



**Queensland University of Technology**  
Brisbane Australia

This may be the author's version of a work that was submitted/accepted for publication in the following source:

[Paxton, Naomi, Lamont, Riki, Brooks-Richards, Trent, & Woodruff, Mia](#)  
(2021)

Expanding material printability for electrowriting.  
*Journal of 3D Printing in Medicine*, 5(2), pp. 61-64.

This file was downloaded from: <https://eprints.qut.edu.au/212904/>

© 2021 Future Medicine Ltd

This work is covered by copyright. Unless the document is being made available under a Creative Commons Licence, you must assume that re-use is limited to personal use and that permission from the copyright owner must be obtained for all other uses. If the document is available under a Creative Commons License (or other specified license) then refer to the Licence for details of permitted re-use. It is a condition of access that users recognise and abide by the legal requirements associated with these rights. If you believe that this work infringes copyright please provide details by email to [qut.copyright@qut.edu.au](mailto:qut.copyright@qut.edu.au)

**License:** Creative Commons: Attribution-Noncommercial 4.0

**Notice:** *Please note that this document may not be the Version of Record (i.e. published version) of the work. Author manuscript versions (as Submitted for peer review or as Accepted for publication after peer review) can be identified by an absence of publisher branding and/or typeset appearance. If there is any doubt, please refer to the published source.*

<https://doi.org/10.2217/3dp-2021-0012>

## Expanding Material Printability for Electrowriting

### **Tweetable Abstract**

Melt electrowriting has grown into a versatile technique in #biofabrication. This article explores how innovations in material characterisation and #3Dprinting hardware are expanding the ability to print novel #biomaterials.

### **Keywords**

Melt electrowriting, electrohydrodynamic jetting, 3D printing, biomaterials, polymers, scaffolds, printability, additive manufacturing

### **Background to Melt Electrowriting**

Melt electrowriting (MEW) is playing an increasingly significant role in the development of biomimetic scaffolds for a range of biofabrication applications [1]. MEW is an extrusion-based additive manufacturing (3D printing) technique using electrohydrodynamic jetting to produce micron-scale fibres from molten materials. This fibre jet is collected onto a motorised, computer-controlled collector plate or rotating mandrel, dictated by 'gcode' to define fibre placement and patterning into 3D structures. The ability to precisely control fibre placement has enabled next-generation biomimetic scaffolds to be fabricated in a highly-controlled manner, recapitulating the native architecture of tissues and has potential to be applied to a range of biomaterials. Recent studies have advanced the complexity of precision fibre placement [2], unit cell designs [3,4] and scaffold geometry [5,6]. However, expanding the availability of printable materials beyond the ever popular polycaprolactone (PCL) remains a challenge [7]. This article will explore innovation in both materials science and engineering to consolidate an understanding of how material properties and novel printing hardware can be leveraged to expand processing capacity for a wider range of materials. We aim to highlight the opportunities for using electrowriting-based fabrication as a promising tool in biofabrication and medical 3D printing research which could revolutionise patient-specific implant manufacturing to enable tissue support and regeneration.

## **Biomaterial Factors Influencing Printability**

It is well-known that the successful extrusion of fluids into controlled structures is largely dependent on material viscosity. Research has investigated the relationship between material viscosity and printability in extrusion-based 3D printing which has informed the processing of novel materials [8,9]. Since the invention of MEW [10], thermoplastics have been preferred both for their suitability in a range of biofabrication applications which require mechanically tuneable and biocompatible tissue scaffolding, but also for their ability to be printed in molten state at low temperature (below  $\sim 100^{\circ}\text{C}$ ) without the need for solvents [7]. PCL is deemed the most favourable polymer for MEW, owing to its low melting point and flow properties under low ( $<1$  bar) pressure, and has been highlighted in over 50 publications [1]. Several properties of thermoplastics have been exploited to demonstrate the optimisation of printability, including molecular weight, chain length, branching, and crosslinking, which strongly dictate the entanglement of polymer chains and resistance to flow in molten state [11]. The use of additives, including bioactive or functionalised nanoparticles to improve mechanical or biological properties has also been explored, whereby particle size and binding between particles and polymer chains have been demonstrated to strongly impact printability [12,13], in conjunction with the material dielectric charge properties which are also critical to a biomaterial's interaction with the electric field [14]. For thermoplastics dissolved in solution, the viscoelastic properties that impact the conditions under which it can be extruded at a MEW-compatible flow rate are largely dependent on the solvent viscosity and polymer concentration, which have been widely explored in the solvent electrospinning literature.

