulation materials (GIMs), nano insulation materials (NIMs) and dynamic insulation materials (DIMs). These materials may have closed pore structures (VIMs and GIMs) or either open or closed pore structures (NIMs). The DIMs aim at controlling the material insulation properties, e.g. solid state thermal conductivity, emissivity and pore gas content. Fundamental theoretical studies aimed at developing an understanding of the basics of thermal conductance in solid state matter at an elementary and atomic level have been addressed. The ultimate goal of these studies is to develop tailor-made novel high performance thermal insulation materials and dynamic insulation materials, the latter one enabling to control and regulate the thermal conductivity in the materials themselves, i.e. from highly insulating to highly conducting. Furthermore, requirements of the future high performance thermal insulation materials and solutions have been proposed. At the moment, the NIM solution seems to represent the best high performance low conductivity thermal solution for the foreseeable future. If robust and practical DIMs can be manufactured, they have great potential due to their thermal insulation regulating abilities.

## 1. INTRODUCTION

### 1.1. The Energy Savings Demand of the World

The world will experience an ever increasing demand for energy savings in various areas. As buildings constitute a substantial part of the total energy consumption, savings within the building sector will be important, both for existing and new buildings. One of the key fields will be the thermal building insulation materials and solutions. Recent studies (McKinsey 2009) show that energy efficiency measures are the most cost-effective ones. Measures like e.g. solar photovoltaics and wind energy are far less cost-effective than insulation retrofit for buildings.

Essentially, the focus has been and still will be to achieve the highest possible thermal insulation values, i.e. the lowest thermal conductivity for the materials and the lowest thermal transmittance, U-value, for the structures and buildings. This usually works well for cold climates, where the building envelopes should have as low U-value as possible. However, even cold climates may experience longer periods with overheating due to solar heat gains and excessive heat loads from miscellaneous indoor equipment and activities. Traditionally, the solution has also been to thermally insulate the building envelopes sufficiently well enough in warm climates due to seasonal variations and periods during the year with a heating demand. Office work spaces with extensive use of electrical equipment and large solar energy gains through large window glass facades are likely to experience overheating.

Today's traditonal thermal insulation materials like mineral wool, expanded polystyrene (EPS) and extruded polystyrene (XPS) have thermal conductivities typically between 33 mW/(mK) to 40 mW/(mK). Polyurethane (PUR) with conductivities typically ranging from 20 mW/(mK) to 30 mW/(mK) is also used as a thermal insulation material, but even if PUR is safe in its intended use it rises serious health concerns and hazards in case of a fire. During a fire PUR will when burning release hydrogen cyanide (HCN) and isocyanates, which is very poisonous. The HCN toxicity stems from the cyanide anion ($CN^-$) which prevents cellular respiration. Generally, hydrogen cyanide may be found in the smoke from nitrogen (N) containing plastics.

### 1.2. The Vacuum Insulation Panel (VIP) Solution

Vacuum insulation panels (VIPs) represent state-of-the-art thermal insulation with thermal conductivities ranging from between 3 to 4 mW/(mK) in fresh condition to typically 8 mW/(mK) after 25 years ageing due to water vapour and air diffusion through the VIP envelope and into the VIP core material which has an open pore structure. Depending on the type of VIP envelope, the aged thermal conductivity after 50 and 100 years will be somewhat or substantially higher than this value. This inevitable increase of thermal conductivity represents a major drawback of all VIPs. Puncturing the VIP envelope, which might be caused by nails and similar, causes an increase in the thermal conductivity to about 20 mW/(mK). As a result, VIPs can not be cut for adjustment at the building site or perforated without loosing a large part of their thermal insulation performance. This represents another major disadvantage of VIPs. Several authors have been studying various aspects of VIPs, ranging from analytical models, thermal bridges and conductivity, air and moisture penetration, ageing and service life, quality control and integration of VIPs in building construction, e.g. Beck et al. (2007), Brunner and Simmler (2007, 2008), Caps and Fricke (2000), Caps (2005), Caps et al. (2008), Feinerman et al. (2007), Fricke (2005), Fricke et al. (2006), Schwab et al. (2005abcde), Simmler and Brunner (2005ab), Simmler et al. (2005c), Tenpierik and Cauberg (2007a), Tenpierik et al. (2007bc, 2008) and Zwerger and Klein (2005),

where a comprehensive review on VIPs for building applications has been made recently by Baetens et al. (2010a).

Nevertheless, despite the large disadvantages of VIPs, including their relatively high costs, they do represent a large leap forward in thermal insulation for building applications. Thermal conductivities between 5 to 10 times, depending on ageing time, lower than traditional thermal insulation materials like mineral wool and polystyrene products will especially be important when trying to achieve the standard and requirements of passive houses and zero energy or zero emission buildings. Thermal insulation thicknesses up to 50 cm or more in walls and roofs are not desired. Such thick building envelopes might require new construction techniques and skills. In addition, transport of thick building elements leads to increased costs. As an example, height restrictions may apply for passing under bridges and through tunnels, i.e. thinner elements will bring about a more efficient transport to a reduced cost. Building restrictions during retrofitting of existing buildings, e.g. by the lawful authorities or practical restrictions concerning windows and other building parts, may also require thinner high performance thermal insulation thicknesses than traditional insulation would be able to solve. Furthermore, in areas with a high living area market value per square meter, a reduced wall thickness may involve large area savings and thus a higher value of the real estate. Simple calculations show that for such areas the application of VIPs may actually result in an economic profit (Grynning et al. 2009).

Therefore, even if the VIPs are not the ultimate solution for the future, they may be the best solution for many thermal building envelopes today and in the near future, both from a thermal energy savings and an economical point of view. VIP research and advances should be concentrated towards developing VIP envelopes far better capable of preventing air and water vapour from entering into the VIP core, and thus being able to maintain a very low thermal conductivity for longer time periods up to at least 50 to 100 years. Besides, the research on and application of VIPs contribute to increased knowledge and idea generation about the thermal insulation solutions of tomorrow.

## 1.3. Gas-Filled Panels (GFP)

Other state-of-the-art thermal insulation solutions do exist. In principle, close to VIPs, is the technology of gas-filled panels (GFPs), studied by Griffith and Arasteh (1992), Griffith et al. (1993, 1995), Mills and Zeller (2008) and Yarbrough et al. (2007) among others. The GFPs apply a gas less thermal conductive than air, e.g. argon (Ar), krypton (Kr) and xenon (Xe), instead of vacuum as in the VIPs. To maintain the low-conductive gas concentration inside the GFPs and avoid air and moisture penetration into the GFPs are crucial to these panel's thermal performance. Naturally, vacuum is a better thermal insulator than the various gases employed in the GFPs. On the other hand, the GFP grid structure does not have to withstand an inner vacuum as the VIPs. Low emissivity surfaces inside the GFPs decreases the radiative heat transfer. Thermal conductivities for prototype GFPs are quite high, e.g. 40 mW/(mK), although much lower theoretical values have been calculated. Hence, the GFPs hold many of the VIPs advantages and disadvantages. Nevertheless, the future of GFPs may be questioned, as compared to them the VIPs seem to be a better choice both for today and tomorrow.

