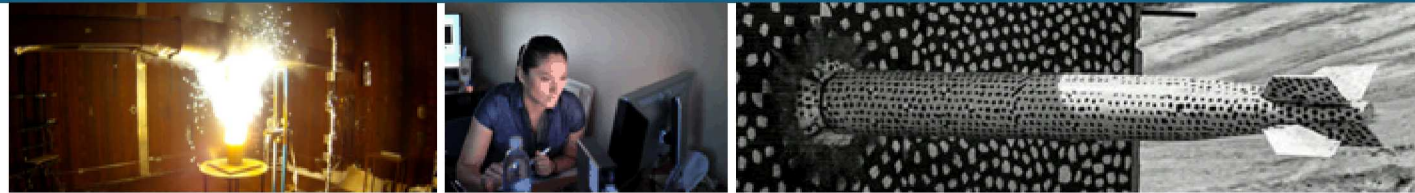


Aerosol jet printing: From fundamentals to graded multimaterial patterning



Ethan B. Secor

Advanced Materials Laboratory, Sandia National Laboratories
Albuquerque, New Mexico, USA

LOPEC Scientific Conference

26 March, 2020

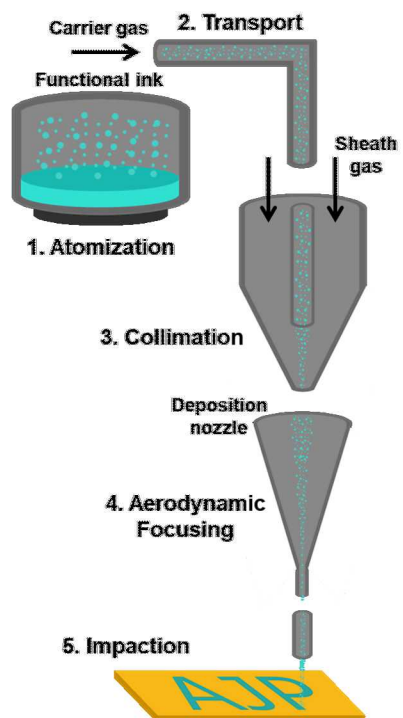


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Aerosol jet printing background/motivation

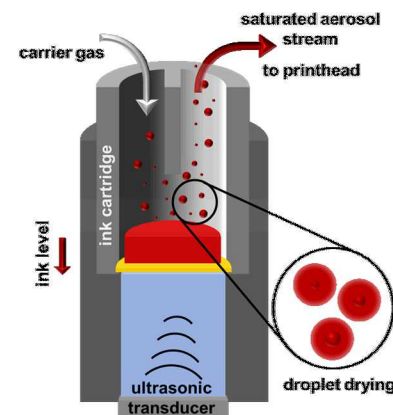
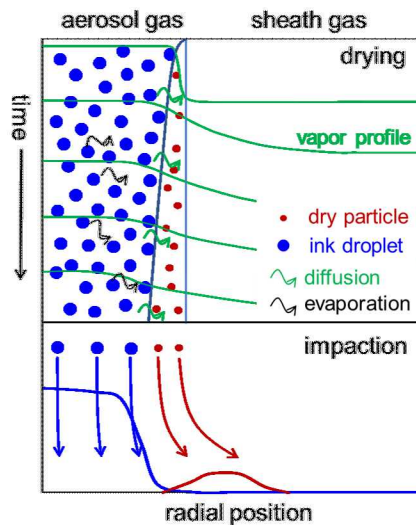
Aerosol physics

- General principles
- Baseline understanding
- Limitations



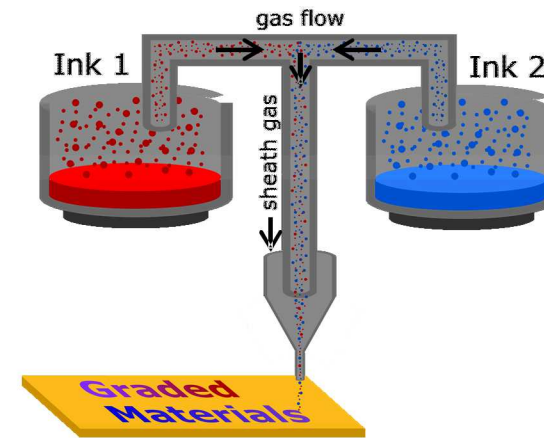
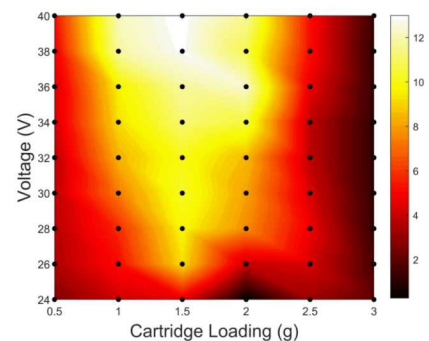
Mass transfer effects

- Conceptual framework
- Ink formulation
- Printhead design



Process drift

- Understanding drift
- Passive strategies
- Closed-loop control

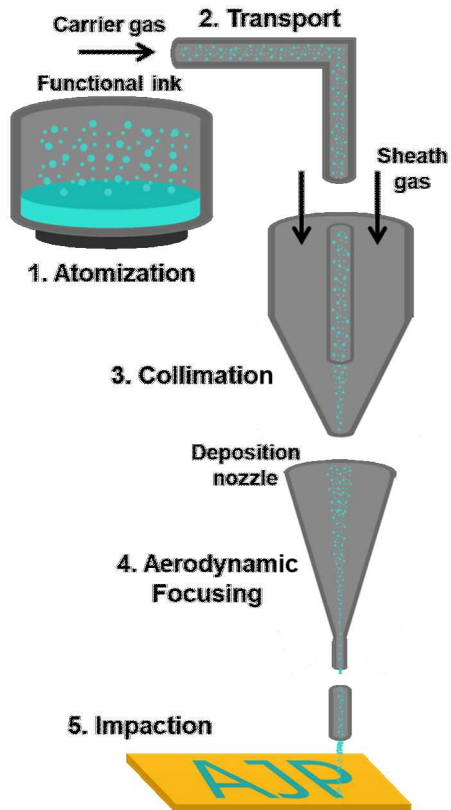


Multimaterial printing

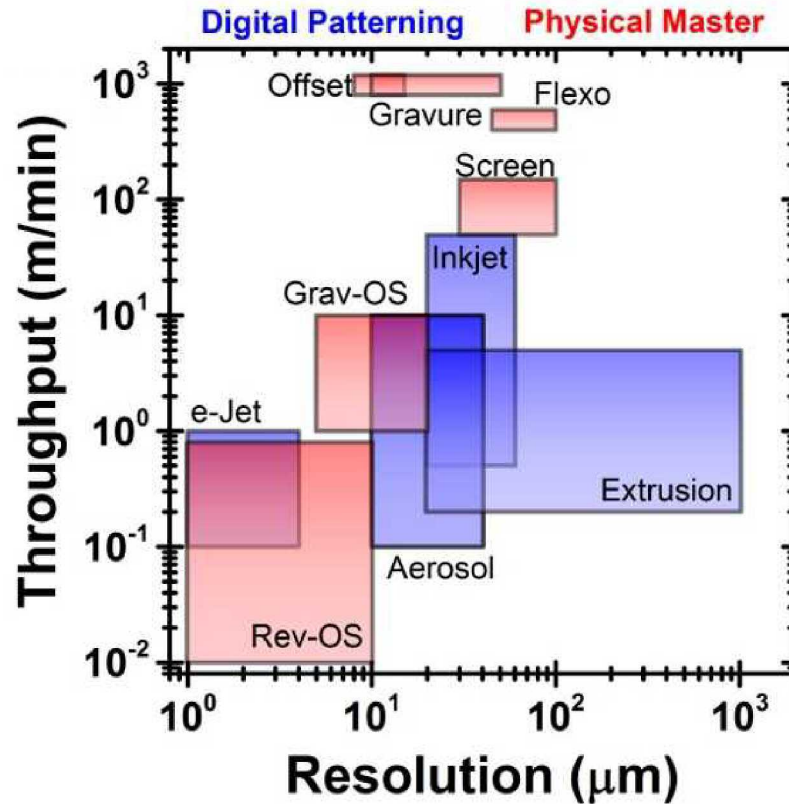
- Proof-of-concept
- Graded functionality
- Application potential

3 Background and motivation for aerosol jet printing

Aerosol jet printing (AJP) uses focused deposition of micron-scale ink droplets suspended in a carrier gas flow



AJP is a versatile technology for prototyping functional devices

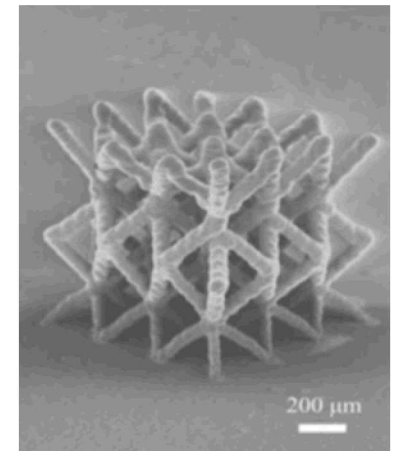
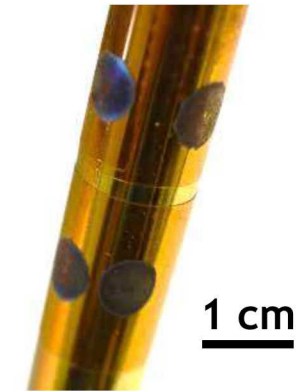
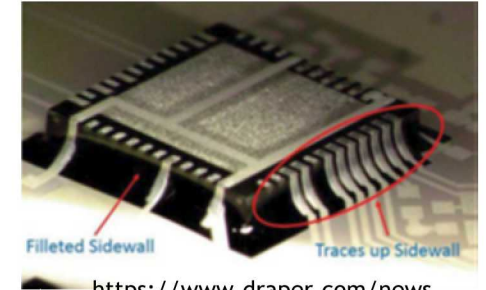


Benefits:

- Digital
- Non-contact
- High-resolution
- Versatile

Limitations:

- Process drift
- Low throughput

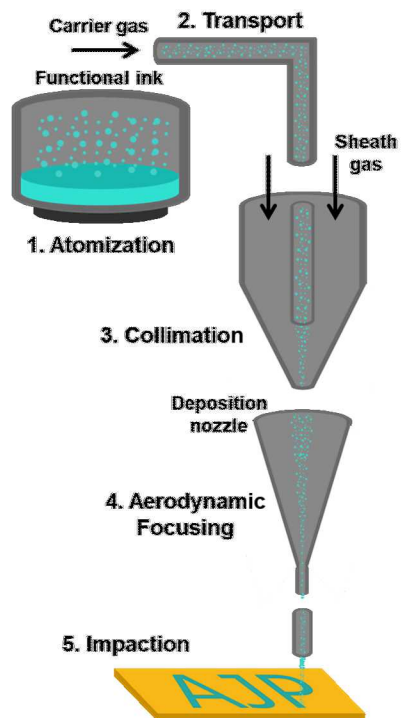


Saleh, H., et al. *Sci. Adv.* (2017) 3, e1601986.

