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Opportunities for silk textiles in reinforced biocomposites: Studying through-thickness compaction behaviour

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ABSTRACT

While it is common knowledge in natural fibre composites manufacture that plant fibre reinforcements are considerably less compactable than synthetic fibre reinforcements, the through-thickness compaction behaviour of animal-fibre silk reinforcements has not been characterised thus far. We find that not only are silk reinforcements significantly more compressible than plant fibre reinforcements, but their compactibility exceeds that of even glass fibre textiles. For instance, the fibre volume fraction (at a compaction pressure of 2.0 bar) of woven biaxial fabrics of silk, plant fibres and E-glass are 54–57%, 30–40% and 49–54%, respectively. Therefore, silks provide an opportunity to manufacture high fibre content natural fibre composites; this is a bottleneck of plant fibre textiles. Analysing the structure of silk textiles through scanning electron microscopy, we show that favourable fibre/yarn/fabric geometry, high degree of fibre alignment and dispersion, and suitable technical fibre properties enable optimal packing and arrangement of silk textiles.

1. Introduction

1.1. Liquid composite moulding

Liquid composite moulding (LCM) processes, such as vacuum infusion (VI) and resin transfer moulding (RTM), are widely used for the cost-effective production of high-performance complex-geometry large components at low-to-medium volumes (e.g. 100–10,000 parts/year) [1]. While numerous acronyms and associated process variations exist [2,3], the basic approach in any LCM process, as illustrated in Fig. 1, is to force a catalyzed thermosetting liquid resin to flow through a stationary dry, and often compacted reinforcement inside a closed mould, by creating a pressure differential (using vacuum and/or injection pressure) between the inlets and outlets.

In general, LCM processes have four stages (Fig. 1): (i) reinforcement lay-up, (ii) mould filling, (iii) post-filling, and (iv) demoulding. In particular, successful implementation of LCM processes involves understanding and optimising the mould filling stage. This stage is affected by numerous factors, including mould/part geometry; (inlet and outlet) gate location and configuration; reinforcement lay-up, orientation, compaction, and permeability; resin temperature, viscosity, and degree of cure (all of which are a function of time); pressure differential in the mould cavity; and tooling temperature. Not surprisingly, computational mould-filling simulations are widely used as a cost-effective and time-saving tool to optimise the LCM process [3]. However, accurate manufacturing process simulations require accurate data, including compaction data.

The through-thickness compaction of a reinforcement directly affects the reinforcement permeability and part fill time in the mould filling process [4]. Importantly, it also dictates the thickness and volumetric composition (i.e. fibre volume fraction) of the final part. Tight control of part thickness (and therefore weight) is a requisite for quality assurance in any composite manufacturing process. In addition, in their uncompressed state, textile reinforcements have a low fibre volume fraction (typically between 10–25% [4]). This must be increased (to up to 70%) during processing to exploit the mechanical properties of the reinforcement. Studying the relationship between compaction pressure P and fibre volume fraction v_f for a given preform also enables determining the maximum (theoretical) fibre volume fraction, which sets the upper limit of reinforcement efficiency. Consequently, compaction plays an important role in not only LCM processes but also in the stamping of textile-reinforced thermoplastic composites. Knowledge of the compaction behaviour of the reinforcement form is therefore critical.

Typically, empirical power-law relationships are used to model compaction [4,5]. However, the compaction response of a reinforcement is governed by various deformation mechanisms (depicted in Fig. 2) and hence is complex and depends on various elements, such

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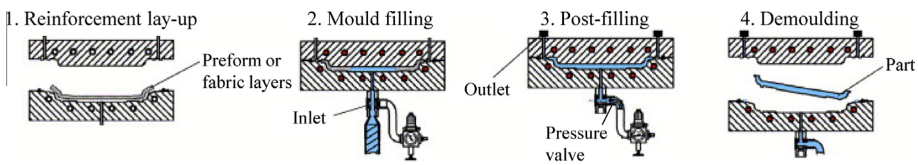

Fig. 1. Schematic of a liquid composite moulding (LCM) process, illustrating the four principal steps: (1) lay-up of the dry reinforcement (either as multiples layers of fabrics or as a prefabricated 'preform') in a mould with a rigid (metal) or semi-rigid (composite) bottom tool and a rigid (metal), semi-rigid (composite) or flexible (silicone or vacuum bag) top tool/surface; (2) compaction of the reinforcement followed by resin impregnation via drawing vacuum and/or injecting resin under pressure; (3) Removal of pressure to allow laminate thickness to equilibrate in cavity followed by curing of resin; and (4) de-moulding of the cured and stiff composite part. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

as: type and form of fibre reinforcement, fibre architecture, number of layers in the preform, preform stacking sequence, history of loading, rate of compaction, tooling temperature, and presence of lubricant (i.e. wet versus dry state) [4–9].

1.2. Natural fibre composites in LCM processes

The increasing consideration of natural fibres as next-generation sustainable composite reinforcements requires tackling the first hurdle which is composite manufacture (reviewed in [10–14]). Due to the commercial applications of natural fibre reinforced composites in principally small-part high-volume low-cycletime markets (e.g. the automotive industry), compression moulding is the widely used manufacturing technique [12]. However, LCM processes are specifically well-suited to natural fibre reinforcements for a variety of reasons [6,12,15], including:

(i) low processing temperatures (often < 120 °C, if not ambient) avoiding thermal degradation of the fibres during composite fabrication,
(ii) minimal fibre damage during composite processing (as opposed to injection/extrusion moulding) allowing retention of high reinforcement properties and efficiencies (i.e. length and orientation),
(iii) use of liquid resins with typically low viscosities (0.1–1 Pas) to allow good preform impregnation with low composite void content even at low compaction/injection pressures,
(iv) use of thermosetting resins with typically polar functional groups, which form a better interface with typically polar natural fibres (than polyolefin-based thermoplastics),
(v) relatively low-cost (and often unsophisticated) tooling, making the process compatible with low-cost plant fibres, particularly when manufacturing in developing countries with an abundance of indigenous natural fibres, and
(vi) LCM processes are close-moulding 'clean' processes which provide worker-friendly conditions.