## 1.4. Aerogels

Aerogels represent another state-of-the-art thermal insulation solution, and maybe the most promising with the highest potential of them all at the moment, studied by Gould et al. (2007), Hostler et al. (2008), Schultz et al. (2005) and Schultz and Jensen (2008) among several others. Using carbon black to suppress the radiative transfer, thermal

conductivities as low as 4 mW/(mK) may be reached at lower pressures (Smith et al. 1998, Ramakrishnan et al. 2008). However, commercially available state-of-the-art aerogels have been reported to have thermal conductivities down to 13 mW/(mK) at ambient pressure (Aspen Aerogels 2008ab). The production costs of aerogels are still high. Aerogels have a relatively high compression strength, but is very fragile due to its very low tensile strength. The tensile strength may be increased by incorporation of a carbon fibre matrix. A very interesting aspect with aerogels is that they can be produced as either opaque, translucent or transparent materials, thus enabling a wide range of possible building applications. Only the future will show how far and extensive the aerogels will be applied in buildings.

### 1.5. Phase Change Materials (PCM)

Phase change materials (PCMs) are not really thermal insulation materials, but since they are interesting for thermal building applications, they are mentioned within this context. PCMs change phase from solid state to liquid when heated, thus absorbing energy in the endothermic process. When the ambient temperature drops again, the liquid PCMs will turn into solid state materials again while giving off the earlier absorbed heat in the exothermic process. Such a phase change cycle stabilizes the indoor building temperature and decreases the heating and cooling loads. Various paraffins are examples of PCMs, but a low thermal conductivity (Farid et al. 2004) and a large volume change during phase transition (Hasnain 1998) limit their building application. An overview of the main PCMs has been given by Demirbas (2006), whereas other reviews on PCMs may be found in works by Farid et al. (2004), Hasnain (1998) and Khudhair and Farid (2004). Further studies on PCMs may be found in the works carried out by Alderman and Yarbrough (2007) and Kosny et al. (2007). A suitable phase change temperature range, depending on climatic conditions and desired comfort temperatures, as well as an ability to absorb and release large amounts of heat, are important properties for the selection of a specific PCM for building applications.

### 1.6. Other Materials and Solutions ?

Despite its several disadvantages, the vacuum insulation panel solution seems at the moment to be the most competitive solution and the one most likely to hit the market first. Though, the aerogels in various forms, without some of the disadvantages of the VIPs, might in the long run change this picture. But might there exist some other materials and solutions which could exhibit as low thermal conductivity as the VIPs in their non-aged pristine condition, and without the several drawbacks of the VIPs? And if not, could they with some effort be envisioned, invented and finally manufactured? And what would be the requirements of these materials and solutions?

## 2. REQUIREMENTS OF THE THERMAL INSULATION OF TOMORROW

The future thermal insulation materials and solutions need to have as low thermal conductivity as possible. In addition, the thermal conductivity should not increase too much over a 100 year or more lifetime span. Furthermore, these materials and solutions should also be able to maintain their low thermal conductivity even if they are perforated by external objects (e.g. nails), except the increase due to the local heat bridges. Technologies based on vacuum may have problems with maintaining a low thermal conductivity over a long time span stretching over several decades, due to loss of vacuum with air and moisture uptake during the years.

A major requirement for the future thermal insulation materials is that they can be cut for adaption at the building site without losing any of their thermal insulation performance. The VIP solution with an envelope barrier around an open pore structure supposed to maintain a vacuum does not satisfy this specific requirement, as cutting a VIP will result in a total loss of vacuum and an increase of thermal conductivity up to typically 20 mW/(mK).

Several other properties also have to be addressed. These include mechanical strength (e.g. compression and tensile strength), fire protection issues either by the thermal insulation material itself or other protection means, fume emissions during fire where preferably no toxic gases should be released, climate ageing durability with various climate exposures, resistance towards freezing/thawing cycles and water in general, dynamic properties (i.e. the ability to regulate the thermal insulation level) and costs which should be competitive versus other thermal insulation materials.

That is, the thermal insulation materials and solutions of tomorrow have to satisfy several crucial requirements. Table 1 summarizes the various properties with their proposed requirements. As it can be seen, the proposed thermal conductivity requirement in the pristine condition is a conductivity less than 4 mW/(mK), which is the typical value for non-aged VIPs. Naturally, the thermal conductivity after a certain period of time or service life, is of vital importance. A conductivity less than 5 mW/(mK) after 100 years is proposed for the future thermal insulation materials and solutions to be developed. Table 1 represents an initial attempt to address the crucial properties and requirements of the future high performance thermal insulation materials and solutions, and will naturally be subject to change in the years to come.

Table 1. Proposed requirements of the future high performance thermal insulation materials and solutions.

| Property | Requirements |
|---|---|
| Thermal conductivity – pristine | < 4 mW/(mK) |
| Thermal conductivity – after 100 years | < 5 mW/(mK) |
| Thermal conductivity – after modest perforation | < 4 mW/(mK) |
| Perforation vulnerability | not to be influenced significantly |
| Possible to cut for adaption at building site | yes |
| Mechanical strength (e.g. compression and tensile) | may vary |
| Fire protection | may vary, depends on other protection |
| Fume emission during fire | any toxic gases to be identified |
| Climate ageing durability | resistant |
| Freezing/thawing cycles | resistant |
| Water | resistant |
| Dynamic thermal insulation | desirable as an ultimate goal |
| Costs vs. other thermal insulation materials | competitive |
| Environmental impact (including energy and material use in production, emission of polluting agents and recycling issues) | low negative impact |

## 3. THERMAL INSULATION MATERIALS AND SOLUTIONS OF TOMORROW

The following subchapters will introduce and define the principles behind what could become the future high performance thermal insulation materials and solutions. An initial presentation of these materials and solutions may be found in Baetens (2009), Jelle et al. (2009) and Baetens et al. (2010a). In short, the *aim* is to investigate the properties, requirements and possibilites of *advanced insulation materials* (AIMs).

### 3.1. Vacuum Insulation Materials (VIM)

A vacuum insulation panel (VIP) consists of an open pore core material with vacuum enveloped by an airtight and water vapour tight foil. If the enveloping foil had been completely air and moisture impermeable for a sufficient long period of time (which is not the case), the VIP thermal insulation technology would have been far better than the current technology. However, even if you could somehow make the VIPs reasonable invulnerable towards any perforations during the building period and the service life, the VIPs still lack one very important property of flexibility. That is, the VIPs can not be cut and adapted at the building site without loosing their vacuum, the very reason for their low thermal conductivity.

