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REVIEW PAPER

Prospect of Thermal Insulation by Silica Aerogel: A Brief Review

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Abstract Silica aerogel is a unique ultra light weight nano porous material which offers superior thermal insulation property as compared to the conventional thermal insulating materials. It can be applied not only for ground and aerospace applications but also in low and high temperatures and pressure regimes. Aerogel granules and monolith are synthesized by the sol–gel route while aerogel based composites are fabricated by the reinforcement of fibers, particle and opacifiers. Due to the characteristic brittleness (i.e., poor mechanical properties) of monolith or bulk aerogel, it is restricted in several applications. To improve the mechanical integrity and flexibility, usually different fibers are reinforced with aerogel and hence it can be used as flexible thermal insulation blankets. Further, to achieve effective thermal insulation behaviour particularly at high temperature, often opacifiers are doped with silica aerogel. In the present brief review, the prospects of bulk aerogel and aerogel based composites are discussed for the application of thermal insulation and thermal stability.

Keywords Aerogel · Composites · Thermal insulation · Thermal conductivity · Thermal stability · Mechanical properties

Introduction

Silica aerogel often known as 'blue smoke' or 'frozen smoke' is basically silica precursors derived nanoporous structure synthesized by the sol–gel route [1]. The pores will be filled with air which is more than 90% of the volume of aerogel. Aerogels are lightest solid material and it possess several unique characteristics such as ultra low density, lowest thermal conductivity, low dielectric constant, with adequate optical transparency and mechanical integrity. The word aerogel signifies that it is derived from gel and finally it is in the form of air or gas trapped nanoporous structure. Generally, the pore size of aerogel is in range of 1–100 nm.

The first ever report of aerogel was published by Samuel Stephens Kistler long back in 1932 [2], where he developed a porous silica structure filled with gas as replaced by liquid in the form of gel. Further, it was interesting to notice that Kistler also introduced a new technique i.e., supercritical drying technique for transformation of gel to the final porous solids.

In general, synthesis of aerogel involves two major stages e.g., (1) preparation of gel from sol–gel process and (2) drying by super critically or ambient pressure. However, several intermediate stages are involved which will be described briefly later.

If one looks into the aerogel market research report, one can find that projected compound annual growth rate (CAGR) for five years (2015–2020) in thermal and acoustic insulation sector is significantly higher [3]. Moreover, the prime driving factor for the growth of business market is due to the demand of industrial thermal insulation. Allied market research [4] also says the major (around 70%) market share of aerogel is mainly held by the commercial insulation blanket product. Now a days, aerogel in the form

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of particle/powder/granules, blocks, panels and composite blankets is experiencing the fastest growth in the market worldwide due to its versatile application in the sectors such as oil, gas, pharmaceuticals, building insulation, aerospace, automotive, health-care, chemical, electronics, apparel, textile etc.

Thermal Conductivity of Common Thermal Insulation Materials and Commercially Available Aerogels

Figure 1 summarizes the thermal conductivity values of common thermally insulating materials and commercially available aerogels [5–9]. The range of thermal conductivity is reported as 0.018–0.048 W/mK for common thermal insulating materials such as polyurethane foam, phenolic foam, mineral wool, polystyrene, fiber glass, rock wool, polystyrene and cork (Fig. 1). On the other hand, commercially available silica aerogel materials offer much lower thermal conductivity value i.e., in the range of 0.0034–0.022 W/mK (Fig. 1) as compared to common thermal insulating materials mentioned earlier. Hence, silica aerogel is found to possess the lowest thermal conductivity value of any insulator so far reported at atmospheric pressure. Further, it is well known that air possesses very low (0.025 W/mK) thermal conductivity. It is worthy to mention that no solid (non porous) is known to possess thermal conductivity as low as air before the invention of silica aerogel by Kistler [10]. The thermal conductivity value of aerogel is significantly lower than air which can be further explained with Knudsen effect [10]. According to the Knudsen effect, nanometric pore size of aerogel, which is comparable or lower than the mean free path of air, hinders the thermal conductivity due to the restriction of movement of the trapped gas inside the pore of aerogel [11].