Not surprisingly, researchers are increasingly investigating different aspects of the LCM process for natural fibre reinforced composites, particularly reinforcement permeability [16–21] and

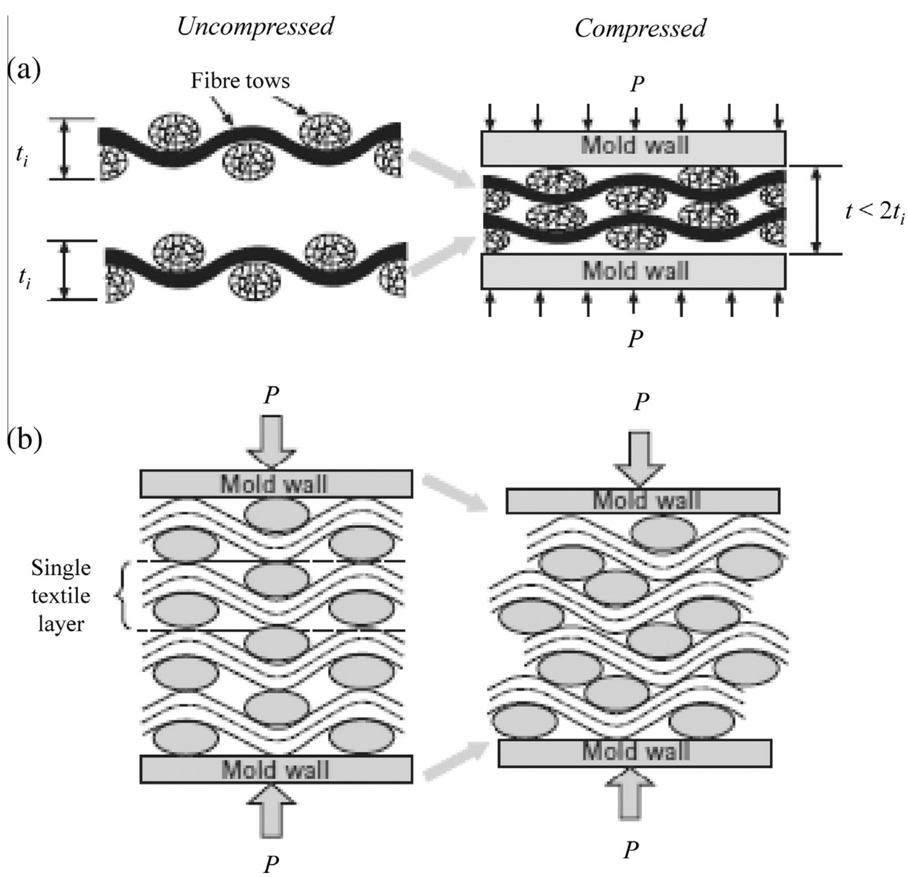

Fig. 2. Key mechanisms that drive reinforcement compaction: (a) yarn cross-section deformation and yarn flattening, and (b) yarn bending deformation, void condensation/reduction, and nesting and packing of layers. Ref. to [42] for detail.

compaction [6,16,18,21–23], and resin flow behaviour [17,18,20,24,25]. In our opinion, there have been two critical findings so far. Firstly, due to the polarity and hollow-structure of natural fibres, during the impregnation stage, the permeability of natural fibre reinforcements is reduced considerably as the fibres absorb polar fluids (including phenolic and vinyl-ester resins) and soften and swell [16,20]. Secondly, the compactibility of natural fibre reinforcements is found to be consistently lower than that of synthetic fibre reinforcements [12,22], principally due to differences in structure of the fibres and their assemblies. For instance, Madsen [22] report that at a constant compaction pressure of 2.2 MPa, unidirectional flax and E-glass preforms have fibre volume fractions of 56% and 71%, while random flax and E-glass preforms have fibre contents of 38% and 52%, respectively. The latter observation, also supported by the works of Goutianos et al. [26] and Shah et al. [15], is a potential bottleneck in the substitution of synthetic fibre composites by natural fibre composites.

Notably, much of the research on natural fibres and their composites is based on fibres derived from plants. Many researchers tend to generalise the outcome of their studies on plant fibres to other natural fibres (for instance [10,27–30]). However, animal fibres like *Bombyx mori* silk are natural fibres that differ significantly in chemical and mechanical properties to plant fibres. Plant fibres, such as flax and hemp, are lignocellulosic in nature and typically exist as technical fibre bundles. Each elementary fibre within the technical fibre bundle is hollow (luminal porosity of 2–16% for bast fibres [12]) and has an irregular polygonal outer cross-section. Elementary fibres have non-uniform width in the range of 10–100 μm and a short length in the range of 4–100 mm [31]. Due to the short fibre length, staple plant fibres are conventionally spun into twisted yarns to produce a continuous product with sufficient strength and handle-ability for the fabrication of woven textiles. On the other hand, protein-based silk fibres are naturally spun through specialised ducts by animals, including spiders and silkworms. Silks, such as those industrially processed from the cocoons of the domesticated *B. mori* silkworm, exist as single continuous filaments/strands of fibroin with lengths of up to 1500 m [31]. The fibroin has an irregular almost-triangular cross-section, with a width in the range of 8–13 μm [31]. As the silk fibre is thin, silk filaments are spun into slivers and rovings to build material thickness for use in woven textiles. The difference in structure of the fibres, their semi-products (i.e. yarns and rovings) and their textiles will have a perceptible effect on their usage as reinforcements for composites.

To our knowledge, the compaction of silk reinforcements, which are also suitable and attractive natural fibre reinforcements for composites [32,33], has so far not been analysed. Using plant fibre textiles as a benchmark, we evaluate the compaction behaviour of silk textiles. Here we examine critical information, such as the maximum fibre volume fraction for a given compaction pressure that would be useful in the manufacture of high quality silk composites processed using LCM processes.

2. Experimental

2.1. Materials

Table 1 lists the different natural fibre textiles used in this work, alongside specifications provided by the supplier. With the exception of Flax UD, which is a non-crimped stitched unidirectional fabric, all other fabrics have balanced plain weave architecture. Optical images of the fabrics used in this study, captured with an Olympus SZ40 microscope equipped with a Canon PC1200 camera, are presented in Fig. 3. To further examine the surface morphology, structure and packing of the fabrics, a JCM-5000 NeoScope (JEOL)

Table 1
Properties of fabrics.

