



Abstract Multifunctional Sintering-Free Composite Scaffolds Developed by Additive Manufacturing [†]

Nilza Ribeiro¹, Paula M. C. Torres¹, Tânia S. S. Carvalho¹, João Horta², João P. Araújo² and Susana M. Olhero^{1,*}

- ¹ Department of Materials and Ceramic Engineering (DEMaC), CICECO-Aveiro Institute of Materials, University of Aveiro, 3810-193 Aveiro, Portugal; nilza.ribeiro@ua.pt (N.R.); ptorres@ua.pt (P.M.C.T.); tssc@ua.pt (T.S.S.C.)
- ² Institute of Physics of Advanced Materials, Nanotechnology and Photonics (IFIMUP), Department of Physics and Astronomy, University of Porto, 4169-007 Porto, Portugal; jhbelo@ua.pt (J.H.); jearaujo@fc.up.pt (J.P.A.)
- * Correspondence: susana.olhero@ua.pt
- + Presented at the Materiais 2022, Marinha Grande, Portugal, 10–13 April 2022.

Keywords: multifunctional composite scaffolds; additive manufacturing; direct write assembly/ robocasting; calcium phosphates; natural polymers; chitosan; silk fibroin

Robocasting (also called direct-write assembling) is an additive manufacturing technology based on the extrusion and deposition of an ink, layer by layer, that has been explored for the manufacture of architectural and compositional 3D porous structures with high precision and reproducibility for bone tissue regeneration. The mechanical integrity of the bioceramic-based scaffolds is commonly obtained by sintering (heat treatment usually above 1000 °C) which precludes the incorporation of bioactive molecules before this step.

The main goal of the present work was the manufacture of multifunctional composite scaffolds by robocasting, suppressing sintering as a post-printing process. The abolition of this step leads to the design of inks comprised by calcium-phosphate-based particles, drugs and/or magnetic particles follow-on in scaffolds with multiple therapy functions as local drug delivery, cancer treatment by hyperthermia and bone tissue regeneration. Natural polymeric aqueous-based solutions of chitosan and/or chitosan/silk fibroin with different ratios were used as a base matrix. The multifunctional inks were characterized by rheological studies in viscometry and oscillatory modes, these being the printable ones selected to produce scaffolds with different macropore sizes (300 μ m and 500 μ m). The inks allowed the fabrication of customizable 3D structures with interconnected pores, able to carrier drugs and/or to respond to an external magnetic field. The mechanical strength and rigidity were achieved through the crosslinking of the system and the amounts of bioceramics in the composition. The scaffolds were characterized by mechanical performance, magnetic behavior, morphological features, degradation rate and biological studies.

The developed sintering-free composite scaffolds represent a strong potential to be applied as bone substitutes. Furthermore, the advances of the evaluated work are opening promising paths for future progresses in materials science and the tissue engineering area.

Author Contributions: Conceptualization: S.M.O., N.R. and P.M.C.T.; methodology: S.M.O., N.R., T.S.S.C., P.M.C.T. and J.H.; software: N.R., T.S.S.C. and J.H.; validation: S.M.O., N.R., T.S.S.C. and J.H.; formal analysis: S.M.O., N.R., T.S.S.C., P.M.C.T. and J.H.; investigation: S.M.O., N.R., T.S.S.C., P.M.C.T. and J.H.; resources: S.M.O. and P.M.C.T.; data curation: S.M.O., N.R., T.S.S.C., P.M.C.T. and J.H.; writing—original draft preparation: S.M.O. and N.R.; writing—review and editing: S.M.O., N.R., T.S.S.C., P.M.C.T., S.S.C., P.M.C.T. and J.H.; writing—original draft preparation: S.M.O. and N.R.; writing—review and editing: S.M.O., N.R., T.S.S.C., P.M.C.T., J.H. and J.P.A.; visualization: S.M.O. and N.R.; supervision: S.M.O. and J.P.A.; project administration: S.M.O.; funding acquisition: S.M.O. and P.M.C.T. All authors have read and agreed to the published version of the manuscript.



Citation: Ribeiro, N.; Torres, P.M.C.; Carvalho, T.S.S.; Horta, J.; Araújo, J.P.; Olhero, S.M. Multifunctional Sintering-Free Composite Scaffolds Developed by Additive Manufacturing. *Mater. Proc.* 2022, *8*, 104. https://doi.org/10.3390/ materproc2022008104

Academic Editors: Geoffrey Mitch-ell, Nuno Alves, Carla Moura and Joana Coutinho

Published: 22 June 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/). **Funding:** This work is funded by FEDER funds through the COMPETE 2020 Programme and National Funds through FCT-Portuguese Foundation for Science and Technology under the projects 2BBone and FlexMicroDerm with references POCI-01-0145-FEDER-029940 (PTDC/CTM-CER/29940/2017) and POCI-01-0145-FEDER-029274 (PTDC/BTM-MAT/29274/2017), respectively. This work was also developed within the scope of the project CICECO-Aveiro Institute of Materials, UIDB/50011/2020, UIDP/50011/2020 and LA/P/0006/2020, financed by national funds through the FCT/MEC (PID-DAC). P. M. C. Torres and S. M. Olhero acknowledge FCT for CEECIND/01891/2017 and CEECIND/03393/2017 contracts, respectively.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available on request from the corresponding authors.

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