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Silk-Based composites and their applications: A comprehensive review

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Abstract

Silk is a very special kind of material that comes from nature. Silk is very strong, can be used in the body without causing problems, can break down easily and can be combined with other materials in different ways. This review explains about silk-based composite materials and it covers where they come from, how they are made and what they can be used for. Silk fibroin is a type of protein that comes from silkworms and some spiders. It is the main part of the material that makes these composites strong and flexible. By combining silk with other materials such as plastics, ceramics, metals, nanomaterials and hydrogels it can create new systems that have specific features and functions looks the use of different materials and technologies to solve problems in different fields. Some of the applications are related to health such as making artificial organs, delivering medicines, healing wounds and making devices that can be put inside the body.

Keywords: Silk fibroin, sericin, composite materials, biomedical applications, drug delivery, biopolymer

Introduction

Silk is a natural protein-based fibre produced by various arthropods most notably the domesticated silkworm *Bombyx mori* and several species of spiders. For millennia, silk has been prized for its lustrous appearance with tensile strength and smooth texture serving as a luxury textile across diverse civilizations. However, in the past three decades there has been a fundamental shift in silk is perceived by the scientific community. Specifically, silk fibroin is recognized as an outstanding biomaterial with properties that few synthetic polymers can match high tensile strength approaching 740 MPa in single fibers extensibility of up to 35% outstanding biocompatibility and an inherent ability to be processed into a variety of morphologies including films, fibers, foams, hydrogels and micro- and nanoparticles (Altman et al., 2003; Kaplan et al., 1998) [2, 12].

The growing demand for advanced functional materials in biomedical, environmental and industrial sectors has driven researchers to explore composite systems that harness the strengths of silk while compensating for its limitations. Silk-based composites are engineered by combining silk fibroin or sericin with secondary phases ranging from synthetic polymers such as polyethylene glycol and polylactic acid to inorganic materials including hydroxyapatite, titanium dioxide, graphene and carbon nanotubes to produce hybrid materials with synergistic properties unattainable by either component alone. (Omenetto & Kaplan, 2010; Hardy & Scheibel, 2010) [28, 8].

The range of silk-based composite applications is broad and continues to expand (Fig 1.). In tissue engineering, silk composites serve as scaffolds that mimic the extracellular matrix supporting cell adhesion with proliferation and differentiation for the regeneration of bone, cartilage, skin, nerve and vascular tissue (Rockwood et al., 2011) [31]. In drug delivery, the controlled release of therapeutics from silk-based vehicles offers unprecedented precision (Seib & Kaplan, 2013) [25]. Beyond biomedicine silk composites have found utility in electronic skin optical waveguides, water purification membranes, sustainable packaging materials and as reinforcing agents in structural composites (Tao et al., 2014; Yucel et al., 2011) [36, 31].

