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Electrohydrodynamic jet printed templates for hot embossing of microfluidic devices

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Abstract

Hot embossing is a scalable method of fabricating microfluidic devices involving precise replication of micrometer-sized features from a master mold onto a thermoplastic substrate. Typically, high-resolution master molds for hot embossing are fabricated using expensive, resource-intensive processes such as photolithography and electron-beam lithography. Here, we present a maskless, cost-effective, and rapid microfabrication process based on electrohydrodynamic jet printing (EJP) for fabricating high-resolution reusable master templates for hot embossing of thermoplastic microfluidic devices. Our method is based on EJP to fabricate intricate polymeric templates, with feature sizes of order $100\ \mu\text{m}$, followed by a double casting process to obtain stiff PDMS master molds. Using these PDMS molds, we demonstrate the hot embossing of microfluidic devices with excellent reproducibility across multiple embossing cycles. In particular, we demonstrate the fabrication of microfluidic devices with simple geometries of cross-shape and Y-shape to complex geometries of flow-focusing droplet generator and tree-shaped gradient generator. Subsequently, we demonstrate the use of hot-embossed microfluidic devices for hydrodynamic focusing, droplet generation, and stable concentration gradient generation. Our method offers a low-cost and rapid alternative to traditional lithographic processes for fabricating master molds for hot embossing with comparable feature resolution.

Supplementary material for this article is available [online](#)

Keywords: electrohydrodynamic jet printing, hot embossing, microfluidics, Lab-on-a-chip, microfabrication

1. Introduction

Thermoplastic polymers, such as polymethylmethacrylate (PMMA), polystyrene (PS), polycarbonate (PC) and cyclic olefin copolymer (COC) have found wide applicability for fabricating microfluidic devices owing to their favorable optical

qualities, thermal and electrical insulation, and biocompatibility [1]. Moreover, thermoplastic polymers are easily fabricated, as opposed to glass, while offering cost-effectiveness, rapid customization, and mass production capabilities. In addition to thermoplastics, polydimethylsiloxane (PDMS) is also a popular alternative to glass for microfluidic device fabrication [2]. However, PDMS structures have lower mechanical strength and rigidity when compared with thermoplastics and are prone to swelling when exposed to non-polar solvents [3]. Therefore, thermoplastics, with PMMA being the most popular option, remain the material of choice for

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fabricating microfluidic devices on laboratory and industrial scales.

Thermoplastic microfluidic devices can be fabricated using direct methods such as laser ablation and micromachining and replication methods such as injection molding, hot embossing, compression molding and casting [4]. For medium to large-volume fabrication of microfluidic devices with high precision and excellent reproducibility, hot embossing is an optimal process given its fast replication rate, low cost of instrumentation, and low cost per device [5]. Hot embossing is based on patterning a heated thermoplastic substrate with a template having positive features through controlled application of temperature, pressure and time [6].

Typically, to fabricate a metallic mold for hot embossing, micrometer-sized features are microfabricated on a glass plate or silicon wafer using standard etching and photolithography techniques [7]. Subsequently, a metallic layer (typically nickel) is electroplated onto the surface of the etched plate or wafer to obtain a metallic mold. The fabrication of micrometer-sized features using photolithography on a glass plate or wafer involves several essential processes, such as preparation of the photomask, coating silicon wafer with photoresist, alignment of the photomask, treatment with UV light, baking and developing with developer solution. As a result, these microfabrication steps make the overall process of fabricating master molds resource-intensive and expensive. In addition, a clean room facility and skilled operators are a must for mask fabrication using photolithography. Consequently, various maskless lithographic techniques, such as electron-beam and laser beam lithography, were introduced to overcome the dependency on photo mask requirements [8]. However, these processes are also expensive, cleanroom-based, and do not favor rapid customization. Consequently, master mold fabrication is typically limited to central facilities, which makes it impractical for rapid customization in a laboratory environment. To overcome the shortcomings of these processes, various alternative techniques to photolithography, including 3D printing [9], laser ablation [10], micro milling [11], and xurography [12] have been explored for rapid fabrication of customized, relatively low-cost master molds. However, these methods fail to achieve the high-resolution features offered by photolithography. Therefore, it is desirable to have a cost-effective master template fabrication technique that is swift and customizable, akin to inkjet printing, and capable of offering high-resolution features like photolithography and electron-beam lithography.

Over the past decade, electrohydrodynamic (EHD) jet printing (EJP), a direct ink writing method originally derived from electrospinning, has shown significant potential for printing high-resolution customized features with polymeric ink solutions [13]. In EJP, a polymeric solution or a molten polymer is flown through a conducting nozzle under the action of an external electric field ($\sim 10^6 \text{ V m}^{-1}$) acting between the nozzle and the substrate. The application of such a high electric field causes the spherical meniscus of the polymeric ink at the nozzle tip to deform into a cone called a Taylor

cone [14]. For a specific range of applied electric field and flow rate, a fine jet emerges from the cone tip, which can be precisely deposited on a moving substrate to obtain micrometer-sized features. Therefore, with the proper selection of operating parameters, EJP can be used to print features ranging from two orders of magnitude smaller than the needle diameter to sizes comparable to the needle diameter. Thus, micrometer resolution features can be precisely printed while using a relatively large diameter nozzle, thereby preventing nozzle clogging. The versatility of EJP has already been demonstrated for printing two-dimensional and three-dimensional patterns, including linear and curvilinear segments [13, 15]. In particular, the use of EJP has been demonstrated in various applications, such as printing polymeric scaffolds, microlens arrays, footpaths for electronics [13], patterned quantum dot LEDs [16], sacrificial templates for Si-based negative mold [17], and PDMS microfluidic devices [18–20]. However, the capability of EJP for fabricating non-sacrificial master templates for hot embossing of microfluidic devices has not been explored yet.

In this work, we demonstrate the use of PDMS-based positive master molds, derived from EHD-jet printed templates, for hot embossing of PMMA microfluidic devices. The first step of fabricating the master mold involves precise EHD-jet printing of polymeric solution of polycaprolactone (PCL) in acetic acid on a silicon wafer. Thereafter, a two-step replica molding process is used to obtain a stiff PDMS-based mold with positive features. The positive PDMS mold so obtained is then used for hot embossing of PMMA microfluidic devices. We demonstrate this approach for high-resolution fabrication of a variety of microfluidic devices such as cross-shaped, Y-shaped devices, flow-focusing droplet generator and tree-shaped gradient generator with channel widths of order $100 \mu\text{m}$. We also demonstrate the use of the fabricated PMMA devices for a wide range of microfluidic applications, such as hydrodynamic flow focusing, droplet generation, and stable concentration gradient generation.

2. Materials and methods

In this section, we provide the details of the experimental setup, the polymeric solution employed, and the operating parameters for EJP of master templates. Following that, we discuss the protocol for fabricating PDMS-based positive master molds using EHD-jet printed templates. Subsequently, we present the methodology for hot embossing of PMMA substrates using the fabricated PDMS-based positive master molds.

