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A review on silica aerogel-based materials for acoustic applications

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ABSTRACT

Silica aerogels are popular in terms of production volume and real-world applications. Although the current market growth rate is driven exclusively by thermal insulation, aerogels may also be attractive for acoustic applications with the potential in aiding sound absorption/insulation. This paper is a summary of the acoustics related studies of silica aerogel-based products. It introduces silica aerogels, some acoustic characterization methods, and reviews systematically the available data on sound absorption/insulation of silica aerogels, polymer-silica aerogel composites, nonwoven-silica aerogel blankets, and aerogel renders/glazing. The work identifies areas where further research is required, including experimental and theoretical work on the physics of sound absorption in mesoporous materials, and more systematic and standardized evaluations of the acoustic properties of aerogel and aerogel-composites. Aside from this call to action, the opportunities and barriers for the commercialization of silica aerogel products for acoustic applications are presented.

1. Silica aerogel

Aerogels are predominantly mesoporous, open-cell solids with large internal porosity and hence low density [1–3]. The microstructure, more than the specifications of the material that makes up the tortuous network of nanoparticles or fibers, is primarily responsible for aerogel's exceptional material properties, such as high surface area, high mesoporosity, and ultra-low thermal conductivity. Aerogels are typically derived from wet gels, themselves prepared by sol-gel processes, and are dried using supercritical fluids, most often CO₂, freeze-drying, or evaporative drying [3]. A wide variety of materials, including polymers, biopolymers, and metal oxides can be turned into aerogels, but silica aerogels are by far the most common, particularly in terms of production volume [4]. The most widely adopted commercial application of silica aerogel capitalizes on the ultra-low thermal conductivity, which is reduced to half of that of standing air because the mesopores are smaller than the mean free path length of the air molecules. Silica aerogel thermal superinsulation constitutes a rapidly globally growing market. Other potential applications of aerogels include acoustic insulation, catalysts and catalyst supports, gas filters and gas storage materials, conducting and dielectric materials, but these have not yet made a significant impact in the market [5, 6].

1.1. Synthesis

1.1.1. Gelation

A silica gel is produced by a sol-gel process where a silica sol, i.e. a stable colloidal suspension of silica nanoparticles, is destabilized, typically through the addition of a gelation catalyst to change pH and thereby surface charge. The gels can be classified according to the pore fluid, e.g. hydrogel (water), organogel (organic solvent), alcogel (alcohol), and aerogel (air) [7, 8]. During the sol-gel transition, the primary particles are formed and then they aggregate into the secondary particles (clusters), and finally interconnect in a pearl necklace morphology [9], as shown in Figure 1. Industrially relevant silica precursors include waterglass, ion-exchanged waterglass, and silicon alkoxides [10–13]. Waterglass is a sodium silicate solution with a Na/Si molar ratio above 1.5. It can be gelled by the addition of acid (partial neutralization). It is arguably the most inexpensive silica precursor. The sodium ions in waterglass can have a strong, negative effect on microstructure and properties. Therefore, ion-exchanged waterglass, a silicic acid solution obtained, e.g. by passing waterglass solution through an ion-exchange resin, is another common precursor. Ion-exchanged waterglass can be gelled by the addition of base (partial neutralization) and it is more flexible in its use compared to non-ion-exchanged

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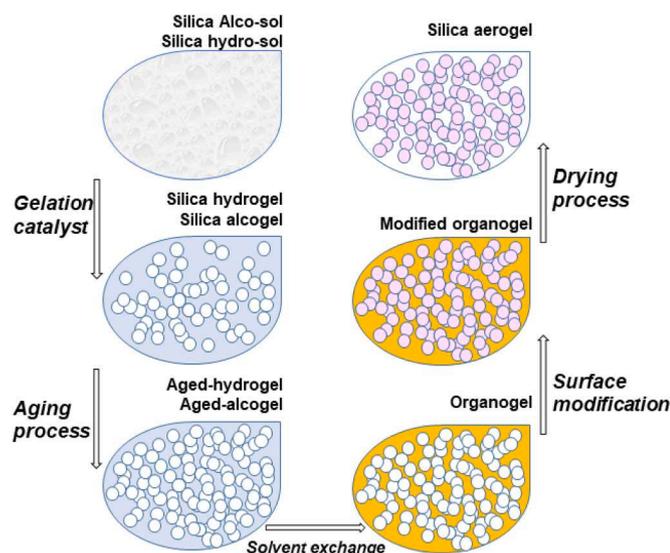
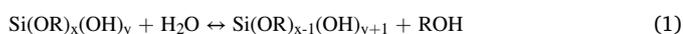


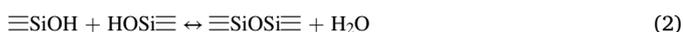
Figure 1. Schematic design of silica aerogels synthesis. The surface modification (typically hydrophobization) is optional. The drying step can be carried out at ambient pressure, using supercritical CO₂ or ethanol drying.

waterglass, but the ion-exchange step adds a considerable cost. The gelation solvent for both regular and ion-exchanged waterglass is typically water, which may necessitate additional solvent exchanges during subsequent processing, although ethanol can be added as a co-solvent [14, 15], and single-step exchanges have been developed [11, 16–19].

Silicon alkoxides, particularly tetraethyl orthosilicate (TEOS) and tetramethyl orthosilicate (TMOS) have transformed the aerogel field. Alkoxide based silica sols are produced through hydrolysis (Eq. 1) and water/alcohol condensation (Eqs. 2 and 3, respectively) reactions of the form:



where x and y the initial number of alkoxy ($x=4,3,2,1$) and silanol groups ($y=0,1,2,3$); and R =methyl/ethyl.



These reactions run simultaneously and are catalyzed by the addition of acid or base. Aside from monomeric TEOS, oligomeric ethyl silicates with higher SiO₂ contents are also available. Alkoxide-based aerogels are often produced through a two-step acid-base synthesis procedure where a stable, acidic silica sol is produced from TEOS or TMOS, followed by base-catalyzed gelation [3, 20, 21]. Inherently hydrophobic silica aerogels derived from methyltri(m)ethoxysilane can have exceptional thermal and mechanical properties [3, 22–25], but are not the topic of this review paper, because they are not yet commercially available in large quantities and should be considered as SiO_{1.5}(CH₃) rather than SiO₂ aerogels.

1.1.2. Aging

The gel prepared in the first step is aged in its mother solution, or less commonly in freshly prepared silica. This aging process strengthens the gel by reinforcing the inter-particle necks and prevents excessive shrinkage during the drying step [26, 27] (Figure 1).

1.1.3. Hydrophobization/surface modification

Because of the extreme susceptibility of hydrophilic silica aerogels to be damaged by liquid water and water vapor, the vast majority of commercially available silica aerogels have been hydrophobized prior to drying [28, 29]. By far the most common strategy is silylation of the

silica gel's surfaces with trimethylsilyl groups by soaking gels in a solution of trimethylchlorosilane, hexamethyldisilazane, or hexamethyldisiloxane. Hydrophobization can be a time- and cost-intensive process and there is extensive scientific and patent literature on increasing the effectiveness of the hydrophobization step [11, 17, 30, 31]. Aside from the increased long-term stability, hydrophobization also enables ambient pressure drying as a potentially more cost-effective drying technique (see below).

1.1.4. Drying

Drying is the final, and arguably most critical, step in aerogel production. The small pore size results in very large capillary stress due to surface tension at the solid-solvent-gas interface during evaporative, ambient pressure drying (APD). This capillary stress leads to pore collapse and densification unless special precautions are taken [32]. The first solution eliminates capillary stress by circumventing the pore fluids boiling curve, either through supercritical fluid drying (SCD) at pressures and temperatures above the supercritical point or by freeze-drying (FD) at temperatures and pressures below the triple point. Ice crystal growth during FD increases macroporosity at the expense of mesoporosity, reduces surface area, and is not particularly relevant for silica aerogel production. Supercritical drying directly from the organic solvent (typically an alcohol), which was developed by Kistler for the first-ever produced aerogels nearly a century ago [1], has some inherent limitations in terms of safety because of the solvent flammability and the high temperature/pressure required to surpass approach the critical point, e.g. 243°C and 63 bar for ethanol [33–35], but the efficiency of the process can be increased by confining the samples in molds to limit the need for excess alcohol through a Rapid Supercritical Extraction process (RSCE) [36–39].

An alternative drying scheme, based on CO₂ with its critical point at 31°C and 73 bar, as the supercritical fluid eliminates the problems of flammability and high process temperature, is now a routine procedure in academic research as well as in industrial production, particularly for silica aerogel blankets [27, 35, 40]. Supercritical drying does not require a prior hydrophobization step, but such a treatment is carried out nonetheless for most commercial products to improve service life stability. Even with CO₂ instead of alcohol as the processing fluid, SCD still requires high-pressure autoclaves and, therefore, it is a batch-type process by definition. However, SCD remains an industrially established production method, particularly for silica aerogel blankets and polymer aerogels. The discovery that silylation of the silica surfaces effectively prevents silanol condensation (see Eq. 2) and irreversible pore collapse during evaporative drying [41] opened up the possibility for ambient pressure drying. Ambient pressure drying has now become a routine process in industrial silica aerogel production, particularly for silica aerogel granulate and powder. During ambient pressure drying, capillary forces and concomitant gel shrinkage do occur, but the gels can spring-back to recover most of the original volume if the samples have the required mechanical stability, e.g. through aging [42].

1.2. Silica aerogel properties

Silica aerogel is available commercially in particulate form (granulate and powder) and as fiber-reinforced blankets, but large monolithic pieces of aerogel are not available in significant quantities. Despite the variety of silica precursors, hydrophobization agents, and drying technologies (see Section 1.1), most high-quality, industrially produced silica aerogels have surprisingly uniform properties when recalculated to the aerogel phase itself, i.e. excluding fiber reinforcement or intergranular macroporosity: envelope or bulk densities of ~0.120 g/cm³ corresponding to porosities of ~95%, high mesopore volumes, surface areas of 700–900 m²/g, and thermal conductivities around 15 mW/mK.

1.2.1. Microstructure

Silica aerogels have fractal structures according to small-angle X-ray

and neutron scattering data, and thus have a similar appearance when observed at different length scales. Primary silica nanoparticles (ca. 5 nm diameter) link up to form a pearl-necklace type skeleton that can be visualized by the higher magnification offered by TEM (Figure 2A). At lower magnification (SEM), secondary particles (ca. 20 nm in diameter), which are porous aggregates of primary particles, enclose the aerogel mesopores (Figure 2B). Nitrogen sorption analysis confirms the high mesopore volume, even though extracting pore size distributions from the isotherms is hampered by the aerogel deformation during the sorption analysis [43].

