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Aerosol jet printing of functionally graded materials

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Advanced Materials Laboratory, Sandia National Laboratories

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Acknowledgements

Advanced Materials Laboratory

Materials chemistry, ink formulation, and advanced manufacturing
Access to UNM to enhance collaboration and promote student engagement

Rebecca R. Tafoya

Bryan J. Kaehr

Adam W. Cook

P. Randall Schunk

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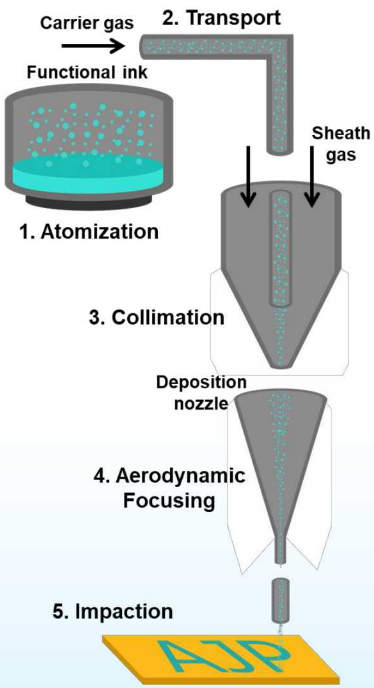
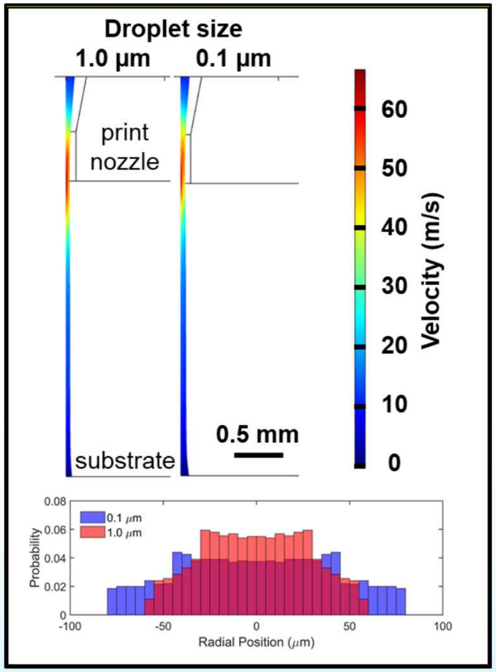
LABORATORY DIRECTED RESEARCH & DEVELOPMENT

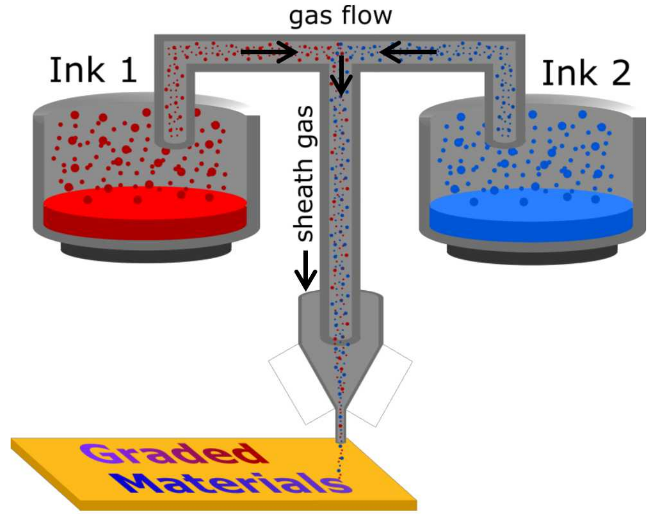
Outline

Basics of Aerosol Jet Printing

1. General mechanism
2. Process models
3. Implications for printer design, ink formulation, process optimization





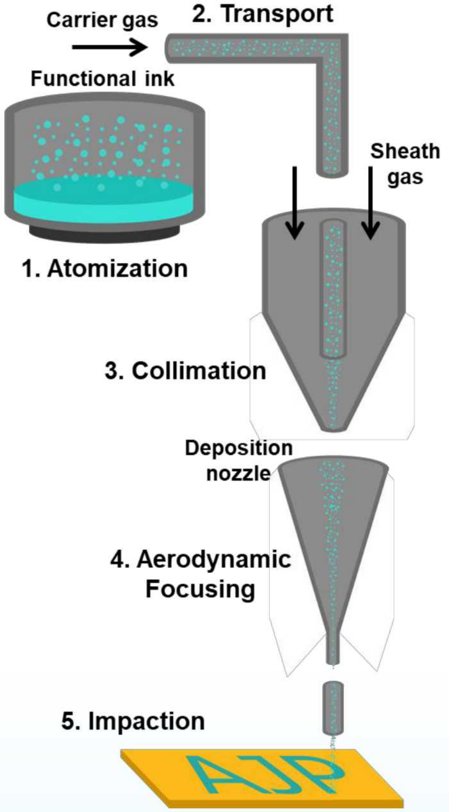


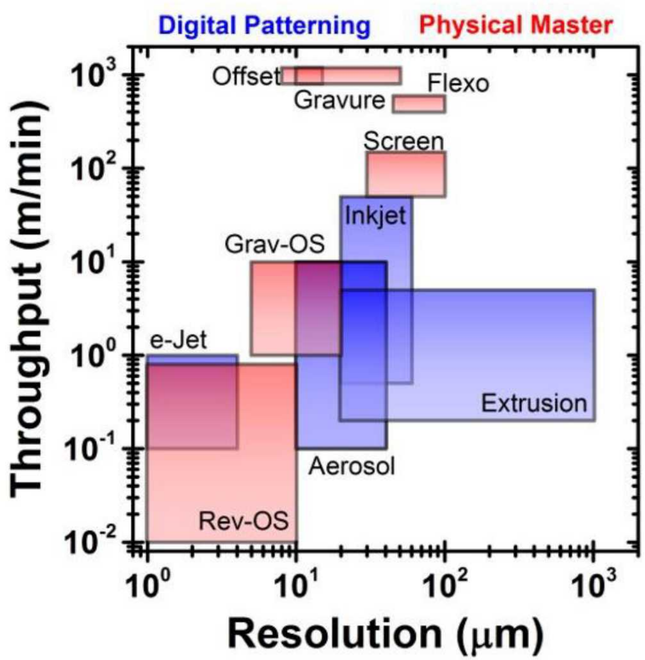
Graded Multimaterial Printing

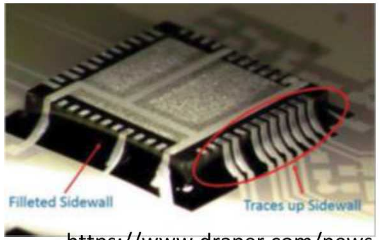
1. Concept
2. Printhead design
3. Proof-of-concept demonstration
4. Characterization and calibration
5. Functional gradient demonstrations
6. Future work

