Additively Manufactured Externally-fed Electrospay Sources

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Sponsorship: MIT-Tecnologico de Monterrey Nanotechnology Program

Additive manufacturing (AM) is a layer-by-layer fabrication technique that creates solid objects by putting material where needed, instead of removing material from stock. Recent advances in AM have made possible the implementation of microsystems that surpass the performance of state-of-the-art counterparts made in a clean room, as well as the demonstration of devices that are challenging or unfeasible to create using standard microfabrication—particularly in the area of microfluidics. In addition, AM is inherently compatible with implementing, with great precision, hierarchical structures with features spanning orders of magnitude in size to accomplish multiple tasks efficiently.

In this project, we are exploring AM to develop, at a low-cost, massively multiplexed externally-fed electrohydrodynamic liquid ionizers (Figure 1) for a wide range of applications such as mass spectrometry, nanosatellite propulsion, species transport, and agile manufacturing. These devices are mesoscaled arrays of high-aspect-ratio, hundreds-of-microns tall, micron-sharp tips that are conformally covered with a nanostructured layer that transports and regulates the flow of liquid from the reservoir to the emission sites. Manufacturing issues such as inter-process compatibility and tip array uniformity need to be addressed to implement devices that operate efficiently successfully. Current work focuses on exploring and optimizing various manufacturing techniques to monolithically create the electrospray source out of different structures made of different materials; future work includes assessment of device performance, e.g., emission characteristics and uniformity.

FURTHER READING

Additive Manufacturing of Microfluidics via Extrusion of Metal Clay

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Sponsorship: MIT-Tecnologico de Monterrey Nanotechnology Program

Most microfluidics uses closed microchannels to efficiently accomplish tasks such as species mixing, heat transfer, and particle sorting by increasing the surface-to-volume ratio of the fluid(s) involved in the process. However, the current manufacturing techniques for microfluidics present disadvantages such as high-cost, long production time, no device customization, elaborated design iteration, restriction in the kinds of structures that can be made, and low fabrication yield.

Recent research results demonstrate that additive manufacturing can readily address the shortcomings outlined, often yielding devices that surpass the state of the art or for which traditional microfabrication creates no counterpart. However, most 3D-printed microfluidics are made of polymeric feedstock, which is not compatible with high-pressure and/or high-temperature applications. Mainstream 3D printing methods for metal include lost-wax micro molding, inkjet binder, and direct metal laser sintering; these processes are unideal to produce monolithic closed-channel microfluidics because they either require internal dummy structures or create internal voids filled in with unprocessed printable material, both of which are challenging to remove from the printed part.

In this project, we are exploring the use of extrusion of metal clay to implement closed-channel microfluidics; the technique is arguably similar to fused filament fabrication and can readily create voids without spurious infill or post-processing required. Via the extrusion of metal clay, leak-tight metal microchannel with monolithic, working ports have been created (Figure 1). A cross-section of the microfluidic shows an unclogged microchannel, evidencing the feasibility of the technique to create closed channels with hydraulic diameters of relevance to microfluidics (Figure 2). Current work focuses on exploring the design space of the technology and demonstrating an application of relevance.

FURTHER READING


▲ Figure 1: Extruded monolithic microfluidic with 350-µm wide and 350-µm tall microchannel and inlet and outlet ports.
▲ Figure 2: Cross section of closed microchannel.
Many compact systems use pumps to precisely set flow rates of liquid or, in general, to manipulate small liquid volumes for effective mass transport, cooling, or momentum transfer. Numerous microfabricated positive displacement pumps for liquids with chamber volumes that are cycled using valves have been proposed. Pumps made via standard (i.e., cleanroom) microfabrication typically cannot deliver large flow rates without integrating hydraulic amplification or operating at high frequency due to their small pump chambers.

Additive manufacturing, i.e., the layer-by-layer fabrication of objects using as template a computer-aided design model, has recently been explored as a processing arena for microsystems. In particular, researchers have reported 3-D printed pumps for liquids and gases with performance on par or better than counterparts made with standard microfabrication. Building upon earlier work on printed MEMS magnetic actuators, we recently developed miniature liquid pumps printed in pure nylon 12 via fused filament fabrication (FFF) whereby a thermoplastic filament is extruded from a hot nozzle to create a solid object layer by layer.