A more generalised understanding of the interaction between material viscosity and printability using modified electrohydrodynamic MEW-based approaches has led to the successful fabrication of non-thermoplastic biomaterial microfibres, namely water-based polymer solutions 'inks' or hydrogels [15]. Recent studies have reported the fabrication of cell-laden bioinks into microfibres, an order of magnitude smaller in diameter than fibres achievable using routine bioprinting [16]. These studies highlight the immense capacity for expanding the range

of materials processable using electrowriting-based techniques through exploitation of a materials' viscoelastic properties both during extrusion to enable controlled extrusion and Taylor cone formation, as well as recovery after extrusion, through gelation, solidification, or other mechanisms to retain the shape fidelity of the printed microfibres. The ability to fabricate microfibre structures using electrowriting techniques using a broader library of thermoplastics, as well as non-thermoplastic biomaterials, offers substantial promise for applications in tissue engineering, regenerative medicine, microfluidics and organ/lab-on-a-chip.

### **Expanding Hardware Capability**

The ability to print novel materials is also strongly dependent on the MEW hardware capacity. Fundamentally, the successful formulation of a stable hydrodynamic jet is established by firstly extruding a fluid at a suitable flow rate from the nozzle, followed by the electric-field induced formation of a Taylor cone, leading to a stable material column formed between the nozzle and collector which is then patterned onto a motorised collector [10]. Materials are typically extruded from the nozzle using a force-driven extrusion mechanism, dispensing a fluid from a reservoir (often a syringe) using syringe pumps or regulated air pressure supplies, as opposed to filament-based extrusion which is common to techniques such as fused deposition modelling (FDM). A common restriction to printing higher viscosity materials is the maximum pressure available in a lab environment, typically ~5 bar, without the use of additional compressors. Many high viscosity materials have been unprintable to-date using the currently-available hardware to extrude them at a suitable flow rate conducive to Taylor cone formation and collection on the motorised plate. However, the use of advanced printer nozzles and extrusion mechanisms may unlock the potential for processing such materials.

For MEW, heating capacity is also a common restriction to suitably reducing the viscosity of a polymer melt to that which is printable within the limit of extrusion force, or available air pressure [17]. High temperature print heads have been successfully utilised to process higher melting temperature polymers than PCL, or those doped with additives leading to increased viscosity [18,19]. However, the thermal stability of polymers, as well as any additives, retained

in the reservoir during the low flow-rate extrusion at high temperatures remains a concern. Heated collector plates have shown to improve the adhesion of printed fibres by reducing the temperature difference between molten polymer and collector [19]. With expansion into higher print temperature experiments, heated collector plates will play a critical role in optimising the morphology and crystallinity of collected fibres.

Although typically undertaken at the end of the MEW optimisation pipeline, the collector plate velocity is typically tuned in inverse proportion to a material's viscosity. Under comparable environmental conditions, higher viscosity materials would extrude slower than low viscosity materials, which would therefore require higher collector plate speeds to collect the fibre in a straight 'written' line. Since a variety of factors can be tuned to optimise the flow rate of the polymer, including pressure and nozzle size, which play a more significant role in material extrusion success, collector plate velocity is seldom identified as the reason materials were deemed 'unprintable' and is readily tuned to optimise jet dynamics and fibre patterning [11].

