



Why Natural-Based Bioactive Coatings?

Arantzazu Santamaria-Echart ^{1,2,*}  and Filomena Barreiro ^{1,2} 

¹ Centro de Investigação de Montanha (CIMO), Instituto Politécnico de Bragança, Campus de Santa Apolónia, 5300-253 Bragança, Portugal

² Laboratório Associado para a Sustentabilidade e Tecnologia em Regiões de Montanha (SusTEC), Instituto Politécnico de Bragança, Campus de Santa Apolónia, 5300-253 Bragança, Portugal

* Correspondence: asantamaria@ipb.pt

The design of bioactive coatings through combining green strategies and natural formulations prevails over conventional synthetic systems. These approaches address the need to provide biofunctional properties through alternative solutions. Green strategies include methodologies that involve lower environmental impact, namely by using green solvents and reagents, or low-cost and low-toxic processes, promoting an optimized life assessment of the product, among other features. It is important to note that most recent methodologies (e.g., 3D printing, electrospinning, layer-by-layer deposition, or dip coating) enable the design of novel and tailor-made structures, whose assemblies/morphology can be essential to guarantee the formulation's functionality [1].

Natural formulations imply using natural polymers to replace total or partial use of petrochemical polymers. Chitin, chitosan, collagen, silk fibroin, gelatin, cellulose and cellulose-derivatives, alginate, starch, pectin, xanthan gum, pullulan, or proteins are some highlightable natural polymers used as bioactive coating matrices [2,3]. Concurrently, formulations with added natural-derived functionalities are increasingly positioned as an attractive pathway to functionalize coatings. The intrinsic properties of these natural compounds, e.g., antioxidant, antibacterial, or anti-inflammatory capacities, can be transferred to the coatings, providing functionalities to the entire system [4].

In this context, bioactives of a diverse nature can be incorporated, attending to the final application. For example, natural extracts, even obtained from agro-industrial residues, have been reported to be attractive candidates in the food industry to produce edible coatings, which are aimed at extending the lifespan of foods. De Carli et al. [5] produced chitosan films from chitin extracted from waste crayfish (*Procambarus clarkii*) shells. These bioactive films were added with propolis bioactive extract, aiming at acting as edible protective coatings in fruits. Aloui et al. [6] developed locust bean gum-based coatings with added cutin monomers and/or cuticular wax from tomatoes to be applied as a postharvest preservation strategy for cherry tomatoes. The results showed the potential of the coatings to delay the respiration rate and fungal decay of tomatoes, while controlling the weight loss and retaining their firmness and antioxidant capacity in cold storage.

Similarly, pectin, hydroxylpropylmethylcellulose, and methylcellulose were employed by Settler-Ramírez et al. [7] to prepare coatings with yeast extract and apple pomace. Apples were coated with the most promising formulations, and the antifungal activity was analyzed during storage against *Penicillium expansum*, particularly the prevention from patulin, the mycotoxin produced by the fungi. Concurrently, the production of bioactive coatings has also been reported for the preservation of other types of foods, including cheese preservation with coatings based on alginate that incorporates bacteriocin-producing lactic acid bacteria [8] and fish products with guar gum-based coatings that contain thyme oil [9] or chitosan coatings with pomegranate peel extract [10].

In the field of biomedicine, the incorporation of growth factors or bioactive compounds can mimic the composition of specific parts of the body. This is the case of hydroxyapatite,



Citation: Santamaria-Echart, A.; Barreiro, F. Why Natural-Based Bioactive Coatings? *Coatings* **2022**, *12*, 1426. <https://doi.org/10.3390/coatings12101426>

Received: 23 June 2022

Accepted: 20 September 2022

Published: 29 September 2022

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

the principal mineral in the body's hard tissues, which is reportedly used in the preparation of bioactive coatings to promote cell proliferation, especially in bone and dental applications [11,12]. Other compounds have been reported to hold diverse roles in the preparation of biomedical coatings, as is the case of polydopamine. Some works reported the potential of polydopamine-based bioinspired coatings to control the release of bioactive compounds (e.g., silver ions and bovine serum albumin) in titanium-based implants [13]. Alternatively, Sharma et al. [14] reported the potential of polydopamine to functionalize polyvinylidene fluoride coatings as piezo-active biomaterials. They demonstrated the potential of coating deposition to tailor the biomineralization and bioactive properties, promoting the regeneration of musculoskeletal damaged areas.

