SANDIA REPORT

SAND2014-17877
Unlimited Release
Printed September 2014

Selective Evaporation of Focusing Fluid in Two-Fluid Hydrodynamic Print Head

David M. Keicher and Adam W. Cook
Advanced Materials Laboratory Department (1815), Sandia National Laboratories

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico 87185 and Livermore, California 94550

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation,
a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's
National Nuclear Security Administration under contract DE-AC04-94AL85000.

Sandia National Laboratories

Issued by Sandia National Laboratories, operated for the United States Department of Energy by Sandia Corporation.

Printed in the United States of America. This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from
U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831

Telephone:	(865) 576-8401
Facsimile:	(865) 576-5728
E-Mail:	reports@adonis.osti.gov
Online ordering:	http://www.osti.gov/bridge

Available to the public from
U.S. Department of Commerce
National Technical Information Service
5285 Port Royal Rd.
Springfield, VA 22161

Telephone:	(800) 553-6847
Facsimile:	(703) 605-6900
E-Mail:	orders@ntis.fedworld.gov
Online order:	http://www.ntis.gov/help/ordermethods.asp?loc=7-4-0#online

SAND2014-17877
Unlimited Release
Printed September 2014

Selective Evaporation of Focusing Fluid in Two-Fluid Hydrodynamic Print Head

David M. Keicher and Adam W. Cook
Advanced Materials Laboratory Department (1815
Sandia National Laboratories
P.O. Box 5800
Albuquerque, New Mexico 87185-MS1349

Abstract

The work performed in this project has demonstrated the feasibility to use hydrodynamic focusing of two fluid steams to create a novel micro printing technology for electronics and other high performance applications. Initial efforts focused solely on selective evaporation of the sheath fluid from print stream provided insight in developing a unique print head geometry allowing excess sheath fluid to be separated from the print flow stream for recycling/reuse. Fluid flow models suggest that >81% of the sheath fluid can be removed without affecting the print stream. Further development and optimization is required to demonstrate this capability in operation. Print results using two-fluid hydrodynamic focusing yielded a 30 µm wide by 0.5 µm tall line that suggests that the cross-section of the printed feature from the print head was approximately 2 µm in diameter. Printing results also demonstrated that complete removal of the sheath fluid is not necessary for all material systems. The two-fluid printing technology could enable printing of insulated conductors and clad optical interconnects. Further development of this concept should be pursued.

Acknowledgments

The authors would like to acknowledge Dr. Marcelino Essien at Castor Technologies for his assistance in completing this work.

CONTENTS

This page intentionally left blank

1. INTRODUCTION

Printed electronics is an emerging area for additive manufacturing. The appeal to directly print electronic features is driven largely by the advantages offered to reduce process steps, capital equipment costs and the need for product specific tooling. By addressing these points, Direct Write (DW) technologies offer the ability to produce low volume, high mix electronics at a per piece cost normally associated with large production volumes where economy of scale aids reducing part cost. There is continuous focus on providing the ability to print finer and finer features using the DW technologies. One leading technology in the DW area is Aerosol Jet (AJ) printing technology where aerodynamic focusing is used to print features down to approximately 10 µm. Although the AJ technology has been demonstrated in a laboratory environment, several technical issues persist that limit the use of this technology for commercial applications. These issues include:

- Shuttering – The mechanical shutter used for AJ printing limits its usefulness for conformal printing and has a tendency to eject excess ink collected in the shutter during printing onto the print surface. Shutter response time (approx. 10 ms) limits high speed processing capabilities.

- Overspray – Both ultrasonic and pneumatic atomization (UA and PA, respectively) methods used for the AJ technology produce a poly dispersed aerosol. Aerodynamic focusing of poly dispersed aerosols leads to overspray on the edges of printed features since smaller droplets focus at a different plane than the larger droplets (an optical analogy is chromatic aberration).

