3D Printing of Multi-Material Hydrogels

IIB Project Final Presentation, Division C

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Presentation Outline

- Introduction
- Theory
- Materials and Methods
- Results
- Applications
- Conclusions

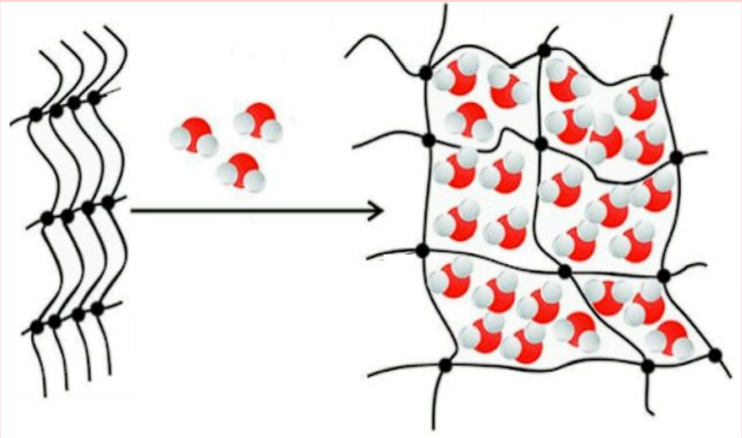
Introduction

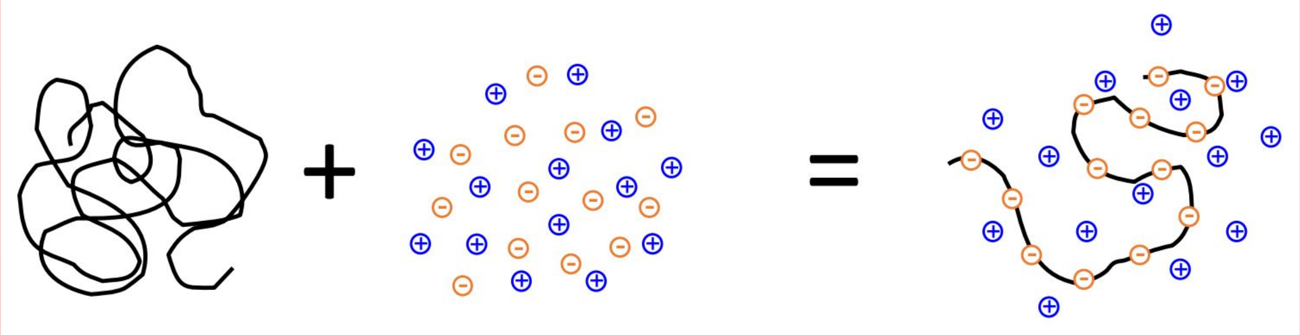
What are multi-material hydrogels?
What has been done so far?
What is new about this work?

Hydrogels

Crosslinked biopolymers swell in water to form a **solid structure**

We aim to make hydrogels that can **change their shape.** How might this be done?





Biopolymer + Aqueous Electrolyte = Ionic Hydrogel (Polyelectrolyte)

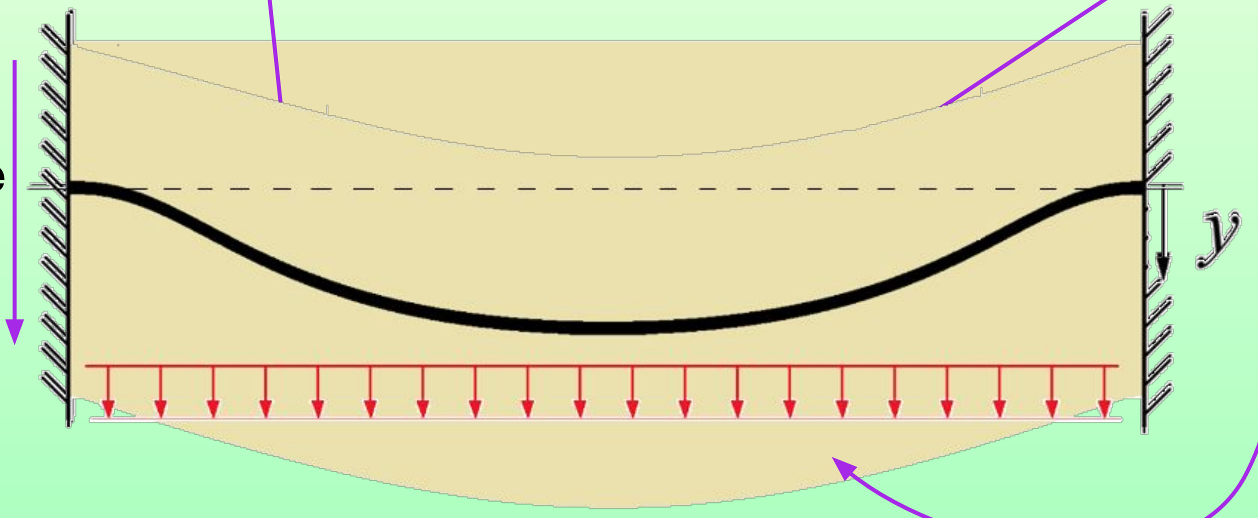
Ionisation is pH dependent
→ allows actuation