1.2.2. Thermal conductivity

Silica aerogels are thermal superinsulators with thermal conductivities as low as half that of standing air (less than 15 mW/mK for aerogel versus 26 mW/mK for air) and this is by far their most unique selling point [2]. Aerogels owe their low thermal conductivity to the small pore sizes (<50 nm) compared to the mean free path length of the gas molecules (~70 nm for air at ambient pressure and temperature), which limits gas-phase conduction through the Knudsen effect. In addition, solid conduction through the silica skeleton is limited by the highly tortuous network structure of nanopores [44].

1.2.3. Mechanical properties

The highly porous and tortuous, pearl necklace structure of silica aerogels is highly effective at reducing thermal conductivity, but it inevitably limits mechanical strength. Therefore, it is a major barrier against the more widespread adoption of aerogels. The mechanical properties of neat silica aerogels display a complex dependence on the bulk or envelope density (Figure 2(C, D)) [16]. At envelope densities below ~0.090 g/cm³, silica aerogels are not brittle but deform plastically and irreversibly upon compression and their Young's modulus (E) is very low. At higher densities, e.g. above ~0.150 g/cm³, silica aerogels are brittle, albeit their Young's modulus is much higher. At the intermediate densities typical for most commercial silica aerogels, around 0.120 g/cm³, silica aerogels behave elastically and recover most of the original volume after decompression. However, they are still relatively brittle with a rather low compressive strength. As for most aerogel materials, Young's modulus displays a power-law behavior on envelope density, with $E \sim \rho^{3.6}$ (Figure 2 (C)) [16].

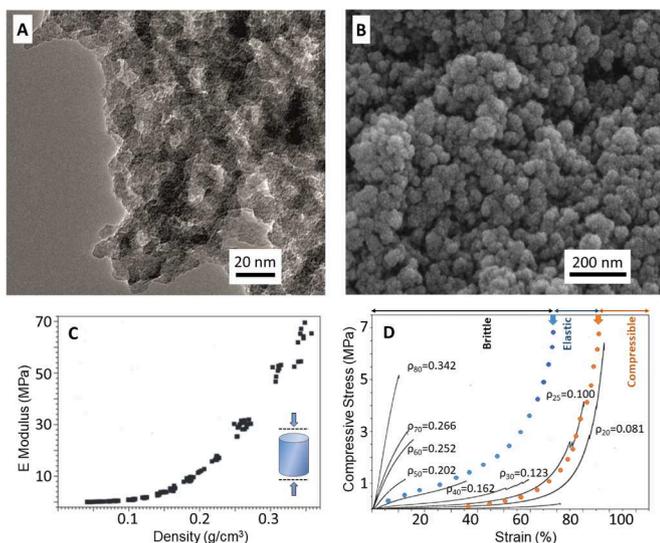


Figure 2. Silica aerogel microstructure and mechanical properties. A) TEM image [31], B) SEM image [51], C) E-modulus as a function of density, D) Stress-strain curves during uniaxial compression: ρ_x represents density (g/cm³) and x corresponds to the volume concentration of polyethoxydisiloxane in the initial colloidal suspension [16]. Revised/reproduced with permission from Ref [16, 31, 51].

Reinforcement of silica aerogels has been a very active field of research and seminal studies by Leventis, Meador and co-workers who have shown impressive improvements in mechanical strength albeit at the cost of higher densities and thermal conductivities and reduced translucency [45-48]. More recent studies have shown that reinforced aerogels can maintain their low thermal conductivity and achieve modest improvements in mechanical strength [49, 50]. None of these stronger silica aerogels are currently available in industrial quantities, but commercialization is ramping up through start-up companies, e.g. Aerogel Technologies (USA) and TIEM Factory (Japan). The aerogel industry overcomes the mechanical drawbacks in two main ways: i) through the production of particulate silica aerogel (powder or granulate) as a semi-finished product that is incorporated into a matrix to impart mechanical strength; and ii) through the incorporation of silica aerogel into a fine fiber blanket that improves handling and mechanical stability, e.g. by impregnating non-wovens with a silica sol or silica aerogel slurry.

1.3. Aerogel Applications

Thermal insulation is by far the dominant application of aerogels that has been successfully transferred to the marketplace [44, 52]. The main markets are pipeline insulation (oil-and-gas), aerospace, industrial insulation, and building insulation. The production ramp-up by Aspen Aerogel (blankets) [53, 54] and Cabot (granulate) in early 2000 has been followed by strong market growth and the entry of new, mostly Asian producers in the last 5 years. Aside from thermal conductivities, a wide variety of other applications that take advantage of the other exceptional properties have been proposed (Table 1), but until now, none of these have made a significant impact on the market.

2. Sound absorption and insulation

2.1. An introduction to sound absorption/insulation

Noise is defined as any perceived sound that is objectionable to a human being [85]. This harmful noise must be controlled to have an acoustically pleasing and safe environment [102]. There are several methods to control the noise's adverse effect either by sound insulation or sound absorption according to the end-use requirements [85]. When a sound wave impinges on a material, it is partially reflected, transmitted, and absorbed.

Sound absorbing materials usually are low density porous materials, with a moderate airflow resistance which can absorb most of the sound energy and prevent sound reflections by allowing sound to penetrate their open cavities or channels [85]. Porous absorbing materials can be classified as cellular, fibrous, or granular materials, e.g. foams, non-wovens, or porous concrete, respectively. Depending on the solid skeleton, and the size and geometry of the pores, the air molecules within the pores/channels of the porous material are forced to vibrate and lose some of their original energy through conversion into heat due to thermal/viscous losses at the walls of the interior pores. In fibrous materials, much of the sound energy can also be absorbed by viscous friction and inertia effects around the individual fibers. Sound absorbing materials are often used in conjunction with barriers to improve their sound insulation effectiveness [4]. Porous media dominate noise control applications in most environments, ranging from factories to homes [103, 104]. Figure 3(A-C) shows porous sound absorbing materials and some of the physical models describing their absorbing mechanisms.

However, there are other types of sound absorbing structures such as Helmholtz and plate resonators as well as membrane (micro-perforated) absorbers [103]. Tuned resonators, like Helmholtz resonance absorbers and plate absorbers are those absorbers that their frequency curve of absorption shows a large absorption peak in a narrow band only like materials having holes. The simplest view of a Helmholtz resonator, as shown in Figure 3(D), is an empty bottle with a neck on top. A volume of

Table 1
Selected silica aerogel applications.

Application	Examples	Technology readiness
Thermal insulation	Low heat transfer materials especially for constructions [52-59], other applications like pipes, appliances, transportation, machinery, space vehicles [55, 60-64], and firefighter and thermal protective clothing [65, 66]	Large-scale industrial production [52-54, 56]
Acoustics	Acoustic matching layers for ultrasonic transducers with low sound velocity and impedance [55, 63, 67-74] Sound absorption/insulation especially for Construction [60,62, 63,75-91] Footfall sound insulation [92, 93]	Pilot production [79, 86, 87]
Filtration, separation, and sorption	Liquid/gas filters and absorbers for paints, varnishes, functional liquids [18, 94]	Academic research
Electrical application	Dielectrics, microwave electronics, electrically conductors, electrodes [55, 60, 88] High voltage insulator, sensor material, impedance adjustment, Cerenkov detectors [63] thermoelectric and piezoelectric materials [92]	Academic research
Optics	Silica glass, mirror backings, laser glass, light source, solar windows, anti-reflective layer for solar cells [55, 61] Cherenkov Counters [55, 92]	Niche applications
Space application	Space dust particles absorber [55, 74, 92] Thermal insulators [55, 74]	Niche applications [95]
Kinetic energy absorbing	Tank baffles, star dust impact, shock absorption [63]	Academic research
Fillers	Fillers for paints, elastomers, thermoplastics, and thermosets [8, 63, 82, 96-99]	Academic research
Carriers	Carrier materials for fungi/herbs/pesticides and drugs [63, 92, 100]	Academic research
Catalysis	Biocatalysts, automobile gas pollutant reducers [92, 101]	Academic research

air in and near the open neck vibrates because of the spring behavior of the air inside the bottle. A mass, either an air mass (Helmholtz absorber) or a solid (plate absorber), are coupled with a soft material or even an air volume acting as spring. When sound excites this system at its particular resonance, it dissipates sound power due to its intrinsic damping. The Helmholtz resonator uses a column of air (cross section S and length l) as mass and volume V as spring, showed in Figure 3 (D) [105]. A very common example is a plate containing holes and slits with a layer of porous absorbing material behind it to reduce the stiffness of the spring

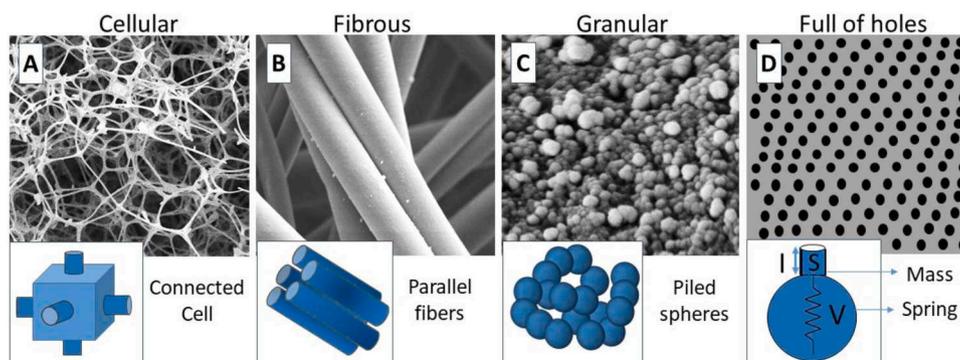


Figure 3. (A-C) The three main types of porous absorbing materials and (D) Resonance absorbers of type Helmholtz.

and increase its damping. In this review article, we see how sound absorption properties of the silica aerogel materials is different from or similar to those of conventional porous or resonance absorbers.

In contrast to sound absorbers, airtight materials, or materials with a high airflow resistance, like steel or composites, are better sound insulators, as sound waves in air have to be coupled to the solid and vice-versa to be transmitted through the layer. In general, the sound insulation effectiveness of a single layer of conventional materials depends upon their stiffness and mass. The mass law states that a material having a high mass per unit area will insulate better. Accordingly, single layers of lightweight materials are not good for sound insulation, but multiple layers of lightweight material with layer of soft material or of air gap in-between are more effective sound insulators in this case. Here, as well porous materials are added in the enclosures between the solid layers to increase sound insulation [4] by reducing the coupling between the solid layers and increase the damping.

However, the open question is whether low-density aerogels can be suitable for acoustic insulation [55, 92]. Sound wave propagation occurs in this type of materials in the fluid of the pores and in the solid skeleton or wave propagation in both phases is coupled. The incident wave is slowed down and attenuated because the wave energy is progressively transferred into heat due to a range of physical phenomena such as inter-particle friction, viscous losses in the material pores, thermal, pressure diffusion and sorption effects. The interstitial gas nature, particle bonds, material properties of the solid, geometry, and distribution of the pores, determine which effect is governing for a particular material. For aerogel-based materials, these effects are not fully understood yet as a wide variety of approaches for material preparation exist. The longitudinal speed of sound in the silica aerogel pores is typically of the order of 100 m/s or less [55, 61] even though this number for non-porous silica is about 5,000 m/s [68].

