# **Supplementary Information**

# Drop Impact Printing

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# **Supplementary Figures**



**Supplementary Figure 1:** Time-lapse images of water droplet when impacting on the superhydrophobic sieve (#0.009) from different heights. (**a**) The drop impact from a height of 2.5 cm (Weber Number (We) - 17), resulting in neither impact penetration nor recoil penetration. **(b)** The drop impact from height of 3.7 cm, resulting in ejection of multiple droplets from single jet. Drop ejection was observed both in impact and recoil penetration, Scale bar  $-1$  mm.



**Supplementary Figure 2:** The comparison of single drop regime for superhydrophobic and hydrophobic sieve (#0.009 and #0.012). The plot shows the range of impact velocities that ensure single drop ejection when water is used as the printing liquid.



**Supplementary Figure 3:** Single drop ejection mechanism for sieve type #0.012 is shown using (**a**) time-lapse images (Scale bar – 1 mm) and (**b**) schematic illustration. When drop impacts on sieve, impact jet is formed. As the drop starts to recoil, impact jet is not able to retract back completely and recoil jet pushes the jet further to eject a single drop. In this case, the collapse of top interface of the drop is responsible for single drop ejection.



 $1000 \,\mu s$  after impact



 $2000 \,\mu s$  after impact



 $1000 \,\mu s$  after impact



 $2000 \,\mu s$  after impact



As the simulations do not capture the recoil cavity formation (due to structural difference with the experimental condition), the dynamics start deviating in later stages.





Despite differences in the simulation and experiments, microdroplets are generated approximately at the same time.



**Supplementary Figure 4:** Simulation and experimental results at different time scales showing the single drop ejection.



**Supplementary Figure 5:** Different collapse dynamics resulting from impact of a drop on sieve to generate single drop is shown. The single drop ejection was possible only in recoil ejection. Under recoil ejection, further distinction was made based on cavity collapsed.



**Supplementary Figure 6:** Time-lapse images of water droplet impacting on sieve (#0.012). This ejection mode comes under impact penetration mode. In this mode, impact jet contributes to single droplet volume since it is not able to retract back to parent drop completely, Scale bar  $-1$ mm.



**Supplementary Figure 7:** Schematic of electroplating process.



**Supplementary Figure 8:** Electroplated mesh characterization. (a) SEM images of the electroplated mesh (pore opening - 32.1  $\mu$ m, wire size - 94.7  $\mu$ m), Scale bar - 20  $\mu$ m. (b) SEM images of etched superhydrophobic electroplated mesh (pore opening -  $25.2 \mu m$ , wire size -101.2  $\mu$ m), Scale bar - 20  $\mu$ m. (c,d) Static contact angle on electroplated mesh and superhydrophobic etched electroplated mesh (Scale bar  $-1$  mm) and (e) Time-lapse photographs showing the ejection of the smallest droplet  $(42 \mu m)$  using superhydrophobic electroplated mesh, Scale bar  $-1$  mm.



**Supplementary Figure 9:** Time-lapse photography of viscoelastic drop when impacted on superhydrophobic sieve (#0.0045). The liquid used was 10% (volume percent  $(v/v)$ ) Xanthum gum-Water solution, Scale bar – 1mm.



**Supplementary Figure 10:** Weber number corresponding to single drop ejection with varying viscosity for different sieves are shown. The liquid used is Glycerol-Water mixture of different concentrations.



**Supplementary Figure 11:** Sieve contamination characterization. (a) pinned nanoparticles on SH sieve (#0.012) after 1000 droplets impact. (b) cleaning process of sieve after contamination (water jet impact and  $N_2$  purging). (c) Mesh after cleaning. (d) Static contact angle on cleaned sieve, Scale bar – 1mm and (e) Water droplet repellence test by impacting drops on superhydrophobic cleaned mesh, Scale bar – 1mm.



**Supplementary Figure 12:** The printed feature droplet size distribution has been shown for sieve #0.0020 and #0.009.



**Supplementary Figure 13:** Position accuracy calculation: (a) 0° angle ejection and 10° angle of ejection for mesh #0.012 (Scale bar – 1mm) (b)  $0^{\circ}$  angle of ejection and  $6^{\circ}$  angle of ejection for mesh #0.0045 (Scale bar – 1mm) and plot between (c) longitudinal deviation (standard deviation) versus ejection angle for mesh #0.012 and #0.0045, (d) lateral deviation (standard deviation) versus ejection angle for mesh #0.012 and #0.0045.



**Supplementary Figure 14:** The sequence of images showing the ejecting silver ink droplet and merging with the neighborhood drop to form a line. The drop was observed to oscillate and then it merged with neighborhood droplet. The droplets were printed on a glass slide embedded with scotch tape. The spacing between the drops was kept between 150  $\mu$ m to 200  $\mu$ m for printing droplet volume of 3  $\mu$ L, Scale bar – 1mm.



**Supplementary Figure 15:** Voltage versus current curves for different silver ink concentrations. The optimization of the silver ink line was performed using mesh type #0.009.