Problem: relies on a **change in chemical environment**
→ totally unsuitable for biomedical applications

Biocompatible Actuation of Hydrogels

True biocompatibility requires **homeostasis**

Integrate **hydrogels** with functional materials like **magnetic fibres**:

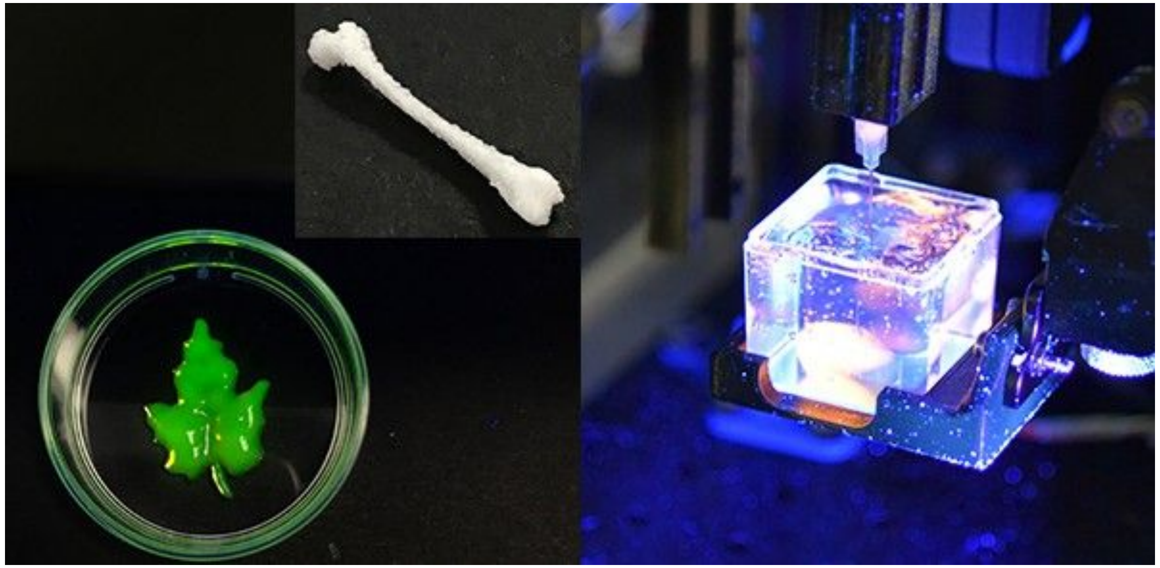


An externally applied magnetic field can **displace** the hydrogel

If the fibres are nontoxic, this system retains biocompatibility

3D Printing of Magnetic Hydrogels

Implants that become part of a person's natural tissues over time



Shery's group has already built a 3D bioprinter (Printer.HM) for extruding soft materials in the Nanoscience centre. [1]

If we can similarly automate the production of magnetic fibres, we can **3D print the composite hydrogels.**

Applications:

1) *In vitro* lung models
2) Smart biomedical textiles

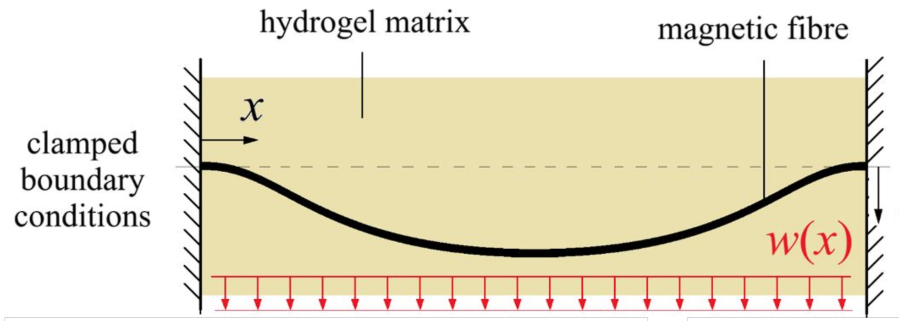
Theory

Structural Analysis
Computational Modelling
Material Selection

Structural Analysis of Magnetic Fibre

Beam theory with a additional contact force from the hydrogel

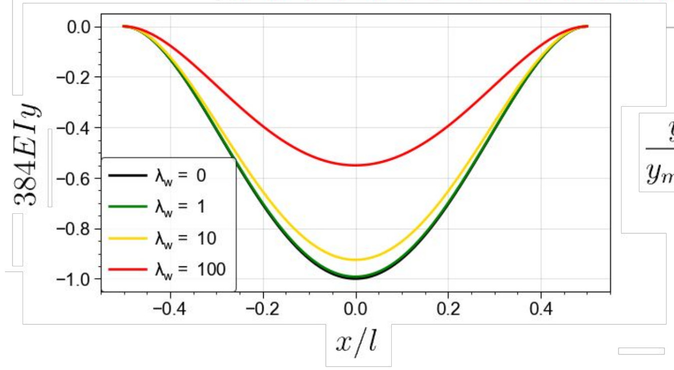
Assume a clamped elastic beam surrounded by a soft hydrogel medium. The hydrogel exerts a viscoelastic reaction force on the displaced fibre **[2]**