- Process Reliability – UA and PA generate aerosol with a broad droplet size distribution and the AJ process only uses the droplets at the small end of the distribution curve. Aerosol usage rate is less than 0.001%. As such, excess energy input to the ink changes ink characteristics over time and affects the output of the printer. Low volatility solvent based inks can extend print time over high volatility solvent based inks; however, in both cases total ink utilization is less than 10% before the ink is degraded. Note that there are many good inks that use volatile solvents. To offset this effect, solvent add back has recently added to all commercial AJ systems. This requires tight temperature control and makes their system more complex.

- Aerosol Transport – The physical sizes of the UAs and PAs necessitate mounting the atomizer some distance from the print head. Aerosol transport distance to the print head allows settling of the droplets causing clogging or pressure pulses in the transport lines. These pressure pulses affect the print quality.

- Multiplexing Print Nozzles – AJ has had some success developing multinozzle print heads but, in addition to the above issues, individual nozzle shuttering and uniform aerosol distribution present additional challenges in scaling the AJ technology.

- Material Output Rate – Aerosol theory shows that diffusion limits the number of droplets that can exist within a given space at any given time and that the number of droplets that can exist is independent of droplet size for the droplet range of interest to AJ printing. This limits the maximum output rate for AJ technology using current UA and PA technology.

Many of the existing issues associated with AJ technology may be overcome with the use of a liquid jet printing approach that can be created using hydrodynamic focusing. Hydrodynamic focusing offers the potential to print electronic features with micron/submicron resolution. Hydrodynamic focusing of fluids occurs in microchannels when multiple fluids flow into the same channel and the flow rate of the focusing fluid is increased with respect to the second focused fluid. This affect is shown graphically in Figure 1. Increasing the flow rate of the focusing fluid decreases the cross-sectional area of the focused fluid causing focusing. Researchers in microfluidics have shown that wires as small as 400 nm in diameter can be produced using hydrodynamic focusing in microfluidic chambers. This phenomenon can be exploited to develop a novel printing technique which uses hydrodynamic focusing to print micron and submicron features. If the focusing fluid can be removed from the fluid stream prior to impacting onto the print substrate, the focused fluid can be deposited to create the desired features. The focusing effect can be further enhanced by flowing the two fluid streams through a converging nozzle.

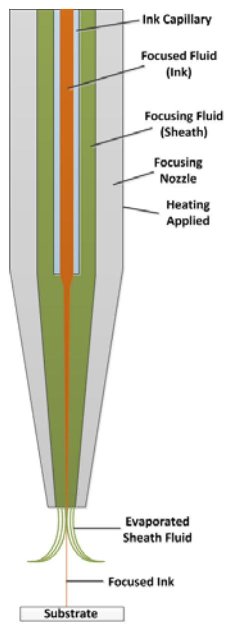


Figure 1. **Schematic representation showing hydrodynamic focusing in coaxial tube assembly.**

The liquid jet has several advantages over its AJ counterpart. These advantages are:

1. Liquid jets are highly stable providing uniform deposition.
2. Fluid output rate can be very high to allow small features to be printed quickly.
3. Liquids are incompressible allowing shuttering to occur in the fluid stream.
4. Microvalves are available to enable shuttering at greater than 1 kHZ enabling high speed printing.
5. Liquid streams have very good edge definition enabling RF application.
6. Liquid jets can be highly focused to produce very fine features.
7. Printed fluid viscosity can have a wide range.
8. Printing of coaxial features can be enabled using two fluid flow.

The objective of this project was to demonstrate that the sheath fluid used for focusing the ink can be selectively vaporized with minimum impact to the central core fluid to enable printing of micron/submicron features. During the early phases of this effort, the objective shifted to focus more broadly on the removal of the sheath fluid which led to a unique solution for a print head geometry as described below.

This page intentionally left blank

2. EXPERIMENTAL/MODELING

2.1 Experimental

The study investigating sheath fluid evaporation was performed using the apparatus shown in Figure 2. The geometric parameters for the critical flow cell features are given in Table 1. An initial study focused on evaporation of a single-component jet consisting of the sheath fluid. The pressure in the apparatus was increased to 69 kPa above the local atmospheric pressure until jetting of the sheath fluid was observed. The temperature of the system was then increased until jet evaporation began. The study used 91% isopropanol (IPA) in water as the sheath fluid.