Therefore, one might ask – Even if we in the future could manufacture VIPs with no degradation over time, could we make a thermal insulation material or solution just as good as the VIPs, but without their disadvantages? One could then ask – Is it possible to make a thermal insulation material with a thermal conductivity as low as for VIPs in the pristine condition, but with no application of an enveloping foil?

That is, a basically homogeneous material with a closed small pore structure filled with vacuum with an overall thermal conductivity of less than 4 mW/(mK) in the pristine condition could be envisioned. Such a high performance thermal insulation material we define as a *vacuum insulation material* (VIM). This material could be cut and adapted at the building site with no loss of low thermal conductivity. In addition, perforating the VIM with a nail or similar would only result in a local heat bridge, i.e. no loss of low thermal conductivity. Figure 1 depicts the distinguished features between VIPs and the desired development of VIMs.
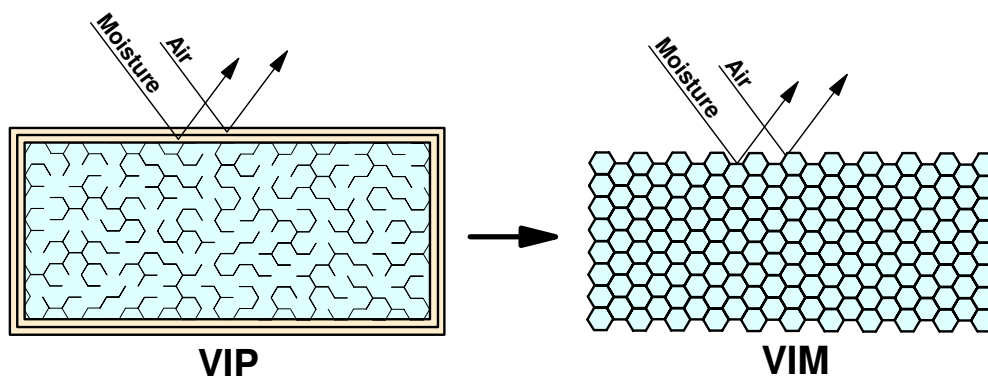


Figure 1.   The development from vacuum insulation panels (VIPs) to vacuum insulation materials (VIMs).

In order to be able to manufacture a VIM a low thermal conductivity of the solid state grid structure has to be made. Furthermore, a closed small pore structure filled with vacuum has to be accomplished. That is, as the pore structure is closed, the vacuum pore structure must be created during the production process of the VIM. One way to accomplish this is to envision a solid state material blowing itself up from within during the formation and subsequent expansion of an inner pore structure, which is illustrated in Fig.2. Another way, although totally different, might be to create a VIM grid structure or inner pore surfaces which will efficiently and completely absorb the pore gas molecules, e.g. by a chemical reaction process.
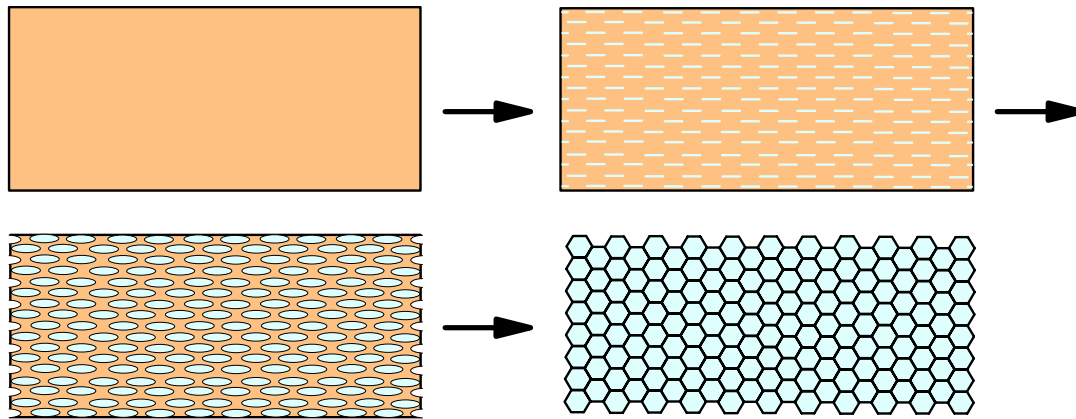
Figure 2.    An envisioned creation of vacuum insulation materials (VIMs); a solid state material blowing itself up from within during the formation and subsequent expansion of an inner pore structure.

The inner surface of the pores, could have as low emissivity as possible in order to decrease the thermal radiation transport. However, this might be in direct contradiction to attaining as low solid state thermal conductivity as possible. The VIM grid structure has to be strong enough to withstand the vacuum inside its pores. Finally, the air and water vapour diffusion or transport through the grid structure and into the vacuum pores have to be sufficient small enough so that the VIMs will maintain their low thermal conductivity below a certain value (e.g. 5 mW/(mK)) for at least 100 years.

Maintaining the vacuum inside the pores during a long service life may be the most difficult or challenging task for the VIMs. That is, the most challenging task after the VIMs have been manufactured, as to make VIMs is a highly challenging task in itself. Nevertheless, when an appropriate VIM production process has been established, such a production might hopefully be found to be both efficient and competitive.

### 3.2.   Gas Insulation Materials (GIM)

A *gas insulation material* (GIM) is basically the same as a VIM, except that the vacuum inside the closed pore structure is substituted with a low-conductance gas. That is, a GIM is basically a homogeneous material with a closed small pore structure filled with a low-conductance gas with an overall thermal conductivity of less than 4 mW/(mK) in the pristine condition. As for the VIMs, the GIMs may also be cut and adapted at the building site with no loss of low thermal conductivity. Likewise, perforating the GIM with a nail or similar would only result in a local heat bridge, i.e. no loss of low thermal conductivity. Figure 3 depicts the distinguished features between VIPs and the desired development of GIMs.
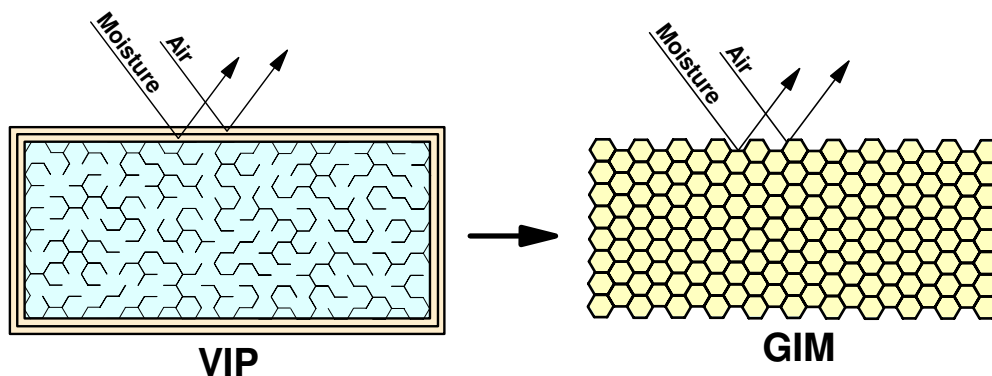


Figure 3.    The development from vacuum insulation panels (VIPs) to gas insulation materials (GIMs).

