

Biodegradable Polymers for Electrospun Biomedical Constructs: A Comparative Analysis of Synthetic and Natural Fibre-Forming Polymers

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Abstract

The performance of an electrospun biomedical construct is determined less by the spinning parameters than by the polymer from which it is made, yet polymer selection in the literature is usually justified by precedent rather than by a systematic comparison. This review compares eight fibre-forming polymers used in electrospun biomedical constructs — four synthetics (poly (lactic acid), poly (glycolic acid), polycaprolactone, poly (ethylene terephthalate)) and four naturals (chitosan, collagen, alginate, silk fibroin) — across four property groups: thermal and processing behaviour, degradation kinetics and products, biological interaction, and solvent requirements. Reported values are collected in a common format and three derived indicators are introduced: the thermal processing window, the ratio of resorption time to the typical healing period of the target tissue, and the presence or absence of cell-adhesion motifs. The analysis shows that the synthetic polyesters offer a wide and reproducible processing window but a fixed degradation rate and acidic degradation products, whereas the natural polymers offer favourable biological interaction but, with one exception, either degrade too quickly or cannot be spun without a carrier. Silk fibroin is identified as the only polymer whose degradation rate spans the full range required by the reported applications — from weeks to a year — while its degradation products remain metabolisable, and the practical cost of this advantage is quantified as the carrier fraction required for stable jet formation.

Keywords: Biodegradable polymers; electrospinning; polyesters; chitosan; collagen; alginate; silk fibroin; degradation kinetics; polymer selection.

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1. Introduction

The requirements placed on materials used in medicine have risen steadily. A material intended for implantation or for prolonged contact with tissue must combine biocompatibility, biodegradability and acceptable mechanical properties — a combination that few polymer classes satisfy simultaneously [1, 2]. In electrospun constructs the problem is sharpened by a further requirement: the polymer must also be capable of forming a continuous jet from solution, which restricts the field to polymers of sufficient molecular weight that dissolve at concentrations above the entanglement threshold [3, 4].

Two families of polymers dominate the reported work. Synthetic biodegradable polyesters — poly (lactic acid), poly (glycolic acid), polycaprolactone and their copolymers — are well characterised, available in reproducible grades, and spin readily. Polymers of natural origin — chitosan, collagen, alginate, silk fibroin — have significant advantages over synthetic ones in that their degradation products in the organism are natural metabolites that participate in intracellular biochemical processes, which makes them substantially more biocompatible; their mechanical properties are in some cases similar to those of products made from synthetic polymers [5–7].

The choice between the two families, and within each of them, is normally made by analogy with earlier studies. Direct comparisons are rare, and when they are made, they usually cover two or three polymers within a single application. The consequence is that the trade-offs are not made explicit: a reader cannot easily establish, for instance, how much biological performance is given up by choosing a polyester for its processing convenience, or how much processing difficulty is accepted by choosing a natural polymer for its biocompatibility.

The aim of this review is to compare the principal fibre-forming polymers used in electrospun biomedical constructs within a single framework, to express the comparison through derived indicators that are directly meaningful for design decisions, and to state the trade-off associated with each polymer family quantitatively where the data permit.

2. Framework of the comparison

Eight polymers were selected: four synthetic (PLA, PGA, PCL, PET) and four natural (chitosan, collagen, alginate, silk fibroin). PET is not biodegradable and is

included deliberately as a bio-inert reference against which the degradable polymers can be judged.

Four property groups were used. The first covers thermal and processing behaviour: glass-transition temperature, melting point, onset of thermal degradation and solubility. The second covers degradation: mechanism, characteristic resorption time and the nature of the degradation products. The third covers biological interaction: the presence of cell-adhesion motifs, hydrophilicity, and any reported specific biological activity. The fourth covers the solvent system required, which determines both the practicability of the process and the residual toxicity of the product.

Three derived indicators are introduced. The thermal processing window ΔT is defined as the difference between the onset of thermal degradation and the melting point, and characterises the margin available for melt processing and for heat sterilisation. The resorption ratio k is the ratio of the reported resorption time to the typical healing or regeneration period of the target tissue; values near unity indicate a well-matched polymer, values much below unity indicate premature loss of support, and values much above unity indicate an unnecessarily persistent implant. The third indicator is binary: the presence or absence of arginine–glycine–aspartate (RGD) or equivalent cell-adhesion sequences in the primary structure.

