Photocrosslinkable Graphene-Enhanced Biomaterial Inks for Improved Printability and Structural Fidelity

V. S. Parihar[1], S. Annurakshita[1,2], H. Jongprasitkul[2], D. A Fulton[2], M. Kellomaki[1]

[1]Biomaterials and Tissue Engineering Group, Faculty of Medicine and Health Technology, Tampere University, 33720, Tampere, Finland, [2]Chemistry-School of Natural Science and Environmental Sciences, Newcastle University, Newcastle-upon-Tyne, NE1 7RU, UK

INTRODUCTION: Bioprinting composite bioinks combine complementary properties for tunable functionality.[1] Graphene contributes electrical conductivity and mechanical reinforcement, while gellan gum provides structural support and biocompatibility.[2] When integrated into photocrosslinkable fabrication methods, these materials form stable, high-resolution constructs with tailored properties.[3] In this work, photocrosslinkable GGMA–graphene composite bioinks were developed for cell-laden bioprinting with human dermal fibroblasts.

METHODS: Gellan gum (GG) was methacrylated under basic conditions to obtain GGMA (≈30% methacrylation, confirmed by ^{1}H-NMR). Bioinks were prepared by dissolving GGMA (1.5% w/v) with 0.5% (w/v) Irgacure and incorporating graphene water dispersions (0.25%, 0.5%, or 1% w/w). The optimum crosslinking conditions and time were evaluated using photorheology. Human dermal fibroblasts (5×10^6 cells/mL) were added for cell-laden formulations. Rheological analyses were performed to assess shear-thinning behavior, viscoelastic properties. Bioinks were extrusion-bioprinted (Brinter BIOX), photocrosslinked, and cell viability was evaluated on Days 1, 3, and 7 using a Live/Dead assay.

RESULTS: Both GGMA and all GGMA–graphene bioinks exhibited shear-thinning with rapid structural recovery and predominantly elastic behavior ($G' > G''$). Added graphene improved viscoelastic properties, storage moduli, and stress relaxation, while photocrosslinking remained rapid and efficient. All bioinks were successfully printed into 6-layer grids and 2 mm cylinders with high resolution and consistent printability. The optimized GGMA-bioink reinforced with 0.5% w/w graphene enabled fabrication of complex anatomically relevant structures, including ear and meniscus models. Cell-laden constructs with human dermal fibroblasts showed uniform distribution and elongated morphology post-printing (Fig. 1).

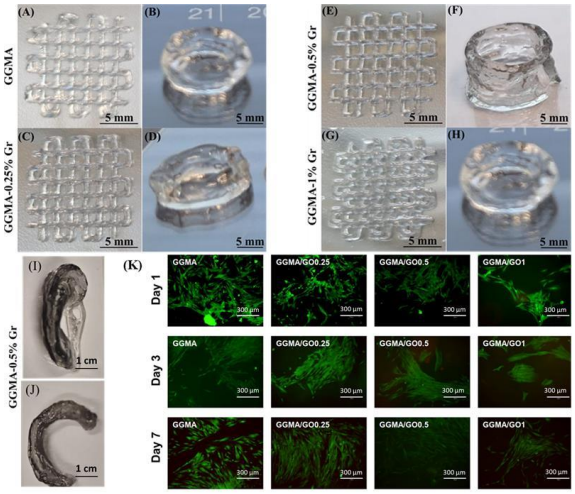

Fig. 1: Extrusion-based bioprinting of six-layer grids and 2 mm cylinders: (A, B) GGMA; (C, D) GGMA–0.25% Gr; (E, F) GGMA–0.5% Gr; (G, H) GGMA–1% Gr. The optimized GGMA–0.5% Gr was printed into complex structures: ear (I) and meniscus (J). Cell viability of bioprinted constructs was assessed at Days 1, 3, and 7 (K).

DISCUSSION & CONCLUSIONS: Graphene-reinforced GGMA bioinks showed improved mechanical and viscoelastic properties while maintaining high printability and rapid photocrosslinking. Complex structures, including ear and meniscus models, were printed with high fidelity, and cell-laden constructs supported uniform distribution and elongated morphology. These results highlight the potential of graphene–GGMA bioinks for creating robust, biologically functional tissue constructs.

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