2.1. Experimental setup for EJP

Figure 1(A) shows a schematic diagram of the experimental set-up used for EJP of non-sacrificial master templates. A high precision syringe pump (KD Scientific Legato 100) was employed to flow the polymeric solution through a stainless steel needle (OctoInkjet, Model: NBMET25G1), with

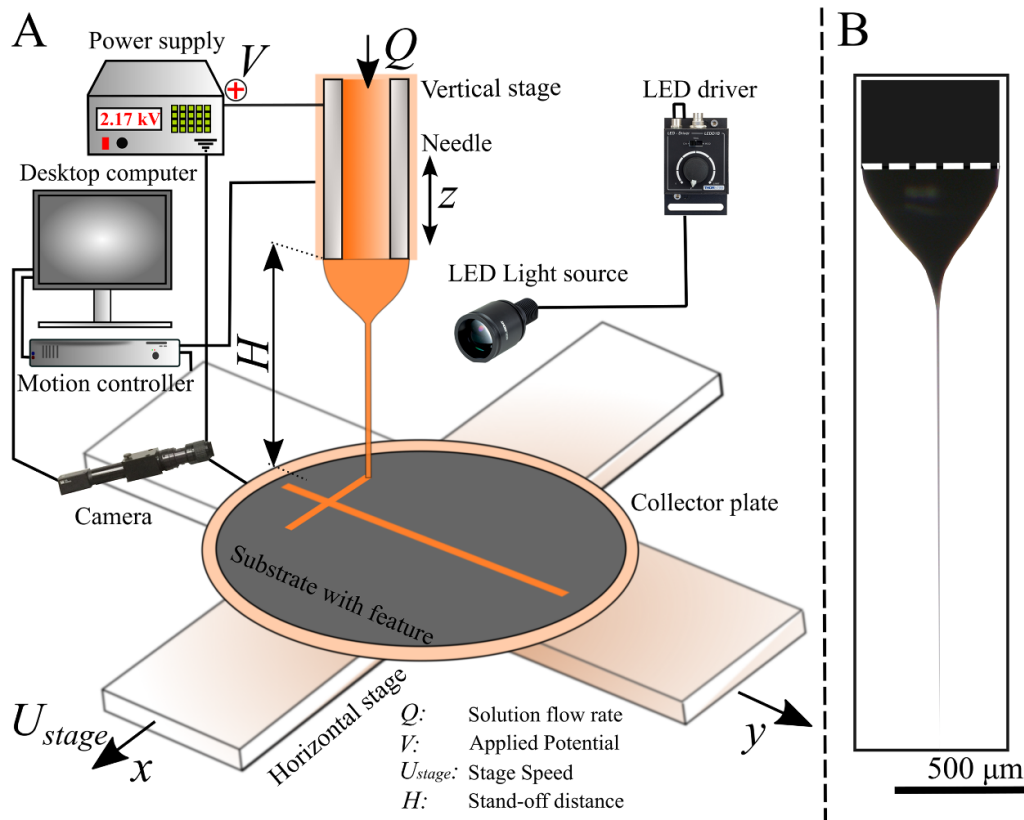


Figure 1. (A) Schematic representation of experimental set-up used for EHD-jet printing of a master template on a silicon wafer. (B) A representative snapshot of a stable cone-jet of PCL 40% (w/V) solution issuing out of a needle with an outer diameter of $500\ \mu\text{m}$. The stable cone-jet mode is used to print the intended feature on a silicon wafer.

a flat end, having inner and outer diameters of $250\ \mu\text{m}$ and $500\ \mu\text{m}$, respectively. A collector plate was mounted on a high-precision motorized horizontal stage (Newport, M-ILS200BPP, S/N: B197835 and B197836, USA) to enable precise translatory motion along x and y directions. Similarly, the needle was mounted on a vertical translation stage (Newport, MFA-PPD, S/N: B195047, USA) to control the needle-substrate gap along the z direction. The precision stages were connected to a motion controller (Newport, XPS-D4, USA), which in turn was connected to the desktop computer through a serial LAN port. A high potential difference was applied between the needle and the metallic collector plate using a high-voltage DC power supply (Ionics, 0–5 kV, 10 mA). The positive terminal of the power supply was connected to the metallic needle, and the collector plate was grounded. The EHD-jet printing was performed on a polished p-type silicon wafer, which was placed over the collector plate. The silicon wafer, having electrical conductivity of $\mathcal{O}(10\ \text{S m}^{-1})$, was two inches in diameter and $500\ \mu\text{m}$ in thickness.

An in-house-developed LABVIEW virtual interface was employed for programming and monitoring of stage position and motion along x , y and z directions, the flow rate of the polymeric solution, and potential difference across the needle and the collector plate throughout the printing process. The printing process was observed and recorded using a CMOS camera (IDS, UI-1220LE-M-HQ, $6\ \mu\text{m}$ pixel size) equipped

with a zoom lens and a collimated LED light source (Thorlabs, MCWHL6-C5, USA).

2.2. Fabrication of silicon-based master template

We used the EJP setup to print polymeric features on a silicon wafer, which were used as templates to fabricate master molds for hot embossing. The printing of templates for microfluidic devices requires (i) the geometry and the dimensions of the microchannels, (ii) the polymeric solution to be used in the printing process, and (iii) the operating parameters for EJP. We chose the microchannel geometries ranging from simple cross-shape and Y-shape designs to complex geometries of flow-focusing droplet generator and tree-shaped gradient generator. Figures 2(A)–(D) show the geometries and dimensions of the chosen master templates to be printed on a silicon wafer.

To prepare the polymeric solution, we chose PCL (average $M_n \sim 45\ 000$ Da, Sigma Aldrich) as the polymeric solute due to its non-toxicity, biocompatibility, biodegradability, and favorable mechanical properties [21]. PCL was dissolved in acetic acid (CH_3COOH , Sigma Aldrich, 99.7% pure) to obtain the polymeric ink. For all experiments, we used a homogeneous polymeric solution of 40% (w/V) PCL dissolved in acetic acid. For the preparation of this homogeneous solution, PCL pellets were dissolved in acetic acid solution and

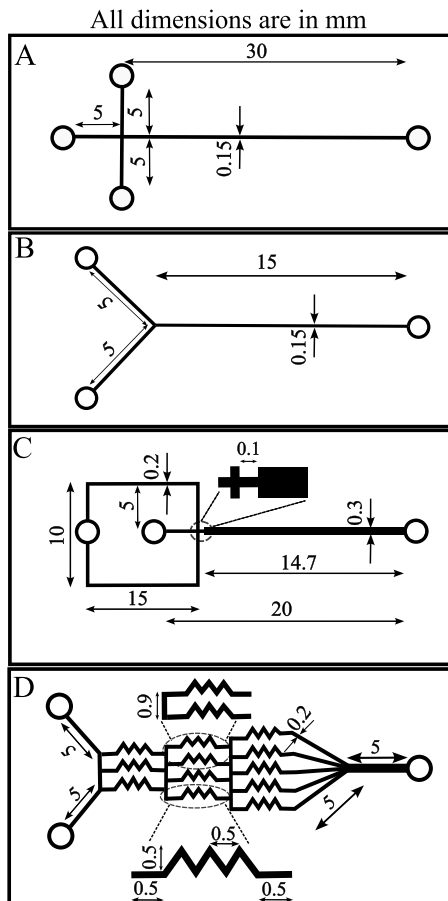


Figure 2. Design and dimensions of master templates to be printed on a silicon wafer using PCL 40% (w/V) solution. (A)–(D) show geometries of templates for fabricating cross-shaped and Y-shaped microfluidic devices, a flow-focusing droplet-generator and a tree-shaped gradient generator, respectively.

stirred at 40 °C for 7–12 h at 700 rpm using a magnetic stirrer. Subsequently, the solution was degassed using a vacuum desiccator to eliminate air bubbles. We did not opt for dielectric polymers like PDMS as the electrical conductivity of PDMS is of $\mathcal{O}(10^{-13} \text{ S m}^{-1})$. Such a low value of electrical conductivity of PDMS precludes steady EHD-jetting, which occurs for leaky-dielectric liquids [22].

