Evacuated Glazing with Silica Aerogel Spacers

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Abstract

Glazing has come a long way and are subject to continuous development, especially under the premise of climate change. Highly porous monolithic silica aerogels feature optical transparency at ultra-low thermal conductivity – well below the conductivity of free gas. Thus they are excellent candidates even at ambient gas pressure for filling the interpane gap in glass windows. Aerogel glazing is known to exhibit amazing thermal properties (U_g -values down to 0.5 Wm⁻²K⁻¹) with the drawback of a high price due to difficulties in large-scale production. An alternative approach is using silica aerogel spacers with an areal coverage of just 10 % in combination with partial evacuation of the interpane volume to suppress convection in between the aerogel spacers and panes. With this, the costs for such a glazing may be greatly reduced; however, the mechanical load on the aerogel is significantly increased.

In this paper, we present silica aerogel for application as spacer in the shape of pillars, investigate their ability to withstand mechanical stress, measure thermal characteristics and perform simulations on how they are expected to perform in a double glazing. With a krypton atmosphere of $1 \cdot 10^4$ Pa in the interpane gap, U_g -values down to 0.5 Wm⁻²K⁻¹ are predicted.

Keywords: aerogel, glazing, window, thermal performance, FEM simulation, demonstrator, solar buildings

1. Introduction

Given the rise in earth's temperature, the reduction of the emission of greenhouse gases is of uttermost importance. Due to that, decisive actions have been taken by many countries. The building sector consumes 40 % of Europe's energy demand with space heating as its main contributor (Artola, 2016). The thermal performance of the building skin is made up by opaque and transparent/translucent elements – the latter so far being the weak spot in terms of energy losses. State-of-the-art developments of triple and even quadruple glazing decreased U_g -values down to 0.6 and 0.5 Wm⁻²K⁻¹, respectively. However, acceptance for quadruple glazing both, from user and production side, is hindered by its enormous thickness, heavy weight and low total solar transmittance. The internal glass panes serve mainly as an obstacle to prevent convection within the gas-filled space between the single glass panes. The suppression thereof is not only possible by choosing an appropriate interpane gap but also by decreasing the gas pressure. However, even a relatively moderate vacuum involves an enormous stress on the outer glass panes causing them to collapse. This behavior can be prevented by mechanically supporting pillars.

Glazing equipped with monolithic silica aerogel boards in the interpane gap can achieve similar or better U_g values down to 0.5 Wm⁻²K⁻¹ (Schultz et al, 2005, 2008) than standard windows and have been underway for a long time (Jensen et al., 2004). Silica aerogels exhibit thermal conductivities of 0.013 Wm⁻¹K⁻¹ at ambient conditions (Reichenauer, 2012). Until now, the production of full-size, high optical quality aerogel boards is cumbersome and thus its high projected costs prevent the entrance into mass production.

In the present work, we combine both ideas – vacuum to prevent convection and silica aerogel pillars for the mechanical support of the glass panes. The small, pillar-sized aerogels exhibit the advantage of a simplified manufacturing process with decreased material usage. We investigate the application of aerogel pillars with a radius in the cm-range and an areal coverage of 10 % in a partially evacuated double glazing. In this approach,

the air gap between the panes is evacuated and subsequently filled with krypton up to low pressures to prevent heat transport by convection and to limit the amount of the pricey noble gas. Within the framework of this study, we present the optical, mechanical and thermal characterization of aerogel pillars synthesized and then evaluate their suitability for glazing application by simulations and a demonstrator.

2. Interpane-gap-dependent Ug-values

In typical insulation glazing, the width of the interpane gap is chosen depending on the used filling gas. In figure 1, the U_{g} -values are calculated according to the European Standard (DIN EN 673:2011). Starting from low interpane gap distances, the U_{g} -value decreases for increasing gap distances – similar to opaque insulation materials. At a specific distance, however, when convection sets in (solid lines), the U_{g} -values increase again. Convection depends on the properties of the fluid and is thus gas-dependent (Linstrom, NIST Webbook). The higher the density of the gas, the lower is the interpane distance for the onset of convection and the lower are the obtainable U_{g} -values.



