Silica Aerogel Incorporated Cementitious Composites

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Silica aerogels are made of 95% air, the rest being silica crystal (SiO2). They have an open porous structure that is composed of particles with diameter less than 10 nm and pores smaller than 50 nm. Lightweight cement composites with silica aerogel in the form of granulate, thanks to their low thermal conductivity and good mechanical performance, may soon find wide application in the construction sector. An important aspect that guarantees this application is the proper design of the composite, proper selection of its components, improvement of the interfacial zone between the silica aerogel and the cement matrix, and ensuring the durability of the material in the long term.

silica aerogel | insulating materials | lightweight cementitious composite | mechanicla properties | durability

1. Silica Aerogel—Synthesis and Properties

Silica aerogel was invented in the 1930s, by Stephan Kistler, but only the last twenty years have brought considerable interest in this material [1]. The intensive development of research on silica aerogel has been initiated by a group that has used organic silica compounds such as TMOS (tetramethylorthosilicate) or TEOS (tetraethylorthosilicate) as precursors. Silica aerogels are synthesized in three stages by the sol–gel process. In the first stage, a gel is prepared by a precursor solution (silica source) with the addition of a catalyst. In the second stage, the gel is aged either in water or the mother solution. The aim of aging is to consolidate the gel and minimize the shrinkage of the gel during drying. The drying step can be realized mainly through three ways, namely supercritical at high or low temperatures, and ambient pressure drying. In the high-temperature supercritical drying method, the gel is put together in an autoclave with an alcohol such as methanol or ethanol, and the temperature is slowly increased until the supercritical temperature and pressure are reached. The fluid is then removed at constant temperature. In the low-temperature supercritical drying method, the alcohol present in the pores of the gel is replaced with another liquid, such as liquid CO2, which has a critical point close to ambient temperature. Here, the wet gel is placed in an autoclave, and liquid CO2 is pumped in at 4–10 °C until the pressure reaches 100 bar. Subsequently, the solvent inside the pores of the gel is extracted, and the autoclave is heated close to 40 °C to reach the supercritical conditions of CO2 [2][3].

Even though the supercritical drying process is the most common process, and is the most suitable for monolithic aerogel production, the cost and safety risks, especially for high-temperature supercritical drying, are limitations. In the ambient pressure drying process, the water–alcohol mixture in the pores of the gel is first exchanged for a water-free solvent. The surface modification is then reacted with a silylating agent so that the Si–OH groups are replaced by methyl silyl groups. The substitution of the H from the Si–OH groups by the hydrolytically stable Si–R groups hinders the adsorption of water, and the aerogel becomes hydrophobic. After solvent exchange, evaporative drying takes place [4][5][6]. The ambient pressure drying procedure is advantageous when compared to the supercritical drying in terms of cost and safety since it does not require high pressures or expensive high-pressure equipment. Nevertheless, there are additional chemicals and solvents employed. Therefore, to make this process suitable for commercialization, minimum amounts of solvent should be used with a minimum number of solvent exchange steps. Nevertheless, laboratory-synthesized silica aerogels are very fragile for sole application in the building sector. Thus, further research should focus on improving the silica aerogel's strength and incorporating it into stronger organic or inorganic, etc., matrixes [7][8][9].

By means of the proper selection of particular parameters of synthesis, precursor and modification method, it is possible to alter the final structural and mechanical properties of silica aerogels at an early stage of the synthesis.

The strength and stiffness of the gel can be improved at the stage of ageing the gel by dissolving and repeatedly precipitating silica from the surface of particles onto the borderline particle–particle and connecting and/or precipitating oligomers that were unreacted during gelling. Another method assumes adding extra amounts of precursor and co-precursor to the solution before and after the moment of gelation, so that it builds into the structure of the gel and, thus, reinforces it [10][11][12].

Apart from altering the parameters of the synthesis, the mechanical properties of silica aerogels can be modified by incorporating various additives into their structure, e.g., nanoparticles and metal nano oxides, or by applying reinforcement in the form of short structural fibers or fiber mats [13][14][15][16][17][18][19]. There is also research carried out on covering the surface of silica aerogels with polymers [20][21][22][23]. This action is taken before the stage of drying the gel; as a result, the surface of the silica aerogel is covered with a layer of polymer that increases the resistance of silica structure to breaking. In addition to the above-mentioned strengthening of the aerogel structure with fibers, an alternative solution may also be to introduce silica aerogel into more durable and stronger structures with a low thermal conductivity coefficient, such as a polymer matrix or concrete.

