

Unique Aerodynamically Driven Methodology for Forming Droplets, Threads to Scaffolds

S. Arumuganathar,¹ S. N. Jayasinghe,¹ N. Suter²

¹Department of Mechanical Engineering, University College London, Torrington Place, London WC1E 7JE, United Kingdom

²Nisco Engineering AG, Dufourstrasse 110, 8008 Zurich, Switzerland

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ABSTRACT: Jet-based methodologies for forming droplets and threads leading to the fabrication of scaffolds are playing a pivotal role in materials science and engineering. These techniques are essentially interfacing engineering with the biological sciences. There are several routes for forming droplets, which are subsequently deposited with precision, namely, ink-jet printing to electrospaying. Fabricating threads/fibers sized from the nano- to the micrometer range are generally carried out by means of electrospinning. These three processes are well-established and have been explored to a great extent. In this article, we

have shown a unique processing technology completely driven by a pressure difference over an orifice possessing the ability to form droplets, threads to scaffolds. Hence, this process now joins the microfabrication race alongside its competing technologies, in the hope to undergo a rapid developmental program arming itself for competing with its rival technologies. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 104: 3844–3848, 2007

Key words: aerodynamically assisted jets; jetting; droplets; threads; scaffolds; patterning

INTRODUCTION

Droplet generation by means of advanced processing methodologies have recently been highlighted in the physical and life sciences as these routes play a major role for handling the desired materials for either placing with precision or for the fabrication of prearranged complex architectures. Two such technologies competing head to head are, namely ink-jet printing¹ and electrospaying.² These two techniques generate droplets through different mechanisms that distinguish them from each other. The former exploits thermal or piezoelectricity within needles, squeezing out a droplet via a needle, subsequently using electrostatics^{3,4} for guiding these droplets to their desired destinations. Ink-jets have been explored in several fields of research and have shown a significant contribution to that respective discipline. However, this technology has a resolution limit which generally is in the micrometer remit. Therefore it could be said that ink-jets have essentially set the scene for “jet-based processing sciences.”

The latter, electrospays, charges media within a needle later exposing it to an electric field, which promotes jet formation and break-up for generating droplets, here a counter electrode, which is grounded, is used for either converging or diverging^{5,6} the spray of droplets to predetermined locations. Electrospays unlike ink-jets have no limitations on the generated droplet sizes allowing the process to form droplets from the nano- to the micrometer size.^{7–9} Till recently electrospinning,^{10,11} a sister technology of electrospays, where the formed jet does not undergo break-up but thins forming a continuous thread from the media cusp, has been shown to have the ability for generating nano- to micrometer sized filaments^{12,13} to scaffolds. These scaffold morphologies could also be controlled,¹⁴ this route has been demonstrated as a versatile methodology for forming continuous fibers. We recently showed that electrospays which forms individual charged droplets could be used to form continuous polymeric threads by electrospaying a tailor made living sol.¹⁵ Hence, not only does this discovery allow us to have greater control, over the fabrication process, unlike in electrospinning but it also enables us to deposit droplets in desired locations where structures are formed, from which interconnects to over hangs could be fabricated linking complex architectures from one to another.¹⁶ This electrospay fabrication process is known as “electrohydrodynamic jet assembly” where the charge in droplets are exploited to assemble structures.

Correspondence to: S. N. Jayasinghe (s.jayasinghe@ucl.ac.uk).

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In all the above three processing methodologies there are obstacles and hazards that limit the fabricated structural resolution to the hazardous nature to the operator as high voltage is employed, respectively. Here we demonstrate a unique and versatile jetting approach which results from aerodynamic forces brought about by a pressure difference over an exit orifice, which is elucidated for generating, droplets and fragments to continuous fibers finally forming controlled scaffolds. Hence this jetting approach now joins the fabrication race.