Fundamentals of Aerosol Jet Printing

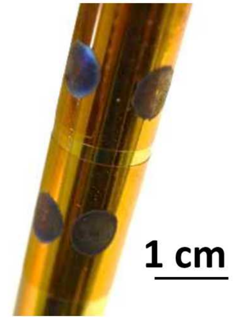
Aerosol jet printing (AJP) uses focused deposition of micron-scale ink droplets suspended in a carrier gas flow

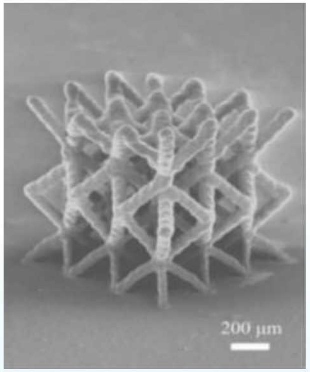






https://www.draper.com/news





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

Benefits:
- Digital
- Non-contact
- High-resolution
- Versatile

Limitations:
- Process drift
- Low throughput

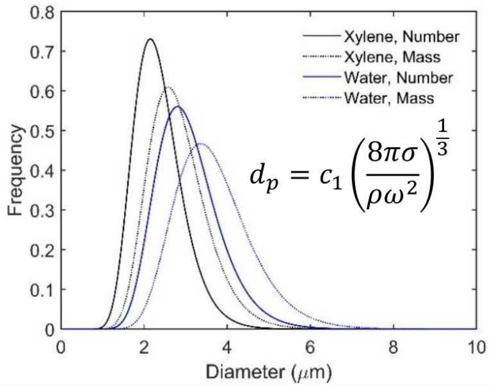
AJP is a versatile technology for prototyping functional devices

Secor, E.B. *Northwestern University* (**2017**)
Secor, E.B. *Flex. Print. Electron.* (**2018**) 3, 035002.

Fundamentals of Aerosol Jet Printing

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

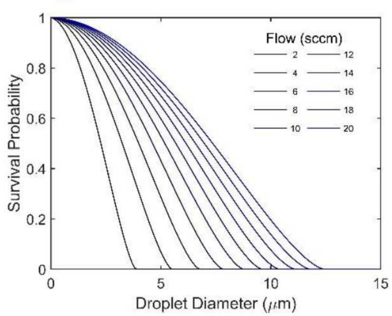
1. Atomization



$$d_p = c_1 \left(\frac{8\pi\sigma}{\rho\omega^2} \right)^{\frac{1}{3}}$$

2. Transport

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

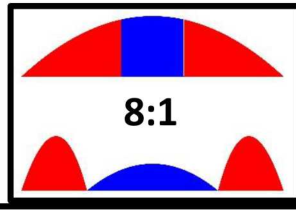


$$where\ \alpha = \left(\frac{3Lu_{TS}}{8u_a R} \right)^{\frac{1}{3}}; \qquad \beta = \sqrt{1-\alpha^2};$$

3. Collimation

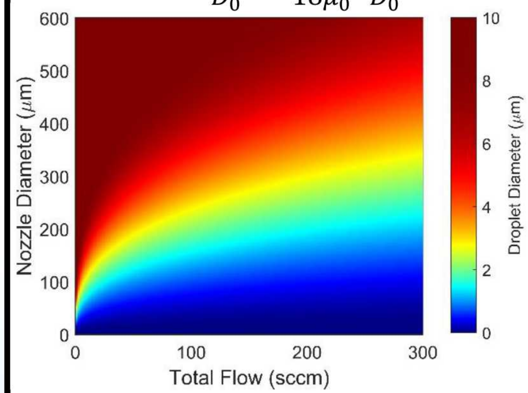
$$f_a = \int_0^{2\pi} d\theta \int_0^{R_a} rdr \left[2u_a \left(1 - \frac{r^2}{R^2} \right) \right] = \pi u_a R_a^2 \left[2 - \frac{R_a^2}{R^2} \right]$$