Concrete, in comparison with other building materials such as stone or steel, is characterized by a relatively low thermal conductivity coefficient, reaching maximum values of roughly 2.0 and 2.5 W/(m·K) for average concrete with a density from 2200 to 2400 kg/m^3 and for reinforced concrete (with steel bars), respectively [24]. The thermal conductivity coefficient of concrete can be easily lowered via air entrainment or the application of a lightweight aggregate characterized by high porosity and a low thermal conductivity coefficient. Unfortunately, very often, high porosity in concrete and lightweight aggregates leads to a significant decrease in composite compressive strength —down to a few Mpa—and eliminates such solutions in terms of construction potential. In addition, the application of a modification of the cement binder with polymers may improve the adhesion of the binder to the aggregate and thus enhance the mechanical parameters of the composite [25][26]. There is, however, a group of lightweight aggregates that enable higher strength parameters to be obtained with a relatively low thermal conductivity coefficient [27][28][29][30][31][32][33][34] (see **Table 1**).

Table 1. The physical and mechanical properties of cementitious lightweight composites.

Aggregate Type/Maximum Size	Dry Density (kg/m^3)	Compressive Strength (MPa)	Thermal Conductivity (W/(m·K))	References
Cenosphere/4 mm	1050–1350	5.0–30.1	0.46–0.60	[27]
Expanded perlite/2–4 mm	354–1833	0.1–28.8	0.06–0.13	[28]
Cenosphere/600 µm	1483–1890	44.3–48.1	0.29–0.37	[29]
Expanded glass/4 mm	1100–1380	23–30	0.49–0.85	[30]
Cenosphere/300 µm	1042–1300	40.9–69.4	0.31–0.40	[31]
Cenosphere/300 µm/GGBS (20–60%) in place of cement	1240–1270	Above 55	0.39–0.45	[32]
Cenosphere/500 µm	1282	52.5	0.6	[33]

Among these aggregates, microspheres have the best strength and insulation parameters. Microspheres (cenospheres) are hollow silica and alumina spheres with a diameter of less than 500 µm that are produced as a by-product of coal combustion in thermal power plants. The most important characteristics of microspheres are low bulk density (about 400 kg/m^3), low thermal conductivity 0.1 W/(m·K) at room temperature, low coefficient of thermal expansion (6.13 × 10^{-6} 1/K) and high melting temperature above 1200 °C (which gives them high temperature resistance) [32][33]. Studies have shown that the use of cenospheres with diameters ranging from 300 to 600 µm in cement composites leads to very high strength parameters (with compressive strengths reaching approximately 40–70 MPa), while low densities are maintained and thermal conductivity coefficients range from 0.29 to 0.60 W/(m·K) [29][31][32][33].

2. Durability and Performance of Silica Aerogel-Based Cementitious Composites

Increased porosity of the cement matrix due to the presence of silica aerogel and poor adhesion at the silica aerogel–cement paste interface are key factors affecting the durability of cement composites. Therefore, it seems very important to study the durability of these materials and the performance over a long service life. There have been a few publications in recent years in which the authors extended the scope of their study and evaluated the durability, fire resistance and exposure to solar radiation of silica aerogel-based cementitious composites [35]. Nevertheless, all authors agree that this is a direction for future research on these materials.

Stefanidou and Pachta [36], for example, looked into the fire resistance properties of cement-based mortar with silica aerogel and perlite. For the purpose of the study, 20% of the aggregate was replaced with silica aerogel and perlite. After curing, the samples were exposed to elevated temperatures of 800–1000 °C. The investigators indicated that the samples containing both silica aerogel and perlite maintained mechanical strength before and after exposure to high temperatures, whereas samples without silica aerogel did not maintain residual mechanical strength [36].

