

Investigation of the effects of silica aerogel particles on thermal and mechanical properties of epoxy composites

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Abstract

Aerogel particle filled epoxy was fabricated in this work by investigating the effect of resin viscosity on pore infiltration and density of resulting composites. Aerogel porous structure was successfully preserved by carefully selecting epoxy viscosity for mixing procedure. The effect of aerogel content and particle size was subsequently investigated on thermal conductivity and compressive properties of epoxy composites. The results showed that incorporating aerogel particles into the resin could lead to over 40% reduction in both density and thermal conductivity. However, there was a linear trade-off between compressive properties and thermal conductivity of silica aerogel/epoxy composites. A wide range of aerogel particle sizes was also studied and it was found that the size effect was dependent on the aerogel content. Overall, the results of the presented work allow conclusions to be drawn regarding the usefulness of delayed wet mixing technique and the impact of particle size and loading on aerogel filled resin composites.

Keywords: Silica aerogel, Particulate-filled composites, Thermal conductivity, Compressive properties

1. Introduction

With growing concern regarding global energy consumption, novel solutions to reducing energy loss are attracting more attention [1, 2]. Silica aerogel is an extremely lightweight and highly porous material, which possesses one of the lowest thermal conductivities documented for solid materials [3]. As a result, aerogel is an arguably ideal insulating material and has multiple applications. Despite the first silica aerogel being produced by Samuel Kistler in 1931, research into these highly efficient insulators was not widespread until much later, due to the lengthy and complex production process required [4]. A great deal of academic and practical research has been undertaken in an attempt to strengthen silica aerogel, which is extremely brittle in nature. This research has generally involved the combination of aerogels with more durable materials, allowing the structural weaknesses to be overcome while providing superior thermal properties. One such approach was based on synthesizing the aerogel with the addition of fibre reinforcement, resulting in the creation of aerogel blankets [5, 6]. Others have attempted to develop the superior thermal characteristics of aerogel-based materials through the addition of silica aerogel particles or hybrid aerogel structures [4, 7-9]. Nevertheless, recent aerogel scholarship has primarily focussed on mechanical properties of aerogel-filled composites and has not exploited its full potential, in terms of thermal properties [10-12].

Gupta et al. analysed the interaction between aerogel particles and epoxy matrix in a mechanical context. It was found that a high viscosity resin resulted in poor binding while a low viscosity resin invaded the aerogel pores [13]. Specimens were created using a heated high viscosity resin, which enabled the production of a composite that had a slightly lower density and improved mechanical characteristics when compared with the plastic matrix [13]. Pore infiltration was also observed by Salimian et al. who investigated the structure-property relationship in epoxy nanocomposites filled with silica aerogel [11]. In their study, no reduction in thermal conductivity of epoxy was found due to fully filled pores, whereas the mechanical properties improved significantly. It was suggested that the introduction of the polymer networks into the aerogel mesopores increased the composite toughness by limiting the crack propagation and deflection of the composite [11]. Different manufacturing techniques were utilised in order to preserve the aerogel pores from resin infiltration, as presented by Kim et al [12]. In their work, aerogel was filled with ethanol prior to mixing with epoxy. Such an approach demonstrated how to prevent pores from being filled and led to a thermal conductivity of 60 and 45 $\text{mWm}^{-1}\text{K}^{-1}$ for samples with 25 vol% and 75 vol% aerogel loading respectively. However, this method resulted in a high cost and hazardous manufacturing conditions [12]. An