**Supplementary Figure 16:** Microscopic images of a printed logo of IISc using drop impact printing technique showing patterned silver ink droplet (a) before annealing and (b) after annealing. The printing was carried out with mesh type #0.012 with droplet volume of approximately 0.35  $\mu$ L. Large area array printing capability of the technique was explored using mesh type #0.0045 showing patterned silver ink droplet from different view, (c) side angled view, and (d) top view. The printed droplet volume was approximately 3 nL. Scale bar  $-500 \mu m$ .



**Supplementary Figure 17:** (a) Schematic showing etching of clean copper sieve followed by silanization to obtain superhydrophobic sieve. SEM characterization of sieve showing for (b) Clean copper sieve, Scale bar  $-100 \mu m$ . (c) Etched superhydrophobic sieve (Scale  $bar - 100 \ \mu m$ ) and inset showing the nanowires (Scale bar – 2  $\mu$ m) that were created over the surface using etching. (d) Contact angle measurement showing the static contact angle on superhydrophobic sieve  $(\sim 159^{\circ})$ , Scale bar – 1mm.



**Supplementary Figure 18:** (a) Drop impact printing setup with recycling unit.

- 1. Ink reservoir for recycling of ink
- 2. Peristaltic pump (pumping ink to the syringe)
- 3. Mother droplet generation using a syringe
- 4. Automated z stage for height manipulation
- 5. Holding platform for mesh
- 6. Substrate holder fixed with XYZ automatic stage
- 7. High-speed camera
- 8. Diffused light
- 9. Sealed cabinet

# **Supplementary Tables**



**Supplementary Table 1: Sieve properties.**



**Supplementary Table 2: Dimensionless numbers of water droplet printing.**



**Supplementary Table 3: Fluid properties**

**A. Newtonian Fluid** – Aqueous Water-Glycerol Solution



**B. Newtonian Fluid** – Ethanol-Water Solution



**C. Non-Newtonian Fluid** – Aqueous PEG Solution



**Supplementary Table 4: Literature study related to** 

**A. Additive manufacturing for different printing techniques.**



**B. Bio-based printing applications for different printing techniques.**



**C. Food and Pharmaceutical applications for different printing techniques.**



**Supplementary Table 5: Literatures citing mass loading and other parameters of ink used by different printing techniques.**



**Supplementary Table 6: Different nanoparticles and size specifications.**



NA - Data is not available, # Resolution of acoustic based drop generation techniques (microfluidics nozzle approach) are higher ( $\sim$ 37  $\mu$ m) and similar resolution can be achieved for acoustophoretic printing by optimizing nozzle diameter and its other parameters.

**Supplementary Table 7: Comparison of different techniques with drop impact technique.**

#### **Supplementary Note 1: Simulation Results**

Drop impact simulation has been performed using the phase-field approach in COMSOL (Supplementary Figure 4). Flow field and phase-field were solved in cylindrical coordinate systems to reduce the computational time. As it is not possible to model the sieve in 2D cylindrical coordinate system, we simplified our simulation by modeling only for the central pore. The pore dimension and the wire size are matched with that of sieve #0.009. We modeled impact of water droplet with radius of  $1250 \mu m$  with an impact velocity of  $0.8 \text{ ms}^{-1}$ . Although the simulation was able to capture several aspects of the impact phenomenon, but we did not observe formation of recoil cavity. Though the liquid in the pores was observed to recoil back, the amount of fluid recoiling back was insufficient for formation of recoil cavity. This was because only one pore was simulated. The color scheme shows pressure with red representing higher values and green representing lower values. The arrows show flow direction. The size of the arrows represents the magnitude of local velocities. The images on the right are snapshots from a high-speed video for an impact on sieve #0.009.

#### **Supplementary Note 2: Preparation of Printing Solutions**

#### **Glycerol-Water Solution and Ethanol-Water solution**

Glycerol and ethanol were purchased from SD Fine Chemicals and distilled water is used for preparing solutions. Different concentrations  $(v/v \%)$  of glycerol-water and ethanolwater solutions were prepared for the experiment. The glycerol water concentration was varied from 10% to 80% and for the ethanol-water solution, it was from 12% to 36%. The solution viscosity, surface tension, and density are measured using Rheolab QC rheometer from Anton Paar, Density meter: DMA™ 4200 M from Anton Paar, and Tensiometer: K20 from Kruss Scientific respectively. The liquid properties of glycerol-water solutions and ethanol-water solutions are listed in Supplementary Table 3A & 3B.

#### **PEG-Water Solution**

Polyethylene glycol 4000 was purchased from Sigma Aldrich. PEG of different weight was mixed in 50 mL of distilled water and stirred, till it completely dispersed. The concentration was varied from 1% to 10% (v/v). The liquid properties like surface tension, density, and viscosities are measured. The liquid properties of PEG-water solutions are listed in Supplementary Table 3C.

