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COMPUTATIONAL FLUID DYNAMICS MODELING AND IN SITU PHYSICS-BASED MONITORING OF AEROSOL JET PRINTING TOWARD FUNCTIONAL ASSURANCE OF ADDITIVELY-MANUFACTURED, FLEXIBLE AND HYBRID ELECTRONICS

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COMPUTATIONAL FLUID DYNAMICS MODELING AND IN SITU PHYSICS-BASED MONITORING OF AEROSOL JET PRINTING TOWARD FUNCTIONAL ASSURANCE OF ADDITIVELY-MANUFACTURED, FLEXIBLE AND HYBRID ELECTRONICS

BY

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DISSERTATION

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Abstract

Aerosol jet printing (AJP) – a direct-write, additive manufacturing technique – has emerged as the process of choice particularly for the fabrication of flexible and hybrid electronics. AJP has paved the way for high-resolution device fabrication with high placement accuracy, edge definition, and adhesion. In addition, AJP accommodates a broad range of ink viscosity, and allows for printing on non-planer surfaces. Despite the unique advantages and host of strategic applications, AJP is a highly unstable and complex process, prone to gradual drifts in machine behavior and deposited material. Hence, real-time monitoring and control of AJP process is a burgeoning need. In pursuit of this goal, the objectives of the work are, as follows:

(i) *In situ* image acquisition from the traces/lines of printed electronic devices right after deposition. To realize this objective, the AJP experimental setup was instrumented with a high-resolution charge-coupled device (CCD) camera, mounted on a variable-magnification lens (in addition to the standard imaging system, already installed on the AJ printer).

(ii) *In situ* image processing and quantification of the trace morphology. In this regard, several customized image processing algorithms were devised to quantify/extract various aspects of the trace morphology from online images. In addition, based on the concept of shape-from-shading (SfS), several other algorithms were introduced, allowing for not only reconstruction of the 3D profile of the AJ-printed electronic traces, but also quantification of 3D morphology traits, such as thickness, cross-sectional area, and surface roughness, among others.
(iii) Development of a supervised multiple-input, single-output (MISO) machine learning model – based on sparse representation for classification (SRC) – with the aim to estimate the device functional properties (e.g., resistance) in near real-time with an accuracy of ≥ 90%.

(iv) Forwarding a computational fluid dynamics (CFD) model to explain the underlying aerodynamic phenomena behind aerosol transport and deposition in AJP process, observed experimentally.

Overall, this doctoral dissertation paves the way for: (i) implementation of physics-based real-time monitoring and control of AJP process toward conformal material deposition and device fabrication; and (ii) optimal design of direct-write components, such as nozzles, deposition heads, virtual impactors, atomizers, etc.
To my wife, Dr. Julia Ludewig,
for the love, support, and constant encouragement I have gotten over the years.
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Chapter 1: Introduction

1. Aerosol Jet Printing (AJP)

1.1. Process:

The AJP process is centered on atomization/nebulization of an ink, which can be implemented pneumatically or ultrasonically. Figure 1 illustrates both the modes of atomization. In the pneumatic configuration, a high-pressure flow of an inert gas is injected into the ink reservoir via a head (see Figure 2(c)) where due to the Venturi effect, the ink is drawn upward through the capillary and then sheared by the jet (see Figure 2(c)); as a result, a non-uniform mist/cloud of aerosols is generated. Therefore, an aerodynamic separator, called the virtual impactor (VI), is utilized to filter out droplets with low linear momentum using an exhaust flow and deliver a uniform stream of aerosols for deposition. In the ultrasonic mode of atomization (UA), a transducer is used for direct aerosol/droplet formation. The temperature of the UA process is kept constant using a chilled water system to ensure uniform ink viscosity and mitigate unavoidable process drifts during the deposition of material. Besides, a bubbler filled with the ink solvent(s) is used to deliver a saturated gas flow for atomization and thus compensate for solvent evaporation. The ultrasonically generated aerosols are more uniform (and to some extent finer) than those generated pneumatically; this precludes the need for a separator such as the VI in the UA mode. The uniform stream of aerosols flows toward the deposition head where a surrounding flow, called the sheath gas flow (ShGF), is introduced to collimate/focus the central aerosol flow to a narrow beam, deposited on free surface. The deposited material eventually experiences the post-deposition phenomena of spreading, receding, relaxation, and coalescence [1].
1.2. Pneumatic Atomizer (PA)

Demonstrated in Figure 2, the pneumatic atomizer is composed of the following components: (i) a head with an internal capillary and an orifice, generating a multi-phase jet of aerosols (based on the Venturing effect); (ii) a long stem, guiding the atomization gas flow toward the atomization head; (iii) a jar/reservoir, containing ink; (iv) a cap which ensures airtight operation and allows for ink temperature measurements via insertion of a thermocouple.
Figure 2 – (a) The assembled PA unit; (b) The main components of the pneumatic atomizer; (c) Obtained using X-ray computed tomography (CT) imaging, a cross-section of the atomization head, showing the mechanism behind the formation of a jet of aerosols, based on the Venturi effect.

1.3. Virtual Impactor (VI)

Figure 3 exhibits the main complements of the virtual impactor, including: (i) an impactor, which is basically a converging nozzle; (ii) a collector, which separates aerosols based on their linear momentum as a function of the exhaust gas flow rate (we note that for the central aerosol flow, not diverted toward the exhaust, this component acts as a diffuser, resulting in a decrease in the flow velocity); (iii) a stem, guiding the aerosol flow toward the impactor; and (iv) a retaining nut and housing. The outlet stream of aerosols (more uniform in particle size) flows toward the deposition head via a polymer tube, known as the mist tube. Accumulation of aerosols within the tube (resulting in droplet formation and ultimately nozzle clog) is one of the main causes of anomalies in the AJP process. Consequently, purging/cleaning the mist tube regularly is a need to maintain continuous AJP operation.
1.4. Deposition Head and Nozzle

The deposition head is the last aerodynamic element in the AJP process, aiding in collimated deposition of aerosols. As demonstrated in Figure 4, it consists of: (i) an upper plenum chamber (UPC); (ii) a lower plenum chamber (LPC); (iii) a combination chamber (CC); (iv) a long converging nozzle/micro-capillary; and (v) a shell together with a retaining cap. The UPC and LPC guide the sheath gas flow toward the CC where it concentrically surrounds the aerosol flow. The collimated aerosol flow then passes through the nozzle – made up of ceramic; see Figure 4(b) – gaining momentum for deposition on a free surface. The exit diameter of the nozzle is an influential factor on the width and final morphology of the deposited material.
1.5. Sensor-Instrumented Experimental Setup

All printing experiments, discussed in this dissertation, were carried out using an Optomec AJ-300 aerosol jet printer (Albuquerque, NM, USA). Shown in Figure 5, the experimental setup was further equipped with a 5 MP CCD camera (Edmund Optics, Grasshopper, GS3-U3-50S5C-C, Barrington, NJ, USA), to acquire online images from the printed structures. The camera was mounted on a 2.5X-10X variable magnification lens (Edmund Optics, VZM 1000i, Barrington, NJ, USA), illuminated by an LED ring light (AmScope, Irvine, CA, USA). This light source has a maximum brightness of 30,000 – 40,000 Lux and a color temperature of 6,000 K.
2. Review of Literature

AJP has been extensively utilized for the fabrication of a broad spectrum of products, such as interconnects, sensors, and transistors in addition to electromechanical, electrochemical, and optoelectronic devices. To a great extent, this can be attributed to the unique capabilities and advantages of AJP, including: (i) high-resolution material deposition, (ii) accommodation of a wide range of ink viscosity, (iii) being of a large standoff distance, which allows for deposition on non-planer surfaces, among others. In this section, a review of literature is performed, related to AJP-based device fabrication, demonstrating the scope and potential areas, engendered by AJP technology.