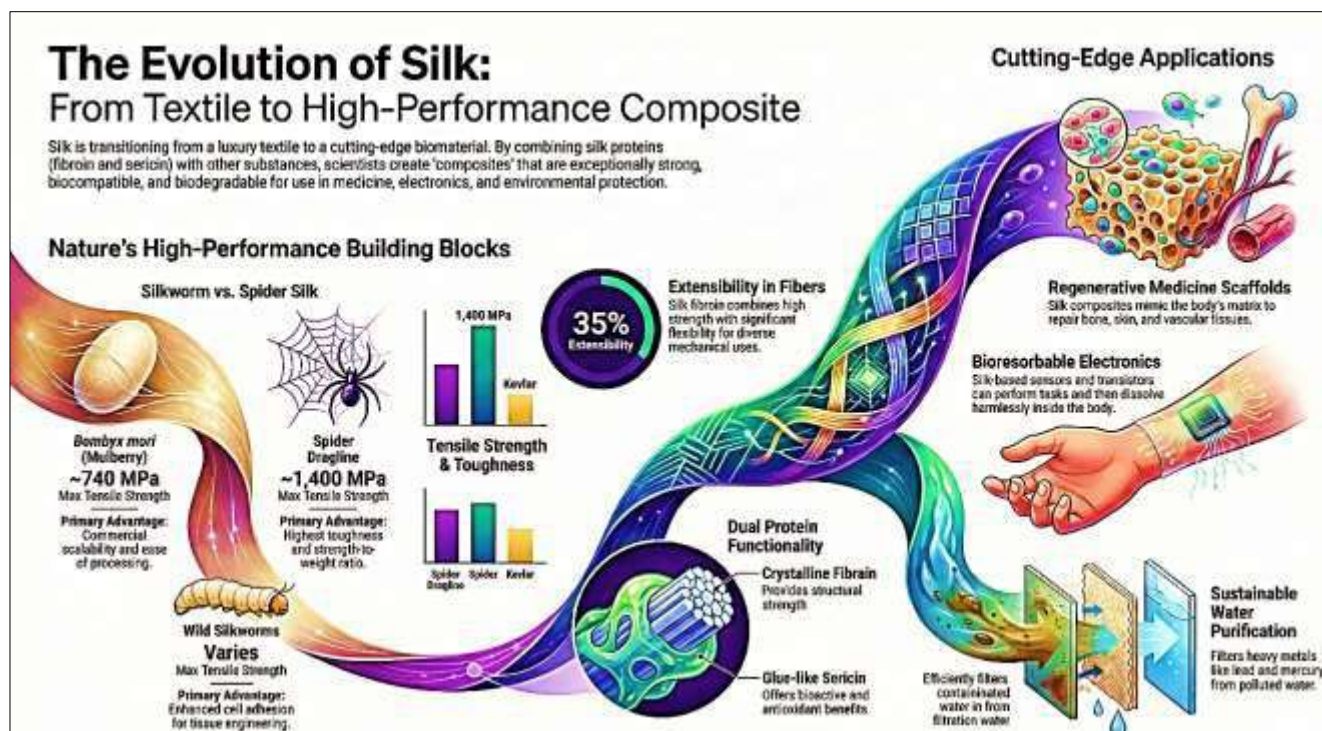


Fig 1: Applications of silk-based composites.

2. Sources and Structural Characteristics of Silk

2.1 Bombyx mori Silk

The most commercially significant silk is produced by the domesticated mulberry silkworm *Bombyx mori*. A single cocoon yields a continuous filament of approximately 1,000 - 1,500 meters in length. Each filament is composed of a fibrous core protein and fibroin sheathed in a glue like protein called sericin. Fibroin itself consists of two chains a heavy chain (~390 kDa) and a light chain (~26 kDa) connected by a single disulfide bond. The heavy chain is the primary structural element characterized by a repetitive sequence of glycine and serine residues that self-assemble into antiparallel β -sheet crystallites embedded within an amorphous matrix. (Zhou et al., 2001; Vepari & Kaplan, 2007) [50, 39].

The crystalline β -sheet domains confer the high tensile strength and stiffness of silk fibroin while the amorphous regions provide flexibility and extensibility. This hierarchical organization from molecular β -sheets to nanofibril bundles to macroscopic fibers is responsible for the remarkable mechanical performance of silk. The crystallinity of silk fibroin can be modulated through processing conditions such as temperature, humidity, organic solvents and mechanical strain enabling fine control over the properties of silk based materials (Jin & Kaplan, 2003) [10].

2.2 Wild Silkworm Silk

In addition to *Bombyx mori* a variety of wild silkworms generate silk that has unique physical and chemical characteristics. Among the most extensively researched are *Antheraea mylitta* (tasar silk), *Antheraea assama* (muga silk), *Philosamia ricini* (eri silk) and *Antheraea pernyi* (Chinese oak silkworm). Typically, wild silks have a higher content of acidic amino acids like aspartic acid and glutamic acid which enhance their cell adhesion properties to a

greater extent than *Bombyx mori* fibroin this feature is particularly beneficial for applications in tissue engineering (Kundu et al., 2008) [16]. Furthermore, wild silk strands usually show higher extensibility, and their sericin components have notably different amino acid profiles from those of *Bombyx mori* sericin affecting their immunogenicity and bioactivity (Patra et al., 2015) [29].

