Long-term Performance of Silica Aerogel and Aerogel Based Composites: A Literature Review Highlighting Pathways for Further Studies

Citation for the original published paper (version of record):
Karim, A., Johansson, P., Sasic Kalagasidis, A. (2019). Long-term Performance of Silica Aerogel and Aerogel Based Composites: A Literature Review
Highlighting Pathways for Further Studies. Proceedings of the 14th International Vacuum Insulation Symposium (IVIS2019): 87-90

(article starts on next page)

Long-term Performance of Silica Aerogel and Aerogel Based Composites: A Literature Review Highlighting Pathways for Further Studies

Ali Naman Karim[1, *], Pär Johansson[1] and Angela Sasic Kalagasidis [1]

[1]Chalmers University of Technology, Gothenburg, Sweden

[*]*Corresponding e-mail: ali.karim@chalmers.se*

ABSTRACT

Aerogels are promising to be used in building materials and insulation products. Among various types of aerogel, silica aerogel is the most commonly type used for building applications. Untreated silica aerogel is hydrophilic, but it can be chemically modified to become hydrophobized. According to the literature review performed in this paper, few studies reporting ageing of hydrophobized silica aerogel under varying hygrothermal (relative humidity and temperature) conditions have been published so far. Among those, studies focused on microstructural evaluation of aerogel during hygrothermal ageing in laboratory conditions reported some reduction of specific surface area and the shift of pore size distribution. This results in a noticeable increase in thermal conductivity of the silica aerogel composite. The objectives of this literature review are to summarize the state-of-the-art studies concerning the impacts of ageing, both accelerated and real-life, in hygrothermal conditions on thermal performance of silica aerogel and aerogel based composite material. The results show that despite the hydrophobic treatment of aerogel, ageing still occur and has, in most cases, a negative impact on thermal performance of aerogel. Relative humidity seems to be the more dominant factor than temperature, concerning the intensity of degradation induced by ageing. Furthermore, a lack of direct correspondence between artificial ageing in laboratory environments and natural ageing is also highlighted.

KEYWORDS
Silica, Aerogel, Accelerated ageing, Durability, Thermal performance

INTRODUCTION

Superinsulation materials (SIM) with the advantage of improved thermal properties and reduced required thicknesses are promising options to the traditional insulation materials (Koebel et al, 2012). Among different types of SIMs, various types of opaque and translucent aerogel-enhanced products have been lately developed and commercially introduced to the market (Baetens et al, 2010). As a consequence of the relatively short real-life history of SIM and in particular aerogel and aerogel-enhanced materials, compared to the normal life time of traditional insulating materials which is about 50 years, the durability and long-term performance of aerogel materials is not fully understood (Chal et al, 2018). Beyond that, due to lack of a common and fully standardized ageing test procedure, various test conditions have been utilized in the (hygrothermal) ageing studies performed on these aerogel materials. Within this work, an attempt to systemize and compare the different studies concerning the durability of aerogel and some aerogel-enhanced products has been done. The aim is to identify possible pathways for further research in this field of science. Focus is on the knowledge of thermal performance of these materials when exposed to various types of accelerated hygrothermal ageing.

METHODS

A literature review was conducted on the published results from studies evaluating the impacts of (accelerated) ageing on the material properties of silica in general and silica aerogel and aerogel-enhanced composites in particular. Generally, an accelerated ageing test is aimed to rapidly evaluate the long-term behavior of a material (Escobar and Meeker, 2006). Among different methods, hygrothermal ageing is a widely used method as weathering is one of the main causes of detorioration. In this artificial ageing process, tested material is exposed to severe hygrothermal conditions, such as elevated temperature, elevated relative humidity (RH), a combination and/or repeated cycles of both elevated temperature and relative humidity. In the studied papers, different authors have been proposed different framework with different hygrothermal conditions. Among the limited number of relevant articles (∼< 20 articles), 5 articles, specifically focusing on accelerated hygrothermal ageing of silica aerogel and aerogel-enhanced materials have been studied closely in the paper at hand.

Overview of published ageing tests

In 1976, Bonsack studied long-term performance of silica powder in terms of surface chemical and physical properties by storing in closed containers during one year (Bonsack, 1976). It was shown that the specific surface area (SSA), the total surface area per unit of mass (Everett, 1972), was reduced by 25-60 %. Morel et al. (2009) studied ageing of pyrogenic (fumed) silica under various temperature and relative humidity conditions. A decrement in surface area and an increment in rigidity and hydrophilic capacity were observed. Another study inspected the changes in properties of mesoporous silica and non-porous pyrogenic silica due to ageing in ambient conditions for 32 years (Collins et al, 2008). The pore size distribution (PSD), i.e. the frequency of different pore sizes, was shifted towards larger sizes and the SSA was decreased by approximately 15 %. Balard et al. (2011) studied the ageing phenomenon of pyrogenic silica for 60 days and in conditions of 25 °C and 95 % RH, resulting in a negligible reduction in SSA. In line with the main objectives of this paper, a summary of the articles focusing on hygrothermal ageing of silica aerogel and aerogel-enhanced materials, studied in this paper, are summarized in Table 1.

