Review of Electrospun Materials from Natural Polymers: Collagen, Keratin and Gelatin

Evelina BRASTAVIČIŪTĖ *, Erika ADOMAVIČIŪTĖ*

**** Kaunas University of Technology, Faculty of Mechanical Engineering and Design, Studentu str. 56, LT-51424 Kaunas, Lithuania, Textile and Clothing Technology, E-mail: evelina.brastaviciute@ktu.edu***

Introduction

Electrospinning is a process which can fabricate ultra-fine fibres with diameters from nanometre to sub-micrometre range. Electrospun materials have unique characteristics, such as high surface area to volume or mass ratio, high density of pores. Because of these unique characteristics of electrospun fibres, they are excellent candidates for various biomedical applications, for example, they can be used as scaffolds for cell/tissue culture, vascular and nerve grafts, carriers for topical/transdermal delivery of drugs [1]. Electrospinning of natural biopolymers had become an important topic because of their excellent biocompatibility, high porosity, better suitability for the human body when comparing to synthetic polymers [2]. In this paper, electrospinning of three natural polymers: keratin, collagen, and gelatine is described.

Electrospinning of keratin

- G. Guidotti and co-authors [4] for the first-time blended keratin with Poly (butylene succinate) (PBS), using 1,1,1,3,3,3-hexafluoro-2-propanol (HFIP) as solvent and loaded with diclofenac sodium salt. By mixing with PBS, keratins mechanical properties were improved, Ker-PBS 50-50 showed a 10 times higher permeation of Diclofenac compared to PBS. Also, G. Guidotti and co-authors [5] spun spun keratin/PBS nanofibers, loaded with rhodamine B (RhB), a model drug, that is easily detectible. Nanofibers, that had more keratin showed a faster release of drug [5].
- M. He and others [3] produced antibacterial composite nanofibers from feather keratin (FK), poly (vinyl alcohol) (PVA), poly (ethylene oxide) (PEO) with incorporated silver nanoparticles (AgNPs). Incorporated AgNPs caused antibacterial activity, the nanofibers showed significant inhibition against E. coli and S. aureus [3].
- X. Wan and co-authors [6] spun PCL/keratin/AuNPs mats with the ability to catalytically generate nitric oxide. When spun with the AuNPs, the release of NO was significantly higher. Also, PCL/keratin/AuNPs mats had the highest cell viability [6].
- H. Zhang an others [7] spun PLGA/wool keratin membranes loaded with antibacterial agent ornidazole (ORN). 1% ORN composite membrane had strong water absorption, mechanical properties, showed suitable drug release and suitable in vitro degradation, antibacterial effects [7].

Electrospinning of gelatin

- S. Ö. Gönen and others [14] electrospun gelatine (Gt) and PCL nanofibers using less toxic and relatively cheaper solvent system of acetic acid and formic acid [14].
- Y. Wang and co-authors[2] spun composite gelatine and pullulan nanofibers. When the solution concentrations were of 20% and 25% w/v produced fibres were bead-free and uniform [2].
- M. A. Al-Baadani and others [15] spun (PCL)/gelatin membranes for bone tissue regeneration. It was determined that PCL had enough mechanical strength to maintain the integrity of the membrane as the new bone forms, while the gelatin enhances the biocompatibility. The membranes could be used as drug carriers, that have tunable drug release, from initial rapid release to prolonged release [15].
- R. Zhang and co-authors [16] electrospun silk fibroin and gelatin nanofibers, modified with graphene oxide-sliver nanoparticles. Films modified with GO-AgNPs had an inhibition effect on E. coli growth [16].
- J. Dulnik and others [17] spun PCL/gelatine and PCL/collagen nanofibers with two different solvents: HFIP and a mixture of acetic (AA) and formic (FA) acids. The samples where not cytotoxic, slightly higher viability was of samples containing polypeptides [17].