Figure 1: U_g -values of double insulation glazing with different noble gas fillings (filling degree = 100 %) as a function of the width of the interpane gap; dashed lines are the respective curves for the same insulation component at a respective partial vacuum which is suppressing convection.

When applying a sufficient partial vacuum, the convection is suppressed effectively (dashed lines). In that case, the U_g -values keep decreasing for larger interpane gaps. Opposing to standard glazing, the interpane gap can – under these conditions - be chosen freely with the benefit of lower U_g -values. With the use of krypton, and neglecting the pillars' conductivity, U_g -values down to below 0.5 W/(m²K) are feasible for a double glazing with an interpane gap of 30 mm. The required partial vacuum for the suppression of convection depends on the filling gas and the interpane gap. In the European Standard (DIN EN 673:2011) the gaseous heat transfer is described by

$$\lambda_{gas} = Nu \cdot \lambda_{0,gas},\tag{eq. 1}$$

with the Nusselt number $Nu = A(Gr \cdot Pr)^n$ or 1, whichever is higher, with Gr and Pr the Grashoff and Prandtlnumber, respectively, and λ_0 , the heat conductivity of the non-convective gas. A and n are parameters which depend on the mounting position. Setting Nu to 1 in eq. (1) yields the convection limit for a given system. For horizontal heat transfer (vertically positioned window), the gas pressure versus interpane gap relationship is given by

$$p = 8.24 \ 10^6 \left(\frac{T_m \ \mu \ \lambda_0}{9.81 \ \Delta T \ c_p}\right)^{1/2} \ \frac{\delta^{-3/2}}{\rho_0} \tag{eq. 2}$$

for a mean temperature $T_{\rm m} = 283$ K, temperature difference $\Delta T = 15$ K, dynamic viscosity μ , specific heat capacity $c_{\rm P}$ and density ρ_0 of krypton at standard conditions (T = 298.15 K, $p = 10^5$ Pa) and the interpane gap δ . The interpane-gap-dependent convection limit is depicted in figure 2. The graph reveals that e.g. for an interpane gap of 0.02 m the krypton gas pressure in between the panes has to be reduced below about 3.5 10⁴ Pa to suppress convection.



Figure 2: Interpane-gap-dependent convection limiting gas pressure for a vertically mounted window with the filling gas krypton at a mean temperature of 283 K.

3. Aerogel Synthesis and Characterization

With an areal coverage of only 10 % and a gas pressure difference of up to 10^5 Pa between the outer atmospheric pressure and the gas filling, the aerogel pillars serving as spacers for the two glass panes will be subject to large mechanical stress. The requirements for the aerogel spacer therefore include – besides optical transparency and low thermal conductivity – a sufficient stiffness in order to prevent major compression by the external load of roughly 10^6 Pa. The physical properties of silica aerogels are usually interdependent and cannot be adjusted separately, thus setting up a challenge in terms of optimization for the given application.

Silica aerogel pillars were synthesized in a sol-gel process with subsequent supercritical CO₂-drying of the gels (Wang et al., 1991). No hydrophobization of the inner surfaces of the aerogels was performed. For the given application, the optical transmittance, mechanical strength and heat conductivity are the most important properties. We used a full-factorial experimental design with three factors (synthesis parameters) on two levels with one center point based on design of experiments (DoE). With this approach, the functional dependency and interaction of synthesis parameters were correlated for the relevant output quantities.



Figure 3: Wavelength-dependent transmittance (left) and visual transmittance according to the European Standard DIN EN 410:2011-04 (right) of the aerogel sample series analyzed in the framework of a DoE study; the dashed line in the right plot shows the set requirement.

The first output quantity is the visual transmittance, determined from the directional hemispherical transmittance value of an 0.02 m thick sample before drying over the visual spectrum from 380 to 780 nm, weighed with the eye sensitivity curve. By use of a grating spectrometer (Perkin-Elmer Lambda 950 with a wolfram halogen lamp as a light source) the transmission was measured with a photo-multiplier in an integrating sphere. As shown in figure 3 all nine silica aerogels analyzed within the DoE study can be described as "transparent" exhibiting a high transmission. Starting at low wavelengths in the blue spectrum, the transmission is lowest and strongly increases to the red spectrum, giving the aerogel its characteristic bluish haze in reflection due to Rayleigh scattering (Emmerling, 1995). We aimed for a high visual transmittance of more than 50 % for aesthetic reasons, a value which was achieved by samples A4, A7 and A8 (figure 3, right plot).