2.2. Types of acoustic measurements

Sound absorption and insulation are complex functions of intrinsic material properties and extrinsic properties such as surface roughness, sample geometry, and thickness. As a result, collecting accurate acoustical data and their interpretation are more difficult than for data on purely intrinsic properties such as density or thermal conductivity. Many studies that report the acoustic properties of aerogels and aerogel composites do not provide sufficient information of the measurement conditions, intrinsic and extrinsic properties of aerogels to enable a useful evaluation or comparison of their ability to work as a sound absorbers or insulator. Here, we briefly review the techniques, instrumentations, and norms for acoustic measurements.

The impedance tube (ISO 10534-1&2) [106, 107] and reverberation room methods (ISO 354) [108] are the most commonly employed technique to measure the sound absorption coefficient. The sound insulation, or airborne sound transmission loss, of material systems, can be measured on small samples via impedance tube [109] or large

samples as a building material or additional lining on them via standard series of ISO 10140: Part 1-5 [110]. Usually, measurements are made with the high-frequency resolution, but presented in $1/3^{\text{rd}}$ octave bands with the following center frequencies (Hz): 100, 125, 160, 200, 250, 315, 400, 500, 630, 800, 1000, 1250, 1600, 2000, 2500, 3150, 4000, 5000, as specified in ISO 266 [111].

2.2.1. Sound absorption measurement

The sound absorption coefficient (α) is a dimensionless number ranging from zero to one. It is the proportion of the incident sound energy absorbed by the boundary:

$$\alpha = I_a/I_i \quad (4)$$

where, I_a and I_i are the absorbed and incident sound intensities in W/m^2 , respectively. To be a good sound absorption material, the value of α should be as close as possible to one which indicates that a high proportion of the energy in the sound wave incident on the material is absorbed (including absorption within the material structure and transmission) [112].

Measurement of the normal incidence acoustic absorption coefficient in the impedance tube is the most common way to determine the ability of a porous material to absorb sound, especially in the research and development stage. An impedance tube is frequently used in the development of new materials because it is a compact set-up that can quickly determine absorption data at relatively low cost and because only small samples are required. The apparatus is essentially a cylindrical tube with a test sample holder at one end and a sound source at the other end (Figure 4). From the sound pressures in the incident and reflected wave, some of the acoustic properties can be calculated [107].

Impedance tubes typically use one of two methods: (1) the standing wave method (ISO 10534-1) [106] and (2) the transfer function method (ISO 10534-2) [107] (Figure 4). The standing wave method is out-dated at present and the transfer function method, which uses the ratio of pressures between two-microphone positions, is the most common method to measure the absorption coefficient. The minimum frequency range for a tube is controlled by the microphone spacing, whereas the maximum frequency range is controlled by the tube diameter [107].

The ISO 10534-2 method [107] directly measures the complex acoustic reflection coefficient at frequency of f (Hz), from which the absorption coefficient spectrum is calculated as:

$$\alpha(f) = 1 - |r(f)|^2 \quad (5)$$

The reflection coefficient is frequency dependent and controlled by material microstructure and geometry [107, 113, 114].

Measuring reverberation time in a room is another method to determine the random incidence absorption coefficient in which decay

of the sound pressure level is measured with several microphones as a function of time after switching off the sound source. Originally the sound pressure level decay curves were directly determined from the sound pressure level spectra measured with a one third octave band analyzer in very short time intervals after a broad band noise emitted by a loudspeaker was switched off. In-between more advanced measurement methods are very common. In this case the room response is measured with microphones at several positions, when the room is excited with a known input signal, such as a sine sweep. During post processing first the room impulse responses are determined using deconvolution techniques, band filtered and backward integrated to obtain the decay curves of the sound pressure level for each frequency band, from which the reverberation times are evaluated. When sound is emitted in a reverberant enclosed space in the presence of a test specimen the rate of decay of this level (reverberation time) after the sound source is switched off depends on the specimen's absorption coefficient and area covered by it. The sound absorption coefficient α_s of the absorbing layer is calculated as

$$\alpha_s = A_T/S \quad (6)$$

where S (m^2) is the area covered by the test specimen and A_T (m^2) is the equivalent sound absorption area of the test specimen [108]. Often, the reverberation room method is used to estimate the weighted absorption coefficient, α_w , which is calculated through a comparison of the absorption spectra, $\alpha_s(f)$, with a reference curve. Based on the value of α_w , absorbers are classified from A to E, where A corresponds to products with an $\alpha_w \geq 0.90$ [112]. In contrast to the impedance tube methods, the reverberation room is not limited to normal incidence absorption coefficient as it covers all possible angles of incidence, but it does require much larger test samples [107, 108].

2.2.2. Sound insulation measurement

The sound insulation performance of a material is defined in terms of the sound reduction index R , which is defined according to Eq. 7.

$$R = 10 \log(W_1 / W_2) \quad (7)$$

where, W_1 and W_2 are incident sound powers of the incident and transmitted waves, respectively. The sound reduction improvement index ΔR is the difference of the sound reduction indices of a basic element with and without the additional acoustic lining for each third-octave band [109, 110]. The frequency-dependent values can be converted into a single number quantity, R_w .

Like for absorption the airborne sound transmission loss (TL) or reduction index (R) in dB for a normal incident sound wave can be measured in the impedance tube using the 'two-load' transfer function method [109], by acquiring the sound pressure in four fixed

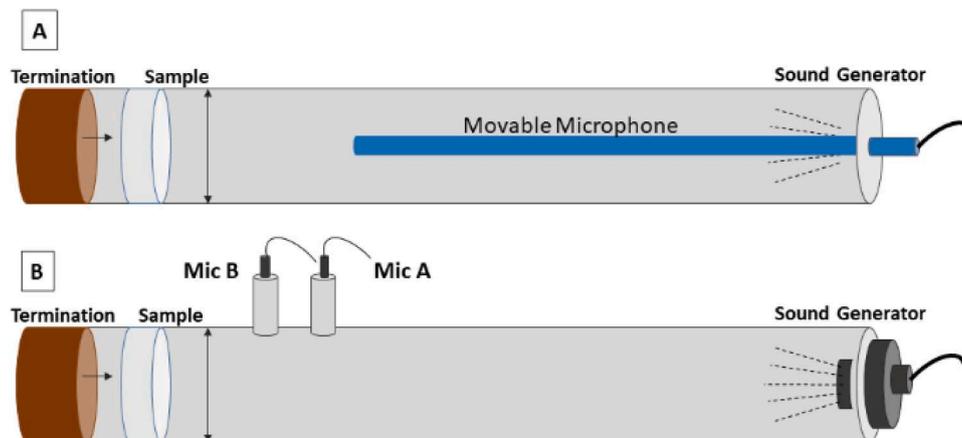


Figure 4. Impedance tubes (A) Standing wave method and (B) Transfer function method.

microphones (two of them between the samples and the sound generator source, and the other two on the back of the sample) positions. Two consecutive acquisitions are carried out for each sample by modifying the characteristics of the tube extremity (a reflective and an absorbing material should be installed) [60]. Unfortunately, this method only is useful for open-cell porous materials with a low to moderate air flow resistance, or limp porous materials with a low bulk modulus of elasticity, where sound propagation in the fluid phase of the material dominates. In most other cases physical effects due to the bending stiffness of material dominate sound insulation, which cannot be captured correctly with the impedance tube.

For these materials, only the measurement of the diffuse field sound reduction index R , for each frequency, according to the standard test series of ISO 10140 provides the correct estimate of the transmission loss [110]. Here, a large sample of the wall or floor separates two sufficiently big rooms. In the source room sound is generated with a loudspeaker and impinges from all possible directions on the specimen that can undergo vibrations like in a real building. The sound reduction index R is determined from the incident and transmitted sound power by measuring the average sound pressure levels in both rooms and, applying diffuse field assumptions, calculating the associated sound power.

Impact sound insulation/reduction is also an important characteristics of resilient layers. Impact sound insulation reduces the sound of footsteps from people walking or falling objects on a floor structure. It is determined by the impact sound pressure level (L_i) in the room below by using a standard tapping machine for generating an impact sound source. A floating floor system in the upper room or a false ceiling in the lower room can be used to improve the impact sound insulation and therefore reduce the impact sound level [115, 116]. The traditional floating floor system can consist of an elastic sound insulation layer and mortar. In addition to the full standardized test, an estimation of the sound insulation performance of a floating floor can be derived from the results of dynamic stiffness measurements performed on small-sized samples (0.04 m^2) [117].

3. Acoustic properties of silica aerogel and aerogel composites

Conventional sound absorbers such as rock wool and open-cell foams are traditionally used for sound absorption and insulation in buildings, but industry and society are looking for alternative, environmentally friendly materials with advantageous sound absorbing/insulating properties [113] and good thermal insulation. Aerogels present an opportunity to combine good thermal insulation performance with useful acoustical properties. Sound absorption and insulation achieved with aerogels strongly depend on the method of material preparation, aerogel density, and pore structure. The sound attenuation in an aerogel relies on the fraction of energy loss of acoustic waves as they are successively emitted from the gas phase to the solid phase, this reduces the amplitude and velocity of the sound waves, causing it to slow down and dissipate faster. This can make aerogels good materials for acoustic insulation [118].

3.1. Sound velocity and impedance in silica aerogels

Silica aerogels [119] display unusual acoustic properties due to their particle network structure, for example extremely low sound velocities in the range of 100-300 m/s for aerogel densities between $0.07\text{-}0.3 \text{ g/m}^3$, compared to 343 m/s in air and $\sim 5000 \text{ m/s}$ for silica glass [120]. Sound velocities as low as 120 m/s have been reported as early as 1984 [67]. Most literature reports indicate that sound velocity increases with increasing density [119], but other factors may also be important. Gross et al. in 1992 [68] suggested that a simple scaling law for sound velocity $u \propto \rho^a$, with $a=1.3$ is true for densities higher than 0.1 g/m^3 , but at lower density, $a=0.8$ is valid. Longitudinal sound velocity in solids is related to the elastic constants (bulk and shear modulus, correlated to

Young's modulus). Heat-treated aerogels display higher sound velocities than untreated aerogels, as heating increases the degree of connectivity of the silica nanoparticle pearl-necklace network structure [121].