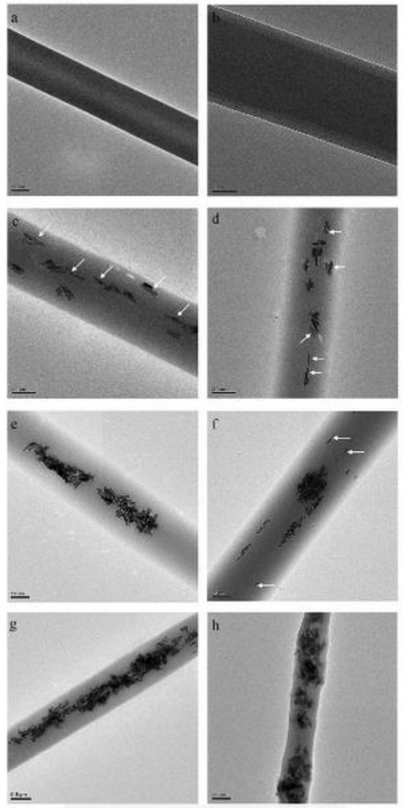

Fig. 1 TEM images of collagen and hydroxyapatite (HA) fibers

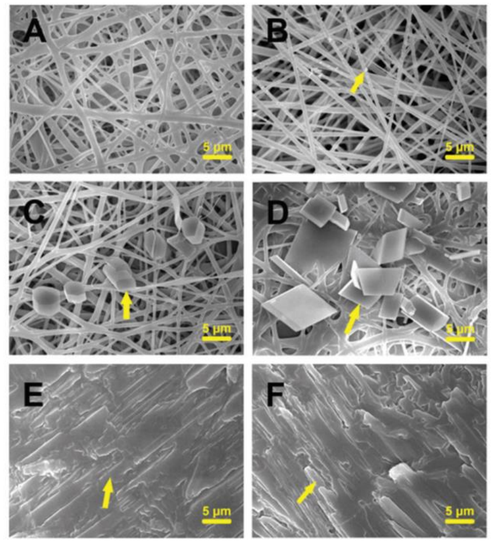

Fig. 2 SEM images of ornidazole-loaded PLGA/wool keratin composite membranes

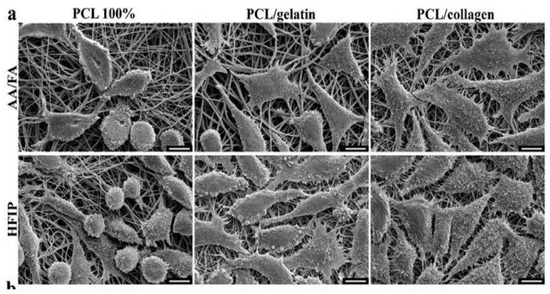

Fig. 3 SEM images of L929 cells cultured for 5 days on the surface of PCL/gelatine and PCL/collagen nanofibers materials

Conclusions

- Electrospun natural polymers: keratin, collagen and gelatine can be utilized for different biomedical applications, such as drug delivery, antibacterial mats, wound dressings, tissue engineering.
- Additional polymers, nanoparticles, solvents can affect the electrospinning process, morphology, mechanical strength, antibacterial activity of these polymers' nanofibers.
- Adding AgNP antibacterial activity of electrospun keratin was increased, PBS improves keratins mechanical properties and release of Diclofenac, AuNPs and PCL made the release of Nitric oxide and cell viability better, PLGA/wool keratin nanofibers loaded with ornidazole have antibacterial properties.
- Incorporation of HA improves collagens mechanical properties, spinning with acetic acid partially preserves collagen's structure.
- Mixture of acetic acid and formic acid is less toxic for spinning gelatine and can improve cell viability, certain pullulan concentrations can improve fibres morphology, PCL and gelatine nanofibers can be used to carry drugs, modification with GO-AgNPs can provide antibacterial properties.

Electrospinning of collagen

- Ji J. and co-authors [10] produced nanofibers of collagen and hydroxyapatite (HA). It was determined that HA significantly improved mechanical properties of collagen nanofibers: elongation and strength [10].
- D. A. Casadiego-Castilla et. al. [9] spun collagen scaffolds using different solvents. Non-oriented fibers with a preserved structure were spun using acetic acid as a solvent at a solution concentration of 20% (v / w) [9].
- J. Bürck and co-authors [13] spun type I collagen nanofibers using two different solvents: acetic acid (99%) and 2,2,2-trifluoroethanol (TFE) (≥ 99%). Collagen nanofibers spun with acetic acid partially preserved the triple helix structure [13].