2.1. Device Fabrication

2.1.1. Interconnects

In a work by Krzeminski et al. [4], the electrical as well as morphological properties of silver nanoparticle-based interconnect structures, deposited using spray coating and AJP,
were compared. Overall, it was concluded that the AJ-printed structures had superior properties. The deposition of the AJP test structures (composed of 4 layers, 15 mm long) was carried out, utilizing an ultrasonic atomizer (having an atomization power of 45V) and a 150 μm nozzle with a speed of 1.5 mm/s and working distance between 2-5 mm. The sheath and atomization gas flow rates were set at 58 sccm and 10 sccm, respectively.

Jabari et al. [5] demonstrated AJ-deposition of silver interconnects on double-sided flexible Cu-PET substrates with the aim to fabricate sensors on chips. This study comprises optimization of the AJP process parameters as well as laser sintering parameters, aiding in achievement of conductive structures free of cracks and delamination. Besides, they developed a coupled thermal and structural finite element model to corroborate the experimental observations regarding the laser sintering of the deposited nanoparticles, and to explain the underlying phenomena behind crack formation.

In a research work by Navratil et al. [6], AJP was utilized to selectively print conductive paths – based on silver and gold nanoparticle inks, as well as PEDOT:PSS (an organic ink) – on a heated flexible substrate (Kapton polyimide). To ensure achievement of highly conductive structures, the main AJP process parameters (i.e., sheath and atomization gas flow rates, ultrasonic current, print speed, and platen temperature) were optimized. The impedance of the printed structures was monitored at various frequencies as a measure of device functional properties. It was demonstrated that optimal deposition of conducive paths would be critical for the fabrication of electronic components such as transistors, sensors, and radio frequency identification (RFID) antennae.

Vogeler et al. [7] employed AJP in a combined manner to deposit conductive (silver nanoparticle-based) high-resolution micro-tracks (having a width in the order of
approximately 100 μm - 1 mm) on extrusion-based 3D-printed substrates. One of the applications of this work would be optimal fabrication of electronic devices such as inductors, capacitors, and strain gauges. They assessed the influence of AJP process parameters on the morphological and functional properties of the printed structures. Sheath gas flow rate, aerosol flow rate (i.e., the difference between the atomization and exhaust gas flow rates in the virtual impactor, VI), ink temperature, substrate temperature, print speed, and working distance were identified as the main process parameters of the AJP process. They observed that line width could be well controlled by controlling the VI-difference. In addition, the sheath gas flow rate could control edge quality significantly. Furthermore, they found that side-by-side deposition of material might be more beneficial than multi-layer deposition.

Seifert et al. [8] investigated AJ-deposition of electrical chip interconnects on an integrated system-in-a-package (also known as SiP), meant to reduce size in terms of volume and footprint. Silver nanoparticle-based aerosols were deposited on a heated substrate, set in the range of 80-120 °C. From the design standpoint, they proposed that the problems of aerodynamic focusing could be overcome using adjustable deposition heads. Nozzle size and working distance were identified as two critical machine parameters, influencing the morphology of the deposited interconnects. They found that crack formation and lift-off effects, detrimental to device functional integrity, would stem from inappropriate/un-tuned surface properties as well as material’s coefficient of thermal expansion (CTE). Furthermore, the overall thermo-mechanical properties of a device would be a function of not only interconnect thickness, but also glass transition temperature, stress relaxation, and CTE. Path optimization, surface treatment, and
identification of extended properties (such as electro-migration) were suggested as means that could further help obtain printed paths with optimal properties.

Gu et al. [9] demonstrated combined AJ-printing of: (i) dielectric fillet structures (used as support structures) onto different leveled surfaces, and (ii) conductive interconnect structures used for circuitization and electrical connections between components. The multi-layer fillet structures were composed of an ultraviolet-curable polymer ink, cured in situ having self-leveled after deposition. Such a printing algorithm ensures formation of smooth surfaces. Silver nanoparticle-based interconnects were subsequently printed on top of the cured fillet structures, thermally sintered in an oven at 150 °C for 30 min. Indicating a satisfactory performance, the reliability and mechanical integrity of the fillet-interconnect structures were assessed using temperature cycling and adhesion tests.

Stoukatch et al. [10] evaluated the suitability of AJ-printed, laser-sintered conductive layers for electronic packaging technologies, such as wire-bonding and surface mount technology. Silver nanoparticle-based layers – deposited on various organic substrates, including polyimide, FR4, polycarbonate, and acrylonitrile butadiene styrene (ABS) – were gold and aluminum wire-bonded. The results illustrated that the wire-bonding of AJP layers might not be a reliable technique for mass production due to poor adhesion and metal delamination. In addition, the suitability of AJ-printed layers for surface-mount technologies (i.e., conductive adhesive and reflow soldering) was also characterized, employing shear tests. The results were consistent with the results of conventional processing of rigid substrates. Furthermore, they demonstrated successful fabrication of a functional prototype of an autonomous wireless sensor node system, based on AJP silver interconnects.
Hoerber et al. [11] utilized AJP for the electrical functionalization of thermoplastic circuit carriers toward the fabrication of molded interconnect devices for automotive applications. Silver nanoparticle tracks were deposited on thermoplastic substrates – including different grades of polyamide (PA6.6, PA6, and special PA6) as well as Polybutylene terephthalate (PBT)/ Acrylonitrile styrene acrylate (ASA) – at various pitches. These substrates were processed to exhibit different levels of surface quality. They found that the pitch setting would be an important factor, influencing the device functional properties. The reliability of the printed structures (in terms of mechanical and electrical properties) was further assessed via thermal shock tests (conducted between -40 °C and +125 °C at 1000 cycles). The results demonstrated that a combination of humidity and heat (e.g., 85% r.h., 85 °C for 504 hours) might significantly influence the adhesion of the deposited interconnects leading to delamination, although having marginal effects on the electrical properties.

Goth et al. [12] investigated potential integration of stereolithography and AJP toward fine-pitch fabrication of molded interconnect devices with consideration of silver inks and a palladium-based catalytic suspension – characterized with respect to their adhesiveness, conductivity, as well as wettability – on stereolithography resins and thermoplastic substrates. They illustrated that surface plasma treatment would: (i) increase line width, (ii) result in formation of even structures with clear morphology features, (iii) improve the adhesion of deposited material. Furthermore, multi-layer (vs. single layer) material deposition on a substrate might lead to crack formation due to different thermal expansion coefficients between the substrate and the ink material. As a complementary process, plating would allow for improving the thickness of the deposited material. Additionally,
substrate hardening time and temperature both would significantly influence the functional properties. Aerosol flow rate, print speed, and the number of passes were identified as critical process parameters.

### 2.1.2. Sensors

Lombardi et al. [13] investigated the AJ-fabrication of sensors (composed of silver nanoparticle- and carbon black-based interdigitated microelectrodes) deposited on multi-layered fibrous membrane paper substrates with the aim to detect chemical and biological species. Having the capability of liquid and vapor sensing, the printed sensors could be used in various applications, such as environmental and human-performance monitoring. They demonstrated that the sensors would not only be mechanically flexible, but also have high surface area (unlike their conventionally manufactured counterparts that are usually expensive and rigid, and also have limited surface area). They found that the paper-based sensors would be more sensitive – which could be, to a great extent, attributed to their permeable framework – than those fabricated based on non-porous polyimide substrates.

In a research work by Li et al. [14], two methods for the fabrication of highly sensitive, flexible strain sensors – composed of: (i) an aligned carbon nanotube network (CNTN), (ii) silver nanoparticle-based microelectrodes, and (iii) a polyimide substrate – were delineated. In the first method, the silver microelectrodes (1 mm wide) are AJ-deposited on top of the CNTN, attached to the polyimide substrate by an epoxy resin which is thermally cured, subsequently. The printed microelectrodes are furnace-sintered at 350 °C for 30 min. In the second method, the silver microelectrodes are AJ-deposited directly on the base polyimide substrate, i.e., being below the CNTN. The sensors fabricated using the
first and second methods demonstrated large positive and negative gauge factors, respectively.

