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Coaxial electrospun aligned tussah silk fibroin nanostructured fiber scaffolds embedded with hydroxyapatite–tussah silk fibroin nanoparticles for bone tissue engineering

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ABSTRACT

The bone is a composite of inorganic and organic materials and possesses a complex hierarchical architecture consisting of mineralized fibrils formed by collagen molecules and coated with oriented hydroxyapatite. To regenerate bone tissue, it is necessary to provide a scaffold that mimics the architecture of the extracellular matrix in native bone. Here, we describe one such scaffold, a nanostructured composite with a core made of a composite of hydroxyapatite and tussah silk fibroin. The core is encased in a shell of tussah silk fibroin. The composite fibers were fabricated by coaxial electrospinning using green water solvent and were characterized using different techniques. In comparison to nanofibers of pure tussah silk, composite notably improved mechanical properties, with 90-fold and 2-fold higher initial modulus and breaking stress, respectively, obtained. Osteoblast-like MG-63 cells were cultivated on the composite to assess its suitability as a scaffold for bone tissue engineering. We found that the fiber scaffold supported cell adhesion and proliferation and functionally promoted alkaline phosphatase and mineral deposition relevant for biomineralization. In addition, the composite were more biocompatible than pure tussah silk fibroin or cover slip. Thus, the nanostructured composite has excellent biomimetic and mechanical properties and is a potential biocompatible scaffold for bone tissue engineering.

1. Introduction

Natural bone forms according to a hierarchical architecture, components of which span nanometer to macroscopic scales. It is produced by natural biomineralization, in which organic templates control the growth of the inorganic phase [1]. The fundamental subunits of bone are mineralized collagen fibrils that consist of self-assembled triple helices of collagen molecules [2–4]. Hydroxyapatite nanocrystals grow on these fibrils with the crystallographic *c*-axis aligned with the axis of the fibril [5].

One strategy to repair or replace bone is to engineer bone tissue through a combination of scaffolds, implanted cells, and biologically active molecules [6–7]. An ideal bone scaffold should be biocompatible, biodegradable, and mechanically robust. This scaffold should facilitate early mineralization and support formation of new bone, while simultaneously allowing replacement of old tissue [8–9]. In other words, the scaffold should have physical architecture and chemical composition similar to that of natural bone.

Silk fibroin has gained significant interest as a scaffold due to excellent biocompatibility and biodegradability. Indeed, various strategies, including solvent casting, freeze drying, and salt leaching, have been developed to create three-dimensional silk scaffolds with high porosity and osteoconductivity. Ceramic components such as hydroxyapatite have been incorporated into these scaffolds to mimic the extracellular matrix [10–12]. To further enhance cell-scaffold interaction, nanofibers of silk fibroin have been used as alternatives to three-dimensional structures, because they more closely resemble the natural extracellular matrix and are also highly porous and interconnected [13–15].

Several approaches, including blending electrospinning and biomimetic mineralization, have been developed to fabricate such silk fibroin nanofibers, usually as a composite with hydroxyapatite, to better imitate natural bone [16–17]. For instance, Kim et al. [18] fabricated composite nanofibers using blending spinning and alternate soaking. Notably, the composite has better mechanical properties than similar scaffolds built from pure silk fibroin. Incorporation of hydroxyapatite into these composites further enhances specific biological activities

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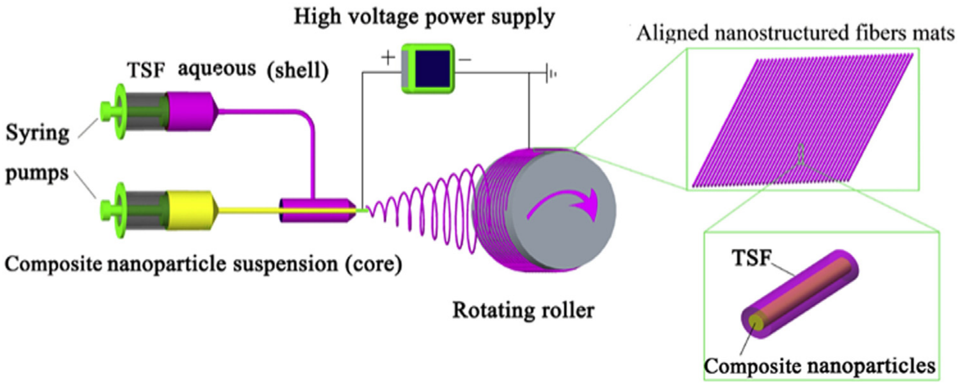

Fig. 1. Schematic diagram of the set-up of coaxial electrospinning.