Aerosol jet printing background/motivation

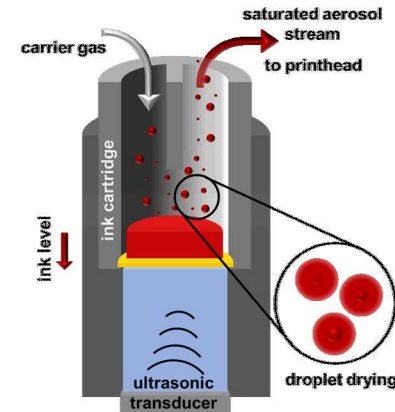
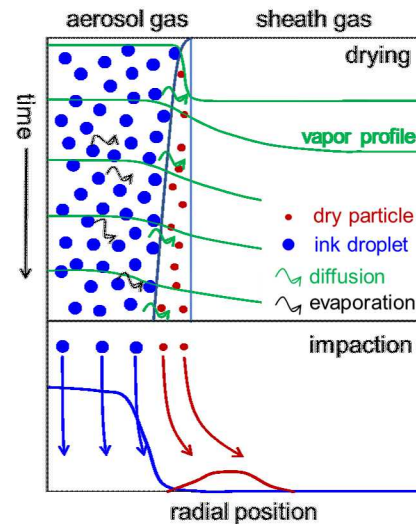
Aerosol physics

- General principles
- Baseline understanding
- Limitations



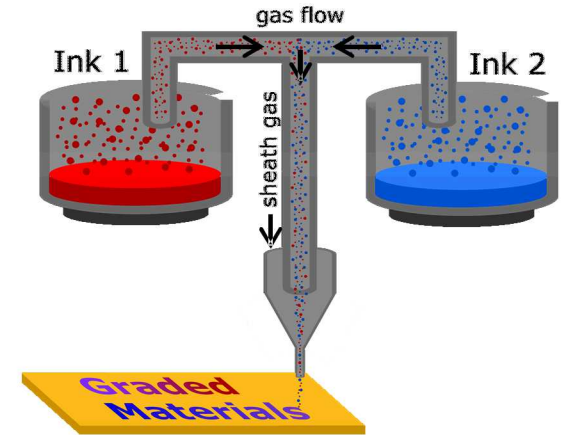
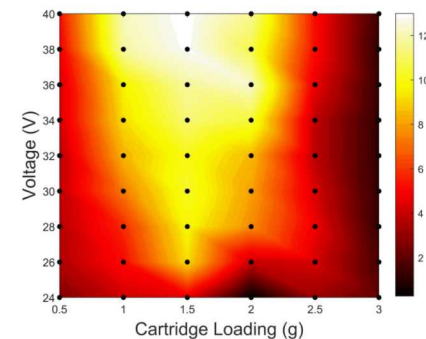
Mass transfer effects

- Conceptual framework
- Ink formulation
- Printhead design



Process drift

- Understanding drift
- Passive strategies
- Closed-loop control



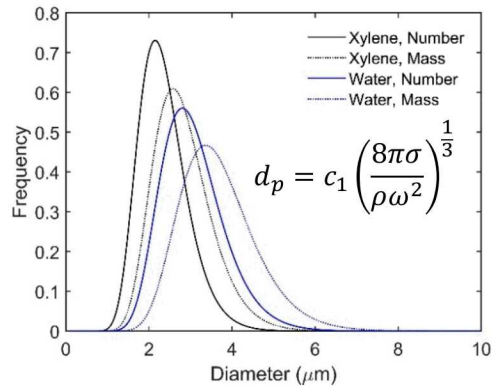
Multimaterial printing

- Proof-of-concept
- Graded functionality
- Application potential

Insight from aerosol physics

Analytical models of the five physical processes can provide insight when coupled

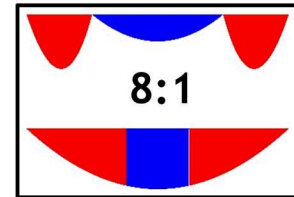
1. Atomization



3. Collimation

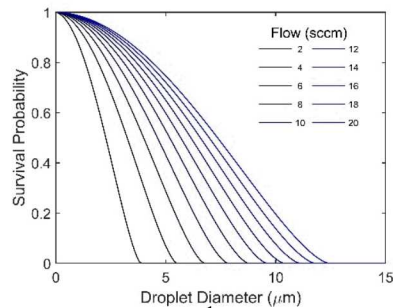
$$f_a = \int_0^{2\pi} d\theta \int_0^{R_a} r dr \left[2u_a \left(1 - \frac{r^2}{R^2} \right) \right] = \pi u_a R_a^2 \left[2 - \frac{R_a^2}{R^2} \right]$$

$$x_c = \sqrt{1 - \sqrt{\frac{FR}{1 + FR}}}$$



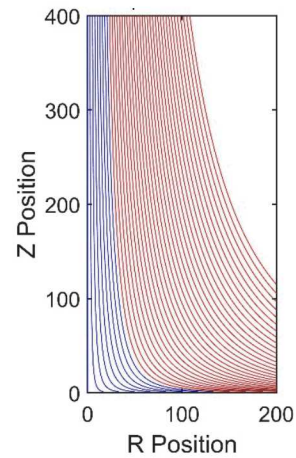
2. Transport

$$F = \frac{2}{\pi} (\alpha\beta + \sin^{-1} \beta - 2\alpha^3 \beta)$$



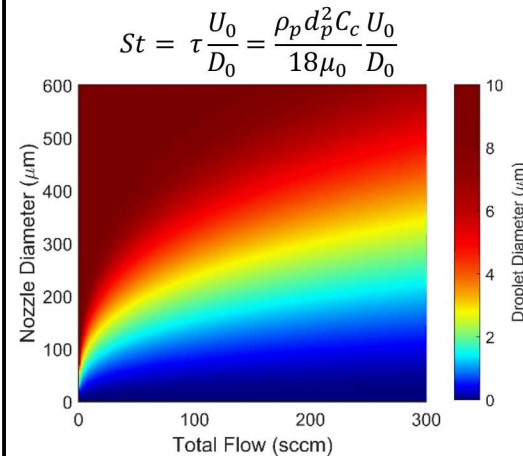
where $\alpha = \left(\frac{3Lu_{TS}}{8u_a R} \right)^{\frac{1}{3}}$; $\beta = \sqrt{1 - \alpha^2}$;

5. Impaction

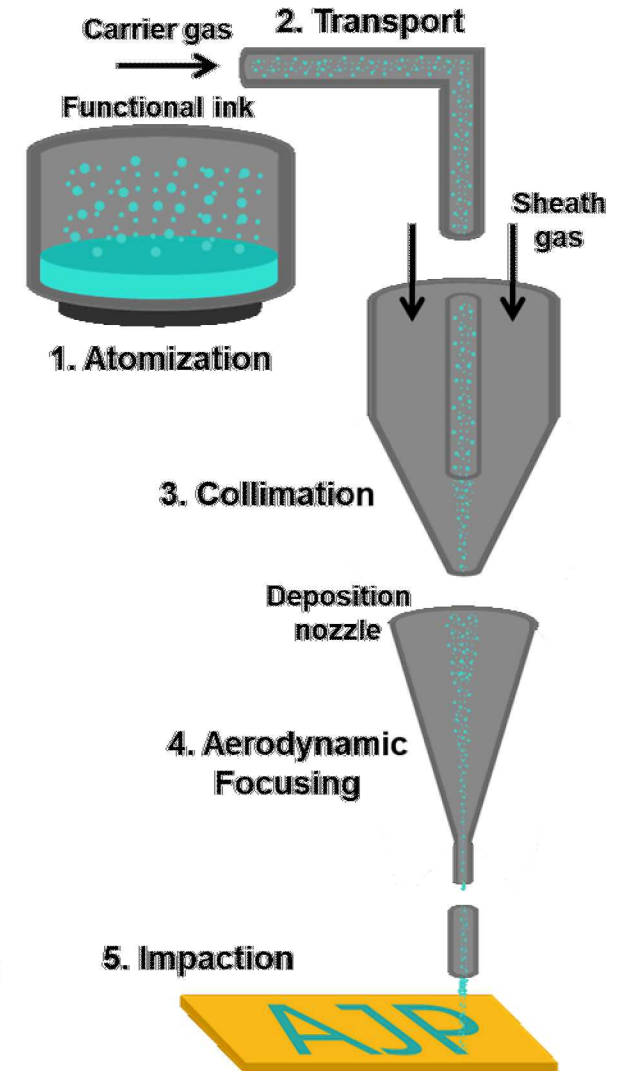


$$r_0^* \exp[-p] * \left\{ \cosh(pq) + \frac{1}{q} \sinh(pq) \right\}$$

4. Focusing

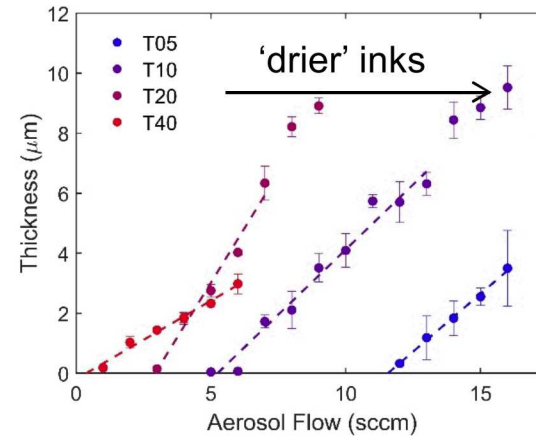
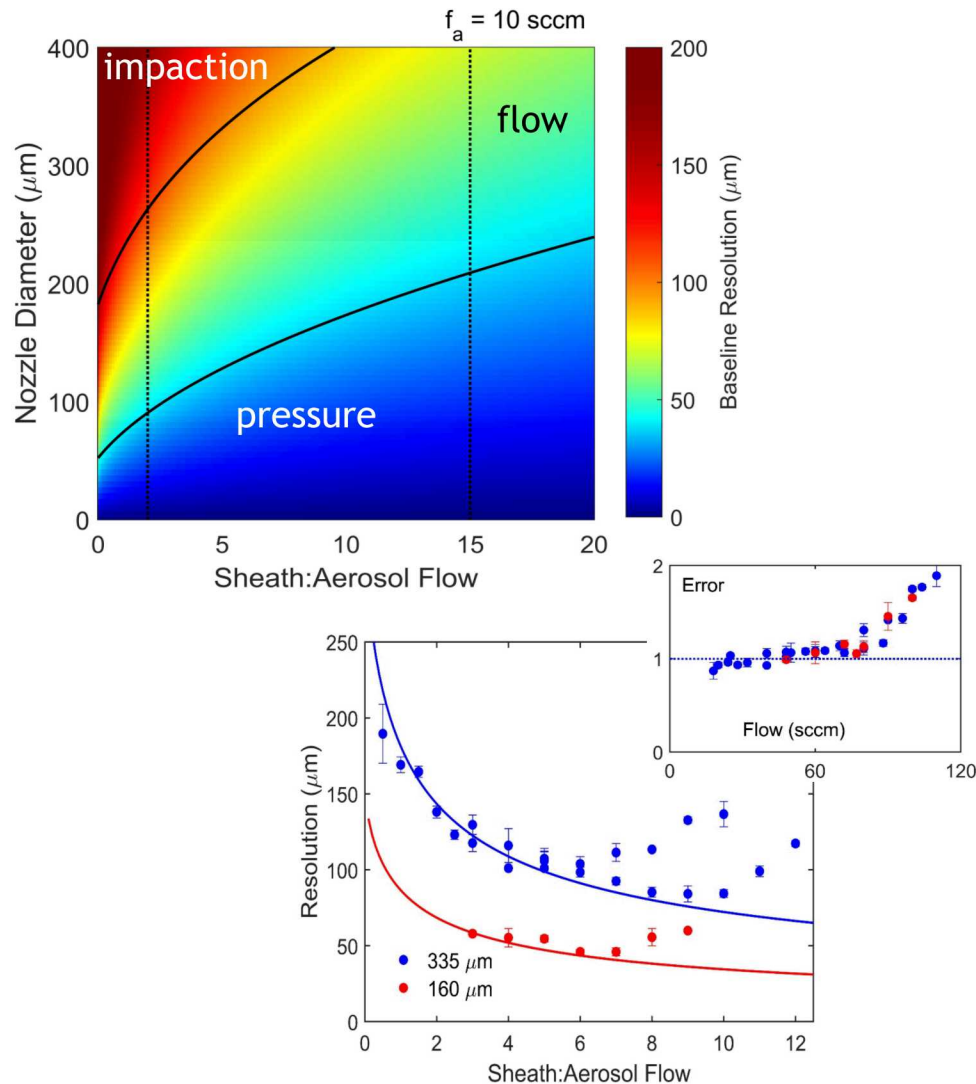


Fundamental description guides
printer and process design

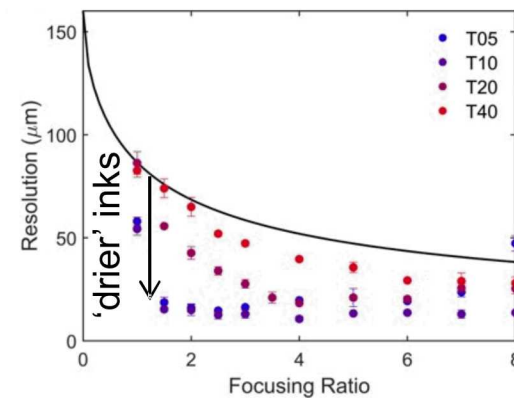


Limitations of aerosol physics perspective

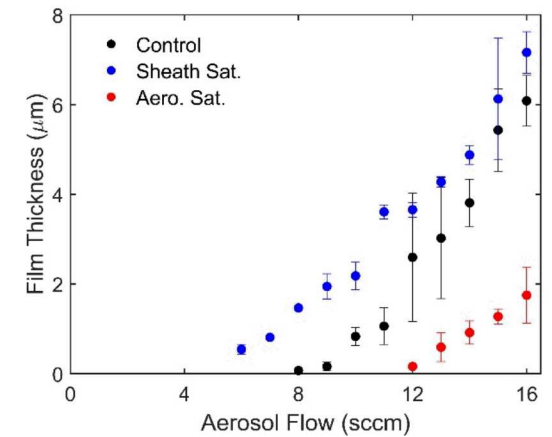
Models identify factors limiting print quality and resolution