Fibre type	Textile architecture	Areal density κ (g m^{-2})	Manufacturer	Layers stacked N	ID
Flax	Plain weave	450	ETS Eyraud (France)	3	Flax
Hemp	Plain weave	233	ETS Eyraud (France)	6	Hemp
Flax	Stitched unidirectional	325	Composites Evolution Ltd. (UK)	4	Flax UD
Silk	Plain weave	87	Stephen Walters & Sons Ltd. (UK)	16	Silk A
Silk	Plain weave	88	Stephen Walters & Sons Ltd. (UK)	16	Silk B

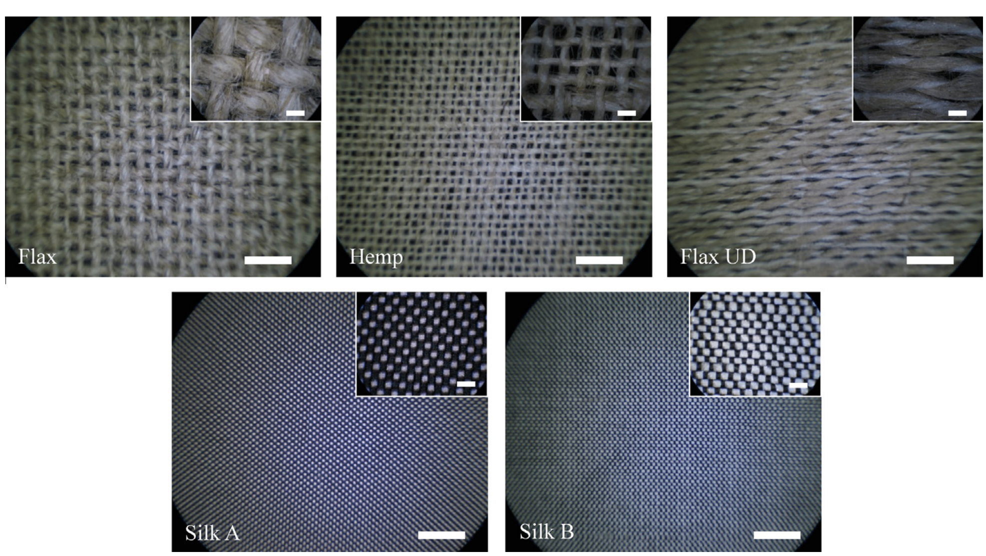

Fig. 3. Optical images of fabrics used for the compaction study. Scale bars are 5 mm for the large images and 1 mm for the zoom images in the top-right corner. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

scanning electron microscope (SEM) was used. Samples, sputter coated with Au/Pd alloy, were viewed at an acceleration voltage of 10 kV.

Fabric areal density κ and yarn linear density are considerably lower in silk textiles than in plant fibre textiles (Table 1). As compaction is dependent on the 'mass' of reinforcement being compressed [9,34], in order to ensure that comparison between different reinforcement types was fair we selected the number of layers N of fabric stacked in such a way as to give a similar reinforcement mass (=$\kappa \cdot N$) in the range of 1300–1400 gm^{-2}. This method is acceptable as Gauvin et al. [34] have demonstrated that the compaction behaviour of a given fabric type is similar for a range of reinforcements whose combination of areal density and fabric layers produce a similar reinforcement mass. The fibre density ρ_f for flax, hemp and silk has been assumed to be 1500, 1500 and 1300 kg m^{-3}, respectively [31].

2.2. Compaction testing

A Zwick/Roell Z0.5 testing machine, equipped with a 500 N load cell, was used to conduct the fabric compaction tests. A circular press area with a d = 0.05 m diameter was used, giving a maximum compaction pressure of ~0.25 MPa (=2.5 bar). The number of layers N of a fabric used is presented in Table 1. For each fabric type, three samples were tested. To determine the initial preform thickness t_0, a pre-load of 1 N was applied. Thereafter, the preform sample was compressed at a constant compaction rate of 5 mm/min. Four repeat cycles were then conducted on the specimen to investigate the effect of multiple compaction cycles on the rearrangement and compactability of the fibre network. Plots of preform thickness t against compaction force F were obtained. Using Eqs. (1) and (2), these curves were then converted into the more conventional plots of fibre volume fraction v_f against compaction pressure P. Using the multiple curve averaging function in OriginPro 9 (linear interpolation over the common compaction load/pressure range), an average curve was obtained for each compaction cycle of a given fabric type

$$v_f = \frac{N\kappa}{\rho_f t} \tag{1}$$

$$P = \frac{4F}{\pi d^2} \tag{2}$$

As observed in Fig. 4, at low compaction pressures (ε < 0.1 bar), the fibre volume fraction increases rapidly (as the preform thickness decreases rapidly). Subsequent increase in compaction pressure pushes the fibre volume fraction and preform thickness to gradually approach an asymptotic maximum and minimum value, respectively. To quantitatively analyse the results, a three-parameter power-law function (Eq. (3)) was fitted to the $v_f - P$ data. Good agreement was found between the regression model and the experimental data, denoted by non-linear least squares regression R^2-values >0.995

$$v_f = v_{f,0} + a \cdot P^b, \quad 0 < b < 1 \tag{3}$$

where $v_{f,0}$ is the initial fibre volume fraction at no compaction pressure, and a and b are fitting parameters related to the 'additional' fibre volume fraction (i.e. $v_f - v_{f,0}$) at P = 1.0 bar, and the compaction stiffening index, respectively. Practically, $v_{f,0}$ is indicative of the uncompressed textile fibre volume fraction, a represents the increase in fibre volume fraction at low compaction pressures, and b denotes the shape of the compaction curve. These three parameters, alongside v_f at an arbitrary value of P = 2.0 bar, were used to characterise the compaction behaviour of a stacked textile reinforcement.