Similarly, Blumenthal et al. [15] demonstrated conformal AJ-based fabrication of sensors and structural health monitoring systems (including strain gauges, circuits, and antennas) on 3D non-planer surfaces with the aim to monitor the performance of campsite structures during operation.

Zhao et al. [16, 17] introduced a novel method for the fabrication of composite structures with satisfactory sensing capabilities where strain sensors were AJ-deposited onto partially pre-cured carbon fiber prepgregs. Uncured resin flow was identified as the primary cause of sensor deformation. They demonstrated that pre-curing of the prepgregs by 10% would result in composite structures with almost no mechanical degradation and preserved interlaminar shear strength. Besides, they illustrated that the resistance change and electrical properties of the sensors (measured under cyclic loading) would be comparable to those of commercial foil strain gauges.

Liu et al. [18] elaborated high-resolution fabrication of hydrogen sensors, based on platinum-decorated single-walled carbon nanotubes (synthesized based on a series of physical as well as chemical processes). AJP was utilized to deposit microelectrode arrays. A process optimization study was conducted to determine the optimal distance between microelectrodes in addition to the optimal number of printing passes. The results exhibited reduced coffee-ring effects and achievement of printed sensors with high sensitivity toward hydrogen.

Zhao et al. [19] introduced a new ink synthesis method to improve the electrical conductivity of AJ-printed structures. They synthesized a novel ink, based on both silver
nanoparticles and carbon nanotubes (CNTs). They illustrated that as long as the CNT concentration remained below the percolation level, the CNTs could bridge/heal the defects of printed silver structures effectively, thus resulting in a reduction in the electrical resistivity by approximately 38%. Nevertheless, as the concentration of the CNTs increased above a threshold, an increasing trend was observed in the resistivity of the deposited structures, stemming from Schottky barrier effect.

Stoukatch et al. [20] forwarded a combined approach, employing AJP as an intermediate method to deposit silver tracks on a temporary transfer film and ultimately on a target vertical surface. The AJ-printed structures had a resistivity of approximately 27 - 33 μOhm.cm (before the pattern transfer), and 18 μOhm.cm (after the transfer). The reduction in the resistivity stemmed from post-cure effects. They fabricated a multi-sensor system, supported with wireless data transfer, that could be used for a broad range of IoT applications.

Vasiliev et al. [21] demonstrated AJ-printing of platinum nanoparticle-based micro-heaters (approximately 50 μm wide, having a particle size of 3–8 nm) as part of the manufacture of high-temperature metal oxide gas sensors with the aim to detect combustible and toxic gases.

Wang et al. [22] discussed fabrication of a thermistor sensor utilizing AJP, based on a synthesized nickel oxide nanoparticle ink, deposited in between two silver tracks on cutting inserts (coated with aluminum oxide layers). The fabricated sensors could operate in a wide range of temperatures (30–250 °C), exhibiting no hysteretic effects.
2.1.3. Transistors

With the utilization of AJP, Liu et al. [23] demonstrated fabrication of flexible, carbon-based field effect transistors (FETs) on polyethylene terephthalate (PET) substrates. The FET components were deposited, based on three carbon-based inks: (i) reduced graphene oxide, used for electrodes and channels; (ii) multi-walled carbon nanotubes, used for gate electrodes; (iii) graphene oxide, used as a dielectric material. All the fabricated FETs exhibited a good mobility.

Zhao et al. [24] demonstrated: (i) synthesis of a single-walled carbon nanotube (SWCNT) ink – tuned to be water-based and printable (in terms of viscosity) by changing the concentrations of its additives and surfactants – and (ii) fabrication of flexible, CoMoCat 76 SWCNT-based thin film transistors (TFTs), employing a combination of printing methods. First, they utilized a hybrid process with nanoimprinting to deposit source and drain electrodes on a polyethylene terephthalate (PET) substrate. Subsequently, inkjet printing was used to deposit the functionalized SWCNT ink on the TFT device channels. Ultimately, with the aid of AJP, the gate electrodes and dielectric layers (composed of silver and ion gel, respectively) were printed. The fabricated TFT devices exhibited effective mobility and on/off ratio.

In a similar work by Jones et al. [25], a surfactant-free SWCNT-based ink was initially synthesized, having a very low amount of metal content and impurities. Then, AJP was employed to fabricate flexible, SWCNT-based field-effect thin-film transistors (FE-TFTs). Unlike the fabrication of silicon-based transistors, the use of AJP eliminates the need for wet/dry chemical processing. The rheological properties of the synthesized SWCNT ink
were in the range that would additionally enable inkjet-based fabrication of FE-TFT device components.

Kim et al. [26] demonstrated AJ-fabrication of poly(3-hexylthiophene)-based electrolyte-gated transistors (EGTs) on polyester substrates. A device optimization study was performed with regard to the deposited material thickness as well as annealing temperature. They found that the transistors, deposited with a film thickness of approximately 50 nm and annealed at 120 °C would have the highest performance. The fabricated devices also exhibited robust mechanical properties (against bias stress and bending strain).

In a research work by Cao et al. [27], AJP was utilized to completely fabricate all components/layers of TFTs on polyimide substrates, including: (i) semiconducting material based on carbon nanotubes, (ii) metal electrodes based on silver nanoparticles, as well as (iii) gate dielectrics based on a two-part ink, i.e., poly(vinylphenol) and poly(methyl silsesquioxane). They observed negligible hysteresis in addition to low threshold voltage in the fabricated TFT devices. Besides, the reliability of the TFTs was assessed using aggressive bending tests, and the results indicated minimal variations in the performance.

2.1.4. Polymer Optical Waveguides (POW)

Reitberger et al. [28] used AJP as a complementary step to flexography to fabricate multimode polymer optical waveguides (POWs) with a focus on the optimization of transmission quality. Flexographic printing was initially used to print conditioning lines – composed of a silicon-based functional polymer, acting as a barrier for the core material that is subsequently AJP-printed – on a poly(methyl methacrylate) substrate, followed by
curing. Pneumatically atomized, the core polymer ink was preheated (between 45-60 °C) to reduce the viscosity. Using an in-line UV light source, the AJ-printed lines were cured layer-by-layer almost in real-time. Furthermore, an AJP process optimization study was conducted, and the results exhibited that setting both sheath and aerosol flow rates at 60 sccm could result in optimal print quality (if print speed and nozzle size were at 50 mm/s and 250 μm, respectively).

In another similar study by Reitberger et al. [29], POWs were fabricated on conditioned, flexible polymer foils of PVC, PMMA, and PI. In this regard, flexographic printing was utilized to deposit hydrophobic silicone-based barriers (100 μm apart, defining the waveguide width) on the polymer substrates. Next, liquid polymer-based core material (composed of transparent epoxy resins) was AJ-deposited between the barriers. The barrier tracks had low surface energy, leading to formation of waveguides with high aspect ratio. Finally, AJP was utilized again to print cladding material. As part of the work, they conducted an optimization study to obtain optimal AJP process parameters as well as surface properties.

Hoffmann et al. [30] similarly adopted a combined approach (based on flexographic printing and AJP) for the fabrication of high-resolution, high aspect ratio POWs (having a width and thickness of approximately 290 μm and 50 μm, respectively). The fabricated POWs exhibited satisfactory multimodal behavior, and could be used for short-range transmission applications.
2.1.5. **Electro-mechanical and -chemical Devices**

Utilizing AJP, Khorramdel *et al.* [31] forwarded a low-cost approach for making silicon-on-insulator (SOI) electrical contacts (also known as vias) in microelectromechanical systems (MEMS). They demonstrated that the AJ-fabricated vias (composed of aluminum nanoparticles) would effectively bridge the silicon substrate to the device layers with satisfactory electrical performance (a line resistance of $< 4\Omega$ was obtained).

