VIP service life assessment: Interactions between barrier laminates and core material, and significance of silica core ageing

Bernard Yrieix*, Benoît Morel, Emmanuelle Pons
EDF, R&D Materials and Mechanics of Components, Avenue des Renardières—Écuelles, F-77818 Moret-sur-Loing Cedex, France

ARTICLE INFO

Article history:
Received 31 January 2014
Received in revised form 9 July 2014
Accepted 13 July 2014
Available online 25 July 2014

Keywords:
Vacuum insulation panels
Barrier laminates
Core material
Ageing
Silica
Permeation
Sorption
Hygric behaviour

ABSTRACT

As a result of temperature and humidity vacuum insulation panels (VIP) age: their thermal resistance decreases due to the combined effect of internal pressure and solid conduction increase. For given conditions this depends on the duration of the exposure and on the permeance of the envelope, as well as on the hygric and thermal behaviour of the core material. This paper begins by assessing the severity of the various building applications with regard to the VIP. It then presents the main behaviours and models needed and explains the life evaluation approach under stationary conditions. Furthermore particular attention is paid to the ageing of silica core material. It concludes that for a realistic estimation of the service life one needs to take into account both the thermal and hygric behaviour of the core material, the permeance of air and water vapour through the envelope, the temperature and humidity service conditions, and the thickness of the panel.

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1. Introduction

As a result of temperature and humidity Vacuum insulation panels (VIPs) age: their thermal resistance decreases due to the double effect of internal pressure and solid conduction increase leading to a finite service life, which has to be evaluated. This paper shows how the application, the core material and the barrier envelope all strongly influence the possible service life and how they interact.

In use, VIPs are stressed during use chemically and physically (oxygen, water, acidity, basicity, air pressure, vapour pressure, temperature, and even UV and IR radiation). VIPs experience two ageing mechanisms. A first set of conditions, typically from 50°C and/or 85%RH to higher temperature and/or humidity for the laminates tested in that study, rapidly leads to failure due to the deterioration of the barrier envelope itself, by delamination, by corrosion of the aluminium, or by hydrolysis of the polymers [1]. The second mechanism, and in fact the subject of this paper, proceeds at a slower pace but at a variable speed and corresponds to a regular aging of the VIP due to the permeation of atmospheric gases without particular aging of the envelope.

Another point to be noted is that the approaches and models cited later in the text are only valid for stationary or pseudo stationary conditions concerning the permeation. Conditions are of stationary type when they can be divided in elementary periods where temperature and humidity are constant during a time greater than the characteristic time of the transitional phase of permeation. Given the very high resistance to permeation of VIP barrier envelopes, this characteristic time (or time lag to diffusion) is long, i.e. typically 48 h at 40°C for an high quality laminate. As a consequence accurate service life assessment of VIP in many real conditions is only possible with dynamic diffusion and solubility models fed with accurate physical data. Determining such data for water vapour is the aim of another paper of the team [2].

It is useful to re-examine some definitions about the life of a component or a product.

The durability, also called life expectancy/time/duration, or service life, is the time during which the VIP has the ability to insulate to the required level until its thermal resistance drops below a required minimum. This notion of life therefore presupposes that one is able to define a criterion such as a property whose impairment renders the component or system unfit for its purpose. Do not confuse durability with reliability which is the probability that
have no failure at a given time, and with the failure rate which is a frequency.

The ageing of the VIP product is the set of irreversible modifications affecting the VIP over time. The concept of aging both covers the mechanism(s) and kinetics.

The nomenclature used is shown in Table 1.

2. Severity of operating conditions

2.1. Operating conditions

The operating conditions in buildings in France have been evaluated by Yrieix et al. [3] and the values for roofs terraces were adjusted according to the measurements of Brunner [4]. They vary strongly because of the variety of climatic conditions and applications. To assess their severity the authors have determined for each one the average operating conditions (T, RH, t) by distinguishing four unequal periods throughout the year (two hot, one cold and one intermediate) on both sides of the VIP. Some of these conditions are listed in Table 2: the maximum (hottest) ones and the nominal ones for the hot face of the VIP without any thermal nor hygric protection.

Table 1

<table>
<thead>
<tr>
<th>Variables</th>
<th>( T ) ((^{\circ})C)</th>
<th>RH (%)</th>
<th>Duration (days/year)</th>
<th>( T ) ((^{\circ})C)</th>
<th>RH (%)</th>
<th>Duration (days/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Internal insulation of wall &amp; floor</td>
<td>30</td>
<td>50–60</td>
<td>60</td>
<td>20–22</td>
<td>65</td>
<td>305</td>
</tr>
<tr>
<td>Heating floor (wet screed)</td>
<td>30–50</td>
<td>50–90</td>
<td>210</td>
<td>22–25</td>
<td>50–65</td>
<td>155</td>
</tr>
<tr>
<td>Heating floor (dry screed)</td>
<td>30–50</td>
<td>50</td>
<td>210</td>
<td>22–30</td>
<td>50–65</td>
<td>155</td>
</tr>
<tr>
<td>External insulation under rendering &amp; ventilated roof</td>
<td>70</td>
<td>70–10</td>
<td>30</td>
<td>20–22</td>
<td>65</td>
<td>335</td>
</tr>
<tr>
<td>Sandwich panel &amp; classical door</td>
<td>70</td>
<td>70–10</td>
<td>30</td>
<td>20–22</td>
<td>65</td>
<td>335</td>
</tr>
<tr>
<td>Wood sandwich panel &amp; door</td>
<td>50</td>
<td>50–80</td>
<td>30</td>
<td>20–22</td>
<td>65</td>
<td>335</td>
</tr>
<tr>
<td>Cladding, roller shutter box</td>
<td>35–40</td>
<td>50–55</td>
<td>30</td>
<td>20–22</td>
<td>65</td>
<td>335</td>
</tr>
<tr>
<td>Roof terrace balcony</td>
<td>45–70</td>
<td>95</td>
<td>60</td>
<td>20–22</td>
<td>65</td>
<td>305</td>
</tr>
<tr>
<td>Domestic hot water tank</td>
<td>60</td>
<td>15</td>
<td>365</td>
<td>60</td>
<td>15</td>
<td>365</td>
</tr>
<tr>
<td>Refrigerator</td>
<td>30</td>
<td>60</td>
<td>60</td>
<td>22</td>
<td>65</td>
<td>305</td>
</tr>
</tbody>
</table>

2.2. Severity criteria

The variability of the conditions depending on the application leads necessarily to variable severities themselves. The difficulty is to quantify these severities. For this purpose authors propose the following three severity criteria:

1. The maximum temperature or moisture level reached on the hot face of a VIP used alone without additional protection (Table 2); this criterion is intended to categorize applications versus the risk of catastrophic aging mentioned in the introduction. Other short-time applications may have higher temperature levels.
2. Two severity indexes \( S_W \) and \( S_0 \) (Eqs. (3) and (4)) which are derived from the permeation flux (Eqs. (1) and (2)) and summed over the four periods on both sides of the VIP:

\[
\Phi = \Delta P \times A \times t \times \Pi \tag{1}
\]

with

\[
\Pi = \Pi_0 e^{-Q_d/RT} \tag{2}
\]
It is interesting to observe that there are not only application parameters in these indexes (P pressure, t duration, T temperature) but also a material parameter from the envelope (Q activation energy of permeation). Figs. 1 and 2 show these indexes on the majority of applications.