2.2.1. Influence of operating parameters. Among various modes of EHD jetting, we used the stable cone-jet mode, as shown in figure 1(B), for printing uniform features (refer to supplementary information Video S1 †). The stable cone-jet mode is the conventional mode of EHD-jet printing for depositing uniform, continuous micro/nano-sized patterns on a substrate that cannot be achieved using standard inkjet printers [13]. For a given composition of the polymeric solution, the stable cone-jet regime occurs for a specific range of applied electric potential (V), flow rate (Q), and stand-off distance (H) [23–25]. Further, the stage speed (U_{stage}) and ambient conditions [15, 26], are crucial in influencing the morphology and

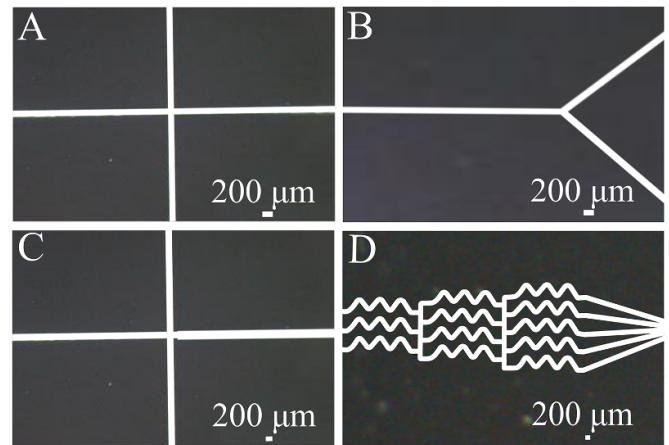


Figure 3. EHD-jet printed master templates with geometries of (A) cross-shaped, (B) Y-shaped, (C) flow-focusing droplet generator and (D) tree-shaped gradient generator microfluidic devices. The high resolution features were printed on a two-inch silicon wafer using a PCL 40% (w/V) solution.

uniformity of the printed features as well as the reproducibility of the printing process.

Several published works have investigated the influence of these operational parameters on the width of the printed feature [13, 15, 20, 26]. In particular, we followed the guidelines for choosing printing parameters provided in our earlier study [20]. Briefly, the width (w) of the printed feature scales with solution flow rate (Q) as $w \sim Q^{1/3}$. Also, the width of the printed feature decreases with an increase in applied potential difference (V) and stage speed (U_{stage}).

To print master templates corresponding to cross-junction and Y-junction devices of width $150\text{ }\mu\text{m}$, we chose $Q = 1.5\text{ }\mu\text{l min}^{-1}$, $V = 2.17\text{ kV}$, $H = 4\text{ mm}$, and $U_{\text{stage}} = 45\text{ mm s}^{-1}$ as the operating parameters. Figures 3(A) and (B) show the printed templates corresponding to cross-shaped and Y-shaped microfluidic devices. Similarly, for printing flow-focusing droplet generator and tree-shaped gradient generator of width $200\text{ }\mu\text{m}$, we chose $Q = 2.0\text{ }\mu\text{l min}^{-1}$, $V = 2.17\text{ kV}$, $H = 4\text{ mm}$, and $U_{\text{stage}} = 30\text{ mm s}^{-1}$ as the operating parameters. A lower value of U_{stage} and a higher value of Q ensured that templates for the droplet and gradient generator were printed with the required broader feature width. The resulting templates for the flow-focusing droplet generator and the tree-shaped gradient generator are shown in figures 3(C) and (D), respectively. We note that all the templates were fabricated using single-pass EHD-jet printing, wherein the deposition of liquid filament at a particular location was done only once. The printing was performed at an ambient temperature range of $20\text{ }^{\circ}\text{C}$ – $25\text{ }^{\circ}\text{C}$ and relative humidity range of 40% – 60% .

To check the repeatability of the process, we printed each feature ten times using the corresponding operating conditions and estimated the uncertainty in the dimensions of the printed features. We measured the widths of the printed features by placing the silicon wafer with the printed features under

an upright measuring microscope (Nikon MM400) coupled with a CMOS camera (Radical Scientific, Procam HS-5). The total uncertainty in the measurement of the width of the printed feature was dominated by random uncertainty. Therefore, we employed t -distribution to estimate the random uncertainty [27], which was less than 2% of the mean value of feature width. The measured values of widths for templates with cross-shaped and Y-shaped features were $150.6 \pm 1.8 \mu\text{m}$ and $149.4 \pm 1.6 \mu\text{m}$, respectively. Similarly, for the flow-focusing droplet generator and tree-shaped gradient generator, the measured widths were $202.2 \pm 1.8 \mu\text{m}$ and $201.6 \pm 1.5 \mu\text{m}$, respectively. This low uncertainty in the widths of the printed features highlights the repeatability and reliability of the EJP process.

Having established the reliability of the printing process, we used the printed templates for the hot embossing process to fabricate PMMA microfluidic devices. We note that the melting point of PCL is 60°C , which restricts the direct usage of the printed templates in the hot embossing process. Therefore, we used the printed templates to fabricate PDMS-based positive molds using the double casting process, which were subsequently used for hot embossing of PMMA microfluidic devices.

2.3. Fabrication of PDMS-based master mold

Here, we describe the replication process to fabricate PDMS molds from EHD-jet printed templates and their subsequent use for hot embossing of PMMA microfluidic devices. The EHD-jet printed PCL templates were used to fabricate PDMS-based positive master molds using the double casting process of Wilson *et al* [11]. Figure 4 provides a step-by-step protocol for the double casting process. First, a 10:1 (w/w) PDMS-curing agent (Sylgard 184, Dow Chemicals) mixture was prepared and then degassed for 25 min in a vacuum desiccator. Next, the 10:1 PDMS mixture was poured over the PCL template and baked at 55°C for 2–3 h in a hot air oven, as shown in figures 4(A) and (B). Finally, the cured PDMS negative replica was carefully peeled off from the non-sacrificial printed template (refer to supplementary information video S2 †), as illustrated in figures 4(C) and (D).