The fundamental capabilities, advantages, and challenges of AJP – particularly concerning the fabrication of electrochemical energy conversion and storage devices – was reviewed in a work by Jay Deiner *et al.* [32]. The authors concluded that despite the capabilities and advantages of AJP, significant development would be required to realize the fabrication of the electrochemical devices. Besides, they emphasized: (i) ink formulation, (ii) material deposition, (iii) film formation, and (iv) consolidation as the main phenomena governing the AJP process, and optimal device fabrication would necessitate implementation of process monitoring and control.

2.1.6. **Solid Oxide Fuel Cell (SOFC)**

Sukeshini *et al.* [33] employed AJP to deposit functionally-graded solid oxide fuel cell (SOFC) anode interlayers, composed of nickel oxide and Yttria-stabilized zirconia (YSZ). Their AJP dual atomizer configuration allowed for deposition of the materials with various compositions. They demonstrated that although the compositional gradation and non-uniform distribution of nickel oxide and YSZ would lead to reduced ohmic resistance, the overall performance of fabricated cells might not be satisfactory, hence require further
investigation and improvement. This could be attributed to phenomena, such as improper mixing and/or aerosolized suspension de-mixing.

Besides, Sukeshini et al. [34] conducted an optimization study to investigate the influence of AJP process parameters on the microstructure of SOFC layers, deposited on nickel oxide/YSZ substrates. The deposited layers include: (i) a dense YSZ-based electrolyte layer, (ii) a porous cathode functional interlayer (composed of strontium doped lanthanum manganite (LSM)/YSZ), and (iii) an LSM-based cathode current collection layer. The fabricated cells demonstrated high electrochemical performance.

2.1.7. Quantum Dot Devices

Utilizing two atomizers, Oh et al. [35] demonstrated AJ-deposition of multicolor luminescent quantum dot patterns on heated substrates (aiding in reduction of the coffee ring effect). Their work has applications from electroluminescent devices to pixilated full-color displays.

With the aid of AJP along with pulsed laser direct-writing, Wang et al. [36] demonstrated fabrication of a photoluminescent quantum dot device, which could be used as a white light emitting source. The pulsed laser ablation method was utilized to create square holes, meant to confine AJ-deposited quantum dot droplets. In addition, the square holes would potentially aid in elimination of the coffee-ring effect. AJP allowed for homogenous deposition of a quantum dot-based solution (having, determined proportional compositions) into the laser-patterned square holes. The high-resolution of AJP would minimize (if not eliminate) the adverse effects of intermixing, and thus result in selective deposition of the quantum dots. Fitting the size of the square holes, individual droplets (30
μm in diameter) were AJ-generated by adjusting print speed as well as shutter on/off frequency. Their fabricated LED device demonstrated stable emission at various currents.

2.1.8. Flexible Electronics

Cao et al. [37] proposed a novel method for the fabrication of stretchable electronics, where functional materials, e.g., carbon nanotube (CNT) and reduced graphene oxide (rGO), would be AJ-deposited on flexible substrates, while stretched. In addition, curing/sintering would be implemented under the strained state. As a result, self-organizing structures, having a wrinkled profile, would form once the stress was released. They observed that the fabricated structures could be expanded and contracted repeatedly without significant loss in the electrical properties.

Renn [38] delineated the AJP-assisted fabrication of flexible and stretchable 3D interconnects (with good functional properties), sensors, as well as passive electronic components. Toward the fabrication of flexible interconnects, Renn demonstrated that AJP could be utilized to deposit not only elastic fillets, serving as a flexible bed between chip and substrate, but also metal nanoparticle-based electronic tracers along the ramp. The use of the supporting fillets would relieve stress on non-planer circuitry and electrical connections.

Elmogi et al. [39] similarly demonstrated the application of AJP for the fabrication of flexible chip interconnects with high frequency capabilities, compared to wire bonding, flip-chip, and through-silicon via technologies. Particularly, they discussed AJ-deposition of transmission interconnects with tunable impedance, with the aim for integration of high-frequency electronic and photonic chips. Utilizing AJP, silver electrical interconnects were
printed between emitting laser arrays and a driver, exhibiting satisfactory functional properties and withstanding standard reliability tests (85°C/85 relative humidity in the span of 700 hours).

In a research work by Mahajan et al. [40], fabrication of flexible substrates with embedded AJ-deposited silver electrodes was investigated. The fabrication process consisted of three steps: (i) insertion, (ii) curing, and (iii) delamination. The AJ-printed tracks were transferred from a low-energy donor surface to a reactive polymer substrate, attached to a flexible support substrate. The results of a bending test (conducted over 1000 cycles) exhibited no significant reduction in the electrical properties of the deposited electrodes.

Khan et al. [41] demonstrated AJ-deposition of power-efficient, high-temperature micro-hotplates (composed of gold nanoparticles) on polyimide substrates. A power consumption of 54 mW and 40 mW at 325 °C was achieved for micro-hotplates, having an area of 500×500 μm² and 300×300 μm², respectively. A numerical model was forwarded for thermal design and optimization. The AJ-deposited hotplates could be used for the fabrication of chemical sensors as well as thermal metrology devices, among others.

Tu et al. [42] utilized AJP to print silver nanowire-based electrodes on an ITO-patterned glass substrate for flexible electronic applications. The number of passes, print speed, and nozzle size were identified as significant process parameters, influencing the performance of the deposited nanowire electrodes. They had a width and sheet resistance of approximately 50 μm and 57.68 Ω/sq, respectively.

Habermehl et al. [43] demonstrated an integrated approach for the fabrication of highly-sensitive lab-on-chip bioanalysis systems, consisting of: (i) roll-to-roll hot
embossing of microfluidic channels on polystyrene foils, (ii) AJ-deposition of gold nanoparticle-based surface-enhanced Raman spectroscopy (SERS) substrates, and (iii) roll-to-roll, thermal bonding of the chip assembly. They conducted an optimization study to investigate the influence of AJP parameters on the functional and morphological properties of the deposited SERS structures.

2.1.9. Circuits

Ha et al. [44] demonstrated AJ-printing of a low-voltage circuit, designed to drive an electrochromic pixel. The main components of the circuit, i.e., transistors, capacitors, as well as resistors, were all AJ-deposited on a polymer substrate along with the electrochromic pixel. Furthermore, to control the circuit current, electrolyte-gated transistors (EGTs) were AJ-printed. The EGT gate insulator layers as well as semiconductor channels were composed of ion gels and poly(3-hexylthiophene), respectively. Only metallic electrodes and electronic tracks (interconnects) were printed with the aid of photolithography.

Sarobol et al. [45] reviewed additive manufacturing technologies – including direct write methods (e.g., inkjet, AJP, and extrusion casting) as well as thermal spray methods (e.g., plasma spray and aerosol deposition) – that could be utilized together with subtractive and ceramic sintering methods in an integrated fashion for the fabrication of multilayer hybrid circuits. Coating materials (such as composites, ceramics, and polymers) can be deposited with the aid of the thermal spray methods. They discussed case studies where additive manufacturing processes integrated with conventional subtractive and material
processing methods (such as sputtering and electroplating) could be used for the fabrication of hybrid devices beyond 2-D planes.

Lan et al. [46] utilized AJP to directly deposit dielectric layers as well as gold nanoparticle-based interconnects on a microwave monolithic integrated circuit, with the aim to connect the gate and ground pads. The fabricated circuit exhibited satisfactory RF performance. Current stress as well as thermal shock and cycling tests were performed to thoroughly assess the reliability of the fabricated circuit device, hosting the AJ-deposited interconnects. No degradation in the performance was observed. The outcome of their work highlights the fact that AJP is a reliable method for direct, high-resolution deposition of on-chip functionalities and features.