alternative method was employed by Maghsoudi and Motahari who used high viscosity resins [14]. Epoxy resin, paired with triethylene tetramine as a curing agent and particles with diameters between 200 and 300 nm, was used to prepare 0.5-3 wt% silica aerogel filled composites. The results showed that the addition of 3 wt% aerogel increased the contact angle from 43° to 118° and decreased the thermal conductivity from 195 to 74 mWm⁻¹K⁻¹ [14]. Similarly, Zhao et al. studied the thermal properties of silica aerogel/epoxy composites and discovered that, at 60 wt% aerogel particles, a thermal conductivity of 105 mWm⁻¹K⁻¹ could be achieved, in addition to an increased serviceability temperature [15]. Two sizes of aerogel particle were used (200 μm and 2000 μm) to produce composites, and it was concluded that the addition of larger aerogel particles resulted in lower thermal conductivity and a higher martens heat distortion temperature [15]. Nevertheless, no mechanical testing was performed in their study. Finally, Krishnaswamy et al. investigated the morphology, optics and thermal character of aerogel/epoxy composites by increasing the resin viscosity to the point where aerogel pores were no longer infiltrated following mixing [16]. The resulting composites presented a 13.3% decrease in thermal conductivity when 3 wt% silica aerogel particles were added. Under microscopy it was found, however, that physical destruction of the aerogel particles had occurred due to epoxy curing shrinkage. Such destruction indicated a larger surface area of the particle through which the resin could now enter the aerogel interior pore network [16].

The effect of silica aerogel particles on other matrix systems has also been measured. Halim et al. investigated the impact of three different particle sizes (powder, granules and beads) with the same 30 vol% loading on thermal and mechanical properties of unsaturated polyester composites [17]. Their work revealed higher effectiveness of larger particles, resulting in higher thermal stability and lower thermal conductivity compared with unfilled resin. However, the authors stressed the need for aerogel pore preservation during manufacturing and that tensile properties of created composites decreased [17]. Conversely, Venkata Prasad et al. investigated the properties of sisal/poly lactic acid (PLA) combined with nanometre size silica aerogel particles up to 2 wt% [18]. This study observed that the addition of particles impacted the interfacial adhesion between fibres and matrix, resulting in tensile and flexural strength increase. This not only improved thermal stability and mechanical properties of created composites, but also identified the opportunity to use the silica aerogel particles as an additive in biodegradable composites [18]. Furthermore, Kim et al. tried a combination of silica aerogel particles (average diameter of 9 nm) and polyimide using a multi-curing process [19]. They

managed to successfully preserve aerogel pores and obtained a thermal conductivity of 0.03 W/mK with 50 vol% aerogel content [19]. Other researches led by Cho and Lee included a mixture of silica aerogel with respectively polyurethane and poly(dimethylsiloxane) (PDMS) matrices, achieving significant decreases in thermal conductivity of both materials and further highlighting the importance of aerogel pore preservation [7, 20]. Moreover, a novel method of silica aerogel incorporation was proposed by YG Kim, who has manufactured polyvinylidene fluoride nanofibers filled with silica aerogel [21]. The fibres with a porous internal structure possessed a low thermal conductivity as well as high flexibility and fire retardation [21]. Furthermore, a more in-depth overview of aerogel particulate composites systems has been prepared by Salimian et al., who has described up-to-date work on nanoporous fillers in polymer based composite [22]. Alternatively, Kim et al. summarised the principles of heat transfer in polymer/aerogel composites and the most important physical factors impacting their thermal conductivity [23]. This paper investigated the effect of the addition of silica aerogel particles to epoxy resin to maximise its thermally insulating properties, while minimising the adverse effect on compressive properties. A delayed mixing method was developed by analysing viscosity changes during the resin curing stages and subsequently evaluating the influence on the composites' thermomechanical properties. This identified a more cost-effective method of introducing aerogel particles to a liquid matrix while maintaining the porous internal structure of aerogel. This was also the first time that a combined effect of varying silica aerogel particle size and loading on the epoxy composite had been studied.

2. Experimental

2.1. Materials

The epoxy resin and hardener were purchased from Easy Composites Ltd. The resin was a mixture of bisphenol-A-epoxy resin and Epichlorohydrin-formaldehydephenol polymer (trade name IN2 Epoxy Infusion Resin) both with molecular weight equal to or less than 700 g mole⁻¹. A cycloaliphatic amine-based hardener (trade name AT30 Slow Hardener) was used as the curing agent. The silica aerogel particles (SiO₂) produced by Enersens were sourced from Aerogel UK Ltd. A total of four batches of different particle sizes were purchased with nominal average particle size quoted as 100 μm, 500 μm, 1250 μm and 3500 μm respectively (trade name of particles was respectively Kwark®XP100, Kwark®XP500, Kwark®GS and

Kwark®GL). These aerogels have a nominal density between 40 and 80 kg/m³, specific surface area of 850 m²/g and pore diameter ranging from 5 to 12 nm.