We examined the effect of (i) fibre type (flax, hemp and silk), (ii) textile architecture (plain weave and stitched unidirectional) and (iii) multiple compaction cycles, on preform compaction behaviour

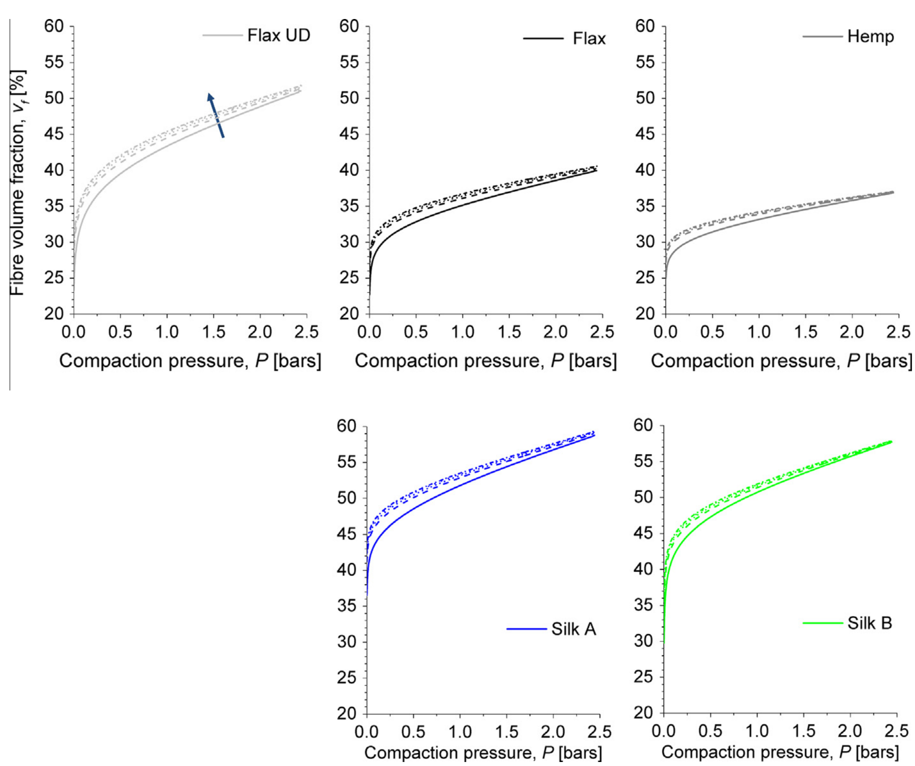

Fig. 4. Compaction behaviour of the five reinforcement types illustrating the effects of fibre type (woven silk textiles vs. woven plant fibre textiles) and textile architecture (woven flax fabric vs. non-crimped unidirectional flax fabric). The arrow indicates the direction in which the compaction curves move for the four successive compaction cycles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

as well as the effect of (iv) number of stacked fabric layers, and (v) compaction rate. For (i)–(iii) tests were conducted on $N = 12$, 16 and 21 layers of Silk B fabrics (at 5 mm/min), while for (iv) and (v) Silk B fabrics (21 layers) were tested at compaction rates of 0.5, 5, and 50 mm/min.

3. Results and discussion

3.1. Effect of fibre type

The compaction behaviour measured for the different reinforcement types is presented in Fig. 4. In particular, we found that woven silk textiles (silk A and B) were significantly more compactable than woven plant fibre textiles (flax and hemp). This suggests that the compaction pressure needed to achieve any fibre volume fraction (within the range studied) is significantly lower in the case of silk reinforcements than for plant fibre reinforcements. In fact, at the same compaction pressure the fibre volume fraction of woven silk textiles was 10–15% (absolute) higher than even unidirectional flax reinforcements. However, as textile architecture has a considerable effect on compaction behaviour, for a fair analysis woven silk textiles will be compared against woven plant fibre textiles in this section. Besides their different vertical positions, the shape of the compaction curves is also different, with woven silk textiles exhibiting a steeper increase in fibre volume fraction with increasing compaction pressure than woven plant fibre textiles.

For quantitative comparison, four parameters (namely, $v_{f,0}$, a, b and v_f at $P = 2.0$ bar) were extracted by fitting the power-law function in Eq. (3) to the experimental data. The determined values are presented in Fig. 5. Firstly, we found that the first-cycle uncompressed textile fibre volume fraction $v_{f,0}$ for silks (33 to 40%) was much higher than that of plant fibre textiles (23–27%). Secondly, the larger (if not comparable) a-values indicate that woven silk textiles exhibited a larger increase in fibre volume fraction at low compaction pressures, than woven plant fibre textiles. Finally, v_f at $P = 2.0$ bar was in the range of 55–58% for silk textiles, but only 35–40% for plant fibre textiles. Notably, the difference in v_f at $P = 2.0$ bar between silk and plant fibre fabrics (15–23%) was larger than the difference in $v_{f,0}$ (6–17%), implying that the structure of silk textiles enables them to capitalise on their high $v_{f,0}$ with increasing compaction pressure. A deeper analysis of the structure and packing of the woven fabrics would allow us to potential sources of this observation.

3.1.1. Structure of the woven textiles

SEM micrographs of the four woven textiles are presented in Fig. 6. While the general plain weave structure was observed in all four fabrics, there were also differences which are likely to account for the differences in fabric compaction behaviour.

Firstly, although silk textiles have a lower areal density than plant fibre textiles (Table 1), the silk textiles are more tightly woven with a lower inter-yarn porosity (Fig. 6(a)–(d)). This would impart high packing density, particularly at low compaction forces, and therefore explain the high values of $v_{f,0}$ and a for silk textiles.

The cross-sectional shape of the yarns in the plant fibre textiles and silk textiles is also different (Fig. 6(a)–(d)). While plant fibre yarns have a circular cross-section, the width-to-thickness ratio in the silk rovings is ~5. The lenticular cross-section shape of silk rovings would enable higher 'inter-yarn' compaction than twisted plant fibre yarns. Moreover, the irregular almost-triangular cross-section shape of single silk fibres [31], observed in Fig. 6(e) and (f), also permit higher 'intra-yarn' packing densities than irregular polygonal cross-section plant fibres [31]. Fig. 6(e) and (f) specifically show how bundles of silk fibres align and pack in an uncompressed roving. Notably, the concave and convex cross-sections of silk fibres would enable them to 'slot' into each other.