Breyfogle [47] discussed potential fabrication of typical electrical components and devices – including antennae and strain gauge sensors in addition to power and signal circuits – via integration of fused filament deposition (FDM) and AJP processes. They found that these printing technologies were compatible and could support emerging applications, although further investigations would be required to address the challenges associated with design rules, performance, etc.

2.1.10. Optoelectronics

Eckstein et al. [48] utilized AJP to deposit current-collecting grids (based on a complex metal-organic silver ink) on transparent electrodes acting as an anode for optoelectronic devices. The electrodes were composed of poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate), known as PEDOT:PSS. The influence of print speed on grid line morphology was explored. Stemming from drying effects, the problem of inhomogeneities
in deposited material at high print speeds was overcome, using high boiling point co-solvents. Finally, they demonstrated fabrication of inverted organic solar cells, utilizing AJ-fabricated hybrid electrodes, on ITO/ZnO-covered glass substrates.

Tait et al. [49] demonstrated AJ-deposition of transparent layers and polymer tracks (having a width and thickness of 30 μm and 30 nm, respectively) for high-resolution fabrication of RGB organic light emitting diodes (OLEDs), based on non-halogenated solvents. An optimization study was conducted with consideration of atomization, exhaust, and sheath gas flow rates in addition to working distance, platen temperature, and print speed.

With the use of AJP, Zhou et al. [50] demonstrated deposition of PEDOT:PSS grids on the ITO-based anode surface of OLEDs. An enhancement in the performance (in terms of power and maximum current efficiency) was achieved, which could be ascribed to conductivity as well as light extraction of the AJ-embedded grids.

Based on experimental observations, Rudorfer et al. [51] elaborated the advantages and potentials, engendered by AJP for the fabrication of LEDs from various perspectives, such as high-precision deposition of: (i) die-attach material, (ii) interconnects, and (iii) phosphor layers (utilized for white light generation as well as color conversion), among others.

2.1.11. Photodetectors and Photoswitches

Aga et al. [52] demonstrated AJ-fabrication of a multilayer, polymer-based photodetector on a paper substrate. The photoactive layer was composed of both poly(3-hexylthiophene) and C61-butyric acid methyl ester (P3HT:PCBM). Besides, the bottom and top electrodes were made up of silver nanoparticles and PEDOT:PSS, respectively. In
addition, a biopolymer interlayer (deoxyribonucleic acid-based) was printed between the photoactive layer and the top electrode (allowing for direct deposition of the top electrode material on the photoactive layer).

Similarly, Wang et al. [53] utilized AJP for deposition of nanowire networks on PET substrates based on a synthesized zinc-octaethylporphyrin (ZnOEP) ink, used for the fabrication of photodetectors (and potentially, photoswitches). The fabricated ZnOEP nanowire-based photodetectors exhibited satisfactory functional and mechanical proprieties (e.g., fast photo-response, fast recovery time, good reversibility, as well as high photo-sensitivity).

Aga et al. [54] employed AJP to precisely deposit PEDOT:PSS (as a coating material) with controlled thickness on an oxygen plasma-treated electrode (composed of Ni(O2-Ni)) with the aim to improve the performance of the electrode for photodetector applications. Subsequently, an infrared photodetector was fabricated using the modified electrodes. A threefold increase in the photodetector response was observed.

2.1.12. Solar Cells/Photovoltaic Devices

Williams et al. [55] demonstrated deposition of nanocrystals for the fabrication of dense polycrystalline semiconductor thin films used in solar cells with the use of AJP (utilizing an ultrasonic atomizer with currents adjusted in the range of 0.30–0.45 mA in addition to two nozzles having diameters of 150 and 200 μm). Sheath and carrier gas flow rates were varied approximately between 20–70 and 20–25 sccm, respectively. Furthermore, print speed was changed in the span of 1-8 mm/s. The ink was composted of copper zinc tin sulfide (CZTS) crystals (~5 nm in diameter) dispersed in toluene. Coating
density and width were identified as two critical morphology traits. It was demonstrated that under certain conditions, line width would be no longer a function of the focusing ratio – i.e. the ratio of the sheath gas flow rate to the carrier gas flow rate – and spreading phenomena after deposition would become dominant over aerodynamic collimation phenomena. Increasing the focusing ratio would lead to continuous but porous deposited structures. High solvent content was noted as a source of crack formation. They found that coating morphology might be affected by aerosol transport time as well as solvent evaporation rate. Besides, particle agglomeration within an aerosol droplet could be a leading cause of discontinuous coating deposition. At short transport times, aerosol droplets were solvent-laden, and coalescence was the dominant phenomenon, resulting in cracked structures; this stemmed from stress relaxation after drying. Modeling of solvent partial pressure and agglomerate formation was proposed as open research areas, which would help address the challenges of conformal material deposition.

Kalio et al. [56] demonstrated AJP-based front side metallization of n-type silicon solar cells (with boron-doped emitters and phosphourous-doped back surface field, based on inks synthesized in house, i.e., pSISC and nSISC, respectively). Subsequently, the cells were furnace-fired to bring the fabrication process (including AJP followed by silver electroplating) close to standard industrial production. With the aid of the AJ-metallization, solar cells with an improved efficiency of approximately 19.6% were fabricated. The improvement in the efficiency could be attributed to better material passivation.

Similarly, in a research work by Platt et al. [57], AJP metallization of silicon photovoltaics was demonstrated, using an aluminum nanoparticle-based ink. The use of AJP eliminated the need for screen-printing metallization of solar cells, being of the
following advantages: (i) high material utilization, and (ii) contactless deposition. Having a thickness in the range of 1-10 μm, the AJ-deposited current-collecting tracks (technically referred to as back contacts) were sintered between 550-800 °C. Subsequently, crystalline silicon solar cells were fabricated (based on the deposited aluminum tracks), exhibiting an efficiency of about 13%.

Kopola et al. [58] utilized AJP for the metallization of indium tin oxide (ITO)-free inverted organic solar cells (using silver nanoparticles). In the proposed configuration, the ITO contact layer would be replaced with a conductive and transparent PEDOT:PSS layer. In addition, because illumination direction is from the grid side, optimization of the grid area coverage as well as conductivity is a bourgeoning need. These characteristics were controlled using three process parameters, i.e., (i) the number of passes, (ii) platen temperature, and (iii) print speed. Line conductivity and aspect ratio could be improved via increasing the number of passes, without compromising on keeping the area coverage minimum. The AJ-printed silver grids had a line width of approximately 60 μm, deposited on the PEDOT:PSS layer. It was demonstrated that the photovoltaic devices fabricated with the embedded AJ-printed grids had similar performance, compared to those fabricated based on evaporated gold grids.

Yang [59] et al. employed AJP for deposition of the active layers of polymer solar cells. In this regard, organic inks were synthesized, based on additives and solvents with high boiling points. They explored the influence of solvent vapor annealing as well as thermal annealing on the device performance. Solvent drying rate was identified as an influential factor on deposited layer surface morphology and molecular ordering during polymer synthesis.
Hörteis et al. [60] studied AJP-assisted formation of front side contact layers as part of the fabrication of silicon solar cells where seed layers were initially AJ-printed (based on silver nanoparticles) and then, laser-fired. Light induced plating was subsequently utilized to grow the front side contact layers. Transfer length model-based measurements were performed to assess the functional properties of the front side contacts. Having an emitter sheet resistance of 110 Ω/sq as well as an area of 4 cm², photovoltaic devices were fabricated, exhibiting an efficiency of 20.3%.