2.2. Silica aerogel/epoxy preparation

Silica aerogel/epoxy composites were fabricated using a delayed wet mixing technique. Initially, a weight ratio of 100 parts resin to 30 parts hardener were combined using manual stirring for 5 minutes. The mixture was subsequently degassed under vacuum for another 10 minutes. The mixture was then left to react under ambient conditions for different time intervals, at which 0-5 wt% of silica aerogel particles of varying sizes were added to the resin. The silica aerogel/epoxy mixture was then poured into silicon moulds coated with Ambersil silicone release agent and was left to cure at room temperature for 24 hours, followed by post curing at 60°C for 6 hours. All aerogel/epoxy composites were machined and polished to the sample dimensions required by ASTM standards [24, 25]. Finished samples for both thermal and mechanical testing are presented in Figure 1. The cylindrical samples for conductivity measurement have a diameter of 50.8 mm and a height of 25.4 mm. The dimensions of the prism samples for compression tests are 12.7 mm × 12.7 mm × 25.4 mm.

2.3. Gas adsorption

To characterise the pore structure inside the silica aerogel particles, adsorption/desorption measurements were conducted using a Micromeritics ASAP2420 machine. Nitrogen gas was used at a temperature of -195.8 °C and the outgassing procedure took 24 hours to complete. The total pore volume was calculated from the amount of gas adsorbed when reaching 0.99 relative pressure, while specific surface area was obtained using the BET (Brunauer-Emmett-Teller) method. Finally, pore size distribution and average pore size were obtained by analysing the adsorption isotherms through the BJH (Barrett–Joyner–Halenda) model.

2.4. Viscosity measurement

The viscosity measurement of epoxy resin is essential for controlling the time intervals for the addition of silica aerogel. Such a measurement was conducted at room conditions using a rotational viscometer at 300 rpm. This constantly monitored the torque required for the spindle (25 mm diameter) to move through the resin.

15 batches of epoxy resin were prepared, each comprising 50 g of epoxy and 15 g of hardener. One sample was placed underneath the viscometer in order to measure viscosity changes as a function of time; simultaneously, the remaining epoxy samples were mixed with 2 wt% 500

μm silica aerogel particles at specific time intervals. After silica/epoxy samples had been cured, as described in 2.2, the samples were tested to obtain the thermal conductivity and their bulk density was calculated through measured sample weight and volume.

2.5. Imaging analysis

As only the maximum diameter of aerogel particles was available from the supplier, a more detailed analysis of particle size was required in order to properly investigate the effects of particle size. As a result, 0.1 g of aerogel particles were dispersed in 100 ml of acetone with additional glycerine and stirred for 5 minutes using a magnetic stirrer at 500 rpm. Afterwards a sample of the solution was transferred onto a glass plate and dried out to avoid particle agglomeration. Finally, the glass plates were scanned in an illuminated dark chamber. The scanned images as presented in Figure 2 were further analysed using Image Pro and Image J software for the comparison purposes. In the case of irregularly shaped particles, the system drew an ellipse around the anomaly and reported its major axis length (ellipse length) and minor axis length (ellipse diameter) which were later used to assign particles to specific categories. The analysis conducted with both software revealed no major differences, however, Image Pro was selected for further analysis due to its ability to measure large quantities of particles at the same time. All available silica aerogel samples were tested under same procedure and at least 800 particles of each nominal size were analysed to obtain a size distribution. Cumulative distribution was presented by particle volume instead of the number of particles in order to avoid skewness in favour of smaller particles when a broad size distribution to be expected.

Finally, scanning electron microscopy (SEM) images were taken to confirm an equal distribution of particles. The samples were initially coated with a layer of gold and images were taken using 15 kV accelerating voltage in a HITACHI SU-6600 machine.