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Subsequently, the mechanical properties were examined. We measured the Young's Modulus of these aerogel pillars with an ultrasonic test equipment determining the longitudinal ultrasonic speed of each specimen (Gross and Fricke, 1992). The corresponding Young's moduli are depicted in figure 4 (left). Samples with a low transmittance tend to yield higher moduli. The dashed line indicates again the goal set for this value in order to limit the uniaxial compression in the application to 5 %. Thus, only samples A4 and A8 meet the two requirements. In addition, for real life stress testing, we used a mechanical testing machine (Zwick Roell TMZ020/TN2S) and exposed the samples A4, A7, and A8, which suffice the set goal for transmittance, to increasing force until rupture or uniaxial compression up to 30 %, whichever came first (see figure 4 (right)). The Young's moduli, ascertained from the linear part of the curve are lower compared to the values of the bulk material in the ultrasonic measurement because of their geometric shape. The versatility of the material becomes obvious by seeing the huge differences the pressure-compression curves. The complex flexible structure of the aerogel leads to a deviation of the linear progression while still exhibiting an elastic behavior. All samples are stable beyond the target value of 10⁶ Pa with concomitant compressions of 4.4, 7.6 and 25 %, respectively. However, sample A8 stands out, since it boasts a compression of below 5 % for the maximum application pressure of 10⁶ Pa.



Figure 4: Young's moduli determined by the ultrasonic runtime method (left) and with a mechanical testing machine (right).

In order to minimize the heat transfer via the supporting pillars, the thermal conductivity was determined in a hotwire setup described in (Ebert et al., 1993). All previously tested, mechanical robust aerogel pillars exhibit heat conductivities between 0.016 and 0.025 W m⁻¹ K⁻¹ in an argon atmosphere and reach values down to approx. 0.013 W m⁻¹ K⁻¹ for low gas pressures (figure 5). As the thermal conductivity under application conditions in krypton atmosphere will also be dominated by the solid and radiative heat transfer, the same or slightly lower conductivities as in argon are expected in krypton. All three candidates shown in figure 5 exhibit thermal conductivities below the conductivity of non-convective air and thus surpass the requirements set for the application at 0.04 Wm⁻¹K⁻¹. In summary, silica aerogel A8 exhibits properties that satisfy all set requirements and is therefore suited to serve as a support pillar in the partially supported insulation glazing.



Figure 5: Gas pressure dependent thermal conductivity in argon atmosphere of three different types of silica aerogel pillars at room temperature.

The long-term stability under ambient conditions and mechanical load is a known issue for the use of silica aerogels in the building sector. In particular in case of non-hydrophobized inner surfaces, ambient humidity leads to creeping under mechanical load. Therefore, sample A8 was subject to a long-term stress test for 7 days – first at zero relative humidity ensured by enclosing the test machine and rinsing the sample volume with dry nitrogen gas. The sample showed no significant deterioration of its mechanical strength (black curve in figure 6). Subsequently, the same test was repeated at a relative humidity of approx. 50 %. Here, a constant creep of the aerogel sample became apparent (red curve). Since the concept of the window element includes the usage of a drying agent in the gap volume, the mechanical stability of the silica aerogel under dry conditions proves the suitability of the supporting aerogel pillars over the lifetime of several years.

In summary of the DoE, we evaluated a recipe to synthesize a silica aerogel fulfilling all given requirements for the application in a double insulation glazing. With the knowledge of the material parameters, we determine the achievable U_g -value of the respective window in the next section.



Figure 6: Long-term mechanical stress test on silica aerogel specimen A8; solid lines represent the measured uniaxial compression and dashed lines the measured humidity during the test(right y-axis); the black curve is derived at 0 % humidity and shows no creep, the red curve is derived at approx. 50 % relative humidity and shows a significant creeping effect.