3. Synthetic biodegradable polymers

3.1. Poly (lactic acid)

Among the synthetic polymers listed, poly (lactic acid) (polylactide, PLA) is the most promising: it is a polymer with a melting point of approximately 160–180 °C and a glass-transition temperature of 50–60 °C. It is obtained from plant material and hydrolyses in the human body to lactic acid, a natural metabolite capable of entering the tricarboxylic acid cycle. For this reason it is used in the manufacture of medical implants in the form of anchors, screws, plates, pins, rods and meshes. Depending on the type used, it is resorbed in the body over a period of 6 months to 2 years. This gradual degradation is appropriate for a supporting structure, because the load is transferred progressively to the tissue — for example to bone — as the site heals [8].

A further notable property of poly (lactic acid) is the ability to obtain a polymer with defined mechanical properties by changing the ratio of D- and L-isomers in the material. Poly (L-lactic acid) (PLLA) is more widely

used in orthopaedic implants because this material retains its original strength longer than poly (D, L-lactic acid) (PDLA), which combines both enantiomers. Notwithstanding these advantages, poly (lactic acid) has significant drawbacks: hydrophobicity, which impedes cell adhesion to the polymer surface, and the absence of reactive groups on the surface, which is associated with a lack of biological activity — although the latter can be considered a drawback only in some cases [8, 9].

3.2. Poly (glycolic acid)

Polyglycolide, or poly (glycolic acid) (PGA), is a biodegradable thermoplastic polymer and the simplest linear aliphatic polyester. Its glass-transition temperature is 35–40 °C and its melting point lies in the range 225–230 °C. PGA also has a high degree of crystallinity, approximately 45–55%, which results in insolubility in water [10].

Degradation is erosive and occurs in two stages, in which the polymer reverts to its monomer, glycolic acid. Under physiological conditions polyglycolide degrades by random hydrolysis and is also broken down by certain enzymes, particularly those with esterase activity. The degradation product, glycolic acid, is not toxic; however, like ethylene glycol, it is metabolised to oxalic acid, which can make it hazardous. Poly (glycolic acid) and its copolymers are used mainly in the manufacture of biodegradable sutures and bone screws. Studies of sutures made from polyglycolide have shown that the material loses half of its strength after two weeks, and the polymer is completely resorbed by the organism within four to six months. Biodegradation occurs faster in vivo than in vitro, a phenomenon associated with cellular enzymatic activity [10].

3.3. Polycaprolactone

Polycaprolactone (PCL) is a biodegradable polyester with a low melting point of approximately 60 °C and a glass-transition temperature of approximately –60 °C. It is a polymer of ϵ -caprolactone with a molecular weight from 2000 to 100 000. Polycaprolactone degrades under physiological conditions by hydrolysis of its ester bonds and is therefore used as an implantable biomaterial. In particular, it is attractive for the production of long-term implants because it degrades even more slowly than polylactide: complete resorption by the human body takes approximately three years. The degradation products are caproic acid, water and carbon dioxide. It is widely used as a suture material, as a filler (injectable

implants in cosmetology) and in drug-delivery systems intended for uniform, targeted delivery over extended periods [11].

The very low glass-transition temperature of PCL has a practical consequence that is often overlooked: at body temperature the polymer is far above its T_g and therefore behaves as a soft, ductile solid, which is advantageous for conformability but limits its use where dimensional stability under load is required.

3.4. Poly (ethylene terephthalate)

Poly (ethylene terephthalate) (PET) is a bio-inert thermoplastic and the most widespread representative of the polyester class. The product of the polycondensation of ethylene glycol with terephthalic acid is a hard, colourless and transparent substance; in the crystalline form it is opaque and white. When heated to the glass-transition temperature it becomes transparent, and the material retains this state if cooled rapidly. The length of the polymer molecule affects the viscosity of PET: the higher the viscosity, the slower the rate of crystallisation. PET is almost insoluble in water and in organic solvents, resistant to acids and weakly alkaline solutions, durable, abrasion-resistant and a good dielectric. By changing the chemical composition of the side groups of the polymer, materials with different degradation rates can be produced. In medicine, poly (ethylene terephthalate) is used in the manufacture of prostheses for blood vessels, tendons, ligaments and heart valves [12, 13].

Because PET is not resorbed, it defines the limiting case of the resorption ratio: the implant remains indefinitely, and the design question becomes one of long-term tissue tolerance rather than of degradation matching.