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

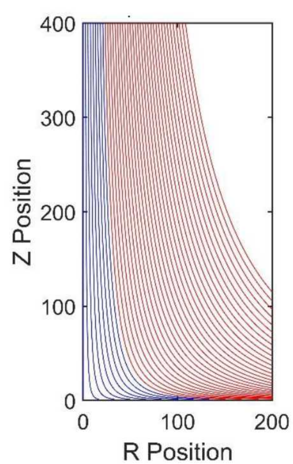


4. Focusing

$$St = \tau \frac{U_0}{D_0} = \frac{\rho_p d_p^2 C_c}{18\mu_0} \frac{U_0}{D_0}$$



5. Impaction

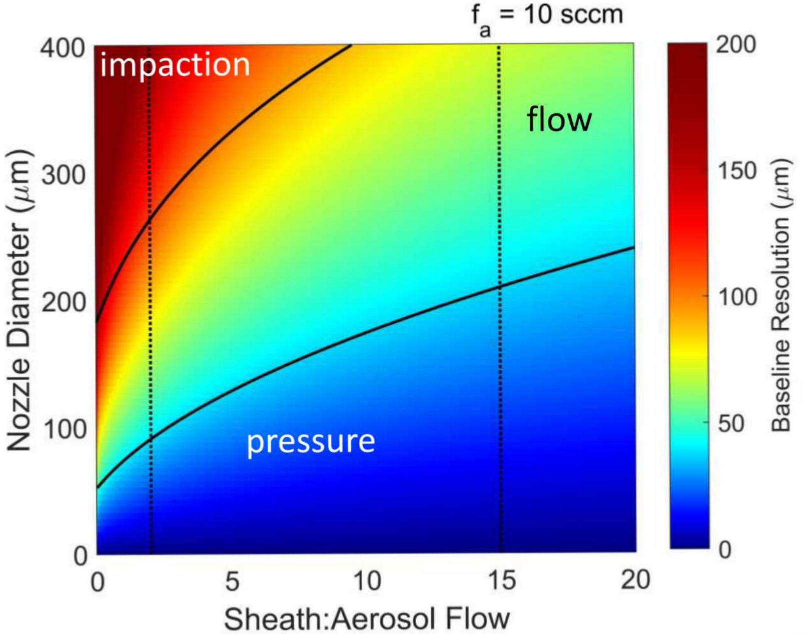


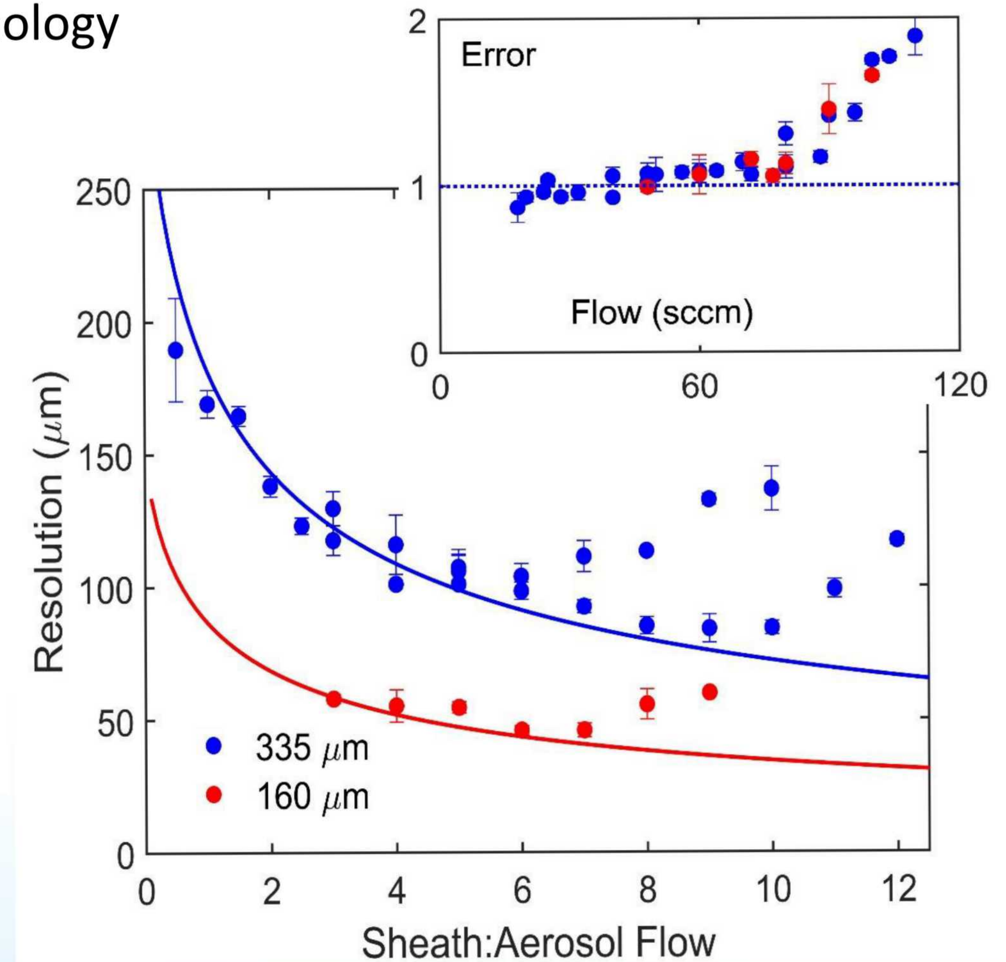
$$r_0^* * \exp[-p] * \left\{ \cosh(pq) + \frac{1}{q}\sinh(pq) \right\}$$

Secor, E.B. *Flex. Print. Electron.* (**2018**) 3, 035002.

Fundamentals of Aerosol Jet Printing

Models based on aerosol physics can aid in process design and help improve the technology

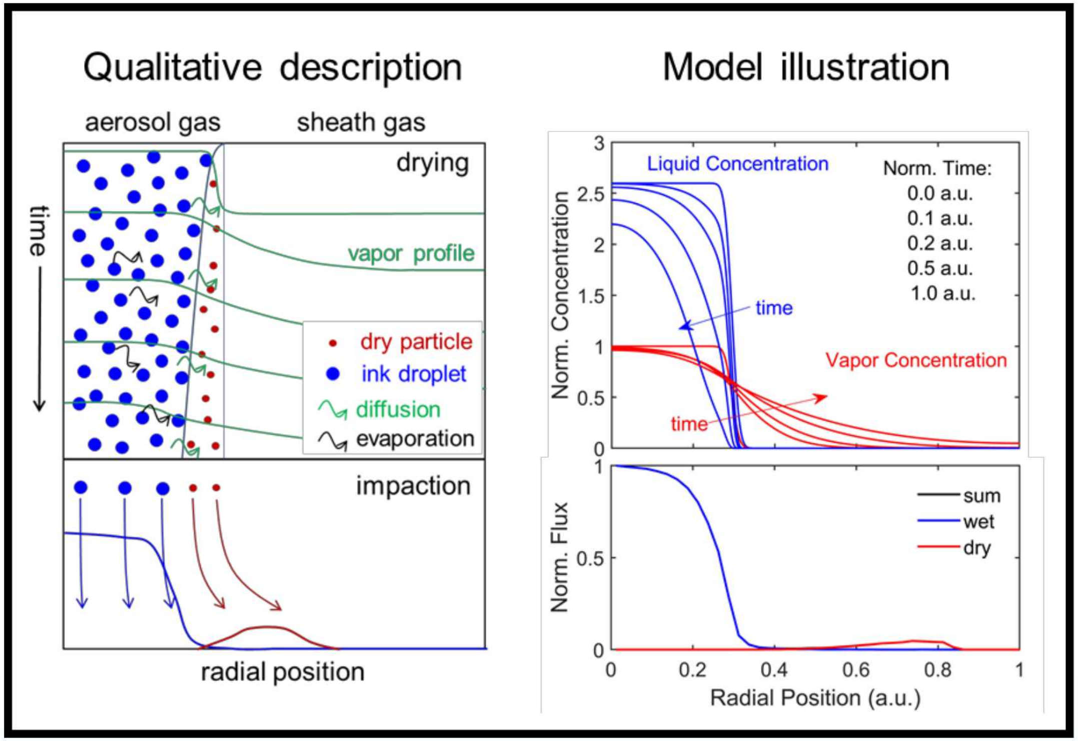


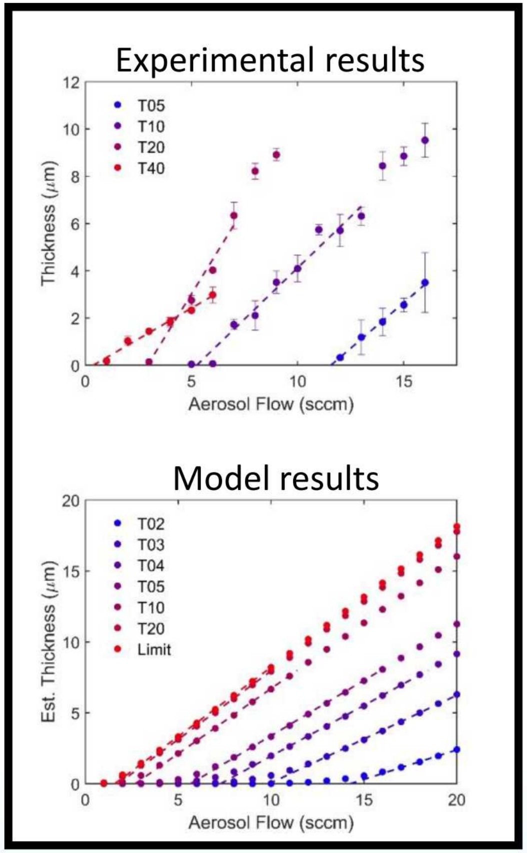


Allows up front description of factors limiting print quality

Fundamentals of Aerosol Jet Printing

Rapid drying within the printhead can have a significant effect on observed performance