It might be easier to create a closed pore structure filled with a low-conductance gas than vacuum. Furthermore, the GIM grid structure does not have to be as strong as the VIM grid structure, as a vacuum pore structure will be prone to collapse before a gas-filled pore structure. In addition, it may be easier to maintain the original low thermal conductivity within a gas-filled pore structure than in a vacuum pore structure.

However, comparing the VIMs and GIMs, the VIMs ultimately have the largest potential of these two as the lowest thermal conductivity is achieved in a vacuum compared to a gas-filled pore structure.

### 3.3. Nano Insulation Materials (NIM)

Both the vacuum insulation material (VIM) and gas insulation material (GIM) solution share two of the same disadvantages. That is, firstly, the VIM and GIM grid structure need to prevent air and moisture penetration into their pore structure during their service life for at least 100 years. Secondly, perforating VIMs and GIMs with nails and alike causes local thermal bridges. Nail penetration and similar is considered to be of less importance for VIMs and GIMs, as this will normally only be minor local heat bridges. Ideally, if a 100 % airtight and water vapour tight VIM or GIM structure could be made, the former disadvantage would not represent any problem. However, we don't live in an ideal world, so some air and water diffusion into the VIM and GIM pore structure are probably bound to occur with time. The question then remains if the VIMs and GIMs are able to maintain their low thermal conductivity below a certain value (e.g. 5 mW/(mK)) for at least 100 years. Next, one may then ask if it is possible to envision and manufacture a high performance thermal insulation material or solution like the VIMs and GIMs, but without their disadvantages?

An answer to that question is the *nano insulation material* (NIM), where the pore size within the material is decreased below a certain level (i.e. 40 nm or below for air), in order to achieve an overall thermal conductivity of less than 4 mW/(mK) in the pristine condition. Figure 4 depicts the distinguished features between VIPs and the desired development of NIMs. Note that in principle, aerogels could be made so they could be considered as NIMs. Naturally, NIMs with an open pore structure will be containing air, where such a structure has to be resistant towards miscellaneous ageing degradation mechanisms as air gases including various pollutions will freely be admitted into the pores of the NIMs. Water condensation in such tiny nano pores has to be prevented, or else the thermal conductivity will increase substantially. The NIM closed pore structure may be containing any gases, e.g. air, Ar, Kr and Xe. A *true* NIM will still maintain a total and local thermal conductivity below 4 mW/(mK) when perforated, including the gas thermal conductivity in a single pore which has been perforated.
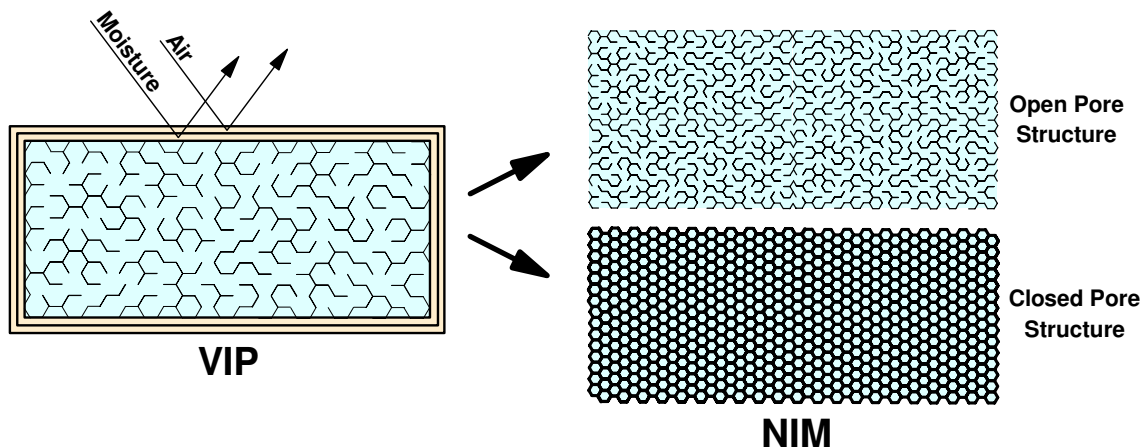


Figure 4.    The development from vacuum insulation panels (VIPs) to nano insulation materials (NIMs).

But how do the NIMs achieve their low thermal conductivity without applying a vacuum in the pores? If we still envision a VIM where we decrease the pore size within the material below a certain level, i.e. a pore diameter of the order of 40 nm or below for air, the gas thermal conductivity, and thereby also the overall thermal conductivity, becomes very low (< 4 mW/(mK) with an adequate low-conductivity grid structure) even with air-filled pores. This is due to the so-called Knudsen effect where the mean free path of the gas molecules is larger than the pore diameter. That is, a gas molecule located inside a pore will ballistically hit the pore wall and not another gas molecule. The gas thermal conductivity $\lambda_{gas}$ taking into account the Knudsen effect may be written in a simplified way as (Handbook of Chemistry and Physics 2003-2004, Simmler et al. 2005c, Schwab et al. 2005b, Raed and Gross 2007, Baetens 2009, Baetens et al. 2010a):

$$\lambda_{gas} = \frac{\lambda_{gas,0}}{1 + 2\beta Kn} = \frac{\lambda_{gas,0}}{1 + \frac{\sqrt{2}\beta k_B T}{\pi d^2 p \delta}} \tag{1}$$

where

$$Kn = \frac{\sigma_{mean}}{\delta} = \frac{k_B T}{\sqrt{2}\pi d^2 p \delta} \tag{2}$$

where

$\lambda_{gas}$ = gas thermal conductivity in the pores (W/(mK))
$\lambda_{gas,0}$ = gas thermal conductivity in the pores at STP
    (standard temperature and pressure) (W/(mK))
$\beta$ = coefficient characterizing the molecule - wall collision
    energy transfer efficiency (between 1.5 – 2.0)
$k_B$ = Boltzmann's constant $\approx 1.38 \cdot 10^{-23}$ J/K
$T$ = temperature (K)
$d$ = gas molecule collision diameter (m)
$p$ = gas pressure in pores (Pa)
$\delta$ = characteristic pore diameter (m)
$\sigma_{mean}$ = mean free path of gas molecules (m)