In 1986, Gronauer and Fricke [67] studied if the low sound velocity in aerogels was due to the air-skeleton interactions or low Young's moduli. Such effects are known to be important from conventional sound absorbers, where the fibrous structures are also small compared to the sound wavelengths in the auditory range. They concluded that sound propagation occurs via the skeleton and not through the air within the porous system, hence the low Young's moduli are the root cause for the sound velocities smaller than that of air. Sound propagation of the aerogel is primarily influenced by elastic properties of the skeleton when densities are above 0.100 g/cm^3 , whilst atmospheric pressure in the enclosed air space is important for aerogels with a density of 0.05 g/cm^3 [121, 122]. In 1998, Forest et al. presented a comparison of acoustic propagation in alcogels and aerogels and showed an interesting difference: for the high porosity alcogels, longitudinal wave velocity remains around the same velocity as in alcohol, while in aerogels, the velocity is significantly lower than that in air. They applied the Biot's model for acoustic wave propagation in porous media to study the velocity and attenuation. Biot considered the problem of the acoustical propagation in a porous elastic solid saturated by viscous fluid by deriving the equations for sound waves through the solid and fluid fractions [71]. This model is used extensively to predict the acoustical properties of porous elastic solids.

3.2. Sound absorption/insulation in pure/hybrid silica aerogels

A majority of studies reporting on the acoustical properties of aerogels preferred the impedance tube method because of the smaller sample size requirements [63, 75-77, 80-84, 119]. The transmission loss, the ability of aerogel to insulate against incident sound, is usually measured with a four-microphone impedance tube method [123]. Some studies employ their own methods for data evaluation [124], which makes the data comparison difficult. In this section, we focus on the acoustic properties of silica aerogel and its composites with polymers mostly obtained with the impedance tube data. We go through these studies in chronological order.

In 1998, Schmidt et al. [63] reported acoustic properties of pure silica aerogels, hot-press formed plates via dry mixing of silica aerogel with polyvinylbutyral (PVB), as well as silica aerogel bound with liquid vinylacetate/ethene. As in the case of a usual porous layer, aerogel displays a 1/4-wavelength resonance peak which maxima shifts towards lower frequency with increased thickness (see Figure 5). An increase in thicknesses in this study was achieved by adding individual layers and the vibration of each layer might have influenced the measurement. The observed absorption coefficient for a $h = 40 \text{ mm}$ thick layer at frequencies above 600 Hz were around 0.6-0.7 which is a promising result [63]. Higher absorption ($\alpha > 0.6$) in thinner layers was achieved at frequencies at which the acoustic wavelength in the material

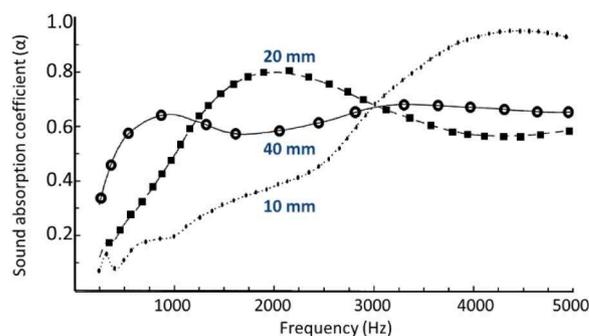


Figure 5. Sound absorption coefficient for different thicknesses of an aerogel particle layer. Revised/reproduced with permission from Ref [63].

was $d = \lambda/4$ [119]. A thermoplastic bound aerogel produced using a polyvinylbutyral binder displayed a lower sound absorption values similar to that of expanded and extruded polystyrene with $\alpha < 0.1$ across all frequencies of interest (see Figure 6). However, relatively thin layers (e.g. 20 mm) of dispersion bound aerogels, made using aqueous vinylacetate-ethene, show a great improvement in sound absorption coefficient particularly at 500 and 800 Hz (see Figure 6).

Schwertfeger and Schmidt, in 2003 [93] patented the use of aerogel-polymer composites for damping of structure-borne or impact sound. In their work, a percentage of aerogel was mixed with PVB or dispersion glue followed with heat treatment which improved impact sound insulation by up to 24 dB for a layer thickness of 18 mm and 90% by volume of hydrophobic aerogel granulate [93]. A substantial acoustic improvement with a relatively low thickness of sound insulation material would be an important tool to solve the significant difficulties of building renovations by separating the mechanical tension of the old building structure from the floor covering [63].

Dong et al. in 2009 [75] produced composite aerogels with variable concentrations of silica and polydimethylsiloxane (PDMS) and evaluated their sound absorption coefficient as a function of composition and average pore size. The acoustic behavior of aerogels developed in this work differed from that in many commercially available porous acoustic absorbers. The behavior of these aerogels resembled that of layer membrane which is controlled by the elastic properties of aerogel. However, Dong's work did not specify the thickness of the studied commercial and aerogel materials. For resonance absorbers, high absorption occurs around a specific frequency, where increasing the amount of PDMS shifts the peak absorption to the higher frequency, ascribed to the reduction in pore size of ormosil aerogel. By some reason, Dong et al. approximated the porous structure of the aerogels with two models of cavities: either with a neck (Helmholtz model, Eq. 8) or a sphere-like shape. They claimed for both models that as the cavity size decreases with the increasing PDMS content, resonance-based absorption occurs at higher frequencies, e.g.

$$f_H = \frac{v}{2\pi} \sqrt{\frac{A}{VL}} \quad (8)$$

where, v , A , V and L , are speed of sound, the cross-sectional area of the neck, volume of the cavity and length of neck, respectively [75].

This resonance behavior is likely to relate to two effects. In the case when the density of aerogel is relatively low and it is granulated to a fine powder (e.g. with particle size to be much less than 100 μm) the incident sound wave causes fluidization of aerogel particles. The aerogel layer behaves like a very light equivalent fluid which causes a multiple peaks in the measured absorption spectra. In the case when a fibreglass blanket is impregnated with this powder, the flow resistivity of the blanket increases dramatically. When exposed to acoustic excitation in the impedance tube the blanket vibrates. This vibration effect together with the circumferential gap effect dominate the measured absorption

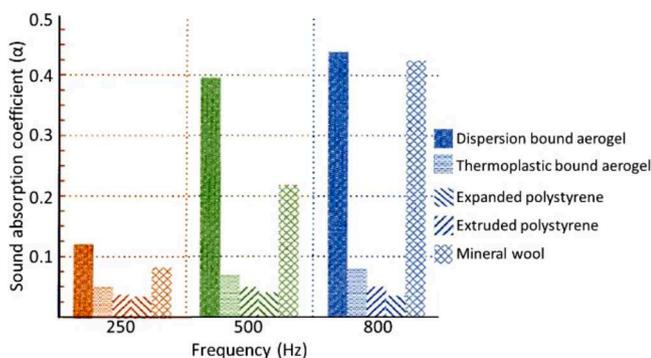


Figure 6. Sound absorption coefficient of different insulation materials at a thickness of 20 mm. Revised/reproduced with permission from Ref [63].

coefficient spectra which tends to have a single resonance peak and limited absorption coefficient beyond this peak. Figure 7 illustrates these sound absorption spectra for a commercial aerogel powder (particle size 2-40 μm , pore diameter 20 nm and particle density 0.12-0.15 g/cm^3) and fibreglass blanket (pore size in the fibers 20 μm) impregnated with this powder. It also provides a comparison against the absorption coefficient for two conventional fibreglass layers. These data were measured by the authors in a 100 mm diameter impedance tube.

Figure 8 illustrates the sound absorption spectra for a commercial aerogel powder (particle size 1-20 μm , pore diameter 20 nm and bulk density 0.04-0.10 g/cm^3) with resonance behaviour, and melamine foam (pore size 115 μm) with porous material behaviour, both with a hard-backed layer thickness of 50 mm. The data was measured by the authors in a 10 mm and 100 mm diameter impedance tube respectively. Comparing Figures 7 and 8 with the previous works [62,73,80], aerogel powder in the micron size range shows continuous peaks across the whole frequency range of 100 to 4000 Hz than aerogels with a larger particle size in the millimetre size range. Resonance behavior of aerogels was also reported by Cai et al. in 2012 [76] for methyltrimethoxysilane (MTMS)-vinyltrimethoxysilane (VTMS) based monolithic aerogels with the thickness of 1.6 cm. Two commercially available acoustic insulation materials, an open-cell polyurethane (PUR) foam with a non-woven scrim (Insulator A, thickness = 2.0) and a non-woven fiber material (Insulator B, thickness = 4.3 cm) were also tested for comparison. Maxima in the absorption coefficient of aerogel material were observed in the frequency range of 540-830 Hz and also at the higher frequency range of 1570-1860 Hz, while insulators A and B exhibit improved acoustic absorption as frequency increases. Strong absorption peaks observed at both low and high-frequency regions may indicate multiple resonances within the aerogel [76].

In 2014 Yan et al. synthesized polyimide-silica aerogel composites and measured their acoustic absorption coefficient in a 16 mm diameter impedance tube [77]. They reported resonance behavior for the sound absorption coefficient with two to three peaks in the 2.5-10 kHz range (see Figure 9). As expected, the positions of the maxima in the absorption coefficient spectra depended on sample thickness: for 10 mm, peaks occur between 5-8 kHz while for a thickness of 30 mm the peaks occur at 2.5, 7, and 10 kHz.

Sachithanadam et al. in 2016 [80] measured the acoustic properties of the silica aerogel granules of various sizes from 0.50 to 3.35 mm, and gelatin-silica hybrid aerogels doped with sodium dodecyl sulfate (GSA-SDS) consisting of 1.2 and 1.7 mm granular size. Absorption coefficients varied across grain size. Larger granules exhibited a somewhat

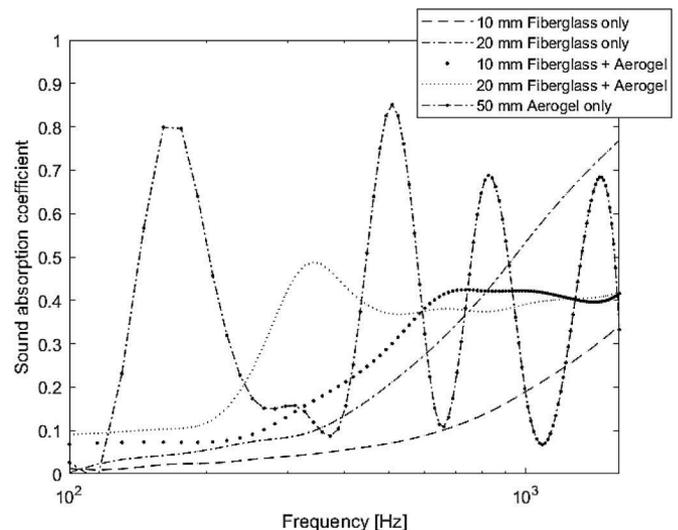


Figure 7. Sound absorption coefficient of silica aerogels and silica aerogel blankets in different thickness, compared to fibreglass sound absorbers.