EXPERIMENTAL

Materials

The polydimethylsiloxane (PDMS) media used in this study had a viscosity of 12,500 mPa s and was supplied by Polymer Systems Technology Limited, High Wycombe, UK. The properties of the PDMS medium were provided by the supplier. The density and surface tension of this media were 970 kg m^{-3} and 21.5 mN m^{-1} , respectively.

Aerodynamic jet device set-up

The aerodynamically assisted jetting device was set-up as previously described,¹⁷ briefly the device has a chamber with an internal height and diameter of 16.2 and 8.2 mm, respectively, made of stainless steel. A needle with threads was fitted firmly in place w.r.t. the chamber, which sealed the unit from above. The needle used in these studies has an internal orifice diameter of 0.35 mm. When the needle is in place the exit orifice is $\sim 0.2 \text{ mm}$ below the jetting needle. The exit orifice was counter sunk externally and has a similar diameter to that of the internal orifice of the needle. Another input exists that is placed on the side of the chamber, which supports the flow of the regulated pressure into the chamber giving rise to the pressure difference over the exit orifice assisting in the formation of a jet. The needle accommodating the flow of media into the chamber has a syringe connected to it via silicone tubing to a hypodermic needle. The syringe is placed on a precision syringe pump capable of modulating consistent low flow rates of up to $10^{-20} \text{ m}^3 \text{ s}^{-1}$ (Model type PHD 4400, HARVARD Apparatus, Edenbridge, UK). The compressed air into the chamber is digitally regulated by means of a precision pressure regulator having a resolution of $\pm 0.01 \text{ bar}$ from a compressed air supply of $\sim 6 \text{ bar}$.

Microscopy

The prepared droplet and thread residues at selected operational conditions in these investigations were

collected onto glass microslides at approximately the same point below the device exit orifice. Collection of these generated residues was carried out by swiftly moving a microslide in and out of the droplet and thread region. The residues as collected were micrographed using a Leica microscope with special add-on optics together with Leica analysis software. This software enabled the generated droplets, fragments, and threads to scaffold residues to be characterized.

RESULTS AND DISCUSSION

It was most important to map an operational guide in which we were able to identify regions where the finest droplets and threads were generated. Hence, we pursued this by first, setting the flow rate to the lowest possible flow that is allowed by our existing equipment ($10^{-20} \text{ m}^3 \text{ s}^{-1}$), subsequently via regulated pressure, aerodynamic forces were applied to the media exiting the needle. At these very low flow rates it was seen that for a correspondingly low applied pressure jetting did not take place but instead we observed that the pressure gave rise to the formation of bubbles in the silicone tube holding the flow of PDMS medium resulting in back flow. Therefore, we increased the flow rate and repeated this protocol till we managed to map a large region, in which the finest possible droplets and threads were formed. The parametric space investigated in this study had a flow rate of 1–10 mL/h via a 10 mL syringe for an applied chamber pressure of 0.01–5 bar (Fig. 1).

It is seen from the operational map that this medium gives rise to the formation of a two phase operational guide having defined regions where droplets and threads are formed. The line demarcating a fence in between these two regions was seen to vary slightly but overall the variation was insignificant.