$$c_0 = \frac{v_{aero} v_2 V_m}{n_{sat}} * \frac{1}{1+\chi}$$

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

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

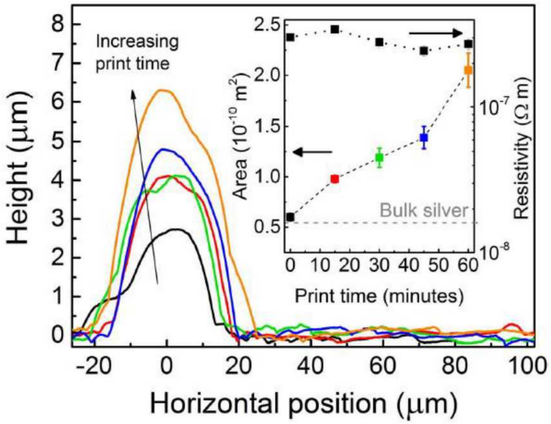
$$\tau_{evap} = \frac{a_0^2}{3 D v_a v_2^{\frac{1}{3}}}$$

Models streamline ink formulation and provide insight for process optimization

Secor, E.B. *Flex. Print. Electron.* (**2018**) 3, 035007.

Fundamentals of Aerosol Jet Printing

Drift in deposition rate a major challenge for aerosol jet printing in general



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

$$\frac{dv_2}{dt} = \frac{f_a v_a}{V_t} v_1 v_2 \left[\frac{1 - c_{pre,1}}{c_{core,1}} - \frac{1 - c_{pre,2}}{c_{core,2}}\right]$$

$$\frac{dc_s}{dt} = \frac{f_a v_a c_s}{V_t}\left[v_1\left(\frac{1 - c_{pre,1}}{c_{core,1}}\right) + v_2\left(\frac{1 - c_{pre,2}}{c_{core,2}}\right)\right]$$

$$\frac{dV_t}{dt} = -f_a\left[v_a + (v_a v_1)\left(\frac{1 - c_{pre,1}}{c_{core,1}}\right) + (v_a v_2)\left(\frac{1 - c_{pre,2}}{c_{core,2}}\right)\right]$$

$$where\ c_{core,i} = \frac{v_a v_i \rho_i\, RT}{m_{w,i}\, p_{sat,i}}\ is\ the\ core\ saturation,$$