Figures 4(E)–(G) show the second step of the double casting process wherein a PDMS-curing agent mixture in the 4:1 (w/w) ratio was prepared and poured onto the negative replica obtained from the previous step. To prevent adhesion between two PDMS parts, before pouring the 4:1 PDMS mixture, three $20 \mu\text{l}$ drops of chlorotrimethylsilane (Sigma Aldrich, P code: 101678597) were applied to the surface of the negative replica. This step ensures seamless detachment of the positive PDMS replica from its negative counterpart. The poured 4:1 PDMS mixture was then cured in a hot air oven maintained at 90°C for 25 min to get a PDMS-based positive master mold. The 4:1 PDMS positive master mold was then placed on a glass slide and subjected to thermal ageing on a hot plate at 275°C for 25 min as shown in figure 4(H).

To ensure that the 4:1 PDMS mold is stiff enough for the hot embossing of PMMA substrates, we measured the Young's modulus of a thermally aged 4:1 PDMS specimen and a 10:1 PDMS-based specimen using a universal testing machine (UTM, SHIMADZU, AG-X plus, Japan). The measured Young's modulus of the specimen with a 4:1 composition was $3.33 \pm 0.2 \text{ MPa}$ in comparison to $1.35 \pm 0.1 \text{ MPa}$ for the 10:1 PDMS specimen (refer to figure S1 † of the supplementary information). This suggests that the 4:1 PDMS mixture results in a significantly stiffer mold. The increased amount of curing agent in the 4:1 PDMS composition ensured higher stiffness, improved thermal stability and reduced thermal expansion, making it suitable for hot embossing [28]. Moreover, the low surface energy of PDMS facilitates a simple and user-friendly demolding process post embossing.

2.4. Hot embossing of PMMA slides

After replicating a positive PDMS mold from the printed PCL template, we used the PDMS mold for the hot embossing of PMMA slides. To this end, we use the simple hot embossing technique of Goral *et al* [29], wherein we sandwiched the PDMS mold and a fresh PMMA slide with two glass slides at an elevated temperature, as shown in figure 4(I). We used four paper binder clips (ODDY, India) to apply the embossing force and a hot air oven to heat the substrate-mold pair. This arrangement is cost-effective and suitable for the swift fabrication of customized PMMA microfluidic devices in the laboratory environment.

2.4.1. Parameters of hot embossing process. The key physical parameters that govern the embossing process are the embossing temperature (oven temperature), embossing force (force applied by the binder clips), embossing time (time of heating the assembly inside the oven), and the surface characteristics of both the mold and the PMMA slide undergoing embossing. The choice of these parameters depends on the type of thermoplastic material selected for embossing.

Among the aforementioned process parameters, the embossing temperature (T_e) is the most critical parameter as it should lie between the glass transition temperature (T_g) and the melting temperature (T_m) of the thermoplastic material. Because PMMA has T_g and T_m at 106°C and 160°C [28], respectively, we found $T_e = 155^\circ\text{C}$ to be suitable for the hot embossing process. Lower temperatures proved inadequate for achieving complete microstructure replication. On the other hand, temperatures above the melting temperature ($T_e \geq T_m$) resulted in defective replication and undesirable deformation of the PMMA substrate. For $T_e = 155^\circ\text{C}$, the embossing time of 20 min was found to be adequate to ensure proper replication of the microfeatures.

Another important parameter in hot embossing is the embossing force on the mold-substrate assembly. Maintaining the correct force is essential to ensure consistent and high-quality replication of micropatterns. Excessive force, which might feel necessary to expedite the embossing process, risks

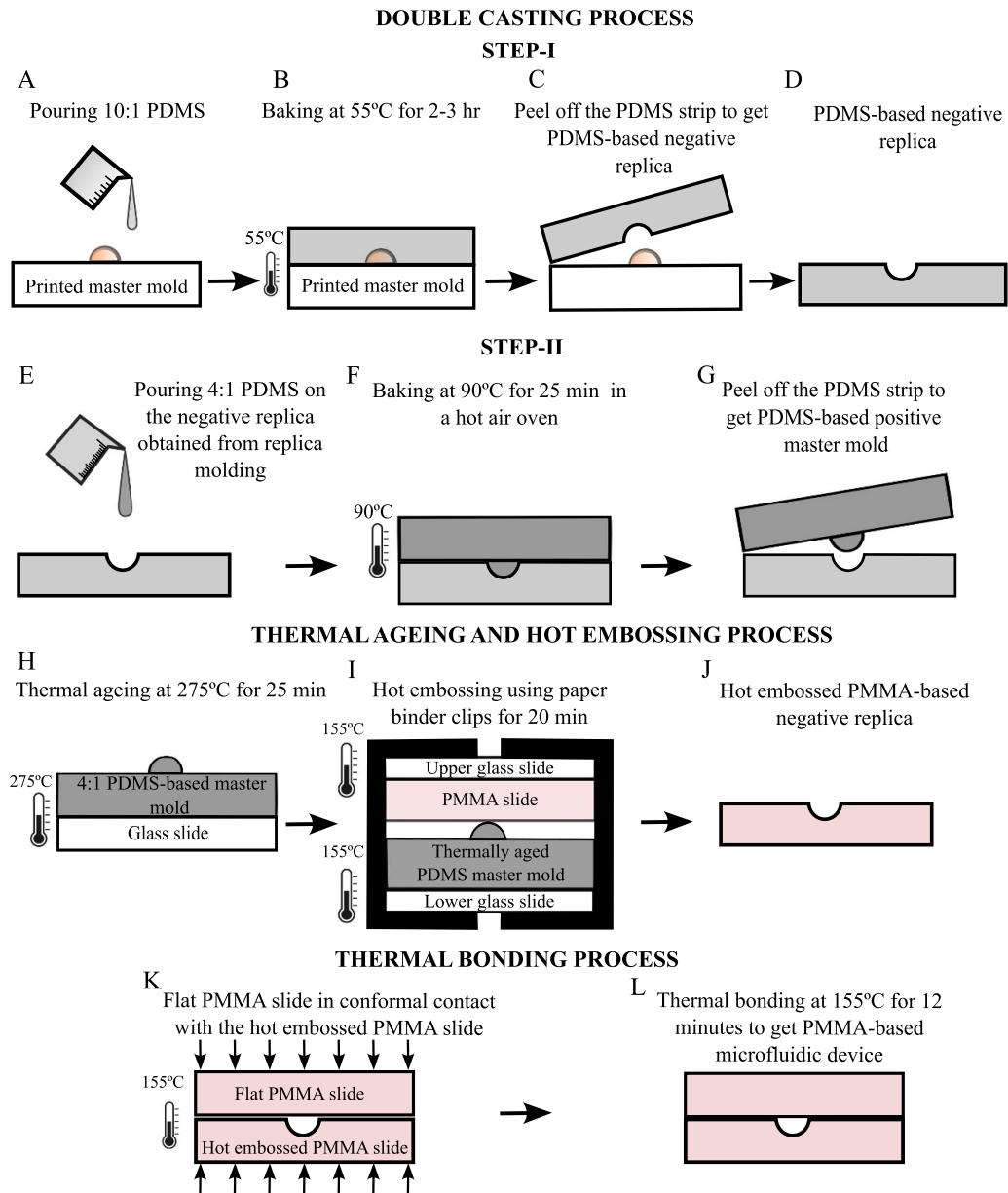


Figure 4. Schematic representation of the double casting and hot embossing processes to fabricate PMMA microfluidic device starting from the EHD-jet printed non-sacrificial master template. (A)–(D) show first step of the double casting process to obtain a PDMS-based (10:1) negative replica from the EHD-jet printed master template. (E)–(G) show the sequence of steps to get a (4:1) PDMS-based positive master mold. This PDMS-based positive master mold was used in the hot embossing process to fabricate a PMMA microfluidic device. (H)–(J) illustrate the thermal ageing process, the hot embossing process using PDMS mold and paper binder clips, and the PMMA replica obtained from the hot embossing process, respectively. (K)–(L) show the thermal bonding process of the PMMA replica with a flat PMMA slide at 155 °C to get the final microfluidic device.