Synthesis of Silica Aerogel Granules/Monolith and Composites

In general, silica aerogel is synthesized by sol–gel route [12–23]. Single-step (gelation via acid or base catalyst) or two-step (gelation via both acid and base catalysts) methods are utilized as reported in literature [12–23]. Silica aerogel synthesis process can be classified into five major stages such as: (1) sol preparation, (2) gelation, (3) ageing/solvent exchange, (4) silylation and (5) drying process.

Usually, metal alkoxides are utilized as starting precursor for the preparation of the sol [12–15, 17, 18, 20–23]. The most common metal alkoxides reported to be utilized in aerogel synthesis are tetraethylorthosilicate [TEOS: $Si(OCH_2CH_3)_4$] [12–15, 17, 18, 20, 23], sodium silicate [16, 19] and tetramethylorthosilicate [TMOS: $Si(OCH_3)_4$] [24, 25]. Though sodium silicate is a very familiar and cheaper material, during reaction it will form corresponding salts within the gel that are required to be removed by many repetitive washes [16, 19]. On the other hand, TMOS is identified as a toxic compound and the fumes may cause severe illness [12]. However, it is reported that comparatively TEOS is a safer reagent as compared to TMOS [12].

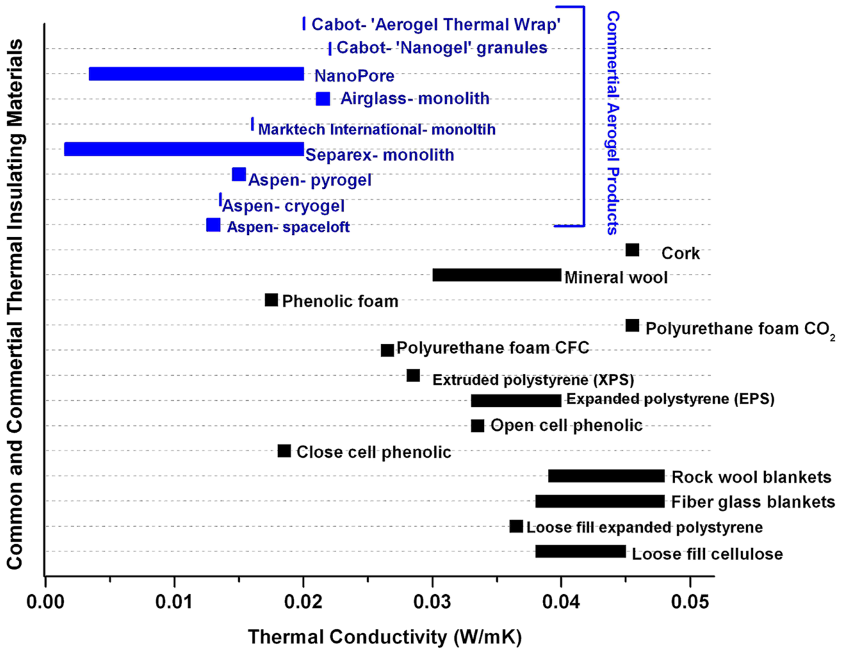

Fig. 1 Thermal conductivity of common and commercial aerogel based thermal insulation materials [5–9]

Figure 2 illustrates the flow chart which shows major stages in the synthesis of silica aerogels. Different stages of aerogel synthesis are now briefly described here after.