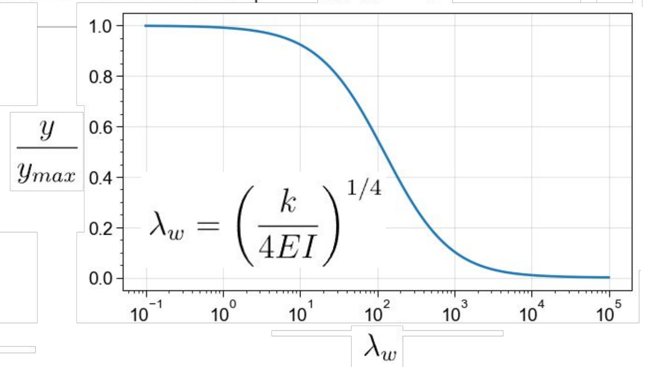


Steady state

$$EI\frac{\partial^4 y}{\partial x^4} + \cancel{\rho\frac{\partial^2 y}{\partial t^2}} = \underbrace{w(x)}_{\text{magnet force}} - \underbrace{ky(x,t) - \cancel{\eta\frac{\partial y}{\partial t}}}_{\text{distributed reaction (Kelvin-Voigt model)}}$$

at $x = 0$





(k: hydrogel stiffness per unit length, E: fibre modulus, I: fibre second moment of area)

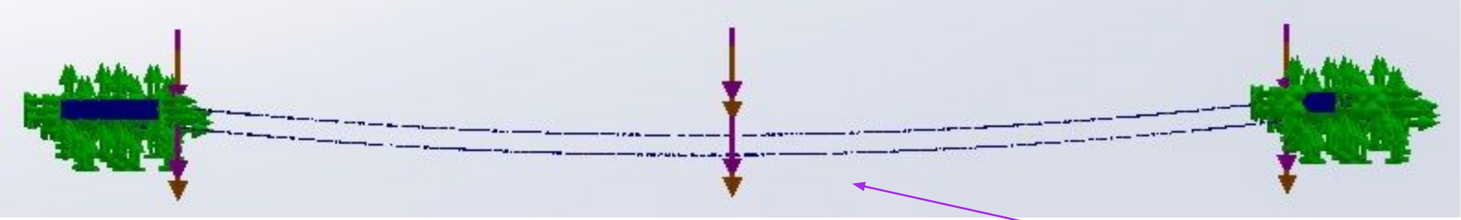
Hydrogel force ky must not cause it to fail