#### **Nanoparticle Suspensions**

Different nanoparticles of varying size (10 nm to 20  $\mu$ m) were used in the experiment and are purchased from US Research Nanomaterials, Inc. Nanoparticles of different weights were mixed in 50 mL of 10% (v/v) PEG-water solution and stirred for 30 minutes. To have dispersed solutions, suspensions are further sonicated for 1 hour before the experiments.

For nanoparticle size variation demonstration, the concentration of nanoparticles was fixed around mass loading of 8.88% (w/w). And for different mass loading demonstrations, 200 nm Zirconium dioxide nanoparticles were used and the concentration (mass loading) was varied from 0.88% to 71%. The different nanoparticles size specifications are listed in Supplementary Table 6.

# **Electronic Inks**

For electronic ink printing applications three main inks are used in the present study: PEDOT: PSS, silver ink, and graphite ink. PEDOT: PSS  $(1.3 \text{ wt\%}$  dispersion in  $H_2O$ ), silver ink (30-35 wt%), and graphite ink (20-30%) were purchased from Sigma Aldrich and. PEDOT: PSS and graphite ink was used as it. The aqueous silver ink solutions of varying concentrations (1% to 4%,  $v/v$ ) were prepared by mixing in a 10% ( $v/v$ ) PEG-water solution. The ink suspensions were sonicated for 1 hour before the experiments.

## **Polymeric Solutions**

Polyacrylic acid (PAA) was purchased from Sigma Aldrich. Polyacrylic acid of 0.5 gm was mixed in 40 mL of distilled water and then stirred for 1 hour. The prepared viscous mixture was used for printing applications. The viscosity of the polymeric solution was measured to be 1.15 mPas.

# **Printing solutions of Red blood cell (RBC) and MDA-MB-231 cell**

Healthy whole blood (procured from RV Diagnostics Centre, Bangalore, India) of 50 μL was suspended in Phosphate-Buffered Saline (PBS) in the ratio,1:20 (v/v) and then the solution was centrifuged at 2000 rpm for 5 minutes. The supernatant part was removed and then the remaining pellet containing RBCs were resuspended in fresh PBS solution (making the total volume of the solution to 1 mL). Further, different volumes of prepared solutions was mixed to PBS (1mL) to prepare solutions of varying cell concentration. Cell counting was performed using hemocytometer.

MDA-MB-231 cells were maintained in Dulbecco's modified Eagle medium (DMEM, HiMedia) supplemented with 10% fetal bovine serum (FBS, Gibco) and 1% antibioticantimycotic solution (Gibco) and incubated in a humid 5%  $CO<sub>2</sub>$  incubator at 37 $^{\circ}$ C. The medium was replenished every 24 h. The cells were used at passage 3-4. To split the cells, they were washed with PBS twice and detached by 0.2% trypsin–EDTA, and the cells were counted using a hemocytometer.

## **Supplementary Note 3: Sieve Contamination**

We observed some degree of contamination during the drop impact printing. To examine that, we performed drop impact experiments using  $ZrO<sub>2</sub>$  nanoparticles (44.4%, weight percent (w/w)) dispersed in a 1:10 mixture of Ethylene Glycol and water. Drop impact using this solution was carried out for approximately 1000 times at one place using the mesh #0.012 coated with Teflon instead of stearic acid (Please refer Supplementary Table

1 for the geometrical parameters of the mesh). After the experiments, we observed some degree of contamination on the spot where drop impacted the mesh as seen in Supplementary Figure 11a. The contamination was easily removed with a mild jet of deionized (DI) water. After washing the samples were purged in  $N_2$  (Supplementary Figure 11b,c). To verify that the superhydrophobicity of the mesh was retained after the wash, we quantified its wettability through measurement of contact angle and contact angle hysteresis (Supplementary Figure 11d,e). Measured contact angle was  $157^{\circ}+3^{\circ}$  and hysteresis was  $\leq 5^{\circ}$  (contact angle hysteresis was calculated from the advancing and receding angle of the droplet at the onset when the droplet starts to slide) satisfying the conditions of superhydrophobicity.

#### **Supplementary Note 4: Description of Supplementary movies**

**Supplementary movie 1.** Single droplet printing during recoil ejection using sieve #0.0045.

**Supplementary movie 2.** Single droplet printing through impact cavity collapsed penetration mode.

**Supplementary movie 3.** Single droplet printing through recoil cavity collapsed penetration mode.

**Supplementary movie 4.** Single droplet printing through impact cavity impact penetration mode.

**Supplementary movie 5.** Ejected droplets with different diameters for different pore openings.

**Supplementary movie 6:** Smallest droplet ejection using electroplated superhydrophobic sieve.

**Supplementary movie 7.** Impacting droplet and moving substrate underneath mesh for single droplet printing.

**Supplementary movie 8.** 3D micropillar printing using sieve #0.012 (pore opening – 533.2 µm).

**Supplementary movie 9.** Multiple droplets impacting and ejecting successive single droplets in a row.

**Supplementary movie 10.** Lab-scale prototype of drop impact printing technique.

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