Sol preparation is the first stage of aerogel synthesis. In this stage, solution of various reactants undergoes hydrolysis and condensation reactions [12]. The reaction of a metal alkoxide (M–OR) with water forms a metal hydroxide (M–OH) as following: M–OR + H_2O = M–OH + H–OR. In contrast, a condensation reaction occurs when two metal hydroxides (M–OH + HO–M) combine to give a metal oxide species (M–O–M). One water molecule is formed in this process. The reaction is as following: M–OH + M–OH = M–O–M + H_2O. After sol preparation, the next stage is known as gelation. Actually here, the 3D network is generated to link oxide particles of the sol. Finally, it gets converted into a wet gel [12–23]. After formation of gel, the gel has to be aged to achieve desired solid structure. The ageing stage/solvent exchange is introduced to strengthen the gel and to eliminate un-reacted entities. Silylation stage is performed subsequently after ageing/solvent exchange stage to change the surface property of aerogel i.e., hydrophilic to hydrophobic surface and helps in retaining final structure of aerogel [13, 15–19, 22, 23]. Hexamethyldisilazane (HMDZ) chemical proved to be comparatively better silylation agent as compared to other chemicals reported in literature [13, 15–19, 22, 23]. The final stage is called as drying. There are two ways to carry out drying process for wet gel e.g., (1) super critical drying and (2) ambient pressure drying. Super critical drying process generally prevents the formation of cracking as reported by Kistler [10]. Here, the formation a liquid–vapor meniscus can be avoided which ultimately results in an interrupted evaporation of solvent from the solid network [22]. Whereas, an ambient pressure drying is less complicated procedure as compared to the aforesaid process to develop un-cracked monolith aerogel by heating the alcogel at ambient pressure above boiling temperature of solvent which is used in sol formation.

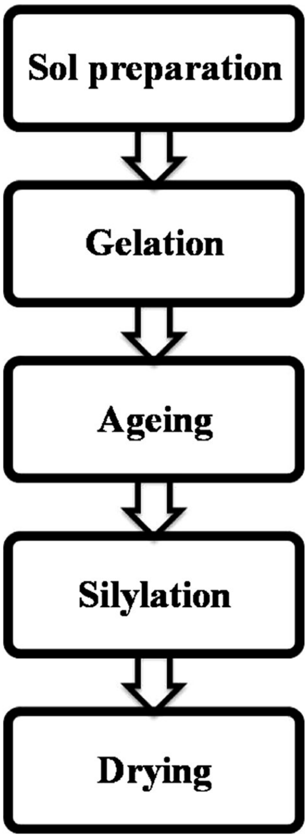

Fig. 2 Major stages in synthesis of silica aerogel

Further, fibers are usually utilized for the fabrication of aerogel composites. Basically, fiber gives the mechanical strength to the composite. In the reported literatures [26–35], most of the fibers are selected to reinforce in aerogels matrix are inorganic fibers, including aramid fibers [27–29], nomex fibers [26], glass fibers [36], carbon fiber [37], epoxy resins [30, 34] and opacifiers such as TiO_2 [33], ZrO_2 [38], carbon black [39] etc. which can improve the strength and flexibility of the aerogel composites as mentioned earlier. To improve further mechanical properties of aerogels cross-linking polymers are also used [40].

Thermal Conductivity and Stability of Monolith Silica Aerogel

Thermal conductivity and thermal stability of monolith or bulk aerogel reported in literature are summarized in Table 1 [12–22]. Thermal conductivity values are reported to be in the range of 0.036–0.417 W/mK [12–22]. On the other hand, thermal stability mentioned in literature is in the range of 100–550 °C [12–22]. Further, it is observed that density and porosity of the bulk/monolith silica aerogel have significant influence on its thermal conductivity shown in Fig. 3a, b, respectively. Thermal conductivity decreases with decrease in density as depicted in Fig. 3a because heat transfer through conduction will be decreased due to decrease in solid volume in aerogel [12]. Conversely, porosity will show opposite trend as illustrated in Fig. 3b. Here, thermal conductivity decreases with increase in porosity. This happens due to the increase in trapped air as in general thermal conductivity of air is much lesser as compared to any solid material [9, 10]. Further, increase in porosity results in decrease of solid network which ultimately hinders the heat conduction as mentioned earlier [16].