Table 1. Ageing studies on aerogel and aerogel-enhanced insulation materials.

Article	Studied product(s)	Ageing factors
1. (Ihara et al. 2015)	Granules	Relative humidity, solar radiation
2. (Chal et al, 2018)	Granules and blankets	Relative humidity, temperature
3. (Bellunato et al, 2004)	Tiles	Irradiation, humidity absorption
4. (Chal et al, 2019)	Granules and blankets	Relative humidity, temperature
5. (Alvey et al, 2017)	Blankets	Relative humidity, temperature

In paper number 1, hygrothermal conditions of 35- 65 °C,100 % RH and a solar radiation simulator with an intensity of 1200 W/m^2 were chosen for the ageing studies. The tests lasted for 3 months including 300 cycles with a duration of 6 hours each (Ihara et al, 2015). By calculating an acceleration factor, the test period was estimated to correspond to 10-20 years in real-world conditions. Shortly, acceleration factors are calculated based on the conditions used, such as temperature and relative humidity range and other material characteristics such as activation energy and reactions rate, to state the correspondence between accelerated and natural ageing. More information about different acceleration factors can be found in (Escobar and Meeker, 2006). It was concluded that thermal conductivity of the aerogel granules could increase by approximately 10 % due to hygrothermal ageing. Ageing by radiation resulted in a negligible deterioration in the hydrophobicity of the aerogel surface. According to the authors of paper 1, occurrence of silica-network breakage in conjunction with the ageing process was the main reason for the observed property changes. In paper 2, 5 different commercial products with hydrophobic-treated aerogels, were studied for periods of 96 to 384 days in ageing conditions of 50 °C, 70% RH and 70 °C, 90% RH (Chal et al, 2018). The aerogel products were either granules or blankets, with different initial SSA and different hydrophobic agents. A decrement in the hydrophobicity was observed at the first period of ageing, followed by a reduction of SSA and a shift of PSD towards higher values at the later stages of ageing. The magnitude of these changes varied broadly depending on the product and the hydrophobic treatment of the aerogels. Thermal conductivities of all products were increased by up to 2.5 mW/mK. They also showed that different commercial products responded differently to the ageing process. Hydrophobizisation quality and initial PSD were identified as the two governing parameters for thermal efficiency in severe hygrothermal conditions. Paper 3 studied the performance of hygroscopic and hydrophobic aerogels when irradiated with Gamma radiation (γ), protons and neutrons, and when absorbing humidity (Bellunato et al, 2004). When the aerogels were exposed to γ and protons no changes were observed, while for neutrons a moderate degradation in term of clarity was observed. For the humidity test, aerogels were aged for one week in the conditions of 22-30 °C and 55-80 % RH, resulting in some degradation of the material properties. However, by baking the samples at approximately 500 °C the initial material properties could be restored. In paper 4, several silicas, aerogel and aerogel-enhanced products were aged in conditions of 50-70 °C and 70-90% RH for 24-96 days (Chal et al, 2019). A reduction of SSA and a shift in PSD were observed for all products but with various intensities. The conclusions were that for the short-term ageing of both precipitated and fumed silica, temperature was the governing parameter, while for long-term ageing, relative humidity was the governing one. Similarly, relative humidity was the decisive parameter for aerogel but in that case the intensity of the degradation due to ageing seemed to be product specified. In paper 5, 3 different aerogel blankets were studied for 5 weeks and in conditions of 65.6 °C, 30/60/90 % RH and 32.2 °C, 90 % RH (Alvey et al, 2017). It was found that the increased moisture content in the blankets increased the thermal conductivities of the blanket differently. It was concluded by the authors that parameters such as volume expansion and hygroscopicity have also an impact on the thermal performance of the aerogel blankets.