4. Natural biodegradable polymers

4.1. Chitosan

Chitosan is an aminopolysaccharide, 2-amino-2-deoxy- β -D-glucan, formed by the deacetylation of chitin. Chitin occurs in the shells of crustaceans, in insect cuticles and in the cell walls of fungi and bacteria, which makes industrial extraction feasible. The chitosan molecule contains a large number of reactive functional groups — hydroxyl and amino groups — which determine its sorption, antioxidant and radioprotective properties as well as its biocompatibility and biodegradability [14].

Chitin and chitosan are broken down by microbial enzymes such as chitinase and chitobiase to N-acetyl-D-glucosamine and D-glucosamine, so that they are

environmentally benign materials. The most common medical applications include devices for the treatment of deep wounds and burns, dental and orthopaedic implants, chitosan derivatives as drug-delivery systems, and substrates for tissue-engineering products. Its structure allows chitosan, or chitosan-based composites, to be produced in the form of films and fibres.

Clinical studies of biotransplants developed for the treatment of burns, trophic ulcers and long-non-healing wounds have shown that the resulting coatings adhere tightly to the wound surface and absorb excess exudate; a significant stimulating effect on regenerative processes has been recorded. The mean healing time for grade II–IIIa burns is reduced from 25–30 days to 10–12 days. Chitosan-based nanocomposites are also of importance for regenerative medicine of bone and cartilage: calcium phosphate or calcium carbonate nanoparticles and hydroxyapatite nanofibres are added to increase mechanical strength [14, 15].

4.2. Collagen

Collagen is a glycoprotein and a fibrillar protein that forms the basis of connective tissue — tendon, bone, cartilage, dermis — and provides its strength and elasticity. It is the principal component of connective tissue and the most abundant protein in mammals, constituting from 25% to 45% of total body protein [16].

The collagen molecule is a left-handed helix formed from three α -chains, a structure known as tropocollagen. Each turn of the α -chain helix contains three amino-acid residues. The molecular weight of collagen is approximately 300 kDa, its length 300 nm and its thickness 1.5 nm. Because of the presence of arginine–glycine–aspartate (RGD) sequences in its primary structure, collagen is a biomaterial that supports cell adhesion. It has low allergenicity and is non-toxic; however, its unregulated and rapid biodegradation significantly shortens the service life of collagen products to about one month. When introduced into the body, collagen stimulates reparative processes and promotes the formation of the organism's own collagen, but does not ensure complete organ regeneration because it is resorbed early and forms scar tissue. Formation of a heterogeneous supramolecular structure in collagen-containing gels helps to slow biodegradation. Collagen has haemostatic properties and is therefore used in the manufacture of artificial valves and vessels [16, 17].

4.3. Alginic acid and alginates

Alginic acid is a polysaccharide obtained from algae of the genus *Laminaria*. It is insoluble in water and in most organic solvents. One millilitre of alginic acid can absorb 300 millilitres of water, which makes it an effective thickening agent. Alginic acid is a heteropolymer formed by residues of two uronic acids, D-mannuronic and L-guluronic, in ratios that vary with the algal species [18].

In dentistry, alginates are used as elastic impression materials. Inorganic fillers (zinc oxide, talc) constitute the main part of the alginate powder and affect the viscosity and strength of the material after setting. Alginates are used in systems for the delivery of biologically active substances and in injectable vehicles for the delivery of cells and various factors, because they form hydrogels on interaction with divalent and trivalent cations (Mg^{2+} , Ca^{2+} , Ba^{2+} , Sr^{2+} , Al^{3+} , Fe^{3+}). Alginates do not contain the RGD sequence, so cell adhesion to their surface is relatively poor. Nevertheless, porous alginate sponges promote the restoration of nerve conduction in rats, and alginate matrices stimulate cartilage regeneration [18, 19].

4.4. Silk fibroin

Fibroin is the principal protein of silk, obtained from the cocoons of *Bombyx mori* and related species. Silk fibre consists mainly of two components: the protein fibroin and the water-soluble adhesive protein sericin, which binds fibroin fibres together within the fibre. Sericin is allergenic and provokes an immune reaction, which restricts the use of sericin-contaminated silk in regenerative medicine; methods of removing sericin from silk are therefore an obligatory preliminary stage [20, 21].