Hence, in order to modify the porosity and density of silica aerogel to achieve desired thermal conductivity various attempts are made by several researchers [12–23] as listed in Table 1. The tailoring of thermal conductivity property can be achieved by using different type of

Table 1 Literature status on thermal conductivity and thermal stability of monolith silica aerogel

Precursors	Drying method	Key features in synthesis	Density (g/cm^3)	Porosity (%)	Thermal conductivity (W/mK)	Thermal stability (°C)	References
TEOS	SCD	Different co-precursors, molar ratio TEOS/CP = 1	0.036–0.163	91–98	0.042–0.086	200–520	[12]
		Different co-precursors, molar ratio TEOS/CP = 0.3	0.049–0.207	89–97	–	–	
TEOS	APD	Acetonitrile: TEOS molar ratio varied from 1.09 to 5.45	0.06–0.15	92–96	0.068–0.099	300	[13]
TEOS	SCD	MTES/TEOS molar ratio varied from 0 to 1	0.103–0.355	–	–	100–300	[14]
TEOS	APD	Multiple surface treatment carried out	0.069–0.624	72–96.8	0.036–0.417		[15]
SS	APD	Various surface modification (silylation) agents	0.152–0.06	92–96	0.091–0.170	~325	[16]
TEOS	APD	TMES/TEOS molar ratio varied from 0.19 to 0.86	0.104–0.06	–	0.077–0.062	~308	[17]
TEOS	APD	PTES/TEOS molar ratio varied from 0 to 0.5	0.23–0.33	84.21–91.16	0.08–0.1	~550	[18]
SS	APD	Effect of two different trimethyl silylation agents					
		TMCS/SS molar ratio varied from 2.4 to 5.4	0.04–0.095	95–97.8	–	450	[19]
		HMDZ/SS molar ratio varied from 2.4 to 5.4	0.093–0.115	93.9–95.1	–	430	
TEOS	APD	Oxalic acid/TEOS molar ratio varied from 3.115×10^{-5} to 3.115×10^{-4}	0.09–0.145	92.3–95.2	0.102–0.125	–	[20]
		NH$_4$OH/TEOS molar ratio varied from 4×10^{-3} to 4×10^{-2}	0.09–0.162	91.47–95.2	0.102–0.135		
		Acidic H$_2$O/TEOS molar ratio varied from 2 to 9	0.095–0.173	90.89–95	0.105–0.125		
		Basic H$_2$O/TEOS molar ratio varied from 1.25 to 5	0.092–0.182	90.42–96.1	0.103–0.153		
MTMS	SCD	NH$_4$OH/TMOS molar ratio varied from 4.25×10^{-2} to 3.5×10^{-1}	0.1–0.35	–	0.09–0.098	~480	[21]
		H$_2$O/TMOS molar ratio varied from 2 to 10	0.18–0.41		0.094–0.109		
		MeOH/TMOS molar ratio varied from 1.75 to 17	0.12–0.30		0.086–0.091		
MTMS	SCD	MeOH/MTMS molar ratio varied from 14 to 39	0.037–0.093	95–98	0.057–0.063		[22]

TEOS tetraethoxysilane, *SS* sodium silicate, *SCD* supercritical drying, *APD* ambient pressure drying, *CP* co precursors, *MTES* methyltriethoxysilane, *MTMS* methyltrimethoxysilane, *TMES* trimethylethoxysilane, *PTES* phenyltriethoxysilane, *TMCS* trimethylchlorosilane, *HMDZ* hexamethyldi silazane, *EtOH* ethanol, *MeOH* methanol