The Knudsen effect, i.e. the correlation between the gas thermal conductivity and the effect of the characteristic pore diameter and the gas pressure in the pores, is visualized in two-dimensional and three-dimensional graphical plots for for air, argon (Ar), krypton (Kr) and xenon (Xe) in Figs.5-6. In addition, the low thermal conductivity value of 4 mW/(mK) has been depicted in the plots in Figs.5-6. Note that these plots are logarithmic with respect to the pore diameter and the pore pressure.
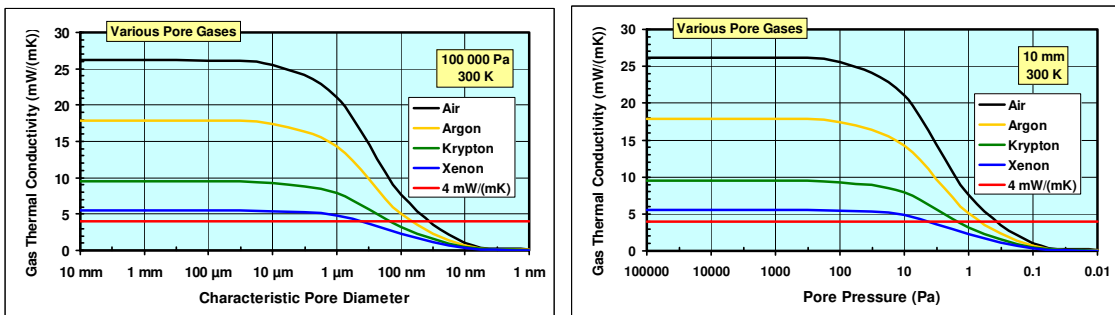


Figure 5.    The effect of pore diameter (left) and gas pressure in pores (right) on the gas thermal conductivity for air, argon, krypton and xenon. From Eq.1 and 2.

The hard sphere collision diameters have been applied for d in the calculations, i.e. 3.66, 3.58, 4.08 and 4.78 Å for air, Ar, Kr and Xe, respectively (given at 298.15 K, Handbook of Chemistry and Physics 2003-2004). That is, the covalent diameters of the gas molecules have not been employed in these calculations. Furthermore, $\beta$ = 1.75 and T = 300 K have been chosen in the calculations. In addition, $\lambda_{gas,0}$ values of 26.2, 17.9, 9.5 and 5.5 mW/(mK) have been applied for air, Ar, Kr and Xe (at 300 K), respectively. In the left plot in Fig.5 a pore gas pressure of 100 000 Pa ($\approx$ 1 atm = 101 325 Pa) has been chosen, whereas in the right plot in Fig.5 a rather large pore diameter of 10 mm has been selected. Note the symmetry these selected values yield in both the two-dimensional (Fig.5) and the three-dimensional (Fig.6) plots versus both pore diameter and pore pressure. Furthermore, for these chosen values in Fig.5, a rapid decrease between 1 $\mu$m - 10 nm and 10 Pa - 0.1 Pa is demonstrated in the gas thermal conductivities for all the four gases. This rapid decrease is also observed in the three-dimensional plots in Fig.6. Note that the gas thermal conductivity with pores filled with xenon is very low (about 5.5 mW/(mK)), even with large macroscopic pores at 1 atm pressure.
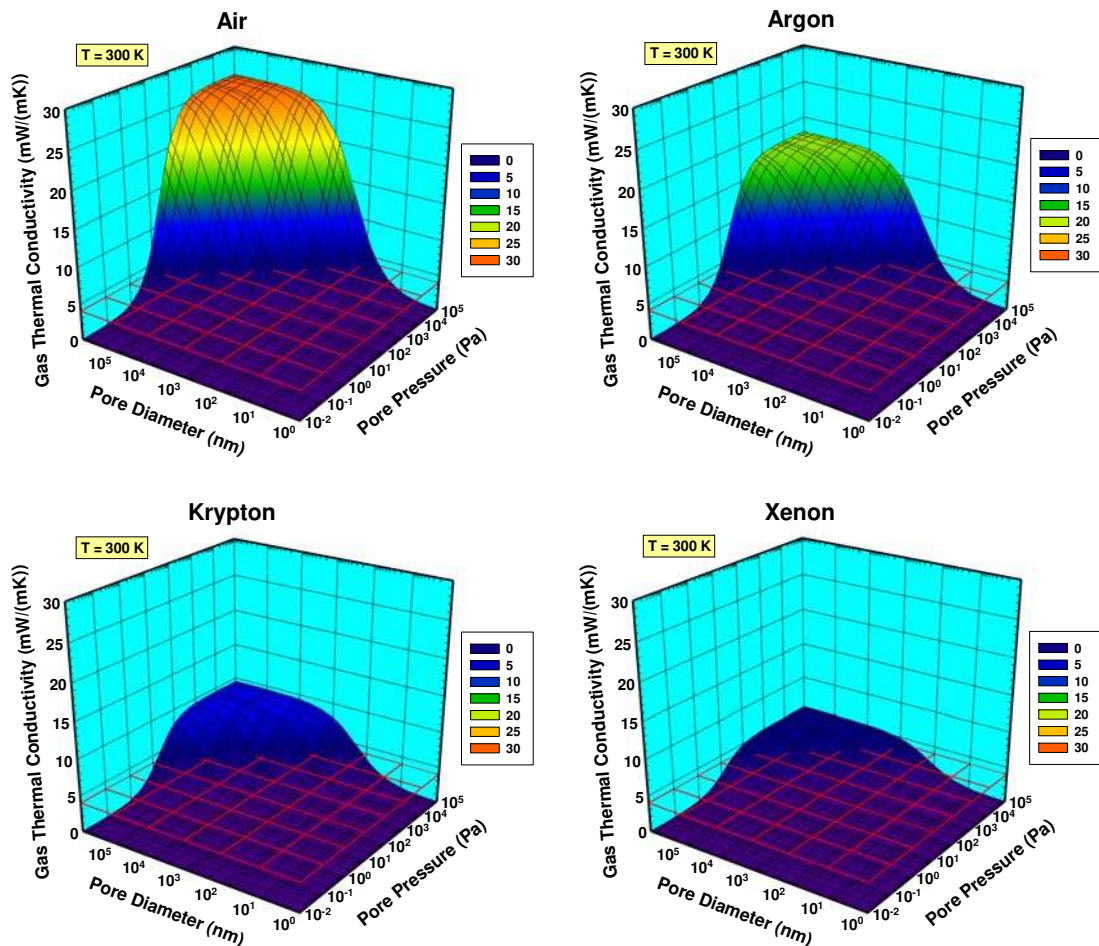


Figure 6.   The effect of pore diameter and gas pressure in pores on the gas thermal conductivity visualized in three-dimensional plots for each of the gases air, argon, krypton and xenon. From Eq.1 and 2.

The solid state lattice conductivity in the NIMs is also of great importance, and has to be kept as low as possible in order to obtain the lowest possible overall thermal conductivity. In addition to heat flows governed by the gas thermal conductivity in the

material pores and the solid state lattice conductivity, heat may also be transported by thermal radiation and thermal convection. The convection part, i.e. the gas (e.g. air) flow inside the pores (convection often driven by temperature differences), is assumed to be small compared to the gas conduction and radiation parts, and is hence not considered further in this context.