Fibroin is a heterodimer consisting of two chains covalently linked by disulfide bridges, a heavy chain (Fib-H) and a light chain (Fib-L). The primary sequence of fibroin consists of glycine (43%), alanine (30%) and serine (12%), with smaller amounts of tyrosine (5%), valine (2%), aspartate, glutamate and cysteine, the last playing the key role in linking the subunits into a single molecule. Glycine, alanine and serine form the principal structural sequence Gly-Ala-Gly-Ala-Gly-Ser of the heavy chain, which constitutes 70% of the total protein sequence. Other sequences include Gly-Ala-Gly-Ala-Gly-Tyr (20%), Gly-Ala-Gly-Tyr-Gly-Ala (6%) and Gly-Ala-Gly-Ala-Gly-Ala (4%); together they form the basis of 12 regular hydrophobic crystalline blocks that make up 95% of the total protein sequence. Each block consists of 413 amino-acid residues, and the blocks are separated

by amorphous intermediate regions of 42–43 residues [20].

This structure gives rise to a uniform secondary structure of antiparallel β -sheets linked by hydrogen bonds, and it is this architecture that provides the properties allowing fibroin to be used as a material for biosynthetic structures. Fibroin is able to pass rapidly from aqueous solution to an insoluble state under the action of alcohols and other agents, which makes it an ideal material for producing durable yet flexible structures. Silk fibroin is also heat-resistant: products made from it can be sterilised at temperatures up to 150 °C.

The decisive property for design purposes is that the biodegradation time of silk fibroin increases with the proportion of β -sheets in its structure. This means that the rate of polymer biodegradation can be regulated by controlling the β -sheet fraction when the product is created. Studies show that fibroin can retain its mechanical properties for up to one year. Silk therefore has a clear advantage over other materials in several

respects related to biodegradation: first, the degradation of some synthetic materials generates acidic by-products that are not included in the metabolic pathways of the organism, whereas fibroin is safe because its biodegradation products are amino acids metabolised by cells; second, some materials cannot retain their mechanical properties for the required period, whereas the slow biodegradation of silk fibroin can be an advantage in tissue engineering. Fibroin is used in regenerative medicine for the production of matrices and films, is incorporated into structures for the delivery of medicinal and biologically active substances, and exhibits antimicrobial activity, so that it can be recommended as a new natural antibacterial biomaterial [20–22].

5. Comparative analysis

Table 1 collects the reported thermal and degradation data in a common format, together with the derived indicators defined in Section 2.

Table 1. Thermal behaviour, degradation and derived indicators of the polymers compared.

Polymer	T _g , °C	T _m , °C	Resorption time	Degradation products	RGD motif
PLA	50–60	160–180	6 months – 2 years	lactic acid (acidic)	no
PGA	35–40	225–230	4–6 months	glycolic → oxalic acid	no
PCL	≈ –60	≈ 60	≈ 3 years	caproic acid, H ₂ O, CO ₂	no
PET	≈ 70	≈ 250	not resorbed	–	no
Chitosan	–	decomposes	weeks – months	glucosamine derivatives	no
Collagen	–	denatures	≈ 1 month	amino acids	yes
Alginate	–	decomposes	weeks (ionic)	uronic acids	no
Silk fibroin	–	stable to 150 °C	weeks – 1 year (tunable)	amino acids	no

Values for T_g and T_m of the natural polymers are omitted where the polymer decomposes or denatures before melting; PET values are given for reference only.

Three patterns emerge from Table 1. First, among the synthetic polyesters the resorption time varies by more than an order of magnitude — from four months for PGA to three years for PCL — but for each individual polymer it is essentially fixed. Adjusting the resorption period therefore requires changing the polymer or forming a copolymer, which simultaneously changes the mechanical and processing behaviour. Second, only two of the eight polymers degrade to products that are

unambiguously benign: PCL and the protein-based polymers. PGA is the clearest counter-example, since its degradation product is metabolised to oxalic acid. Third, only collagen among the natural polymers carries the RGD adhesion motif, which explains why it continues to be used despite its very short service life.

Table 2 expresses the same comparison in terms of the resorption ratio k for three representative applications: a skin wound dressing (healing period taken as 2–4 weeks), a cartilage scaffold (regeneration period 3–6 months) and a bone scaffold (6–12 months).

Table 2. Resorption ratio k (resorption time divided by the typical regeneration period of the target tissue).

Polymer	Skin dressing (2–4 weeks)	Cartilage scaffold (3–6 months)	Bone scaffold (6–12 months)
PLA	$\gg 1$ (persists)	1.3–8	0.5–4
PGA	4–9	0.7–2	0.3–1
PCL	$\gg 1$ (persists)	6–12	3–6
Collagen	1–2 (matched)	0.2–0.3 (too fast)	0.1 (too fast)
Chitosan	1–4	0.2–1	≤ 0.5
Alginate	0.5–2	≤ 0.3	≤ 0.2
Silk fibroin	tunable to ≈ 1	tunable to ≈ 1	tunable to ≈ 1

Table 2 makes the central trade-off explicit. No polymer other than silk fibroin achieves $k \approx 1$ across all three applications: the polyesters are matched to the slower applications and persist unnecessarily in the faster ones, while collagen and alginate are matched to dressings and resorb prematurely in scaffolds. The ability to tune the β -sheet fraction allows silk fibroin to be configured for each case from a single feedstock — which is the

practical meaning of the statement, frequently made but rarely quantified, that its degradation rate is controllable.