More volatile inks deviate from expected behavior



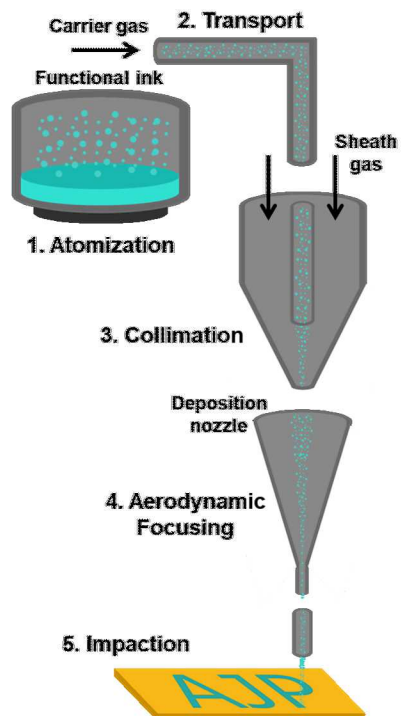
Transport effects within the printhead can strongly affect process outcomes



Aerosol jet printing background/motivation

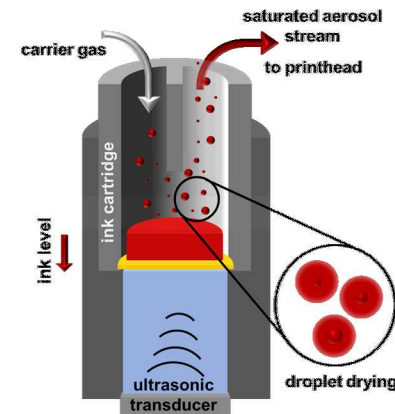
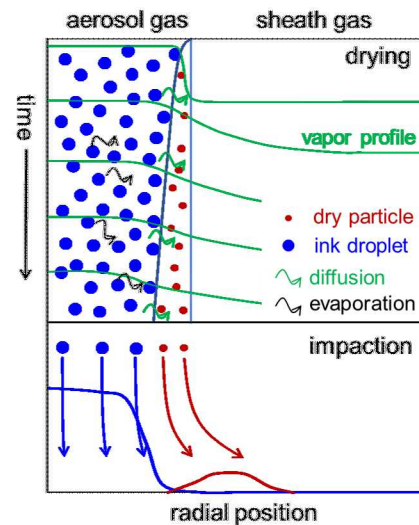
Aerosol physics

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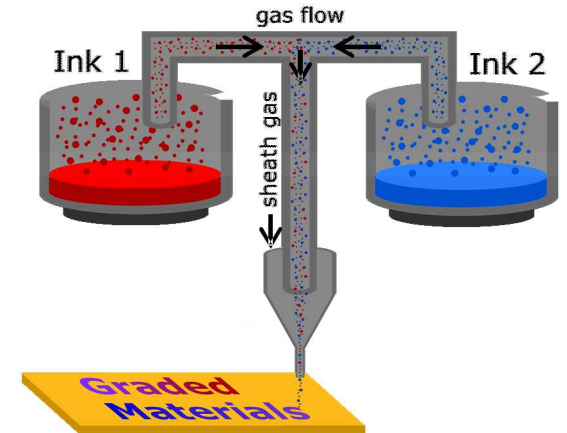
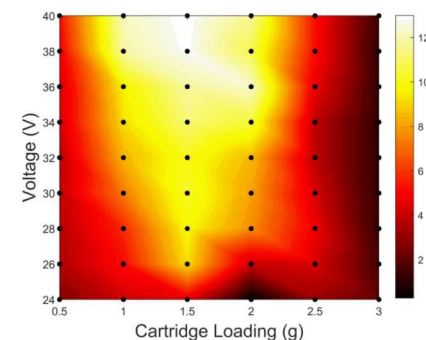
Mass transfer effects

- Conceptual framework
- Ink formulation
- Printhead design



Process drift

- Understanding drift
- Passive strategies
- Closed-loop control

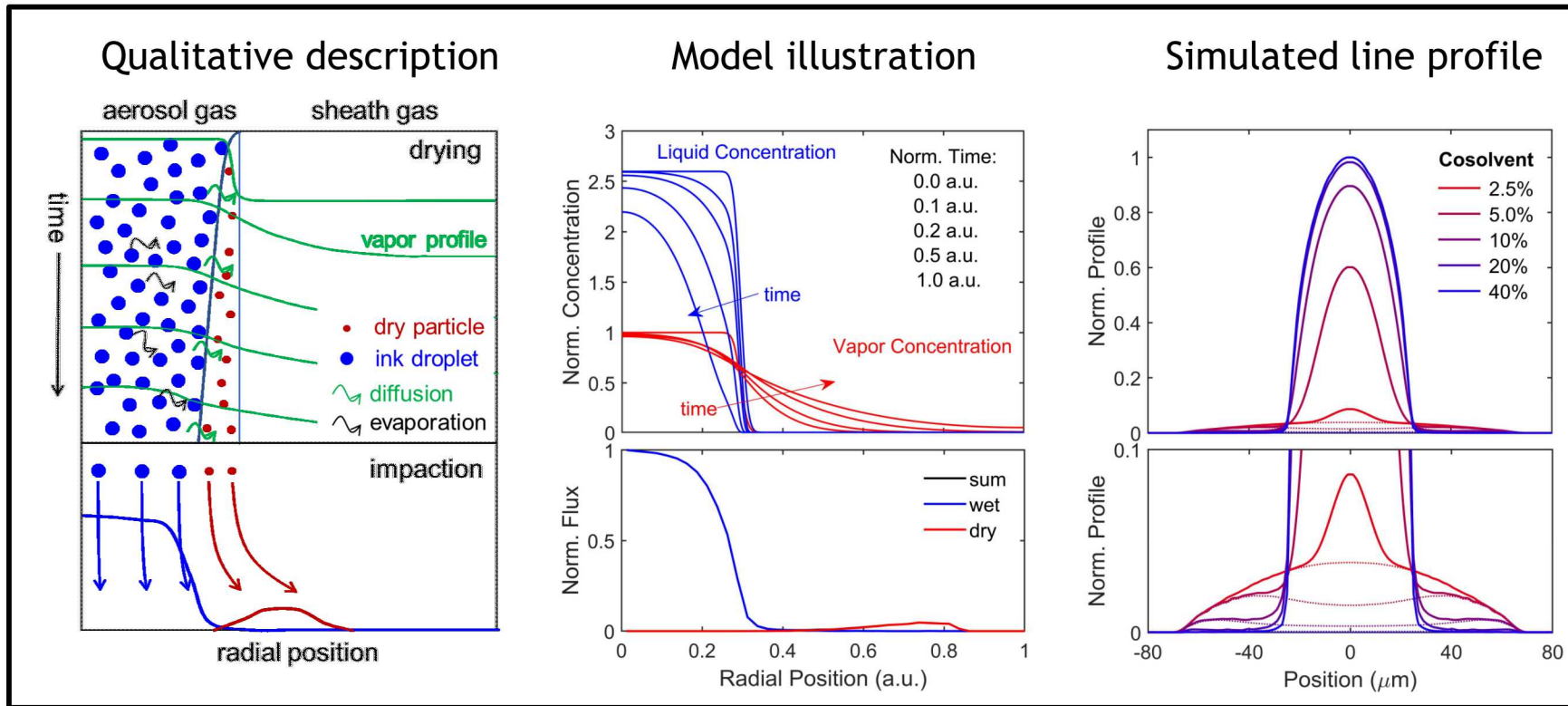


Multimaterial printing

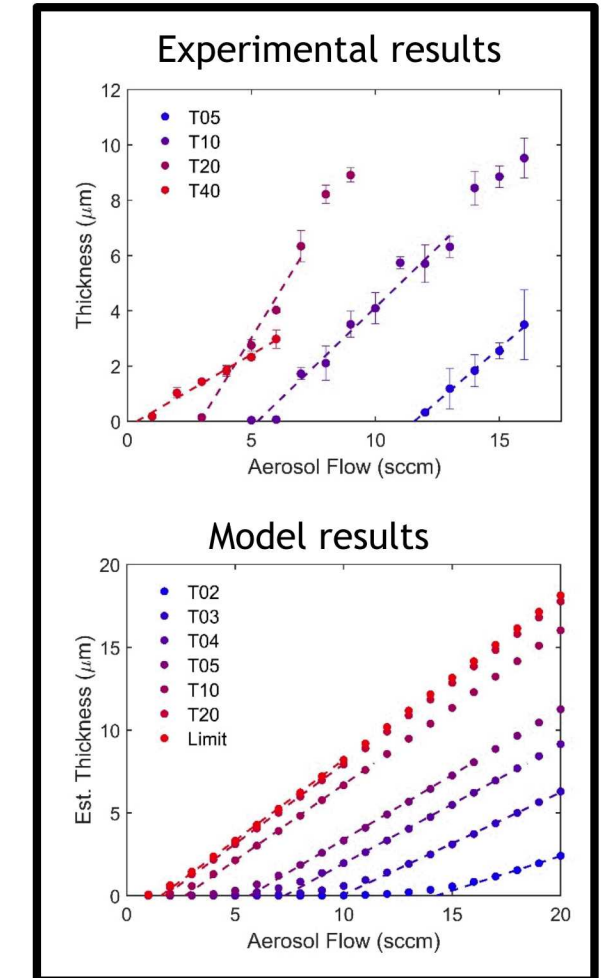
- Proof-of-concept
- Graded functionality
- Application potential

Describing mass transfer effects during printing

A dry sheath gas can induce spatially nonuniform drying within the printhead on timescales of relevance



- Radial drying within the printhead is induced by the sheath gas
- Peripheral droplets can be completely dried before vapor saturates
- Models guide ink formulation, printer design, and process optimization



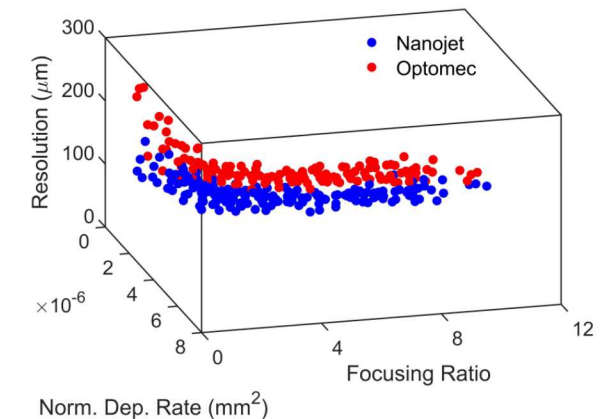
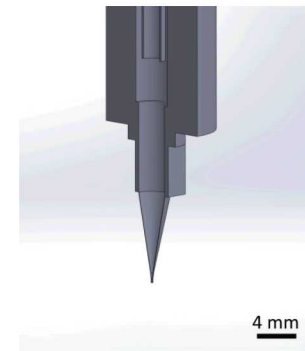
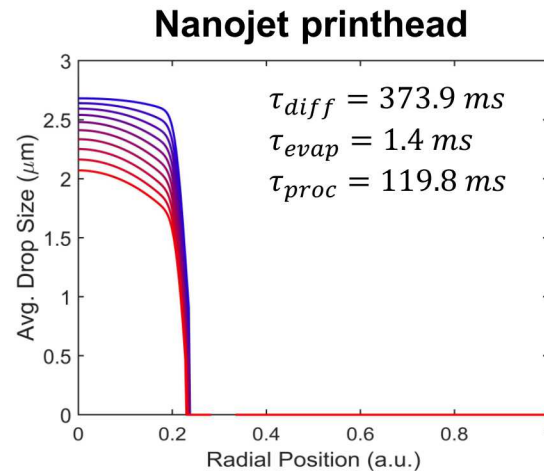
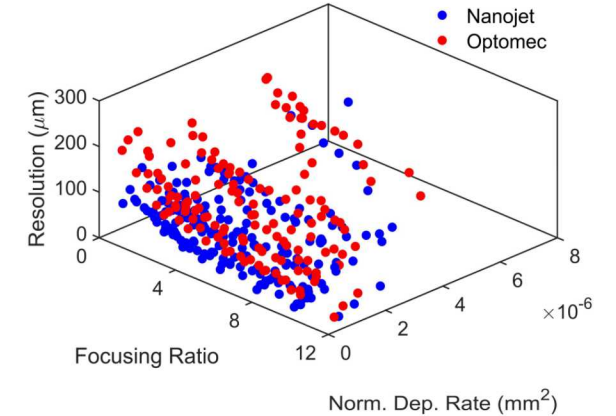
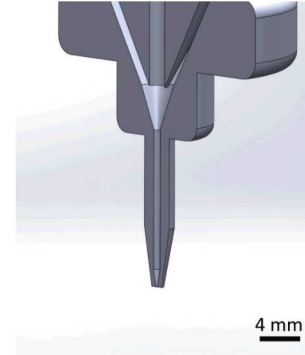
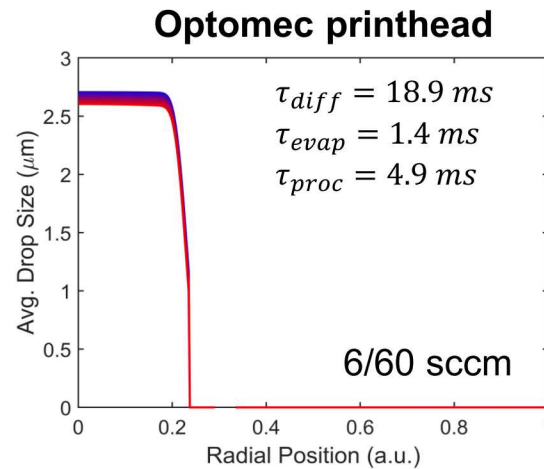
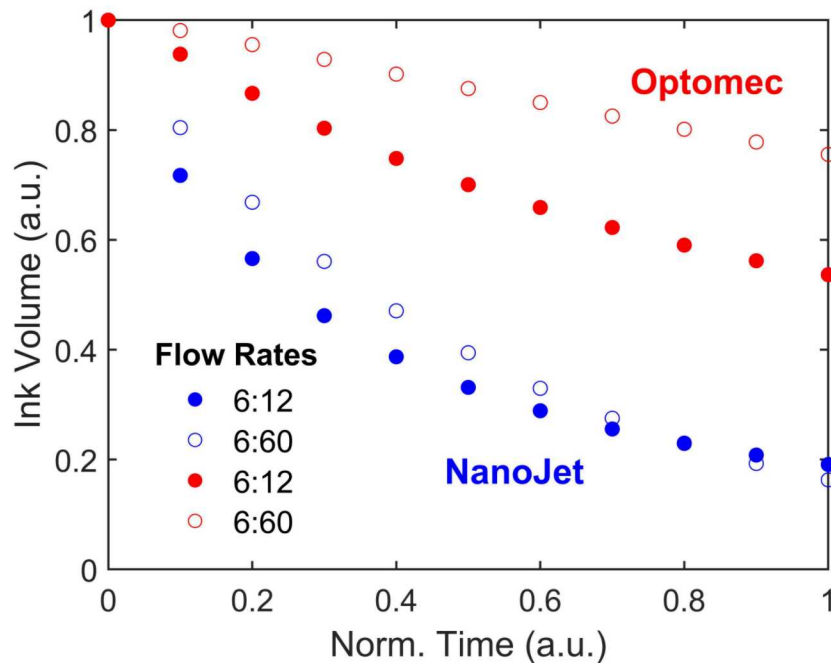
9 Implications for ink formulation and printhead design