the geometric deformation of the PDMS mold. At the same time, the embossing force should be sufficient for the mold to overcome the viscous resistance offered by the thermoplastic substrate during the embossing process, thereby ensuring good replication. After multiple iterations of experiments, we found that four 19 mm paper binder clips were sufficient to apply the requisite embossing force. Figure S2 † of the supplementary information shows the variation of the compressive force (F_c) of the binder clip with the increase in the gap between its two jaws (y). The compressive force F_c increases from

17 N to 47 N for the corresponding increase in y from 1 mm to 9 mm. In our experiments, the assembly had a total height of 7 mm. Figure S2 shows that a single paper binder clip exerts a compressive force of 40 ± 3 N corresponding to a 7 mm gap between its two jaws. Therefore, a combination of four binder clips exerted a net compressive force of 160 ± 6 N and correspondingly a compressive stress of 130 ± 7 kPa.

The flatness and surface roughness of the cured PDMS mixture also affect the quality of the embossed structures. A non-uniform thickness of the PDMS mold can lead to uneven

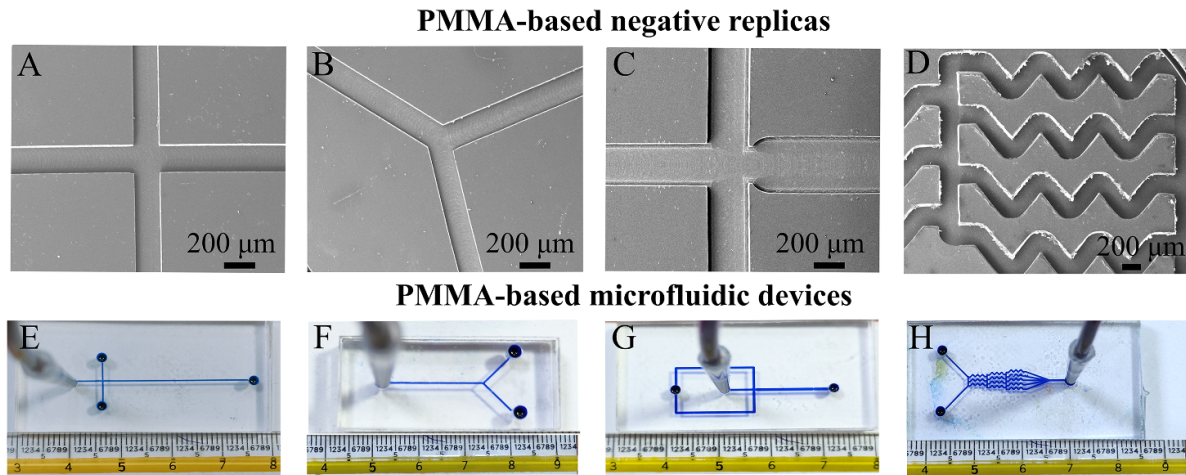


Figure 5. PMMA replicas obtained from the hot embossing process and thermally bonded PMMA microfluidic devices. (A)–(D) show SEM images of PMMA replicas with geometries of cross-shaped, Y-shaped, droplet generator and tree-shaped gradient generator microfluidic devices, respectively. (E)–(H) show the thermally bonded PMMA microfluidic devices filled with blue-colored dye solution.

force distribution, leading to partial replication of the micro-patterns. To ensure uniform thickness, the petri dish containing PDMS was carefully placed on a leveled surface while curing. Furthermore, a leveled and smooth PMMA substrate with no sharp edges at the corner was chosen for the hot embossing process, ensuring uniform force application during embossing.

2.4.2. Protocol for hot embossing process. Figures 4(I) and (J) illustrate the process of hot-embossing PMMA substrate using PDMS-based positive master molds. The cured 4:1 PDMS-based master mold (50 mm × 25 mm × 2 mm) was positioned on a glass slide (75 mm × 25 mm × 1.5 mm) with the microfeatures pointing upward. The PMMA substrate (50 mm × 25 mm × 2 mm) was precision-cut using a laser cutting machine (CO₂ laser, Epilog Helix 24, 80 W). Subsequently, the PMMA substrate was cleaned thoroughly with DI water and IPA and then dried with compressed N₂ gas. The PMMA substrate was brought into conformal contact with the PDMS master mold, with an additional glass slide to sandwich the PDMS mold and the PMMA substrate. The assembly was carefully clamped using four 19 mm binder clips, as depicted in figure 4(I). The entire assembly was then placed inside the preheated oven at 155 °C for a duration of 20 min. Subsequently, the assembly was removed from the oven and allowed to cool for another 10 min to room temperature while keeping the binder clips clamped to the slides. Finally, the PDMS mold was carefully peeled off from the embossed PMMA slide. The embossed PMMA slide was then cleaned with IPA and DI water, followed by drying with compressed N₂ gas.

2.5. Characterization of the master molds and negative replicas

Figures 5(A)–(D) show the images of hot-embossed PMMA negative replicas with geometries of cross-shaped, Y-shaped,

flow-focusing droplet generator and tree-shaped gradient generator microfluidic devices, respectively. To characterize the PDMS negative replica, PDMS positive master mold and PMMA negative replica, a cut section of the respective components was placed vertically under the 10× objective lens (CFI TU Plan Flour, NA = 0.30) of an upright measuring microscope (Nikon MM400) coupled with a CMOS camera. Subsequently, the width (w) and height/depth (h) of the cut-sections of the respective components were measured and recorded. Figures 6(A)–(C) and 6(D)–(F) show the snapshots of the cross-sections of 10:1 PDMS-based negative replica, 4:1 PDMS-based positive mold and the hot-embossed PMMA slide of cross-shaped geometry and droplet generator geometry, respectively. As in the current work we have employed single-pass EJP, the aspect ratio of the printed feature and the PMMA negative replica is governed by the contact angle of the polymeric ink on the substrate. In the current experiments, the contact angle was in the range of 51 °–55 °, and correspondingly, the aspect ratio ranged between 0.24–0.25. However, several methods have been reported in the literature to increase the aspect ratio of the EHD-jet printed features. These include surface treatment [20], multi-pass printing [30], and enhancing the evaporation rate of the solvent [31]. In the supplementary information, we demonstrate that the aspect ratio of the printed features can be increased up to 0.34 by treating the substrate with chlorotrimethylsilane prior to EJP, which increases the contact angle to 70 ° (refer to figure S3 † of the supplementary information). In addition, we infer from figures 6(C) and (F) that the microchannels obtained from the EHD-jet printed templates are of semi-circular cross-section, similar to other inkjet printing processes. The semi-circular cross-sectional shape of the microchannel is a characteristic of this fabrication process, just as wet etching, laser ablation and photolithography are characterized by D-shaped, Gaussian-shaped and rectangular cross-sections, respectively [4]. Furthermore, it is important to note that the cross-sectional shape of the microchannel does not limit its functionality, as fluid flow in narrow microfluidic