4. 3D-simulations of the center-of-pane U-value of the proposed window glazing

The performance of the glazing is crucial for its suitability in the market. In particular, the U_g value must show a significant improvement over the value of double glazing with comparable weight. Since convection is suppressed, the contributions to the heat transfer are given by conduction and radiation. With the supporting pillars and their high emissivity in between the glass panes, there is a possible shortcut for radiative transport, which needs to be quantified and – in case for a high contribution - reduced by antireflective coatings.

Simulations of the intended double glazing were performed using the FEM simulation software COMSOL Multiphysics[®] Version 5.3 to quantify their thermal performance considering heat transport by conduction and radiation. The component consists of two glass panes with an interpane air gap of $2 \cdot 10^{-2}$ m and $3 \cdot 10^{-2}$ m. Aerogel pillars with diameters in the cm-range were evaluated. The spacing between the pillars was set according to an areal coverage of 10 %. The goal was to determine the influence of the thermal radiation contribution in the window component since, compared to full aerogel boards, the radiative heat transport is not intrinsically suppressed. Multiple simulations were performed for two pillar diameters, different emittances of the components and varied heat conductivities.

The U_g -value was first determined for a standard configuration. Subsequently, the effects of different parameters were studied. The standard configuration exhibits an emissivity of the outer pane ε_e of an untreated float glass (0.837), an aerogel conductivity of 0.035 Wm⁻¹K⁻¹, an aerogel emissivity of 0.9 (all values for room temperature), and an interpane gap of 0.02 m. The inner pane emissivity ε_i was set to the state-of-the-art value 0.03 for a low-emissivity (low-e) coating. The U_g -value calculated for this parameter set equals 0.91 Wm⁻¹K⁻¹ and is slightly better than the expected value for conventional double insulation glazing. Better values are limited by the radiative transport between the pillars and the inner pane.

				U_{g} -value of standard model [Wm ⁻² K ⁻¹]	0.91
standard model		measure		ΔU_g -value [Wm ⁻² K ⁻¹]	
ε _e	0.837	→	0.03	-0.19	
Interpane gap [m]	0.02	→	0.03	-0.12	
$\lambda_{pillars}$ [Wm ⁻¹ K ⁻¹]	0.035	→	0.020	-0.05	
Ø _{pillars} [m]	13.1 10-3	→	19.6 10 ⁻³	-0.05	
spacing [m]	36.7 10-3	→	54.9 10-3		
ε _e & ε _i	0.03	→	0.01	-0.04	

Table 1: U_g -value of the standard configuration and effect of various measures in order to reduce the U_g -value. The parameters of the standard model are shown on the left hand, the measures in the middle and the respective effect on the right side.

The application of a low-e coating to the outer pane suppresses the radiative transport strongly thus significantly reducing the U_g -value to values below 0.75 W m⁻² K⁻¹. The second largest effect is the increase of the interpane gap from 20 to 30 mm diminishing the U_g -value further to 0.60 Wm⁻²K⁻¹. Inserting the thermal conductivity for sample A8 from the previous section with its diameter of 19.6 10⁻³ m and a corresponding spacing of 54.9 10⁻³ m yields a total U_g -value of 0.50 W m⁻² K⁻¹. A further decrease in the emissivity to 0.01 of both outer and inner pane reduces the U_g -value down to 0.46 W m⁻² K⁻¹. The simulated values for the different configurations are shown in figure 7. The upper set of four curves assumes an uncoated outer pane whereas the next four curves correspond to an outer pane with a low-e coating. The red dashed curve shows values for an interpane gap of 30 mm with otherwise the same parameters as the blue dashed curve.



Figure 7: Simulated Ug-values for different emissivities, pillar diameters, interpane gaps and thermal conductivities of the pillars.

5. Demonstrator

In order to evaluate the appearance and behavior under typical stress conditions due to atmospheric pressure, two test window elements of size $0.25 \text{ m} \cdot 0.25 \text{ m}$ with 16 and 12 pillars, an interpane gas pressure lower than 10^2 Pa and a gas-tight edge bond was fabricated (see figure 8). The pillars were ordered in a cubic and hexagonal spacing as two different design solutions. The interpane gaps were evacuated to pressures below 10^2 Pa; both demonstrators withstood the mechanical stress applied by the atmospheric pressure until the venting of the demonstrator after two weeks of testing.