the relative solvent vapor concentration

Two principle causes to consider

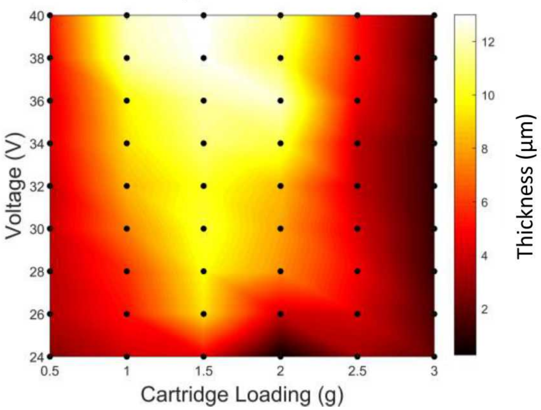
- Ink composition change

Change in cosolvent fractions

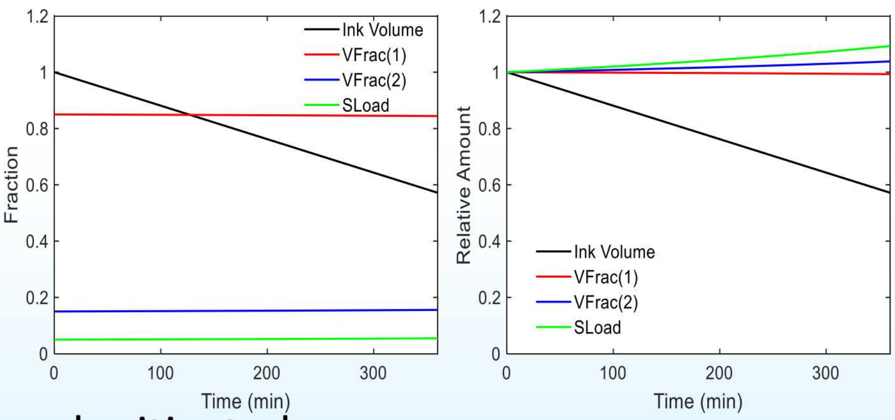
Change in solute concentration

- Ink level change

Change in atomization



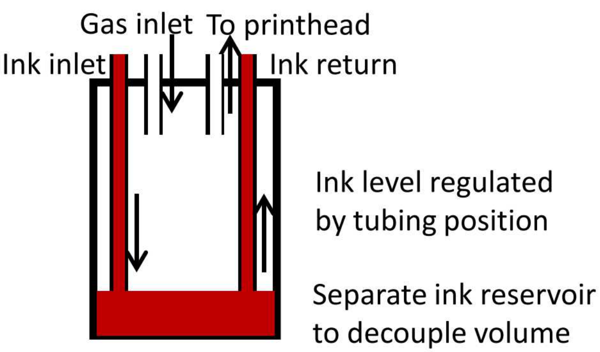
estimated time evolution

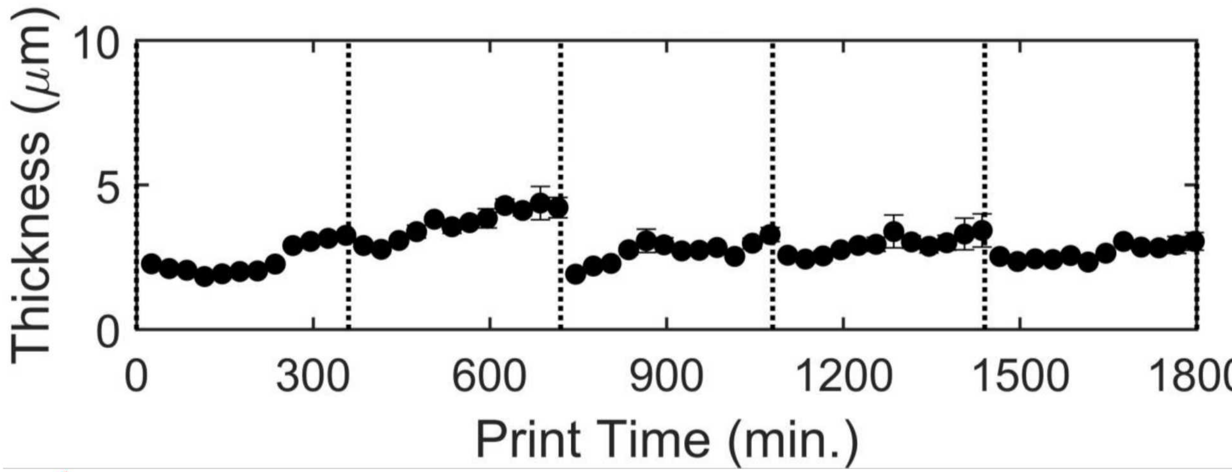


Several causes of drift can be understood and mitigated

Fundamentals of Aerosol Jet Printing

Custom system design provides flexibility to address causes of printing drift

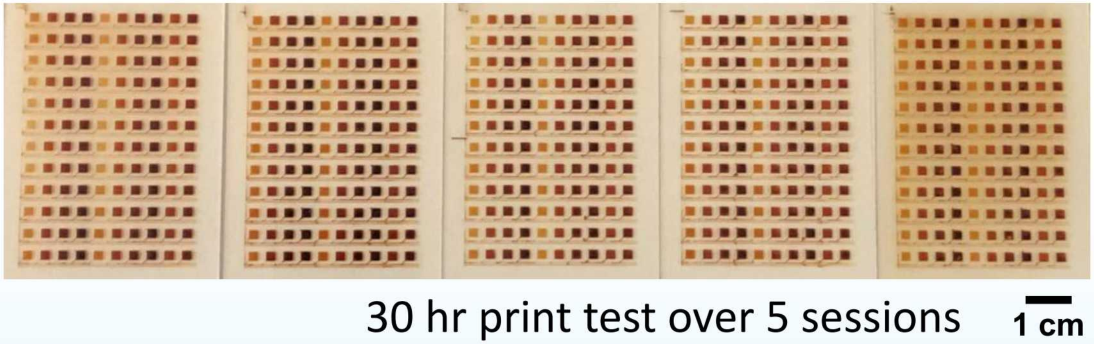




$$\frac{dv_2}{dt} = \frac{f_a v_a}{V_t} v_1 v_2 \left[\frac{1 - c_{pre,1}}{c_{core,1}} - \frac{1 - c_{pre,2}}{c_{core,2}}\right]$$

$$\frac{dc_s}{dt} = \frac{f_a v_a c_s}{V_t} \left[v_1 \left(\frac{1 - c_{pre,1}}{c_{core,1}}\right) + v_2 \left(\frac{1 - c_{pre,2}}{c_{core,2}}\right)\right]$$

$$\frac{dV_t}{dt} = -f_a \left[v_a + (v_a v_1) \left(\frac{1 - c_{pre,1}}{c_{core,1}}\right) + (v_a v_2) \left(\frac{1 - c_{pre,2}}{c_{core,2}}\right)\right]$$

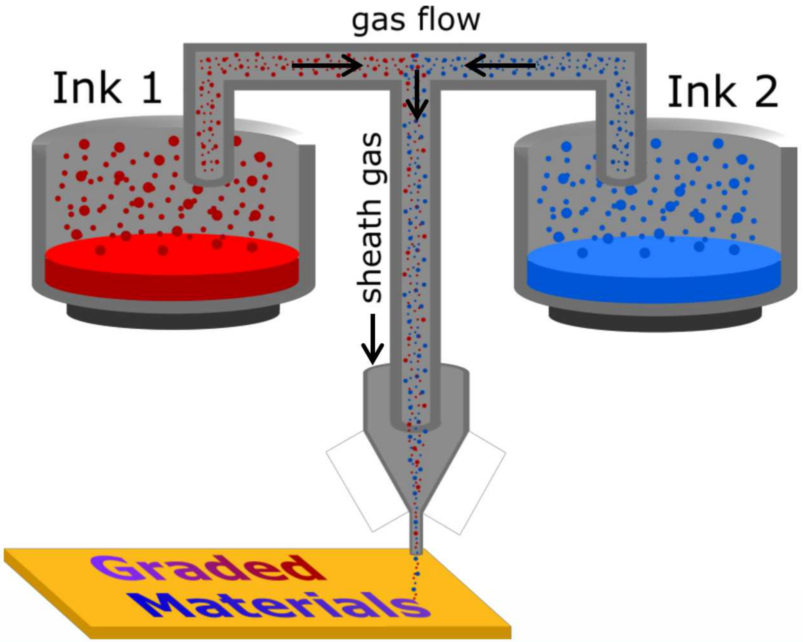


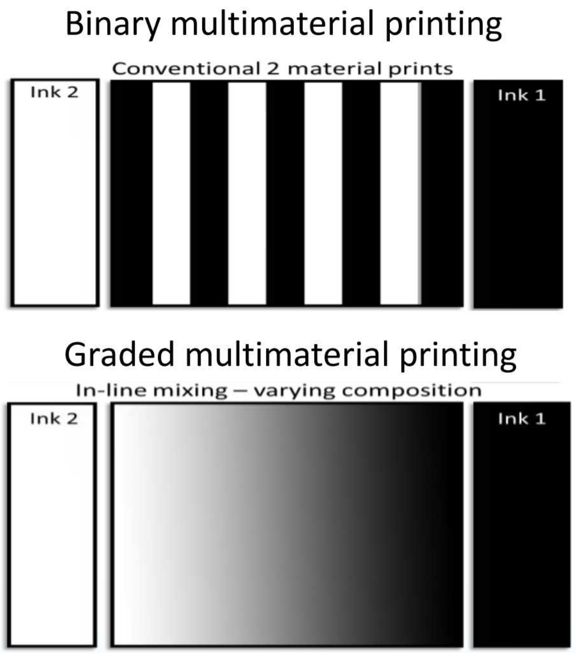
30 hr print test over 5 sessions demonstrates long-duration printing

