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# PROCEEDINGS

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# Symposia

**S32 – O4**

# Engineered supramolecular hydrogels with tunable self-healing for embedded bioprinting applications

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## Introduction

Supramolecular hydrogels are emerging as attractive support materials for embedded 3D Bioprinting due to shear-thinning and self-healing behaviour. However, current support baths often show limited tunability, poor stability under culture conditions, or require harsh removal procedures that may compromise printed constructs. In this study, we developed a supramolecular hydrogel based on custom poly(ether urethane)s (PEUs) and α-cyclodextrins (CDs), as support baths for embedded bioprinting.

## Materials and Methods

Two high molecular weight PEUs were synthesized from different macrodiols: poly(ethylene glycol) (3350 Da) and Poloxamer® 407 (12600 Da), yielding SHE and SHP, respectively[1,2]. By combining SHE and SHP at different weight ratios (total PEU 4% w/v) with CDs (9% w/v), we obtained physically crosslinked hydrogels driven by polypseudorotaxanes formation. The hydrogel with optimal composition was enriched with microbial transglutaminase (mTG) (5-10% w/v) to enable in situ enzymatic crosslinking of porcine gelatin bioinks (3-5% w/v) during printing.

## Results

All formulations exhibited reproducible gelation (10-24 h), and stability up to 4 days, with more pronounced stability observed for hydrogels with higher SHP content. Rheological analyses confirmed gel-like behavior, with storage modulus G' consistently exceeding loss modulus G'' during frequency sweep tests, while self-healing strain tests revealed a G' recovery between 50% and 95% depending on the SHE/SHP ratio. Cytotoxicity tests on human fibroblasts confirmed high biocompatibility (80-100% cell viability). Among the tested compositions, the 30:70 SHE/SHP provided the best balance between mechanical recovery (87%), stability and cell viability (95%), and was selected as the optimal support bath. When enriched with mTG, the system enabled in situ enzymatic crosslinking of gelatin bioinks during printing, ensuring enhanced shape fidelity and structural retention of complex geometries (Fig. 1). Diffusion studies performed with fluorescein isothiocyanate–dextran, as model molecules for nutrients transport, confirmed the network permeability. Importantly, printed structures could be gently retrieved through washing with CD solution, enabling bath removal under mild, cell-compatible conditions.




**Figure 1**: Gelatin construct printed within support bath that maintains its structure after the bath removal.

## Conclusion

This tunable supramolecular platform represents a versatile and cytocompatible alternative to conventional support baths, offering controlled network recovery, culture stability, nutrient permeability, and facile removal. These features make the system particularly promising for embedded bioprinting of complex, functional tissue constructs for regenerative medicine.

## Acknowledgements

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## References

[1]Boffito M. et al. Novel Polyurethane-based Thermosensitive Hydrogels as Drug Release and Tissue Engineering Platforms: Design and in Vitro Characterization. *Polymer International* 2016;65; 756-769.

[2]Pappalardo R., Boffito M. Schiff-base crosslinked hydrogels based on properly synthesized poly(ether urethane)s as potential drug delivery vehicles in the biomedical field: design and characterization;. *ACS Omega* 2024; 9(46); 45774-45788.