2.6. Conductivity measurement

A TA FOX 50 heat flow meter was used to measure the thermal conductivity of the silica aerogel/epoxy composites in accordance to ASTM C518 [24]. All test specimens were cast using a silicone rubber mould and had a cylindrical geometry with a diameter of 51 mm and a thickness of 25.4 mm. In order to minimise the effect of any surface defects, a silicon thermal paste was applied to both horizontal surfaces of each sample. The temperature difference was set to be 25 °C and 15° C respectively. The thermal conductivity was calculated using Fourier's law for one-dimensional heat conduction at steady state. At least two samples for each silica aerogel/epoxy blend were tested.

2.7. Mechanical testing

Compression tests in accordance to ASTM D695 were undertaken to evaluate the effects of the silica aerogel addition on compressive properties of epoxy specimens [25]. An Instron 5969 series universal testing system, fitted with 50 kN load cell and equipped with a video extensometer, was used to load rectangular samples with the dimensions of 12.7×12.7×25.4 mm at a constant rate of 1.3 mm/min at 20 °C. Following load application, the compressive strength was calculated by dividing the maximum compressive load experienced by the sample by its' smallest initial cross-sectional area [25]. On the other hand, the modulus of elasticity was obtained by inserting a line tangent to the linear section of the load deformation curve [25]. Followed with selection of a point on the linear part and dividing compressive stress of this point by the corresponding strain which was measured from the crossing point between extended tangent line and the strain-axis [25]. At least 5 samples for each silica aerogel/epoxy blend were tested.

3. Results and discussion

3.1. Particle size distribution

Figure 3 presents the size distribution of silica aerogel particles as received from the supplier. The results show that the single value of the particle size given by the supplier only covers the upper range of particles. In addition, due to transportation and handling it is anticipated the particles size was further reduced. It can also be noticed that the size of most tested particles follows a normal distribution with the exception of the largest particles exhibiting negatively skewed distribution. As seen from the results in Figure 3, the measured average particle diameters are approximately 85, 200, 600 and 1230 μm with respect to the corresponding 100, 500, 1250 and 3500 μm as received. These average values obtained from the measurement were subsequently chosen to differentiate four corresponding batches of silica aerogel particles.

3.2. Silica aerogel pore structure

Nitrogen adsorption and desorption has been used to characterise the internal pore structure of silica aerogel samples. The isotherms obtained are presented in Figure 4. All aerogel/epoxy blends present a similar shape of type IV isotherm which indicates mesoporous material with pore diameter in a 2 – 50 nm range. The shape also suggests condensation of the gas inside pores at low pressure ranges, with further formation of mono and multi layers at higher pressure. In addition, the type IV hysteresis in Figure 4 implies the presence of spherical pores with walls made of mesoporous silica.

The BET surface area and average pore size of the specimens are summarised in Table 1. The specific surface area of aerogels is significant with an average value of 801 m²/g indicating high porosity of the material. The adsorption pore width of all samples varies between 1.7 nm to 70 nm, with an average of 17.5 nm and a cumulative pore volume of 3.55 g/cm³. These pore characteristics suggest that a high quality of the aerogel was obtained and varying particle sizes had little impact on porous structure.

3.3. Epoxy viscosity

Figure 5 presents the measured shear viscosity of pure epoxy resin as a function of time after the addition of the hardener. An initial average viscosity growth of 0.078 Pa·s per minute was detected before gelation point, followed with a rapid increase at a rate of 0.831 Pa·s per minute. This resin system provided sufficient time for incorporating aerogel particles.

Figure 6 shows both the measured density and thermal conductivity of cured silica/epoxy composites with a 2 wt% of 1250 µm silica aerogel particles added at different epoxy curing times. The following section will focus on the impact of viscosity on composite density, while the impact on thermal conductivity will be discussed in Section 3.4.