Compliant hydrogels do not impede the motion

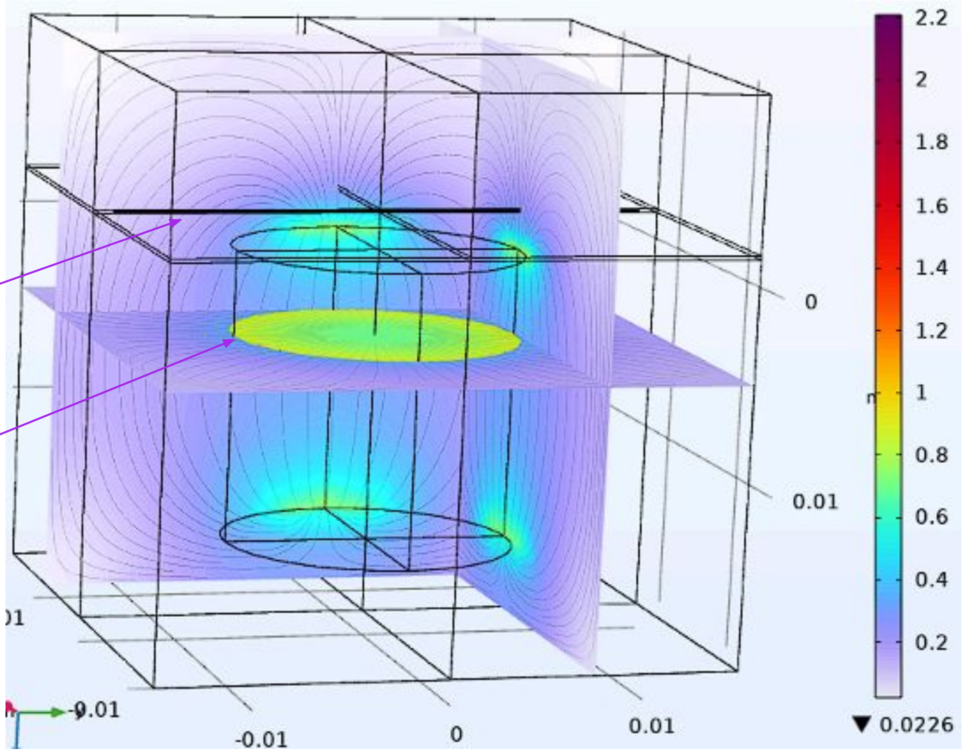
Computational Modelling

Captures the nonlinear magnetomechanical coupling

Solidworks: simple structural FEA



COMSOL: complex multiphysics



fibre

electromagnet

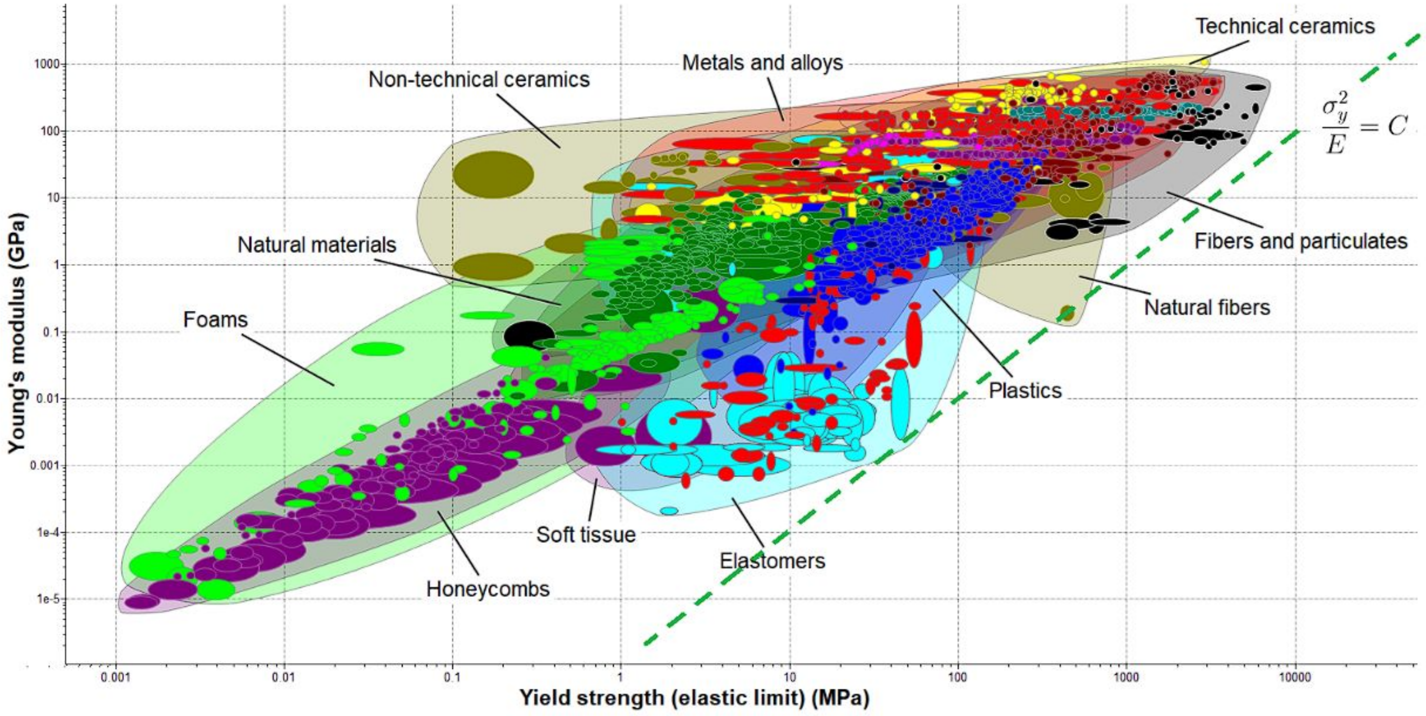
Useful for checking **feasibility** of setup

But precise calculations are hard because **material properties are highly variable**!

Material Selection

Stiffness-limited design, with many secondary constraints

The relevant **failure criterion** for this application is **fatigue loading**



Material property chart, using **Ansys Granta** L3 database

Rule of thumb: ensuring peak stress is **less than half of yield stress** will protect against high-cycle fatigue

Materials and Methods

What materials were chosen?
How are the fibres made?
How are the composites made and tested?