The calculated severity indexes, reported in Table 3, use the activation energies:

- $Q_{aw}$ derived from the authors’ measurements on commercial barrier laminates: 26 kJ/mol [2].
- $Q_{a,o}$ of air permeation in crystallized PET chosen equal to that of oxygen: 30 kJ/mol [5]. This last value is very close to the one reported by Schwab et al. [6] on a comparable multilayer (28 kJ/mol) or identified by Bouquerel [7] on the same raw data (25.5 to 27.4 kJ/mol).

\begin{align*}
SI_w &= 2 \sum_{f=1}^{2} \sum_{i=1}^{4} P_{aw,f} \times t_i \times e^{-Q_{aw}/RT_{f,i}} \\
SI_a &= 2 \sum_{f=1}^{2} \sum_{i=1}^{4} P_{a,f} \times t_i \times e^{-Q_{a,o}/RT_{f,i}}
\end{align*}

Regarding the water vapour permeation, roof terrace/balcony and domestic hot water tank are much more severe application while the less severe ones include the internal insulation, the external insulation under cladding and the refrigerator.

Regarding the air permeation, domestic hot water tank is much more severe than other applications while the less severe ones once again include the internal insulation, the external insulation under cladding and the refrigerator.

For the building envelope authors conclude that the flat roof without any protection is the riskiest implementation, and even excluding this application, there is less difference between applications with respect to the permeation of air (+35%) than that of water (+80%).

In the next two paragraphs the main behaviours and models needed are given.

3. Behaviour of the barrier envelope

The main behaviour of the barrier envelope composed of metalized multilayer is the permeance to air and vapour (in the high temperature and humidity domain its own resistance against degradation in these conditions is also of importance). It is described by models describing solubility, diffusion and ideal layers. In the case of the VIP barrier the only force driving the permeation for each gas is the gradient of chemical potential, so the flux through the barrier is given by Eq. (5).

\begin{equation}
\Phi = A \times \left( \frac{D}{kT} \right) \times \left( \frac{\partial \mu}{\partial z} \right) \times X = A \times \left( \frac{D}{kT} \right) \times \left( \frac{\partial \mu}{\partial X} \times \frac{\partial X}{\partial z} \right) \times X
\end{equation}

with

\begin{equation}
\mu = \mu_0 + R \times T \times \ln \left( \frac{X}{X_0} \right)
\end{equation}

The first heavy hypothesis is that the activity coefficient $\gamma$ is equal to 1.

Then

\begin{equation}
\Phi = A \times \left( -D \frac{\partial X}{\partial z} \right) \quad \text{Fick’s law}
\end{equation}

For a one dimensional flux:

\begin{equation}
\Phi = A \times \left( -D \frac{X_{\text{out}} - X_{\text{in}}}{X} \right)
\end{equation}

The second heavy hypothesis is the validity of Henry’s law:

\begin{equation}
X = S \times P
\end{equation}

leading to the classical Eq. (1) where

\begin{equation}
\frac{\delta}{X} = \frac{D \times S}{X}
\end{equation}
Because both solubility and diffusion are thermally activated, permeability and permeance are also thermal dependent:

\[
\delta(T) = \delta_0 \times e^{-Q_d/RT} = \delta_0 \times e^{-Q_d/RT} \times S_0 \times e^{-Q_w/RT}
\]  

Like polyamide and ethylene vinyl alcohol polymers with water, some polymers are plasticized by solute, leading to an increase of permeability of water or other gas. A very simple way to express this for water vapour as plasticizer is given for each gas by the following equation:

\[
\delta = \delta_0 \times e^{\eta \times X_w} = \delta_0 \times e^{\eta \times S \times P_{sat} \times RH}
\]

where \(\eta\) the plasticizing coefficient depends on the permeate gas (here could be air or water vapour itself).

The behaviour of an ideal layer-stacking is given by Eq. (13):

\[
\Pi = \frac{1}{\sum_{j=1}^{n} 1/P_j}
\]  

Finally for a laminate consisting of \(n\) identical layers, this can be expressed for each gas as Eq. (14) (coming from Eqs. (11) to (13)).

\[
\Pi = \frac{1}{n} \times \frac{1}{\delta_0} \times e^{-Q/RT} \times e^{\eta \times X_w}
\]

\[
= \frac{1}{n} \times \frac{1}{\delta_0} \times e^{-Q_w/RT} \times S_0 \times e^{-Q_w/RT} \times e^{\eta \times S \times P_{sat} \times RH}
\]

Note that in Eq. (13), “layer” should be understood either virgin polymer film or metallized polymer film. The metallizations are considered not to be layers within the meaning of this equation. This approach has already been explained by Langowski [8]; it does call for a permeance homogenized across virgin film + metallization which can be accounted by the models of defects [8–10] of the metallic deposit, otherwise coherent and impermeable.

4. Behaviour of the core material

The whole behaviour of the core material, especially made of fumed silica is very complex: both the thermal and the sorption behaviours have to be taken into account as well as of course the influence of water and air incoming but also the ageing of the silica.

