



Silica aerogel infused hierarchical glass fiber polymer composites

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ABSTRACT

Hierarchical systems can address the matrix-dominated failures of structural fiber polymer composites. Here, a new synergistic hierarchical structure combines conventional structural glass fibers with a bi-continuous silica-based aerogel matrix; both pure-silica and organically-modified silicate aerogels are demonstrated. When infused with an epoxy matrix, this type of hierarchical architecture showed a marked improvement in mechanical properties: without any loss in modulus, both the compressive strength and the interlaminar shear strength increased by up to 27%, relative to the equivalent glass-fiber reinforced epoxy composite baseline. The bi-continuous network modification strategy uses industrially-relevant infusion techniques, at or near room temperature, and retains a similar final composite density (within 2%). The strategy presented here provides a versatile and readily applicable means to improve state-of-the-art continuous fiber reinforced composite systems in compression and offers an opportunity to develop a new generation of composite materials.

1. Introduction

Ten million tons of glass fibers are produced per year, and glass fiber reinforced polymers (GFRPs) make up 95% of the composites market [1, 2], due to their lower cost than higher performance carbon fiber systems. However, such composites are often limited in application by the inferior properties of the matrix or the fiber-matrix interface [3]. Poor matrix properties can be mitigated by reinforcing the polymer matrix with nanomaterials, forming a hierarchical reinforced composite. Such systems are often inspired by biological examples, including those based on silicates [4] or other minerals [1]. A wide range of strategies for hierarchical composites have been studied, including simple infusion of low loading, discrete nanoparticle-filled resin systems [5], and other processing techniques designed to increase the nanomaterial content [6]. The usual goal is to maximize the dispersion quality of individual reinforcements, to avoid any stress concentrations associated with agglomerates. One of the most successful commercial resins containing

discrete particle reinforcements [7] uses silica nanoparticles synthesized *in-situ* within the resin to maximize processability. An alternative strategy locates the nano-reinforcements specifically at the fiber-matrix interface, via directed growth or grafting reactions, to address debonding or delamination issues [8,9]. This paper focuses on a new approach to forming hierarchical, fiber-reinforced nanocomposites, exploiting aerogel technology in a new context, to provide a bicontinuous, fully connected reinforcement throughout matrix phase.

Pure silica aerogels have been developed for their low density, high transparency, large specific surface area, low thermal conductivity, low refractive index and high porosity [10], with wide application in space [11], construction [12], bioengineering [13], and medicine [14]. Aerogels consist of a highly porous network of fused nanoparticles, typically with a primary particle size around 10 nm. Due to their low density (often much less than 0.1 g cm⁻³), they typically have low strength (compression strength on the order of 3 MPa or less [15,16]). Large freestanding structures can be hard to manufacture due to shrinkage effects, related to surface tension during drying. On small samples,

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Nomenclature

BET	Brunauer, Emmett and Teller
ESI	electronic supplementary information
GFRP	glass fiber reinforced polymer
ILSS	interlaminar shear strength
ormosil	organically-modified silicates
PDMS	polydimethylsiloxane
RIFT	resin infusion under flexible tooling
SEM	scanning electron microscopy
TEOS	tetraethyl orthosilicate
3D	three-dimensional

capillary effects can be avoided via supercritical drying; ambient drying typically produces denser, more collapsed 'xerogels' [17]. On small specimens, aerogel networks have been partially coated with polymers (often referred to 'X-aerogels') to produce aerogel-polymer, two component systems, with retained porosity; despite the incomplete filling, this approach can significantly increase compressive properties (compressive strength up to 300 MPa [18]), depending on the infused polymer [15–17]. To create larger specimens, aerogels are combined with nanofibers or nanotubes, to reinforce their primary nanostructure [15,19–21], or hybridized with low volume fraction, discontinuous fiber

felts, arrays, or mats [21–28]. These aerogel-fiber structures (two component systems) are generally designed to retain the low density and porosity of the aerogel, whilst improving the thermal, electrical and wetting properties relevant to typical aerogel applications [29–32]. There is limited literature on the mechanical properties of silica aerogel reinforced fiber hybrids [22–25,27,33,34], with compressive and tensile strengths remaining very low, on the order of 50 MPa and 15 MPa [25], respectively, due to the low fiber content and/or their random, discontinuous arrangement. Modelling of these, low volume fraction, high porosity (ca. 95%), short fiber silica aerogel hybrids shows that mechanical performance depends on aerogel particle size, fiber stiffness and fiber orientation [35].