The effect of the radiation transfer inside the pores may be calculated approximately by applying the Stefan-Boltzmann equation to find the total radiation heat flux $J_{rad,tot}$ (W/m$^2$) through a material with n air gaps in series with infinite parallel surfaces of equal emissivity, which may be approximated as n pores along a given horizontal line in the material:

$$J_{rad,tot} = \frac{\sigma}{n\left[\frac{2}{\varepsilon} - 1\right]}(T_i^4 - T_e^4)$$

(3)

in order to find the radiation thermal conductivity $\lambda_{rad}$:

$$\lambda_{rad} = \frac{\sigma\delta}{\left[\frac{2}{\varepsilon} - 1\right]}\frac{(T_i^4 - T_e^4)}{(T_i - T_e)}$$

(4)

or

$$\lambda_{rad} = \frac{\pi^2 k_B^4 \delta}{60\hbar^3 c^2\left[\frac{2}{\varepsilon} - 1\right]}\frac{(T_i^4 - T_e^4)}{(T_i - T_e)}$$

(5)

where

$\lambda_{rad}$ = radiation thermal conductivity in the pores (W/(mK))
$\sigma = \pi^2 k_B^4/(60\hbar^3 c^2)$ = Stefan-Boltzmann's constant $\approx 5.67\cdot10^{-8}$ W/(m$^2$K$^4$)
$k_B$ = Boltzmann's constant $\approx 1.38\cdot10^{-23}$ J/K
$\hbar$ = h/(2$\pi$) $\approx 1.05\cdot10^{-34}$ Js = reduced Planck's constant (h = Planck's constant)
c = velocity of light $\approx 3.00\cdot10^8$ m/s
$\delta$ = pore diameter (m)
$\varepsilon$ = emissivity of inner pore walls (assumed all identical)
$T_i$ = interior (indoor) temperature (K)
$T_e$ = exterior (outdoor) temperature (K)
$J_{rad,tot}$ = total radiation heat flux (W/m$^2$)
n = number of pores along a given horizontal line in the material

In the above calculations the radiation thermal conductivity given by the expression $\lambda_{rad} = J_{rad,tot}\delta/(T_{k-1} - T_k)$ is found by applying the approximation $(T_{k-1} - T_k) = (T_i - T_e)/n$, i.e. assuming a linear relationship between temperature versus distance (indoor to outdoor) in the thermal insulation material, where $T_{k-1}$ and $T_k$ are the surface temperatures on two opposite inner pore walls within the same pore along the horizontal indoor to outdoor axis, respectively. This also implies that the whole temperature difference is assumed to be over all the pores and not over the thin solid state lattice walls connecting two neighbour pores.

From Eq.4 and 5 it is seen that the radiation thermal conductivity $\lambda_{rad}$ decreases linearly with decreasing pore diameter $\delta$, i.e. the smaller the pores the lower the radiation thermal conductivity. The linear relationship between $\lambda_{rad}$ and $\delta$ expressed by Eq.4 and 5 is visualized as a linear (left) and a logarithmic (right) plot with respect to the pore

diameter $\delta$ in Fig.7 for various emissivities $\varepsilon$ of the inner pore walls. Also note that the gas thermal conductivity $\lambda_{gas}$ in Figs.5-6 is plotted logarithmically with respect to $\delta$ for the whole pore diameter 10 mm to 1 nm range, whereas the radiation thermal conductivity $\lambda_{rad}$ logarithmic plot with respect to $\delta$ in Fig.7 only cover the pore diameter 10 mm to 0.1 mm (100 $\mu$m) range. An indoor and an outdoor temperature of 20°C and 0°C are applied in the grahical plots in Fig.7.

The strong $\lambda_{rad}$ dependence of both $\delta$ and $\varepsilon$ is observed in Fig.7. It is also seen that for materials with a pore diameter $\delta < 1$ mm and an emissivity $\varepsilon < 0.8$ the radiation thermal conductivity $\lambda_{rad}$ drops below 4 mW/(mK), i.e. only a relatively high $\varepsilon$ is required to achieve a low $\lambda_{rad}$ when $\delta < 1$ mm. Furthermore, it is observed that when $\delta = 1$ mm the radiation thermal conductivity is still as low as about 4 mW/(mK) and 5 mW/(mK) when the emissivity is at its maximum of 0.9 and 1, respectively. However, as $\delta$ is increasing above 1 mm the radiation thermal conductivity is increasing rapidly (linearly with $\delta$) and attains high values for high $\varepsilon$ values. The $\lambda_{rad}$ dependence of the indoor and the outdoor temperature is found to be rather small for normal temperature ranges, and is thus not plotted as graphs as the differences from the graphs in Fig.7 would only be marginal.
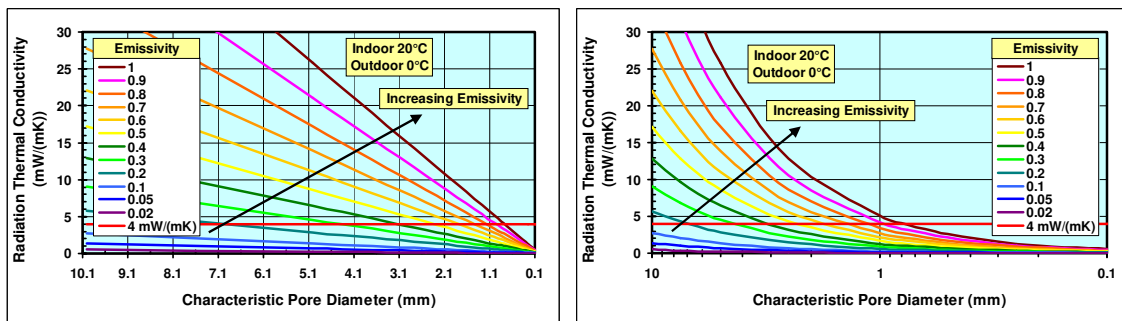


Figure 7.    The radiation thermal conductivity versus pore diameter for various emissivities of the inner pore walls. Linear (left) and logarithmic (right) plot with respect to the pore diameter. From Eq.4 and 5.

However, various works (Ben-Abdallah et al. 2009ab, Boehm and Tien 1970, Cravalho et al. 1967, Fu and Zhang 2006, Hargreaves 1969, Joulain et al. 2005, Loomis and Maris 1994, Mulet et al. 2002, Pan et al. 2000, Rousseau et al. 2009, Volokitin and Persson 2001, Volokitin and Persson 2007, Zhang 2007) describe a large increase in the thermal radiation as the pore diameter decreases below the wavelength of the thermal (infrared) radiation (e.g. 10 $\mu$m), where tunneling of evanescent waves may play an important role. A pore diameter of 10 $\mu$m is several orders of magnitude larger than the small pore diameter (e.g. 40 nm for air-filled pores) required to obtain a very low gas thermal conductivity according to the Knudsen effect (without applying a vacuum) as seen from Figs.5-6. Note that in Fig.7 the radiation thermal conductivity is only plotted down to a pore diameter of 0.1 mm (100 $\mu$m), which is about 10 times larger than the mean infrared wavelength at room temperature (10 $\mu$m). Work by Mulet et al. (2002) and Joulain et al. (2005) indicate that the large thermal radiation is only centered around a specific wavelength (or a few). That is, this might suggest that the total thermal radiation integrated over all wavelengths is not that large. How much this actually contributes to the total (overall) thermal conductivity is not known by the authors at the moment, although we believe it is at least rather moderate. Nevertheless, these topics are currently being addressed in on-going research activities.