As can be seen, the addition of aerogel particles resulted in no reduction in the density of silica aerogel/epoxy composites within the first 150 minutes of curing time. Thereafter, the density of composite samples gradually decreased by 28% with aerogel addition at 280 minutes of curing time. Afterwards, the addition of silica aerogel particles became impossible due to high viscosity, restricting uniform distribution of particles across the samples and trapping a significant volume of air pockets during mixing. The onset of density change corresponds to resin viscosity of 14 Pa·s, which is marked by an ‘open circle’ in Figure 5. It is clear from Figure 5 and 6 that there is a strong correlation between composite density and epoxy viscosity.

Figure 7 presents the relation between silica aerogel/epoxy density, thermal conductivity and epoxy viscosity for the chosen time of aerogel addition. Similar to the results in Figure 6, there was little density change when epoxy viscosity was below 14 Pa·s. However, a sharp density drop was found between 14 Pa·s and 18 Pa·s, which signified the approach to the resin gelation point as shown in Figure 5. The composite density was left largely unaltered after this abrupt drop. The change in composite density in Figure 7 could be explained by the effect of resin viscosity on infiltration aerogel pores. It seems reasonable to assume that the low viscosity epoxy may have caused open aerogel pores to be filled with resin during mixing and consequently led to higher composites density. As resin viscosity increases, resin molecules

grow larger and gradually lose their mobility. This hinders pore infiltration and preserves open pores inside the silica aerogels embedded in the resin. Consequently, the density of cured composites decreased during this stage as shown in Figure 7. It is also possible that a higher viscosity can cause more air bubbles to be trapped in the resin which in turn can reduce the density. In order to rule out this possibility for the observation in Figure 7, resin was stirred at different time intervals without adding any aerogel particles. The results showed no measurable change in the density of cured resin within 280 minutes of mechanical stirring. This supports the suggestion that the decrease of the composites density in Figure 7 was most likely caused by preserving the pores inside the aerogel particles due to increased resin viscosity. Figure 8 compares SEM images of aerogel particles embedded in the epoxy that had aerogel incorporation at the beginning of curing and at the 240th minute of curing. It can be seen that the former does not show detestable porous whereas the latter is still highly porous. To further investigate the degree of pore infiltration, the specific volume (V) of filled pores was calculated from the measured composites density using equation (1). As a result, the volume fraction (V_{inf}) of filled pores was obtained from equation (2) with respect to the specific volume (V_o) of original unfilled pores. This can help to assess the quantity of pores filled by the resin during aerogel/epoxy processing, with $V_{inf} = 100\%$ or 0% indicating complete or no pore infiltration, respectively.

$$V = \left(\frac{100}{wt\%}\right) \times \left(\frac{1}{\rho_{composite}} - \frac{1}{\rho_{resin}}\right) + \frac{1}{\rho_{resin}} - \frac{1}{\rho_{silica}} \quad (1)$$

$$V_{inf} = \frac{V_o - V}{V_o} \quad (2)$$

where, $\rho_{composite}$, ρ_{resin} and ρ_{silica} are densities of composites (as in Figure 7), epoxy resin (1.312 g cm^{-3}) and β -Cristobalite silica (2.18 g cm^{-3}) [26]. Additionally, $wt\%$ represents the aerogel weight fraction of the composite and V_o was obtained from gas adsorption analysis as shown in Table 1. As the only variable in the equation above is the composites' density, the variation of V_{inf} is determined by the measured composites' density. However, since it is aerogel pores that affect the composites' density, it is more meaningful to plot the composites' density normalised to the unfilled resin density as a function of calculated volume fraction of filled pores as shown in Figure 9. It can be deduced from Figure 9 that a percentage of unfilled resin weight of nearly 73% would be achieved by incorporating the epoxy with 2 wt% addition of silica aerogel particles, whose pores had not been filled. Notably, if using the rule of mixture to calculate normalised composite density for each mixture, a result of 76% can be obtained.

Since the rule of mixture estimates an upper boundary of the composite properties, a close fit to the experimental data collected was achieved.