From the results of the above-mentioned work, it can be inferred that the fire-resisting properties of silica aerogel might find application in the field of insulation materials. Such properties were investigated by several researchers [35][37][38][39][40]. In the publication of Ismail et al. [38], for example, the investigators presented an experimental study on the energy efficiency of cement-based thermal cladding with silica aerogel amendment. For the purpose of the research, mechanical strength, thermal conductivity and exposure to solar radiation were analyzed, and the insulating capability of the renders was tested under exposure to various climate conditions. The researchers indicated that suitable energy efficiency and insulating capability were achieved [38]. Morgado et al. [39], in turn, investigated the durability parameters of thermal renders with silica aerogel and other eco-friendly materials. The scope of the experiment was to expose samples to long-duration freeze/thaw cycles and to hygrothermal accelerated aging cycles. Between and after exposures, mechanical strengths and thermal conductivity were measured. According to the final results, the long-duration freeze/thaw cycle and the accelerated aging cycle led to an increase in the compression strength of renders with re-granulated cork and renders with expanded polystyrene. Moreover, renders with silica aerogel maintained their mechanical strength, whereas the thermal conductivity of the renders with silica aerogel was reduced from 0.20 W/(m·K) to 0.09 W/(m·K) before and after exposure to several freeze/thaw cycles [39]. The work of Morgado et al. shows the significant relationship between the porosity of the cement matrix and the thermal–moisture properties of potential coating materials.

Hygrothermal testing under different climatic conditions has also been studied by other researchers [41][42][43]. The results of Sakiyama et al., for instance, showed high water absorption in the analyzed renders during weathering; this was especially evident in the deepest layers of the thermal insulation [41]. The test program used included the following approaches: heat–rain cycles for 20 days, heat–cold cycles for 5 days and rain–cold cycles for 20 days [41]. The high water absorption in the aerogel-based render caused its damage after undergoing the aforementioned freezing cycles, so an important issue in the future is to strengthen this layer of insulation and protect it from external moisture access. The applied ageing method did not affect the thermal conductivity coefficient, and no significant changes in it were recorded during the examined time. Similar relationships and conclusions were also presented by Berardi et al., who also studied the accelerated aging of lime-based aerogel composites under cyclic temperature changes, negative to −30 °C and positive to +40 °C, with different moisture content [44]. Other researchers also point out various moisture problems in the outer insulation layers, some of which seem to be important, such as the inability to dry completely over a long period of time or the phenomenon of condensation. A study by Ibrahim et al. revealed that the application of an additional insulation layer in the form of a silica aerogel-based render on an uninsulated building or on a building with existing interior insulation results in a reduction in or complete removal of the moisture problem [42]. Moreover, Maia et al. demonstrated that the application of an additional protective layer to aerogel reduces the negative effects of accelerated aging and results in increased durability of the mortars over a longer service life [45].

A summary of recent trends in aerogel cementitious composites depending on the potential use and the factors for durable high-performance materials that guarantee long service life is shown in **Figure 1**.