$$c_o = \frac{v_a v_2 V_m}{n_{sat}} * \frac{1}{1 + \chi}$$

$$\tau_{proc} = \frac{\pi L R^2}{2Q} \sqrt{\frac{1 + \chi}{\chi}}$$

$$\tau_{diff} = \frac{R^2}{D} \left[1 - \sqrt{1 - \sqrt{\frac{\chi}{1 + \chi}}} \right]^2$$

$$\tau_{evap} = \frac{a_0^2}{3D v_a v_2^{\frac{1}{3}}}$$



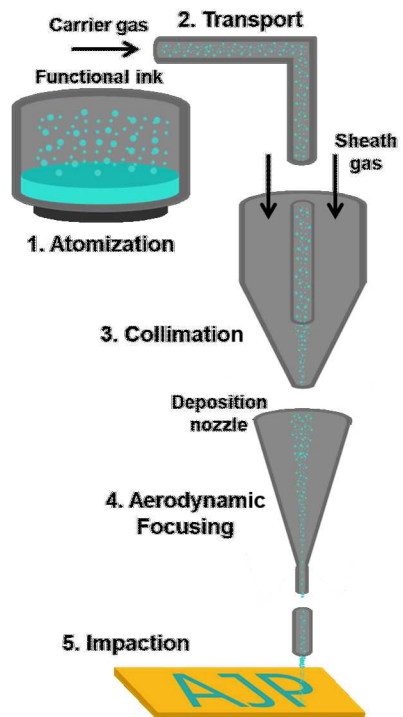
- Ink formulation should account for solvent vapor saturation
- Printhead design affects drying timescales

- Clear difference in printing characteristics aligns with predicted wet/dry deposition

Aerosol jet printing background/motivation

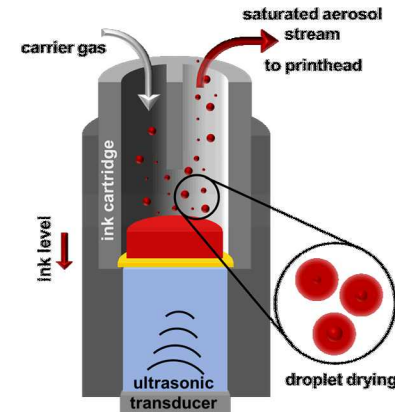
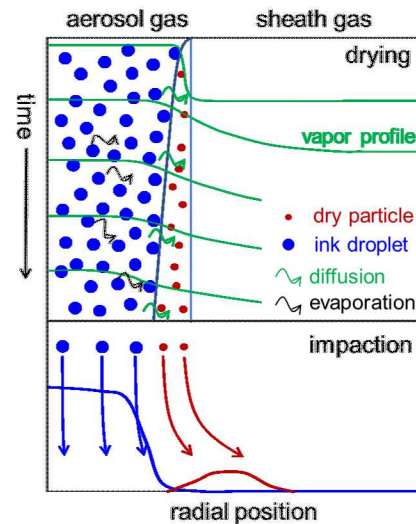
Aerosol physics

- General principles
- Baseline understanding
- Limitations



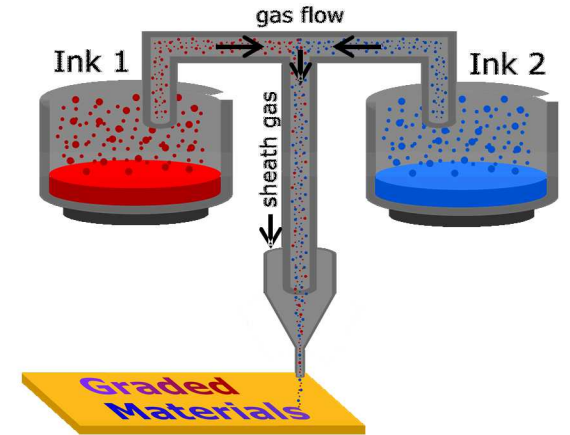
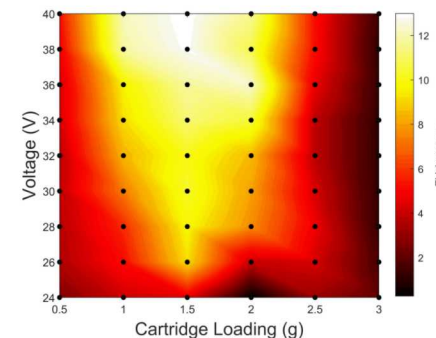
Mass transfer effects

- Conceptual framework
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Process drift

- Understanding drift
- Passive strategies
- Closed-loop control



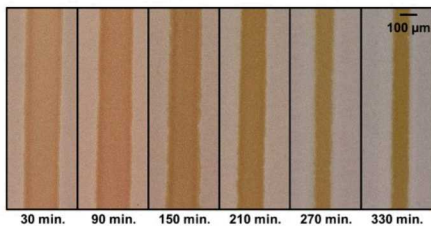
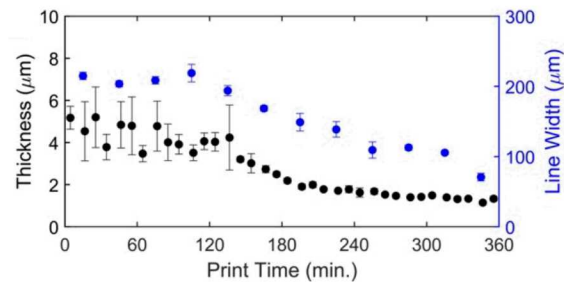
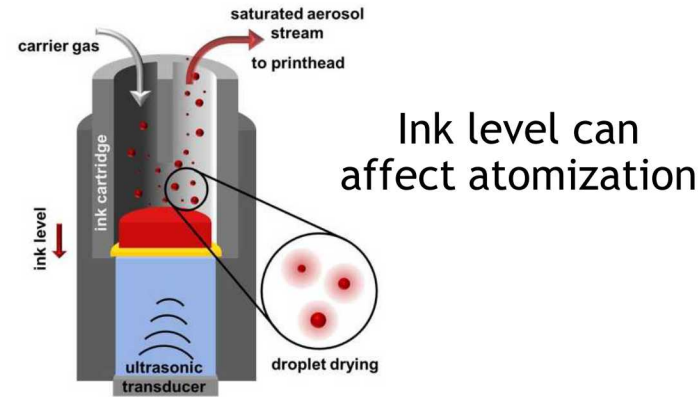
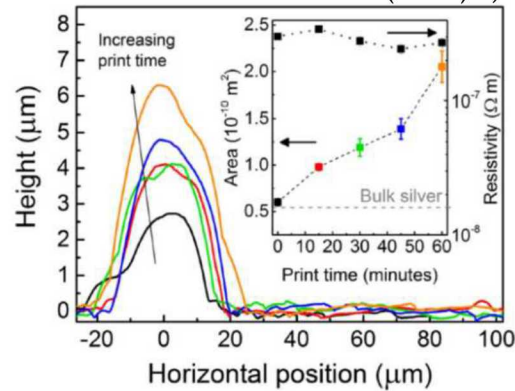
Multimaterial printing

- Proof-of-concept
- Graded functionality
- Application potential

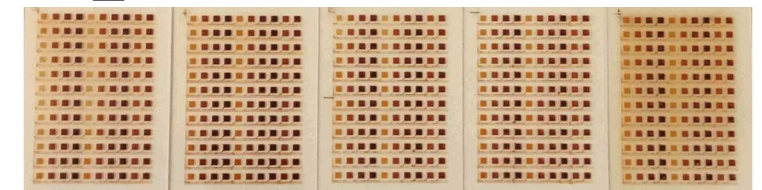
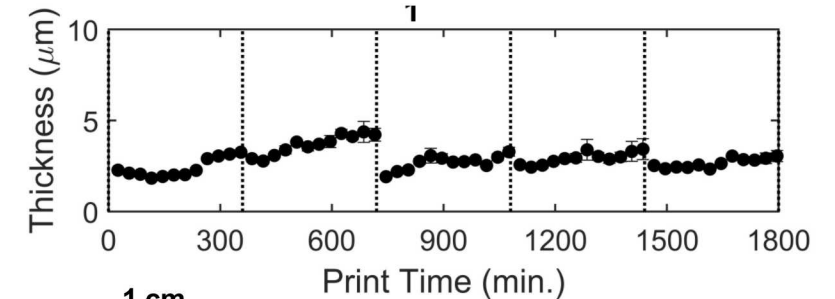
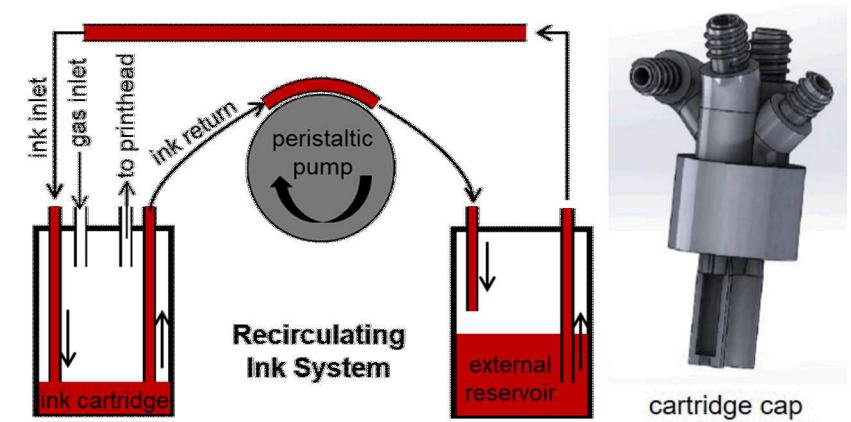
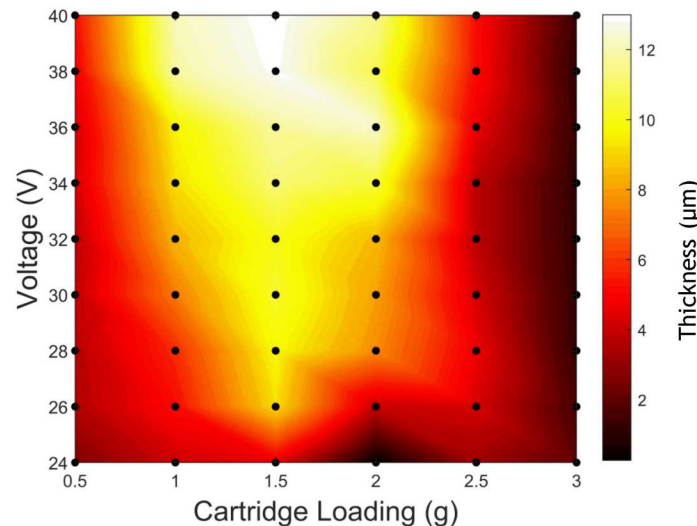
Leveraging understanding to mitigate process drift

Fundamental mechanisms support understanding of process drift, a major limitation of aerosol jet printing

Smith, M., et al. *Flex. Print. Electron.* (2017) 2, 015004.