For mechanical purposes, a matrix presenting a continuous, interconnected and rigid three-dimensional (3D) nanoscaled network appeals as a means to transfer load between the primary structural fibers in a polymer matrix composites. In particular, longitudinal lamina compression performance is known to depend crucially on the matrix shear/compression modulus [36]; hence, we hypothesize that a stiff continuous silica network matrix may be especially relevant to structural fiber composites in compression. However, as noted, silica networks have limited mechanical performance, as synthesized, due to their low densities. Our objective, therefore, was first to create a monolithic silica aerogel network/matrix around a conventional structural, high volume fraction woven glass fiber pre-form (Fig. 1 (a)), then infuse a conventional structural resin/matrix, to form an organized, bi-continuous nanocomposite matrix *in-situ* (Fig. 1 (d)). The resulting

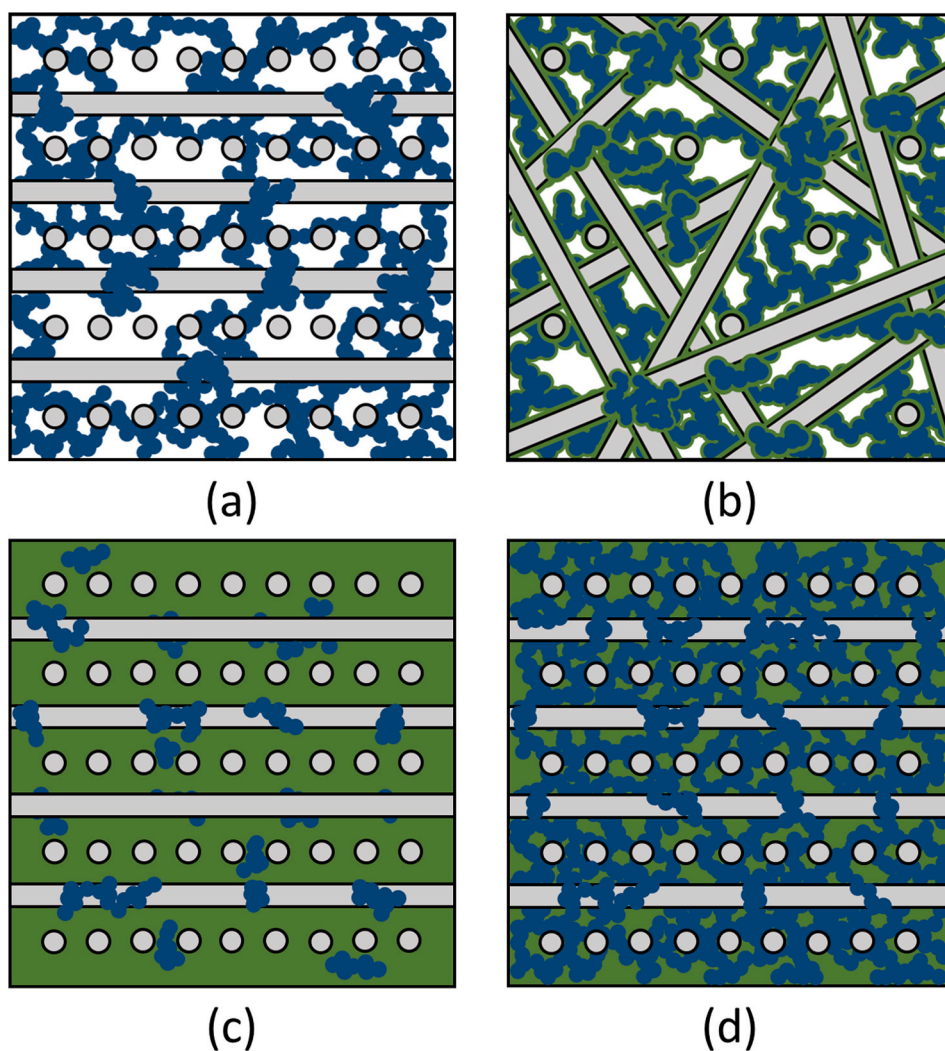


Fig. 1. Schematic representations of alternative fiber, aerogel, and matrix hierarchical composite architectures. (a) Continuous silica aerogel network [porous] with continuous ordered (woven) fiber reinforcement [34]. (b) Continuous porous silica aerogel network with [39] or without [40] matrix-crosslinking (typically epoxy coating, "X-aerogel", as shown) with continuous disordered glass fiber reinforcement (wool), note that the matrix cross-linking is only coating the reinforcing fibers and aerogel and does not fill the whole volume [not fully back-filled and remaining porous]). (c) Discontinuous aerogel dispersed in matrix with continuous ordered (woven) glass fiber reinforcement [38] [all volume in system filled, no porosity]. (d) Continuous matrix-filled silica aerogel (bicontinuous matrix) network with continuous ordered (woven) glass fiber reinforcement (this work) [all volume in system filled, no porosity]. Fibers shown in grey, aerogel in blue, fully backfilled matrix regions shown as green fill, fiber/matrix-crosslinked regions shown with green border, and porous regions shown in white. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

hierarchical fiber matrix composite contains three phases (fiber, aerogel, resin) with three different characteristic lengthscales (fiber diameter $\sim 10\ \mu\text{m}$, primary silica particles $\sim 10\ \text{nm}$, and molecular epoxy $\sim 1\ \text{nm}$). This system is distinguished from existing glass fiber, silica aerogel network, resin/matrix materials by its unique combination of a continuous aligned fiber reinforcement with a continuous silica network throughout the matrix, fully back-filled with resin.