Hörteis et al. [61] similarly utilized AJP for the front side metallization of silicon solar cells. Having been fired and plated, the AJ-deposited lines covered 70% of a screen-printed contact’s area. The AJP-based metallization was accomplished, using two inks: (i) a synthesized, chemically-stable ink, and (ii) a diluted screen printing paste. The synthesized ink had a particle loading of approximately 70 wt%, allowing for an increase in print speed, and thus a reduction in line width (without compromising on functional properties). Conductive lines having a width of 20 μm and 50 μm were obtained, based on the synthesized ink and the diluted screen printing paste, respectively. Likewise, utilizing these ink materials, photovoltaic devices were fabricated, exhibiting efficiencies of 16.5% and 18.3%, respectively.

In a similar research work by Mette et al. [62], AJP-based front side metallization of solar cell devices was demonstrated. In the proposed method, as discussed earlier, the seed layer (approximately 15 μm wide) of a contact structure is initially AJP-deposited on a silicon surface, based on a metal-organic ink. In the second step, the conductivity of the AJP-deposited seed layer is further increased using light-induced silver plating. In addition, they proposed use of modified screen printing pastes to improve the seed layer adhesion.
and other mechanical properties. Monocrystalline silicon solar cells with embedded AJ-printed layers as well as aluminum back surface fields were fabricated, exhibiting an efficiency of 17.8%.

Binder *et al.* [63] demonstrated AJP-assisted, reliable formation of silver contacts on lowly-doped boron and phosphorus emitters (on silicon substrates with pyramid textures) for photovoltaic devices. A silver ink with a density tuned in the range of 6 to 23 g/m$^3$ was used to print seed layers (approximately 40 μm wide) on the textured surfaces. The thickness of the deposited layers was further increased using electro-plating. They found that the influence of post firing temperature would be more significant than that of the amount of deposited material.

Rodriguez *et al.* [64] discussed selective etching of dielectric layers using AJP (as a high-resolution and high-throughput direct write method vs. already utilized inkjet printing) for photovoltaic applications. Grooves as small as 20 μm were etched in pre-patterned dielectric layers, designed for surface passivation of photovoltaic cells. Furthermore, AJP was utilized to etch point openings for fabrication of metal contacts with the aim to enhance rear surface passivation. The number of passes, print speed, in addition to sheath and carrier gas flow rates were identified as influential process parameters, allowing AJP to be a flexible process for various applications.

### 2.2. Ink and Substrate Materials

Umrani *et al.* [65] demonstrated AJ-fabrication of high-aspect ratio ceramic micro-pillar arrays, using a formulated Yttria Stabilized Zirconia (YSZ) ink, for MEMS, SOFC, and imaging applications. As a preliminary step, they explored the effect of particle loading
on the ink viscosity. Subsequently, via designed experiments, the influence of the main parameters of AJP process on both the diameter and the height of deposited pillars was studied (with target values of 50 µm and 1000 µm, respectively). In addition, the optimum distance between the pillars was identified; this is a critical factor, affecting the functional properties of the deposited pillar arrays.

In a research work by Jabari et al. [66], AJP was utilized to print crack-free, flexible, and conductive graphene/silver structures (synthesized with a ratio of 3:1 by volume) for the fabrication of flexible electronic devices. The graphene/silver patterns exhibited not only less resistivity than those made of individual graphene or silver inks, but also the same level of flexibility/reliability as highly flexible graphene structures.

Besides, Jabari et al. [67] employed AJP to deposit conductive interconnects based on a graphene ink (having viscosity, concentration, and particle size of approximately 21 cP, 3 mg/ml, and 200 nm, respectively) on silicon/silicon-dioxide wafers. Atomization power/current, atomization gas flow rate, and the number of passes were noted as influential process parameters on print morphology. They demonstrated that the AJ-printed graphene structures had width and resistivity in the range of 10-90 µm and 0.018 Ω cm, respectively. The graphene ink was synthesized using chemical exfoliation of graphene flakes (via long term sonication in ethanol) followed by adding ethyl cellulose (EC) as a stabilizer to increase the graphene concentration. Particle size, viscosity, and surface tension were mentioned as critical characteristics, which would make an ink suitable for aerosol jet printing. Increasing the substrate temperature could improve the surface energy, adhesion, and as a result, uniformity of the printed structures. It was found that annealing processing (used to remove the ink solvents as well as stabilizing polymers) would not
have significant influence on the width of the deposited structures (provided that the surface energy was kept high). Furthermore, the width would be significantly influenced by the atomization current and carrier gas flow rate. They measured the thickness, resistance, and resistivity of the AJ-printed structures, using AFM, 4-points probe, and Van der Pauw methods, respectively.

Rahman et al. [68] showed that unlike conventional subtractive manufacturing methods (such as lithography), AJP could be used for the fabrication of 3D dielectric-metal microstructures such as passive electronic devices for wearable and smart electronic applications (with comparable dimensional accuracy). A curable dielectric ink as well as a metal (silver) nanoparticle-based ink were used separately, having a viscosity of 300 and 160 cP, respectively. Their AJ-printing system was additionally equipped with a titled head (used for the deposition of the silver nanoparticle ink), a UV light source (for curing the polymer ink), as well as a laser system (for sintering the deposited metallic structures). Standoff distance and flow parameters (i.e., sheath, carrier, and exhaust gas flow rates) were identified critical to print morphology.

Lai et al. [69] utilized AJP for the fabrication of large area, defect-free molybdenum disulfide (MoS$_2$)-based films. An ink was synthesized, composed of self-assembling peptides, bonding to 2D flakes of MoS$_2$ dispersed in a solvent system. They found that the following factors would significantly influence the assembly of the AJ-deposited MoS$_2$ films: (i) the age of the peptides, (ii) MoS$_2$ solvent type, (iii) excessive deposition, (iv) low aerosol gas flow pressure, (v) drop deposition, and (vi) peptide sit time. It was concluded that changing the functionalization of the peptides as well as the peptide concentration both would not have significant effects on the properties of the deposited films.
In a research work by Ahn et al. [70], amphiphilic silver microparticles (composed of poly(acrylic acid), known as PAA) were prepared for the synthesis of conductive inks with tunable wettability. The surface chemistry of the particles was modified using Amidation reaction. Conductive structures (36 μm wide and 1.2 μm thick) were AJ-printed (utilizing ultrasonic atomization and a nozzle being of a diameter of 150 μm) on polyimide and glass substrates. Print speed was changed in the range of 1-10 mm/s. Having a viscosity of 2.5 cP, the ink was based on 27.5 wt% solids dispersed in a 1:1 water:IPA solvent system. Thermal and photonic annealing were used to improve the electrical conductivity of the deposited structures (measured by the 4-point probe method). With the use of a goniometer, the contact angle as well as the surface tension of the synthesized inks were measured.

Vunnam et al. [71] investigated various surface treatment techniques (such as air plasma, dodecyltrichlorosilane-based chemical treatment, and ultrasonic treatment) to modify indium tin oxide (ITO) surfaces grown on glass substrates. The aim was to obtain an optimal interface between AJ-deposited silver nanoparticle tracks and the ITO surfaces. The treated surfaces were assessed with respect to their surface roughness, homogeneity, and polarity. Using van Oss-Chaudhury-Good method, the components of surface free energy (e.g., contact angle hysteresis, Lewis acid-base, as well as Lifshitz-van der Waals) were compared and contrasted against each treatment technique.