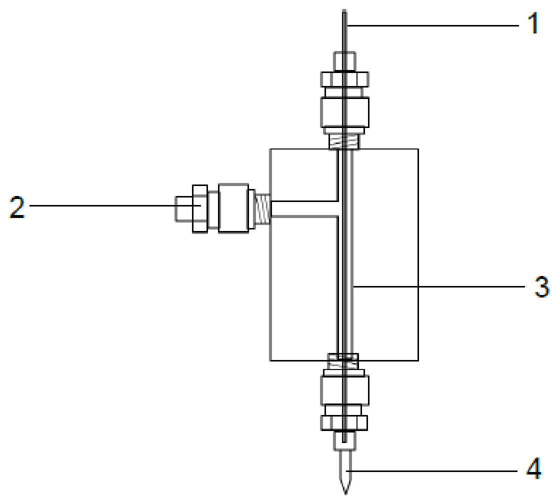
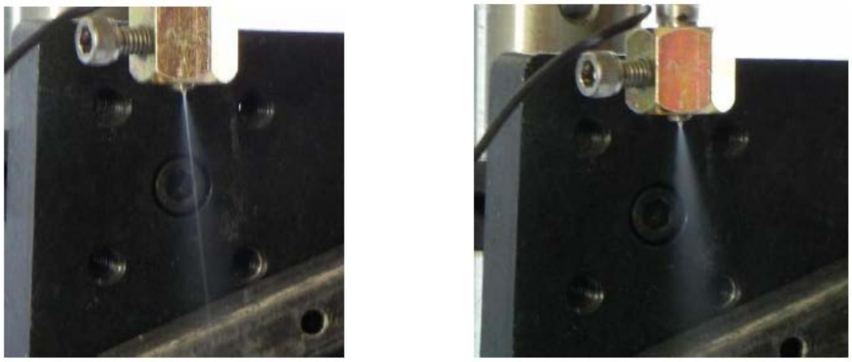


Figure 2. Schematic of the Liquid Jet apparatus for developing coaxial sheath and ink flows.

Table 1.

Item	Description
1	Ink input capillary (760 µm)
2	Sheath gas input
3	Main flow channel
4	Output nozzle (40 µm)

Figure 3 shows a real-time image of the sheath fluid evaporation from the liquid jet apparatus. Figure 3a shows a partially shattered jet, in which the outer layer of the liquid jet is evaporated, leaving a liquid core. Figure 3b shows a completely evaporated jet. In both cases the flow from the jet is sporadic, askew from the normal, downward fluid flow direction.



(a) **(b)**

Figure 3. Images of sheath fluid evaporation for (a) partially evaporated liquid stream and (b) fully evaporated fluid stream.

2.2 Modeling

In investigating the feasibility of using superheating to selectively evaporate the sheath fluid, an alternative print head design was developed that would enhance the ability to vaporize the sheath fluid. This head geometry is shown in Figure 4. Note that although this is a cross-sectional view, quarter symmetry was used in modeling fluid flows and so only the top half of the channels are shown in color. In this geometry, several features were added to the flow cell design to enhance the focusability of the ink and to significantly reduce the total volume of sheath fluid that is expended during the printing process. For this geometry, the sheath channel diameter transitions to a smaller diameter in the focusing section to maintain laminar flow. After focusing occurs, the channel transitions back to a larger diameter to allow the sheath fluid to be removed from the print flow stream prior to exiting the exit orifice. As such, only a fraction of the sheath fluid must be dealt with as part of the printing process.

Fluid flow modeling was performed on the head design shown in Figure 5 using the Fluid Analysis plug in for SolidWorks. Since the model is cylindrically symmetrical, quarter symmetry of the model was used to reduce the calculation time in the simulations. Model conditions are given in Table 2.