Our low-cost, leak-tight, miniature devices are microfabricated using 150- to 300-µm layers with a multi-step printing process (Figure 1) that monolithically creates all key features with <13-µm in-plane misalignment. Each pump has a rigid frame, a 21-mm-diameter, 150-µm-thick membrane connected at its center to a piston with an embedded magnet, chamber, passive ball valves, and two barbed fluidic connectors (Figure 2). Pump fabrication under 2 hours and costs less than $4.65 are achieved. Finite element analysis of the actuator predicts a maximum stress of 18.7 MPa @ 2-mm deflection, about the fatigue limit of nylon 12 (i.e., 19 MPa). A maximum water flow rate of 1.37 ml/min at 15.1 Hz actuation frequency is calculated, comparable to reported values of miniature liquid pumps with up to 200X higher actuation frequency.

**FURTHER READING**

Microfluidic devices are conceptually an ideal platform for the provision of personalized medical evaluations as they require small analyte volumes and facilitate rapid and sensitive investigations. However, inherent challenges in device fabrication have impeded the widespread adoption of microfluidic technologies in the clinical setting. Additive manufacturing could address the constraints associated with traditional microfabrication, enabling greater microfluidic design complexity, fabrication simplification (e.g., removal of alignment and bonding process steps), manufacturing scalability, and rapid and inexpensive design iterations.

We have developed an entirely 3D-printed microfluidic platform that enables modeling of interactions between tumors and immune cells, providing a microenvironment for testing the efficacy of immunotherapy treatment. The monolithic platform allows for real-time analysis of interactions between a resected tumor fragment and resident or circulating lymphocytes in the presence of immunotherapy agents. Our high-resolution, non-cytotoxic, transparent device monolithically integrates a variety of microfluidic components into a single chip, greatly simplifying device operation vs. traditionally-fabricated microfluidic systems. The 3D-printed device sustains viability of biopsied tissue fragments under dynamic perfusion for at least 72 hours while enabling simultaneous administration of drug treatments, illustrating a useful tool for drug development and precision medicine for immunotherapy. Confocal microscopy of the tumor tissue and resident lymphocytes in the presence of fluorescent tracers provides real-time monitoring of tumor response to various immunotherapy. The platform and accompanying analysis methods distinguish between a positive immune response and a lack of tissue response in the presence of immunotherapeutic agents.

This platform introduces novel methodologies in modeling and analyzing tumor response to improve prediction of patient-specific immunotherapy efficacy. To the best of our knowledge, this is the first report of human tumor fragments cultured in a dynamic perfusion system capable of testing the effect of circulating immune checkpoint inhibitors on resident tumor-infiltrating lymphocytes.

FURTHER READING


▲ Figure 1: An optical picture of a 3D-printed, transparent, non-cytotoxic microfluidic platform for analysis of the efficacy of immunotherapy, with features labeled.

▲ Figure 2: Overlaid bright-field and fluorescence images enable visualization of device geometries in addition to the stained tumor fragment. Single cells are visible.
Electrohydrodynamic Printing of Ceramic Piezoelectric Films for High-frequency Applications

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Sponsorship: MIT–Tecnológico de Monterrey Nanotechnology Program

The high operating frequencies that ceramic piezoelectric ultra-thin films attain have made possible exciting applications such as energy harvesting, telecommunications’ filters, high sensitivity biosensors, and acousto-fluidic devices; however, the inherently high cost and complexity of current manufacturing methods limit, in general, their widespread use. Additive manufacturing (AM), which has proven successful in creating complex devices and components of relevance to micro and nanosystems, could overcome these disadvantages; nevertheless, AM of piezoelectrics has been achieved only with polymer-based materials—unsuitable for said applications.

We report the first additively manufactured ceramic ultra-thin piezoelectric films compatible with high-frequency applications using electrohydrodynamic deposition (EHD) at room temperature. The films were made by electrospaying a zinc oxide (ZnO) nanoparticle liquid feedstock, directly writing line imprints as thin as 213 nm and as narrow as 198 µm.

We harness a previously unreported effect to align the polar axis of the imprint and obtain overall piezoelectricity. As Figure 1a shows, the (100) orientation monotonically increases as the linear density of the deposition is reduced by increasing the raster speed or reducing the feedstock flow rate (Q)—provided two conditions are met: the feedstock is ionized (via EHD), and a small separation between emitter and substrate is used. Notably, the orienting effect directly acts on the direction of the polar axis by means of the rastering direction (Figure 2a), allowing for vibration modes and resonator configurations that were previously unfeasible. The macroscopic piezoelectric behavior is shown through piezoforce response microscopy (PFM) (Figure 2b) and the suitability for high-frequency applications was demonstrated by testing thin-film bulk acoustic resonators (FBAR) on a flexible polymer substrate, where the resonant frequency of ~5 GHz was used to calculate the acoustic speed of the films (~2,000 m/s), which is close to the transversal wave speed of ZnO.