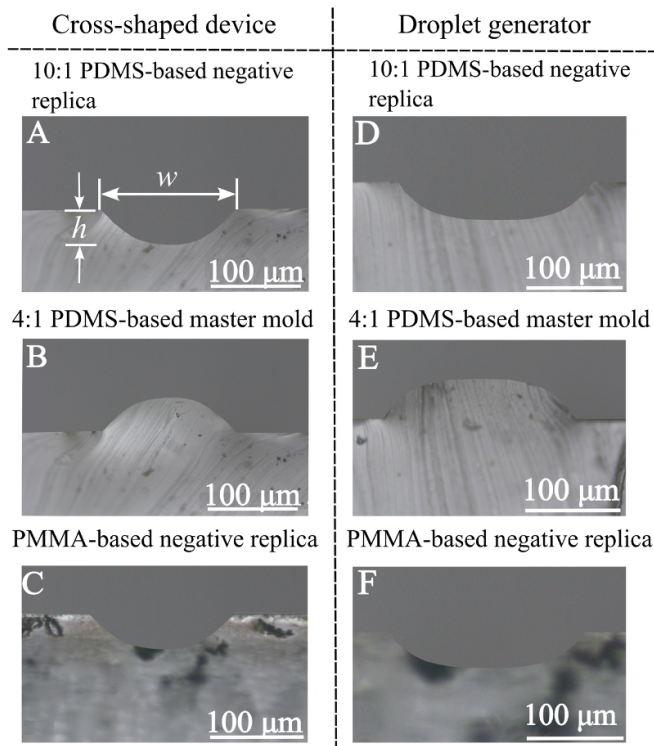


Figure 6. Cross-sections of 10:1 PDMS-based negative replica, 4:1 PDMS-based master mold and PMMA-based negative replica for (A)–(C) cross-shaped microfluidic device and (D)–(F) droplet generator, respectively.

channels is primarily governed by axial variations in channel geometry. For the same reason, microfluidic devices with D-shaped and Gaussian-shaped cross-sections fabricated using wet etching and laser ablation methods are interchangeably used with devices fabricated using the conventional photolithography process. Next, to get an estimate of the roughness of hot-embossed PMMA microchannels, we placed a cut section of a cross-shaped microchannel under a 3D optical profilometer (Zygo, NewView™ 9000) (refer to figure S4 † of the supplementary information). We observed that the average roughness of the microchannel was 80 nm, which is negligibly small compared to the dimensions of the microchannel.

To assess the uniformity of the printed master templates, PDMS-based master molds and PMMA replicas, we measured the line width at ten different locations along the length of the respective components following the methodology discussed above. The uncertainty in the measured width, estimated using t -distribution [27], was found to be less than 2% at 95% confidence level for each of the printed master templates, PDMS master molds, and PMMA replicas. Specifically, at the cross-over points of the printed templates, such as in cross-shaped geometries, we observed that the uncertainty in dimensions remained within the acceptable limit of 5% at a 95% confidence level. This consistency is attributed to the relatively short time interval between printing two perpendicular lines in our experiments. Consequently, the intersecting liquid filament deposited on the previously printed filament

locally merged, and the potential dimensional non-uniformity was minimized due to surface tension. Subsequently, we compared the widths of the printed master template, PDMS master mold, and PMMA replica after fifteen iterations of replica molding and hot embossing for each geometry. For the cross-shaped PCL-based template, the measured mean width was $150.2 \pm 1.6 \mu\text{m}$, and the corresponding mean widths of the PDMS master mold and PMMA replica were $151.2 \pm 1.9 \mu\text{m}$ and $151.6 \pm 2.1 \mu\text{m}$, respectively. Similarly, the mean height of the PDMS master mold was $37.5 \pm 0.7 \mu\text{m}$ signifying a 99% replication [32] along the depth-wise direction. Furthermore, the cavity volume of the PMMA replica was nearly identical to the feature volume of the PDMS mold at the embossing temperature [33]. In the current study, the differences between the mean widths of the printed feature, the PDMS mold, and the hot-embossed microfluidic channel in PMMA were statistically insignificant, confirming the non-sacrificial nature of the EHD-jet printed template. Also, the near-unity values for both the ratio of the depth of PMMA replica to the height of the PDMS mold and the ratio of cavity volume of the PMMA replica to the feature volume of the PDMS mold indicate high replication fidelity of the microfabrication process [32, 33].

Having established the reliability and replication fidelity of the hot embossing process, we bonded the hot-embossed open microfluidic channels in PMMA with flat PMMA slides to obtain the respective final microfluidic devices. To this end, a flat PMMA slide was thoroughly cleaned with DI water and IPA, followed by drying with compressed N_2 gas. Subsequently, inlet and outlet holes, each with a diameter of 2 mm, were laser cut on the embossed PMMA slide using a laser machine. The PMMA slides were then carefully brought into conformal contact with each other and placed between two steel plates covered with PDMS pads. The assembly was then subjected to a force of 0.7 kN at a temperature of 155°C for 12 min to successfully perform thermal bonding. This process results in a sealed and leakproof PMMA microfluidic device. Alternatively, two hot-embossed PMMA slides can be thermally bonded to achieve microfluidic devices with elliptic cross-section. Finally, to check for leakages and clogging of the fabricated PMMA microfluidic devices, a blue-colored food dye solution in DI water (50 mg ml^{-1}) was flown through the inlets of the microfluidic devices. As illustrated by figures 5(E)–(H) the microchannels are visibly filled with the blue-colored dye solutions without any leakages, confirming the integrity of the fabricated microfluidic devices.

3. Results and discussion

In this section, we demonstrate the application of our hot-embossed PMMA microfluidic devices through a set of typical microfluidic experiments. Specifically, we demonstrated the use of the cross-shaped device for hydrodynamic flow focusing, the Y-shaped device and the flow-focusing droplet generator for droplet generation, and the tree-shaped gradient generator for generating stable concentration gradients.