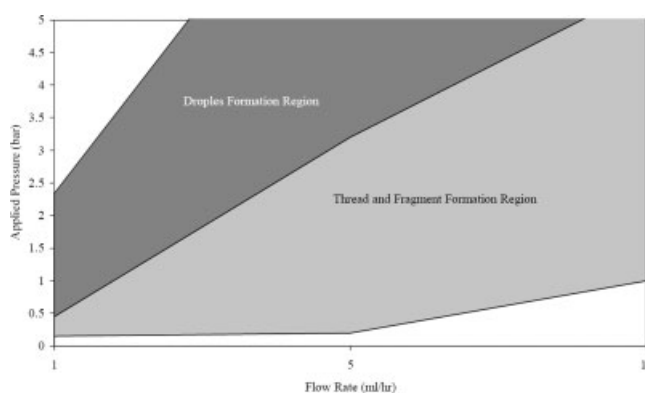
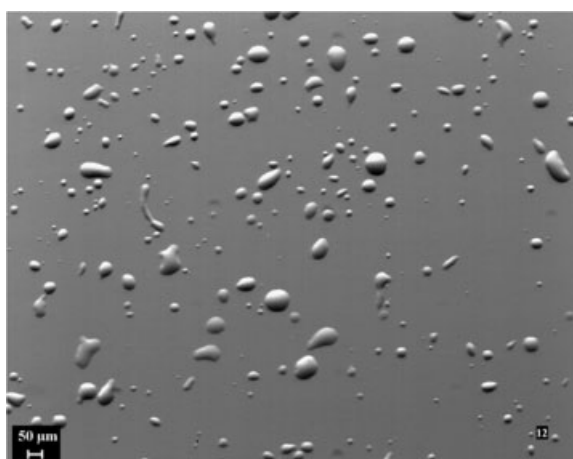
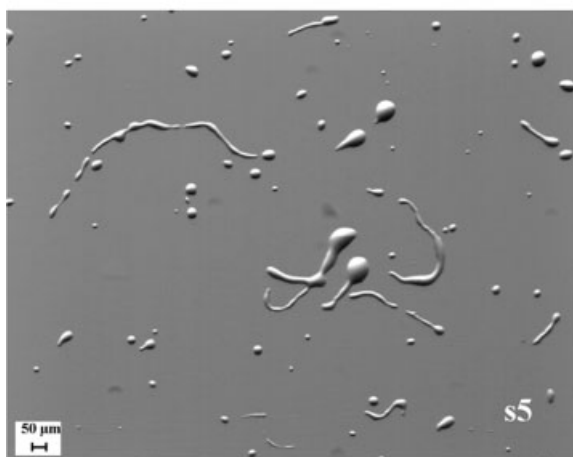


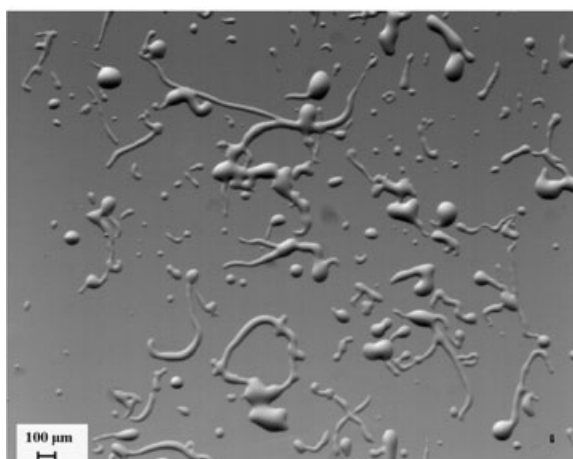
Figure 1 Operational map generated for the polydimethylsiloxane medium jetted for a parametric space of flow rate 1–10 mL/h via a 10 mL syringe for an applied chamber pressure of 0.01–5 bar.