Previous examples combining long fibers, silica aerogel, and resin do not address our underlying hypothesis as they contain either discontinuous (powdered and discrete) silica aerogel particles [37,38] (Fig. 1 (c)), or disordered fibers (Fig. 1 (b) in this instance not fully back-filled with resin akin to the X-aerogels described previously [39], but also produced without resin [40]); furthermore, none of these silica systems consider composite compression performance. A related example using a low temperature graphene oxide-based aerogel formed *in-situ* around random short aramid fibers, back-filled with epoxy, was studied in compression [19]; however, the compressive strength (127 MPa) and modulus (2.1 GPa) were limited by the lack of alignment and low primary fiber fraction. In contrast, traditional structural fiber composites exploit high volume fractions of aligned, continuous fibers to provide significant structural performance [3].

We have previously pioneered the use of related, hierarchical, structural composite systems, using combinations of carbon aerogel and woven carbon fiber with resins. Originally, we developed 3D bi-continuous carbon aerogel matrices for multifunctional composites in structural energy storage applications [41]. However, the carbon system involves high temperature processing which is energy intensive, and tends to degrade both the intrinsic tensile properties of the fibers, and their interface chemistry [42]. The mechanical performance of these systems is currently limited by poor interface compatibility, which leads to poor load transfer and premature fiber buckling in compression.

A new, pure glass system is developed here, combining aligned continuous structural glass fibers with a bi-continuous silica aerogel epoxy matrix. Silica and organically-modified silicate (ormosil) aerogel networks were used to reinforce a structural epoxy matrix to form a three-phase hierarchical GFRP composite (Fig. 2). The silica-based chemistry is compatible with near ambient, low pressure processing and can be readily combined with conventional infusion processing. Compared to the carbonaceous system, the chemistry is also much more suited to developing the required favorable fiber/aerogel/matrix interfacial interactions. An effective interface is essential to prevent premature fiber debonding leading to the onset micro-buckling, without

exploiting the shear support provided by the aerogel network. After developing a new double infusion protocol, the interfacial shear strength and compressive properties of the resulting epoxy-filled bicontinuous aerogel matrix GFRPs were determined. Full experimental details for all material preparations and analyses are included in the electronic supplementary information (ESI).