precursors such as TEOS [12–15, 17, 18, 20, 23], TMOS [24, 25], SS [16, 19], and co-precursor [12–14, 17, 18] as well, various chemicals e.g., ethanol [20, 23], methanol [21, 22] used for ageing, silylation agents such as methyltrimethoxysilane (MTMS), methyltriethoxysilane (MTES), vinyltrimethoxysilane (VTMS), phenyltrimethoxysilane (PTMS), phenyl triethoxysilane (PTES), dimethyldimethoxysilane (DMDMS), trimethylmethoxy silane (TMMS), trimethylchlorosilane (TMCS), bis(trimethylsilyl)acetamide (BTSA) and hexamethyldisilazane (HMDZ) [16] and alteration of molar ratio of precursors [20, 23], ageing/solvent exchange [20–23] and silylation agents [16–20]. TMOS increases health hazards due to its toxic release of fumes which restricts its popularity [16]. On the other hand, comparatively cheaper sodium silicate (SS) produces impurities in the form Na ion and removal of it requires additional washing steps [16, 19]. Among the other precursors, TEOS is reported to be most popular one [12–15, 17, 18, 20, 23]. Further decrease in thermal conductivity e.g., 0.042–0.086 W/mK [12] can be achieved by using various combination of co-precursors and variation in molar ratio as well [12, 13, 17, 18].

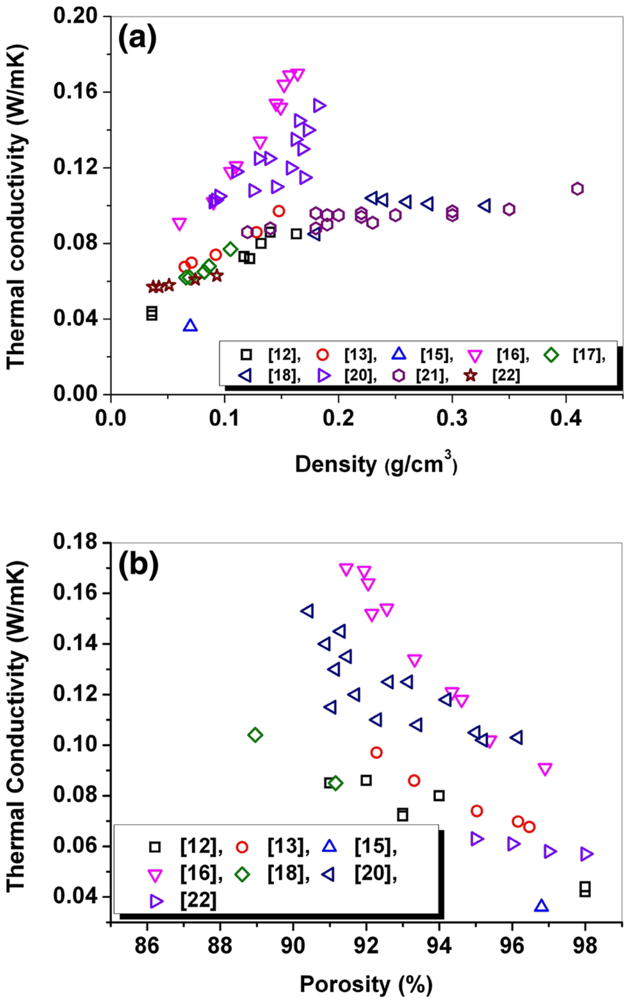

Fig. 3 Variation of thermal conductivity as a function of **a** density and **b** porosity of monolith silica aerogel

Thermal Conductivity and Stability of Silica Aerogel Composites

Thermal conductivity and thermal stability of composite aerogel reported in literature are summarized in Table 2 [26–35]. Thermal conductivity values are reported to be in the range of 0.0221–0.19 W/mK [26–35]. Further, thermal stability is documented in literature in the range 280–470 °C [26–28, 34].

It is observed that thermal conductivity and thermal stability properties are strongly dependent upon the fiber content [26–29], fiber alignment [28, 29] and type of opacifier/whisker [30–35]. Though monolith or bulk silica aerogel possesses low thermal conductivity (in particular at low temperature application) as compared to aerogel composites. However, it lacks in mechanical flexibility which restricts their applications for insulation purpose in particular.

The density and porosity of the aerogel composites are reported to be in the range of 0.05–1.35(g/cm^3) [26–35] and 65.3–93.6% [26–35], respectively. In general, the density is increased for aerogel composites as compared to bulk silica aerogel while porosity shows opposite trend. Similar to bulk aerogel, the thermal conductivity of aerogel composite decreases with decrease in density as summarized in Fig. 4.