2.3 Spider Silk

Spider silk is often regarded as the most remarkable natural fiber available, merging incredible strength with outstanding toughness. The dragline silk from *Nephila clavipes* and *Araneus diadematus* has been the subject of extensive research, showing tensile strengths reaching as high as 1,400 MPa and an extensibility ranging from 30% to 40% which provides it with toughness that exceeds that of both Kevlar and high tensile steel when compared by weight (Gosline et al., 1999) [7]. The proteins that compose spider silk is identified as spidroins are produced by genes belonging to the Major Ampullate Spidroin (MaSp) group which is similar to the fibroin found in *Bombyx mori*, spidroins feature repetitive structural patterns polyalanine segments that create β -sheets alongside glycine-heavy areas that enhance elasticity.

The scarcity of spider silk, stemming from the cannibalistic and territorial behaviors of these arachnids, has driven the need for alternative production methods through recombinant techniques. Spidroins have been successfully generated in various organisms which including bacteria, yeast, plants, mammalian cells and genetically modified silkworms each demonstrating different levels of success in mimicking the mechanical properties of natural silk (Scheibel, 2004; Tokareva et al., 2014) [34, 38]. The production of recombinant silk proteins presents thrilling possibilities for crafting engineered composites with customizable mechanical and biological characteristics.

Table 1: Major Sources of Silk and Their Key Structural Characteristics

Silk Source	Organism	Key Structural Components	Notable Mechanical / Biological Properties	Major Applications	References
Mulberry silk	<i>Bombyx mori</i>	Fibroin core (heavy and light chains) coated with sericin	High tensile strength, biocompatibility, tunable biodegradation	Tissue engineering scaffolds, drug delivery systems, sutures	Altman et al., 2003; Vepari & Kaplan, 2007 ^[2, 39]
Tasar silk	<i>Antheraea mylitta</i>	Fibroin with higher acidic amino acids	Improved cell adhesion and bioactivity	Tissue engineering and biomaterials	Kundu et al., 2008 ^[16]
Muga silk	<i>Antheraea assama</i>	Fibroin with distinctive amino acid composition	High durability and thermal stability	Biomedical materials and textiles	Kundu et al., 2012 ^[17]
Eri silk	<i>Philosamia ricini</i>	Non-mulberry fibroin proteins	Enhanced elasticity and biocompatibility	Biomedical scaffolds and composites	Kundu et al., 2012 ^[17]
Spider silk	<i>Nephila clavipes</i> , <i>Araneus diadematus</i>	Spidroin proteins with β -sheet crystallites	Extremely high strength (~1400 MPa) and toughness	High-performance biomaterials, recombinant protein composites	Gosline et al., 1999; ^[7] Tokareva et al., 2014 ^[38]

3. Processing and Fabrication of Silk-Based Composites

3.1 Dissolution and Regeneration

The creation of silk-based composites generally starts with the dissolution of natural silk fibers. In the case of *Bombyx mori* fibroin this process includes degumming eliminating sericin through boiling in weak alkaline solutions followed by dissolving in chaotropic substances like lithium bromide or a combination of calcium chloride and water with subsequent dialysis in water to yield a regenerated silk fibroin (RSF) solution (Rockwood et al., 2011)^[31]. RSF acts as a multifunctional precursor for most silk-based composite production. By integrating RSF with other materials either before or during processing, it is possible to create composite like films, fibers gels and particles.

