3D Printing of Multi-Material Hydrogels

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Summary

The advent of hydrogels has revolutionised biomedical science, propelling research in bioengineering and nanoscience due to their mechanically flexible, soft, and biocompatible properties. While functional materials like magnets are ubiquitous, their integration with hydrogels into composite materials remains underdeveloped despite significant potential. This project presents the synthesis, optimisation, characterisation, and applications of 3D-printable magnetic nanofiber-hydrogel composites optimised for fatigue resistance. Two applications for these composites include substrates for in vitro lung organoids to study mechanobiological interactions of lung epithelial cells during inspiration via programmable electromagnets, and as tissue-mimetic fillers for post-surgical cavities to prevent internal tissue adhesions. Although these applications were outside the scope of the experimentals in this project, they are a key focus of the conclusions on this project's potential applications.

Report Structure

The report begins with a literature review of current magnetic soft actuator technology and microfiber synthesis, followed by a theoretical analysis of the target composite system. It details the experimental procedures for modular synthesis and provides a quantitative comparison of magnetic responsiveness across various material compositions. The report concludes with a discussion on the applications, advantages, and challenges of these materials.

Approach

Although stimuli-responsive pure hydrogels have been known in the literature for decades, these systems rely on their intrinsic physicochemical properties, such as the effect of environmental pH, temperature and light sensitivity on the hydrogel crosslinking and ionisation. These environmental parameters must be tightly controlled in the biomedical domain to maintain cell viability both *in vitro* and *in vivo*, limiting the utilisation of this mode of actuation in soft biocompatible materials. Magnetism, as a straightforward method for applying external, no-contact forces on soft materials, with manageable cytotoxicity, was chosen as a suitable functional material to enable actuation while maintaining a homeostatic environment. This led to the development of a composite of flexible magnetic-polymer microfibers within a hydrogel matrix for soft biocompatible actuators.

A theoretical framework based on structural analysis and multiphysics simulations guided the material selection process. Experiments identified fibres with optimal magnetic responsiveness and hydrogels with suitable viscoelastic and rheological properties. Wet spinning was used to produce the fibres, and 3D bioprinting to incorporate them into the hydrogel matrix (although manual extrusion is also possible and was simpler for demonstration purposes). The fibres were encapsulated within the hydrogel in its liquid phase and solidified by cooling. The composites were tested for deflection under a switching magnetic field, evaluating sensitivity and fatigue resistance.

Results

Automated wet spinning, using a robotic arm and motor-controlled spinner, produced fibres with consistent morphology. Light microscopy revealed an average fibre diameter of 15 ± 9 microns, largely independent of the synthesis formula. Iron oxide (Fe_3O_4) and polystyrene were used to create the magnetic-polymer fibres. A binary solvent of 85% *N*,*N*-dimethylformamide (DMF) and 15% acetone dissolved the mixture, producing fibres spun to an initial length of 28 mm. Variations in iron oxide and polystyrene concentrations (10%, 20%, 30% by weight) identified that a 40-50 wt% solid fraction yielded fully spinnable solutions, with highest magnetic responsiveness at the highest iron content. Deflection tests showed peak displacements of ~2 mm with virtually zero time lag or hysteresis, retained with the hydrogel. The 20% polystyrene and 30% iron oxide composition was optimal for fatigue resistance and hydrogel adherence, making it the optimal choice for soft actuators in remotely stimulated applications.

Conclusions

This project has demonstrated a novel synthesis of microfibre technology within soft biocompatible hydrogels with key challenges of such composites resolved, including fatigue resistance, prevention of delamination between the fibres, and prevention of cutting through the hydrogel during fibre actuation. The physical construction of the presented composite is straightforward and feasible with open-source technology, and is moreover expected to be highly biocompatible due to the separation between the inorganic magnetic fibre layer and the hydrogel in which cells may proliferate freely. This paves the way for soft actuator technology in a range of biomedical and nanoscience applications leveraging the inherent biocompatibility of the system.