2.3. Sintering

Jabari et al. [72] forwarded a new laser-based sintering method (using a 1550 nm continuous-wave Erbium fiber laser) for the thermal treatment of AJ-printed graphene structures (typically carried out at a temperature of less than 250 °C). The aim was to
employ laser sintering as an in situ localized post-processing method, compared to conventional methods, such as thermal treatment in vacuum ovens and tube furnaces. They demonstrated that the laser sintering method would form structures with minimal compressive stress (otherwise leading to formation of defects in the graphene structures). Besides, the resulting thickness and resistivity were comparable to those of conventionally sintered structures. Consequently, this method would have the capability of being integrated with micro-fabrication systems (such as AJP), allowing for in situ localized low-temperature sintering, protecting low-melting temperature substrates from warpage. Using the laser system introduced additional corresponding process parameters, i.e., laser power (1-10 W), beam diameter (fixed at 50 μm), and sintering speed (fixed at 0.03 mm/s). The range of each parameter was optimally chosen, using heat transfer simulations (setup based on the properties of the deposited graphene structures, e.g., absorptivity and thermal diffusivity). In addition, the thermal model allowed for prediction of the temperature of the graphene structures during laser sintering as a function of laser power. They found that a substrate temperature of 150 °C could improve surface energy and thus adhesion, leading to formation of reproducible high-quality printed structures. An online imaging system – consisting of a complementary metal-oxide semiconductor (CMOS) camera mounted on a variable magnification lens (VZM 1000i) – was utilized to contrast the morphology of sintered and unsintered structures. In addition, offline techniques such as optical microscopy, SEM, Raman spectroscopy, and XPS\textsuperscript{1} were employed to further characterize the morphology of the sintered structures. The thickness and resistivity were measured with the air of AFM\textsuperscript{2} and the 2-point probe method, respectively. The presence of ink solvent as

\textsuperscript{1} X-ray Photoelectron Spectroscopy
\textsuperscript{2} Atomic Force Microscopy
well as stabilizing materials in the printed structures appeared as active defects, which could be removed substantially using the laser sintering method.

Utilizing AJP and photonic sintering, Mahajan et al. [73] demonstrated low-cost fabrication of biodegradable electronic devices, using an ink composed of polyvinylpyrrolidone (PVP)-coated Zn nanoparticles (leading to formation of conductive Zn matrix after sintering). Their study showed that: (i) the ink compositions, (ii) photonic energy, (iii) film thickness, and (iv) ventilation conditions would influence the effects of the photonic sintering. In addition, it was demonstrated that employing a combined, cascaded approach of photonic and laser sintering could further improve the conductivity of the AJ-deposited structures. Similarly, an improvement in the conductivity could be obtained with the aid of acid-reduction of surface oxidation. They indicated that the photonic sintering process would be implemented rapidly (in the order of a few microseconds).

2.4. Process Development

Wang et al. [74] introduced a new AJP-based dual-material (DM) direct-write method (called DM-AJP) primarily for the fabrication of smart nanocomposites. In the DM-AJP process, utilizing both ultrasonic and pneumatic atomizers, two independently controlled streams of materials are mixed before aerodynamic collimation and deposition; this allows for not only deposition of composite structures but also tuning the functional properties in near real-time. Particularly, they investigated the fabrication of freestanding nanocomposites, made based on a carbon nanotube (CNT) ink as well as a poly amic acid (PAA) solution aerosolized ultrasonically and pneumatically, respectively. In this hybrid
method, unlike conventional fabrication techniques, since the conductive components (i.e., CNTs) and the hosting polymer matrix (i.e., polyimide) are deposited together, robust and reliable structures are achieved.

Efimov et al. utilized AJP equipped with a spark discharge generator to deposit dry silver nanoparticles on a heated silicon substrate (being at 200-300 °C) [75] as well as on a glass substrate [76]. The use of the heated substrate eliminates the need for post-deposition sintering. Using the spark generator (working based on electrical erosion of silver electrodes), silver nanoparticles were produced with a size of 15-100 nm. This method allows for generation of an environmentally friendly source of nanoparticles with zero solvent content. In the study, employing the heated silicon substrate, the authors obtained deposited silver structures (200 μm wide and 20 μm thick) – printed in 7 layers with a speed of 0.12 mm/s, focusing ration of 3, and working distance of 0.5 mm – having a resistivity of 35 μΩ·cm (obtained at 250 °C). However, the samples printed on the glass substrate, were furnace-sintered at 450°C for 1 hour; they showed a resistivity of 7.5 μΩ·cm. They identified porosity as well as interparticle contact resistance as two main factors, contributing to the larger resistivity of the printed structures compared to bulk silver.

Chang [77] addressed the lack of quality control in AJP by forwarding a quality control framework, including a vibration-based wavelet method (used to monitor atomization/nebulization process, estimating a stable process time) as well as some image processing techniques (used to monitor the morphology of AJ-printed lines). A process model was developed, based on experimental observations using central composite design (CDD), to understand the influence of process parameters on printed line morphology. A
20% reduction in process variation in addition to a 50% improvement in completion rate (in AJ-fabrication of sensors and interconnects) was reported in the author’s work. Furthermore, Chang developed the following two methods: (i) print-stick-peel (PSP), allowing for integration of sensors on 3D printed objects; (ii) dual material aerosol jet printing (DMAJP) for in situ mixing of polymer and filler materials with the aim to fabricate conductive nanocomposites.

3. Goal and Objectives

As delineated in this chapter, despite all unique advantages and engendered strategic applications, AJP is a highly unstable and complex process, prone to gradual drifts. Consequently, real-time process monitoring seems to be inevitable. The goal of this doctoral research work is to forward a robust, image-based framework for real-time functional monitoring and control of AJP process. In pursuit of this goal, the following objectives are defined:

(i) *In situ* image acquisition from the traces/lines of printed electronic devices right after deposition. To realize this objective, as discussed in Chapter 2, the AJP experimental setup is instrumented with a high-resolution imaging system (consisting of a charge-coupled device (CCD) camera and a variable-magnification telescopic lens), which allows for acquisition of RGB images of deposited material in near real-time.

(ii) *In situ* image processing and quantification of line morphology. Several image processing algorithms are introduced to quantify various 2D and 3D aspects of printed line morphology – e.g., line width, density, overspray, thickness, cross-
sectional area, and surface roughness – as discussed in Chapter 2 and Chapter 3, respectively. The 3D morphology characterization is based on the concept of shape-from-shading (SfS) image analysis.

(iii) Development of a supervised multiple-input, single-output (MISO) machine learning model, based on the concept of sparse representation for classification (SRC), discussed in detail in Chapter 4. The aim is to estimate device functional properties in near real-time with an accuracy of $\geq 90\%$. The forwarded model formulates a classification problem as a combination of: (i) Absolute Shrinkage and Selection Operator (LASSO), (ii) Elastic Net, and (iii) Ridge regression.

(iv) Development of a computational fluid dynamics (CFD) model to explain the underlying aerodynamic phenomena behind material transport and deposition in AJP process. A 2D-CFD model is initially forwarded to corroborate the experimental results presented in Chapter 2. Furthermore, in Chapter 5, the development of a 3D-CFD model – as a compressible, turbulent multi-phase model for the flow of aerosols within the deposition head as well as after deposition on a free surface – is discussed. The CFD model allows for: (i) investigation of post-deposition phenomena of receding, relaxation, and wetting equilibrium; and (ii) simulation of the aerosol flow behavior in critical conditions, where conducting experiments is time-consuming and/or expensive.
References


Chapter 2: Computational Fluid Dynamics (CFD) Modeling and Online Monitoring of Aerosol Jet Printing (AJP) Process

1. Introduction

1.1. Goal and Objectives

The goal of this work is to quantify the link between process parameters and the print quality in terms of the morphology of printed lines (electronic traces) in aerosol jet printing (AJP) additive manufacturing (AM) process; and subsequently elucidate the underlying causal aerodynamic interactions that lead to certain trends in line morphology vs. process parameters as observed from experimental data. In pursuit of this goal, the specific objectives of this work are to:

(1) Quantify the line morphology using online images acquired by an in situ charge coupled device (CCD) camera as a function of two AJP process variables, namely, sheath gas flow rate (ShGFR) and carrier gas flow rate (CGFR).

(2) Formulate a two dimensional computational fluid dynamics (2D-CFD) model to explain the experimental trends in terms of ShGFR and CGFR, and thereby, elucidate the underlying aerodynamic phenomena that influence line morphology in AJP. This will lead to an understanding of how and why ShGFR and CGFR interact to influence line morphology.