Further, it is reported [26, 27] that thermal conductivity increases (up to 5%) with increase in fiber content. However, the increase in fiber content provides significant enhancement (more than 150%) of mechanical properties such as bending modulus and bending strength. This information signifies that the flexibility of the aerogel composites has significantly improved without compromising thermal insulating property. The improvement of flexibility of aerogel composites can be achieved by altering fiber alignments [28, 29]. However, there is no noticeable degradation in thermal insulation property [28, 29]. Further, enhancements of mechanical properties are also reported with incorporation of particulate hard ceramic such as Si_3N_4 [35].

It is reported that pure silica aerogel is highly transparent in IR region particularly from 3 to 8 µm which increases the possibility of radiative heat transfer [33]. So, to improve thermal insulation property of aerogels for high temperature applications they are reported to be doped with opacifiers such as TiO_2 [33], $K_2Ti_6O_{13}$ whisker [33], carbon [39] etc. Thus, transparent aerogel can be fabricated in the form of opaque and it can be useful in spacecraft application because heat transfer by radiation is dominated in space rather than other two modes of heat transfer i.e., conduction and convection.

Thermal stability is also depending upon the reinforcing materials with the aerogel matrix. In general, aramid fiber [27, 28] and glass fiber [36] show comparatively higher (285–600 °C) melting temperature as compared to copolyolefin fiber (110–150 °C) [41].

Prospect of Aerogel Based Thermal Insulation and Its Challenges

The aerogel composite blankets are widely used as thermal insulation for various ground and aerospace applications [42, 43]. The aerogel composite thermal insulator blanket or wrap are commercially available from vendors such as Aspen Inc., Cabot etc. and they are mainly used in industrial pipeline for gas and oil. Further, in construction and building fields [4, 44, 45] aerogel based thermal insulation elements are often used in the current decade. Aerogel thermally insulated composite panels are now widely used,

Table 2 Literature status on thermal conductivity and thermal stability of silica aerogel composite

Precursors	Key features in synthesis	Drying method	Density (g/cm^3)	Porosity (%)	Thermal conductivity (W/mK)	Thermal stability (°C)	Mechanical properties		References
							Strength (MPa)	Modulus (MPa)	
TEOS	Varying concentrations of TMCS 2–10% volume in hexane (nomex fiber + modacrylic cotton)	APD	0.09–0.11	–	0.022–0.036	~470	–	–	[26]
TEOS	Varying fiber content (2.5–7 vol%)	APD	0.142–0.152	82.6–83.7	0.0221–0.0235	285	BS: 0.059–0.137	BM: 0.528–1.1415	[27]
TEOS	Varying fiber content (3–9 vol%)	APD	0.185–0.225	80.5–84.5	00.025–0.032	370	CS: 1.81–21.03	BM (longitudinal): 1.2–12.1 BM (transverse): 2.87–12.8	[28]
	Varying fiber alignment of 4 layers (fiber content 3 vol%)		0.163–0.186	84.29–86.2	0.0233–0.0274	370	CS: 1.71–2.49	BM (longitudinal): 1.58–5.84 BM (transverse): 0.44–3.03	
TEOS	Varying fiber alignment of 4 layers	APD	0.159–0.186	–	0.0233–0.0274	–	CS: 1.71–2.49	BM (longitudinal): 1.58–5.84 BM (transverse): 0.44–3.03	[29]
TEOS	Varying aerogel in epoxy (0–60 vol%)	APD		–	0.105 (60 wt% silica aerogel)	–	–	–	[30]
TEOS	Different wt% of SiO_2 nanoparticle (0–0.25%)	APD	0.27–0.35	–	0.078–0.082	–	–	–	[31]
–	–	–	–	–	0.004 (For 'MLIB/aerogel/MLIB' architecture at high vacuum)	–	–	–	[32]
TEOS	Varying TiO_2 (0–10 wt%)	APD	–	–	0.060–0.072	–	–	–	[33]
	Varying $K_2Ti_6O_{13}$ whisker (0–8 wt%)		–	–	0.051–0.060	–	–	–	
–	Aerogel in epoxy (40–90 vol%)	–	0.25–0.73	46–85	0.04–0.15	300	–	–	[34]
TEOS	Varying vol. fractions of Si_3N_4 (5–20%)	APD	0.32–0.82	65.3–85.88	0.024–0.072	–	CS: 3.24–12.12 FS: 0.36–2.45	–	[35]