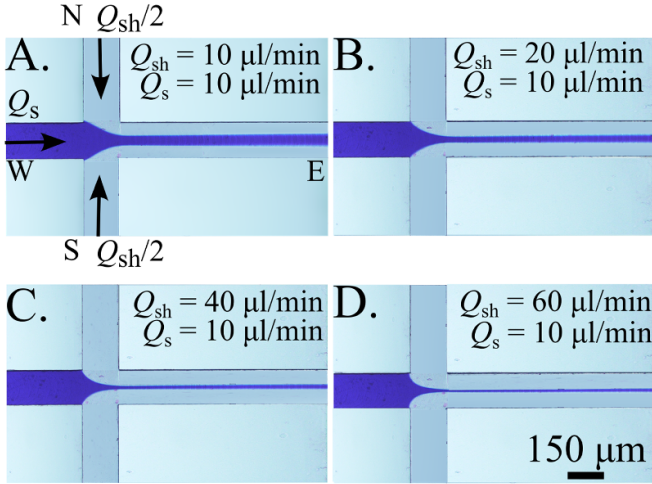


Figure 7. Visualization of steady hydrodynamic focusing in the cross-junction microfluidic device using a blue-colored dye solution as the sample solution and DI water as the sheath solution. (A)–(D) show hydrodynamic focusing corresponding to $Q_s/Q_{sh} = 1$, $Q_s/Q_{sh} = 1/2$, $Q_s/Q_{sh} = 1/4$, and $Q_s/Q_{sh} = 1/6$, respectively. These experiments show that, for a fixed value of Q_s , the width of the focused stream decreases upon increasing Q_{sh} .

For all the experiments, we used syringe pumps (KD Scientific Legato 100) to flow liquids through the fabricated microfluidic devices. For visualization of hydrodynamic flow focusing, we used an inverted microscope (Nikon Eclipse, TS100, Japan) equipped with a collimated LED light source (Thorlabs, USA) and a high-definition CCD color camera (Nikon DS Fi-2, 21 fps). The droplet generation process was observed using the same microscope but with a high-speed CMOS camera (Basler acA720–520 μm, 500 fps). For visualizing the gradient generation experiments, a stereozoom microscope (Radical Scientific, 8–45 × zoom) equipped with a CMOS camera (Radical Scientific, Procam HS–5) was employed. The raw images corresponding to each experiment were processed to remove the background in MATLAB.

3.1. Hydrodynamic focusing experiment

First, we present hydrodynamic focusing experiments in the cross-shaped microfluidic device as shown in figures 7(A)–(D). Such focusing of sample stream is typically used for microfluidic flow cytometry. For these experiments, we flowed a blue-colored dye solution (sample fluid) from the west ('W') inlet with a flow rate of $Q_s = 10 \mu\text{l min}^{-1}$. Simultaneously, we flowed the sheath fluid (DI water) from the north ('N') and south ('S') inlets with flow rates of $Q_{sh}/2 = 5 \mu\text{l min}^{-1}$ each. As shown in figure 7(A), the dyed stream hydrodynamically focuses downstream of the junction as the flow velocity downstream of the junction is higher than the upstream velocity of the dyed solution. Thereafter, we increased the sheath flow rate sequentially while keeping the flow rate of the dyed stream fixed at $Q_s = 10 \mu\text{l min}^{-1}$. Figures 7(B)–(D) show that the width of the focused sample stream decreases with a decrease in

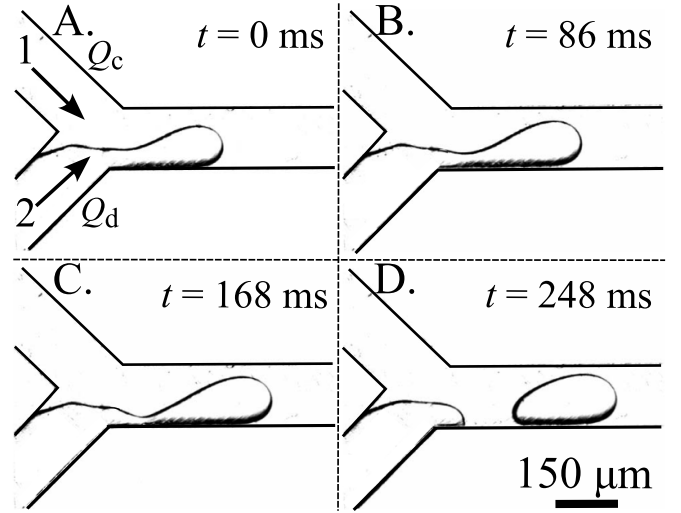


Figure 8. Various stages of droplet generation process in the Y-junction microfluidic device. Here, silicone oil is the continuous phase, and DI water is the dispersed phase. The flow parameters are $Q_d/Q_c = 1/8$, $\mu_d/\mu_c = 1/285$, $\text{Ca} = 0.17$.

Q_s/Q_{sh} , in accordance with the mass conservation of the dyed stream. The performance of our cross-shaped microfluidic device for hydrodynamic flow focusing is similar to hydrodynamic flow focusing devices in the literature [34].

3.2. Droplet generation experiment

Next, we demonstrate monodisperse droplet generation in the Y-shaped microfluidic device and the flow-focusing droplet generator. For these experiments, silicone oil (CDH, India) was the continuous phase, and DI water (CDH, India) was the dispersed phase. In the Y-shaped device, silicone oil was flown from inlet '1' at a flow rate of $Q_c = 4 \mu\text{l min}^{-1}$, and DI water was flown from inlet '2' at a flow rate of $Q_d = 0.5 \mu\text{l min}^{-1}$, as shown in figure 8(A). The stream of dispersed phase breaks up into monodispersed droplets at the junction. Figures 8(A)–(D) show the various stages of the droplet formation at the Y-junction. As the shear force exceeds the interfacial tension force between the two phases, necking is observed near the junction at $t = 168 \text{ ms}$, subsequently leading to the formation of a droplet of DI water at $t = 248 \text{ ms}$. The experiments shown in figures 8(A)–(D) correspond to capillary number of $\text{Ca} = \mu_c U_c / \gamma = 0.17$, flow rate ratio of $Q_d/Q_c = 1/8$, and viscosity ratio of $\mu_d/\mu_c = 1/285$. In the expression of Ca , U_c denotes the average velocity of the continuous phase, and $\gamma = 28 \text{ mN m}^{-1}$ denotes the interfacial tension between the continuous and the dispersed phases. The droplet generation corresponding to these dimensionless numbers occurs in the dripping regime [35], as observed in our experiments.

Next, we demonstrate the droplet generation using the flow-focusing droplet generator. We flowed silicone oil, which is the continuous phase, from the north ('N') and south ('S') directions with an equal flow rate of $Q_c/2 = 10 \mu\text{l min}^{-1}$, as shown in figure 9(A). Simultaneously, we flowed DI water,

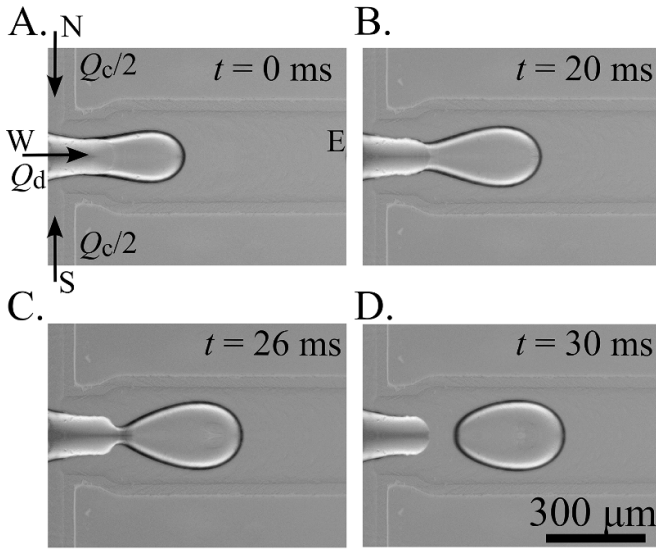


Figure 9. Various stages of droplet generation process in the flow-focusing droplet generator. Here, silicone oil is the continuous phase and DI water is the dispersed phase. In these experiments $Q_d/Q_c = 1/4$, $\mu_d/\mu_c = 1/285$ and $Ca = 0.11$.

