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High volume-fraction silk fabric reinforcements can improve the key mechanical properties of epoxy resin composites

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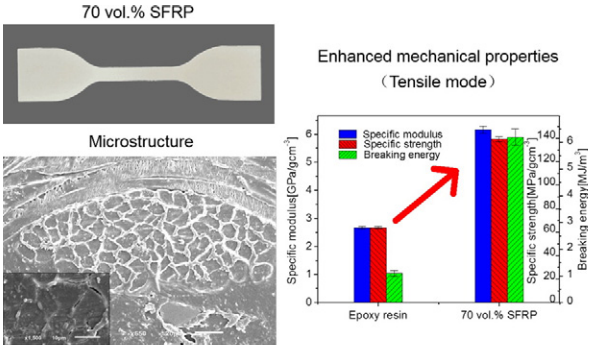
HIGHLIGHTS

- Environmentally friendly silk fibres were used to reinforce a widely used commercial epoxy resin.
- The effect of volume fraction of the reinforcement silk on the mechanical properties of the composite was studied.
- A highest volume fraction of the silk reinforcement in this work was 70 vol.%.

GRAPHICAL ABSTRACT



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ABSTRACT

Silk fabric reinforced epoxy composites (SFRPs) were prepared by simple hot-press and vacuum treatment, to achieve a maximum reinforcement fraction of 70 vol.%-silk. Mechanical behaviour, specifically tensile, flexural, interlaminar shear, impact, dynamic and thermal properties of the SFRPs, was investigated. It was shown that reinforcement by silk fabric can greatly enhance the mechanical performance of SFRPs. In particular, the tensile modulus and breaking energy of 70 vol.%-silk SFRP were 145% and 467% higher than the pristine epoxy resin. Moreover, the flexural modulus, ultimate strength and breaking energy were also markedly increased for SFRPs. The flexural strength increased linearly with increasing silk volume fraction from 30 to 60 vol.% but diminished slightly at 70 vol.%. Additionally, interlaminar shear results showed that the silk and the matrix epoxy resin had better adhesion properties than plain woven flax fibre. Of most significance is that the impact strength reached a maximum of ~71 kJ m^{-2} for the 60 vol.%-silk SFRP, which demonstrates the potential of silk reinforcements in impact-resistant composites for applications such as wind turbine blades. Our study may shed light on improving the strength and toughness of engineering composites by incorporating high volume fractions of natural fibres.

1. Introduction

Natural fibre reinforced composites have recently been the subject of extensive research [1–3], propelled firstly by their potential

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applications due to their low density, high specific mechanical properties and promising biodegradability and secondly by the environmental and social demands for a sustainable world. Silk fibre reinforced epoxy materials embody one such class of composites. In this regard, compared with the flax-represented plant fibres, the best-known fashion silk from silkworm *Bombyx mori* (*B. mori*) represents the only natural fibre which can be used as a continuous filament of fibrous protein. Despite this, silk reeled from cocoons and fed to the textile industry in the silk commercial value chain is rarely utilized for practical engineering. However, this strong and tough protein fibre may endow modern engineering composites with unprecedented mechanical properties, as has been suggested in several recent studies on silk-composites [1,3].

Shah et al. [4], for example, prepared nonwoven (from natural cocoon walls) and woven (from silk fabric) silk fibre reinforced plastics (SFRPs) via vacuum-driven resin transfer moulding. The fibre volume fraction and density of the SFRPs were, respectively, 36.2% and 1.20 g cm^{-3} for the nonwoven and 45.2% and 1.22 g cm^{-3} for the biaxial-woven. These SFRPs displayed much greater tensile and flexural specific strengths (~90 MPa/g cm^{-3} and ~13 MPa$^{1/2}$/g cm^{-3}), impact strength (115 kJ m^{-2}) and inter-laminar shear strength (~92 kJ m^{-2}/g cm^{-3}) than those of flax fibre reinforced epoxy resin plastics; indeed, they are comparable, although not necessarily superior, to those of glass fibre reinforced epoxy resin composites. In particular, the plain woven SFRPs demonstrated the high ductility (e.g., tensile fracture strain of 7%) of silk-fibre composites.

With respect to biodegradable-polymer based silk composites, Ho et al. [5] manufactured a silk fibre reinforced biodegradable plastic poly(lactic acid) (PLA) with 5 wt.% silk by small scale injection moulding; this material had tensile and flexural moduli that were, respectively, 27% and 2% higher than pure PLA. Similarly, Zhao et al. [6] showed with as little as 5 wt.% incorporation of silk the dynamic mechanical, thermal and biodegradable properties of silk/PLA biocomposites could be improved markedly. Shubhra et al. [7] studied the mechanical and degradation characteristic of silk fibre reinforced gelatine composites, and measured tensile strengths, tensile modulus, bending strengths, bending modulus and impact strengths that were, respectively, ~260%, 400%, 320%, 450% and 260% higher than the unreinforced matrix material. A series of novel silk fibroin fibre/poly(ε-caprolactone) (PCL) biocomposites with different silk contents, prepared by Li et al. [8] showed the highest strength with 35 to 45 wt.% silk contents; the 35% silk composite had the highest tensile strength whereas the 45% silk composite showed the best flexural strength. These results provide evidence of the important role of silk fibres as a reinforcement phase to improve the mechanical properties of PCL. In similar vein, an all-silk composite made of silk fibre embedded in a reconstituted silk fibroin matrix with various weight fractions of silk fibre, prepared by Yuan et al. [9], showed the best tensile strength (151 ± 5 MPa) and breaking strain (27.1 ± 1.4%) with 25 wt.% silk fibre reinforcement.