Nevertheless, if a low-conductivity solid state lattice ($\lambda_{lattice}$ or $\lambda_{solid\text{-}state}$) and a low gas thermal conductivity ($\lambda_{gas}$) are achieved, and which still dominate the heat transport, i.e. larger than the thermal radiation part ($\lambda_{rad}$), then NIMs may become the high performance thermal insulation material of tomorrow. It has to be emphasized that the red borderline or border grid of 4 mW/(mK) depicted in Figs.5-7 is the prevailing aim and upper limit for the *total* (overall) thermal conductivity of the future advanced insulation materials (e.g. Table 1), whereas Figs.5-7 depict solely the gas thermal conductivity $\lambda_{gas}$ and the radiation thermal conductivity $\lambda_{rad}$ in their respective figures. That is, if more than one transport mechanism dominate the heat flow through the specific NIM, the respective $\lambda_{gas}$ and $\lambda_{rad}$ values have to be chosen so far below 4 mW/(mK) that the total thermal conductivity ($\lambda_{tot}$) is lower than the upper requirement limit of 4 mW/(mK) (Table 1).

As seen above, the NIMs may obtain a very low thermal conductivity with either an open or a closed pore structure. That is, firstly, the NIM grid structure does not need to prevent air and water vapour to diffuse into the pores. Secondly, perforating the NIMs do not create any local thermal bridges induced by air and water vapour leakage into the pore structure locally around the intrusion, except the thermal bridges caused by the perforating agents (e.g. nails) themselves. Naturally, a nail or similar through the whole NIM might lead to air leakages through the NIM and thereby increased heat loss from a building, as with all other thermal insulation materials, both traditional ones and new high performance materials.

Hence, as a conclusion, the NIMs obviously represent what could become the high performance thermal insulation materials for the future.

## 3.4. Dynamic Insulation Materials (DIM)

Usually we think about thermal insulation materials as something static, i.e. the insulation materials have a constant value which does not change. The normal choice is then to choose as low thermal conductivity as physically possible and within prevailing economical and building (e.g. area and height) restrictions. However, one might envision a building envelope which can change its thermal properties according to energy and user comfort demands. This might be solved by a *dynamic insulation material* (DIM) where the thermal conductivity can be controlled within a desirable range. Another name might have been controllable insulation material (CIM).

DIMs may constitute a part of an intelligent building envelope which includes the emerging solutions phase change materials (PCMs) and electrochromic materials (ECMs), i.e. for day/night storage/release of accumulated solar energy and dynamic control of solar energy through glazing systems, respectively. PCMs as described briefly in the above are basically adaptive materials, whereas ECMs as used in electrochromic windows (ECWs) are materials which can be controlled by applying an external electrical voltage, aiming at regulating as much of the solar radiation throughput in windows as possible (Baetens et al. 2010b, Granqvist 2005, Granqvist et al. 2010, Jelle 1993c, Jelle and Hagen 1999, Lampert 1998, Lampert 2004). Extensive investigations and research have already been carried out on smart windows, e.g. ECWs, where the first products are commercially available (Baetens et al. 2010b).

Among the ECMs there is also a group of materials called (electrical) conducting polymers, which in addition to their ability to dynamically change the solar radiation transmittance, e.g. their absorptive and reflective properties, are also able to change their electrical conductivity by several decades from electrical insulating materials till materials which are often denoted as synthetic metals. One of these ECMs and

conducting polymers is polyaniline (Chiang and MacDiarmid 1986, Jelle et al. 1993a, Jelle and Hagen 1993b, Jelle 1993c, Jelle et al. 1998, Jelle and Hagen 1999, Jelle et al. 2007), which by application of an external voltage may undergo colour changes from a clear, transparent state to a violet colour, with yellow, green and blue as intermediate colours, the unique doping mechanisms including both redox processes and proton doping. The electrical conductivity of polyaniline may be increased with a factor of $\sim 10^{10}$ by decreasing the pH from $\sim 4$ to $\sim 0$, reaching a maximum electrical conductivity of $\sim 500$ $\Omega^{-1}m^{-1}$ (Chiang and MacDiarmid 1986). Could these materials with such dynamic properties give us ideas as how to create dynamic thermal insulation materials?

How may DIMs achieve their thermal conductivity regulating abilities? In fact, this might be accomplished in several ways, i.e. by controlling the (a) inner pore gas content or concentration including the mean free path of the gas molecules and the gas-surface interaction, (b) the emissivity of the inner surfaces of the pores, and finally (c) the solid state thermal conductivity of the lattice.

Dynamically changing the inner pore gas content or concentration might be envisioned in several ways, e.g. by chemical reactions or inner pore surface adsorption processes, e.g. controlled by an applied electrical field. The emissivity of the inner surfaces of the pores might be controlled by changing the plasma wavelength $\lambda_p$ and thus the free electron density. The plasma wavelength may be defined as (Goldner et al. 1985, Jelle 1993c):

$$\lambda_p = (2\pi c/q_e)(m_e\varepsilon_0/n_e)^{1/2} \tag{6}$$

where $c$, $q_e$, $m_e$, $n_e$ and $\varepsilon_0$ denote the velocity of light, the electron charge, the free electron effective mass, the free electron density and the dielectric coefficient of vacuum, respectively. Materials with a high free electron density (short $\lambda_p$) will be highly reflecting materials with a low emissivity, whereas a low free electron density (long $\lambda_p$) corresponds to a low reflectivity and thus a high emissivity. The solid state thermal conductivity may be changed by dynamically controlling the phonon thermal conductivity or the free electron thermal conductivity, see further details in the following chapter concerning the thermal conductivity.

The process of changing the thermal conductivity back and forth within a desirable range may require a certain amount of energy. Naturally, this energy consumption should be kept as low as possible in DIMs.

The thermal insulation regulating abilities of DIMs give these materials a great potential. However, first it has to be demonstrated that such robust and practical DIMs can be manufactured.

## 3.5. What Is Really Thermal Conductance ?

In order to be able to invent and design the thermal insulation materials and solutions of tomorrow, one should initiate fundamental theoretical studies aimed at developing an understanding of the basics of thermal conductance in solid state matter at an elementary and atomic level. The ultimate goal of these studies will be to develop tailor-made novel high performance thermal insulation materials and dynamic insulation materials, the latter one enabling to control and regulate the thermal conductivity in the materials themselves, i.e. from highly insulating to highly conducting.