Recirculating ink cartridge shows promise for extending print duration

Graded Multimaterial Printing

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

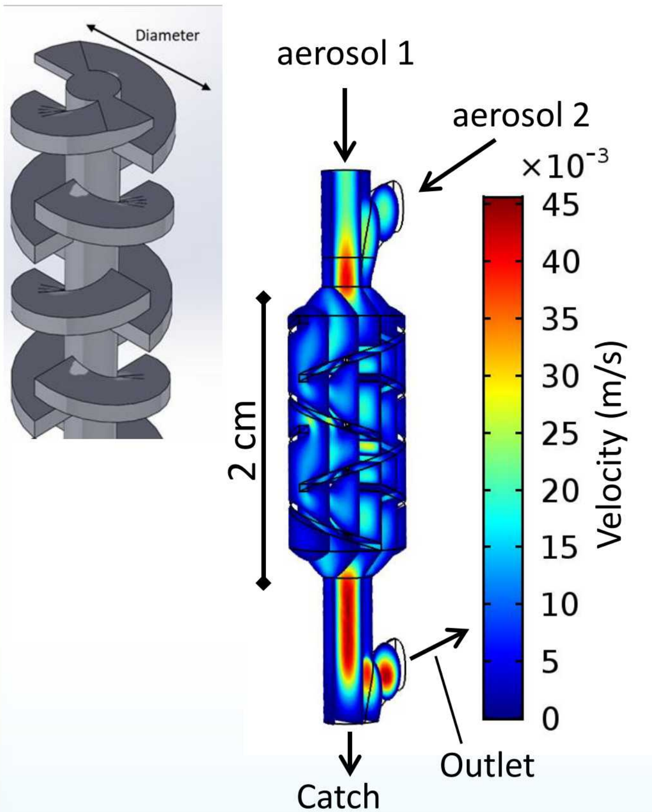


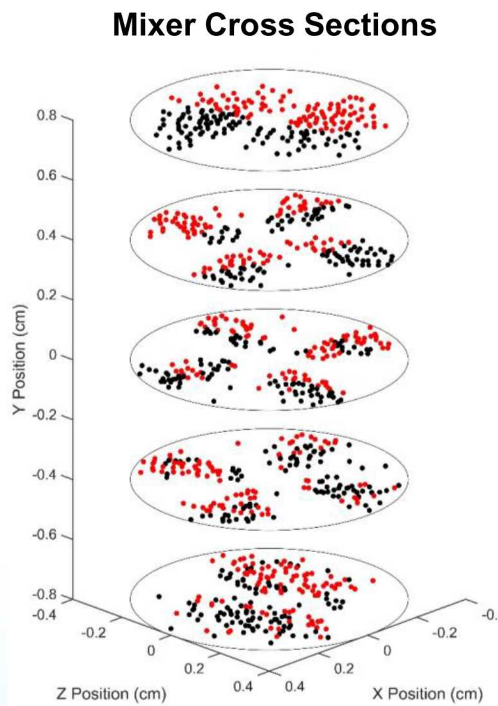


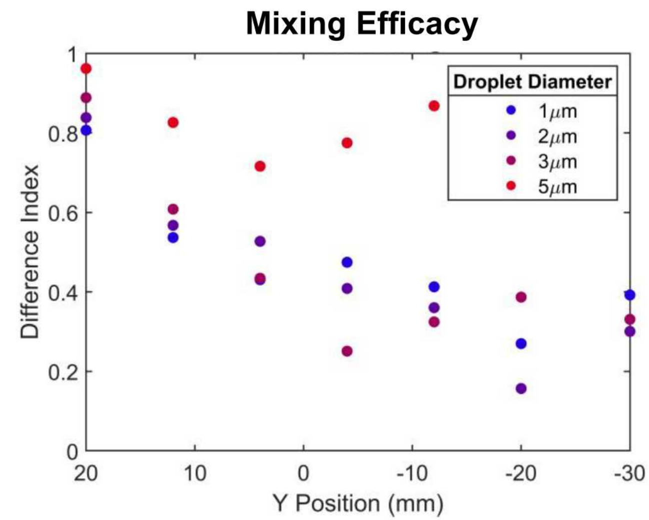
Unique multimaterial printing platform for graded patterning with micron-scale mixing

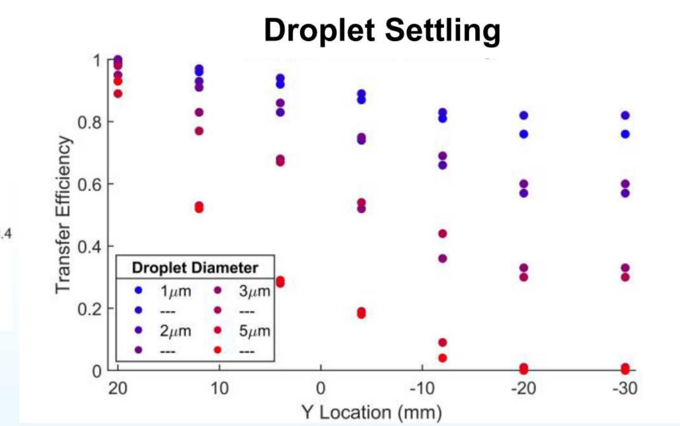
Graded Multimaterial Printing

Laminar static mixers modeled for aerosol applications using COMSOL to design custom mixing printhead









A laminar static mixer provides effective mixing of micron-scale aerosol droplets

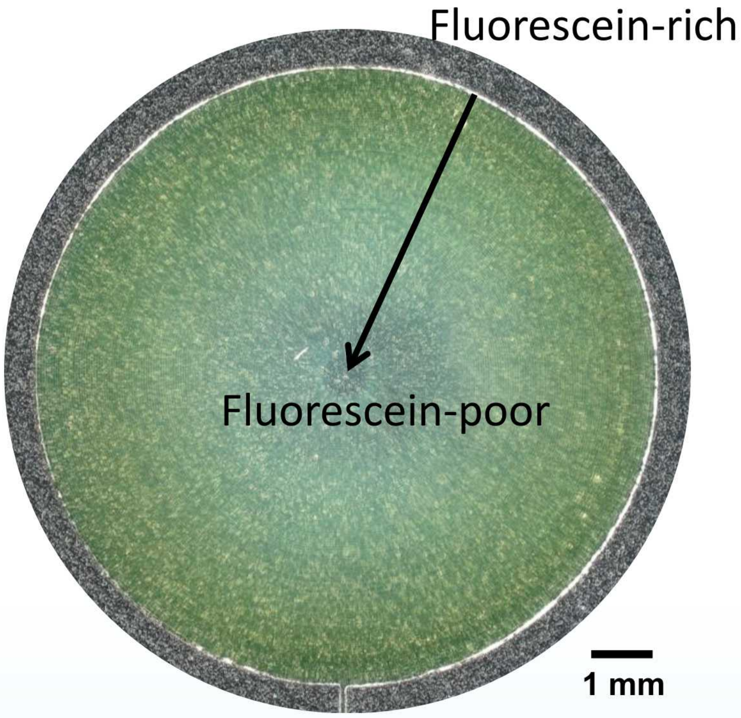
Graded Multimaterial Printing

Initial demonstration focused on optical materials for straightforward quantitative characterization

Ink 1: Norland optical adhesive NOA 61

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

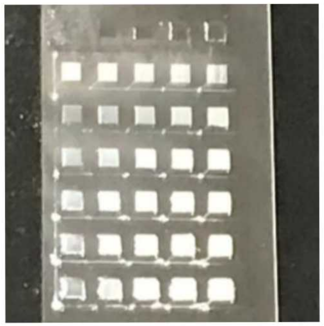
- Fluorescence microscopy measurement to detect fluorescein-containing ink
- Thickness measurement for overall deposition rate
- Calibration required to deconvolute thickness and composition