In addition, fibre alignment is likely to affect the compactibility of a preform [8,22]. In terms of orientation, the arrangement of yarns in the fabrics is clearly more ordered and uniform in the silk textiles (Fig. 6(a)–(d)). Studies on the compaction of both E-glass and plant fibre reinforcements have reported that rovings are significantly more compactable than twisted yarns [8,22]. This is because yarn twist induces fibre misorientation and transverse pressure, both of which are detrimental to effective packing [35]. Note that twist is a 3D phenomenon and that yarn twist (and yarn

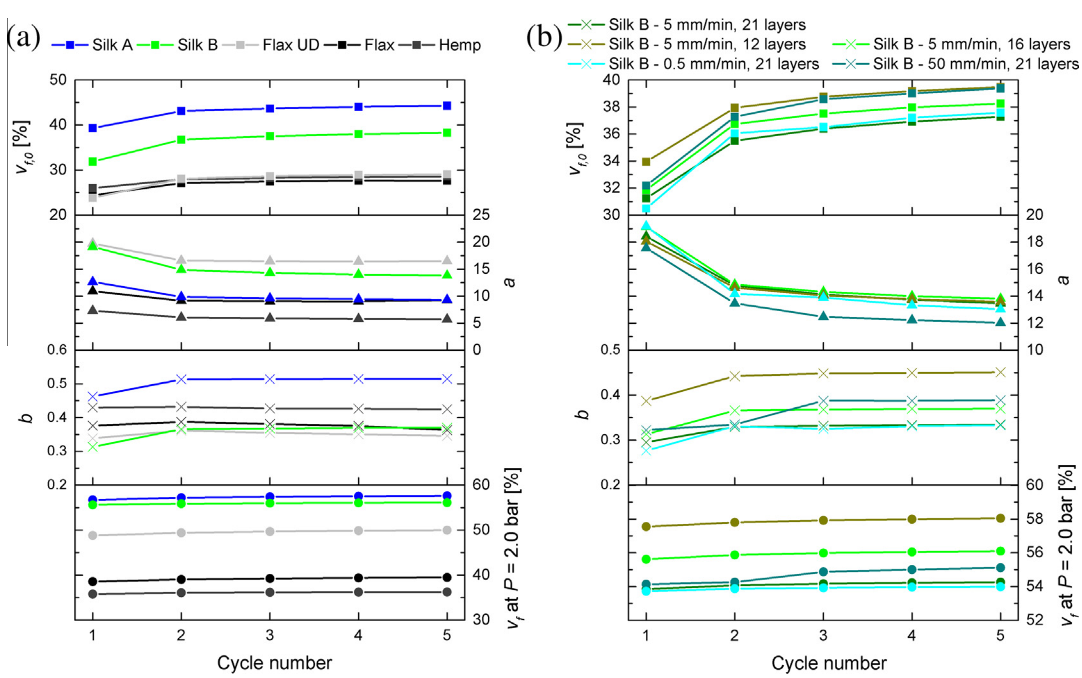

Fig. 5. Comparison of parameters which characterise the compaction response of (a) various natural fibre reinforcements, and (b) woven silk textiles for different number of stacked layers and compaction speeds, for multiple compaction cycles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

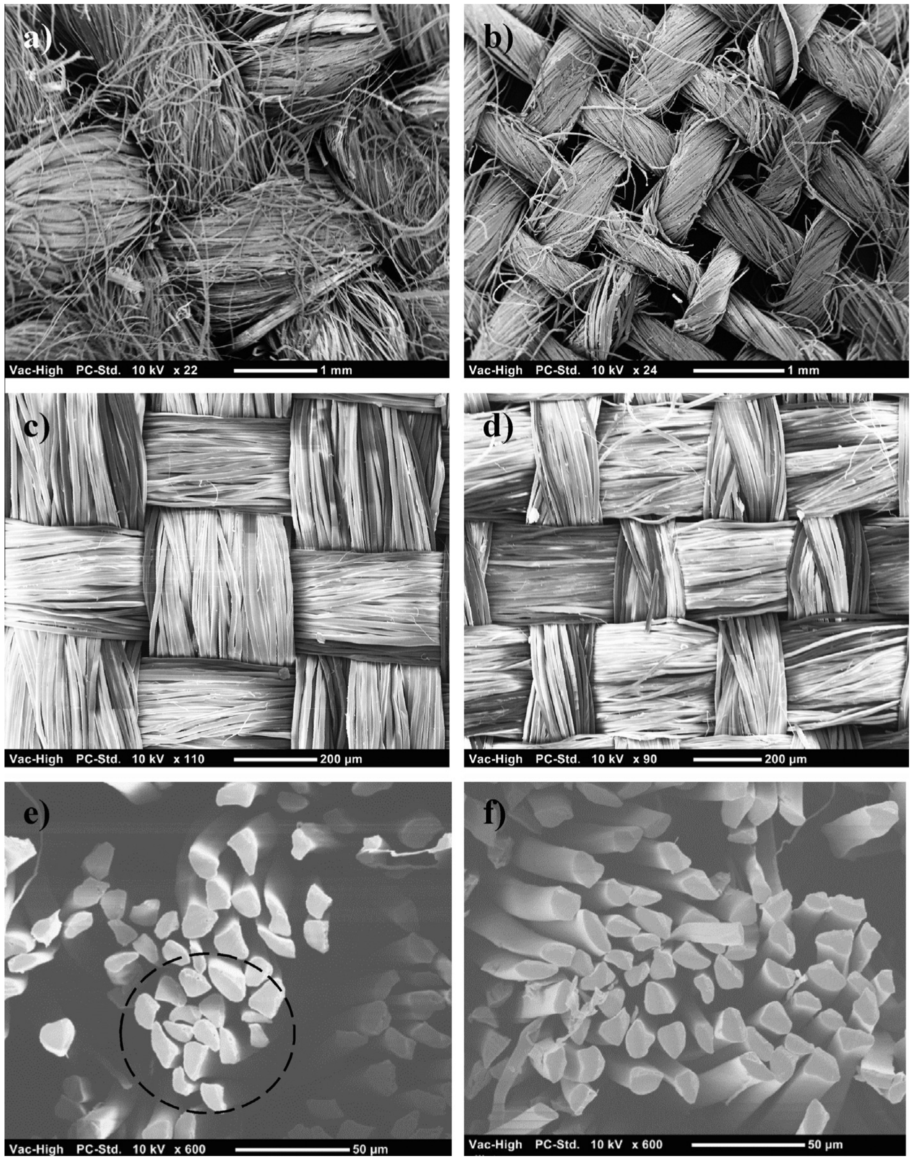

Fig. 6. SEM images of the plain weave textiles: (a) flax, (b) hemp, (c) silk A, and (d) silk B. SEM images of cross-sections of silk rovings and fibres in (e) and (f).

packing fraction) are a function of yarn radius; the twist angle and packing fraction are highest and lowest, respectively, at the yarn surface [35]. As observed in Fig. 6(a)–(d), while the plant fibre textiles employ yarns that are twisted (surface twist angle of 20–30°), the silk textiles employ rovings of well-aligned filaments.

