

# BIOINSPIRED TRANSPARENT IMPACT-ABSORBING COMPOSITE MADE WITH A FLUID MECHANICAL INSTABILITY

Gosselin, FP<sup>1\*</sup>, Zou, S<sup>12</sup>, and Therriault, D<sup>1</sup>

<sup>1</sup> Laboratory for Multiscale Mechanics (LM2), Research Center for High Performance Polymer and Composite Systems (CREPEC), Polytechnique Montreal, Montreal, QC

<sup>2</sup> AMOLF, Amsterdam, Netherlands

\* Corresponding author (frederick.gosselin@polymtl.ca)

**Keywords:** *3D Printing, Biomimetics, Liquid Rope Coiling Instability*

## ABSTRACT

Spider capture silk outperforms most synthetic materials in terms of specific toughness. We present an instability-assisted 3D printing technique to fabricate tough microstructured fibers bioinspired by the structure of spider silk. Upon embedding these polycarbonate microstructured fibers into an elastomer (PDMS) matrix with a matching refractive index, we obtain a transparent composite with interesting impact-absorbing properties. The exact geometry of the fibers results from the liquid rope coiling instability which occurs when a fused filament fabrication (FFF) printer extrudes material faster than its printhead moves, similarly to a viscous honey thread buckling and wiggling as it hits a plate. Changing the printing parameters changes the looping topology of the fibers as well as how it unwinds when pulled in tension. We embed these micro-structured fibers in an elastomer matrix with matching refractive indices. We test the ability of the resulting composite to dissipate the energy of a dart impact. We compare a pure elastomer sheet, a composite with straight fibers, one with alternating fibers, and a mixed composite with both straight and alternating fibers. The composites containing alternating fibers exhibit very large hysteresis, dissipating more than 95% of the energy of the falling dart. Our approach introduces a new class of impact-absorbing composites, which can even be designed to be transparent.

## 1 INTRODUCTION

Tough materials absorb a lot of energy when they deform. They are necessary to manufacture light products which resist impact and wear. For example, on an aircraft, the jet engine fan containment system must be made of a light material which will absorb the energy of the fast-flying debris of an unlikely engine explosion. Similarly, harnesses and fall-arrest lanyards for construction workers and circus performers require materials which deform to absorb the energy of a fall. Another example is the windshield of an aircraft which must be transparent, light, and tough to absorb the energy of a debris or a bird impact. When thinking about tough materials, 3D-printed parts are not exactly what comes to mind as they are more often brittle and weak. 3D printers are ubiquitous in schools, labs, companies and even the homes of hobbyists. Most basic models are typically limited to printing only a handful of materials (often only one). Improving the mechanical properties of the material of parts made with these 3D printers already out on the market could greatly increase their functionality. New tough materials are desirable, but considering issues of material certification, printability, biocompatibility, etc., it would be even more desirable to render the materials already used in the cited applications extremely tough.

Solutions to improve the mechanical properties of 3D printed parts typically come from printing parameter optimisation or by including reinforcements. Another approach, inspired by nature and building on the strength of 3D printing for making complex geometries, would be to modify the microstructure of the printed parts while using the same bulk material to achieve the same goal. In this project, we follow this latter approach to use existing 3D

fuse-filament fabrication (FFF) technology outside of its intended parameter range to fabricate tough microstructured fibres. A fluid flow instability which normally destroys the accuracy of prints is here used as an advantage to improve the mechanical properties of the 3D-printed material. This instability-assisted 3D-printing concept is used to fabricate a web of microstructured fibres out of polycarbonate. Upon embedding these polycarbonate microstructured fibers into an elastomer (PDMS) matrix with a matching refractive index, we obtain a transparent composite with interesting impact-absorbing properties.

## 2 Instability-assisted 3D printing

We developed a novel approach of instability-assisted 3D printing of thermoplastics and demonstrated its use to make microstructured fibres [1]. We use FFF 3D printing outside its conventional parameter range, yielding control of the geometry of the fibres produced to the liquid rope coiling instability[2], [3]. We flow a filament of molten polymer towards a substrate moving perpendicularly at a slower velocity than the filament flows (Figure 1). The filament buckles repetitively giving rise to periodic meanders and stitch patterns. As the polymer cools, the filament solidifies into a fibre with a geometry bestowed by the instability. Some coiling patterns give rise to a toughening mechanism due to the sacrificial bonds created when the viscous filament loops over itself and fuse. In order to stretch the fibre under uniaxial tension, the sacrificial bonds must be broken, hence adding to the energy required for breaking the fibre. The sacrificial bonds in the microstructured fibre play an analogous role to that of the hydrogen bonds present in the molecular structure of the silk protein which give its toughness to spider silk.