Further Reading

A corona discharge is a high-electric field ionization phenomenon caused by the development of a self-sustained electron avalanche between a sharp electrode (i.e., corona electrode) and a blunt electrode; the ions create a plasma region around the corona electrode and in their travel to the opposite electrode transfer momentum to the surrounding fluid. In this project, we are harnessing advanced metal inkjet printing technology to demonstrate massively multiplexed MEMS corona discharge ionizers (Figure 1), with the aim to increase greatly their ionization throughput and optimize their transduction mechanism to be able to implement exciting applications such as no-moving-parts pumps for gases and compact ion mobility spectrometers.

A 1D electrohydrodynamic coaxial cylinder model was implemented in COMSOL Multiphysics to study the ionization and collision processes in air at atmospheric pressure and room temperature of a 1-tip device, predicting a 400-µm-thick corona region surrounding the corona tip. The onset voltage estimated from the simulation is 5.849 kV, which is close to the theoretical value from Peek’s formula of 6.416 kV. In addition, current over voltage (I/V) versus bias voltage minus the onset voltage (V-V0) characteristics were collected for different ionizer array designs while varying the separation between the corona electrode and the collector electrode; the data follow the Townsend current-voltage model (Figure 2). Moreover, the data show that the corona current decreases with increased spacing of the corona electrode-to-collector electrode due to the decrease of the electric field on the tips; however, a smaller separation between the corona electrode and the collector electrode results in larger fluctuations in the corona discharge current. Devices with different numbers of tips tend to generate the same total corona current at the same bias voltage although more tips are set to discharge as the number of tips increases; this increase can be ascribed to the stronger interference between adjacent tips when the tip-to-tip spacing decreases. Current research efforts focus on optimizing the array design to minimize electric field shadowing and sharpening the tips to achieve operation at a lower bias voltage.

**FURTHER READING**

Mass spectrometers are powerful chemical analytical instruments used to quantitatively characterize the composition of unknown samples via ionization and mass-to-charge ratio species sorting. However, mainstream mass spectrometers are large, heavy, power hungry, and expensive, limiting their applicability in real-time and in-situ applications. Gas molecules can be ionized via electron impact ionization (EI), for which a source of electrons, i.e., a cathode, is required. Cold cathodes emit electrons into a vacuum via quantum tunneling due to high surface electric fields that lower and narrow the barrier that traps electrons within the material; typically, high-aspect-ratio, nano-sharp tips are used to produce such fields with moderate bias voltages. Compared to thermionic cathodes, field emission electron sources have faster response and less power consumption. Compared to other field emitters, carbon nanotubes (CNTs) are less affected by back-ion bombardment and chemical degradation. There are numerous reports of gas ionizers with CNT cathodes EIIs; however, these devices are microfabricated using clean-room technology and/or use ion-generating structures machined with standard technologies, affecting their cost and size.

In this project, we are harnessing additive manufacturing (AM) to develop novel electron impact ionizers that circumvent these challenges. AM has unique advantages over traditional manufacturing methods including compatibility with creating complex 3D geometries, print customization, and waste reduction. Our design (Figure 1) uses inkjet binder printing of SS 316L to create electrodes to efficiently generate ions and steer charged species, stereolithography of polymer resin to define the dielectric structures that electrically isolate the different electrodes, and an additively manufactured CNT electron source. We have successfully characterized the ionizers at pressures as high as 5 mTorr while achieving ionization efficiencies as high as 8.5% (Figure 2).

**FURTHER READING**

Field emission cathodes are promising electron sources for exciting applications such as flat-panel displays, free-electron lasers, and portable mass spectrometry where fast switching, low-pressure operation, and low power consumption are favored metrics. A field emitter quantum tunnels electrons to a vacuum due to the high electrostatic fields at its surface; this tunneling is typically done at low voltage using a whisker-like structure. Carbon nanotubes (CNTs) are attractive structures to produce electron field emission due to their ultrasharp tip diameter, high aspect ratio, high electrical conductivity, and excellent mechanical and chemical stability. Although CNT-based cold cathodes have been widely reported in the literature, their manufacture could be quite expensive (e.g., devices partially or fully made in a semiconductor cleanroom), or the extractor electrode of the cathode is an external mesh, causing high-beam interception (e.g., in screen-printed devices) or requiring an advanced method of assembly to the emitting component to achieve high transmission.