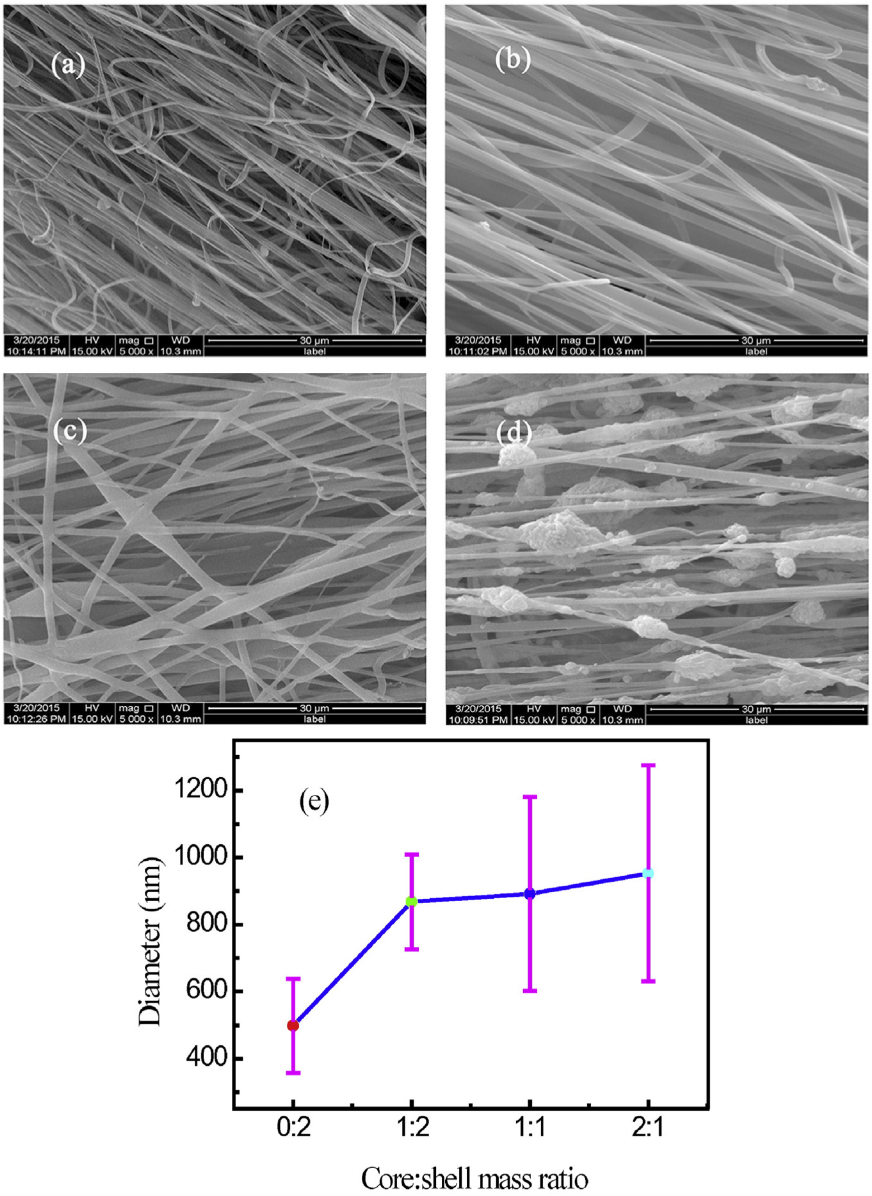

Fig. 2.

Fig. 2. SEM images (a–d) and diameter distribution (e) of nanostructured fibers with different core:shell mass ratios. (a) pure TSF; (b) 1:2; (c) 1:1; (d) 2:1.