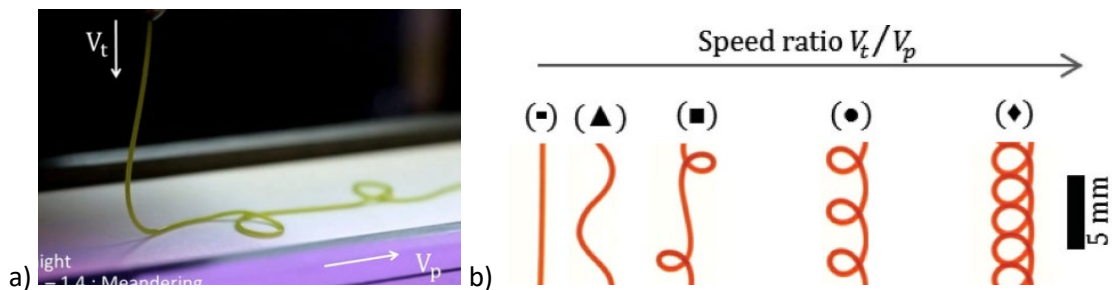


Figure 1 Instability-assisted 3D-printing: a) a fuse-filament fabrication 3D printer extrudes a filament at velocity  $V_t$  faster than the platform below is moving  $V_p$ ; b) the filament buckles and coils into different instability patterns depending on the speed ratio.

## 3 Tuning the parameters to optimize the fiber properties

In order to better understand how microstructured fibers unwind and stretch when pulled upon, we performed an experimental campaign of tensile tests together with finite element simulations [4]. We fabricated polylactic acid (PLA) fibers while varying multiple parameters: the printing platform speed, extrusion speed, nozzle height, and nozzle size. These different printing parameters gave rise to various instability patterns (Figure 1) with varying degrees of sacrificial bond strength when the fiber loops over and fuses to itself. We then pulled these fibres in quasi-static tension with a universal testing machine and measured their stiffness, strength, and energy at break (Figure 2). We paid special attention to early failures: fibres that break upon a slight extension. Finite element analysis simulations done with a combination of beam and solid elements with a multilinear isotropic hardening plasticity and von Mises yield criterion allowed better understanding how the fibres broke. We found five different types of failures that occurred in the various fibres depending on their patterns and their printing parameters.

Overall, we found that for maximal energy absorption, microstructured fibres should be slender and made of a ductile material. This means that their diameter should be small with respect to their pattern wavelength and the material should exhibit drawing before failing.

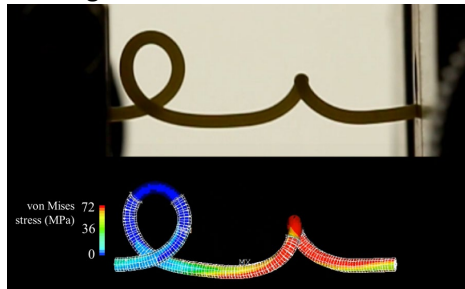


Figure 2 Comparison of an experimental tensile test of a microstructured FFF fiber with the corresponding finite element simulation. These simulations helped us understand the best parameters for making tough fibers.

## 4 Impact-absorbing transparent composite

The instability-assisted 3D printing technique works for any polymer that can be melted [1], dissolved into a viscous solution [5], [6], or even for molten glass [7]. To demonstrate how versatile the technique is, we fabricated a web of microstructured fibres out of transparent polycarbonate and embedded it in a refractive-index matching elastomer matrix to create an impact-absorbing transparent composite [8], [9] (Figure 3). We tested the ability of the resulting composite to dissipate the energy of a dart impact. We compare a pure elastomer sheet, a composite with straight fibers, one with alternating fibers, and a mixed composite with both straight and alternating fibers. The composites containing alternating fibers exhibit very large hysteresis, dissipating more than 95% of the energy of the falling dart. Our approach introduces a new class of impact-absorbing composites, which can even be designed to be transparent.