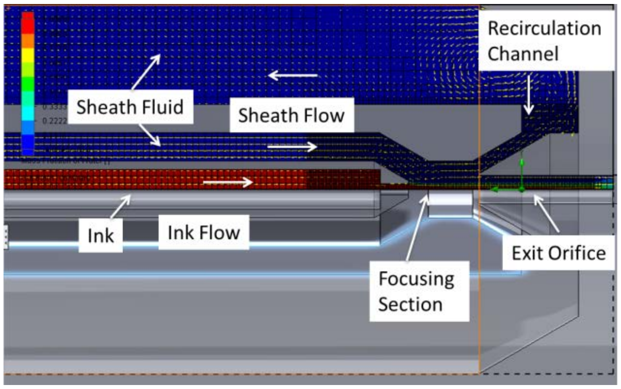


Figure 4. Cross-sectional view of improved flow cell (print head) geometry to minimize sheath vaporization requirements.

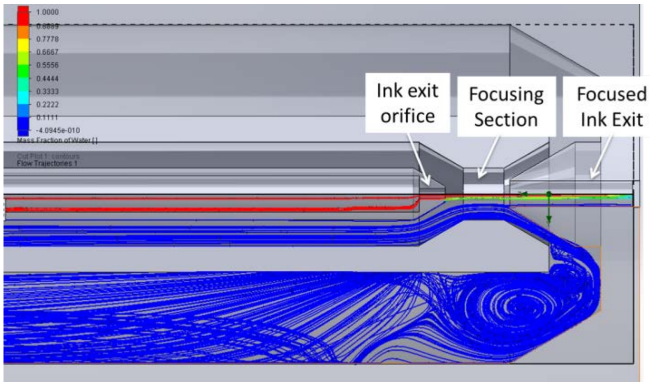


Figure 5. Cross-sectional view of print head geometry showing fluid flow within the head. Note that the ink is shown in red and the sheath fluid is shown in blue.

Table 2.

Item	Description
Ink exit orifice diameter	100µm
Focusing section diameter	500µm
Focused ink exit diameter	250µm
Ink flow rate	$1x10^{-9}m^3/s$
Sheath flow rate	$5x10^{-8}m^3/s$
Ink composition	Water
Sheath composition	Ethanol

2.3 Printing

A study was also performed to investigate liquid jet deposition using a coaxial jet consisting of an evaporating sheath and a core ink fluid. The print head used in this printing work was shown previously in Figure 2. The sheath fluid was a solution of isopropanol in distilled water as described previously. The ink used was a 7% by volume solution of polyvinylpyrrolidone (PVP) in distilled water with green dye. Figure 6 shows a PVP traces deposited on glass using a coaxial liquid jet. The trace width is approximately 30 µm wide and approximately 0.5 µm thick. The trace was printed at 100 mm/s.

This page intentionally left blank

3. RESULTS/DISCUSSION

The stated objective of this project was to demonstrate that the sheath fluid used for focusing the ink could be selectively vaporized with minimum impact to the central core fluid to enable printing of micron/submicron features. Results from experiments involving evaporation of only sheath fluid as it passed through a deposition head demonstrated that this concept is feasible; however, it also demonstrated the difficulty associated with this approach. The images shown in Figure 3 show that in the case of a partially evaporated jet and a fully evaporated jet, the mist or jet is ejected from the head at an angle that is skewed from the normally downward jet direction. This deviation is the result of trapped gas bubbles within the print head. Gas bubbles in the print head were produced when the heating caused vaporization of the sheath fluid in the head before the fluid exited the head. Additional heating of the head resulted in complete stoppage of the printing process where no fluid flow occurred.

These early investigations of superheating the sheath fluid alone lead to the conclusion this approach would not likely yield the desired result. The basis for this conclusion is as follows: Experimental results showed that in order to achieve full vaporization of the sheath fluid from the print head, the head needed to be heated to the point where vaporization was occurring inside of the head which significantly affected the ability to print. If one considers the pressure versus boiling point curve shown in Figure 7, increasing superheating would require the internal head pressure to be increased. Increasing the fluid flow rate in the head would increase the pressure but would also result in the need for more power to vaporize the increased fluid flow. Decreasing the exit orifice from the print head would also increase the internal head pressure which would affect the ability to further superheat the sheath fluid. However, the exit orifice diameter was already 40µm and so reducing the orifice diameter really wasn't preferred.