2. Results and discussion

Silica aerogel properties are affected by a number of factors during synthesis, including the precursor, sol gel medium, choice of catalyst, catalyst concentration, the time for hydrolysis (time between acid-to-base addition), pH, temperature, aging time, surface modification (from hydrophilic to hydrophobic), and drying temperature/methodology [43]. To produce a silica aerogel network at a suitable scale and simplicity for infusion into (aligned) woven glass fabrics, there are a number of constraints: the process should operate at ambient temperatures and pressures, avoiding the use of supercritical drying [44], and with a sufficient handling time for low viscosity liquid infusion techniques. Porosity is required to back-fill with polymer resin, but a higher density provides more reinforcement and a more robust aerogel suitable for handling. The silica aerogel monoliths were produced from a tetraethyl orthosilicate (TEOS) precursor, and hydrolysis times and catalyst concentrations varied to optimise the structure. Short gelation times, Fig. 3, increased the surface area, and reduced the pore widths and primary particle diameters (from ca. 80 nm to ca. 25 nm), with the resultant silica aerogel monolith becoming transparent but more fragile. A relatively dense silica aerogel (skeletal density $2.05\ \text{g cm}^{-3}$, synthesized with 30 min hydrolysis duration using diluted hydrochloric acid (aq) at 1.55 mM concentration), was chosen for the glass fiber infusion experiments, as it had large pore width (12.4 nm) and was relatively robust to handling. The experimental conditions, micrographs and tabulated surface area properties for the silica aerogel monoliths can be found in the electronic supplementary information (ESI).

Polydimethylsiloxane (PDMS) was added to the silica system, to form an ormosil aerogel, as it has been shown to have improved compressive properties as a monolith [45,46] and reduced shrinkage. The addition of an organic polymer, like PDMS, in the sol-gel promotes a homogeneous product [47]. Adding PDMS initially increased gel times ($>25\ \text{h}$), but with higher acidic catalyst concentration, the gelation time reduced to a convenient 30 min. Altering the proportion of PDMS, between 15 wt % to 25 wt % relative to the TEOS, did not significantly alter the ormosil

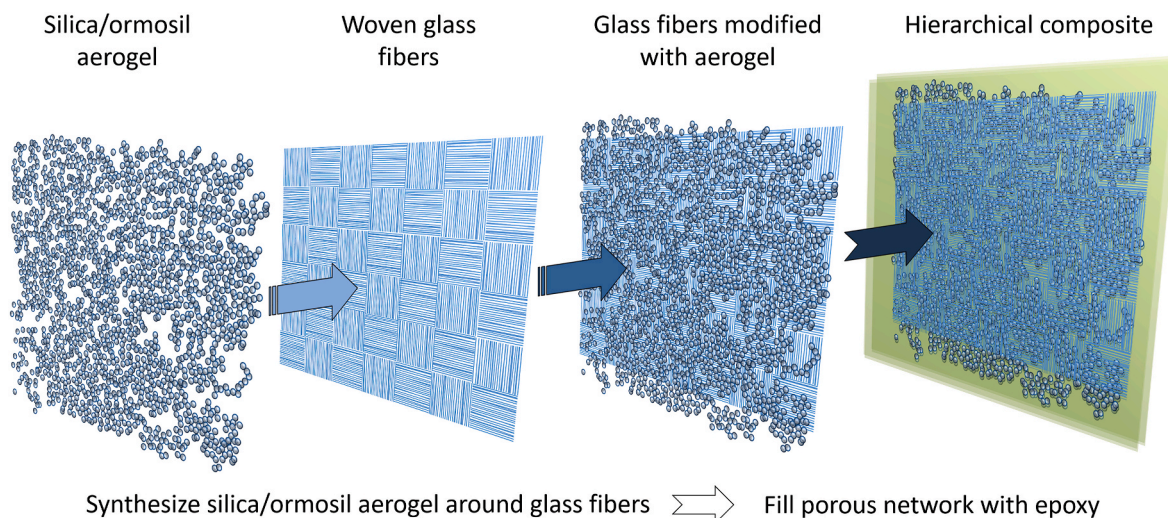


Fig. 2. Schematic of sample manufacture; Silica/ormosil based aerogel precursors are infused via resin infusion flexible tooling (RIFT) and gelled *in-situ* within a woven glass fabric. The woven glass fiber modified aerogel plies are then dried and infused with epoxy, once more through RIFT, to produce a hierarchical composite structure (not to scale). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