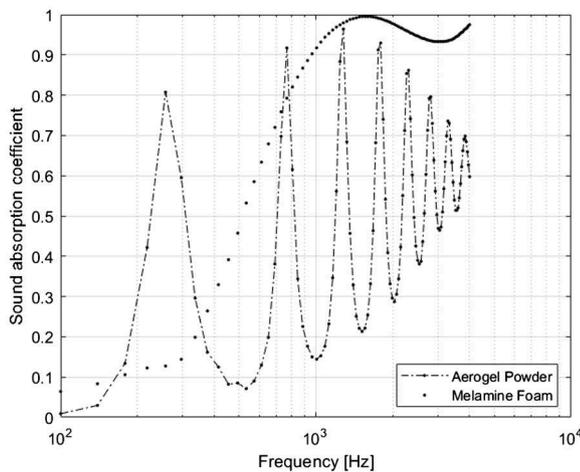


Figure 8. Sound absorption coefficient of silica aerogel, compared to a melamine foam (thickness 50 mm).

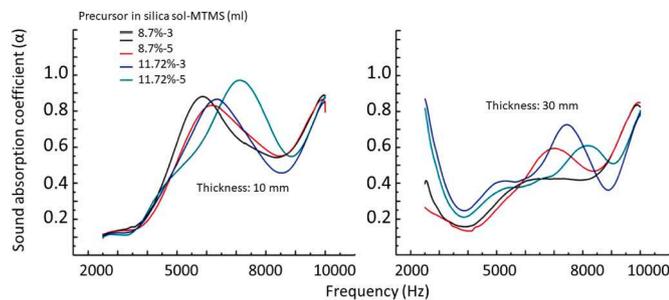


Figure 9. Sound absorption coefficient of polyimide-silica aerogels. Revised/reproduced with permission from Ref [177].

lower absorption coefficient than that measured for smaller granules. This result demonstrated the importance of the visco-thermal and pressure diffusion effects which are controlled by the inter-particle pores. The second aspect of their research was the use of the “Inferential Transmission Loss method” (InTLM) to determine the transmission loss using a three-microphone impedance tube. The approach was a modification to the usual transfer function method by inferring the transmission coefficient with and without the rigid wall. The sound velocity in the GSA-SDS aerogels was found very low at 70 to 78 m/s. Both the silica and GSA-SDS aerogels exhibited resonance behavior in the transmission loss data (see Figure 10) and thus, a different kind of graphs in comparison with the other research [60] with increasing trend graph with one dip in the resonance frequency.

Moretti [60] and Buratti [62] et al., in 2017, experimentally

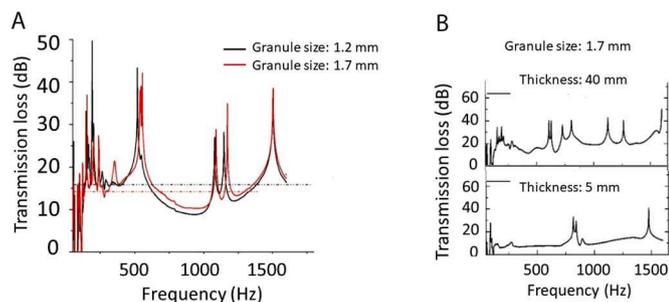


Figure 10. Transmission loss for (A) aerogel granules and (B) GSA-AG composites (granular size: 1.7 mm) via the proposed “Inferential Method” using a two-microphone impedance tube. Revised/reproduced with permission from Ref [80].

investigated the influence of granule size of silica aerogels on thermal and acoustic performance. They used a 100 mm diameter, 4-microphone impedance tube to find that smaller grain sizes, which are accompanied by higher densities, and thicker samples improved the sound transmission loss (see Figure 11).

In 2018, Geslain et al. [83] presented a novel signal processing method for retrieving the viscoelastic properties of a silica aerogel clamped plate. This method is based on a genetic algorithm optimization with two objective functions resulting from two acoustic configurations, a reflection problem, and a transmission problem in an acoustic impedance tube. The two objective functions are the differences between the experimental and modeled acoustic problems around the plate resonance frequency. The estimated aerogel viscoelastic properties were $\rho=80 \text{ kg/m}^3$, $\nu=0.12$, $E_r=197.92 \text{ kPa}$, where ρ is density, ν is the Poisson ratio and E_r is the real part of the Young’s modulus. The absorptive properties of aerogels are encouraging and can be applied to design more complex artificial structures (metasurfaces) for the broad-band absorption of sound [83].

Merli et al. in 2018 [118] used 29 mm and 100 mm diameter impedance tubes to study the sound absorption and transmission losses of monolithic and granular aerogels. In their research, they used small aerogel granules of 0.01–1.2 mm (density of 0.08–0.085 g/m^3), intermediate grain size aerogels of 0.7–4.0 mm (density of 0.065–0.075 g/m^3), larger grain size aerogels of 4.0 mm (density of 0.065–0.070 g/m^3) and monolithic aerogel sample thicknesses of 12.7, 19.1 and 25.4 mm. Monolithic aerogels showed peak absorption coefficient values in 0.54 to 0.88 in the frequency range of 1.1 to 1.5 kHz whereas granular aerogels showed peak absorption coefficient values of 0.9 to 1.0 in the frequency range of 1.7 to 4.1 kHz. They found monolithic aerogels to have a transmission loss of 10–15 dB for sound waves attenuating at a frequency of 100–1600 Hz, when compared to granular aerogels which had a lower transmission loss of 5–7 dB [118].

From the limited number of papers reporting the sound absorption and transmission loss of the pure/hybrid silica aerogels, it can be clear that silica aerogels, in contrast to the other porous sound absorbers,

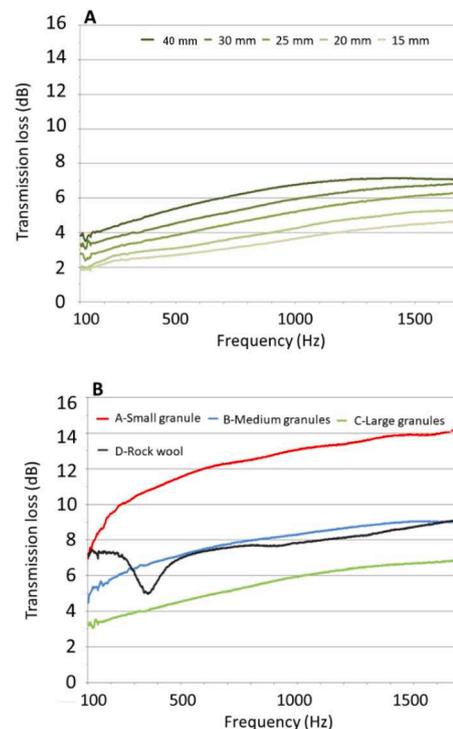


Figure 11. (A) Transmission loss of 1–4 mm silica aerogel granule layers for different thicknesses and (B) Comparison between aerogels and rock wool panel in 30 mm thickness. Revised/reproduced with permission from Ref [60].

display a resonance behavior with one or more resonance frequencies in their sound absorption spectra. The resonance frequencies and width of the resonance peaks depend on their mechanical properties, thickness, sample mounting conditions and potential presence of circumferential air gap. The transmission loss typically rises with an increasing frequency, with a dip in the lower frequency range which corresponds to a layer resonance or resonance in the circumferential air gap. It is possible to qualitatively compare the transmission loss among various aerogels to evaluate the effect of different parameters, but relying on the absolute numbers is not recommended from data collected with the impedance tube.

A key problem to good understanding of the acoustical properties of aerogels is to have accurate data on its pore size distribution and pore connectivity. The physics of the effects which control the observed acoustical behavior is markedly different for different scales of pores. For pore which size is close to free mean path (around 70 nm) the sorption effects dominate. For pores which size is similar or larger than the viscous boundary layer thickness ($\delta = \left(\frac{\eta}{\omega\rho_0}\right)^{0.5}$, where η is the dynamic viscosity and ρ_0 is the ambient density of air) the viscous friction and inertia absorption will dominate. In granulated aerogels with millimeter size particles pressure diffusion absorption can be pronounced. This effect is controlled by the contrast between the relatively large inter-particle pores and relatively small (transport) pores in the surface of aerogel grains. All these effects are difficult to describe in a single theoretical model. Accurate data on the pore size and connectivity are difficult to acquire. As a result, there is a clear lack of publications which study and account for these effects in aerogels.

Last, but not least, there is a clear lack of modeling and simulation of key acoustical properties of monolithic and granulated aerogels, which are the complex, frequency-dependent bulk modulus, dynamic density of the air trapped in the material pores, and complex Young's modulus of the frame supporting aerogel. These properties control the sound speed, attenuation, and acoustical characteristic impedance of aerogel. Data on these key properties are difficult to find in published literature. This issue is complicated by the liberal use of a range of experimental procedures for measuring the acoustical properties of aerogels which seems to be parochial to a particular layer thickness and experimental setup.

3.3. Sound absorption/insulation of silica aerogel-polymer composites

For practical applications, aerogels are usually impregnated in foams or fibrous matrix. Only a few recent studies have investigated the acoustic properties of foam type of aerogel composites. In 2017, Dourbash et al. [81] prepared and acoustically investigated silica aerogel/PUR foam composites and silica aerogel/elastomeric PU. They showed that adding silica aerogel to the polyol did not improve the thermal and acoustical properties of the PUR foams, but did result in elastomeric PU results in better sound insulation characteristics, particularly transmission loss (see Figure 12), which is related to stiffness, absorption potential, and sound attenuation. Increasing the aerogel amount increases the transmission loss in the higher frequency range while adding 1% aerogel material resulted in the highest transmission loss in the frequency range of less than 3 kHz as shown in Figure 12 (B). Adding more silica aerogel to the polyol substantially reduces the density and increases the number of voids present, this leads to lower values of the transmission loss [81].

Eskandari et al. [82], in 2017, investigated the effect of silica aerogel on Unplasticized Polyvinyl Chloride (UPVC) on acoustic and thermal properties. While a melt procedure was used for mixing, they did not investigate the possibility of (partial) infiltration of polymer into the aerogel pores. Sound absorption values of up to 40% were observed above 2500 Hz. At frequencies below 1000 Hz neat UPVC and all of its composites displayed a downtrend due to the resonance phenomenon, which causes a reduction in the reflection coefficient and increases

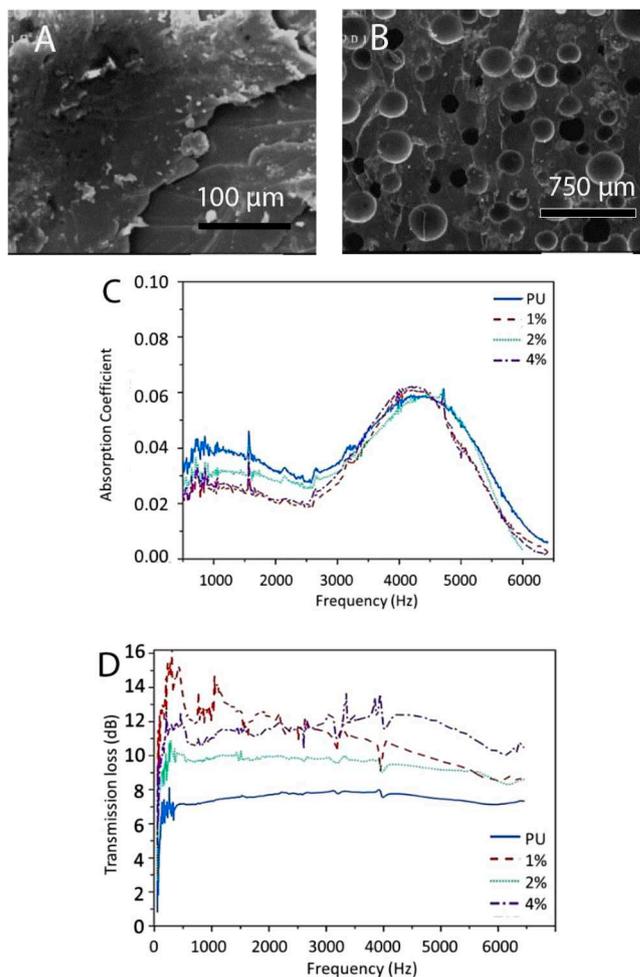


Figure 12. (A) SEM image with no aerogel content, (B) SEM image with 2% aerogel content, (C) Sound absorption coefficient, and (D) Transmission loss, for coating grade PU and its composites with 1-4% aerogel. Revised/reproduced with permission from Ref [81].

transmission [82].