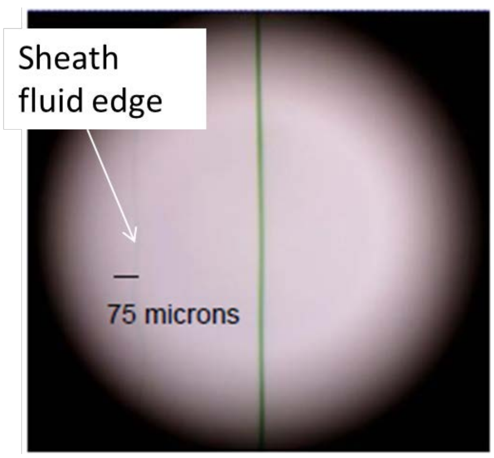


Figure 6. PVP ink deposited on glass using a coaxial liquid jet.

Since the unstated yet underlying objective to this project was to remove the sheath fluid from the print stream another option was considered. This approach was to redesign the print head to physically remove the sheath fluid from the print stream before it was allowed to exit the print head. The model for a conceptual design was developed and analyzed using fluid flow analysis software (see Figure 4). Analytical results from the fluid flow analysis of the conceptual print head geometry demonstrated that the sheath fluid could in fact be removed from the print flow stream before it is allowed to exit the print head. As shown Figure 5, the flow fields are fully laminar in the areas of interest and the portion of the sheath fluid that is stripped from the print stream is not mixed with the ink and so it can be recycled and reused for subsequent focusing operations. The total fluid flow rate (sheath + ink) into the print head was 3.06 sccm and the fluid flow rate out the head print nozzle was 0.58 sccm. The percentage of sheath fluid that is recycled is approximately 81%. Additionally, the print orifice was 250 µm which if preferred to minimize clogging and other reliability issues.