4.1. Core material conductivity

The classical model used is that called with parallel flux (Eq. (15a)) where the radiative contribution (Eq. (16)), solid conduction and gaseous conduction (Eq. (17a)) are added. Another literature axis [11] uses a modified model which includes a coupling term (Eq. (15b)). For a more representative model in the case of granular compacted silica one may use a double pore size distribution instead of single one (Eq. (17b)) [12] where it is relevant to chose for the pore diameters the mean pore sizes of the mesopores and the macropores.

\[
\lambda = \lambda_r + \lambda_{cs} + \lambda_{cg}
\]  

\[
\lambda = \lambda_r + \lambda_{cs} + \lambda_{cg} + \lambda_{coupling}
\]  

\[
\lambda_r = \frac{16 \sigma T^3}{3 \varepsilon}
\]  

\[
\lambda_{cg} = \frac{\lambda_{cg0}}{1 + CT/\left(\phi \times P_g\right)}
\]  

\[
\lambda_{cg} = \frac{\lambda_{cg0}}{1 + CT/\left(\phi_1 \times P_g\right)} + \frac{1}{1 + CT/\left(\phi_2 \times P_g\right)}
\]

After VIP ageing (namely water and air income) the apparent conductivity of the core material is very often described by Eq. (18) or equivalent [6,13,14] where two terms depend on ageing:

\[
\lambda = \lambda_0 + B \times \tau_w + G \times P_g
\]

\[
\lambda = \lambda_r + (\lambda_{cs0} + B \times \tau_w) + \left(\frac{\lambda_{cg0}}{1 + CT/\left(\phi_1 \times P_g\right)} + \frac{\lambda_{W0}}{1 + CT/\left(\phi_2 \times P_g\right)}\right)
\]

We have to note that this expression assumes only one effect of air on the inner pressure and a double effect of water on the “solid” conduction and on the inner pressure. In the case of a core made of silica, because of its water sorption behaviour, this last effect is often neglected or hidden behind the total pressure treated as (dry) air and behind a non purely conductive impact of moisture (cf. Section 5.3).

The global thermal behaviour can also be expressed by graphs like Fig. 3 proposed by Quenard et al. [16] that show the apparent conductivity of the core as a function of the inner pressure for different relative humidity or water content. In these results [16] the conductivity is measured by the hot wire method but the guarded hot plate or fluxmeter steady state methods can be used preferentially. In this expression, the two effects of water are taken into account but, as in Eqs. (18) and (19), the ageing of the core is not considered. It is very convenient to describe the core behaviour on a global basis, but it is a bit tricky to perform the measurement; three possibilities exist for this. The first one is the hot plate or fluxmeter under vacuum with potentially a difficulty to manage humidity. The second possibility is to use a small device for conductivity measurement in a vacuum climatic chamber with the inherent difficulty to master such small device like hot wire, hot disk, flash loading, etc. . . . The third one use opening/ice introduction/re-welding of the VIP at a given pressure to obtain VIPs with different humidity and total inner pressure; the conductivity measurement can then be made conventionally on the VIP by guarded hot plate or flux meter in a real mechanical loading condition much more preferable than the other methods.

4.2. Core material hygric behaviour

4.2.1. Macroscopic behaviour

The second point about the core is its hygric behaviour (water sorption isotherm) which gives the equilibrium between the water content and the water vapour partial pressure. An example of isotherms recorded at 25 °C on three kinds of silica is given Fig. 4, showing that even in a same core material family the behaviours in
the three domains of Langmuir, Henry and capillarity could be very different. Depending on the considered core many equations for the sorption isotherm are available [17]; for the silica an effective one is Eq. (20) [18]:

\[ \tau_W = \frac{RH}{C_1 \times RH^2 + C_2 \times RH + C_3} \]  

(20)

In practice the knowledge of the water content allows to deduce the partial water pressure and vice versa. The difficulty is that isotherms strongly depend on the real state of the silica which ages during service but potentially also during testing. These ageing phenomena and their consequences are described in the following paragraphs.

Note that another open question concerns the temperature dependence of the isotherm which remains unknown. All the literature about VIP made the implicit hypothesis of a negligible effect, which has to be verified.

4.2.2. Origin of the different hygric behaviours

The influence of the hygric behaviour of the core material on the evaluation of the VIP lifetime is very decisive. This is well known when the cores are very different like mineral wool versus silica or versus cellular polymers. But inside the family of the silica core, there exists little knowledge with respect to the initial state and virtually none in the aged state. To understand the influence of these behaviours on the VIP lifetime assessment, it is useful to remind some basic knowledge about hygric behaviour of silica before presenting the results of ageing cores outside and inside VIPs.

The hygric behaviour of silica for VIPs is a function of two parameters: the surface chemistry and the material microstructure.

4.2.2.1. Surface chemistry. The water adsorption may occur either by chemisorption or physisorption (cf. Fig. 5). Several authors described those mechanisms as a function of the silica nature and of the ambient temperature [19–25]. Considering physisorption only, and the water liquefaction energy \( Q_{\text{liq}} = 44 \text{kJ/mol} \) as reference, the value of the adsorption energy of water \( (E_{\text{ads}}) \) defines the nature of the sites, i.e. hydrophobic \( (E_{\text{ads}} < 44 \text{kJ/mol}) \) or hydrophilic \( (E_{\text{ads}} > 44 \text{kJ/mol}) \). On a non functionalized silica surface, three main adsorption sites exist:

- the siloxanes Si–O–Si which are hydrophobic
- the silanols Si–OH which are hydrophilic
- the physisorbed water molecules which are hydrophilic or neutral \( (E_{\text{ads}} = 44 \text{kJ/mol}) \).

As silanol is the first hydrophilic site available on dried silica, the higher is its surface concentration the higher is the adsorption capacity of the silica. At the initial state, the silanol surface concentration varies according to the synthesis process. Thus, in the case of precipitated silica, synthesized in a water solution, its surface is consequently fully hydroxylated and in consequence fully hydrophilic. Furthermore, it may contain a significant concentration of “inner” silanols inside the silica particle structure [20]. On the contrary fumed silica, produced in a hot flame at a temperature close to 1000 °C, has a surface very little hydroxylated and mainly non hydrophilic with a negligible inner silanols concentration.

One of the most detailed models is the one provided by Zhuravlev [20]. This model indicates the remarkable condition of 190 ± 10 °C in dry atmosphere where no physisorbed water exists on silica surfaces without affecting the existing silanols. Based on this, it is possible to quantify the different adsorption states of water on silica [19, 26] as illustrated by Fig. 6. Indeed, according to Zhuravlev’s model [20], the concentration of chemisorbed water \( (\theta_{\text{ChemH}_2\text{O}}) \) may be quantified by Thermo Gravimetric Analysis (TGA) after in-situ pre-treatment at 190 °C under dry atmosphere. Then physisorbed concentration \( (\theta_{\text{physH}_2\text{O}}) \) may be deduced from the variation in mass of a second sample beforehand stabilized in a reference humid atmosphere and after TGA. Based on this procedure, it is also possible to quantify two concentrations of physisorbed water differentiated in their temperature range of desorption. The main one in quantity desorbs in first step in the

![Water adsorption mechanisms](image-url)
lowest temperatures (<150 °C) and is associated to low energies. This concentration is noted $\theta_{\text{LEP H}_2\text{O}}$ in Fig. 6 ("LEP" for low energy physisorbed water). The second one desorbs in higher temperature range indicating higher energy bonds and this concentration is noted $\theta_{\text{HEP H}_2\text{O}}$ ("HEP" for high energy physisorbed). The existence of various high energy adsorption sites on silica surface is confirmed by various studies which a resume is provided in [19]. It appears that such adsorption sites are associated to special silanol configurations increasing the polarization of some ones among other.