3.2 Electrospinning

Electrospinning is a commonly utilized method for creating nanofibers composed of silk and other materials. This technique involves the use of an electric field to expel a silk solution from the end of a needle, resulting in a charged jet that stretches and narrows into very fine fibers collected as a nonwoven sheet. Silk fibroin can be electrospun independently or mixed with other polymers such as polyethylene oxide (PEO), poly (lactic-co-glycolic acid) (PLGA), polyvinyl alcohol (PVA), collagen and chitosan to enhance its ability to spin and modify the characteristics of the fibers (Jin et al., 2002; Bhardwaj & Kundu, 2010)^[11, 3]. Mats made from electrospun silk composites possess a large surface area, high porosity and a structure that closely mimics the extracellular matrix making them appropriate for use as scaffolds in tissue engineering and as wound care materials.

3.3 Freeze-Drying and Scaffold Fabrication

Freeze-drying is also known as lyophilization of silk fibroin solutions or mixtures results in three-dimensional structures that are extremely porous and contain interconnected networks of pores. By adjusting the freezing rate and temperature dimensions and design of the pores can be

modified. Scaffolds created by freeze drying silk in collaboration with hydroxyapatite and gelatin or carbon nanotubes have shown enhanced mechanical characteristics and compatibility with cells for applications in bone and cartilage tissue engineering (Gharivi, M et al).

3.4 Hydrogel Formation

Silk fibroin undergoes sol-gel transition via β -sheet-mediated physical crosslinking producing hydrogels without the need for toxic chemical crosslinkers. Gelation can be induced by vortexing and sonication changes in pH or temperature or addition of organic solvents. Silk hydrogel composites incorporating growth factors and nanoparticles or cells have been extensively explored for injectable drug delivery and minimally invasive tissue engineering (Wang et al., 2008)^[40]. The mechanical properties of silk hydrogels can be tuned over a wide range from tens of pascals to hundreds of kilopascals by adjusting silk concentration, gelation conditions and composite composition.

3.5 Silk-Reinforced Polymer and Ceramic Composites

Silk fibers have been utilized as reinforcing materials within polymer matrices similar to composites reinforced with glass or carbon fibers. Composites made with silk fiber reinforced epoxy, polyester and biopolymer matrices have been evaluated for their strength resistance to impact that ability to dampen vibrations (Cheung et al., 2009)^[5]. Surface modification on silk fibers with silane coupling agents like alkali treatment or plasmas hugely strengthens the interfacial bonding strength between the silk and polymer matrix. Similarly, silk ceramic composites have been developed for bone tissue engineering and coatings for orthopedic implants more specifically silk with hydroxyapatite (HA) to impart the HA potential to support bone outgrowth that silk offers in terms of its strength as well as compatibility with biological systems. (Kim et al., 2005)^[14]. The different types of composites are presented in the table 2.

Table 2: Types of Silk-Based Composite Materials

Composite Type	Secondary Material	Key Functional Properties	Example Applications	References
Silk-polymer composites	PVA, PCL, PLGA, PEG	Improved mechanical strength and flexibility	Tissue scaffolds, wound healing materials	Bhardwaj et al., 2012 ^[4]
Silk-ceramic composites	Hydroxyapatite, bioglass	Osteoconductivity and bone integration	Bone regeneration scaffolds	Kim et al., 2005 ^[14]
Silk-nanomaterial composites	Graphene oxide, carbon nanotubes	Electrical conductivity and mechanical reinforcement	Biosensors, flexible electronics	Wu et al., 2017 ^[51]
Silk-metal nanoparticle composites	Silver nanoparticles	Antibacterial activity	Wound dressings	Rujitanaroj et al., 2008 ^[33]
Silk-hydrogel composites	Growth factors, nanoparticles	Controlled drug release	Injectable drug delivery systems	Wang et al., 2008 ^[40]

4. Biomedical Applications

4.1 Tissue Engineering Scaffolds

Tissue engineering aims to regenerate damaged or diseased tissues through the combined use of scaffolds, cells and biological signals. Silk based composite scaffolds have been developed for virtually every tissue type owing to silk fibroin's biocompatibility with slow biodegradation matching tissue ingrowth and mechanical properties. For tissue engineering hydroxyapatite, bioglass and calcium phosphate composites have demonstrated outstanding osteo conductivity and ability to support osteoblast differentiation *in vitro* and bone defect healing (Meinel et al., 2005; Marolt et al., 2012) [25, 24]. The incorporation of bone morphogenetic proteins (BMPs) and vascular endothelial growth factors (VEGFs) into silk composite scaffolds further enhances their regenerative capacity.