References

1. Songchotikunpan, P. et al. (2008). Extraction and electrospinning of gelatin from fish skin. *International Journal of Biological Macromolecules*, Vol. 42, no. 3, p. 247–255.
2. Wang, Y. et al. (2019). Study on the Electrospinning of Gelatin/Pullulan Composite Nanofibers. *Polymers*, Vol. 11, no. 9, p. 1424.
3. He, M. et al. (2020). Electrospun Silver Nanoparticles-Embedded Feather Keratin/Poly (vinyl alcohol) /Poly (ethylene oxide) Antibacterial Composite Nanofibers. *Polymers*, Vol. 12, no. 2, p. 305.
4. Guidotti, G. et al. (2020). Regenerated wool keratin-polybutylene succinate nanofibrous mats for drug delivery and cells culture. *Polymer Degradation and Stability*, Vol. 179, p. 109272.
5. Guidotti, G., et al. (2021). Effects of the Blending Ratio on the Design of Keratin/Poly (Butylene Succinate) Nanofibers for Drug Delivery Applications. *Biomolecules (Basel, Switzerland)*, vol. 11, no. 8, p. 1194.
6. Wan, X. et al. (2018). Electrospun PCL/keratin/AuNPs mats with the catalytic generation of nitric oxide for potential of vascular tissue engineering. *Journal of biomedical materials research. Part A*, Vol. 106, no. 12, p. 3239–3247.
7. Zhang, H., et al. (2020). Construction and Characterization of Antibacterial PLGA/Wool Keratin/Ornidazole Composite Membranes for Periodontal Guided Tissue Regeneration. *Journal of Biomaterials Applications*, Vol. 34, p. 1267–1281.
8. Kazanci, M. et al. (2021). Synchrotron Fourier transform infrared microspectroscopy (sFTIRM) analysis of unfolding behavior of electrospun collagen nanofibers. *Spectrochimica acta. Part A, Molecular and biomolecular spectroscopy*, Vol. 251, p. 119420.
9. Castilla-Casadiego, D.A. et al. (2016). Engineering of a Stable Collagen Nanofibrous Scaffold with Tunable Fiber Diameter, Alignment, and Mechanical Properties. *Macromolecular Materials and Engineering*, Vol. 301, 9, p. 1064.
10. Ji, J. et al. (2012). Mechanics of electrospun collagen and hydroxyapatite/collagen nanofibers. *Journal of the mechanical behavior of biomedical materials*, Vol. 13, p. 185–193.
11. Khadka, D. B. et al. (2012). Protein- and peptide-based electrospun nanofibers in medical biomaterials. *Nanomedicine*, Vol. 8, no. 8, p. 1242–1262.
12. Law, J. et al. (2017). Electrospun Collagen Nanofibers and Their Applications in Skin Tissue Engineering. *Tissue Engineering and Regenerative Medicine*, Vol. 14, no. 6, p. 699–718.
13. Bürck, J. et al. (2018). Observation of triple helix motif on electrospun collagen nanofibers and its effect on the physical and structural properties. *Journal of molecular structure*, Vol. 1151, p. 73–80.
14. Gönen, S.Ö. et al. (2015). Effects of Electrospinning Parameters on Gelatin/Poly(ε-Caprolactone) Nanofiber Diameter. *Chemical engineering & technology*, Vol. 38, no. 5, p. 844–850.
15. Al-Baadani, M.A., et al. (2021). Co-Electrospinning Polycaprolactone/Gelatin Membrane as a Tunable Drug Delivery System for Bone Tissue Regeneration. *Materials & Design*, vol. 209. p. 109962.
16. Zhang, R., et al. (2019). High Antibacterial Performance of Electrospinning Silk Fibroin/Gelatin Film Modified with Graphene Oxide-sliver Nanoparticles. *Journal of Applied Polymer Science*, vol. 136, no. 35, p. 47904.
17. Dulnik, J. et al. (2018). The effect of a solvent on cellular response to PCL/gelatin and PCL/collagen electrospun nanofibres. *European polymer journal*, Vol. 104, p. 147–156.