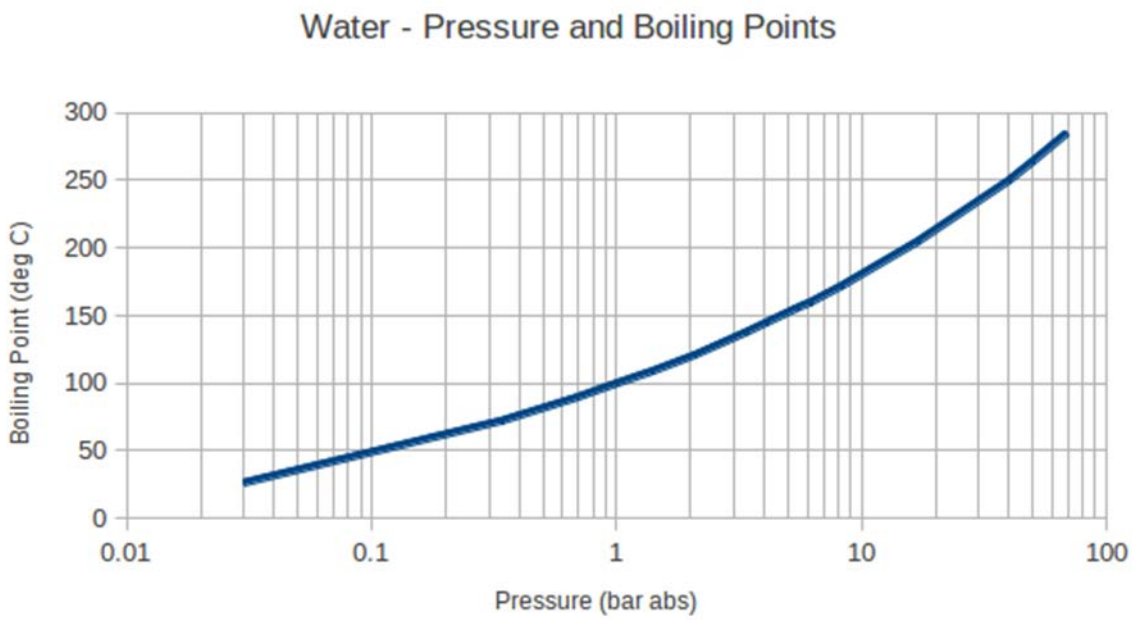


Figure 7. **Pressure versus boiling point curve for water (Ref: "The Engineering ToolBox; www.EngineeringToolBox.com")**

Finally, to understand the need to fully removed the sheath fluid from the print stream, printing tests were performed using the prototype print head shown in Figure 2. A representative printed trace is shown in Figure 6. The deposition of PVP shows good substrate wetting and good edge definition. Profiles of liquid jet deposited PVP traces were measured using a contact profilometer. A representative profile is shown in Figure 8. The profile shows a trace height of approximately 0.5 microns. The trace was deposited at a speed of approximately 100 mm/s. The very wide and flat profile of the trace suggests that significant spreading of the trace occurred upon contacting the print substrate. This is related to surface tension effects. Since the cross-section of the ink trace is approximately 15 μm^2, then, assuming a circular cross-section upon exiting the print head, an estimate of the ink filament diameter from the print nozzle approximately 2 µm. This demonstrates the feasibility of using hydrodynamic focusing to create a fine filament of the core fluid. Successful printing without fully eliminating the sheath fluid also provides support that technology could be used to create a two or more material structure. Examples could include core and clad optical interconnects and insulated conductive traces. Subsequent post processing could also be used to remove the remaining sheath fluid from the print surface as needed.

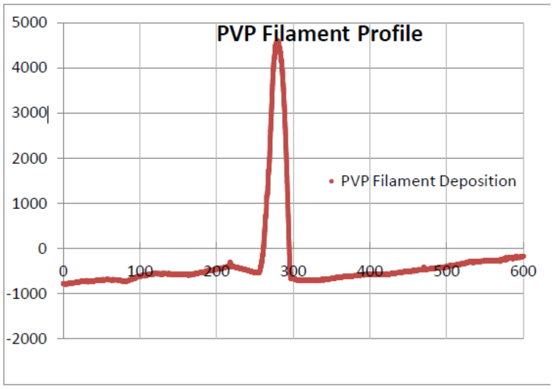


Figure 8. **Profile for printed PVP line shown in Figure 6.**

This page intentionally left blank

4. CONCLUSIONS

Early experimental efforts focused on superheating and evaporation of the two-fluid print head sheath fluid demonstrated feasibility of the evaporation concept and, more importantly, provided insight to clarify the objective for sheath fluid removal in the development of a two-fluid print head technology. This insight led to identification and development of a design concept to efficiently remove the sheath fluid from the print stream prior to exiting the print head and minimized the need for sheath fluid evaporation. This concept provides a unique solution using hydrodynamic focusing to create a print head technology for micro printing applications. Modeling results suggest the recycling feature within the print head provides a method to remove at least 81% of the sheath fluid from the print stream. Printing results using the two-fluid approach with the sheath fluid present also demonstrated that printing of fine features can occur even if the sheath fluid is not completely removed from the print stream. Printing results suggest that the cross-section of the ink stream diameter upon exit of the print head was approximately 2 µm. Results of this effort support further development of this technology.

This page intentionally left blank

4. DISTRIBUTION

1	MS0161	Legal Technology Transfer Center	11500
1	MS0359	D. Chavez, LDRD Office	01911
1	MS0665	H. Tran	02523
1	MS0678	A. Claudette	02617
1	MS0885	M. Smith	01830
1	MS0958	B. Jared	01832
1	MS0968	S. Mani	05771
1	MS1349	A. Cook	01815
1	MS1349	B. Hammetter	01815
1	MS1349	D. Keicher	01815

1	MS0899	Technical Library	9536 (electronic copy)