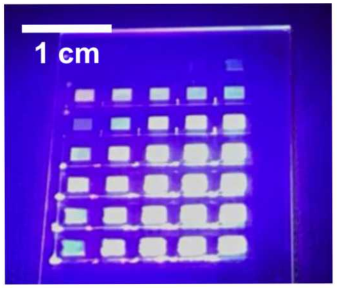


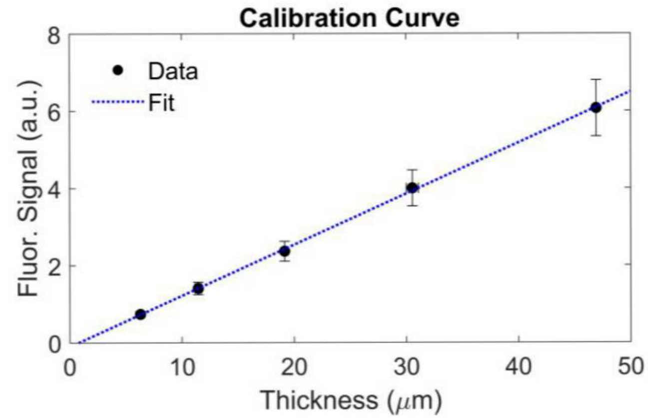
A fluorescent tracer in an optical matrix provides a useful proof-of-concept experiment

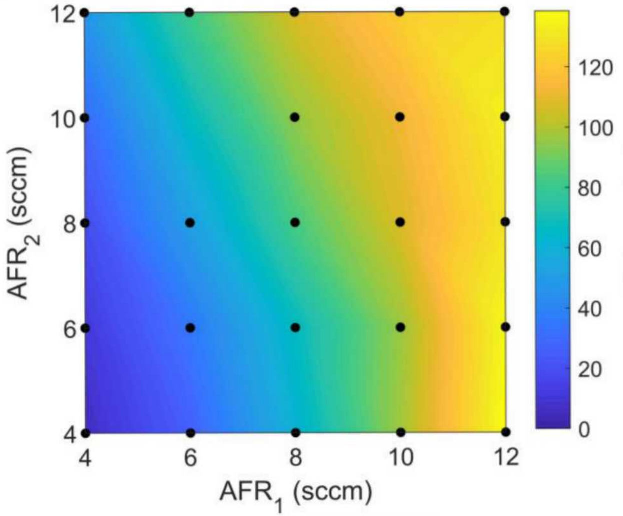
Graded Multimaterial Printing

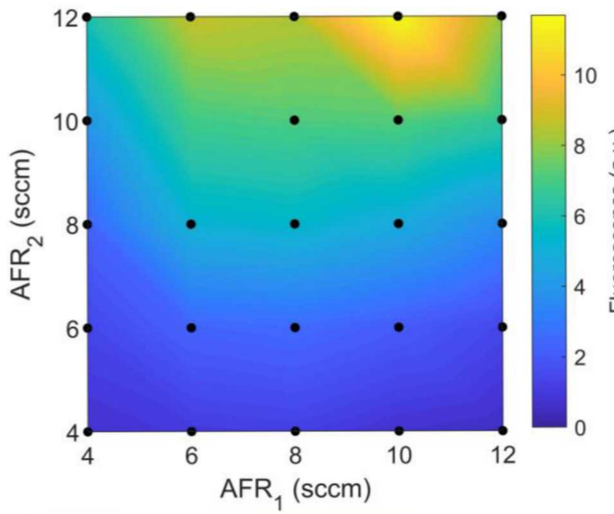
Characterization/calibration

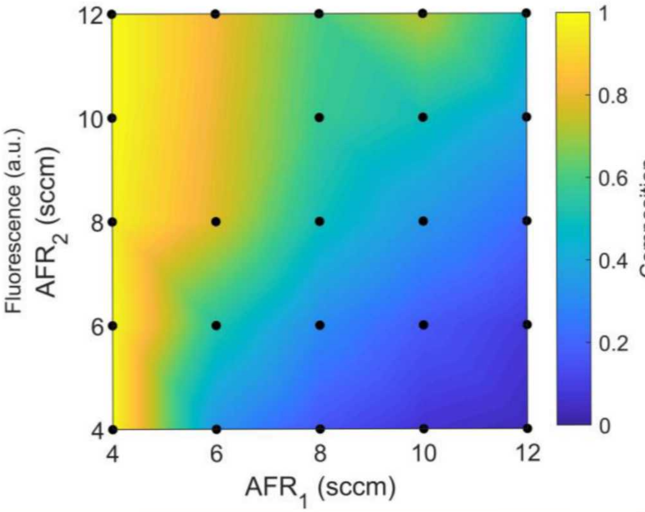












Thickness and fluorescence data allow composition to be estimated for various deposition conditions

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NNSA National Nuclear Security Administration
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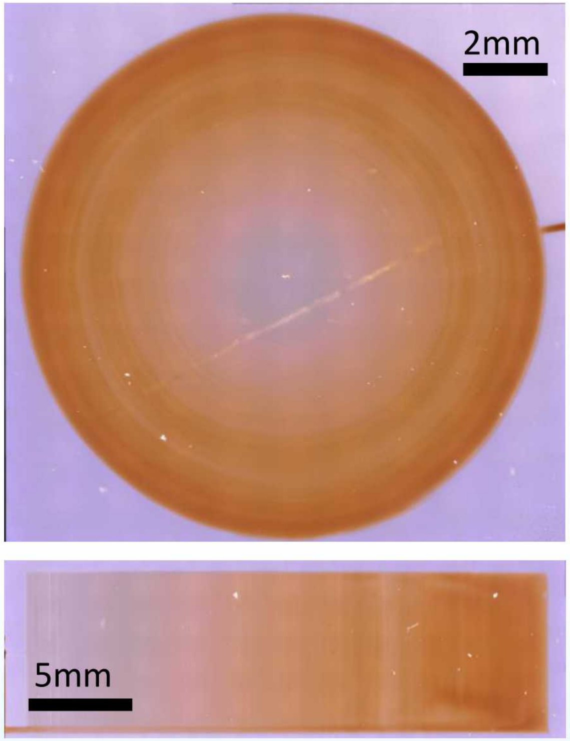
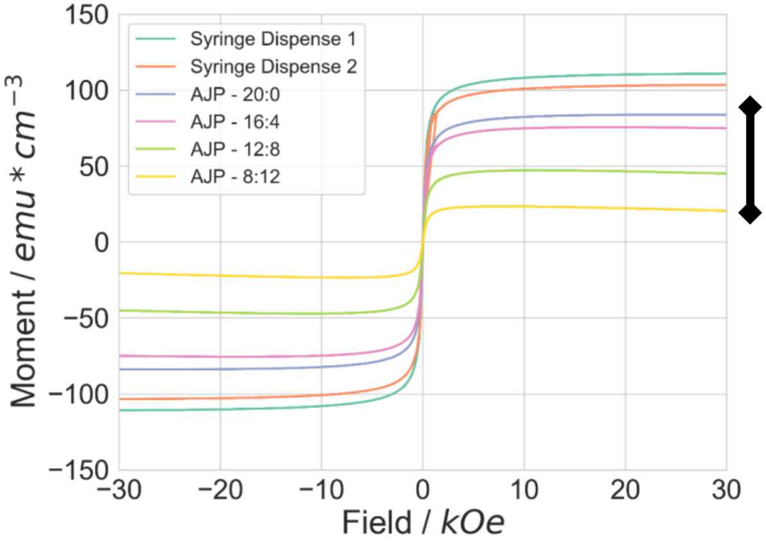
Graded Multimaterial Printing