The family of growth factors is also considered as essential compounds in tissue engineering. For example, several works have demonstrated the potential of adding growth factors into bioactive coatings to promote bone healing. Different growth factors, including morphogenic proteins, fibroblast growth factors, and vascular endothelial growth factors, were reported to show promising results for osteoconduction and osteoinduction, favoring bone healing processes [15].

In agriculture, microorganisms or biocatalysts are added to the coatings to promote soil fertilization. Weiß et al. [16] developed soil improver bio-based and biocompatible coatings based on lignosulfonates biocatalyzed by laccase and plasticized with glycerol, xylitol, or sorbitol. The spray-coated formulations were added with plant growth promoter microorganisms (*Bacillus* species) and tested in diverse model plants, including corn, wheat, tomato, and salad and demonstrated no toxic effects on their germination and growth process. Majaron et al. [17] designed a novel approach to improve the availability of unprocessed (raw) nutrient sources (e.g., elemental sulfur and mineral oxides) to plants, aiming at promoting sustainable agriculture as an alternative to conventional fertilizers. The coatings were prepared with maize starch and the unprocessed nutrient sources with a microbial source (*Aspergillus niger* or *Acidithiobacillus thiooxidans*), which is responsible for ensuring that the nutrients are accessible to the plants. The acidification effect of the microorganisms improved the oxide and elemental sulfur oxidation, validating the potential of these biocoatings as nutrient suppliers' co-adjutants.

It is worth noting that in the aforementioned application fields, the activity of the coating solution does not only depend on the polymer and bioactive compounds, but it is also influenced by the coating preparation strategy and its final structure. The surface activation of hydrophobic coatings through the high-frequency plasma at low pressure in a nitrogen atmosphere was presented as a strategy to improve the adhesion and reactivity of the side chains of poly (lactic acid)-based coatings [18]. Other tailoring strategies comprise the selection of specific and advanced coating preparation techniques to modulate the bioactive profile of the coatings. Coatings produced by electrospinning can tailor the biocompatibility and antimicrobial activity. In contrast, dip coatings can promote mechanical and adhesion properties, and layer-by-layer deposition enables the design of specific drug release profiles [1].

Overall, when asked the question "Why natural-based bioactive coatings?", one must consider the broad possibilities (in terms of formulation and preparation strategies) offered by the bioactive coatings to improve and modulate the bioactivity of various products and their environmental compatible fabrication strategies.