Fibre Compositions

Solvent + Polymer + Magnets

+

+

+

→

DMF
(*N*,*N*-dimethylformamide)
85% weight

Acetone
(propanone)
15% weight

Solvent

Polystyrene
sets spinnability

Polymer Filler

Iron oxide
sets magnetism

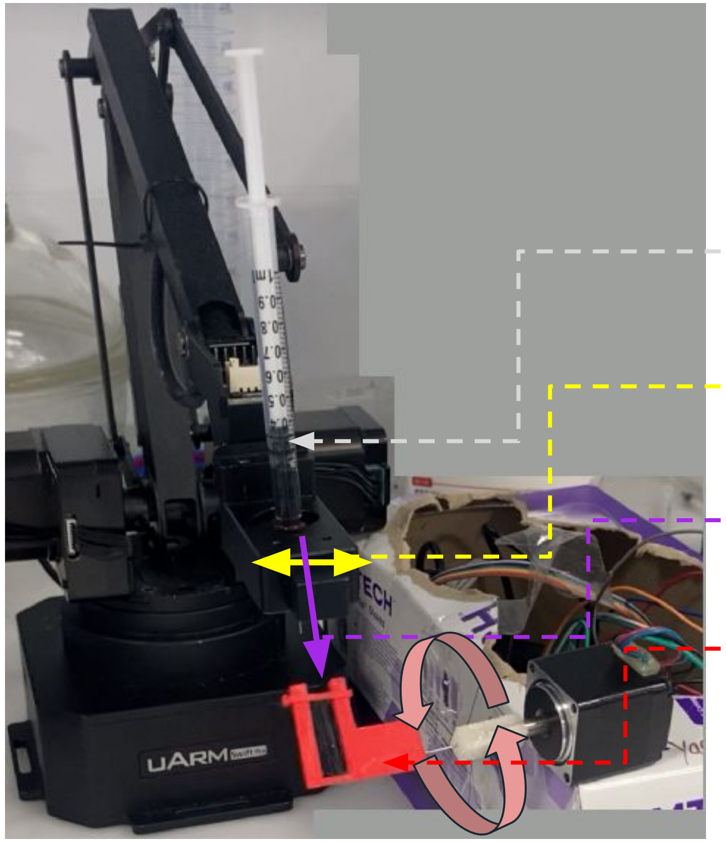
Magnets

Magnetic solution

Ready for nanospinning process

Fibre Spinning

Using a robot arm, syringe and spinning frame



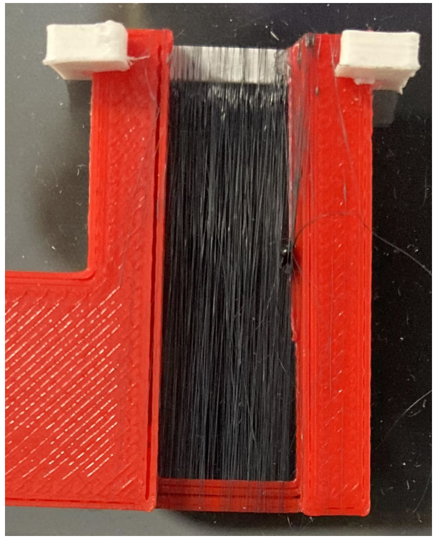
Automated nanospinning

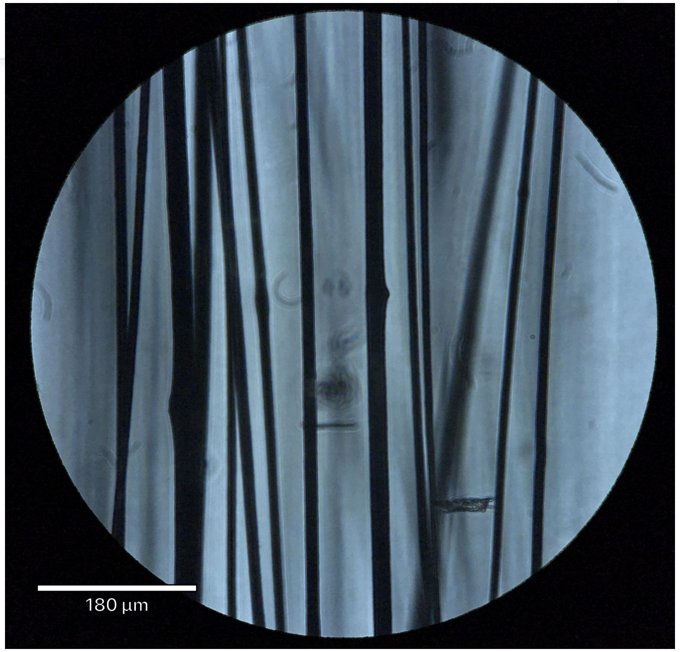
syringe full of fibre solution

robot arm

extruded fibres

spinning frame





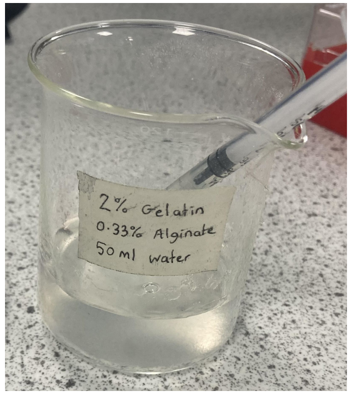
diameter ~ 20 µm

Produces this in about 2 mins of spinning

Hydrogel Compositions

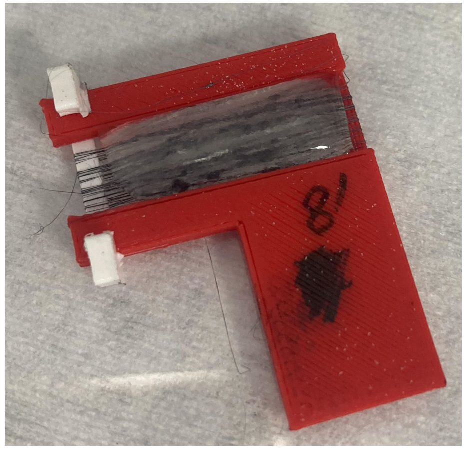
Gelatin/Alginate as a Self-Crosslinking UCST Hydrogel

Making gelatin hydrogels is **very simple**: just mix the powders together in water



Printer.HM (3D bioprinter) extrudes hot liquid phase hydrogel which **solidifies** on the stage as it cools below its **upper critical solution temperature (UCST)**