Decrease in both deposition rate and line width over 6 hours of printing



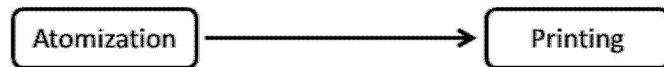
Custom recirculating ink system improves consistency in 30 hr print run

Working towards closed loop process control

Closed-loop control is highly sought across additive manufacturing processes

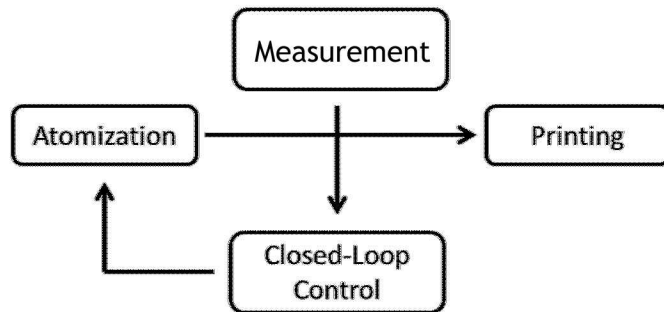
Custom system developed to assess aerosol density within the printhead

Standard Printing

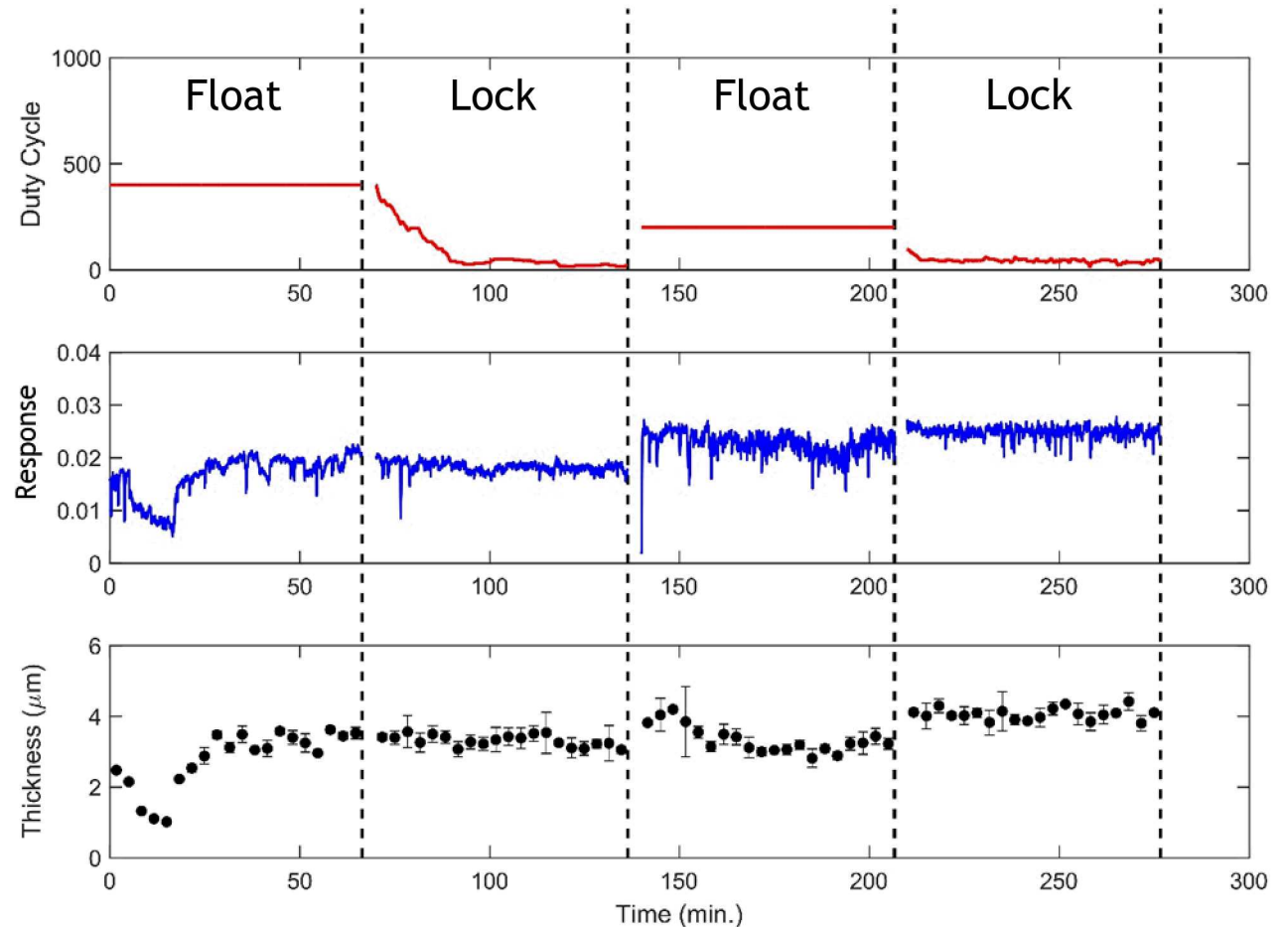


Susceptible to drift in atomization yield due to temperature changes, electronic drift, atomizer aging, ink composition change, and ink volume change

Closed-Loop Control



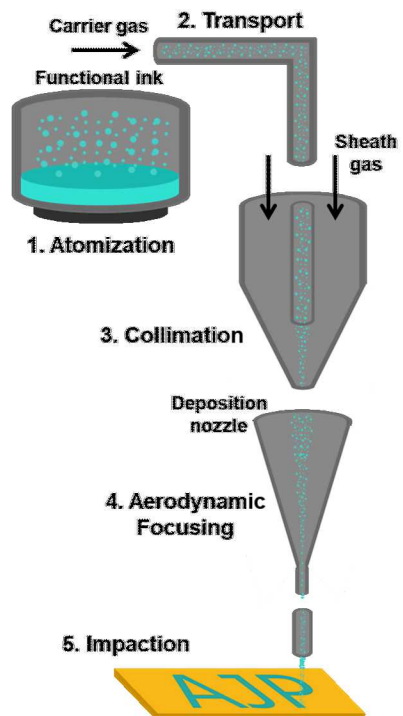
Changes in atomization are detected in real time and compensated for if possible; if not possible, allows automatic shutdown or alert to indicate potential problems



Aerosol jet printing background/motivation

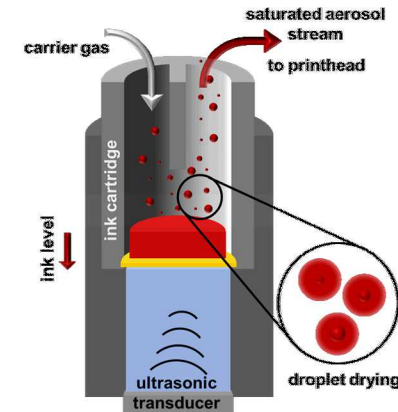
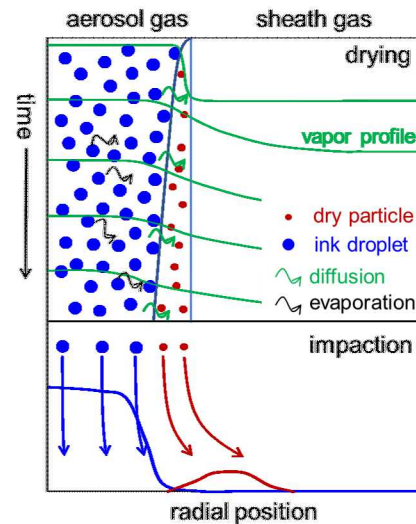
Aerosol physics

- General principles
- Baseline understanding
- Limitations



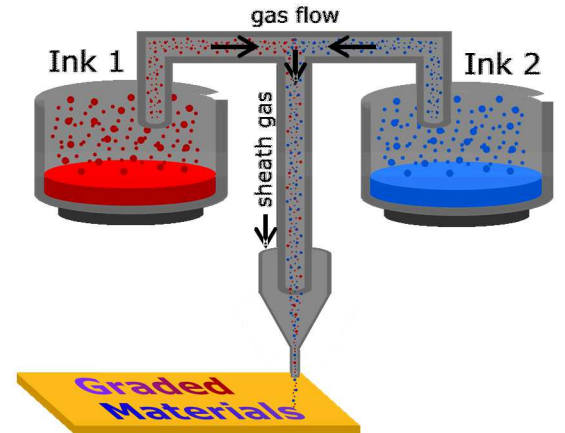
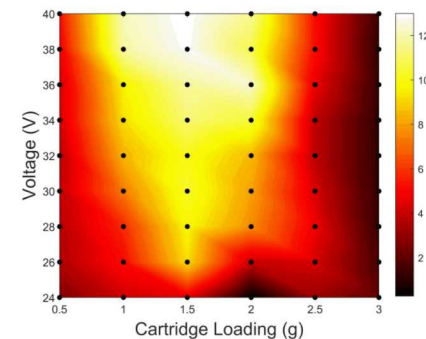
Mass transfer effects

- Conceptual framework
- Ink formulation
- Printhead design



Process drift

- Understanding drift
- Passive strategies
- Closed-loop control

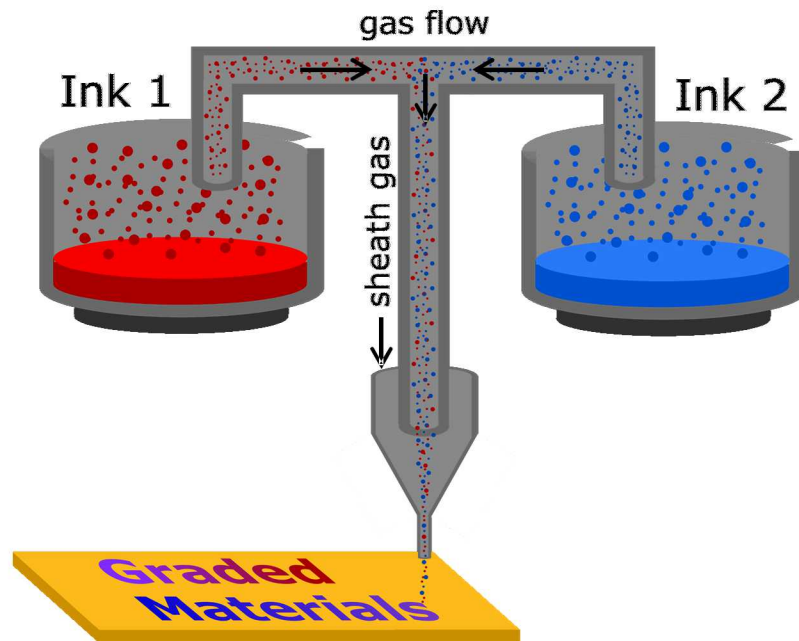


Multimaterial printing

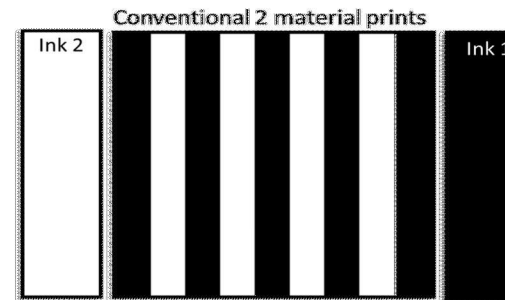
- Proof-of-concept
- Graded functionality
- Application potential

Extending versatility to graded multimaterial patterning

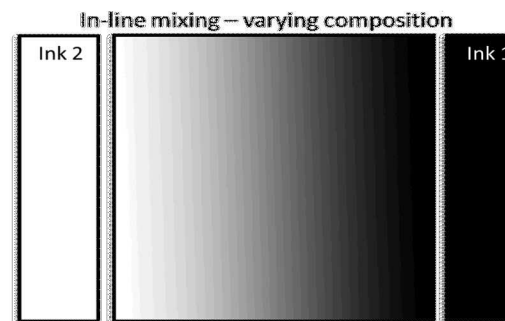
In-line mixing of aerosols upstream of the printhead for simultaneous codeposition of distinct inks