4.2.2.2. Material microstructure. It is described first by its specific surface area and secondly by the sizes and volumes of its pores. Indeed, the higher is the area the higher is the global adsorption capacity of the silica. Then at higher humidity (RH > 50%), water adsorption by capillary condensation could occur for mesoporous or with small macropores materials (2 nm < pore diameter < 200 nm). Therefore, the lower are the pore sizes and the higher is the whole pore volume; the higher is the adsorption by capillary condensation.

Considering the high specific surface area range (100 to 500 m²/g) of the various silica for VIPs and the effective water permeation through the membranes, two studies were completed in order to evaluate the potential impact of the moisture on the VIP ageing. The first study consisted in pure silica ageing in various humid atmospheres and following their mass and specific surface area. For the second study, the specific surface area and hygric properties of core materials aged under vacuum and under the protection of commercial VIP membranes were characterized.

4.3. Thermo hygric ageing of the silica core

4.3.1. Pure silica ageing in climatic chamber (study 1)

The results summarized in this part are extracted from Morel’s PhD thesis concerning the thermo–hygric behaviour of nanometric silica [19].

A fumed silica (HDK T30® from Wacker) and a precipitated silica (Sipernat 500LS® from Evonik) conditioned in pellets of 200 kg/m³ were exposed for several months to humid atmospheres with varying temperature and relative humidity. The evolution of the pellet mass after re-stabilization in a reference condition and of the specific surface area $A_{\text{BET}}$ were followed as indicators of the evolution of their surface chemistry and of their microstructure. The reference condition 20 °C and 44%RH was chosen to avoid capillary condensation. It was generated by a salt solution of K₂CO₃ in desiccators. As the initial adsorbed mass is only composed of water, it was quantified according to the method presented previously in Fig. 6 [19,26]. During ageing, the adsorbed mass concentration was deduced from the mass variation.

Results and discussion. Depending on the silica two opposite trends were observed concerning the evolution of their global adsorbed matter $\theta_{\text{ads}}$ (cf. Fig. 7). While a systematic and continuous increase occurred for HDK T30 when $T > 20$ °C and RH > 44%, $\theta_{\text{ads}}$ did not vary significantly for Sipernat 500LS when $T \leq 35$ °C and greatly decreased for both conditions at $T = 60$ °C and RH ≥ 80 °C. On the other side, a similar and general decrease of the specific surface area was observed for both silica (cf. Fig. 8). This phenomenon was the most pronounced for the precipitated silica with a decrease higher than 65% after 125 days at 60 °C and 80%RH. A remarkable point concerns the dimensional variations of the pellets: the highest thickness variations amounted to −0.4% in the conditions at 80%RH for both silica, −2.5% for HDK T30 and −1.3% for Sipernat 500LS in the conditions at 95%RH. This result confirms a macroscopic stability of the pellets despite the high area decreases measured.

The examination of the microstructure by TEM revealed that the specific surface area decrease is associated to a coalescence of the primary particles without any modification of the aggregates.
architecture (cf. Fig. 9). Despite some area evolution crossings, we assume that the following trend exists: the higher the temperature and the humidity the higher the specific surface area decrease. Such an evolution is consistent with a dissolution and re-precipitation mechanism governed by Oswald’s law (Eq. (21)) [27,28] and amplified by the temperature, by the curvature of the nanosize silica primary particles and by the physisorbed water concentration. This hypothesis explains also well the faster decrease of $A_{BET}$ for the precipitated silica. But the fact it decreases down to a value lower than for the fumed silica in a shorter time, shows that the Sipernat 500LS structure is more sensitive than HDK T30. The hypothesis of difference in inner silanols concentration provides a possible explanation for this observation.

$$S = S_0 \exp \left( \frac{2 \times \gamma_{SL} \times V_m}{R \times T \times r} \right)$$  \hspace{1cm} (21)

where $S$ is the effective solubility, $S_0$ is the flat surface solubility, $\gamma_{SL}$ is the solid/liquid surface tension, $V_m$ is the molar volume, $r$ is the curvature radius.

Based on mass concentration and specific surface area data, it is possible to calculate the surface concentration $\psi_{ads}$ at 20\% RH of the adsorbed matters according to the relation (22):

$$\psi_{ads} = \frac{\theta_{ads}}{A_{BET}}$$  \hspace{1cm} (22)

The surface concentration after ageing is shown Fig. 10. For both silicas: the higher the temperature and the humidity the higher the increase of $\psi_{ads}$. In addition, it is observed that for high relative humidities (≥80\%), this increase occurs mainly during the first 20 ageing days. Based on these results, the decrease in the adsorbed mass concentrations noted for the precipitated silica is in fact due to the significant reduction of its specific surface area which impacts more than the increase of adsorbed matter surface concentrations.

It should be noted that a non expected adsorption of hydrophobic aliphatic and silicone volatile organic compounds (VOC) occurred on all samples aged in the climatic chambers. This contributed to the mass adsorbed on silica. After the tests, the chambers themselves were identified as being the emission source of those VOC. Based on the VOC molecular formulas and relative proportions determined by nuclear magnetic resonance an analysis combined with carbon quantifications, an estimation of the evolution of the

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**Fig. 8.** Specific surface areas after ageing under the conditions mentioned.

**Fig. 9.** HDK T30 microstructure observed with TEM before (A), and after 205 days ageing at 60\% C-80\%RH (B).
adsorbed water effective surface concentrations was carried out (cf. Fig. 11). Considering only the first 20 days, the impact of the temperature and of the humidity on the surface concentration remains the same as observed previously. Beyond this point, decreases of the adsorbed water are noted for various conditions. This fact is attributed to the VOC adsorption rendering the silica surface partially hydrophobic. In the case of HDK T30 silica at 20 °C, and at 35 °C and 55%RH, it is interesting to notice that water concentrations would have been lower than initial for the first 50 days.