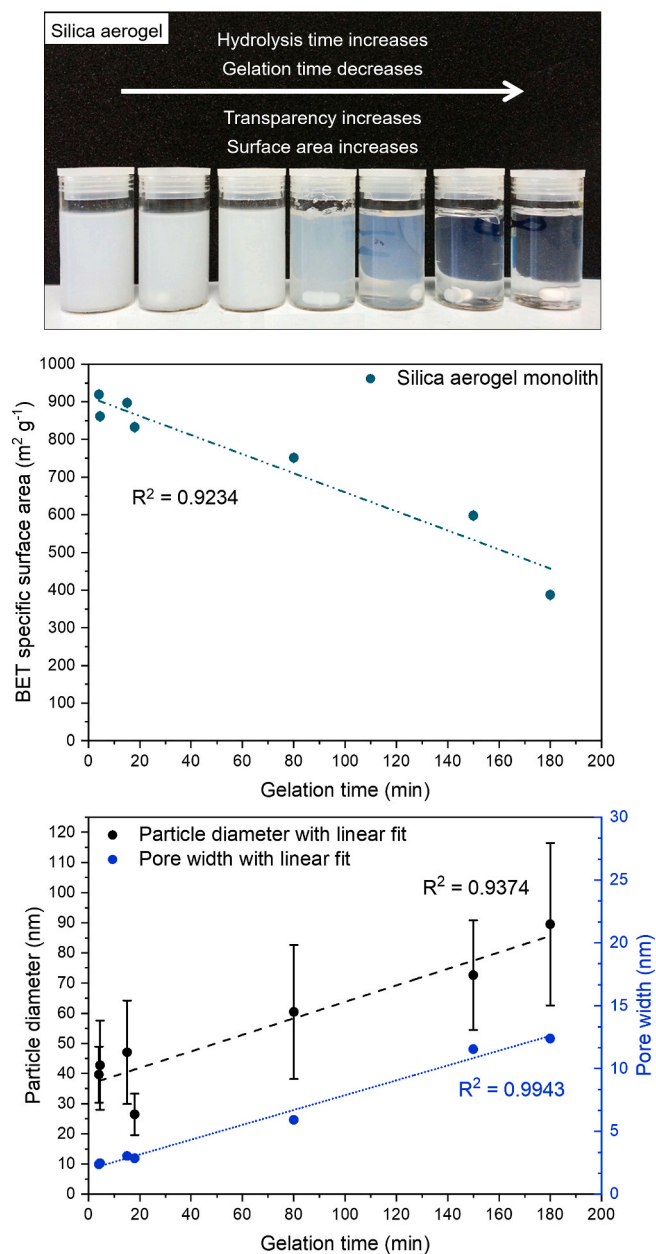


Fig. 3. Top: A photograph illustrating the effect of altering gelation time on the transparency of synthesized silica aerogel monoliths. Middle: Silica aerogel monolith BET (Brunauer, Emmett and Teller) specific surface area against gelation time. Bottom: Silica aerogel monolith pore width and particle diameter against gelation time. Error bars indicate standard deviation from the mean. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

aerogel product; the morphology, surface area (ca. 835 m² g⁻¹ to 815 m² g⁻¹) and porosity (diameter ca. 30 nm ± 7 nm) remained similar. Hierarchical composites were prepared with 20 wt % PDMS ormosils.

Woven continuous glass fibers (ca. 10 μm in diameter, 0.1 m² g⁻¹ surface area, shown in Fig. 4 (A) left and (B) left) were successfully embedded in a matrix network of either, nano-scale silica or ormosil aerogel (Fig. 4 (A) and (B) center and right, respectively). The infusion of the silica or ormosil aerogel precursor around the glass fabric was achieved by adapting a conventional flexible tooling method (see ESI); aging was carried out at an ambient temperature and pressure which produced aerogels with primary particle diameters 63 ± 22 nm, and 41 ± 12 nm for silica aerogel and ormosil aerogels synthesized in the

presence of the fibers, respectively, with standard deviation given n ≥ 250 for each. As expected, there was a dramatic increase in specific surface area for the glass fibers modified with silica aerogel and ormosil aerogel to 12.7 m² g⁻¹ and 54.0 m² g⁻¹, respectively. Using the current aerogel formulations there was an increased loading of ormosil aerogel (9.6 wt %) compared to silica aerogel (4.4 wt %) on the glass fabric, however the skeletal density for the ormosil modified glass fabric was around 12% lower than the as-received glass fabric and the silica modified glass fabric. Specific surface area isotherms and other modified glass fiber properties can be found in the ESI.