The properties including physical, tensile mechanical and flexural mechanical properties, from the abovementioned references are consolidated and compared with those of typical flax fibre reinforced epoxy resin (thermosets) and glass fibre reinforced epoxy resin (thermosets) in Table 1.

Shah et al. [1] also examined the through-thickness compaction behaviour of three fibre reinforcements, namely, silk, plant fibre and glass fibre, and under the same compaction pressure biaxial silk fabric was more compressible than flax plant fibre and glass fibre. This indicates that silk fabric can reinforce fibre composites that require higher volume fractions, a problem which until now has been a “show stopper” for plant fibre reinforcements.

Of all the thermosetting resins, epoxy resin is one of the most widely used owing to its many credentials such as easy processibility, low cost, reasonable mechanical properties, good adhesive performance, good chemical resistance and great high-temperature tolerance [10–13]. Epoxy resin-based composites have been used extensively in electronics, aerospace and civil engineering [14–17]. However, due to the high degree of cross-linking, many products of epoxy resin are intrinsically very brittle, and the impact strength is low (shown as a catastrophic failure). This weakness in the mechanical performance has compromised the use of epoxy resin for many structural applications. As a result, modifications of the epoxy resin products for enhanced mechanical properties, especially impact strength and toughness, have become an important area for research [18–20], with the use of natural fibre reinforcements to improve their mechanical performance proving a popular approach [21–24].

Table 1

Summary of the properties of the silk fibre reinforced composites (SFRPs), as compared to flax fibre reinforced epoxy resin composites (FFRPs) and glass fibre reinforced epoxy resin composites (GFRPs). Interlaminar shear strength is shortened as ILSS in the unit of MPa.

Composite	Physical properties		Tensile mechanical properties			Flexural mechanical properties			ILSS (MPa)	Impact strength (kJ·m^{-2})
	Fibre volume fraction (%)	Density (g·cm^{-3})	Stiffness (GPa)	Ultimate strength (MPa)	Ultimate strain (%)	Stiffness (GPa)	Ultimate strength (MPa)	Ultimate strain (%)		
Nonwoven silk–epoxy [4]	36.2	1.20	5.4 ± 0.2	60 ± 5	1.3 ± 0.1	5.2 ± 0.2	143 ± 10	3.4 ± 0.4	31.0 ± 3.7	16 ± 1
Plain woven silk–epoxy [4]	45.2	1.22	6.5 ± 0.1	111 ± 2	5.2 ± 0.2	6.4 ± 0.4	250 ± 4	6.9 ± 0.2	42.6 ± 5.9	115 ± 7
Nonwoven flax–epoxy [4,22]	15–35	1.20–1.26	5.8–9.8	37–75	0.8–1.6	4.8–6.7	55–91	2.1–3.2	13.6–26.7	8–15
Plain woven flax–epoxy [4,22]	30–55	1.24–1.32	7.3–11.2	63–89	1.5–2.9	2.1–10.1	57–195	3.3–4.9	9.7–23.3	23–36
Nonwoven glass–epoxy [4,22]	15–45	1.36–1.80	10.2–16.7	123–241	1.0–2.1	9.0–11.4	192–325	3.0–4.0	25.0–35.0	73–107
Plain woven glass–epoxy [4,22]	30–65	1.58–2.09	17.0–24.0	350–500	2.1–2.5	13.2–22.0	370–560	3.5–4.0	38.0–52.0	165–280
Silk-PLA [5]	5	–	4.08 ± 0.05	70.6 ± 1.1	3.8 ± 0.5	4.06 ± 0.2	97.4 ± 21.8	2.9 ± 0.9	–	–
10% silk fibre/fibroin [9]	10	–	3.1 ± 0.2	83 ± 7	11.2 ± 1.3	–	–	–	–	–
20% silk fibre/fibroin [9]	20	–	3.0 ± 0.2	142 ± 7	23.5 ± 17	–	–	–	–	–
25% silk fibre/fibroin [9]	25	–	2.8 ± 0.1	151 ± 5	27.1 ± 1.4	–	–	–	–	–
Silk fibre/gelatin [7]	20	–	0.65	44.5	8.2	3.7	63	–	–	5.1
Silk fibre/PCL [8]	35	–	–	26.5	12.0	1.8	49	–	–	–
Silk fibre/PCL [8]	45	–	–	22.5	5.0	2.1	59.5	–	–	–