Despite the prevalence of custom-built 3D printers amongst MEW research teams, there is a surprisingly limited consensus on equipment safety which is of vital importance when considering the intrinsic hazards associated with such equipment upgrades, particularly involving high voltage equipment to increase the range of printable materials. This is further compounded by the substantial variation in electrical safety standards between jurisdictions, and a lack of relevant standards surrounding the threshold for stored capacitor energy to become hazardous [20]. Standards have provided some guidance on best practice for minimising the risk of shock or arc-flash hazards, including implementation of grounding on all equipment contact points exposed to personnel, isolation of hazardous voltages at safe distances with adequate insulation, safe discharge of capacitive devices and interlocking devices to disable high voltage supplies, in addition to rigorous process control practices [21]. However, routine voltage operating ranges (>1.5 kV DC) either lie outside the scope of electrical safety standards or relate to large scale power infrastructure rather than small-scale laboratory and medical equipment [22]. With the growing interest in MEW amongst the

biofabrication and tissue engineering communities leading to the development of more custom-built machines, combined with the drive to push the limits of MEW hardware to be able to process novel materials under expanded hardware capacity, it is critical to consult with experts in electrical safety to ensure the highest standards of equipment safety are met.

### **Expanding Electrowriting Capacity through Multi-Disciplinary Research**

Analytical models for consolidating fluid behaviour and electrodynamics have yet to be realised to fully understand the relationship between material properties such as viscosity, molecular weight distribution, chain length and branching, concentration, melt and dielectric properties. In response, machine learning has been proposed as an alternative modelling tool to predict material printability based on a range of input experimental and modelling data sets [23]. This 'black-box' approach to establishing highly accurate models for predicting and guiding the optimal material and printing parameters required for successful electrowriting will be invaluable to the field for circumventing the highly complex material and process constraints using analytical modelling.

Substantial investment in novel biomaterials research has driven the development of more biomimetic, bioactive and sustainable materials for a range of biofabrication applications. Regulatory approval for materials has also been an essential component of discourse surrounding the potential clinical translation of electrowritten biomaterial structures [7]. This innovation in biomaterial development is coupled with strong mechatronics research engagement in the development of the next generation of electrowriting hardware to expand the capacity for materials with a greater breadth of viscoelastic profiles. In response, the biofabrication research community is only just beginning to unlock the full potential of electrowriting through a strong multidisciplinary approach towards personalised, biofabricated healthcare interventions.