a



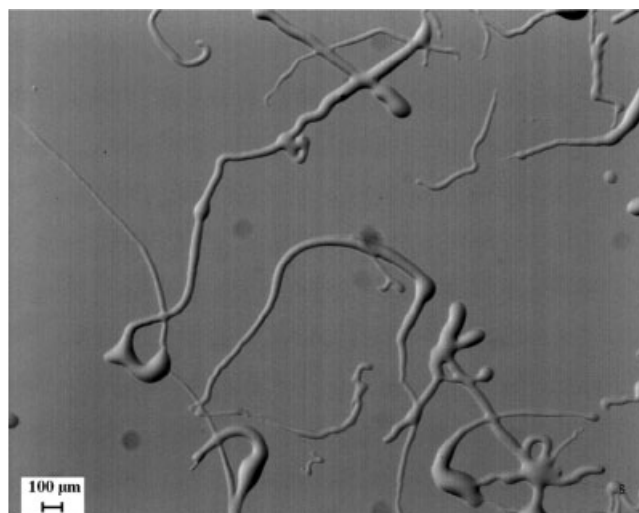
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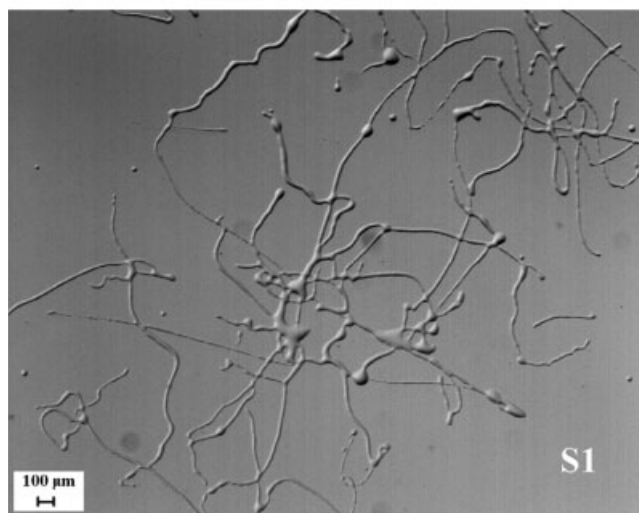
c

Figure 2 (a–c) Typical optical micrographs of collected residues on crossing from the droplet to threaded region in the generated operational guide depicted in Figure 1.

On selecting an applied pressure to flow rate in which we crossed the fenced region we clearly observed that droplets [Fig. 2(a)] are formed in the demarked region and on nearing the fenced area crossing over to the threading region the collected droplet residues had an elongated component attached to the droplet residues which we refer to as tails [Fig. 2(b)]. On crossing over to the thread region but still very close to the demarking fence we find elongated fragments being formed [Fig. 2(c)]. On moving beyond this point we found the generation of continuous threads which varied in diameters in the micrometer range as a function of both the applied pressure and flow rate [Fig. 3(a,b)]. This droplet to thread generation is reversible.



a



b

Figure 3 (a, b) Characteristic optical microscope images showing effect on the diameter of the formed microthreads as a function of applied pressure to flow rate.

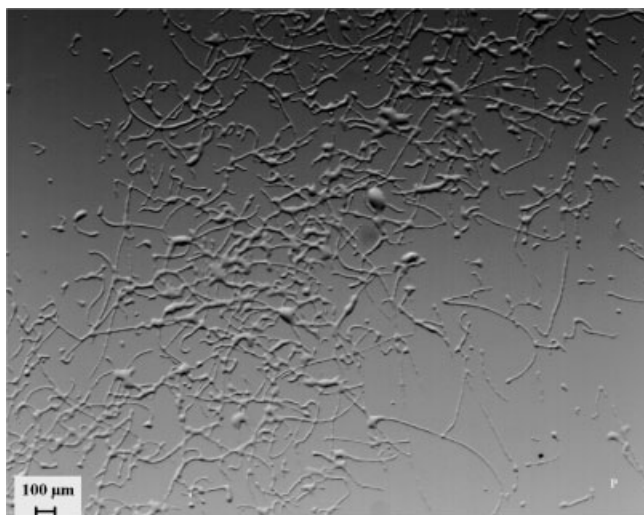


Figure 4 Typical collected residue of a formed scaffold.

On the achievement of forming continuous threads [Fig. 3(a,b)], we collected these residues and formed a scaffold (Fig. 4). Such scaffolds are most important for biologically-related research. We previously mentioned that threads and scaffolds are generally formed by means of electrospinning although recently electrosprays have also been shown to have the ability. Scaffolds formed by means of electrospinning have been shown to have much control over the fiber alignment. However, these electrospun fibers have not been investigated in detail with respect to their precision deposition to a preorganized architecture. We show here that using this continuous thread generation technique, we could not only form threads but also confine them to selected regions hence much like a two- [Fig. 5(a)] and three-dimensional pattern, like those fabricated by means of fused deposition modeling.¹⁸ The authors are vigorously pursuing the construction of a three-axes plotting device [Fig. 5(b)], which will hold the jetting device in place while the substrate will be moved in three-dimensions for the fabrication by deposition of prearranged scaffolds forming a porous track. Such structures when fabricated with bio-inspired materials would have huge implications in cell proliferation type studies to the formation of bio-foams and porous architectures required for harnessing or breeding cells.