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

Magnetite composite





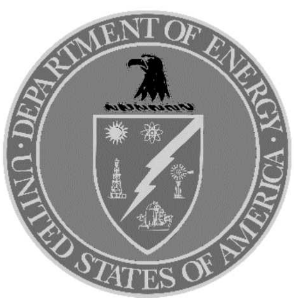
Changing composition of magnetite allows tuning of magnetic properties

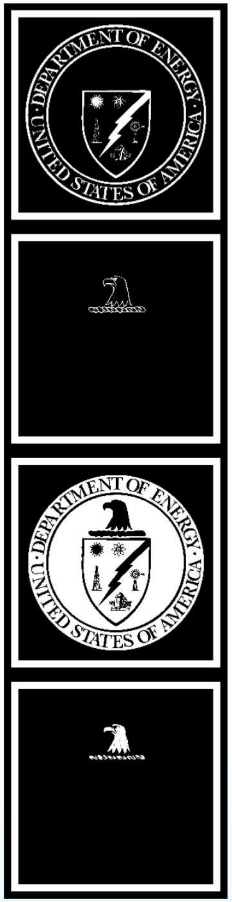
Sample (Fe_3O_4: PMMA)	Fe_3O_4 Loading (mg/cm^2)	St. Dev. (mg/cm^2)
AJP – 20:0	2.39	0.16
AJP – 16:4	1.55	0.08
AJP – 12:8	0.64	0.03
AJP – 8:12	0.35	0.02

Thanks to Dr. Lok-kun Tsui (UNM) for magnetic measurements

Graded Multimaterial Printing

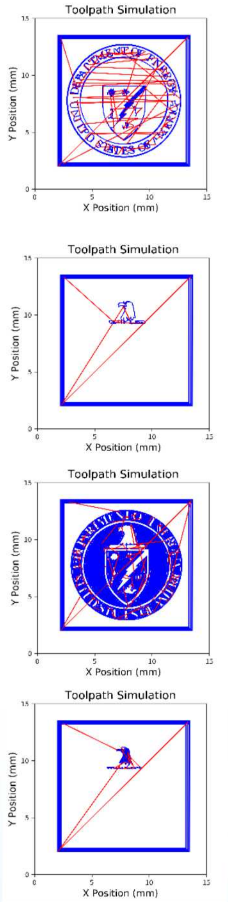
New fabrication capability requires corresponding software for print file generation

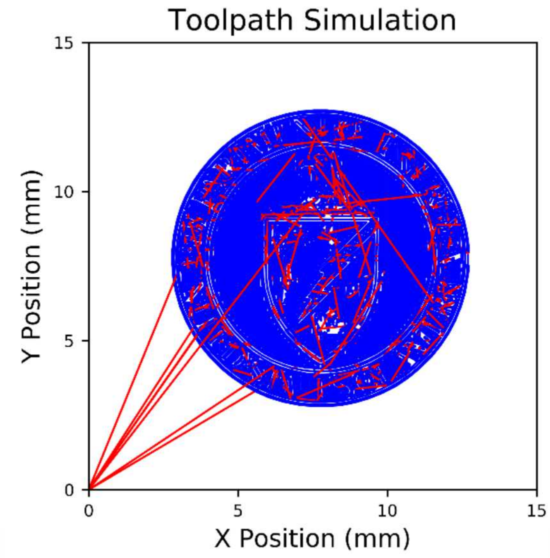




PNG to DXF
DXF to STL

STL to G-Code
G-Code to FGC





Custom application processes grayscale images for complex toolpath generation

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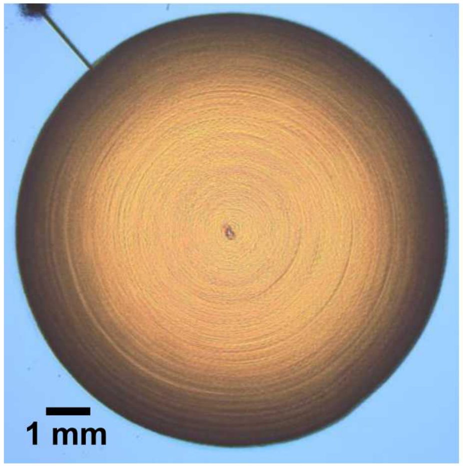
Graded Multimaterial Printing

Widespread opportunities in research and applications for advanced fabrication of graded materials

Graphene/magnetite composite



Theoretical understanding of ink design accelerates development of new inks

$$c_0 = \frac{v_{aero} v_2 V_m}{n_{sat}} * \frac{1}{1+\chi}$$

Broad scope of compatible materials

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

Mixing only upon deposition relaxes requirements for miscibility of precursors

Graded Multimaterial Printing

Widespread opportunities in research and applications for advanced fabrication of graded materials

EM wave manipulation

- Gradient index of refraction lenses and waveguides for optical applications
- GRIN materials for RF communications and sensing with graded dielectric and magnetic properties

Active electronics

- Graded semiconductors for, ie, diodes and photovoltaics
- Graded thermoelectric materials

Catalysis and Electrochemistry

- Tailor porosity/composition in mixed ion/electron conducting systems
- Engineer electronic band structure for catalysis and sensing

Passive electronics

- Reduce electric and magnetic field gradients at interfaces of magnets and capacitors
- Reduce eddy currents in magnetic materials

Mechanical components

- Thermal expansion at diffuse interfaces
- Graded modulus to reduce stress concentrations at interfaces
- Graded adhesives for bonding dissimilar materials

Multimaterial AJP offers a simple and efficient experimental platform to test a wide range of graded material concepts

Rebecca R. Tafoya

Thanks for your attention!

Bryan J. Kaehr

Adam W. Cook

If interested in more details, check out the poster presentation by Rebecca Tafoya later today

P. Randall Schunk

Contact: ebsecor@sandia.gov

Thanks to Rebecca R. Tafoya, Adam W. Cook, Bryan J. Kaehr, and P. Randall Schunk

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