## References

- 1 Robinson TM, Hutmacher DW, Dalton PD. The Next Frontier in Melt Electrospinning: Taming the Jet. *Adv. Funct. Mater.* 29(44), 1904664 (2019).
- 2 Liashenko I, Hrynevich A, Dalton PD. Designing Outside the Box: Unlocking the Geometric Freedom of Melt Electrowriting using Microscale Layer Shifting. *Adv. Mater.* 32(28), 2001874 (2020).
- 3 Saidy N, Wolf F, Bas O, Hutmacher DWDW, Mela P, De-Juan-Pardo EM. Biologically-inspired scaffolds for heart valve tissue engineering via melt electrowriting. *Small* 15(24), 1900873 (2019).
- 4 Paxton NC, Daley R, Forrestal DP, Allenby MC, Woodruff MA. Auxetic tubular scaffolds via melt electrowriting. *Mater. Des.* 193, 108787 (2020).
- 5 Saidy NT, Shabab T, Bas O *et al.* Melt Electrowriting of Complex 3D Anatomically Relevant Scaffolds. *Front. Bioeng. Biotechnol.* 8, 793 (2020).
- 6 Paxton NC, Lanaro M, Bo A *et al.* Design tools for patient specific and highly controlled melt electrowritten scaffolds. *J. Mech. Behav. Biomed. Mater.* 105, 103695 (2020).
- 7 Kade JC, Dalton PD. Polymers for Melt Electrowriting. *Adv. Healthc. Mater.* 10(1), 2001232 (2021).
- 8 Zhang Z, Jin Y, Yin J *et al.* Evaluation of bioink printability for bioprinting applications. *Appl. Phys. Rev.* 5(4), 41304 (2018).
- 9 Turlomousis F, Ding H, Kalyon DM, Chang RC. Melt Electrospinning Writing Process Guided by a “Printability Number”. *J. Manuf. Sci. Eng.* 139(8), 081004 (2017).
- 10 Brown TD, Dalton PD, Hutmacher DW. Direct writing by way of melt electrospinning. *Adv. Mater.* 23(47), 5651–5657 (2011).
- 11 Daneshfar A, Edwards SL, Dumée LF, Kong L, Hughes TC. Predicting Operating Rules for Successful Melt Electrowriting. *ACS Appl. Polym. Mater.* acsapm.0c01421 (2021).
- 12 Pang L, Paxton NC, Ren J *et al.* Development of mechanically enhanced polycaprolactone composites by a functionalized titanate nanofiller for melt electrowriting in 3D printing. *ACS Appl. Mater. Interfaces* 12(42), 47993–48006 (2020).
- 13 Hochleitner G, Kessler M, Schmitz M, Boccaccini AR, Teßmar J, Groll J. Melt electrospinning writing of defined scaffolds using polylactide-poly(ethylene glycol) blends with 45S5 bioactive glass particles. *Mater. Lett.* 205, 257–260 (2017).
- 14 Ding H, Cao K, Zhang F, Boettcher W, Chang RC. A Fundamental Study of Charge Effects on Melt Electrowritten Polymer Fibers. *Mater. Des.* 178, 107857 (2019).
- 15 Nahm D, Weigl F, Schaefer N *et al.* A versatile biomaterial ink platform for the melt electrowriting of chemically-crosslinked hydrogels. *Mater. Horizons* 7(3), 928–933 (2020).
- 16 Castilho M, Levato R, Bernal PN *et al.* Hydrogel-Based Bioinks for Cell Electrowriting of Well-Organized Living Structures with Micrometer-Scale Resolution. *Biomacromolecules* (2021).
- 17 Paxton NC, Ren J, Ainsworth MJ *et al.* Rheological Characterization of Biomaterials Directs Additive Manufacturing of Strontium-Substituted Bioactive

- Glass/Polycaprolactone Microfibers. *Macromol. Rapid Commun.* 1900019 (2019).
- 18 Hochleitner G, Hümmer JF, Luxenhofer R, Groll J. High definition fibrous poly(2-ethyl-2-oxazoline) scaffolds through melt electrospinning writing. *Polym. (United Kingdom)* 55(20), 5017–5023 (2014).
  - 19 Haigh JN, Dargaville TR, Dalton PD. Additive manufacturing with polypropylene microfibers. *Mater. Sci. Eng. C* 77, 883–887 (2017).
  - 20 Scott MA. Working Safely with Hazardous Capacitors: Establishing Practical Thresholds for Risks Associated with Stored Capacitor Energy. *IEEE Ind. Appl. Mag.* 25(3), 44–53 (2019).
  - 21 Lanaro M, Lu A, Lightbody-Gee A *et al.* Systematic Design of an Advanced Open Source 30 Bioprinter for Extrusion and Electrohydrodynamic based Processes. *Int. J. Adv. Manuf. Technol.* in press (2021).
  - 22 Petretta M, Desando G, Grigolo B, Roseti L. 3D printing of musculoskeletal tissues: impact on safety and health at work. *J. Toxicol. Environ. Heal. - Part A Curr. Issues* 82(16), 891–912 (2019).
  - 23 Ruberu K, Senadeera M, Rana S *et al.* Coupling machine learning with 3D bioprinting to fast track optimisation of extrusion printing. *Appl. Mater. Today* 22, 100914 (2021).