Cartilage tissue engineering has benefited from glycosaminoglycan (GAG), collagen and chondroitin sulfate composites that mimic the biochemical and mechanical microenvironment of native cartilage promoting chondrogenic differentiation of mesenchymal stem cells (MSCs) (Wang et al., 2008) [40]. For skin tissue engineering electrospun silk or collagen and silk or poly(ϵ -caprolactone) (PCL) composite mats have shown accelerated wound closure and epidermal regeneration in preclinical models (Bhardwaj et al., 2012) [4]. Vascular tissue engineering with tubular elastin, PCL and polyurethane composites has yielded grafts with burst pressures and suture retention strengths meeting clinical requirements (Lovett et al., 2011) [31].

4.2 Drug Delivery Systems

The ability of silk fibroin to encapsulate diverse drug classes like small molecules, proteins nucleic acids and nanoparticles with high loading efficiency and release them in a controlled sustained manner makes it an attractive carrier platform. Drug release from silk matrices occurs through diffusion and enzymatic biodegradation and the rates of which can be tuned by adjusting β -sheet crystallinity and composite composition (Seib & Kaplan, 2013) [25]. Silk fibroin nanoparticles prepared by nanoprecipitation or desolvation methods have been loaded with anticancer agents such as doxorubicin, paclitaxel and tamoxifen achieving tumor targeted delivery and reduced systemic toxicity in animal models (Wongpinyochit et al., 2015) [43]. Silk hydrogel composites incorporating growth factors such as BMP-2 fibroblast growth factor (FGF) and insulin have been investigated as local delivery depots following orthopedic surgery or wound care. Instead of a single heavy dose that the body might process too quickly then silk structures allow for a steady gradual release of healing molecules. (Wang et al., 2008) [40] suggests this "slow and steady" approach is far better at helping tissues repair themselves.

4.3 Wound Dressings

Wound dressings must fulfill multiple functions that provide a moist wound environment, absorb excess exudate protect against the infection and ideally promote wound healing. Silk based composite wound dressings have been developed to meet these requirements. Electrospun silk or silver nanoparticle composites exhibit potent antibacterial activity against both Gram-positive and Gram-negative bacteria while promoting fibroblast migration and proliferation

(Rujitanaroj et al., 2008) [33]. Silk or chitosan composite films combine the antimicrobial properties of chitosan with the mechanical strength of silk and have demonstrated efficacy in chronic wound models. Sericin is historically discarded as a processing byproduct has gained recognition as a bioactive component that promotes skin cell proliferation and exhibits antioxidant and anti-inflammatory properties when incorporated into composite wound dressings (Kundu et al., 2016) [19].

4.4 Implantable Devices and Biosensors

The optical transparency which mechanical flexibility and biocompatibility of silk fibroin films make them attractive substrates for implantable electronic and optical devices. Silk based bioresorbable electronics including transistors, antennas, sensors and drug delivery platforms have been demonstrated in animal models. Where, the devices perform their intended function and then dissolve harmlessly after a programmed period (Tao et al., 2014) [36]. Silk optical waveguides capable of transmitting light through tissue have been fabricated for phototherapy and optogenetic applications (Huby et al., 2013) [9]. In the field of biosensing, silk fibroin composite films functionalized with enzymes or antibodies or aptamers have been developed for the detection of cancer biomarkers and environmental pollutants with high sensitivity and specificity.