Once aged, washed, and dried, the glass fiber-aerogel preforms were infused with epoxy, again through the resin infusion under flexible tooling (RIFT) method, to create dense ternary hybrid GFRP composites, featuring glass fiber reinforcements embedded in a bi-continuous aerogel epoxy matrix (Fig. 4 (C)). Further process details can be found in the Experimental Section in the ESI. The compression strength, moduli and interlaminar shear strengths (ILSS) for GFRP, GFRP modified with silica aerogel, and GFRP modified with ormosil aerogel epoxy composites (with densities ~ 1.81 g cm⁻³) were determined using standard methods (Fig. 5 (A) and (B), with tabulated data contained in the ESI). Improved compressive properties were observed for both silica and ormosil aerogel modified GFRP composites over the GFRP baseline, with increases in fiber volume normalized compression strength of 27% and 15%, respectively, and enhancements in fiber volume normalized compression modulus of 1.7% and 9.7%, respectively. Matrix shear modulus is a critical factor in determining long fiber composite compression strength [36]. Both aerogels likely delay the onset of micro-buckling, the primary failure mode for many fiber reinforced composites in compression, by providing local lateral matrix stiffening and mechanical interlocking [48]. At the same time, the aerogel network reinforcements contribute to enhanced fiber/matrix interfacial properties [49], including improving resistance to delamination, as inferred from an increase in ILSS of 27% for silica aerogel modified GFRP as compared to that of the GFRP baseline (Fig. 5 (B)). The ormosil aerogel-modified GFRP had a slight increase in ILSS when compared to the GFRP baseline, but it was lower (ca. 10%) when compared to the silica aerogel-modified GFRP. The absolute aerogel fraction for the silica aerogel-modified GFRP was only 1.7 vol %, which was lower than the corresponding ormosil aerogel-modified GFRP content (6.5 vol % aerogel). This greater efficacy of the silica system can be attributed to a higher anticipated stiffness associated with the pure inorganic composition and larger characteristic lengthscale.

3. Conclusion

In summary, nanostructured silica-based aerogels can reinforce the matrix of aligned continuous micro-scale glass fiber reinforced epoxy composites, forming a three-phase hierarchical structure. The silica aerogel chemistry is readily compatible with the silicate fibers, forming a strong interface as evidenced by the improved interlaminar properties (up to 27%). The bi-continuous aerogel reinforcements provided efficient load transfer and local stiffening and lateral support to the fibers, likely delaying micro-buckling in compression, improving the compressive strength (up to 27%) without a loss in stiffness. This hierarchical approach provides improved mechanical performance with a minimal increase in composite density (~1.3%). The formation of a continuous rigid reinforcing network, is much more effective than dispersed particulates, whilst the nanoscale structure avoids unwanted stress concentrations.

The use of silica or ormosil aerogel, a material which is traditionally viewed as weak and brittle, as a composite matrix reinforcement expands its already wide range of applications. Silica or ormosil aerogels are already mass-produced, affordably; by using resin infusion flexible tooling infusion techniques that are widely used in the composite sector, the new hierarchical composites can potentially be applied on a commercial scale. A low temperature and pressure processing route is less