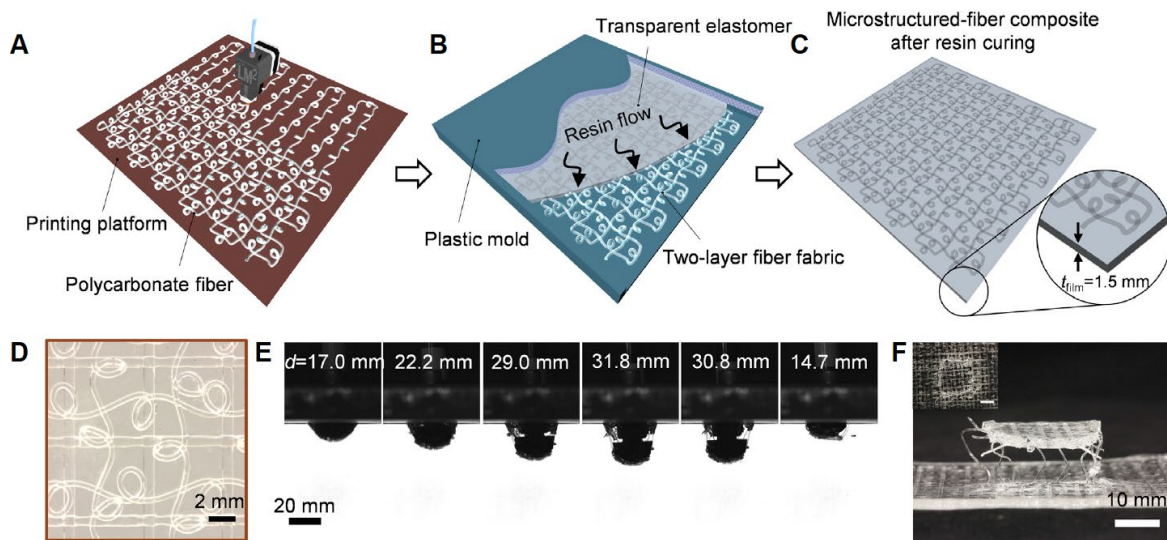


Figure 3: Fabrication and testing of transparent impact-absorbing composites: (A) instability-assisted 3D printing of a mat of polycarbonate fibers; (B) PDMS resin transfer moulding; (C) resulting microstructured-fiber composite after curing; (D) top-view photograph of the composite; (E) snapshots from a high speed camera of the composite absorbing the impact of a rough dart; and (F) photograph of the damaged composite after impact.

## 5 Conclusion

Instability-assisted 3D printing is a versatile, easy-to-implement, but hard-to-master fabrication technique (it can be finicky). All that is required is an FFF 3D printer which allows for enough freedom in setting its parameters and custom-coding its G-code. By changing the shape of the printed material fibres, we can control its mechanical behaviour. By microstructuring fibres to have sacrificial bonds like spider silk, and arranging them in a web, like a spider web, we can design the mechanical behaviour of a component, and also how it fails.

Whereas real biological materials are truly hierarchical, with structures at multiple length scales, the microstructured fibres we present here only have two length scales: their diameter and their wavelength. In future work, we could improve the mechanical behaviour of these fibres by adding another length scale to their construction by doping them with short carbon fibres or nanotubes.

## 6 Acknowledgements

We acknowledge the support of the Fonds de Recherche du Québec: Nature et Technologies (FRQNT), Canada, [funding reference number 63014], the Natural Sciences and Engineering Research Council of Canada (NSERC), [funding reference number 175791953], and the Canadian Foundation for Innovation.

## 7 REFERENCES

- [1] R. Passieux, L. Guthrie, S. H. Rad, M. Lévesque, D. Therriault, and F. P. Gosselin, 'Instability-Assisted Direct Writing of Microstructured Fibers Featuring Sacrificial Bonds', *Advanced Materials*, vol. 27, no. 24, pp. 3676–3680, Jun. 2015, doi: 10.1002/adma.201500603.
- [2] N. M. Ribe, M. Habibi, and D. Bonn, 'Liquid rope coiling', *Annual Review of Fluid Mechanics*, vol. 44, pp. 249–266, 2012.
- [3] P.-T. Brun, N. M. Ribe, and B. Audoly, 'A numerical investigation of the fluid mechanical sewing machine', *Physics of Fluids*, vol. 24, no. 4, p. 043102, 2012, doi: 10.1063/1.3703316.
- [4] S. Zou, D. Therriault, and F. P. Gosselin, 'Failure mechanisms of coiling fibers with sacrificial bonds made by instability-assisted fused deposition modeling', *Soft matter*, vol. 14, no. 48, pp. 9777–9785, 2018.
- [5] S. Guo, F. Gosselin, N. Guerin, A. Lanouette, M. Heuzey, and D. Therriault, 'Solvent-Cast Three-Dimensional Printing of Multifunctional Microsystems', *Small*, vol. 9, no. 24, pp. 4118–4122, 2013.
- [6] Q. Wu, S. Zou, F. P. Gosselin, D. Therriault, and M.-C. Heuzey, '3D printing of a self-healing nanocomposite for stretchable sensors', *Journal of Materials Chemistry C*, vol. 6, no. 45, pp. 12180–12186, 2018.
- [7] P.-T. Brun *et al.*, 'The molten glass sewing machine', *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 375, no. 2093, p. 20160156, May 2017, doi: 10.1098/rsta.2016.0156.
- [8] S. Zou, D. Therriault, and F. P. Gosselin, 'Spiderweb-Inspired, Transparent, Impact-Absorbing Composite', *Cell Reports Physical Science*, vol. 1, no. 11, p. 100240, 2020.
- [9] S. Zou, D. Therriault, and F. P. Gosselin, 'Toughening elastomers via microstructured thermoplastic fibers with sacrificial bonds and hidden lengths', *Extreme Mechanics Letters*, vol. 43, p. 101208, 2021.