TEOS tetraethoxysilane, *TMCS* trimethylchlorosilane, *APD* ambient pressure drying, *BS* bending strength, *CS* compressive strength, *FS* flexural strength, *BM* bending modulus, *MLIB* multilayer insulation blanket

particularly in European countries and North America [4], which saves energy and thus global warming can be minimized. Recently, aerogel is also reported to be used as thermal insulation in textile industry [46].

Apart from the ground thermal insulation application, bulk aerogel and aerogel composite blankets are also used in space applications [42, 47]. For example, aerogel based thermal insulation blankets are successfully implemented in cryogenic propulsion tank [48, 49], Mars rovers e.g., Sojourner in 1997, Spirit and Opportunity in 2003 by NASA [42]. Further, the possibility of replacement of Kapton based multilayer insulation (MLI) blanket is also studied and reported by several researchers [32, 50, 51]. The aforesaid research results [32, 50, 51] concluded that MLI shows much effective thermal insulation behaviour in high vacuum environment as compared to aerogel blanket. However, because of the presence of an environment rich in CO_2 at Martian surface, aerogel based thermal insulation

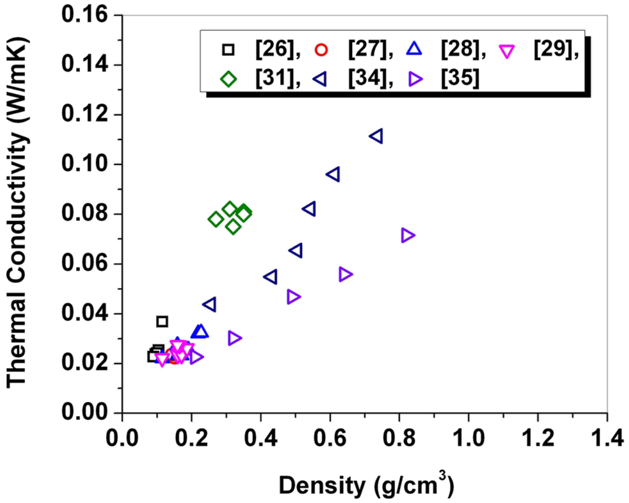

Fig. 4 Variation of thermal conductivity as a function of density of silica aerogel composite

may perform better than conventional MLI [32, 50, 51]. Further, due to light weight, superior outgassing behaviour and flexibility made aerogel blankets more attractive for spacecraft thermal control applications. It is also reported that aerogel blankets can be applied for micrometeorite protection particularly for the spacecraft in low earth orbit (LEO) besides its thermal protection [32, 50, 51].

Though nowadays aerogel has become popular for its super insulating behaviour with ultra low weight, but it suffers from high production cost and poor mechanical properties [43]. However, several research and developmental attempts are ongoing for economic scale up with improved mechanical properties [52].

Conclusion

Based on the above brief literature review, it can be concluded that both silica based aerogel monolith and composites are promising material for thermal insulation as compared to the present insulating materials available. This review provides an idea about the major synthesis stages and chemistry behind silica aerogel and it's composite. To achieve desired thermal insulation properties with adequate mechanical flexibility, the process parameters of monolith and composite aerogel can be tailored. Thermal conductivity and thermal stability of different silica aerogel monolith and composites reported in literature are summarized thoroughly. Further, the alteration of thermal conductivity of aerogel materials with porosity and density are also illustrated. Also, uses of aerogel for ground and space applications and market analysis are discussed in the present review.

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