As a first thought, one might think that everything is well understood about thermal conductivity with a sound underlying theory. Restricting ourselves to solid state thermal

conductivity, the heat flow process seems to be rather straightforward. That is, one atom bouncing into the next one and so on in a solid state lattice structure, thus transferring heat from atom to atom. The term bouncing might be more appropriate concerning heat conduction in gases than solid state matter though, as the thermal energy is transferred through the chemical bonds in the solid state structure.

*Then we might ask ourselves, could we change these chemical bonds between the various atoms and thereby change the thermal conductivity? What kind of materials could be appropriate? What is characterizing a thermally highly conducting versus a highly insulating material? Could we utilize this information to tailor-make materials with the desired low thermal conductivity? What kind of physical model could describe and explain thermal conductivity? Investigating these fields closely, could it be possible to dynamically change the thermal conductivity from very low to very high, i.e. making a dynamic insulation material (DIM)?*

There exist two models for describing solid state thermal conductivity, i.e. the phonon model (atom lattice vibrations) and the free electron model. The origin of both these models might be seen as an analogue to the thermal conduction of molecules in gases with mean free path calculations, i.e. a gas of phonons and a gas of free electrons. It has to be emphasized that these thermal conduction models are just that, i.e. *models*. Be reminded that the atom model is also just that, i.e. a model.

Analytical expressions for the thermal conductivity are derived from both the phonon model and the free electron model. However, the utilization of these models on real materials for quantitative practical applications might be considered as rather limited. For example, the free electron thermal conduction model applies strictly to metals, not other materials, and is further complicated by the electron distribution being dependent on both the thermal gradient and the electrical field. Just the fact that there exists two models explaining only parts of the observed thermal conductance properties, tells us that thermal conductivity is not really understood on an atomistic level for solid state matter. A single model explaining all observed properties will always be the ultimate goal.

To what extent quantum mechanics, which is also merely a model, might be applied into the field of thermodynamics remains to be seen. One might also bear in mind the sudden and unexpected experimental discoveries and breakthroughs within the field of electrical superconductivity, where several models and theories have been proposed without being able to fully explain the observed phenomenon. The field of electrical superconductivity includes the early work by London (London and London 1935, London 1948), investigations throughout the 1950s including the original BCS theory by Bardeen, Cooper and Schrieffer from 1957 (Bardeen et al. 1957ab) and the urgent need for new explanations and theories when high-temperature electrical superconductivity suddenly was discovered by Bednorz and Müller in 1986 (Bednorz and Müller 1986) with subsequent rapid experimental discoveries the next few following years, e.g. Wu et al. (1987), Sheng and Hermann (1988) and Dai et al. (1995). Analogously and ultimately, one might also envision the whole span from thermal insulator to thermal supraconductor. We may for now end this section with the following words of wisdom: *The more we know the more we know we don't know.* And the more we want to know… and that's the whole fun of it, scientific research included!

## 3.6. Materials and Solutions to be Discovered

The ultimate thermal solution will always be subject to change as time is progressing. That is, the thermal solution we are searching might very well, also for the near future,

be found in solutions governed by hitherto unknown principles and which has yet to be discovered or invented. With other words, the thermal solution of tomorrow might be found in materials and solutions not yet thought of, which requires that we may have to *think thoughts not yet thought of*.

## 4. THE POTENTIAL OF THE STATE-OF-THE-ART AND BEYOND

In Table 2 there is given a short summary of the potential of the state-of-the-art and beyond with respect to becoming the high performance thermal insulation materials and solutions of tomorrow. Traditional thermal insulations like mineral wool and expanded or extruded polystyrene (EPS or XPS) are included as a reference for comparison. In addition to being a summary, Table 2 might be used to initiate a chain of thoughts of how to proceed beyond today's state-of-the-art thermal solutions. It should also be noted that Table 2 expresses the current status for the state-of-the-art solutions of today and the foreseen status for the beyond state-of-the-art solutions, where certain items in the table might be subject both to discussion and change.

At the moment, the NIM solution seems to represent the best high performance low conductivity thermal solution for the foreseeable future. In order to be able to regulate the thermal conductivity, the DIM solution may accomplish such a feat. However, how long time it would take before DIMs with satisfactory properties could be made, is rather questionable and difficult to predict.

Table 2. The potential of todays and beyond state-of-the-art solutions for becoming the high performance thermal insulation materials and solutions of tomorrow.

| Thermal Insulation Materials and Solutions | Low Pristine Thermal Conductivity | Low Long-Term Thermal Conductivity | Perforation Robustness | Possible Building Site Adaption Cutting | A Thermal Insulation Material and Solution of Tomorrow ? |
|---|---|---|---|---|---|
| *Traditional* | | | | | |
| Mineral Wool and Polystyrene | no | no | yes | yes | no |
| *Todays State-of-the-Art* | | | | | |
| Vacuum Insulation Panels (VIP) | yes | maybe | no | no | today and near future |
| Gas-Filled Panels (GFP) | maybe | maybe | no | no | probably not, near future |
| Aerogels | maybe | maybe | yes | yes | maybe |
| Phase Change Materials (PCM) | - | - | - | - | heat storage and release |
| *Beyond State-of-the-Art – Advanced Insulation Materials (AIM)* | | | | | |
| Vacuum Insulation Materials (VIM) | yes | maybe | yes | yes | yes |
| Gas Insulation Materials (GIM) | yes | maybe | yes | yes | maybe |
| Nano Insulation Materials (NIM) | yes | yes | yes, excellent | yes, excellent | yes, excellent |
| Dynamic Insulation Materials (DIM) | maybe | maybe | not known | not known | yes, excellent |
| Others ? | - | - | - | - | maybe |

## 5. CONCLUSIONS

In search for the development of high performance thermal materials and solutions beyond the state-of-the-art of today, new concepts have been introduced, i.e. advanced insulation materials (AIMs) as vacuum insulation materials (VIMs), nano insulation materials (NIMs), gas insulation materials (GIMs) and dynamic insulation materials (DIMs). These materials may have closed pore structures (VIMs and GIMs) or either open or closed pore structures (NIMs). The DIMs aim at controlling the material insulation properties, e.g. solid state core conductivity, emissivity and pore gas content. Fundamental theoretical studies aimed at developing an understanding of the basics of thermal conductance in solid state matter at an elementary and atomic level have also been addressed. The ultimate goal of these studies is to develop tailor-made novel high performance thermal insulation materials and dynamic insulation materials, the latter one enabling to control and regulate the thermal conductivity in the materials themselves, i.e. from highly insulating to highly conducting. Furthermore, requirements of the future high performance thermal insulation materials and solutions have been proposed. At the moment, the NIM solution seems to represent the best high performance low conductivity thermal solution for the foreseeable future. If robust and practical DIMs can be made, they have great potential due to their controllable thermal insulating abilities.

## ACKNOWLEDGEMENTS

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