which is the dispersed phase, from the west ('W') inlet with a flow rate of $Q_d = 5 \mu\text{l min}^{-1}$. Figures 9(A)–(D) show various stages of droplet generation in the flow-focusing droplet generator microfluidic device. In the present flow configuration, the competition between the viscous stresses and the capillary pressure determines the droplet breakup process and is quantified by the capillary number (Ca). We used the expression provided by Anna and Mayer [36] for evaluating the value of Ca for the flow-focusing droplet generator. For the current set of experiments $Ca = 0.11$, $Q_d/Q_c = 1/4$ and $\mu_d/\mu_c = 1/285$, which correspond to droplet generation in the dripping regime [36]. Overall, the sequence of droplet formation in both Y-shaped microchannel and droplet generator are similar to droplet generation experiments in the literature [36, 37].

3.3. Tree-shaped gradient generator

Finally, we present the use of the hot-embossed tree-shaped gradient generator to generate stable concentration gradients. Such gradient generators are typically used to analyze the cell chemotaxis in a controlled microenvironment [38]. We flowed yellow colored food dye solution in DI water (50 mg ml^{-1}) and blue colored food dye solution (50 mg ml^{-1}) in DI water from the inlets marked as '1' and '2', as shown in figures 10(A) and (B). We allowed the system to reach a steady state and then captured the data in the form of images. In the first experiment, as shown in figure 10(A), the flow rates of both the solutions were kept equal at $0.8 \mu\text{l min}^{-1}$. As the dye streams flow through the microchannel network, they are divided sequentially at various junctions and undergo mixing with the neighboring streams through diffusion within the serpentine channels. Towards

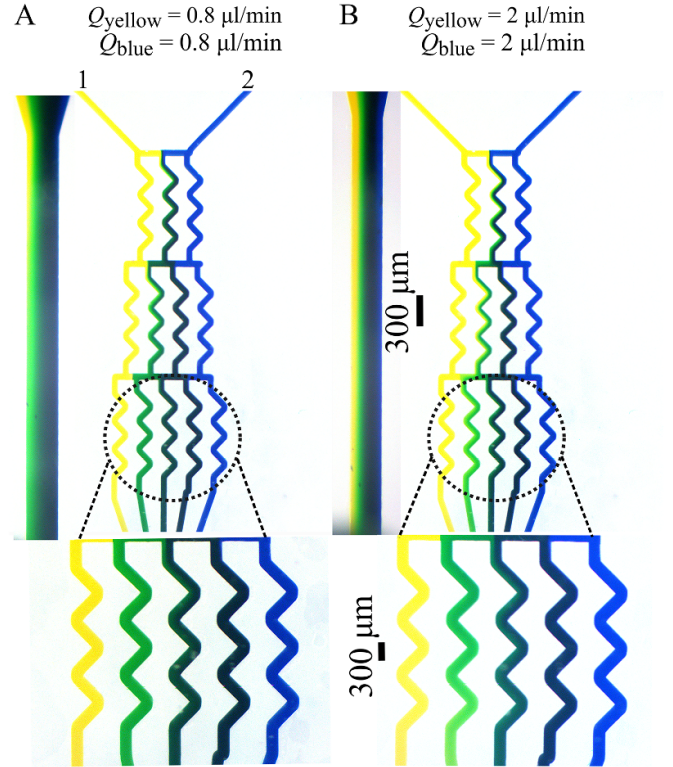


Figure 10. Photograph showing the tree-shaped gradient generator used for generating gradients of yellow and blue dyes in solution. (A)–(B) show steady state snapshots of tree-shaped gradient generator for $Q_{\text{yellow}} = Q_{\text{blue}} = 0.8 \mu\text{l min}^{-1}$ and $Q_{\text{yellow}} = Q_{\text{blue}} = 2 \mu\text{l min}^{-1}$, respectively. It is evident from the snapshots that for a flow rate value of $0.8 \mu\text{l min}^{-1}$, the mixing is better, resulting in different shades of green color at the outlet segment of the device. Whereas for a flow rate value of $2 \mu\text{l min}^{-1}$, the parallel streams of yellow and blue colors are clearly visible at the outlet segment of the tree-shaped gradient generator, signifying comparatively poor mixing.

the end of the network, all streams carrying varying concentrations of blue and yellow dye solutions combine in the broader channel (the straight segment of the gradient generator). Figure 10(B) shows the gradient generator for a higher flow rate $Q = 2 \mu\text{l min}^{-1}$ of the dye solution.

The mixing of the two streams in the gradient generator is governed by the competition of diffusion and convection. The relative importance of convection and diffusion is quantified by the Peclet number (Pe). In the current experiments, the dye transport in the tree-shaped gradient generator is dominated by convection, with $Pe \gg 1$ [39]. For $Q = 0.8 \mu\text{l min}^{-1}$, $Pe \approx 3 \times 10^3$ and for $Q = 2 \mu\text{l min}^{-1}$ $Pe \approx 8 \times 10^3$. Figures 10(A) and (B) show that the diffusive mixing in the straight segment of the gradient generator is relatively higher for $Q = 0.8 \mu\text{l min}^{-1}$ than for $Q = 2 \mu\text{l min}^{-1}$. This is attributed to the lower value of Pe for the first case as compared to the second case. The performance of the fabricated tree-shaped gradient generator in generating steady concentration gradients of a mixture of dye solutions is similar to that observed in the literature [38, 39]. Overall, the experimental demonstration

of the hot-embossed microfluidic devices in section 3 shows that our microfabrication method yields reliable microfluidic devices with performances comparable to those described in the literature.

4. Conclusion

We have demonstrated a novel approach for fabricating high-resolution master templates for hot embossing of thermoplastic microfluidic devices using EJP. Specifically, we have demonstrated rapid prototyping of intricate polymeric templates for various microfluidic devices using EJP with feature sizes of order 100 μm . We used these templates to develop stiff PDMS molds, using a double casting process, which were subsequently used for the hot embossing of PMMA devices. We demonstrated the versatility of our approach by repeatably fabricating PMMA microfluidic devices for applications such as hydrodynamic flow focusing, droplet generation and concentration gradient generation. In contrast to conventional photolithography-based methods for mold fabrication, our method, based on EJP and double casting, allows rapid customization of low-cost master molds with comparable resolution. Moreover, our approach for mold fabrication offers significantly better resolution than other prototyping methods such as micromilling, laser ablation and 3D printing. Overall, our microfabrication method allows rapid prototyping of customized high-resolution master molds for hot embossing of microfluidic devices at a significantly lower cost and faster speed than the comparable microfabrication techniques.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Conflicts of interest

There are no conflicts to declare.

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