All the data acquired through this study demonstrate that silicas used in VIPs are not stable when exposed to humid atmospheres. Their water mass adsorption capacity increases or decreases as a function of their nature and microstructure. Thus as fumed silica is produced at high temperatures, its partially hydroxilated surface can become more hydrophilic by siloxane hydroxylation. Due to the fact that the silica used for VIP cores is composed of very strongly bounded nanosize particles, their reactivity increases exponentially according to relation (20). Indeed, transformation of their microstructure may occur due to a modification of the silicon and oxygen atoms at the particle surface. This modification may be easily demonstrated by the measurement of the specific surface area as a function of the ageing time. The kinetics of the modifications of surface chemistry and of microstructure are function of the temperature and of the relative humidity. Furthermore the fact that there may be or not capillary condensation during ageing has to be considered as well. Because the adsorbed water concentration may greatly vary as a function of relative humidity and silica density, this can strongly affect the ageing of the materials.

4.3.2. Ageing in commercial VIPs (study 2)

Three commercial VIP references with the same core material were exposed to high temperature and high humidity atmospheres for more than one year. The core material is composed of 90% fumed silica (noted as FSa) + 5% PET fibbers + 5% SiC (mass percent).

The three VIPs chosen for characterization (see Table 4) suffered a significant mass uptake during ageing but they were clearly not ventilated. Water vapour uptake capacity at 25 °C of the core materials and of the fumed silica itself was measured by a volumetric adsorption method in an automatic very accurate apparatus (BELSORP AQUA3, Bel Japan) after degassing between 140 °C and 150 °C in vacuum. The volumetric adsorption method takes typically 1.5 days for a complete adsorption run. The specific area was also measured by nitrogen adsorption.

Results and discussions. Table 4 and Fig. 12 present the results. First the cores of the aged VIPs exhibit water adsorption capacities (about 2.3% at 25 °C-50%RH) close to that of aged pure silica (2.9%), very far from the capacity in the initial state (0.9%). Because PET fibbers and SiC have negligible and stable vapour adsorption
capacities compared to FSx, the observed evolution is clearly due to
the ageing of the silica itself.
By the way it also shows that moderate duration of exposure (one month) at ambient temperature but at high relative humidity (80%) leads to a hardly aged state with a completely different hygrometric
behaviour of the silica core than in initial state.
Next a significant decrease of the specific area is observed, from
200 to around 140 m²/g. The specific area of pure aged silica also
decreased but in a less pronounced way: 170 m²/g. This is consistent
with the lower temperature and shorter duration of the ageing.
This is not due to the humidity reached which is the same for pure silica and in aged VIPs (roughly 80%RH for 5% of water uptake).
These results demonstrate that the same evolution of silica powder occurs inside VIPs as well (i.e. an increase of the surface hygroscopicity and a decrease of the specific area). This evidence of ageing in the VIP and in short time at ambient temperature and quite high humidity has three consequences addressed in the following paragraphs:

- one on the determination of isotherms;
- another on the humidity at equilibrium \( \tau_{w,x} \) (Eq. (23)), thus on the ageing kinetics from the conductivity point of view;
- and finally one on the final and real conductivity of ventilated VIPs.

### 4.3.3. Impact on the determination of isotherms

Most of the studies (as in Annex 39 [13]) used the cup/gravimetric
method in desiccators with salt solutions or climatic chambers as
required by the NF EN ISO 12571. This method implies very long exposure times to humidity, potentially high, to obtain values at
high partial pressure or to obtain the desorption isotherm. These
long exposures (typically 4 months or more) to humidity allow
modifications of the highly reactive surface of silica, first hydroly-
ation of the siloxanes and then an increase of the hygrophilicity,
and secondly decrease of the specific area. A third impact could
also occur, namely the shrinkage of the sample corresponding to
the collapse of the structure due to the capillary condensation. The
collapse of the structure leads to an entry in the capillary condensation
domain of the isotherm at a lower water vapour pressure than expected.

Therefore ageing of the silica occurs during the test itself, so
obtained data are not those of pristine silica but of silica aged during
the characterization.

The differences between the isotherms obtained by the cup method and those obtained by a fast method such as the automatic volumetric apparatus could be significant as shown on Fig. 13 for two kinds of silica. In this figure the two impacts, i.e., an increase of specific hygrophilicity and a shift of the capillary condensation domain, are clearly visible. Depending on the temperature and the duration of each stabilization stage, the intensity of these modifications can vary as shown in the data from Annex 39 [13].

The short measurement time (about 3 days for complete adsorption and desorption curves) the volumetric adsorption method driven by automatic systems allows to assess the effective vapour sorption isotherm of silica powders at a state given.

Another comment about the determination of isotherms concerns the choice of the drying conditions before starting the measurements. Because of the hygrometric behaviour described in
the paragraph "origin of the hygrometric behaviours" and especially illustrated by Fig. 6, the classical drying at atmospheric pressure in an oven at 105 °C like for mineral wool is not relevant. The silica core needs to release at least all the physisorbed water. So a drying under vacuum at 190 °C is recommended, where 140 °C is a minimum drying temperature.

### 4.3.4. Impact on the assessment of VIPs ageing

For the majority of the approaches to assess VIPs ageing, the core
material is not supposed to age. Authors have shown that the silica
core ages (Sections 4.3.1 and 4.3.2), even at ambient temperature (Section 4.3.1), and this phenomenon strongly depends on the
conditions of ageing and on the formulation of the core. According to Sections 4.2 and 4.3 different mechanisms could occur:

**Fig. 12.** Water vapour adsorption isotherms at 25 °C of cores of aged VIP and of pure FSx silica measured by volumetric adsorption method.

**Fig. 13.** Influence of the method on the determined water vapour adsorption isotherm of two types of silica (respective precipitated/fumed initial \( A_{BET} = 450 \) m²/g) compare to data from Annex 39 [13].