DISCUSSIONS

Among the limited number of studies focusing on ageing and long-term performance of aerogels, some characteristic trends can be identified. An increase in SSA and shift in PSD towards larger sizes, negatively affecting the hygrothermal properties, i.e. increasing the moisture capacity and thermal conductivity seem to occur repeatably in different studies. Looking at the ageing effects on thermal property of aerogel it is also clear that ageing can have a negative impact. However, due to the diversities in the results from these studies, in terms of the magnitude of the ageing-induced degradation, it is difficult to state a concrete relation between ageing and aerogel properties. Also, due to the variety of the chosen conditions for the laboratory experiments performed, namely different time intervals, hygrothermal conditions, irradiations (solar radiation) and different products, different production techniques, conditioning, etc., it is difficult to make a direct comparison between the different studies. It is also noted that in some studies, the time of the accelerated ageing is interpreted to real life time by estimating an acceleration factor for the test. However, accuracy and validity of these estimations for materials in nanoscale, such as aerogel seems to be not fully elaborated.

CONCLUSIONS

Based on the literature review performed, it is concluded that ageing of aerogel can affect the material properties and result in a worsen thermal performance. Variations in relative humidity seems to be one of the most crucial parameters, even if the aerogel particles are hydrophobically treated. The magnitude of the deterioration by ageing seems to be product depended. It can be concluded that there is a lack of knowledge concerning ageing of aerogel and correlation between accelerated ageing and real time ageing. To fully capture the performance of aerogel-enhanced product and to increase the market penetration of these insulation products, ageing of aerogel needs to be fully understood. Further studies focusing on ageing of aerogel particles, based on standardized framworks, is therefore recommended.

ACKNOWLEDGEMENT

The financial support from the Swedish Energy Agency (46822-1) is gratefully acknowledged.

REFERENCES

Alvey, Jedediah B., Jignesh Patel, and Larry D. Stephenson. 2017. “Experimental Study on the Effects of Humidity and Temperature on Aerogel Composite and Foam Insulations.” *Energy and Buildings* 144: 358–371.

Baetens, Ruben, Bjørn Petter Jelle, Jan Vincent Thue, Martin J. Tenpierik, Steinar Grynning, Sivert Uvsløkk, and Arild Gustavsen. 2010. “Vacuum Insulation Panels for Building Applications: A Review and Beyond.” *Energy and Buildings* 42 (2): 147–172.

Balard, H., J.-B. Donnet, H. Oulanti, T. Gottschalk-Gaudig, and H. Barthel. 2011. “Study of Aging of Pyrogenic Silicas by Gravimetry and Microcalorimetry.” *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 378 (1–3): 38–43.

Bellunato, T, M Calvi, C Coluzza, G Longo, C Matteuzzi, M Musy, P Negri, and D.L Perego. 2004. “Study of Ageing Effects in Aerogel.” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 527 (3): 319–328.

Bonsack, J.P. 1976. “Effect of Aging on Surface Properties of Silica Powders Produced by Plasma-Arc Vaporization.” *Powder Technology* 15 (1): 53–62.

Chal, B, G Foray, B Yrieix, … K Masenelli-Varlot - Microporous and, and undefined 2018. n.d. “Durability of Silica Aerogels Dedicated to Superinsulation Measured under Hygrothermal Conditions.” *Elsevier*.

Chal, Bruno, Bernard Yrieix, Lucian Roiban, Karine Masenelli-Varlot, Jean-Marc Chenal, and Geneviève Foray. 2019. “Nanostructured Silica Used in Super-Insulation Materials (SIM), Hygrothermal Ageing Followed by Sorption Characterizations.” *Energy and Buildings* 183 (January): 626–638.

Collins, K.E., M.C. Gonçalves, R.B. Romero, R.F. Conz, V.R. de Camargo, and C.H. Collins. 2008. “Low Temperature Ageing of Silicas Gasil-I and TK800.” *Applied Surface Science* 254 (13): 4029–35.

Escobar, Luis A., and William Q. Meeker. 2006. “A Review of Accelerated Test Models.” *Statistical Science* 21 (4): 552–577.

Everett, D. H. 1972. “Manual of Symbols and Terminology for Physicochemical Quantities and Units, Appendix II: Definitions, Terminology and Symbols in Colloid and Surface Chemistry.” *Pure and Applied Chemistry* 31 (4): 577–638.

Ihara, Takeshi, Bjørn Petter Jelle, Tao Gao, and Arild Gustavsen. 2015. “Aerogel Granule Aging Driven by Moisture and Solar Radiation.” *Energy and Buildings* 103: 238–248.

Jelle, Bjørn Petter. 2012. “Accelerated Climate Ageing of Building Materials, Components and Structures in the Laboratory.” *Journal of Materials Science* 47 (18): 6475–6496.

Koebel, Matthias, Arnaud Rigacci, and Patrick Achard. 2012. “Aerogel-Based Thermal Superinsulation: An Overview.” *Journal of Sol-Gel Science and Technology* 63: 315–339.

Morel, B, L Autissier, D Autissier, D Lemordant - Powder Technology, 2009. “Pyrogenic Silica Ageing under Humid Atmosphere.” *Elsevier*.