By **depositing the hydrogel onto the fibres**, the composite can be prepared as the hydrogel surrounds the fibres



Testing the Magnetic Responsiveness

Use an electromagnet and measure the deflection

Electromagnet programmed to **switch on and off**

With a small contraction using the slider on the frame, the fibres will show **large deflections in the magnetic field**

Results

Make Magnetic Polymer Solution
Program Spinner
Extrude Fibres
Observe Under Microscope
Assess Quality and Deformation

Best Compositions for Magnetic Fibres

Need ~50% solid in fibre solution, with most compliant hydrogel

‘Best’ means 1) good spinnability, 2) homogeneous fibres, 3) high deflection

Combinations of these variables were tested:

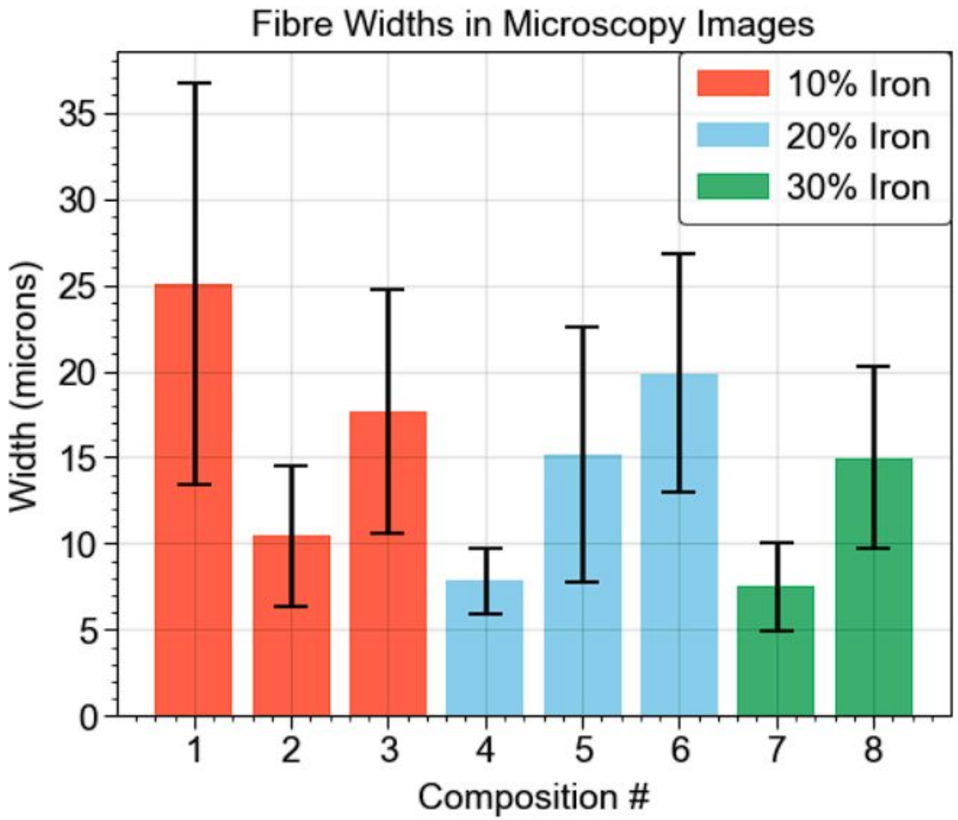
- Polystyrene mass fraction: {10%, 20%, 30%}
- Iron oxide mass fraction: {10%, 20%, 30%}
- Gelatin/Alginate (6:1) mass fraction: {7%, 2.33%, 0.33%}

