TENDON FASCICLE-INSPIRED COLLAGEN MULTIFILAMENT BUNDLE PRODUCED BY MULTI-PIN CONTACT DRAWING OF AN AQUEOUS COLLAGEN-POLYETHYLENE OXIDE SOLUTION

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ABSTRACT

Polyethylene oxide (PEO)-collagen monofilaments are prepared using multi-pin contact drawing, which is a bio-friendly and cost-effective method that uses an entangled polymer fluid to aid in monofilament formation[1]. Using this bottom-up approach, thousands of monofilaments containing well-aligned collagen triple helices are assembled into a structure that recapitulates the architecture of a tendon fascicle. In this approach, a series of hydration steps are applied to remove the PEO, and promote the self-assembly of the collagen triple helices into fibrils within each monofilament, while preserving the hierarchical structure of the collagen multifilament bundle. Attenuated total reflectance Fourier-transform infrared (ATR-FTIR) spectroscopy and wide-angle X-ray scattering (WAXS) are used to confirm the presence and alignment of collagen triple helices in the resulting collagen multifilament bundle. Scanning electron microscopy (SEM) reveals that each monofilament contains collagen fibrils with a diameter around 15 nm. Small angle X-ray scattering (SAXS) patterns are consistent with fibrils containing microfibrils, with the characteristic D-band spacing observed *in vivo*. However, these microfibrils are axially staggered by one-sixth of the D-band spacing. This is a well-known polymorphic form of collagen that has been observed *in vitro*[2], albeit at significantly lower collagen concentration than what is used here. These polymorphic collagen fibrils can be effectively crosslinked with ultraviolet (UV) radiation in the dry state giving rise to collagen multifilament bundles with an average ultimate tensile strength (UTS) of 38.5 MPa, which is comparable to that of wet spun, glutaraldehyde crosslinked collagen multifilaments (40 MPa)[3], and also to that of rat tail tendon fibers (39 MPa)[4]. The effect of UV crosslinking on the chemical and molecular structure of collagen multifilament bundles is also demonstrated through ATR-FTIR spectroscopy and WAXS, which shows that the triple-helical structure of the collagen molecule remains unchanged. This fabrication method leverages extensional flow for molecular alignment, decouples the molecular alignment from the molecular assembly, recapitulates the structure of a tendon and offers tunability in tensile properties, using only

collagen molecules and no other chemical additives in addition to PEO, which is removed during the hydration process.

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