5. Environmental Applications

5.1 Water Purification

The increasing burden of water pollution by heavy metals and pathogens has driven the search for efficient sustainable adsorbent and membrane materials. Silk based composites have emerged as promising candidates for water remediation. Silk sericin or polyacrylamide hydrogel composites and silk fibroin or graphene oxide membranes have demonstrated exceptional adsorption capacities for heavy metals such as lead, cadmium, chromium and mercury as well as cationic and anionic dyes (Zhao et al., 2015; Li et al., 2019) [49, 21]. The abundant amino hydroxyl and carboxyl functional groups on silk protein chains provide numerous binding sites for pollutant molecules and regeneration of spent adsorbents using mild acidic or basic solutions is feasible without significant loss of capacity over multiple cycles.

Silk-based nanofiltration membranes have been fabricated by cross-linking silk fibroin with polyethyleneimine or glutaraldehyde achieving high water flux and effective rejection of divalent ions and organic micropollutants. The inherent biodegradability of silk ensures that end of life disposal of these membranes does not introduce persistent pollutants into the environment a critical advantage over conventional polymer-based membranes (Deng et al., 2020) [6].

5.2 Agricultural Applications

Silk-based composites have found innovative applications in the agricultural sector particularly as controlled release carriers for agrochemicals and as soil conditioners. Silk fibroin encapsulation of pesticides, herbicides and fertilizers enables slow sustained release that reduces the frequency of applications which minimizes leaching into groundwater and decreases environmental toxicity to non-target organisms (Zhang et al., 2018) [48]. Silk sericin has been investigated as a plant growth promoter and biostimulant

with demonstrated efficacy in enhancing germination rate, root elongation and crop yield of various agricultural plants.

6. Electronic and Photonic Applications

6.1 Flexible Electronics

The emergence of flexible and wearable electronics has created demand for substrate materials that are mechanically compliant, biocompatible and amenable to precise patterning. Silk fibroin films meet these requirements exceptionally well. Transistors diodes, organic light-emitting devices (OLEDs) and sensors fabricated on silk substrates have demonstrated comparable performance to those on conventional rigid substrates with the additional advantage of biodegradability (Rogers et al., 2010; Kim et al., 2010) [32, 18]. Silk composite films incorporating metallic nanowires or carbon nanotubes or conductive polymers serve as stretchable electrodes and interconnects in electronic skin applications with capable of detecting temperature pressure and chemical signals (Wu et al., 2013) [44].

6.2 Optical and Photonic Devices

Silk fibroin is optically transparent across the visible spectrum with low scattering losses enabling its application in passive and active photonic structures. Silk-based diffraction gratings, holograms and photonic crystals have been fabricated by nano-imprinting and soft lithography (Lawrence et al., 2008) [20]. Doped silk composite films incorporating fluorescent nanoparticles and quantum dots or rare earth elements have been developed for luminescent applications in anti-counterfeiting imaging and lighting. Silk based optical waveguides capable of guiding light over centimeter scale distances have been demonstrated for *in vivo* phototherapy and biosensing (Huby et al., 2013) [9].

7. Structural and Mechanical Applications

The high specific strength and toughness of silk fibers make them attractive reinforcements in lightweight structural composites. Silk fiber reinforced polymer matrix composites have been evaluated as sustainable alternatives to glass fiber composites in applications such as automotive panels, sports equipment and construction elements (Cheung et al., 2009) [5]. Studies have demonstrated that alkali-treated silk fiber or epoxy composites exhibit specific tensile and flexural strengths competitive with glass fiber composites at reduced weight and environmental impact. The energy absorbing capacity of silk-reinforced composites attributable to the hierarchical fiber fracture mechanism is particularly valuable in impact resistant structures.

Silk has also been incorporated into cement and geopolymer matrices as a reinforcing fiber improving fracture toughness and crack resistance. Bio inspired structural materials that mimic the nacre-like architecture of laminated silk or clay nanocomposites have been fabricated by sequential assembly achieving outstanding stiffness toughness combinations that challenge synthetic nacre analogy (Yao et al., 2010) [45].