Studies have shown that not only yarn twist, but yarn hairiness (or fluffiness) is also an important source of misorientation [8]. Due to the short length of staple plant fibres, during the spinning process not all fibre ends are integrated into the yarn structure. The distribution (length and frequency) of fibre ends protruding from the fibre surface is referred to as yarn 'hairiness' by textile engineers. These protruding fibres have a negative impact on the packing of a fabric preform. Kim et al. [8] report that the compaction response of a fluffy roving may be as poor as that of a twisted spun yarn, relative to a straight roving; for instance, straight roving preforms have fibre volume fractions of 70% at $P = 2$ MPa, compared to fibre volume fractions of only 50% for both fluffy roving and spun yarn preforms. As observed in Fig. 6, numerous fibres are protruding from the structure of the yarns in the flax textiles, while filaments in the silk fibre textiles are well-integrated into the roving. It is noteworthy that the sources of misorientation (yarn twist and yarn hairiness) are lacking in silk textiles because silks exist as long fibres (i.e. filaments), unlike staple plant fibres. Put it simply, it is easier to align longer fibres than shorter fibres.

Yet another source of fibre misorientation is crimp. Due to the higher yarn linear density and circular yarn cross-section, the plant fibre textiles have a high degree of crimp (Fig. 6(a) and (b)). It would be expected that yarn bending deformation and nesting would be more important compaction mechanisms in plant fibre textiles than silk textiles. On a side note, as the longitudinal and transverse elastic tensile modulii of silk and flax are 7–17 GPa and 0.5–0.7 GPa, and 50–70 GPa and 4–9 GPa [36], respectively, and as the ultimate failure strain of silk and flax is 15–30% and 2–4% [36], respectively, silks can undergo much greater bending deformation without fibre breakage to enable efficient nesting and inter-layer packing. In fact, fibre breakage and lumen collapse have been reported to be two critical mechanisms responsible for the permanent deformation of plant fibre preforms during compaction [6]; both would have an expectedly detrimental effect on composite mechanical properties.

The degree of fibre separation also affects preform compaction [22]. Preforms with well-separated fibres compact better due to

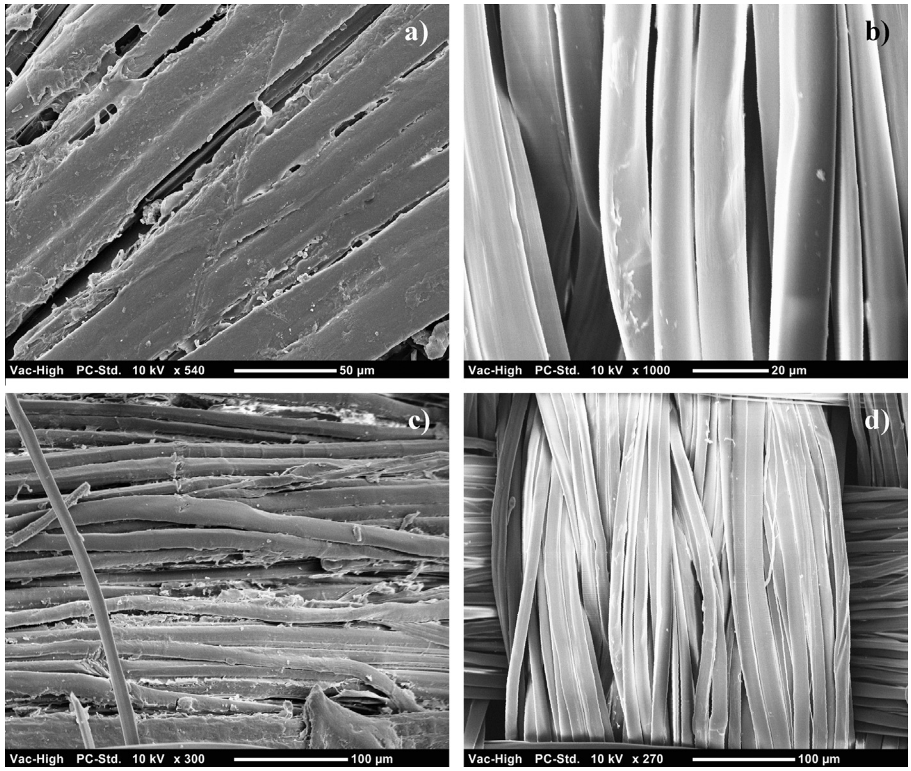

Fig. 7. Micrographs depicting (a) the bundled nature of plant fibres, and (b) the fibrillated nature of silk fibres. In addition, (c) plant fibres have a rougher surface than (d) silk fibres. Pectin, the binding agent between elementary plant fibres to form a technical plant fibre bundle, is usually only partially decomposed during the fibre extraction process. On the other hand, the fibroin strand in silk is usually well-degummed from sericin (the binding agent between two fibroin strands).

increased degrees of freedom. While plant fibres typically exist as bundles, silk fibres are well-separated from one another. This is shown in the micrographs of Fig. 7. Notably, it is the residue of the natural binding agent (pectin in plant fibres and sericin in silk), that dictates whether the fibres are separated or bundled.

During compaction, layers within the stacked fabric may move (i.e. slide against an adjacent layer) to dissipate energy. Therefore, the friction between fibre to fibre contacts is a relevant factor [3]. On a micro-scale (Fig. 7) and macro-scale (Fig. 6), silk fibres and their textiles have a lower surface roughness than plant fibre textiles. Therefore, silk textiles may find it easier to adjust to a more efficient packing arrangement due to lower friction forces. A similar explanation has been provided by Francucci et al. [16] when comparing rough sisal/jute fibre preforms with smooth glass fibre preforms.