3.4. Effect of Resin viscosity on composites' thermal conductivity

The results regarding thermal conductivity presented in Figure 6 and 7 mostly follow a similar trend to that observed in density variation. The thermal conductivity showed little variation around the initial value of $190 \text{ mWm}^{-1}\text{K}^{-1}$ (the same conductivity for pure epoxy) when the aerogel particles were mixed with the resin at the viscosity below $14 \text{ Pa}\cdot\text{s}$. Between 14 and $18 \text{ Pa}\cdot\text{s}$ a sharp conductivity drop was followed by another independent stage of resin viscosity. As a result, it is reasonable to assume that the delayed mixing technique or the direct control of resin viscosity is a viable route to tailoring the density and the thermal conductivity of aerogel/resin composites. These results clearly demonstrated that, in order to significantly reduce thermal conductivity of resin materials, the porous structure of aerogels need to be preserved. Similar behaviour was also observed by Lee et al., who noted that a decrease in thermal conductivity only took place when pores were preserved [7]. Based on the above results, it was decided that all silica aerogel/epoxy samples used for the following analysis would be created by mixing the aerogel particles at the 240th minute during epoxy curing cycle.

3.5. Effect of silica aerogels on composites' thermal conductivity

Figure 10 presents the thermal conductivity of silica aerogel/epoxy composites as a function of different aerogel weight fractions. The incorporation of silica aerogel in epoxy matrix reduced the composites' thermal conductivity as aerogel loading increased. The correlation between these seems to be linear for the different particle sizes investigated in this work. However, there appears to be a somewhat different behaviour from the smallest particles when compared with the rest. As seen in Figure 10, the addition of $85\mu\text{m}$ aerogel particles at lower content results in more conductivity decrease than that caused by much larger particles. This trend gradually shifts inverts as aerogel content increases up to $5 \text{ wt}\%$.

Two competing mechanisms may explain this behaviour. Firstly, the number of particles introduced to the resin can affect heat transfer in filled resin. Dispersed particles create a barrier for energy transfer and reduce the number of direct paths through the composite, hence lowering the thermal conductivity. Smaller particles can better implement such benefit as larger quantities of particles are present in the system for the same weight fraction. In contrast, larger particles possess a lower number of granules in the composite in the same volume, resulting in more aerogel-free paths through the epoxy. This leads to a lesser reduction in thermal

conductivity, however the effect is negated by usage of higher loadings of aerogel [27]. Another important factor negating aerogel properties is the infiltration of aerogel pores by the resin. Despite delaying addition of particles until reaching the desired viscosity of epoxy, a degree of pore infiltration still occurs. However, this effect is greater with smaller particle sizes, due to their higher surface-to-volume ratio. As a result, smaller aerogel particles tend to have more filled pores than larger particles at a given weight fraction. Thus, as aerogel content increases, the effect of this factor may gradually outweigh the effect of the absolute number of aerogel particles in the resin and cause smaller particles to bring about less impact on decreasing conductivity. While this hypothesis requires further work to verify, the results shown in Figure 10 seem to suggest that aerogel particle size should at least be considered as one of the important parameters for designing aerogel filled composites.

Figure 11 presents the thermal conductivity of silica aerogel/epoxy composite as a function of density. As expected, the conductivity rises as the density increases and they correlate well in a linear fashion with an r-squared value of 0.97.

3.6. Effect of silica aerogels on composites compressive properties

Figure 12 plots compressive stress-strain curves of 85 μm silica aerogel/epoxy composites at different weight fractions. It is clear that the addition of aerogel particle can significantly affect mechanical behaviour of the resin. The neat resin exhibits a distinct yield point after initial linear behaviour. After yield, the resin experiences strain hardening at large deformation. In contrast, the resins doped with silica aerogel particles tend to develop a less distinct yield point. It can also be noticed that incorporating the resin with the silica aerogel can make the resin become more brittle and less stiff simultaneously.