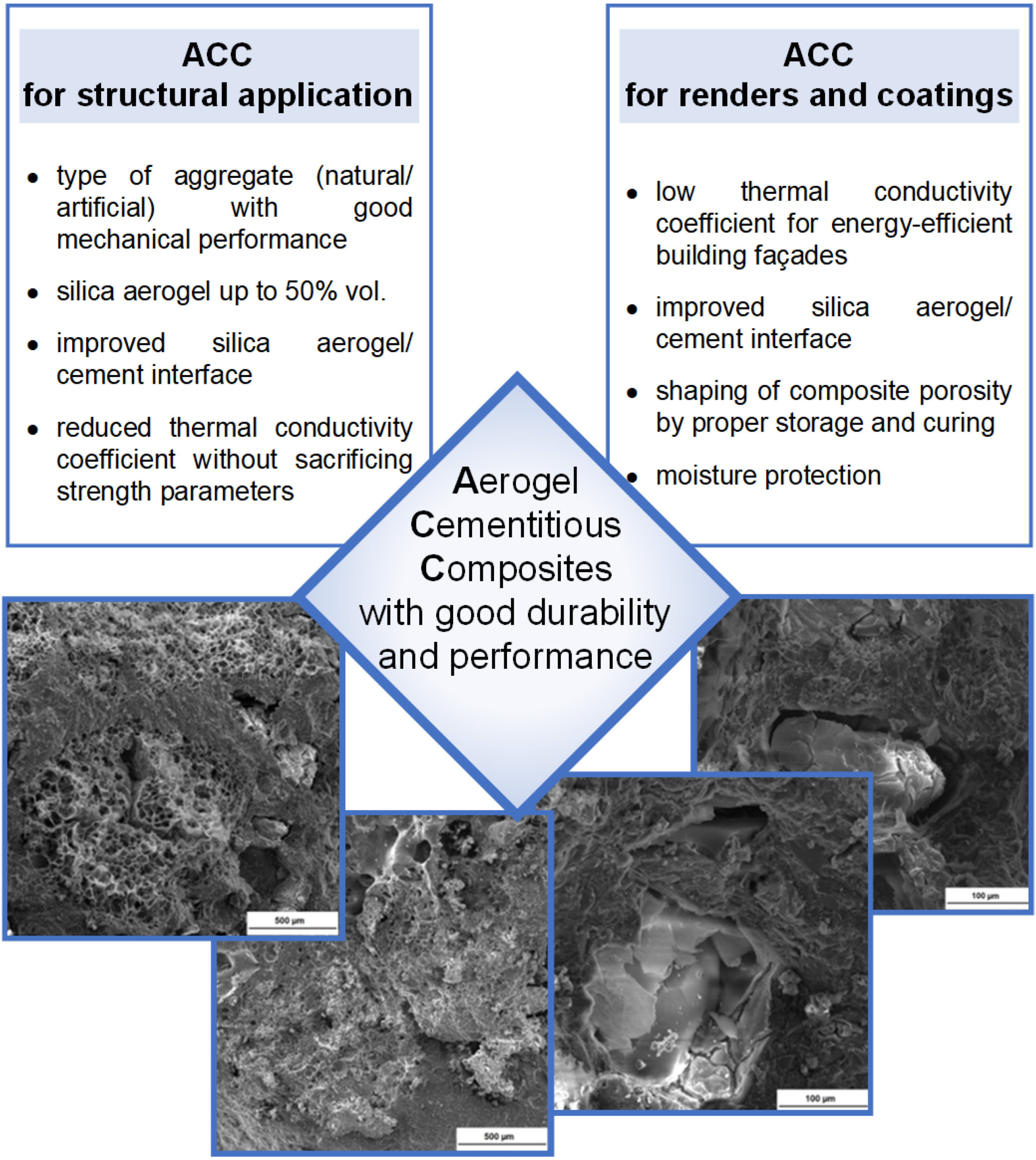

Figure 1. Development pathways for aerogel cement composites.

3. Conclusions

Researchers collect and discusses recent publications on cementitious composites with silica aerogel. The unique properties of silica aerogel, such as its transparency, low density and high porosity, make this material an interesting solution for lowering the thermal conductivity of the cement matrix and creating new, eco-efficient future mortar and concrete materials that meet stringent energy conditions. The listed studies clearly indicate two directions in the development of aerogel cement composites. In the first, research is conducted with the aim of creating ultralight cement composites for lightweight façade systems, such as thermal renders with silica aerogel as an aggregate. These hold very good insulation parameters but sacrifice strength parameters. In the second, research is directed towards the creation of green, lightweight cement composites based on lightweight aggregates that come with good thermal conductivity and good strength parameters. Among these are cenospheres, fly ash-based aggregates and foam concrete. In these solutions, the aggregates are partially replaced by silica aerogel to further reduce the thermal conductivity while achieving satisfactory mechanical performance of the cement matrix.

Researchers provide an analysis of the results of studies wherein cementitious composites were produced in the form of mortars and concretes in which silica aerogel was added as a replacement for natural or lightweight artificial aggregates. The density, strength and thermal conductivity relationships were assessed as a function of the amount of silica aerogel used. In addition, attention was paid to the aspects of water transport and porosity, which directly affect the durability of the composites studied. Based on the investigation, the following conclusions can be drawn: the introduction of silica aerogel into the cement matrix, usually at the expense of natural aggregate, contributes to a reduction in the density of the material by increasing the porosity, which in turn contributes to a significant reduction in the thermal conductivity coefficient. Nevertheless, the results show that large volume proportions of silica aerogel bring about significant decreases in compressive strength and increase the water absorbability of the cement composite. Therefore, an important aspect of future research on cement composites with silica aerogel will be to improve adhesion at the silica aerogel–cement matrix interface. Moreover, the issues of water transport and the durability of cement composites should be particularly studied in the coming years. These aspects will certainly contribute to the wider applicability of these materials in the construction industry.

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