Binary multimaterial printing

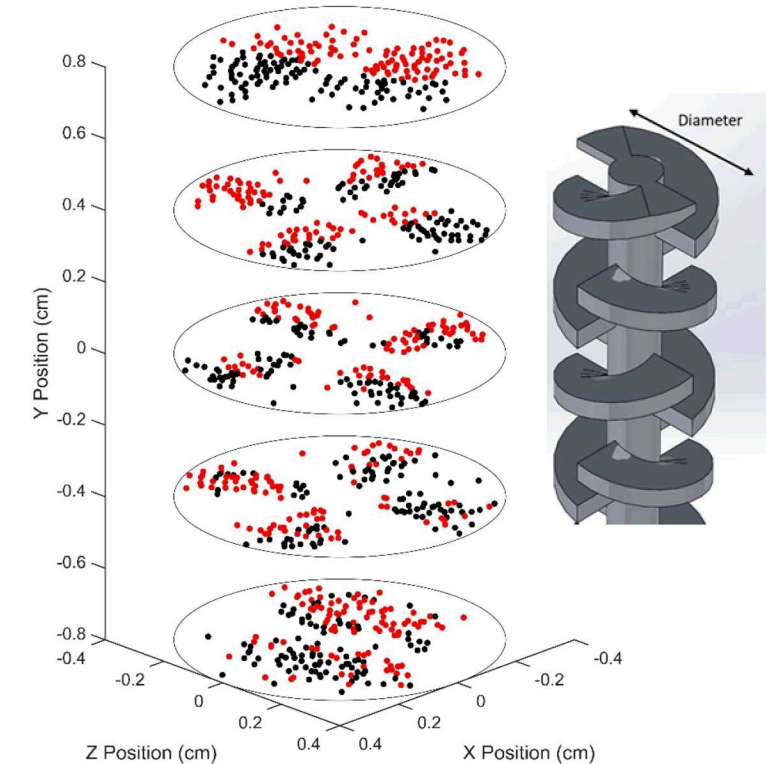


Graded multimaterial printing



Unique multimaterial printing platform for graded patterning with micron-scale mixing and digital control

Mixer Cross Sections



Laminar static mixer improves droplet homogeneity

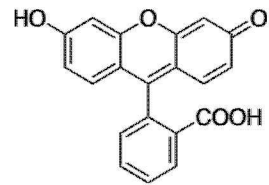
Validation of multimaterial printing concept

Initial demonstration with optical materials for straightforward characterization

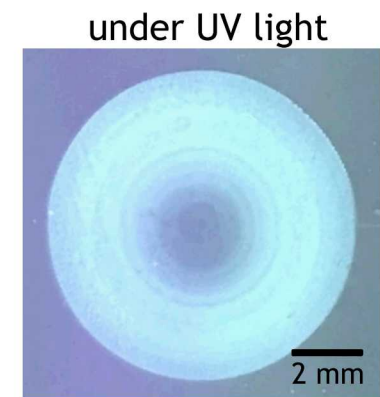
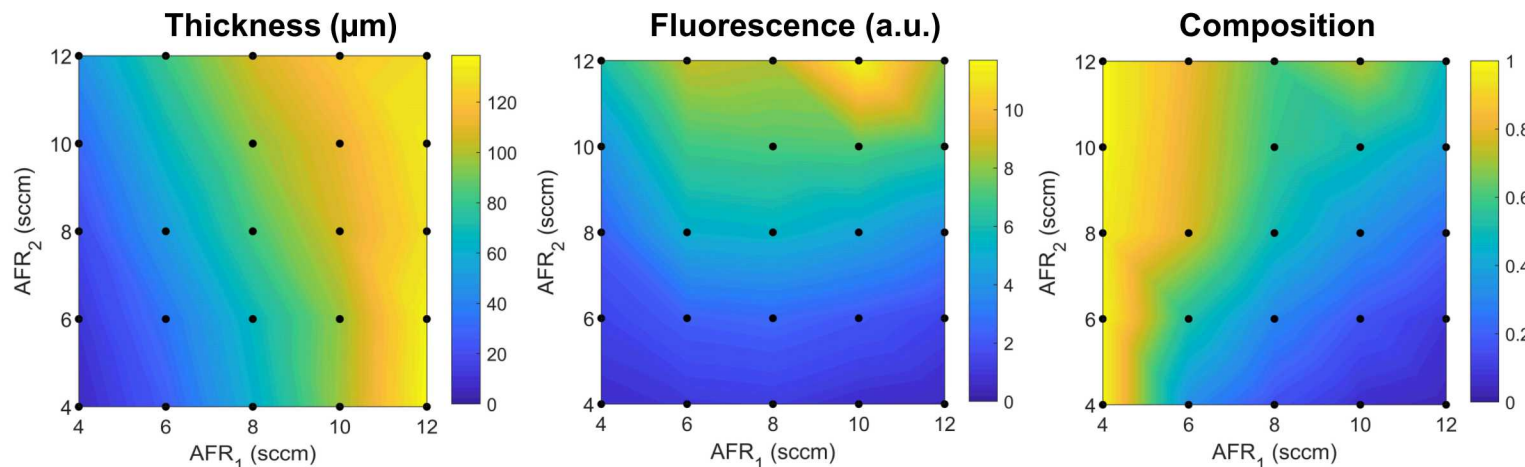
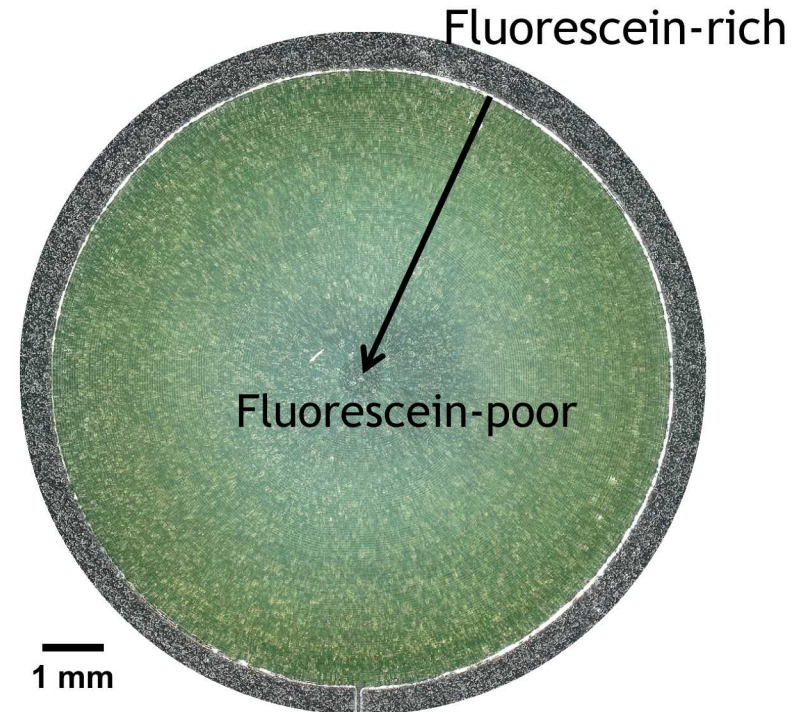
Ink 1: Norland optical adhesive NOA 61

Ink 2: Norland optical adhesive NOA 61 with fluorescein and ZrO_2

- Fluorescence microscopy to measure amount of fluorescein-containing ink
- Thickness measurement for overall deposition rate
- Calibration required to deconvolute thickness and composition



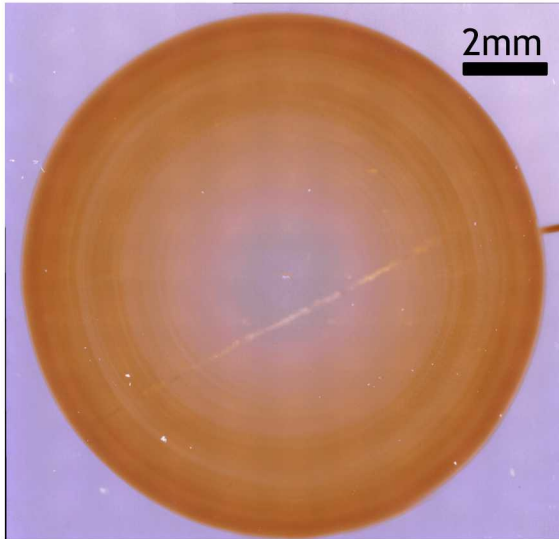
Fluorescein dye



Promise for functionally-graded patterning

Widespread opportunities for functionally graded materials based on broad compatibility of aerosol jet printing

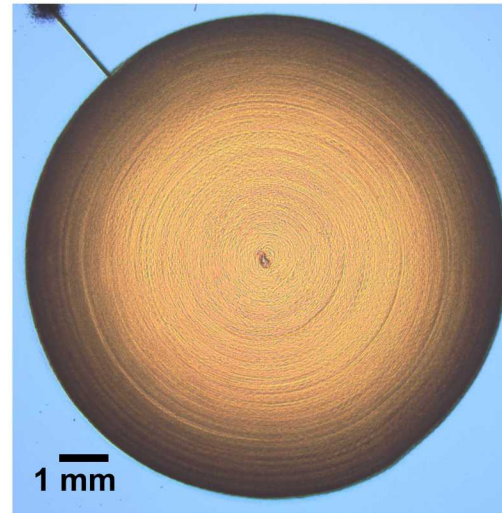
Magnetite composite



Broad scope of compatible materials

Epoxies
Acrylates
Polyimide
PMMA
Magnetite
Silver
Graphene
Gold
Metal oxides
Metal hydrides

Graphene/magnetite composite



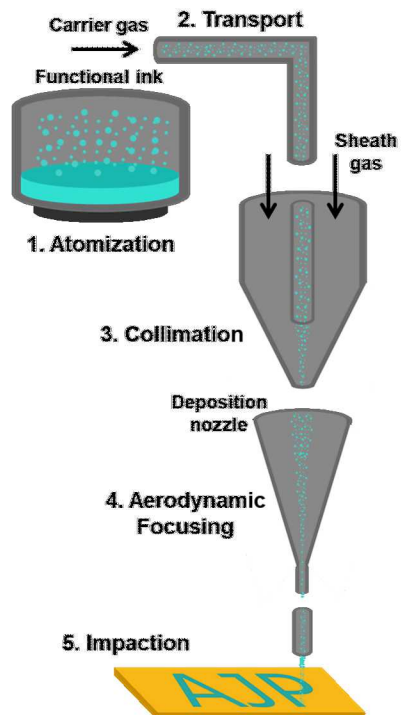
- Potential application areas
- Mechanical interfaces
 - Gradient index optics/RF
 - Electronic devices
 - Electrochemical systems

Platform for fabricating arbitrary gradients offers extensive design flexibility for widespread applications in mechanical, electronic, magnetic, and thermal systems

Aerosol jet printing background/motivation

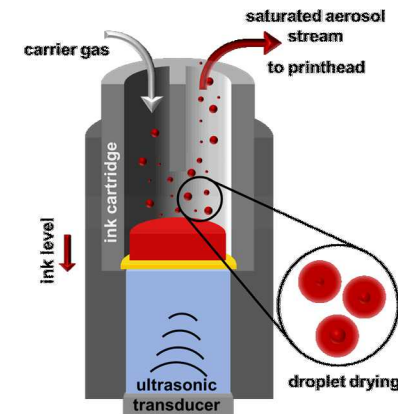
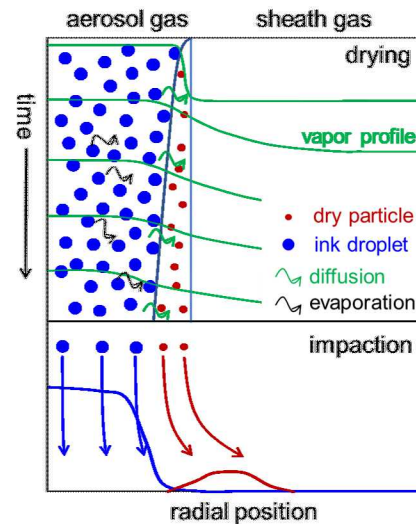
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- Limitations



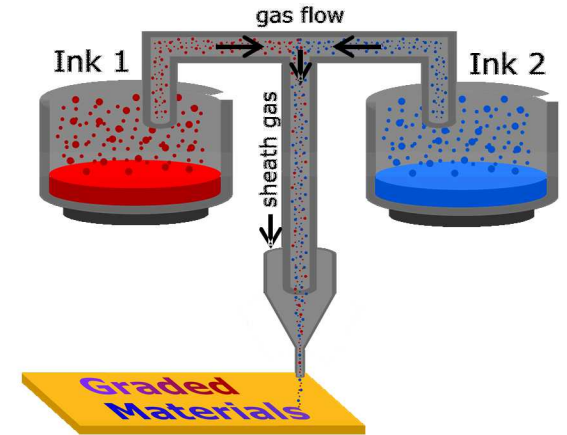
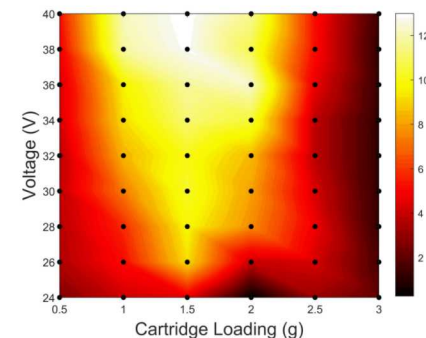
Mass transfer effects

- Conceptual framework
- Ink formulation
- Printhead design



Process drift

- Understanding drift
- Passive strategies
- Closed-loop control



Multimaterial printing

- Proof-of-concept
- Graded functionality
- Application potential



Rebecca R. Tafoya



Adam W. Cook



Bryan J. Kaehr



P. Randall Schunk

Thanks for your attention!