<table>
<thead>
<tr>
<th>Material</th>
<th>Treatment</th>
<th>VIP mass variation (%)</th>
<th>( A_{BET} ) (m²/g)</th>
<th>( \tau_{w} ) (% at 25 °C 50%RH)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Core 1</td>
<td>VIP aged 70 °C 90%RH, 540 days</td>
<td>+5.0</td>
<td>134</td>
<td>2.24</td>
</tr>
<tr>
<td>Core 2</td>
<td>VIP aged 70 °C 90%RH, 400 days</td>
<td>+5.3</td>
<td>155</td>
<td>2.41</td>
</tr>
<tr>
<td>Core 3</td>
<td>VIP aged 50 °C 90%RH, 540 days</td>
<td>+5.0</td>
<td>142</td>
<td>2.23</td>
</tr>
<tr>
<td>FSx</td>
<td>Initial state</td>
<td>–</td>
<td>200</td>
<td>0.94</td>
</tr>
<tr>
<td>FSx</td>
<td>23 °C 80%RH, 30 days</td>
<td>–</td>
<td>170</td>
<td>2.89</td>
</tr>
</tbody>
</table>
• First one needs to consider the hydroxylation on the silica surface which leads to the increase of the mass humidity stage of the sorption isotherm, up to an equilibrium with the water partial pressure different that the initial one.
• Secondly one observes a slight decrease of the specific area with a subsequent additional sintering of the silica particles which leads to an increase of the solid conduction of the core.
• Thirdly the amount of physisorbed water could increase. Its mobility is clearly detrimental regarding the apparent conductivity of the VIP under transient thermal loading.

In conclusion the simple linear effect of the mass humidity content used in many approaches to assess VIPs lifetime which does not consider the silica core ageing is to simple and leads to errors. Some recent work [29] presents results showing an underestimation of the linear relationship of the conductivity in aged states. Real behaviour after the given ageing has to be considered to do relevant time life assessment.

Another consequence is the evolution of the humidity at equilibrium $\tau_{W,\infty}$ during the ageing of the VIP. A continuous shift occurs towards higher values in a first step, potentially followed in a second step by a relative decrease if duration, temperature and humidity are high enough to lead to a strong reduction of the specific area.

4.3.5. Impact on the conductivity of ventilated VIP

For numerous certifications, the thermal resistance/conductivity of a ventilated VIP is required. The measurement is made after a short stabilization of the opened VIP. This short stabilization is insufficient to reach the equilibrium state leading to a significant under estimation of the real conductivity. Because of the silica core age, this measurement has to be performed after an ageing corresponding to the service conditions. Temperature and humidity must be chosen at or over the service conditions and the duration of the treatment must be long enough to reach the equilibrium. Authors have identified several satisfactory treatments ranging from 24 h to 90%RH to 30 days at 23 °C 80%RH. In these conditions it is clear that the conductivity of standard European ventilated VIPS is well over the 20 mW/(mK) declared at the moment: values up to 24 mW/(mK) on commercial products from different suppliers have been measured.

4.3.6. Proposals for silica core ageing study

The potential impacts of silica core ageing on the life assessment of VIP or more generally on the characteristics of the core should be studied. Several possibilities for a simplified evaluation of their ageing exist:

• With only two conditions the consequences of ageing with and without capillarity condensation can be determined: high temperature to accelerate the phenomena and, respectively, 50 and 80%RH to avoid and encourage the capillarity condensation.
• With some additional ageing conditions (different temperatures) to obtain the temperature dependence, potentially including the apparent activation energy of silica ageing at low humidity (<50%RH) (mechanism of surface hydroxylation and specific area increase) where only the surface modifications without microstructural evolutions occur i.e. without capillary condensation.

To perform this tests authors propose to age cores in sealed bags quite permeable to water vapour but tight against volatile carbon organics.

5. Evaluation of life time

Many methods exist; they can be classified according to different criteria:

• Approaches from global to step by step;
• With one or two gases;
• With coupled effect between gases or not;
• More or less physical or empirical;
• With ageing of the core material or not;
• With ageing of the envelope or not;
• Stationary sequences or actual dynamics.

This shows the variety of the possibilities of lifetime assessment protocols. Next some examples to illustrate some benefits, difficulties or shortcomings are presented.

5.1. Example 1: Global assessment

Ideally the assessment of life can be done as schematically represented in Fig. 14: by determining which values of pressure and humidity match the required value $\lambda_{\text{max}}$ (relationship $\lambda = f(RH, P)$), knowing the loading conditions and the permeances of the envelope the duration to reach $\lambda_{\text{max}}$ can be calculated. Depending on the conditions in which $\lambda = f(RH, P)$ and the sorption isotherm is obtained, the ageing of the silica core is taken into account or not.

In practice it is difficult to have all the characteristics enabling to implement this approach. Other, simpler approaches are possible.

5.2. Example 2: Simplified approach

A possible simplified approach in particular for a user who lacks the means to determine the law of permeation of dry air is to consider only the permeation of water vapour that is very easy to determine by weighting. The prior construction (1) of the evolution curve $\Delta \lambda = f(\Delta T_W)$ on VIP (example Fig. 15) which indeed includes the contribution of air dry and (2) the water vapour permeance of the VIP, allow to deduce (Eq. (1)) the duration of exposure to reach the permissible maximum conductivity $\lambda_{\text{max}}$. This approach could also be summarized by Eq. (32) presented Section 6. In this approach, the ageing of the silica core is taken into account because it occurs during the test performed to obtain the $\Delta \lambda = f(\Delta T_W)$ relationship.
5.3. Example 3: Most common practice

The third example uses the decoupled lambda model (Eq. (15)) with a linear relation where the moisture impact is added to the dry air impact (Eq. (18)) [14]. The corresponding kinetic is given in Eq. (23).

\[ \lambda = \lambda_0 + B \times \tau_{W} + G \times P_a \]  
\[ \lambda(t) = \lambda_0 + B \times \tau_{W,\infty}(1 - e^{-t \times \Delta m_{W}(t)/\tau_{W,\infty}}) + G \times \Delta P_a(t) \times t \]  

In this approach, recommended in the ISO project 16478, the coefficients B and G are determined by conductivity measurements of VIPs whose envelope is cut open and re-welded at different humidity (ice input) for B and at different pressure of dry air for G. The short comings of this method are:

- The ageing of the core silica material is not considered because the duration and the temperature are not sufficient to let the ageing to proceed. Some improvements could consider B(\text{T}) and/or \text{T}_{W,\infty} (T, \text{RH}).
- The contribution of the inner water vapour pressure on gaseous conduction is not individual but it is included in the B coefficient where dominates the contribution of solid conduction.
- This could be justified for the VIP with a silica core but not for those with mineral wool or cellular polymers. For these cores Eq. (18) must be used and if there are getters their capacity has to be considered and the extrapolation after some month is not usable.