Funding: The authors are grateful to the Foundation for Science and Technology (FCT, Portugal) for financial support through national funds FCT/MCTES (PIDDAC) to CIMO ((UIDB/00690/2020 and UIDP/00690/2020)) and SusTEC (LA/P/0007/2021). National funding by FCT, through the institutional scientific employment program-contract with Arantzazu Santamaria-Echart.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Kravanja, K.A.; Finšgar, M. A review of techniques for the application of bioactive coatings on metal-based implants to achieve controlled release of active ingredients. *Mater. Des.* **2022**, *217*, 110653. [[CrossRef](#)]
2. Iñiguez-Moreno, M.; Ragazzo-Sánchez, J.A.; Calderón-Santoyo, M. An extensive review of natural polymers used as coatings for postharvest shelf-life extension: Trends and challenges. *Polymers* **2021**, *13*, 3271. [[CrossRef](#)] [[PubMed](#)]
3. Guo, L.; Liang, Z.; Yang, L.; Du, W.; Yu, T.; Tang, H.; Li, C.; Qiu, H. The role of natural polymers in bone tissue engineering. *J. Control. Release* **2021**, *338*, 571–582. [[CrossRef](#)] [[PubMed](#)]
4. Kravanja, K.A.; Finšgar, M.; Knez, Z.; Knez Marevci, M. Evaluation of natural extracts as promising components of bioactive coatings for orthopedic implants. *Front. Mater.* **2022**, *9*, 878176. [[CrossRef](#)]
5. De Carli, C.; Aylanc, V.; Mouffok, K.M.; Santamaria-Echart, A.; Barreiro, F.; Tomás, A.; Pereira, C.; Rodrigues, P.; Vilas-Boas, M.; Falcão, S.I. Production of chitosan-based biodegradable active films using bio-waste enriched with polyphenol propolis extract envisaging food packaging applications. *Int. J. Biol. Macromol.* **2022**, *213*, 486–497. [[CrossRef](#)] [[PubMed](#)]
6. Aloui, H.; Ghazouani, Z.; Khwaldia, K. Bioactive coatings enriched with cuticle components from tomato wastes for cherry tomatoes preservation. *Waste Biomass Valorization* **2021**, *12*, 6155–6163. [[CrossRef](#)]
7. Settler-Ramírez, L.; López-Carballo, G.; Hernández-Muñoz, P.; Fontana-Tachon, A.; Strub, C.; Schorr-Galindo, S. Apple-based coatings incorporated with wild apple isolated yeast to reduce *Penicillium expansum* postharvest decay of apples. *Postharvest Biol. Technol.* **2022**, *185*, 111805. [[CrossRef](#)]
8. Silva, S.P.M.; Ribeiro, S.C.; Teixeira, J.A.; Silva, C.C.G. Application of an alginate-based edible coating with bacteriocin-producing *Lactococcus* strains in fresh cheese preservation. *LWT* **2022**, *153*, 112486. [[CrossRef](#)]
9. Ruelas-Chacon, X.; Aguilar-González, A.; de la Luz Reyes-Vega, M.; Peralta-Rodríguez, R.D.; Corona-Flores, J.; Reboloso-Padilla, O.N.; Aguilera-Carbo, A.F. Bioactive protecting coating of guar gum with thyme oil to extend shelf life of tilapia (*Oreochromis niloticus*) fillets. *Polymers* **2020**, *12*, 3019. [[CrossRef](#)] [[PubMed](#)]
10. Yu, D.; Zhao, W.; Dong, J.; Zang, J.; Regenstein, J.M.; Jiang, Q.; Xia, W. Multifunctional bioactive coatings based on water-soluble chitosan with pomegranate peel extract for fish flesh preservation. *Food Chem.* **2022**, *374*, 131619. [[CrossRef](#)] [[PubMed](#)]
11. Liu, Z.; Kataoka, T.; Samitsu, S.; Kawagoe, D.; Tagaya, M. Nanostructural control of transparent hydroxyapatite nanoparticle films using a citric acid coordination technique. *J. Mater. Chem. B* **2022**, *10*, 396–405. [[CrossRef](#)] [[PubMed](#)]
12. Singh, J.; Chatha, S.S.; Singh, H. Microstructural and in-vitro characteristics of functional calcium silicate topcoat on hydroxyapatite coating for bio-implant applications. *Prog. Biomater.* **2022**, *11*, 95–108. [[CrossRef](#)] [[PubMed](#)]
13. Xu, K.; Zhou, M.; Chen, W.; Zhu, Y.; Wang, X.; Zhang, Y.; Zhang, Q. Bioinspired polydopamine/graphene oxide/collagen nanofilms as a controlled release carrier of bioactive substances. *Chem. Eng. J.* **2021**, *405*, 126930. [[CrossRef](#)]
14. Sharma, V.; Chowdhury, S.; Bose, S.; Basu, B. Polydopamine codoped BaTiO₃-functionalized polyvinylidene fluoride coating as a piezo-biomaterial platform for an enhanced cellular response and bioactivity. *ACS Biomater. Sci. Eng.* **2022**, *8*, 170–184. [[CrossRef](#)] [[PubMed](#)]
15. Bjelić, D.; Finšgar, M. The role of growth factors in bioactive coatings. *Pharmaceutics* **2021**, *13*, 1083. [[CrossRef](#)] [[PubMed](#)]
16. Weiß, R.; Gritsch, S.; Brader, G.; Nikolic, B.; Spiller, M.; Santolin, J.; Weber, H.K.; Schwaiger, N.; Pluchon, S.; Dietel, K.; et al. A biobased, bioactive, low CO₂ impact coating for soil improvers. *Green Chem.* **2021**, *23*, 6501–6514. [[CrossRef](#)]
17. Majaron, V.F.; da Silva, M.G.; Bortoletto-Santos, R.; Klaic, R.; Ribeiro, S.J.L.; Polito, W.L.; Bevilacqua, D.; Farinas, C.S.; Ribeiro, C. Bioactive material with microorganisms can enhance the micronutrients solubilization and sulfate availability from low reactive sources: Insight for application as coating fertilizer granules. *J. Polym. Environ.* **2022**, *30*, 2602–2613. [[CrossRef](#)]
18. Stoleru, E.; Vasile, C.; Irimia, A.; Brebu, M. Towards a bioactive food packaging: Poly (lactic acid) surface functionalized by chitosan coating embedding clove and argan oils. *Molecules* **2021**, *26*, 4500. [[CrossRef](#)] [[PubMed](#)]