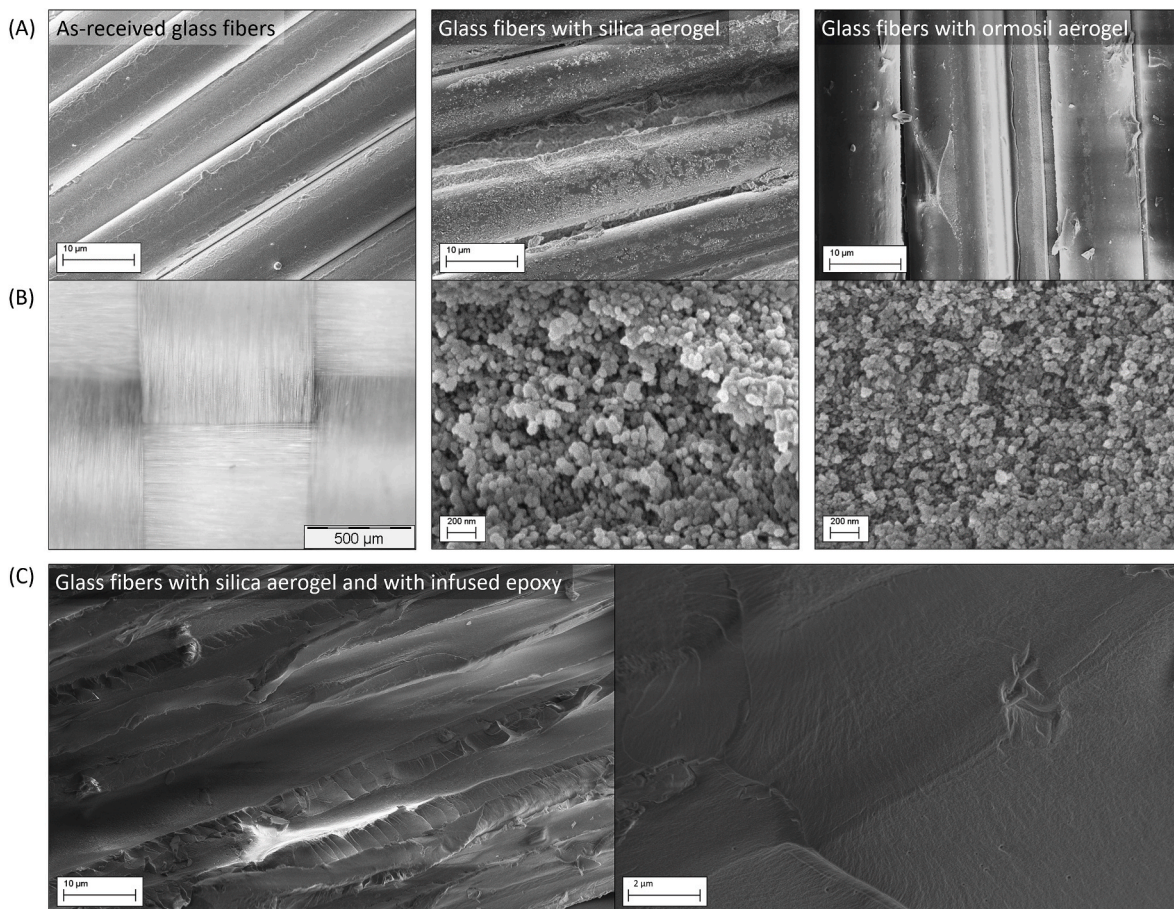


Fig. 4. (A) Left to right: Scanning electron microscopy (SEM) micrographs of as-received glass fibers, glass fibers modified with silica aerogel, and glass fibers modified with ormosil. (B) Left to right: Optical microscopy image of as received glass fiber weave, SEM micrographs of silica aerogel rich region, and ormosil aerogel rich region. (C) SEM micrographs of glass fibers infused with a bicontinuous silica aerogel epoxy matrix (left) and a higher magnification image of the bi-continuous matrix region (right).

energy intensive than the analogous carbon system and circumvents the cost and damage associated with high carbonization temperatures. There is considerable scope for developing the glass fiber-modified silica aerogel composite system through optimizing the density and fiber volume fraction, adjustment of the chemical interface (for example, using alternative organically-modified silicates) and further selection of the aerogel precursors. The use of a hybrid carbon fiber-modified silica/ormosil aerogel-based system might also be attractive, combining the specific properties of carbon fibers with easily tailored silica/ormosil aerogel. The development of rigid bi-continuous reinforced composite architectures, to improve load transfer compared to using particulate stiffening modifiers, may mark the advent of a new generation of hierarchical continuous structural fiber composite systems.

Electronic supplementary information (ESI)

Electronic Supplementary Information is provided for this manuscript. It includes full experimental methods including the material preparation and analysis. In addition, it contains micrographs and tabulated surface area properties for the silica aerogel monoliths.

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Ductile Composite Technology (HiPerDuCT) in collaboration with the University of Bristol, EP/P007465/1 Beyond structural - Multifunctional composites that store electrical energy, and EPSRC programme Grant EP/T011653/1 Next Generation Fibre-Reinforced Composites: a Full Scale Redesign for Compression (NextCOMP) in collaboration with the University of Bristol. All underlying data to support the conclusions are provided within the manuscript or ESI. For the purpose of open access, the author has applied a Creative Commons Attribution (CC BY) license to any Author Accepted Manuscript version arising.

CRediT authorship contribution statement

David B. Anthony: Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization, Supervision. **Sang N. Nguyen:** Formal analysis, Writing – original draft, Writing – review & editing, Supervision. **Hui Qian:** Conceptualization, Methodology, Validation, Investigation, Supervision. **Shi Xu:** Methodology, Validation, Formal analysis, Investigation. **Charles M.D. Shaw:** Visualization, Investigation, Writing – review & editing. **Emile S. Greenhalgh:** Conceptualization, Writing – review & editing, Funding acquisition. **Alexander Bismarck:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition. **Milo S.P. Shaffer:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition.