A few studies on the acoustic properties of the other kinds of aerogel materials like recycled polyethylene terephthalate (rPET) aerogels have also been carried out, but this paper focuses on silica-based aerogels and their composites [84].

3.4. Sound absorption/insulation of aerogel-textile compounds

There are a few possible ways for how aerogels can be combined with textiles. As mentioned in Section 3.4.1, nonwovens can be impregnated with a silica aerogel, to produce fiber-reinforced silica aerogel blankets. The often observed dustiness of such blankets motivated investigations on the use of aerogel particles as a porous filler inside the polymer phase, where it is completely protected [64]. Several reports are available where aerogel particles are either mixed with electrospinning PET solution to make aerogel-filled fibers [64], mixed with binder and knife-oated [125], padded [126] or electrospayed [51] onto textiles, or thermally bonded in nonwoven textile [127], but the applications are not related to sound insulation. Here, we review those studies that reported on acoustic properties.

A limited number of reports are available where electrospinning was used to combine aerogel and fibers. A typical approach is to add electrospun nanofibers into a silica precursor before aerogel synthesis to produce aerogel coated fibers [128, 129]. No literature was available regarding the incorporation of aerogel particles in electrospun polymer

fibers, until 2015, when Mazrouei-Sebdani et al. [64] added aerogel particles in a PET electrospinning polymer solution to produce PET-aerogel composite fibers. The addition of silica aerogel particles to the electrospun PET fibers increases the sound absorption coefficient in the frequency range of 250 to 4000 Hz for a sub-millimeter thick layer of fibrous membrane. While 0.5 wt% aerogel-filled electrospun webs exhibited higher sound absorption at low frequencies of 250 and 500 Hz, 4 wt% aerogel-filled samples displayed the highest sound absorption coefficient at intermediate frequencies of 1000 and 2000 Hz. Mazrouei et al. suggested that the effects on sound absorption are related not only to the aerogel porous structure itself, but also to the effects of aerogel addition on the fiber structures and properties. Because the lower aerogel content resulted in larger fiber spacings, low-frequency range sound waves could be absorbed more efficiently, as suggested by other researchers [130]. Because the acoustic property of silica aerogel itself is not fully examined (Section 3.2), more experimental and modeling insights on pure silica aerogel are needed before the effect on aerogel-polymer nanofibers can be fully understood.

A nonwoven fabric is a manufactured web of fibers, bonded/entangled together by mechanical, thermal, or chemical processes [131]. Nonwoven fabrics with high bulkiness and resilience, great compressional resistance, and good filling properties have been extensively studied as one of the most common porous thermal insulating/sound absorbing materials [102, 132]. Since both nonwovens and aerogels have impressive thermal and acoustic properties on their own, a combination of these two materials is widely used in various environments because of their flexible structure. When the aerogel blankets are not encapsulated, problems with the migration of the aerogel particles and dust release exist [56]. Despite these potential drawbacks, fiber-reinforced silica aerogel blankets make up well over 50% of the silica aerogel market, and the thermal conductivity of such blankets has been studied extensively. However, it is also essential to understand the sound absorption behavior of nonwoven-aerogel compounds, which have been studied to a much lesser extent [102].

Oh et al. in 2009 [133] selected two methods for the acoustic optimization of polyethylene terephthalate (PET) nonwoven-silica aerogel blankets by dipping the nonwoven in a TEOS based sol (method I) or by ultrasonication of a silica hydrogel dispersion (method II), followed by solvent exchange/surface modification with TMCS/n-hexane and ambient pressure drying. All measurements were done for the samples with a thickness of 5 mm. Sound absorption coefficients of the method I and II blankets were low and below 0.1 at frequencies below 1000 Hz. At frequencies above 1000 Hz, the sound absorption coefficient of the method I prepared blankets with a density of 0.184 g/cm³ increased steadily and showed a wide bell-shape graph (see Figure 13) [133] in line with that expected from traditional porous sound absorption materials [8, 9, 134].

The presence of silica aerogel had a positive effect on energy

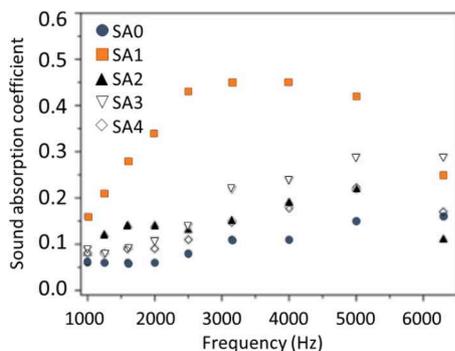


Figure 13. Sound absorption coefficient of the 5 mm thickened PET nonwoven-silica aerogel blankets prepared by dipping a PET nonwoven in a TEOS based sol. Revised/reproduced with permission from Ref [133].

absorption. Therefore, PET nonwoven-silica aerogel blankets, prepared under optimum conditions, can be considered as a good sound absorbing material [133] particularly in the frequency range for which the human ear is most sensitive (2.5 to 5 kHz) [9]. However, a majority of conventional porous insulation materials are also good absorbers in this frequency range. Küçük and Korkmaz, in 2012 [104] investigated the effect of bonding, thickness, composition, permeability, and fiber thickness on sound absorption properties of nonwoven-aerogel composites. They varied the layer thickness from sub-millimeter thick to 35 mm to confirm that the increase in the layer thickness and carefully chosen permeability results in an increase in sound absorption [104].

In another work on PET nonwoven-silica aerogel blankets, Ramamoorthy et al. in 2017 [85] focused on silica aerogel blankets prepared from sol with variable silica content (110 to 28 molar ratio of methanol/TEOS). A molar ratio of 55 was recommended to maximize sound absorption and hydrophobicity. Similar to the work done by Oh et al. (2009) [133], silica aerogel was synthesized with solvent exchange/surface modification with TMCS/n-hexane and ambient pressure drying, but here, structural properties of aerogel composites have also been studied to determine their durability for long term noise control applications [85]. The PET nonwoven-silica aerogel blankets exhibited a higher sound absorption coefficient than the control sample for the entire frequency range of 50–6300 Hz. The silica aerogel present in the nonwoven structures (Figure 14) increases the absorption coefficients due to reduced average pore sizes, which cause stronger visco-thermal effects [85]. Also, airflow resistance has increased in the mesopores due to the presence of aerogel leading to higher sound absorption [85, 102].

In 2015 Motahari et al. [135] used cotton instead of PET to produce cotton nonwoven (1 cm)-silica aerogel blankets through a single-stage sol-gel process with different MeOH/TEOS molar ratios and aging times, with the aim to produce an efficient sound absorber. The results showed that for high MeOH/TEOS molar ratios (11) and a long aging time (24 hours), significantly higher sound absorption coefficients below 3 kHz can be achieved with blankets with a relatively low bulk density of 0.088 g/cm³. This could be due to the increase of fiber diameter after coating with silica aerogel particles, which decreases the space in between fibers in the mat and increases viscous stress caused by shearing and friction of air across the pores of silica aerogels. A further increase of the MeOH/TEOS molar ratio to 14 caused an increase in bulk density and speed of sound, and a subsequent reduction in sound absorption. In this work, the thickness of resulted cotton nonwoven-aerogel mats is missing which is make the results

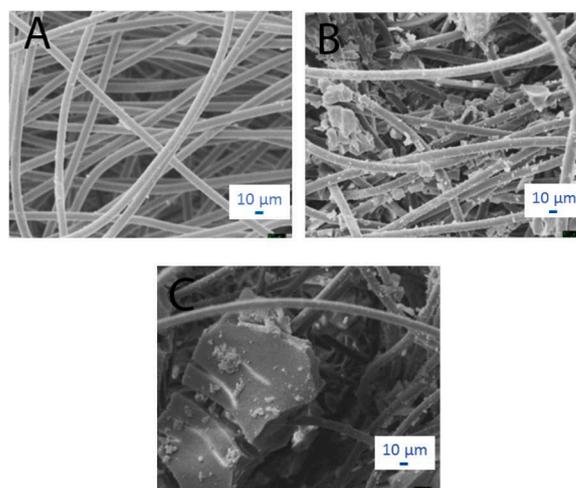


Figure 14. SEM images of (A) control PET, (B) PET nonwoven-silica aerogel blankets (MeOH:TEOS molar ratio: 110), and (C) PET nonwoven-silica aerogel blankets (MeOH:TEOS molar ratio: 28). Revised/reproduced with permission from Ref [85].

incomparable with the other researches.

In 2019 Yang et al. [102] examined the sound absorption properties of the aerogel-(Polyethylene/polyester) nonwoven bonded blankets, prepared with the method of Venkataramana et al. [127] through the addition of aerogel granules/particles during the thermal bonding of the fibers (see Figure 15). The sound absorption coefficient linearly increased with increasing frequency (see Figure 16A). Although there was no reference nonwoven sample without aerogel for comparison [102], Yang et al. concluded that the aerogel content is not a major factor in determining the sound absorption ability because the sample with the lowest aerogel content showed the higher sound absorption value. Important to note is that the density and the thickness of the samples were different for different aerogel contents and this may have affected the properties. Lamination of between 2 to 6 sample layers increased the sound absorption coefficient as expected with diminishing returns after more than 3 layers were laminated (see Figure 16B).

In the other study on the sound absorption behavior of in-situ synthesized silica aerogel/PET blankets in 2019 by Talebi et al. [90], at all frequency levels aerogel blankets exhibited higher sound absorption than the neat nonwoven samples, attributing to the low bulk density of silica aerogel, reduction in pore size and increase in tortuosity of blankets. It was also found that aerogel coating of the nonwoven fabrics makes them particularly adapted to low/medium frequency sound control where space is a concern, since lower thickened aerogel blankets showed a little higher sound absorption in comparison with the higher thickened neat nonwoven in the frequencies of less than 2000 Hz [90].