Thanks to Rebecca R. Tafoya, Adam W. Cook, Bryan J. Kaehr, and P. Randall Schunk
Funding from the Sandia LDRD program through the Truman Fellowship

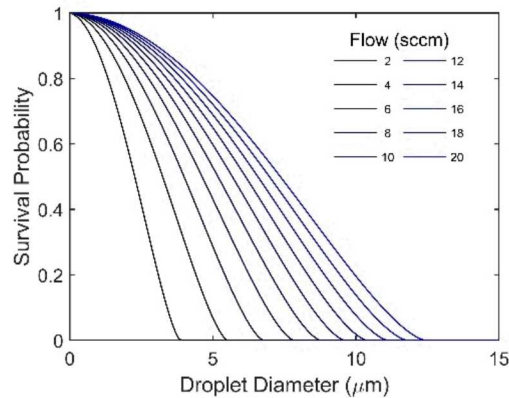


**LABORATORY DIRECTED
RESEARCH & DEVELOPMENT**

Support Slide 1: Aerosol Physics

2. Transport

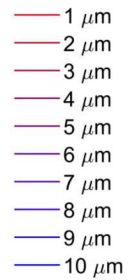
$$F = \frac{2}{\pi} (\alpha\beta + \sin^{-1} \beta - 2\alpha^3\beta)$$



$$\text{where } \alpha = \left(\frac{3Lu_{TS}}{8u_a R} \right)^{\frac{1}{3}}; \quad \beta = \sqrt{1 - \alpha^2};$$

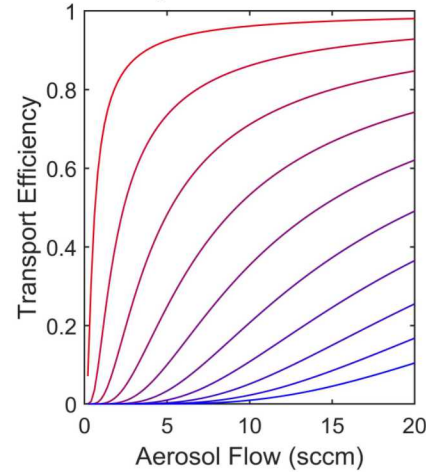
Thomas, J.W. JAPCA J. Air Waste Manage. Assoc. (1958) 8, 32.

Median Diameter

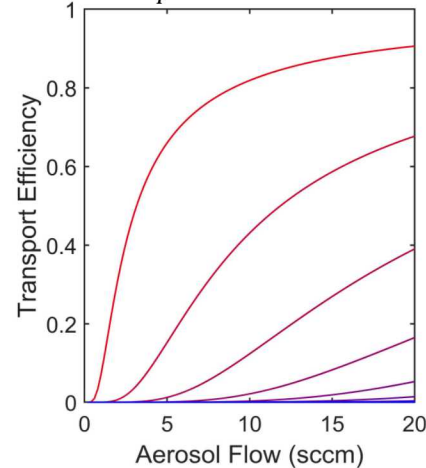


- Large droplets settle under gravity
- System design is important for droplet loss
- Droplet settling can limit print time

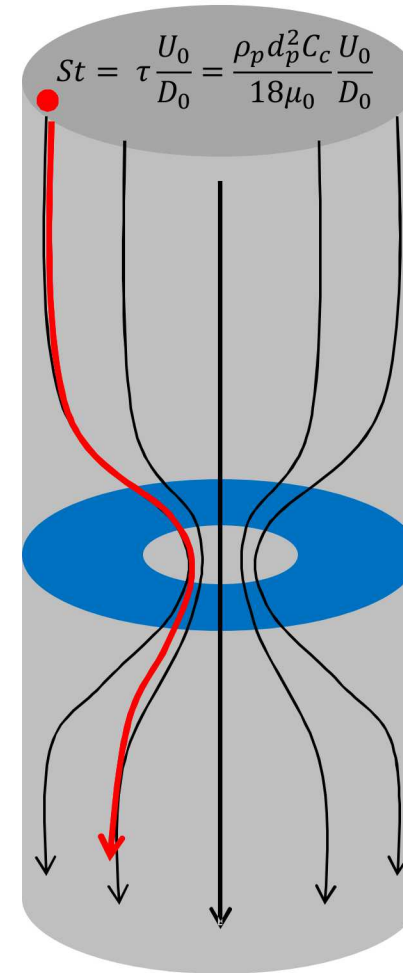
$R_H = 3 \text{ mm}, L_H = 20 \text{ mm},$
 $L_T = 100 \text{ mm}$



$R_H = 1 \text{ mm}, L_H = 200 \text{ mm},$
 $L_T = 500 \text{ mm}$

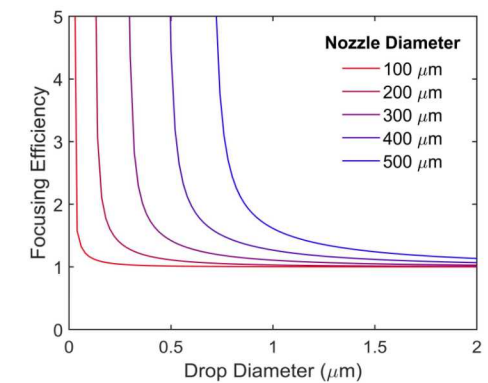
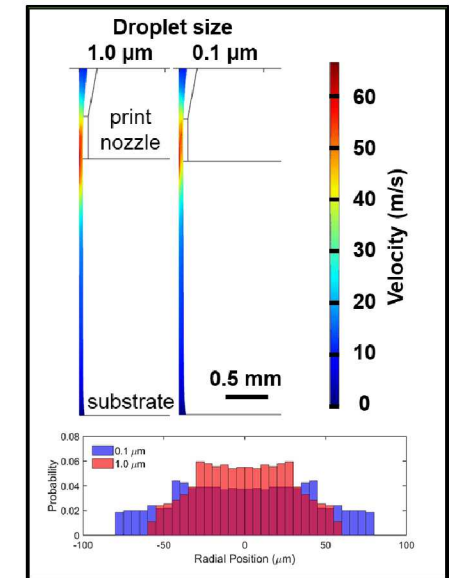


4. Focusing



Israel, G.W. and Friedlander, S.K. J. Colloid Interface Sci. (1967) 24, 330.

5. Impaction



- Impaction of droplets on substrate dependent on inertia
- Droplet size, flow rates, and nozzle diameter all affect the process

Support Slide 2: Mass Transfer

$$c_o = \frac{v_a v_2 V_m}{n_{sat}} * \frac{1}{1 + \chi}$$

$$\tau_{proc} = \frac{\pi L R^2}{2Q} \sqrt{\frac{1 + \chi}{\chi}}$$

$$\tau_{diff} = \frac{R^2}{D} \left[1 - \sqrt{1 - \sqrt{\frac{\chi}{1 + \chi}}} \right]^2$$

$$\tau_{evap} = \frac{a_0^2}{3Dv_a v_2^{\frac{1}{3}}}$$

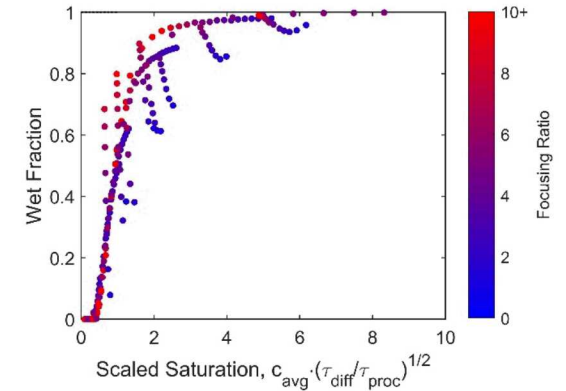
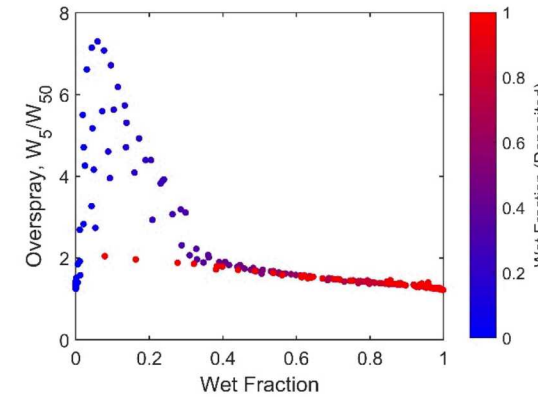
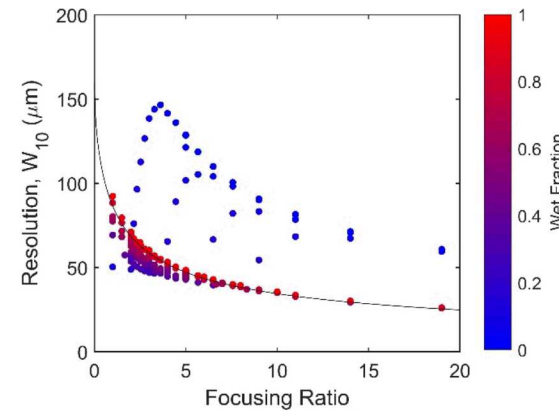
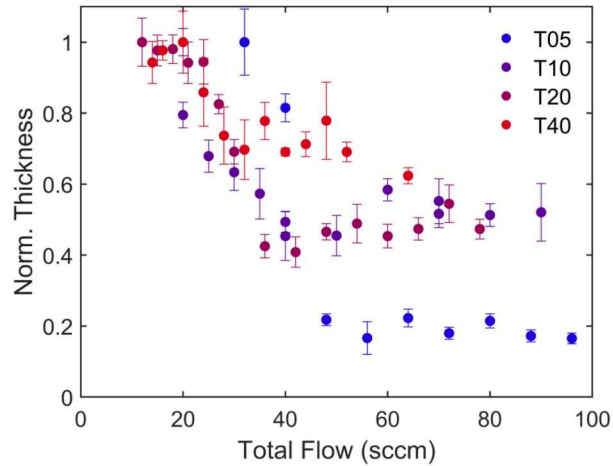
Machine parameters: L, R
(length, radius)

Process parameters: Q, χ
(flow, focusing ratio)

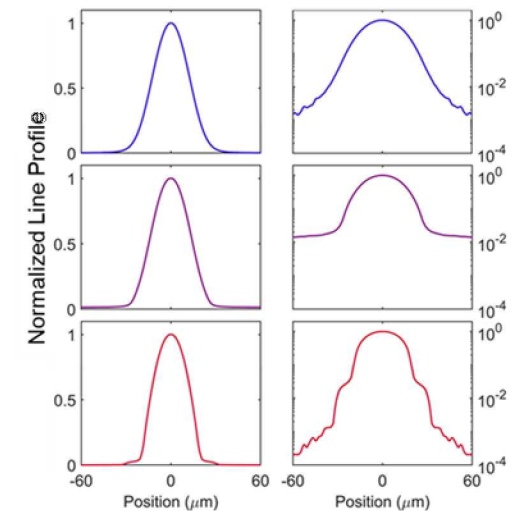
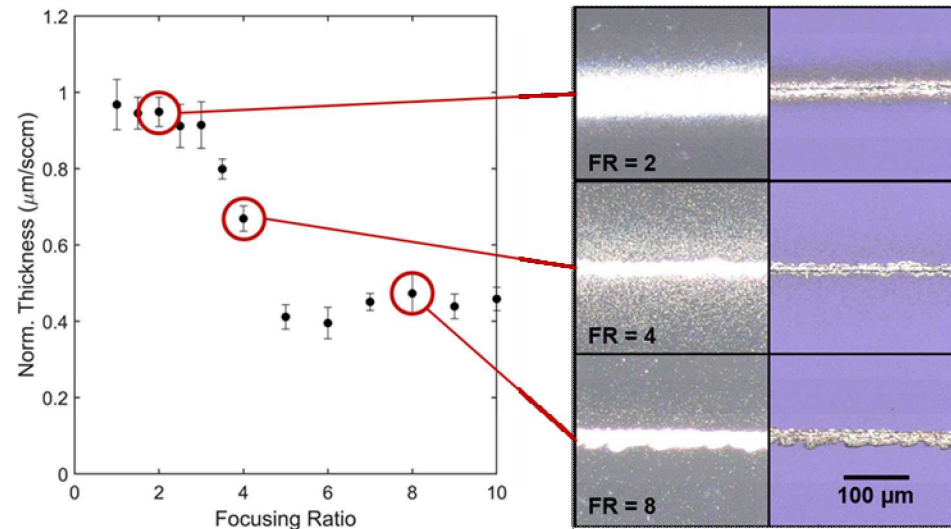
Ink parameters: n_{sat}, v_2
(vapor saturation conc., volume fraction)

Mixed or poor control: v_a, a_0
(aerosol fraction, initial droplet size)