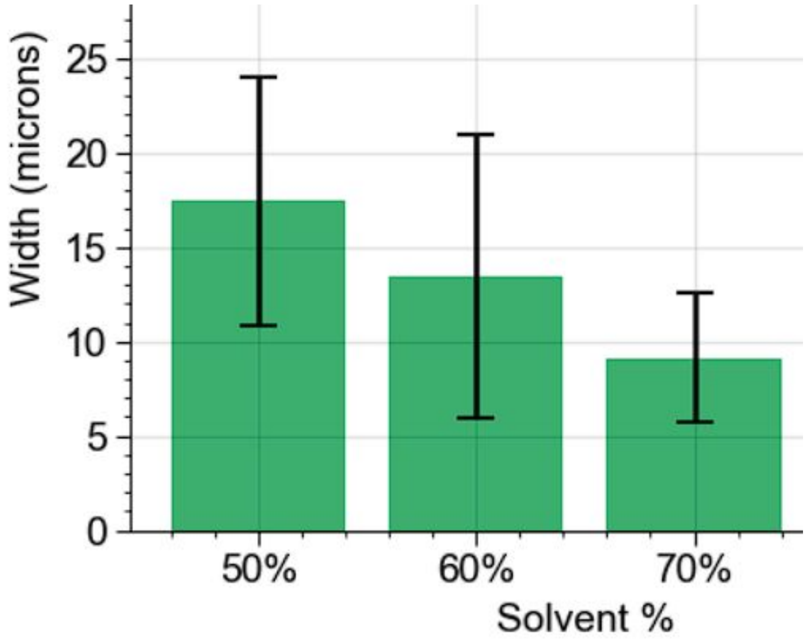
Best fibre solution: using 20% polystyrene and 30% iron oxide

Best hydrogel: 2% gelatin and 0.33% sodium alginate in DI water

Homogeneity of Fibre Dimensions

Over the range of tested compositions, size was quite consistent





More polymer/iron in the solution usually gives thicker fibres but correlation is weak

Homogeneity of Fibre Dimensions

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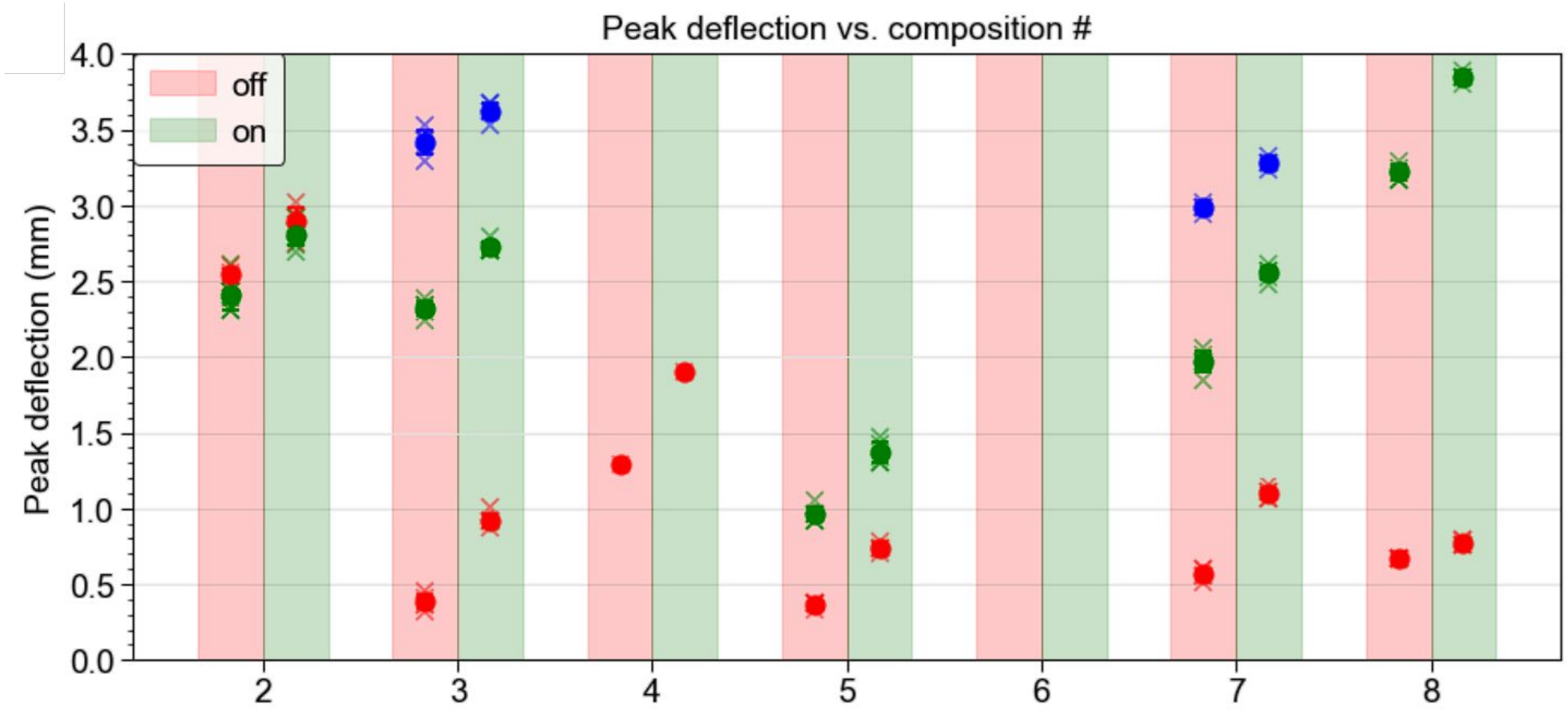