It is interesting to note that silk fibres like plant fibres are highly anisotropic. While the transverse compressive properties of silkworm silk fibres have not been investigated in literature thus far, spider silk fibres have been subjected to such an analysis. Spider silk has a low transverse compressive modulus of 0.5–0.7 GPa [37] and a high Poisson's ratio of 0.4–0.5 [38]. Consequently, it undergoes no change in cross-sectional area, but experiences substantial deformation (i.e. flattening in cross-section shape) with increasing compressive force [37]. A substantial change in fibre cross-section shape is not unimportant in the context of preform compaction, as the former may alter the potential for fibre relative motion and yarn reorganisation and could possibly lead to hindered impregnation in localised inter-fibre zones. However, while a 20% reduction in thickness has been recorded for spider silk for compressive stresses of about 20 MPa (200 bar) [37], for the compaction pressures we are considering (up to 0.25 MPa) the expected change in cross-section is negligible. It is expected that silkworm silk would behave in a similar manner to spider silk, noting the similarity in the proteinaceous materials. Indeed, our preliminary studies on the SEM showed no visible change in cross-sectional shape of the silk fibres for low compaction pressures (of up to 0.1 MPa = 1 bar), thereby deeming the effects of change in cross-section shape unimportant for these low compaction pressures.

We conclude that the combination of all these structural and geometric factors should explain why silk textiles have a more favourable compaction response than plant fibre textiles.

3.1.2. Comparison with synthetic fibre textiles

While it is established that plant fibre reinforcements have lower compactability than synthetic fibre reinforcements [12,22] (and Table 2), the relative position of silk reinforcements has never been studied. For comparative purposes, Table 2 lists the representative fibre volume fraction v_f at $P = 2.0$ bar for plant fibre and synthetic

Table 2
Compaction of plant fibre and glass fibre preforms reported in literature.

Fibre type	Textile architecture	Areal density κ (g m^{-2})	v_f at $P = 2.0$ bar (%)	Source
Silk	Woven biaxial	87	57	This study
Silk	Woven biaxial	88	54	This study
Jute	Woven biaxial	300	29	[16]
Flax	Unidirectional	–	42	[22]
Hemp	Random mat	320	18	[22]
E-glass	Knitted biaxial	808	65	[39]
E-glass	Stitched biaxial	1000	61	[9]
E-glass	Stitched biaxial	618	54	[9]
E-glass	Woven biaxial	814	50	[9]
E-glass	Woven biaxial	–	54	[8]
E-glass	Woven biaxial	–	49	[8]
E-glass	Stitched unidirectional	–	57	[9]
E-glass	Random mat	450	26–31	[9]

fibre preforms. The previous sub-sections and literature comparisons in Table 2 make it abundantly clear that woven silk reinforcements are significantly more compactable than plant fibre reinforcements in of various forms (e.g. woven, unidirectional and randomly mats).

More interestingly, we note that the compactibility of silk textiles is comparable to, if not higher than, that of glass fibre textiles. For instance, woven silk and E-glass fabrics have a fibre volume fraction of 54–57% and 49–54% respectively, at $P = 2.0$ bar. This observation indicates the potential of silk textile reinforcements to match the performance of synthetic textiles, and perhaps even substitute them in reinforced plastics.

The potential sources of the comparable compactibility of silk and glass fibre reinforcements may lie in the fact that unlike plant fibres, silk, the only natural fibre to exist as long filaments (rather than short fibres), and its semi-products and textiles have much resemblance with synthetic fibres and their semi-products and textiles. Observing the structure of silk textiles in the previous section, it can be said that these similarities include: (i) fibres are long filaments, (ii) fibres have a smooth fibre surface, (iii) typically, low twist yarns or rovings are used for textiles and composites, (iv) roving cross-section shape is lenticular, and (v) fibres are well-separated and dispersed.

3.2. Effect of textile architecture

The effect of textile architecture on synthetic fibre preform compaction has been studied in some detail [8,9,39] and similar trends have been reported in the limited articles investigating plant fibre reinforcements [16,22]. The results presented in Figs. 4 and 5 are in agreement with the literature. Compared with unidirectional flax fabric, the curves for woven flax fabric are shifted downwards, and are more flat at higher compaction pressures. This is because compaction nesting is easier in unidirectional reinforcements than in multiaxial reinforcements, indicated by the higher a values of the former. In unidirectional fabrics, fibres in adjacent layers are parallel to each other and can therefore fill gaps by dislodging other fibres to produce a compact structure. In multiaxial fabrics, however, fibres/yarns leave voids during cross-over (referred to as inter-yarn porosity in Fig. 6) which cannot be filled by cross-over fibres in other layers. Essentially, fibre packing between layers with the same fibre/yarn orientation is easier than packing between layers with different fibre/yarn orientation. This trend is concurrent with the reported literature values in Table 2 as well.

While the effect of textile architecture on the compaction of silk textiles has not been specifically considered here, noting the general trends, we assert that unidirectional silk reinforcements would show even higher compactibility than the woven silk textiles that have been studied.

3.3. Effect of number of layers

The effect of number of layers on the compaction of silk woven textiles is shown in Fig. 8. It is observed that reducing the number of layers shifts the compaction curves upwards. Moreover, the curves become steeper at higher compaction pressures. The determined compaction parameters in Fig. 5 reveal that both $v_{f,0}$ and v_f at $P = 2.0$ bar increase by 0.6–1.8% (absolute) when fabric layers are increased from 12 to 16 to 21. Notably, the increase in v_f at $P = 2.0$ bar is linear. These are indicative of the fact that stacks made of less fabric layers are easier to compact to a given fibre volume fraction, particularly at higher compaction pressures, because nesting, the primary compaction mechanism of woven textiles, becomes progressively difficult with more layers [4], potentially due to the non-uniform transmission of the applied compressive force through the textile layers.

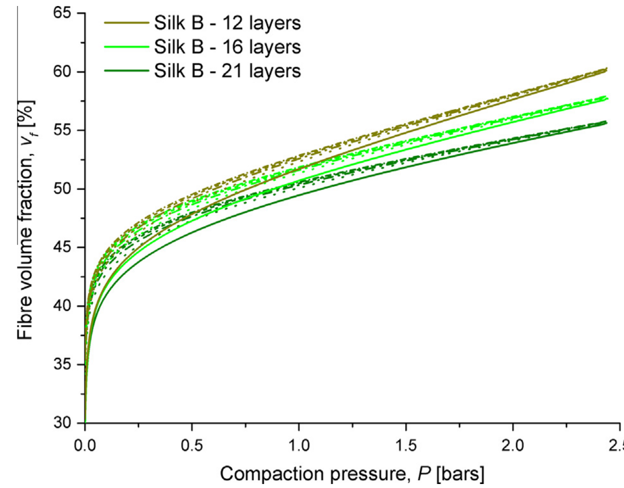

Fig. 8. Effect of number of layers on the compressibility of silk textiles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.4. Effect of compaction rate