Another difficulty is the determination of the rates of water vapour uptake \( \Delta m_{W}(t) \) and pressure increase \( P_a(t) \). Measuring this accurately for 6 months at 23 °C-50%RH is possible but difficult because the pressure increase and the water uptake remain very small. For example the water vapour uptake of a panel (0.02 x 1 x 1 m³) made with a good current laminate is only 0.15 g. Of course these conditions are not relevant for applications where the temperature or the humidity are higher.

Doing that in a shorter duration, and/or with greater evolutions, or for applications with higher temperature and/or humidity needs an accelerated ageing, so we have to deal with the limitations mentioned for this in the paragraph describing the lump sum method.

We also have to know the equivalence between the accelerated ageing and the real climatic loading (Eqs. (24) and (25)), it needs also to determine the acceleration factors \( F_W \) and \( F_a \) obtained in the ageing test with respect to the real application.

\[ \frac{\Delta m_{W}(t)}{F_W} = \frac{[\Delta m_{W}(t)]_{\text{ACC}}}{F_W} \]  
\[ \frac{\Delta P_a(t)}{F_a} = \frac{[\Delta P_a(t)]_{\text{ACC}}}{F_a} \]  

To calculate them we must know for what application the evaluation is done, in other words what are the successive conditions of temperature and humidity imposed to the VIP. The acceleration factors are given by:

\[ F_W = \frac{\Phi_{W,\text{ACC}}}{\Phi_{W,\text{U}}} \times \frac{t_{W,U}}{t_{W,\text{ACC}}} = e^{-\frac{Q_{a,W}}{R/TU,f,i} \times \sum_{i=1}^{f} P_{W,\text{ACC}} \times t_{U,i} - \frac{Q_{a,W}}{R/TU,f,i}} \]  
\[ F_a = \frac{\Phi_{a,\text{ACC}}}{\Phi_{a,\text{U}}} \times \frac{t_{a,U}}{t_{a,\text{ACC}}} = e^{-\frac{Q_{a,a}}{R/TU,f,i} \times \sum_{i=1}^{f} P_{W,\text{ACC}} \times t_{U,i} - \frac{Q_{a,a}}{R/TU,f,i}} \]  

There is no difficulty for the conditions (t, P) but appropriate activation energies of permeation \( Q_{a,W} \) and \( Q_{a,a} \) are needed. These values strongly depend on the polymers and on the nature of the defects in the barrier layers; the relevance of the chosen values has to be verified. Note that using the service mean temperature instead of the sum over all periods is erroneous because of the thermo-activation.

In practice, because the permeation activation energies for water and air are unequal, it is impossible to reach the same acceleration factors except to strongly reduce the humidity or to increase the air pressure during the test. Reducing humidity is not relevant because we have to generate the thermo-hygroscopic ageing of the core during the test, and increasing the air pressure needs an autoclave device which is not easily available.

Two possibilities of exploitation exist depending on the knowledge or not of the coefficients B and G. If they are known, the steps to follow are summarized on the flowchart Fig. 16. If they are not known, the approach is described in the next paragraph.

5.4. Example 4: Lump sum method

The fourth and last example in this section of assessment methods, allows the authors to draw the attention to the risks of the popular lump sum method of assessment where a short laboratory test (at high temperature and/or humidity) is used as a method equivalent to the long service use (lower temperature and/or humidity).

Defining a representative accelerated ageing protocol is very difficult, a well known fact in materials science but absolutely not in product technology. So a lot of mistakes are generated when taking this approach. Some very common ones are:
Perform tests at temperatures too close or above a transition temperature like the glass transition ($T_g$) of the polymers. This is the case with the ageing at $80\, ^\circ C$ of VIP where the barrier multilayer contains PET film with a $T_g$ around $70\, ^\circ C$.

Perform tests at humidity conditions over $90\%$RH, conditions leading to micro-condensation in micro-voids thereby enhance corrosion of aluminium layers. This could be relevant or not depending on the actual conditions loading the VIP.

Perform test at too high temperatures and humidity. Such conditions lead to the hydrolysis of the PET layers and the laminating adhesives.

It is interesting to note that the same problem is encountered in the photovoltaic lifetime assessment; mainly because there are fundamentally two different needs addressed by accelerated ageing tests. On one hand the qualification of raw materials, components and fabrications where some very rapid technologic tests are useful even if they are not scientifically relevant. On the other hand the evaluation of lifetime and certification of the suitability for use during very long time where the tests have to be absolutely representative.

To be representative accelerated tests must fulfil the following conditions:

- They should cover a range of temperature and humidity where the same mechanisms as in real life are activated;
- The acceleration coefficients of each mechanism (i.e. vapoour and air permeation) should be kept as close as possible;
- It should be kept in mind that the activation energies of permeation are very low, leading to a very strong effect of increasing temperature, as in uncoated polymers an increase of $10\, ^\circ C$ is efficient and an increase of $20\, ^\circ C$ is very strong.
- Finally, basic qualification of products should not be confused with an accurate assessment of lifetime.

Therefore it is not possible to define conditions for the accelerated test equivalent to a service life of years, i.e. 25 to 50 years. However it is possible to define a moderately accelerated test during some months to determine the conductivity increase rate in these conditions; then by using the acceleration factors (Eqs. (26) and (27)) the real conductivity increase rate in service can be estimated. This approach is summarized in the flowchart of Fig. 17. Because of the unequal factors $F_W$ and $F_a$ one has the choice to use a mean value (Eq. (28)) or a conservative one (Eq. (29))

$$[\Delta \lambda(t)_{mean}]_U = \frac{2 \times [\Delta \lambda(t)]_{ACC}^{\text{middle}}}{F_W + F_a}$$  \hspace{1cm} (28)

$$[\Delta \lambda(t)_{max}]_U = \frac{[\Delta \lambda(t)]_{ACC}^{\text{middle}}}{\text{MIN}(F_W; F_a)}$$  \hspace{1cm} (29)

Table 5 shows examples of calculated acceleration factors based on (i) the exposure conditions for unprotected VIP given in the paragraph “Operating conditions”, (ii) the activation energies of the classical laminates made with three metalized PET layers given in the paragraph “Severity criteria”. The intended test postulated here is a proposal trying to reach a compromise between the maximum allowed temperature, the sensitivity of the involved increase of conductivity and the mismatch between the two factors $F_W$ and $F_a$. This leads to the following proposal:

- Two exposure temperatures ($50$ or $70\, ^\circ C$) depending on the maximum service temperature of the application.
- A moderate water vapour pressure: around $6200\, \text{Pa}$ corresponding to ($50\, ^\circ C 50\%$RH or $70\, ^\circ C 20\%$RH), except for the most severe application identified (roof terrace/balcony) which needs a higher pressure to keep $F_W$ high enough ($70\, ^\circ C 50\%$RH suggested).
- For these conditions, the acceleration factors remains quite low ($F_a$ around 5–7 and $F_W$ around 2–3) but with a high confidence in the representativity of the accelerated tests.
- Another positive point is the ratio $F_W/F_a$ about 2.5 which is very homogeneous and close to the respective influence of humidity and air in the increase of conductivity noted by Brunner [29] in the return on operating experience shown in Fig. 18.