8. Food Packaging Applications

Consumer demand for sustainable biodegradable food packaging has spurred research into biopolymer-based films and coatings. Silk fibroin and sericin offer excellent film forming properties with optical clarity and gas barrier characteristics suitable for food packaging (Teramoto et al.,

2004) [37]. Silk composite films with antimicrobial agents such as silver nanoparticles and zinc oxide nanoparticles or natural antimicrobial extracts have demonstrated efficacy in extending the shelf life of fresh produce like meat and fish. The inherent biodegradability of silk based packaging films ensures that they do not contribute to the global plastic waste crisis and their amino acid rich composition makes them potentially compostable as soil nutrients.

9. Challenges and Limitations

Despite the remarkable versatility of silk based composites with several challenges limit their widespread adoption. The scalability of silk production particularly recombinant spider silk remains a significant bottleneck. Although *Bombyx mori* silk is produced on a commercial scale, the supply of wild silkworm silk and the cost of purified recombinant silk proteins remain high relative to synthetic polymers (Scheibel, 2004) [34]. Variability in silk composition and properties across different breeds and geographic sources introduces batch to batch inconsistencies that are challenging to control in composite fabrication.

The hydrophilic nature of silk proteins makes silk composites susceptible to moisture induced softening and limiting their utility in wet or humid environments outside biomedical contexts (Hardy & Scheibel, 2010) [10]. The immunogenicity of residual sericin in silk based materials has been a historical concern though extensive degumming and purification largely mitigate this issue for fibroin based composites. Regulatory pathways for silk based biomedical devices remain complex with requirements for extensive biocompatibility testing and demonstration of batch reproducibility.

10. Future Perspectives

The future of silk-based composites is poised for significant advances driven by convergent developments in synthetic biology, nanotechnology additive manufacturing and artificial intelligence. Recombinant silk technology, including the design of fusion proteins combining silk structural domains with bioactive peptide sequences and growth factors or enzyme-responsive motifs will enable next generation composite materials with programmable biological functions (Tokareva et al., 2014) [38]. Metabolic engineering of silkworms to produce chimeric silk fibers containing heterologous protein sequences represents an emerging approach to diversifying silk composite building blocks.

Additive manufacturing includes bioprinting of silk composite inks and melt electrowriting of silk composite scaffolds is expected to enable the fabrication of patient specific implants and tissue constructs with complex anatomically accurate geometries (Nguyen et al., 2019) [27]. Machine learning approaches are increasingly being applied to predict structure property relationships in silk composites accelerating the discovery of optimal formulations for target applications. The integration of silk composites with digital health technologies including wearable biosensors and smart wound dressings will further expand their clinical utility.

Sustainability considerations will increasingly drive the adoption of silk based composites as drop in replacements for petroleum derived polymers in packaging textiles and construction materials. Life cycle assessment studies consistently demonstrate favorable environmental profiles for silk composites relative to glass or carbon fiber

composites particularly when end of life biodegradation is accounted (Rauer et al., 2021)^[30]. Circular economy models incorporating silk sericin as a co product with fibroin will improve the economic viability of silk based composite manufacturing.

11. Conclusions

Silk based composites represent a fascinating and rapidly evolving class of functional materials that bridge the gap between natural biomaterials and high performance engineered systems. The unique combination of mechanical strength, biocompatibility, biodegradability and processability that silk proteins offer is unrivaled among natural materials and the ability to fine-tune composite properties through choice of secondary phase fabrication technique and post processing treatment provides virtually unlimited design space for materials engineers.

While challenges related to scalability cost regulatory pathways and long-term *in vivo* performance must be addressed the trajectory of the field is unmistakably toward broader adoption and more sophisticated application of silk based composites. As synthetic biology expands the structural and functional repertoire of silk proteins beyond what nature has provided and as advanced manufacturing techniques enable increasingly complex composite architectures and the impact of silk-based materials on human health sustainability and technology is expected to grow substantially.

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Competing interests

Authors have declared that no competing interests exist.

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