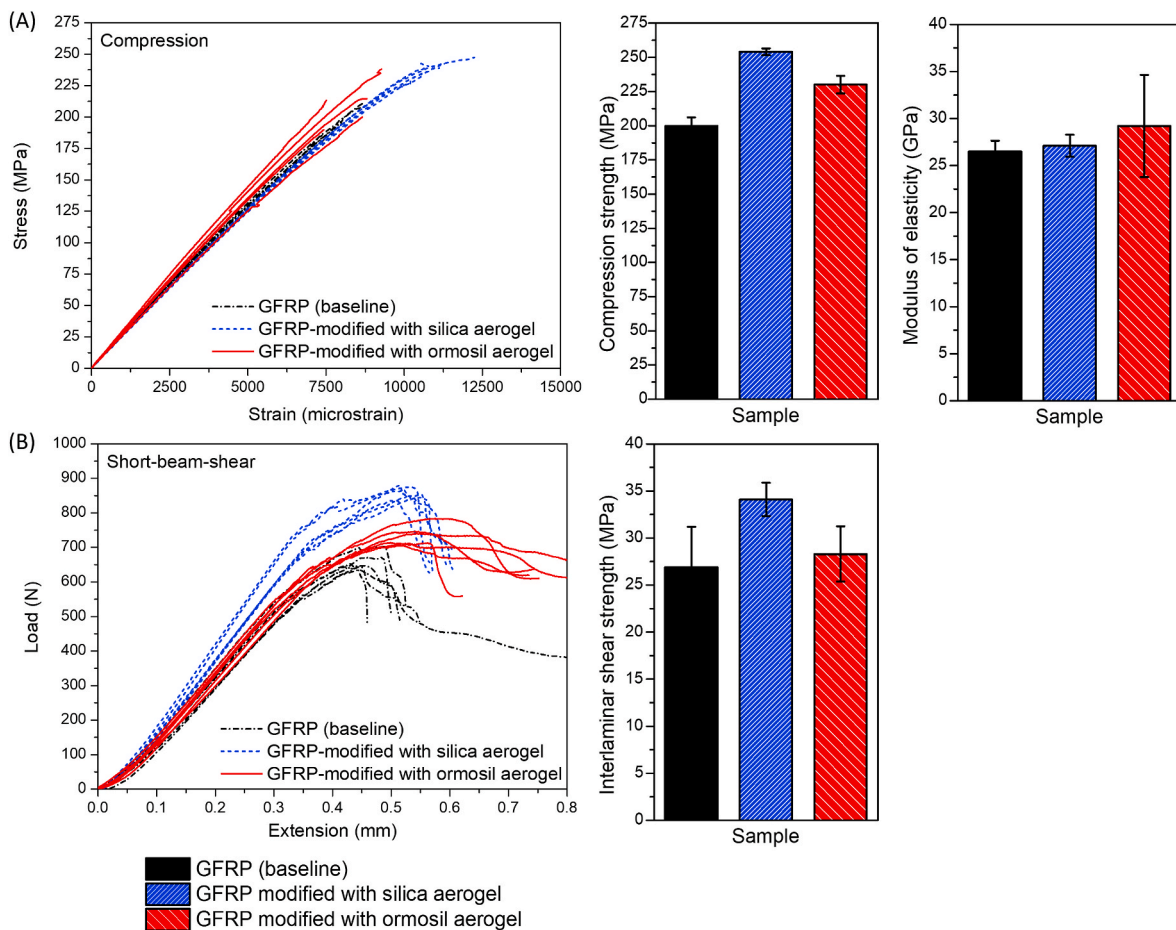


Fig. 5. Epoxy composites reinforced with glass fibers (GFRP), glass-fibers modified with silica aerogel, and glass fibers modified with ormosil aerogel; (A) Left: Representative compression stress-strain curves, Centre: Compression strength, Right: Compression modulus of elasticity, both normalized to 50% fiber volume fraction. (B) Left: Representative short beam-shear load extension responses, Right: Interlaminar shear strength. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.coco.2023.101531>.

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