Figure 8: Demonstrators for evacuated double insulated glazing with silica aerogel pillars positioned in hexagonal (left) and cubic (right) arrangement.

6. Conclusions

In summary, we developed a process for synthesizing silica aerogel pillars with high optical transmittance, increased mechanical stiffness at a low thermal conductivity of 0.020 W m⁻¹ K⁻¹. Simulations reveal, that the implementation of these silica aerogels as pillars inside a partially evacuated double insulation glazing results in exceptional U_{g} -values down to 0.5 W m⁻² K⁻¹. The window element possesses furthermore high transmittance in the visual spectrum and provides high solar gains. For production of the novel window elements, only 10 % of the inter pane volume has to be filled with noble gas, enabling the use of the more expensive krypton, while the standard gas filling process consumes about two times the interpane volume.

Acknowledgements

The results in this paper are a part of the research project HFV (HFV 2016), which is funded by the german Federal Ministry for Economic Affairs and Energy by resolution of the German Federal Parliament.

References

Artola, I., Rademaekers, K., Williams, R., & Yearwood, J. (2016), Directorate General for Energy-European Commission, Boosting Building Renovation: What potential and value for Europe? http://www.europarl.europa.eu/RegData/etudes/STUD/2016/587326/IPOL_STU(2016)587326_EN.pdf. Accessed online 6 September 2018.

DIN EN 673:2011-04: Glas im Bauwesen; Bestimmung des Wärmedurchgangskoeffizienten (U-Wert); Berechnungsverfahren.

Ebert, H.-P., Bock, V., Nilsson, O., & Fricke, J. 1993. The hot-wire method applied to porous materials of low thermal conductivity. High Temperatures-High Pressures, 25(4), 391-402.

Emmerling, A., Petricevic, R., Beck, A., Wang, P., Scheller, H., & Fricke, J. (1995). Relationship between optical transparency and nanostructural features of silica aerogels. Journal of non-crystalline solids, 185(3), 240-248. https://doi.org/10.1016/0022-3093(95)00021-6

Gross, J., & Fricke, J. (1992). Ultrasonic velocity measurements in silica, carbon and organic aerogels. Journal of Non-Crystalline Solids, 145, 217-222. <u>https://doi.org/10.1016/S0022-3093(05)80459-4</u>

Jensen, K. I., Schultz, J. M., & Kristiansen, F. H. (2004). Development of windows based on highly insulating aerogel glazings. Journal of Non-Crystalline Solids, 350, 351-357. https://doi.org/10.1016/j.jnoncrysol.2004.06.047

Linstrom, P. J., Mallard, W. G., Eds., NIST Chemistry WebBook, NIST Standard Reference Database Number 69 (National Institute of Standards and Technology); <u>https://webbook.nist.gov/chemistry/</u>.

Reichenauer, G. 2012. Aerogels: Porous Sol-Gel-Derived Solids for Applications in Energy Technologies, in Lambauer, J., Fahl, U., Voss, A. (Eds.), Nanotechnology and Energy. Science, Promises, and Limits. Pan Stanford, pp. 90-114. ISBN: 9789814310819 9814310816

Schultz, J. M., Jensen, K. I., & Kristiansen, F. H. (2005). Super insulating aerogel glazing. *Solar energy materials and solar cells*, 89(2-3), 275-285. <u>https://doi.org/10.1016/j.solmat.2005.01.016</u>

Schultz, J. M., & Jensen, K. I. (2008). Evacuated aerogel glazings. Vacuum, 82(7), 723-729.

Hochwärmedämmende Fassadensysteme auf Vakuumbasis (HFV) (2016); FKZ 03ET1324A; https://projektinfos.energiewendebauen.de/foerderkennzeichen/03et1324a/.

Wang, P., Emmerling, A., Tappert, W., Spormann, O., Fricke, J., & Haubold, H. G. (1991). High-Temperature and Low-Temperature Supercritical Drying of Aerogels - Structural Investigations with Saxs. *Journal of Applied Crystallography*, *24*, 777-780. <u>https://doi.org/10.1107/S0021889891002327</u>