In summary, the sound absorption behavior of nonwoven-silica aerogel blankets depends on silica precursors and synthesis conditions, and different effects are observed in different studies. The addition of silica aerogel generally, but not always, improves the sound absorption, particularly in the medium frequency range, but the extent to which this adds a benefit will depend on the layer thickness and composite arrangement. Nonwovens themselves without silica aerogel are good porous sound absorbers efficient in a broad, but especially higher, frequency range. The addition of nonwoven-silica aerogel into a blanket typically results in reduced permeability and increased absorption in a lower frequency range. Higher concentrations of aerogels in a blanket can result in the resonance behavior in the absorption coefficient of the composite similar to that seen in neat silica aerogel without fibers (Sections 3.2 and 3.3).

A summary of works have done on the acoustic properties of the silica aerogels and their composition with the polymers could be found in Table 2.

3.5. Sound absorption/insulation of aerogel renders

Recently aerogels granules have been incorporated into high-

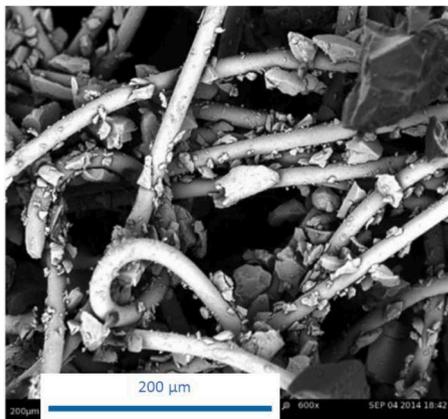


Figure 15. SEM images of an aerogel-nonwoven bonded compound. Revised/reproduced with permission from [102].

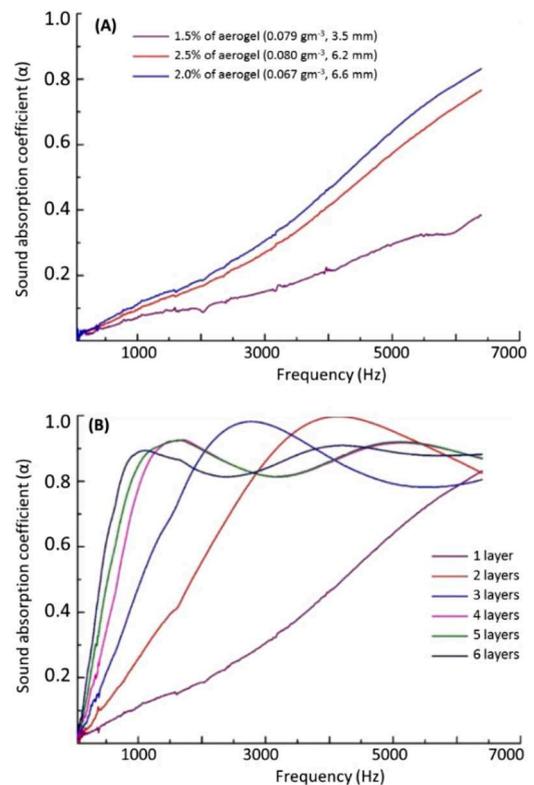


Figure 16. Sound absorption coefficient of the aerogel-nonwoven bonded compounds (A) with different aerogel contents and (B) the different number of layers for the sample with 2% of aerogel (0.067 g/m^3 , 6.6 mm). Revised/reproduced with permission from [102].

performance concrete and render for building applications [56, 79, 136]. Silica aerogel filled renders in particular have established themselves in the market place particularly for thermal insulation during the renovation of historical buildings [137]. Aerogel-based insulating renders, which can be applied to external and interior walls, are manufactured by mixing the conventional ingredients, additives and water with granular aerogel and coated on the wall (Figure 17) [78, 79].

In 2014 Buratti et al. [78] reported the sound absorption coefficient of the two samples composed of plasterboard support, an aerogel insulation plaster with aerogel of two different thicknesses and a final plaster coat (2 mm). Tests were carried out on small samples with an impedance tube. The results showed a strong dependence on the acoustic behavior on the final coat, which negatively influenced the acoustic properties. The acoustic absorption coefficients of the aerogel embedded samples were not very high for the proposed plasters [78].

Preikss et al. [138] in 2018 applied silica aerogel granules in a foamed gypsum (5-30%) and attached an aerogel blanket to one side of the foamed gypsum intending to improve the thermal and acoustic properties. However, no significant increase of the sound absorption coefficient in the 1 kHz frequency could be obtained for the foamed gypsum itself. On the other hand, attaching the silica aerogel blanket to one side of the solid gypsum specimen did lead to an increase of the sound absorption coefficient from 0.05 to 0.3 [56, 138].

In summary, very few studies investigated the acoustic properties of silica aerogel filled renders and products. These preliminary studies did not observe a clear acoustic benefit of adding silica aerogel to renders and boards, but this question cannot be settled with the currently available data. Also, this point that aerogels' pores are filled and/or cloaked with the plaster should be discussed here.

Table 2
Silica and silica-polymer related aerogels acoustic application related papers.

Material	t (cm)	Method (Device)	Frequency (Hz)	Sound Quantity measured	Author (Year)
Silica and silica-polymer derived aerogel					
- SA*	1-4	Kundt's tube (unknown)	250-5000	Absorption Impact insulation	Schmidt [63] (1998)
- SA/PVB					
- SA/VAE					
SA/PVB	1.8	-	-	Impact insulation	Schwertfeger [93] (2003)
Silica-PDMS derived aerogel	-	Impedance tube (home-made)	250-4000	Absorption	Dong [75] (2009)
MTMS-VTMS derived aerogel	1.6	Impedance tube (unknown)	200-2500	Absorption	Cai [76] (2012)
Polyimide-silica derived aerogel	1-3	Impedance tube (unknown)	2500-10000	Absorption	Yan [77] (2014)
- SA	1-5	Impedance tube (Brüel & Kjær)	50-1500	Absorption	Sachithanadam [80] (2016)
- GSA-SDS aerogel				Transmission loss	
SA	1.5-4	Impedance tube (Brüel & Kjær)	100-1700	Transmission loss	Moretti [60] (2017)
SA	1.1	Signal processing model based on impedance tube	Audible range	Absorption Transmission loss	Geslain [83] (2018)
Monolithic/granular SA	1.3-2.5	Impedance tube (Brüel & Kjær)	100-4900	Absorption Transmission loss	Merli [118] (2018)
Silica aerogel-polymer composite					
- SA/PUR foams	1.2-3.7	Impedance tube/ (Brüel & Kjær)	250-6400	Absorption Transmission loss	Dourbash [81] (2017)
- SA/elastomeric PU					
SA/UPVC composites	0.1	Impedance tube/ (Brüel & Kjær)	63-6300	Absorption Transmission loss	Eskandari [82] (2017)
Silica aerogel-textiles composite					
PET nonwoven-SA blankets	0.5	Impedance tube (unknown)	250-6400	Absorption	Oh [133] (2009)
Cotton nonwoven (1 cm)-SA blankets	-	Impedance tube (Brüel & Kjær)	100-6300	Absorption	Motahari [135] (2015)
SA-filled superfine PET fibers	0.02-0.024	Impedance tube/ Home-made	250-4000	Absorption	Mazrouei [130] (2015)
PET nonwoven-SA blankets	0.5-0.6	Impedance tube/ (Brüel & Kjær)	50-6300	Sound absorption	Ramamoorthy [85] (2017)
SA-(PE/PET) nonwoven bonded blankets	0.4-0.7 (1-6 layers)	Impedance tube/ (Brüel & Kjær)	50-6400	Sound absorption	Yang [102] (2019)
PET nonwoven-SA blankets	2.7 and 6.4 (nonwoven)	Impedance tube/ (BSWA)	50-6100	Sound absorption	Talebi [90] (2019)

* SA: Silica aerogel, PVB: Polyvinylbutyral, VAE: vinylacetate/ethene, PDMS: polydimethylsiloxane, MTMS: Methyltrimethoxysilane, VTMS: Vinyltrimethoxysilane, GSA: gelatin-silica aerogels, SDS: Sodium dodecyl sulfate, PU: Polyurethane, UPVC: Unplasticized Polyvinyl Chloride, PE: Polyethylene.

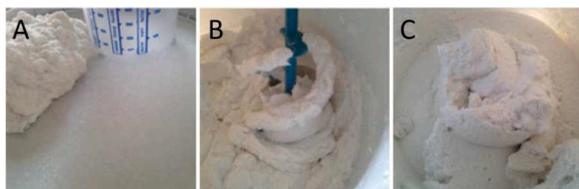


Figure 17. Mixing and applying of an aerogel-based plaster. (A) original components, (B) mixing phase, (C) final composition. Revised/reproduced with permission from [78].

3.6. Sound absorption/insulation of aerogel glazing systems

Because of their ultra-low thermal conductivity and high translucency and good solar-optical properties, silica aerogels have been investigated for their application in window systems. It is thus also of interest to study the sound insulation behavior of aerogel-based glazing systems. As early as 1991 Narang provided a detailed study of the diffuse field sound-transmission behavior of aerogel-based glazing systems with different glass thicknesses and interspace, and compared the results to those for conventional double glazing [139].

Two kinds of frequencies, considerable in the study of acoustic properties of the glazing systems, windows, and walls in the buildings, are critical frequency and resonance frequency. When the wavelength of sound air projected on the plate equals the wavelength of the bending

waves, the movement of the panel increases, leading to low sound insulation. The lowest frequency at which this wave coincidence occurs for flat homogenous plates is the critical frequency and can be approximated with

$$f_c = \frac{c^2}{1.81h} \sqrt{\frac{\rho(1-\nu^2)}{E}}, \quad (9)$$

where, c , h , ρ , ν , and E are the sound velocity in the air, thickness, the density of the material, Poisson's ratio, and Young's modulus, respectively. The resonance dip due to the coincident effect usually begins about an octave below the critical frequency. Below the frequency range of the coincidence, the mass law determines the sound reduction index. Above the coincidence zone, the sound reduction index depends on the frequency only for double systems. For a temperature of 20°C and normal sound incidence, the resonance frequency is

$$f_{res} = 59.8 \sqrt{\frac{1}{d} \left(\frac{1}{m_1} + \frac{1}{m_2} \right)}, \quad (10)$$

where, d is the distance between the inner surfaces of the glazing, and m_1 and m_2 (kg) are the mass of each pane. If the frequency of the sound incident on a double element is higher than the resonance frequency, the air chamber absorbs part of the sound energy, resulting in greater acoustic insulation than is observed in a single element with the same mass [139, 140].