### 6. Interactions between barrier films and core material

In all these examples of assessment methods there were in fact some cross influences of the core material, the barrier envelope and
other parameters. This paragraph has a look on these cross links.
In order to show these links, even they occur really simultaneously
the permeations of water vapour and dry air will be considered separately.

For both the lifetime is reached when the conductivity reaches
the maximum allowable conductivity $\lambda_{\text{max}}$:

- for vapour permeation only, the lifetime $t_{W,\text{max}}$ is reached when
the critical maximum moisture content $t_{W,\text{max}}$ is reached (could
be deduced from data like Fig. 3 or from return of experience like
Fig. 15);
- for air permeation only, the lifetime $t_{a,\text{max}}$ is reached when
the critical maximum pressure $p_{a,\text{max}}$ corresponding to $\lambda_{a,\text{max}}$,
reached (from Eq. (15))

$$\lambda_{\text{max}} = \lambda_t + \lambda_{c,s} + \lambda_{c,g,\text{max}}$$

(30)

For the water vapour, by combining Eq. (1) and

$$t_{W} = m_{W} / m_{\text{core}} + m_{W},$$

one obtains:

$$t_{W,\text{max}} = \frac{1}{\Delta P_{W}} \times x \times \frac{t_{W,\text{max}}}{1 - t_{W,\text{max}}} \times \rho \times \frac{1}{P_{W}} \times \frac{1}{2}$$

(32)

For air, by combining Eq. (1) and the Knudsen relationship (Eq.
(17)), considered at constant temperature and for one pore size:

$$\lambda_{c,a} = \frac{\lambda_{c,s,0}}{1 + (c/P_{a})}$$

(33)

one obtains:

$$t_{a,\text{max}} = \frac{1}{P_{a}} \times x \times \frac{\lambda_{a,\text{max}}}{\lambda_{a,\text{atm}} - \lambda_{a,\text{max}}} \times \frac{1}{\phi} \times \frac{1}{P_{a}} \times \frac{C \times M_{a}}{2R}$$

(34)

Both Eqs. (32) and (34) appear similar with the following terms
(in the same order as in the equations):

- solicitation (outside pressure: water vapour difference Eq. (32)
and atmospheric pressure Eq. (34))
- geometry (thickness)
- criterion of end of life (respectively, maximum humidity content
and maximum gaseous conductivity)
- core material (respectively, density and pore size)
- barrier (permeance at the VIP scale)
- and constants.

In both cases authors find the lifetimes $t_{W,\text{max}}$ and $t_{a,\text{max}}$ related
to the core material and to the barrier film. It is therefore obvious
that some technical or economical optimizations of the VIPs with
respect to the intended applications could be made. It is also obvi-
ous that any lifetime assessment method must consider the four
kinds of characteristics: those of the core material and of the bar-
rier envelope as well as the service conditions and the thickness
of the panels.

We can note that no influence of length and width come from
these equations unlike other works [6,14]. This is because we con-
sider here for educational purpose only permeation through the
current surface. If we consider also the especial permeation through
the peripheral potential defects (welds, interfaces, folds, . . .), these
influences appear in the equations by the length and width parameters.

In practice if the conditions are such that one of the mecha-
nisms is very dominant, it is possible to neglect the other and to
evaluate the lifetime with Eqs. (32) or (34). But if the two gas con-
tributions are significant the lifetime cannot be calculated by taking
the smaller duration between the two previous individual $t_{W,\text{max}}$
and $t_{a,\text{max}}$. In that case one can use Eq. (35) based on the above
equations to obtain the relation $\Delta \lambda(t)$.

$$\Delta \lambda = \Delta \lambda_{c,s,W} + \Delta \lambda_{c,g,a}$$

(35)

with

$$\Delta \lambda_{c,s,W} = \frac{2P_{W} \times P_{W}}{x \times \rho} \times B \times \tau$$

(36)

$$\Delta \lambda_{c,g,a} = \frac{\lambda_{a,\text{atm}}}{1 + (C \times M_{a} \times x)/(2P_{a} \times \tau \times R \times P_{a} \times \tau)}$$

(37)

To explain the cross influences of the different parameters, the
potential relationship which gives $t_{max}$ here from Eq. (35) to (37),
is not so educational than the split cases.

7. Conclusions

It has been shown that the usual applications of VIP in buildings
are not equivalent with respect to ageing. Even without considering
both extremes (roof terrace/balcony and domestic hot water tanks),
and without corrective measures, some are 50 to 115% more severe
than others.

The authors have also seen that four components should be
considered with regard to lifetime: they explained how (1) the
characteristics of the core material and (2) those of the barrier enve-
lope interacted with (3) service conditions and (4) the thickness
of the panels when estimating the life expectancy. This allows to
conclude that any assessment method of lifetime or declared resis-
tance or conductivity which does not take into account the reality
of these four components would be at best unclear and at worst
partially wrong and thus not as reliable as expected.

Another conclusion is that the ageing of the core silica mate-
rial during characterization and during service in VIPs in a severe
application has been proven. Hydroxylsation of the silica leads to
an increase of the specific hydrophilicity while the decrease of the
specific area leads to an increase of the solid conduction of the
silica skeleton. This evolution is often wrongly omitted. The con-
sequences can be very important on the estimation of the lifetime
of the VIP.

References

The authors acknowledge the ANR (French National Research
Agency) and the ADEME (ADEME convention with EDF : 0604C0090)
(French Agency for Environment and Energy Management)
for their financial support of the BARILOS (ANR number
ANR-06-PBAT-0002), ECOISIL (ADEME convention n° 0604C0075)
and EMMA-PIV (ANR-12-VBUDU-0004) projects. Special thanks to
S. Kherrout (ADEME), L. Heymans (Microtherm), C. Pompéo and D.
Quénéhard (CSTB).

through high performance laminates for VIPs and physical characterization of
eries of copolymers based on ethylene terephthalate, Journal of Polymer Science
gases through foils used as envelopes for vacuum insulation panels, Journal of