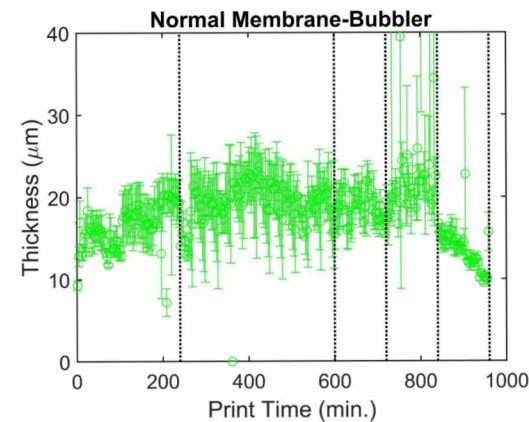
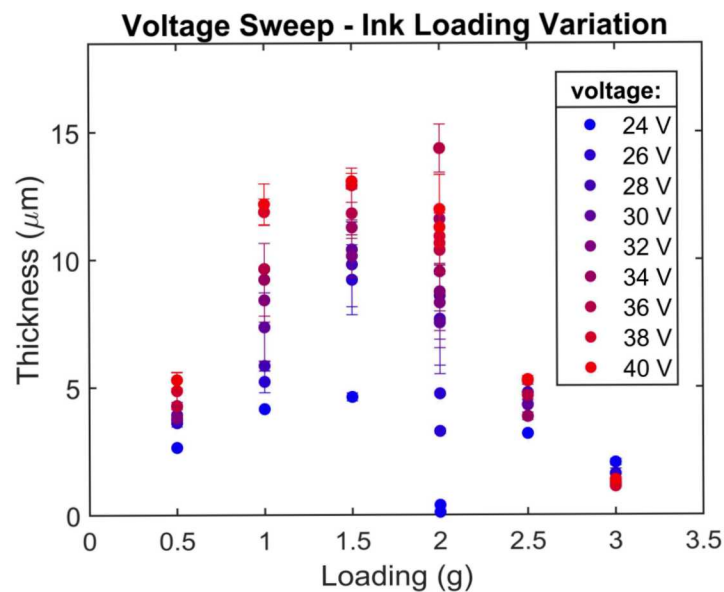
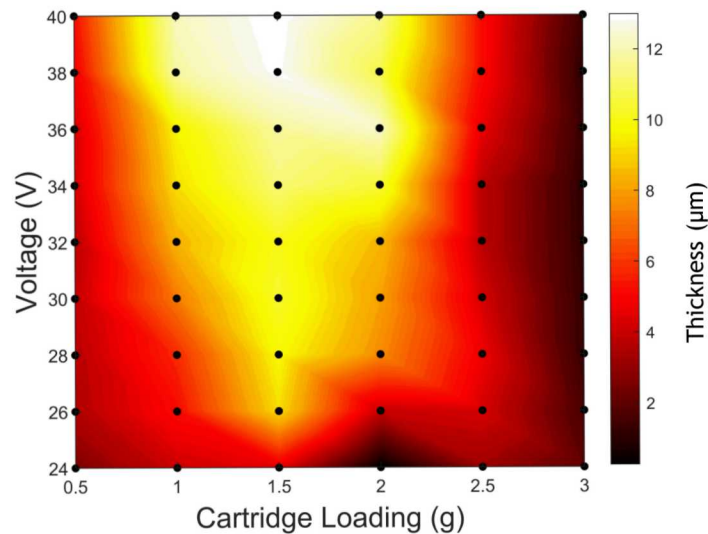
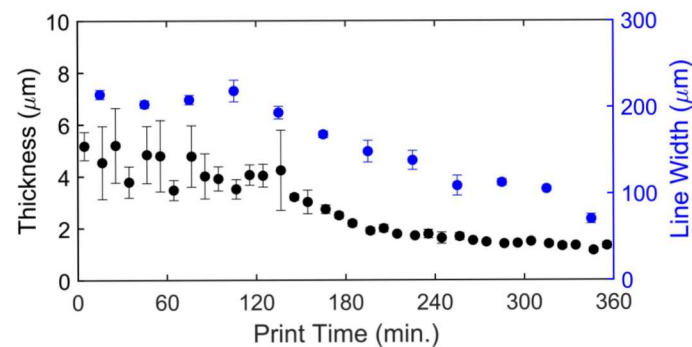
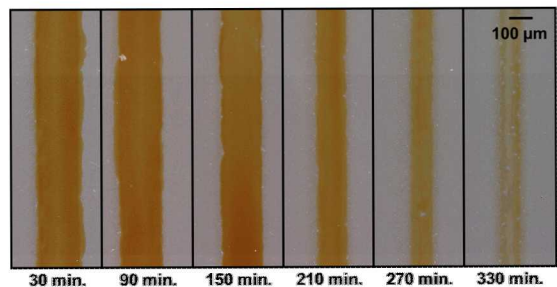
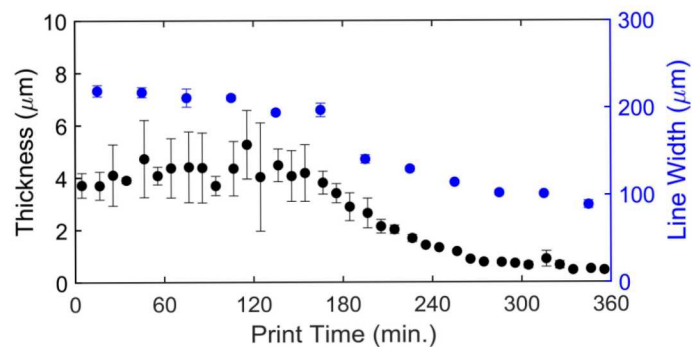
Drying induced by the sheath gas affects deposition rate



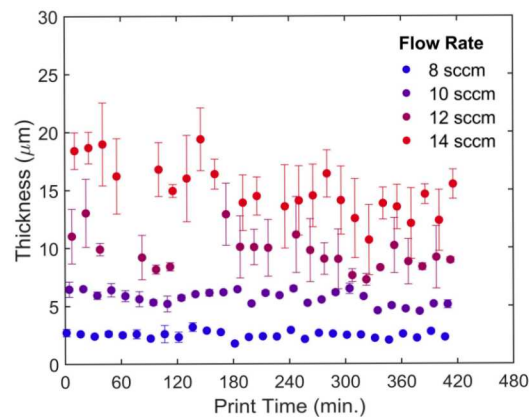
- Understanding from models helps explain some unusual observed print characteristics
- Rebound of dry particles may have a role under some conditions



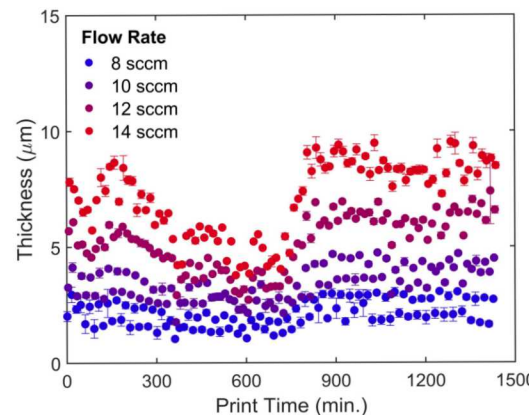
Support Slide 3: Process Drift



Polyimide

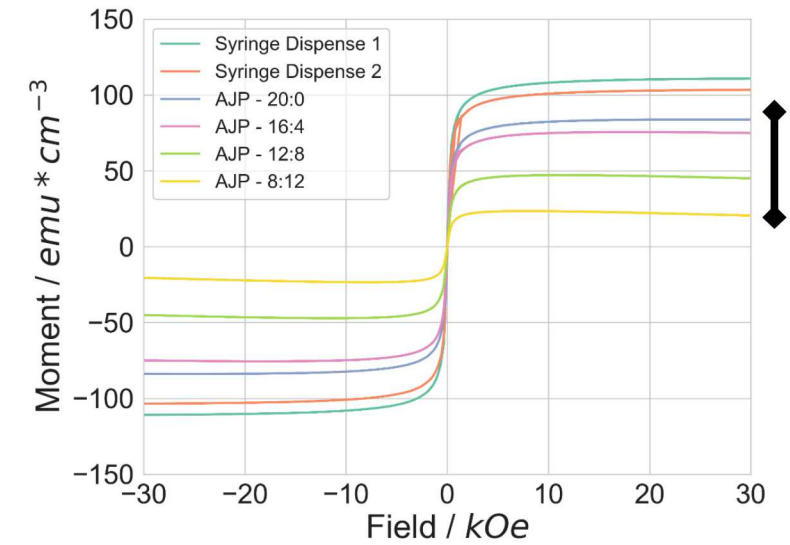
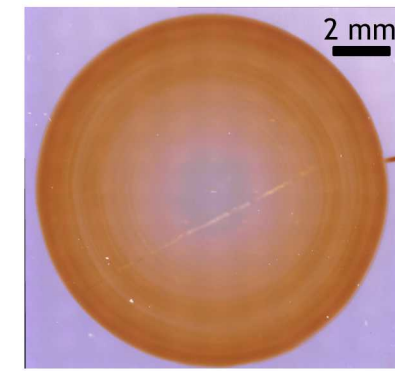
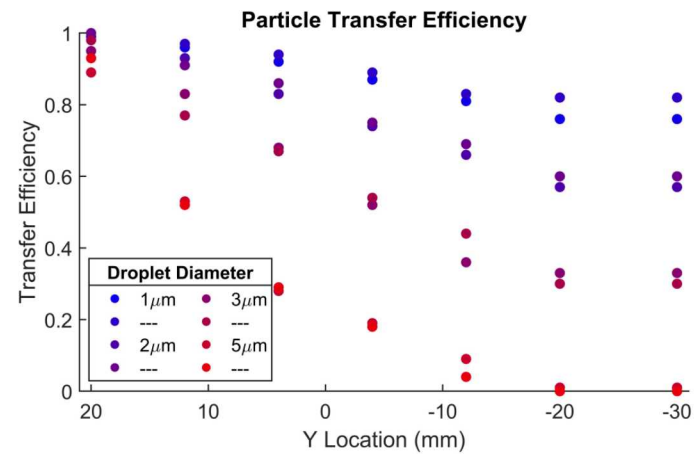
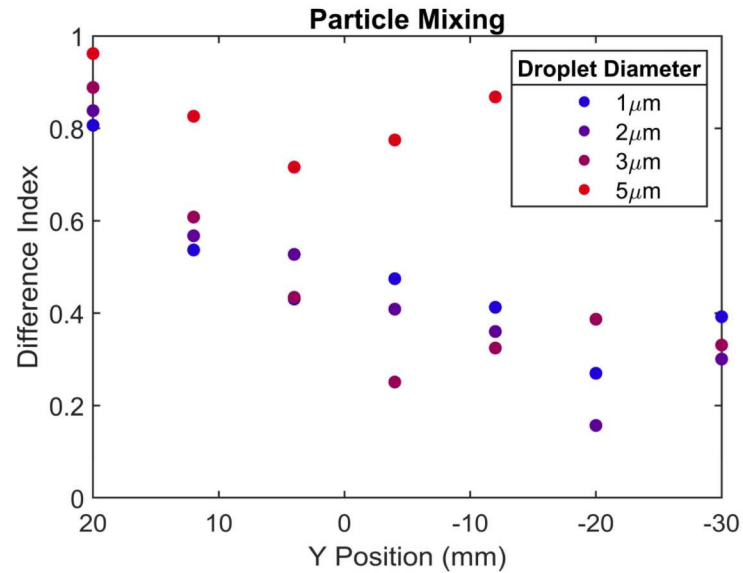
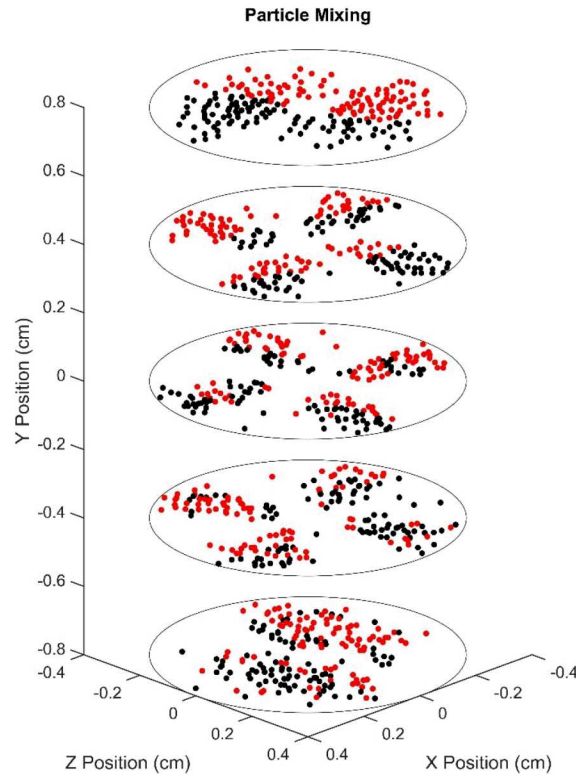


PMMA (44 V)



PMMA (40 V)

Support Slide 4: Multimaterial Printing



Sample (Fe_3O_4 : PMMA)	Fe_3O_4 Loading (mg/cm^2)	St. Dev. (mg/cm^2)
AJP - 20:0	2.39	0.16
AJP - 16:4	1.55	0.08
AJP - 12:8	0.64	0.03
AJP - 8:12	0.35	0.02