Table 3 completes the comparison with the processing and solvent requirements, which determine whether the polymer can be electrospun at all and what residues the product will carry.

Table 3. Processing and solvent requirements.

Polymer	Spinnable alone	Typical solvent system	Residue concern
PLA / PGA / PCL	yes	chloroform, dichloromethane, HFIP	chlorinated/fluorinated residues
PET	yes	TFA/dichloromethane	strong acid residues
Chitosan	poorly; carrier needed	dilute acetic or formic acid	acid residues
Collagen	yes	HFIP, TFE (fluorinated)	denaturation; fluorinated residues
Alginate	poorly; carrier needed	water	none significant
Silk fibroin	no; carrier needed	CaCl ₂ / H ₂ O / ethanol	residual salt (pharmacologically benign)

The cost of the silk fibroin advantage can now be stated quantitatively. Because fibroin solutions obtained in a calcium chloride based system reach concentrations of only about 1.8–3.5 wt%, which is below the entanglement concentration needed for continuous jet formation, a carrier polymer must be blended in. In a representative formulation the carrier (poly(vinyl alcohol)) accounts for 9.6 wt% of the final solution against 1.4 wt% of fibroin, that is a carrier-to-fibroin ratio of about 6.8 : 1, so that fibroin constitutes only some 13% of the polymer in the finished web. Any biological benefit attributed to fibroin must therefore be weighed against this dilution.

6. Discussion

The comparison supports a simple decision rule. Where the dominant requirement is reproducible processing and a defined mechanical performance over a period longer than six months, the synthetic polyesters remain the rational choice, and the acidic degradation products can be managed by buffering additives or by copolymer design. Where the dominant requirement is biological interaction over a short period — a dressing, a transient cosmetic film — collagen, chitosan or alginate are appropriate, and their rapid resorption becomes an advantage rather than a defect. It is only in the intermediate region, where a matched degradation rate must be combined with retained mechanical properties and benign degradation products, that silk fibroin has no direct competitor.

Two qualifications should be attached to this rule. The

first is the carrier dilution described above: a fibroin construct is in practice a fibroin–carrier composite, and the reported biological performance of such constructs cannot be attributed to fibroin alone. The second is that the RGD motif, present only in collagen among the polymers compared, can be introduced into other polymers by surface functionalisation with defined oligopeptides; where this route is available, the absence of a native adhesion motif ceases to be a decisive disadvantage.

The evidence base has clear limitations. Resorption times are collected from studies using different animal models, implantation sites and specimen geometries, and are therefore comparable only to within a factor of about two; the resorption ratios of Table 2 should be read as order-of-magnitude indicators rather than as design values. Glass-transition and melting data for the natural polymers are largely unavailable because these polymers decompose or denature before melting. Finally, the residual-solvent contents that Table 3 identifies as the principal translational concern are almost never reported quantitatively in the primary literature.

7. Conclusions

Eight fibre-forming polymers used in electrospun biomedical constructs were compared within a single framework covering thermal behaviour, degradation, biological interaction and solvent requirements, and three derived indicators were introduced to make the comparison usable for design decisions.

The synthetic polyesters offer a wide and reproducible processing window but a degradation rate that is fixed for each polymer and, in the case of PLA and PGA, acidic degradation products. The natural polymers offer favourable biological interaction and metabolisable degradation products, but collagen and alginate resorb prematurely for scaffold applications and chitosan and alginate cannot be spun without a carrier.

Expressed through the resorption ratio k , no polymer except silk fibroin achieves a matched degradation rate across dressing, cartilage and bone applications; the tunability of the β -sheet fraction allows a single fibroin feedstock to be configured for each. The quantified cost of this advantage is the carrier fraction required for stable jet formation — a carrier-to-fibroin ratio of about 6.8 : 1 in a representative formulation, corresponding to roughly 13% fibroin in the polymer of the finished web.

The principal gaps in the evidence base are the incomparability of reported resorption times across models and geometries, the absence of thermal data for the natural polymers, and the almost complete absence of quantitative residual-solvent reporting.

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