In this project, we are developing novel field emission sources that are fully additively manufactured to circumvent the aforementioned challenges. The devices are made via direct ink write (DIW) printing, which is one of the least expensive and most versatile additive manufacturing methods as is capable of monolithic multi-material printing. Compared to screen printing, DIW does not involve static masks to transfer patterns and produces significantly less waste. The fully-printed field emission electron source is composed of two continuous imprints: a spiral trace made of a CNT compound, acting as an emitting electrode, symmetrically surrounded on both sides by a spiral trace made of silver nanoparticles, acting as in-plane extractor electrode (Figure 1). After printing, the CNT spiral receives a mechanical treatment that releases the CNT tips from the bulk of the imprint (Figure 2), enabling field emission from the CNT imprint. Characterization of the printed CNT field emission sources in triode configuration (i.e., using an external anode) shows low turn-on voltage and low interception of the emitted current by the extractor electrode. Current work focuses on design optimization and experimental characterization of the devices.

**FURTHER READING**

Controlling the Nanostructure in Room-temperature-microsputtered Metal

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Sponsorship: MIT Lincoln Laboratory

Sputter deposition involves the ejection of atoms from a target and the atoms’ subsequent deposition on a nearby substrate. Because the deposition is done on the atomic level, the nanostructure of the deposit can vary significantly. This variance is of concern, as it can greatly affect material strength and conductivity. Traditional sputtering relies on vacuum and thermal annealing to ensure dense, highly conductive deposits. However, agile manufacturing on temperature-sensitive substrates is not compatible with these two solutions.

To enable high-quality material without heating the material or requiring a vacuum, we performed a statistically-motivated set of experiments to determine what deposition parameters improve the material quality. We developed an empirical model and found that an appropriate electrical bias voltage, applied either to the substrate or to a conductive plate under the substrate, has the greatest impact on the material quality. This is due to the presence of charged nanoparticles, formed by collisions between sputtered atoms in the dense plasma around the sputter target. The applied electric field attracts positively charged nanoparticles, allowing the nanoparticles to strike the substrate with more energy than their temperature alone would dictate. This extra energy enhances the mobility of the sputtered metal, allowing it to form denser, more energetically favorable coatings (Figure 1) without significant substrate heating. With this technique, we have improved the conductivity of the sputter coating to 5x bulk metal (15 µΩ·cm) at room temperature.

Applied electric fields also improve the coating’s thickness. In the absence of electric fields, the sputtering process is self-limiting. As the positively charged sputtered material reaches the substrate, charge builds upon the substrate, repelling charged sputtered material and preventing the deposit from thickening. However, biasing the substrate with a negative voltage prevents this charge from accumulating, allowing for thicker (> 200 nm) films.

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**FURTHER READING**

Neutron radiation is widely used in various applications, ranging from the analysis of the composition and structure of materials and cancer therapy to neutron imaging for security. However, most applications require a large neutron flux that is often achieved only in large infrastructures such as nuclear reactors and accelerators. Neutrons are generated by ionizing deuterium ($D_2$) to produce deuterium ions ($D^+$) that can be accelerated towards a target loaded with either $D$ or tritium ($T$). The reaction generates neutrons and isotopes of He, with the D-T reaction producing the higher neutron yield. Classic ion sources require extremely high positive electric fields, on the order of $10^8$ volts per centimeter (10 V/nm). Such a field is achievable only in the vicinity of sharp electrodes under a large bias, and consequently, ion sources for neutron generation are bulky.

This work explores, as an alternative, highly scalable and compact Si field ionization arrays (FIAs) with a unique device architecture that uses self-aligned gates and a high-aspect-ratio (~40:1) silicon nanowire current limiter to regulate electron flow to each field emitter tip in the array (Figure 1). The tip radius has a log-normal distribution with a mean of 5 nm and a standard deviation of 1.5 nm, while the gate aperture is ~350 nm in diameter and is within 200 nm of the tip. Field factors, $\beta$, > $1 \times 10^6$ cm$^{-1}$ can be achieved with these SI FIAs, implying that gate-emitter voltages of 250-300 V (if not less) can produce $D^+$ based on the tip field of 25-30 V/nm. In this work, our devices achieve ionization current of up to 5 nA at ~140 V for $D_2$ at pressures of 10 mTorr. Gases such as He and Ar can also be ionized at voltages (<100 V) with these compact Si FIAs (Figure 2).

**Further Reading**


**Figure 1:** Schematic of gated field ionization array, with SEM cross-section of a single field ionizer and photograph of a packaged chip with arrays of different sizes for neutron generation.

**Figure 2:** Ion current measured for different gases (He, Ar, and $D_2$) at 1 mTorr pressure demonstrating low ionization voltages using 1000 by 1000 Si FIAs.
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