Figures 13 and 14 give the compressive stress at yield and modulus against aerogel content and different particle sizes. It shows that both compressive yield strength and modulus of the silica aerogel/epoxy composite decrease as aerogel content increases for the range of particle sizes investigated in this work. Such correlation is anticipated as porous silica aerogels have much lower mechanical properties than the epoxy resin. Similar to what is observed in Figure 10, it is also interesting to note that the small particles tend to display little effect at high aerogel content ($> 3\text{wt}\%$). On the contrary, large aerogel particles tend to monotonically reduce yield strength and modulus up to 5wt%.

The addition of porous particles can introduce stress concentration, causing a weakening effect [28]. This is more detrimental with irregular particles than their rounded counterparts [28]. Due

to the fact that silica aerogels are mostly composed of air, their presence inside the resin, to some extent, resemble voids [29]. As a result, larger silica aerogels have more influential weakening effects, causing premature failure of the parent material [28]. At small aerogel content, there could be a small probability of finding a critically large and irregular particle in the resin. As increasing aerogel content is likely to increase such probability, more detrimental effect associated with larger particles tend to reveal at large aerogel content as shown in Figures 13 and 14. In addition, small particles form large interfacial areas with the resin due to the large surface-to-volume ratio. As mentioned earlier, there is still a small degree of pore infiltration occurring during aerogel/epoxy mixing. It follows that the total volume effectively occupied by the aerogel is probably smaller for smaller particles. This can potentially mitigate the weakening effect as aerogel content increases as seen in Figure 13 and 14. It is interesting to see that the compressive yield strength and modulus even reach a plateau at high aerogel content. Further work is needed to understand this observation.

Figure 15 shows the compressive yield strength against the composite density. As expected, the yield strength increases with material density. Figure 16 attempts to correlate the compressive yield strength with thermal conductivity of silica/epoxy composites with all four groups of particle sizes. A strong linear relation (r-squared value of 0.96) was found between these two parameters despite particle size differences, however caution should be taken before further extrapolation of this relationship outside the investigated data range. Comparing the overall trend in Figures 10 and 13, one can see the effect of particle size on both properties coincide with each other. This paper's results clearly indicate the possibility of enhancing the thermal insulating properties of epoxy resin by introducing silica aerogel particles. An incorporated manufacturing method allows for simple and low-cost production of such composites, making them accessible to broad range of applications. On the other hand, addition of aerogel deteriorates compressive properties of epoxy resin. As a result, those materials are more likely to be used in the scenarios with limited external loading applied to them. However, by investigating both thermal and mechanical properties, the opportunity for careful balancing of those properties has been created.

4. Conclusions

In this work, the effect of silica aerogel content and particle size was investigated on thermal conductivity and compressive properties of epoxy composites. The influence of resin viscosity

on pore infiltration was investigated and the results suggested that combining silica aerogel and the resin at a carefully selected viscosity can successfully prevent pores from being filled by the resin. By doing so, it has been clearly shown that incorporation of silica aerogel particles into the resin can lead to significant reduction in both resin density and thermal conductivity. However, there is a linear trade-off with compressive properties of silica aerogel/epoxy composites. A wide range of silica aerogel particle sizes were also studied; it was found that the size effect does not seem to be remarkably evident at small aerogel content. At large aerogel contents (>3 wt%), however, small aerogel particles tend to have a lesser effect on the thermal and mechanical properties of the resin. For the investigated composite system, there is even a tendency for thermal conductivity and compressive properties to be unaffected by further addition of small aerogel particles once a certain concentration is reached. In order to fundamentally understand thermomechanical behaviour of aerogel/resin systems, more work is required. This could focus on the aerogel/epoxy interface region and its influence on composite properties or different types of aerogel and matrix allowing a more general conclusion to be reached. Nevertheless, the results in this study provide insight into both fabrication methods and key design parameters of aerogel filled resin composites.

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Table 1. Pore characteristics of silica aerogel particles obtained from BET analysis

Nominal particle size [μm]	100	500	1250	3500
BET surface area [m^2/g]	761.42	811.49	750.19	881.52
BJH adsorption average pore width [nm]	17.16	16.58	19.63	16.44
BJH desorption average pore width [nm]	13.43	12.84	16.17	14.27
Cumulative pore volume (cm^3/g)	3.47	3.55	3.88	3.65