

3D Printed Gellan/Gelatin Scaffolds Reinforced with Cellulose Nanofibrils and Graphene Derivatives for Bone Tissue Engineering Applications

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The growing demand for efficient solutions regarding bone substitution has led to significant advances in fabrication technologies, with a particular emphasis on 3D printing, enabling the development of scaffolds tailored for the personalized needs of each patient. Despite technological progress, printable formulations derived from natural polymers typically exhibit limitations in terms of printing fidelity, structural integrity, mechanical properties, and mineralization stimulation [1-3]. To address these challenges innovative inks employing a gellan gum/gelatin polymeric mixture, enhanced with different reinforcement agents, including cellulose nanofibrils (CNF) and various functionalized graphene derivatives, such as aminated reduced graphene oxide (NH₂-rGO), carboxylated graphene oxide (COOH-GO), and a combination of both were designed (Figure 1). Based on aforementioned formulations, scaffolds for bone substitution were successfully fabricated with high shape fidelity employing extrusion-based 3D printing, followed by a double cross-linking procedure, with calcium chloride and genipin. All structures were freeze-dried and evaluated from a structural, morphological, mechanical, and biological point of view.

Structures reinforced with NH₂-rGO presented enhanced biological properties, while the ones reinforced with COOH-GO exhibited improved tensile and compressive strength. Our findings indicated that scaffolds comprising both types of graphene derivatives demonstrated an overall superior performance with highest printing fidelity, enhanced shape retention, increased structural reproducibility, adequate degradation rate and swelling kinetics, improved compressive and tensile strength, biocompatibility, and highest mineralization capacity. Therefore, our results indicated a combined effect of the two graphene derivatives, providing the most promising candidate for bone tissue engineering applications.

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References

- [1] Elena Chiticaru, Mariana Ioniță, Materials Today Bio, 29 (2024) 101341.
- [2] Mihaela-Raluca Dobrisan, Adriana Lungu, Mariana Ioniță, Virtual and Physical Prototyping, 19 (2024) e2378003.
- [3] Mingxin Qiao, Weimin Wu, Wen Tang, Yifan Zhao, Jian Wang, Xibo Pei, Bowen Zhang and Qianbing Wan, Biomaterials Science, 13 (2025) 587-605.

Figures

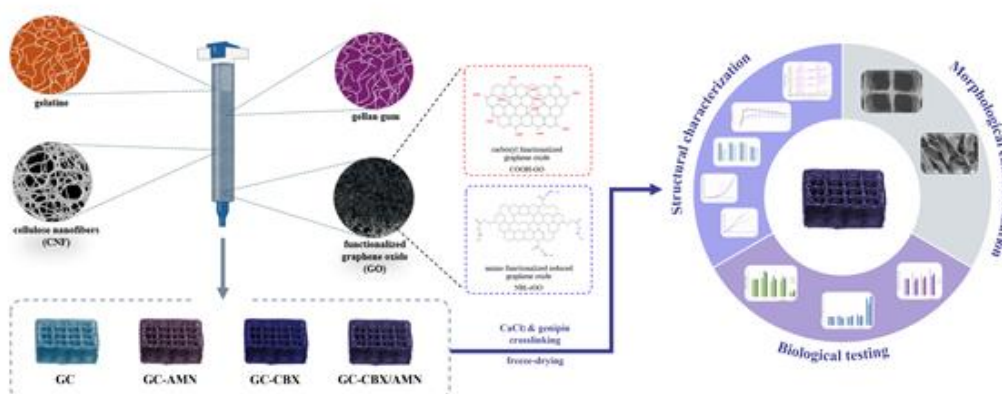


Figure 1: Schematic representation of the 3D printing ink formulations, fabrication, and characterization process.