CONCLUSIONS

We have shown a novel route for forming droplets, threads, and scaffolds from high viscosity media using aerodynamic forces brought about by an applied pressure over a chamber orifice. Our investigations have mapped an operational guide indicating regions where droplets and threads are formed and

their transformation. The article also highlights this processing route as a versatile technique for process fabricating scaffolds, which have widespread applications in the bio-world. Finally, we elucidate the ability to control deposit these scaffolds to predetermined patterns as porous tracks which other competing technologies have not been shown to possess. The use of PDMS in this study was essentially carried out for highlighting the ability of processing high viscosity media, this could be changed to a biopolymer or a polymer composite (having the desired nanoparticles suspended in the polymer) having a high molecular weight mimicking the viscosity investigated here for forming such architectures. Currently we are investigating the formation of a single continuous thread where on deposition into a reactive solution (assisting in rapid solidification) on impact will maintain thread integrity in terms of cylindrical cross-section to forming scaffolds having overlaying threads which will not fuse or attached to one another. The authors also wish to follow this work with a detailed investigation in to the processing

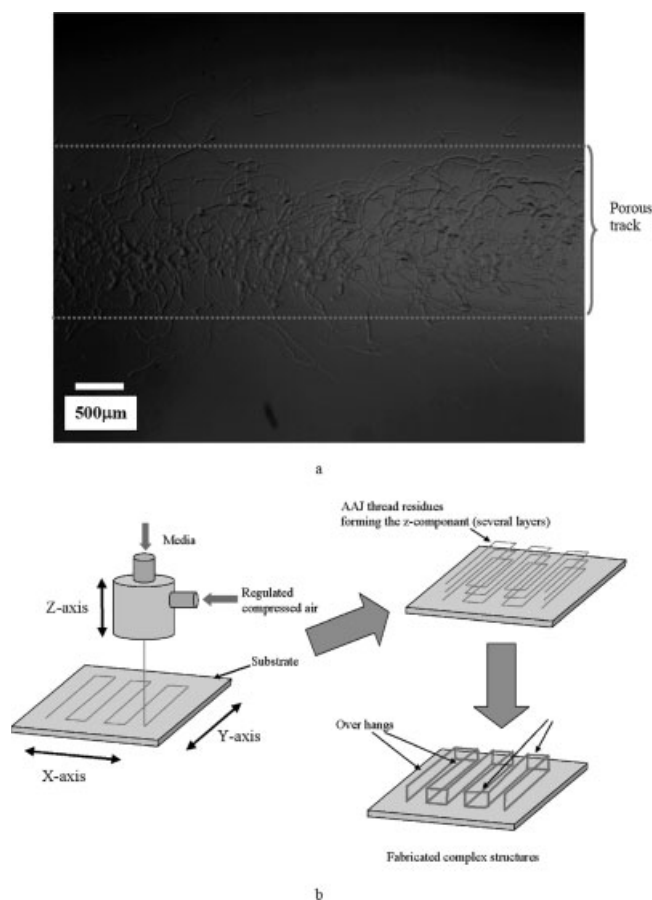


Figure 5 (a) Representative optical micrograph of a patterned porous scaffold having a width of $\sim 1500 \mu\text{m}$ and (b) schematic representation of the solid freeform fabrication device under construction for the creation of three-dimensional porous structures.

of a matrix having a combination of a biopolymer (for e.g., alginate, carrageenin or pectine) together with living organisms for the fabrication of a three-dimensional bio-inspired structure.

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