Fibre length ~ 28 mm

Deflection ~ 1 mm

Strain ~ 5%

Reduced to **about half** when combined with the hydrogel

Applications

In vitro lung model organoids
Smart biomedical textiles

Applications of Soft Actuators

Combines remote movement with biocompatibility

Can **combine with other projects** demonstrated from Shery's group (cryoprinting, fibre printing robot…) to **fully realise 3D printing capabilities**

Application #1: Lung Organoids
Hydrogel loaded with cells could experience **actuation** to simulate lung tissue **breathing** *in vitro*, study cell-matrix mechanobiological interactions

Application #2: Smart Biomedical Textiles (+ Regenerative Medicine?)
Using bioresorbable hydrogels as post-surgical cavity fillers to **prevent internal adhesions** by moving into place, **reducing patient complications**

Conclusions

Magnetic hydrogel composites can be 3D printed in a modular fashion

Key Takeaways from this Project

Magnetic Hydrogels

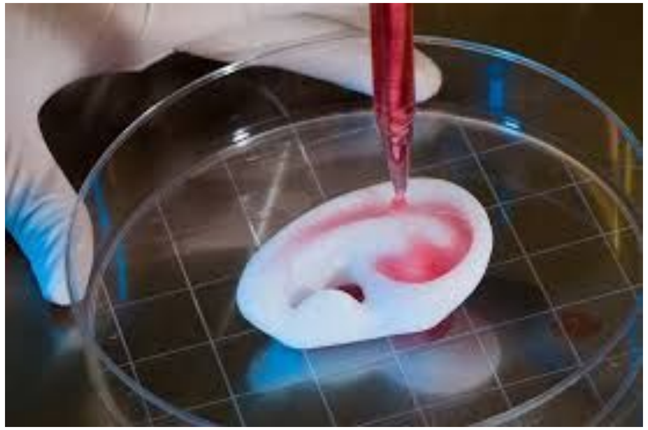
★ It is easy to **functionalise** fibres with **magnetic materials**

★ The nanospinning process can be **automated** for **high-throughput** production

★ The fibres and hydrogels can be printed separately

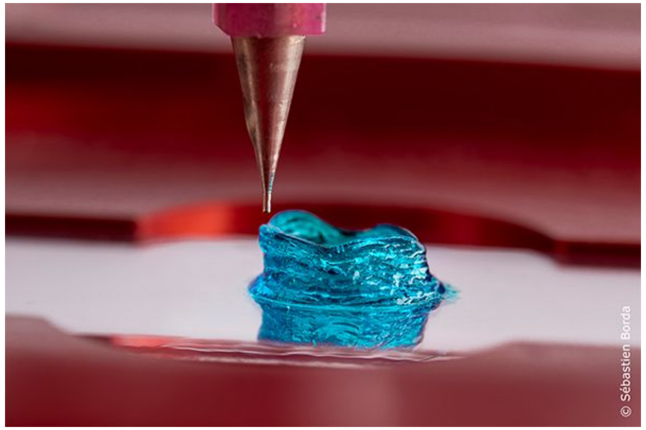
★ Using (**30% iron oxide, 20% polystyrene**) fibres with (**2% gelatin, 0.33% alginate**) hydrogel was found to be optimal across a variety of metrics

★ The fibres are **resilient** and **do not delaminate or cut** the hydrogel during operation



Thank You

Q & A



Sources cited in this presentation

[1] Lei, I. M., Sheng, Y., Lei, C. L., Leow, C., & Huang, Y. Y. (2022). A hackable, multi-functional, and modular extrusion 3D printer for soft materials. Scientific Reports, 12(1). https://doi.org/10.1038/s41598-022-16008-6

[2] Dillard, D. A., Mukherjee, B., Karnal, P., Batra, R. C., & Frechette, J. (2018). A review of Winkler's foundation and its profound influence on adhesion and soft matter applications. Soft Matter, 14(19), 3669–3683. https://doi.org/10.1039/c7sm02062g

Own content: photographs of experimental results not referenced