Fig. 9 illustrates the effect of increasing compaction speed on the compressibility of woven silk textiles demonstrating that a marginal increase in fibre volume fraction was achieved for the same compaction pressure. In fact, an increase in the compaction speed from 0.5 mm/min to 50 mm/min increased $v_{f,0}$ from 30.5% to 32.2%, and increased the representative fibre volume fraction v_f at $P = 2.0$ bar from 53.7% to 54.1% (Fig. 5). However, the effect of speed on compaction behaviour may be regarded as negligible as the changes in compaction response were weak even for a 100 fold increase in compaction speed, particularly in comparison to the effect of fibre type, textile architecture and number of layers of fabric stacked for instance. Because some studies found conflicting trends [4,8,34], Robitaille et al. [4,9] argue that although compaction speed may have a small, if not negligible, effect on compaction, the effect of speed on relaxation is significant and detrimental. That is, increasing compaction speed may lead to less time for fibre/yarn rearrangement but induce greater fibre/yarn deformation [8]. Therefore, lower compaction speeds are preferred.

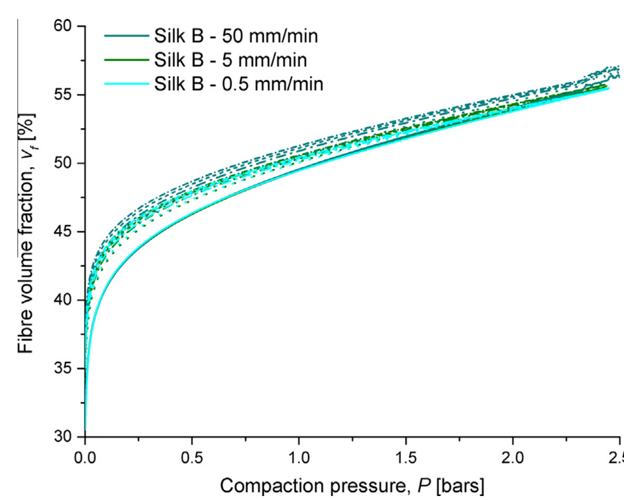

Fig. 9. Effect of compaction speed on the compressibility of silk textiles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.5. Effect of cyclic compaction

Although the speed of compaction may have little effect on compaction behaviour, the number of cycles of compaction has a noticeable and well-documented effect on preform compaction [4,8,9]. In agreement with the literature, our studies confirmed the general trend that the compaction curve almost stabilises after the second compaction cycle. In effect, subsequent compaction cycles (i.e. third, fourth and fifth) do not cause further significant shifts, although the compaction pressure required to achieve a given fibre volume fraction is considerably lower for the second compaction cycle in comparison to the first cycle, particularly at lower compaction pressures. This is shown qualitatively in Figs. 4, 8 and 9 and quantitatively in Fig. 5. In particular, the uncompressed fibre volume fraction $v_{f,0}$ tends to increase, a (indicative of the increase in fibre volume fraction at low compaction pressures) tends to decrease, b tends to increase, and the representative fibre volume fraction v_f at $P = 2.0$ bar (i.e. high pressure) tends to remain fairly constant, when the reinforcement is reloaded with a second compaction cycle. Subsequent compaction cycles have negligible effect on these parameters.

Our results and interpretation are of particular importance in the context of developing high fibre content silk reinforced composites. Maximising the fibre content is appealing for natural fibre composites, not only to improve composite mechanical performance by increasing the content of the reinforcing fibre [12,40], but also to increase the content of bio-based material and reduce the amount of polymer use. The latter improves sustainability credentials as natural fibres require significantly less energy for production than both synthetic fibres and polymer matrices [12,41]. If high compaction pressures (e.g. >2 bar), such as those realised using an autoclave for LCM processes (up to 10 bar) or a mechanical press for compression moulding (up to 100 bar), are used, a single compaction cycle is sufficient. That is, pre-compaction is not necessary. However, if low compaction processes are employed, such as vacuum infusion, pre-compaction of the reinforcement is an attractive technique. For instance, the uncompressed fibre volume fraction $v_{f,0}$ of silk A and B textiles would increase from 39% and 31% to 43% and 35%, respectively if a pre-compaction cycle is employed.

The reorganisation and permanent deformation of fibres/yarns within the textiles, driven through both elastic and irreversible mechanisms such as those illustrated in Fig. 2, are thought to be principal causes of the dependency of textile compaction behaviour on number of cycles. However, it should be noted that compaction mechanisms such as yarn cross-section deformation, yarn flattening and nesting become more significant at higher compaction pressures (and thus fibre volume fractions) [6]; therefore, the maximum compaction pressure of the previous cycle has an effect on the compaction in the next cycle. In addition, stacked textiles have a time-dependent visco-elastic behaviour [6], therefore studying preform relaxation is as important as studying preform compaction.

4. Conclusions

Our main conclusion asserts that silk textile reinforcements are significantly more compactable than plant fibre reinforcements, and even glass fibre textiles. For instance, the fibre volume fraction (at a compaction pressure of 2.0 bar) of woven biaxial fabrics of silk, plant fibres and E-glass are 54–57%, 30–40% and 49–54%, respectively. Consequently, we propose that silk fibre reinforcements offer a unique opportunity in the production of high fibre volume fraction natural fibre composites, which has been difficult to achieve so far with plant fibre reinforcements. Thus silk might become a key player in the generic development of bio-based composite materials not least because high fibre content generally imparts higher mechanical performance and improved sustainability (lower embodied energy).

The microscopic analysis of silk textiles revealed the primary sources of optimal packing ability of this material as: (i) favourable fibre/yarn/fabric geometry, (ii) high degree of fibre alignment and dispersion, and (iii) suitable technical fibre properties. Silk is the only natural fibre to exist as a filament and its textiles are structurally comparable to synthetic fibre reinforcements.

While the effects of compaction rate, number of layers, and multiple compaction cycle on the compaction response of silk textiles has been systematically studied, more data needs to be collected, particularly on the effect of lubrication on compaction behaviour. Studies on the relaxation behaviour and permeability of silk fabrics will be important for improved understanding on the manufacture of silk fibre composites using liquid composite moulding processes.

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