Because mass-air-mass resonance is a major drawback in

conventional double-glazed windows for insulation against traffic noise, it appears that by choosing appropriate physical parameters of an aerogel filling of the interspace of the double glazed system, one should be able to design a suitable window system free of the undesirable resonance dip in the low to the medium frequency range. Figure 18 shows that the transmission loss increases for an aerogel filled double glazing system [139]. However, compromises with respect to aerogel layer thickness may need to be made concerning acoustic/thermal performance versus cost, transparency, and solar gains.

Buratti et al. in 2012 evaluated a prototype of an aluminum frame window with granular aerogel in the interspace [141]. Not only was thermal transmittance of the innovative glazing system lower than 1 W/m^2 , but also the acoustic properties were improved with $R_w=37 \text{ dB}$ which is 3 dB higher than the one of a conventional window with air in the interspace [141]. Importantly, this number was achieved according to the standard series of ISO 10140 which is the test used on large-scale samples which is more reliable than the impedance tube method. To improve further the acoustic performance of the window ($R_w>40 \text{ dB}$), the granular aerogel could be assembled into laminated glasses with a special acoustic polyvinyl butyral layer. Figure 19 shows the R-curve of the samples with and without aerogel [125]. In 2014 these systems were tested in-field monitoring campaigns to validate the capability of aerogels in improving the acoustic performance of the glazing systems [142].

3.7. Aerogel-based products versus conventional acoustic materials

In this section, we compare silica aerogel-based materials to four classes of conventional acoustic materials. This comparison is complicated by the wide variability of aerogel-based products (describe above) and, perhaps more critically, the wide knowledge gaps about their acoustic properties. As discussed in the previous sections, silica aerogel can be incorporated in a wide variety of composite materials (fiber mats, polymers, inorganic renders and concrete, glazing systems) and the nature of the composite can be the dominant control on the acoustic properties, sometimes more so than the properties of the aerogel itself. In addition, sound absorption properties depend on the composition, thickness and surface pattern of the absorbers, method of mounting (air gap) and frequency of the incident sound are also so important.

Table 3 compares the most important, application-relevant physical properties of aerogel-based to conventional acoustic materials. In terms of sound absorption, porous fibrous and foamed materials display absorption over a wide range in frequency, while perforated materials show high sound absorption, but in a narrow frequency range. The pore size of foamed materials, and holes' size and holes' surface density for perforated systems determines their frequency range and percentage of absorption. Silica aerogels on their own resemble more closely the behaviour of the perforated materials with resonance absorption in a narrow frequency range, but in combination with fibers and textiles behave more similar to fibrous materials and foams.

All tabulated materials, aside from the perforated plates, display low

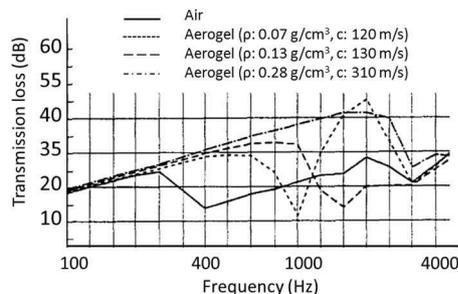


Figure 18. Sound transmission loss for a glazing system (glass-aerogel-glass of 4-6-4 mm). Revised/reproduced with permission from [139].

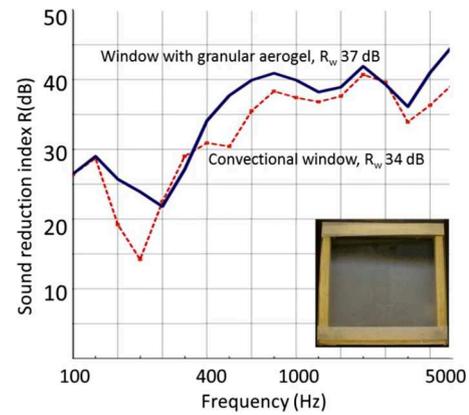


Figure 19. Sound reduction index (R) for the conventional window and the glazing prototype with aerogel in the interspace. Revised/reproduced with permission from [141].

densities and low thermal conductivities. Among the conventional materials, closed-cell, pentane and/or CO_2 filled polymer foams (often polyurethane based) have the lowest thermal conductivity, but closed-cell foams tend to perform worse acoustically compared to open-cell foams because these materials do not allow for viscous, inertia and thermal absorption effects to develop in the material pores. Silica aerogels have even lower thermal conductivities due to their mesoporous structures. Silica aerogels also stand out for their hydrophobicity. In the absence of fire retardants, polymer based materials (foams and fibrous materials) naturally display worse fire properties compared to mineral wool or silica aerogel. Silica aerogel based products are more expensive by up to an order of magnitude compared to fibrous mats and most polymer foams, although high performance polyurethane foams engineered for impact noise control and vibration isolation also can be very expensive. Perforated materials are usually used for very specific applications, e.g. as ceiling tiles in auditoria, theatres or concert halls, while fibrous and foamed materials are also used in standard applications, such as residential, commercial and public buildings, automotive and other transportation thanks to their low cost and ease of installation. Mineral wool is particularly cost-effective, but cannot be used as a final cover or design in terms of their health and environmental problems, whereas other materials can target the acoustical/architectural designs, specially foams for internal designs and perforated systems for internal/external designs. Aerogels, when mixed with the other materials, have a potential of being used as the final coating related to their minimized dust releasing behaviour.

4. Summary, open questions, and road to market

From the literature survey above it is clear that the physics of how sound propagates through silica aerogel is poorly understood. Although there is literature on the absorption and transmission properties of aerogels and effects of thickness [63, 77, 93], pore size [75], and granule size [60, 80], these studies lack mathematical modeling to understand key physical mechanisms which can explain the observed acoustical behavior of aerogels. A majority of these works are particular to the adopted layer thickness and do not discuss any generic acoustical quantities such as frequency-dependent bulk modulus of the air trapped in an aerogel, its dynamic density and complex Young's modulus of the aerogel's frame which can exhibit viscoelastic behavior. These properties are frequency-dependent and should be related to the aerogel's chemistry and pore morphology. There is a wide variation in how experimental data were collected and interpreted. Therefore, systematic efforts are necessary to fully discern and realize the potential of silica aerogel and its composites as acoustic materials.

Since the physics of sound absorption/transmission is not fully

Table 3
Comparison of conventional and aerogel-based acoustic insulation materials.

Material	Fibrous mats	Polymeric fibers	Polymer foam	Perforated panels	Silica aerogel
Density (kg/m ³)	Mineral wool Low (30-150)	Low-medium (50-1200)	Low (8-400)	Medium to high	Low (20-200)
Pore sizes	Macroporous	Macroporous	Macro/mesoporous	Macroporous	Mesoporous
Hydrophobicity	Low- medium	Low-medium	Low-medium	Low-medium	High (tunable)
Fire resistance	Good	Poor	Poor	Poor to good	Good
Structure	Fibrous	Fibrous	Cellular (open/closed)	Perforations	Nanoparticle networks
λ (mW m ⁻¹ K ⁻¹)	Low (30-36)	Low (30-36)	Low (20-36)	High	Ultra-low (12-36)
Cost	Low	Low-medium	Low-high	High	High
Acoustic	Absorption	Absorption Vibration Isolation	Absorption Insulation	Resonant	Absorption Insulation
Applications	Insulation	(less)	Vibration	absorbers	
Absorption	Wide range	Wide range	Wide range	Narrow range	Narrow range, wide if combined with textiles
Frequency					

understood even for pure silica aerogel, the situation for the composites is even more dramatic. Because of the wide variety of composites (polymers, textiles, renders), each type of composite will require its own detailed experimental and theoretical investigations. Unfortunately, the effect of silica aerogel on the acoustic properties of the final products is typically investigated as a side-thought, and only a few studies focus on this matter. In contrast to neat silica aerogel, aerogel-fiber blankets typically display smoother dependencies of the sound absorption coefficient on frequency, with broad bell-shaped curves reminiscent of the aerogel-free nonwovens [102, 133, 135]. Although thermal insulation and fire resistance remain the market driver for aerogel blankets for pipeline and industrial insulation, commercial products have passed the industry's acoustic standards, e.g. ISO 15665 [143]. These systems are offered by Armacell (Germany) and ULVA Insulation Systems (UK). The main users are in the oil, gas and petrochemical sectors. For building materials other than aerogel blankets, an improvement in sound transmission loss was observed for the aerogel-filled double glazing systems in some case studies [139, 141], but there is no clear data if an improvement is possible for aerogel filled renders and gypsums. From the limited data available, it is clear that the combination of aerogels with other building materials can significantly affect the aerogel's acoustic performance, but sometimes also in an adverse way [138]. Hence, the method used for the integration of the aerogel with other aqueous/liquid phase materials needs to be considered carefully and the effect on acoustic properties evaluated through rigorous measurements.

In the current market place, silica aerogels' unique selling point is the ultra-low thermal conductivity, which enables thinner thermal insulation layers. Based on the current state-of-the-art, it is clear that silica aerogel and its derivative products often have good acoustic properties, but they do not necessarily outperform conventional materials and products. Only a few studies have targeted acoustic properties as a selling point for silica aerogel [93]. As large-scale production is concerned, only limited data are available on the use of the transparent silica aerogel in the interspace of the double glazing systems in the pilot-scale which showed the sound transmission loss was increased in comparison with the glazing system with the air in the interspace [139, 141]. However, also in the field of transparent insulation, it is not clear if the benefits are sufficient to offset the added cost of an aerogel solution. Whether acoustics can become a market driver for silica aerogel remains an open question, particularly from the viewpoint of silica aerogel's high cost and low mechanical properties, compared to conventional materials. Additionally, the combination of aerogels with polymers and other materials should be carried out thoughtfully to prevent infiltration of the aerogel mesoporosity and to avoid adverse effects on the acoustic performance.

Silica aerogels' absorption behavior has the potential for interior room acoustics to absorb specific frequencies (e.g. absorbing layer combined with typical porous absorbers of rock wools and foams). In addition, the relatively high sound absorption coefficients and transmission loss, especially at low to intermediate frequencies, combined with the low density and thickness can play an important role in

reducing the weight and thickness of construction elements. However, the silica aerogel-based products currently on the market have been developed for thermal insulation as their main application. Hence, the composite properties have been optimized to minimize thermal conductivity with mechanical strength, durability and ease of installation as important secondary parameters. The acoustic performance has at best been considered of minor importance, or more commonly, has not been considered at all. The question about how well an aerogel-based product can perform as acoustic insulation, if it were to be optimized for its acoustic properties first and foremost, thus remains open.

The silica aerogel thermal insulation market is rapidly growing. The use of thinner insulation layers, compared for example to mineral wool, inevitably has consequences for the acoustic performance of the façade. Lingering uncertainty on the direction and size of the acoustic effects of using aerogel thermal insulation could negatively impact this market. Thus, although the feasibility of acoustics as a market driver remains an open question, it is clear that a better understanding of the acoustic performance of silica aerogel and its products is also imperative to support the main thermal insulation application.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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