Accordingly, this work seeks to connect the physical process interactions in AJP with morphological integrity of deposited lines through online image analysis and CFD modeling, and thus answers the following open research questions:

- What are the process-machine interactions in AJP that influence the line morphology, and how to quantify the line morphology from online images?
- What are the causal aerodynamic phenomena that govern line morphology in AJP?
The practical aim is to use the CCD camera-based image analysis to detect incipient drifts in line morphology and, subsequently, invoke CFD model-derived predictions to suggest the appropriate corrective action for future closed-loop process control in AJP.

1.2. Motivation

Aerosol jet printing (AJP) facilitates a host of strategic applications, such as manufacture of complex geometry antennae, organic photovoltaics, embedded flexible electronics, etc. [1, 2]. In such emerging applications, line morphology attributes, e.g., line width, overspray, line continuity, etc., are critical for ensuring device functional integrity [3-5]. Currently, AJP lines are characterized using offline techniques, such as optical microscopy and profilometry as exemplified in Figure 1(a-c), and Figure 2, respectively. However, these techniques are ex situ, and are therefore not amenable for online rectification in case of a process drift. Instances of deleterious process drifts in AJP are exemplified in Figure 5 (Sec. 1.5).

![Offline Images taken with a Microscope](image1)

**Figure 1:** (a-c) Offline images of AJP-printed electronic traces captured by an optical microscope (model: Carl Zeiss M1M); (d-f) Online images captured by a high-resolution CCD color camera installed on our experimental setup.
Consequently, it is practically expedient to suggest avenues for sensor-based online monitoring of line morphology in AJP process. Furthermore, physical models are required to elucidate how the aerodynamics of aerosol flow influences line morphology. This will facilitate model-based, closed-loop control of AJP process in the future.

1.3. Feasibility of Online Monitoring in AJP Process

In this work, we show that online monitoring of line morphology is feasible by integrating high-resolution optical imaging hardware and novel image data analytics approaches. For instance, Figure 1(d-f) shows that online images acquired using an *in situ* CCD color camera (Point Grey, Grasshopper) installed on our experimental testbed (Optomec – AJ-300) are comparable to the offline images shown in Figure 1(a-c) captured using an optical microscope (Carl Zeiss M1M Axio Imager). The experimental setup is explained in detail later in Sec. 2 (shown in Figure 6).

We note that the three-dimensional line morphology corresponding to Figure 1(a-c) observed using an optical profiler (Figure 2(a-c)) also depicts facets seen in the top view with two-dimensional imaging (Figure 1). This justifies that the use of two-dimensional imaging for gauging line morphology is not impractical. Although there is a loss of information in the vertical direction, line facets, such as overspray are nonetheless captured with 2D images. Figure 1 and Figure 2 thus demonstrate the feasibility of using the CCD camera-based optical imaging as a key data acquisition step, which paves the way for online monitoring of AJP process.
1.4. Overview of Aerosol Jet Printing (AJP) Process

Aerosol jet printing (AJP) is a non-contact, droplet-based, direct write (DW), additive manufacturing (AM) technique [1]. AJP is used in the manufacture of flexible electronics, molded interconnect devices, high aspect ratio interconnects, conformal antennas, biosensors, among others such emerging applications [6-10] (see, e.g., Figure 3). In these aforementioned applications, conventional electronics manufacturing techniques, e.g., photolithography, electroplating, etc., are limited due to geometry, harsh operating conditions, and material properties [11].

Novel solution-based materials, such as metal nanoparticles, graphene, and PEDOT:PSS can be AJP-deposited. Furthermore, low temperature, ambient fabrication of electronics is possible [7-10]. Figure 3 exemplifies some of the electronic devices and structures printed using AJP process at the authors’ laboratories in Binghamton University (SUNY). These include an antenna printed on a flexible glass substrate (Figure 3(a)), reduced graphene oxide (rGO) supercapacitors (SCs) and silver interdigitated electrodes (IDEs) printed on a slim glass substrate (Figure 3(b)), silver test lines printed on a polymer substrate (Figure 3(c)), and IDEs printed on polyimide (Figure 3(d)), respectively.
Figure 3: Electronic devices and structures AJP-printed at the authors’ facilities in Binghamton University. (a) An antenna printed on a flexible glass substrate; (b) Reduced graphene oxide (rGO) supercapacitors (SCs) and silver interdigitated electrodes (IDEs) printed on a slim glass substrate; (c) Silver test lines printed on a polymer substrate; and (d) Silver interdigitated electrodes (IDEs) printed on polyimide.

Key advantages of AJP, over competing processes, e.g., inkjet printing, are that the process accommodates a wider range of ink viscosities (0.7-2500 cPs), operates at larger standoff distances (~1-5 mm) allowing printing over non-planar substrates, and has the capability of printing geometries with higher resolution (≤10 μm) [1, 11]. Salient aspects of inkjet printing and AJP are juxtaposed in Table 1 (data synthesized from Ref. [6, 12-17]). A schematic of the AJP process is shown in Figure 6 (Sec. 2). A liquid ink, consisting of dispersed nanoparticles, such as silver or copper (particle size approximately in the range of 30 nm to 50 nm), is nebulized/aerosolized using an ultrasonic or pneumatic atomizer. The type of nebulization technique is predicated by the application area and ink properties.

Table 1: A comparison between inkjet and aerosol jet printing (AJP) [6, 12-17].

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Inkjet Printing</th>
<th>Aerosol Jet Printing (AJP)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Deposition Mechanism</td>
<td>Electrostatic</td>
<td>Aerodynamic</td>
</tr>
<tr>
<td>Viscosity (cP)</td>
<td>10-20</td>
<td>1-2500</td>
</tr>
<tr>
<td>Working Distance (mm)</td>
<td>1</td>
<td>1-5</td>
</tr>
<tr>
<td>Print Speed (mm/s)</td>
<td>Up to 5000</td>
<td>Up to 200</td>
</tr>
<tr>
<td>Dynamic Accuracy (μm)</td>
<td>N/A</td>
<td>±6</td>
</tr>
<tr>
<td>Line Width (μm)</td>
<td>2-200</td>
<td>5-30</td>
</tr>
</tbody>
</table>
In the ultrasonic atomization technique, a transducer which has a frequency, typically, in the range of 1.6–2.4 MHz is used for droplet generation [2]. Inks having high vapor pressure and low boiling point loaded with particles of less than 50 nm in diameter can be ultrasonically atomized [17]. In addition, the range of viscosity for such inks is limited to 0.5–15 cP [17]. On the other hand, in pneumatic atomization, a high-pressure flow of a gas (e.g., Nitrogen) disperses the ink into droplets. Pneumatic atomization is usually accompanied with an aerodynamic separator called virtual impactor (VI). Separation is carried out with the aid of an exhaust flow which eliminates small-medium sized droplets [17]. As a result, only droplets with larger size and momentum enter the deposition head. In general, inks having vapor pressure less than 0.1 mm (Hg), boiling point larger than 180 °C, viscosity in the range of 0.7–1000 cP, and particle size larger than 50 nm are amenable for pneumatic nebulization [17].

A carrier gas flow conveys the aerosolized ink (mist) to a deposition head. The carrier gas flow is saturated by passing through a bath (bubbler) of solvent material. This impedes changes to the ink chemistry stemming from evaporation of the solvent. The solvent used in the bubbler is contingent upon the solvent(s) used in the ink. The saturated carrier gas, containing the suspended ink droplets, then flows to a deposition head, where the carrier gas is surrounded by another gaseous stream, called sheath gas. The sheath gas flow aids in focusing/collimating the carrier gas flow.
In our experimental testbed, a pure and dry stream of nitrogen is used as both carrier and sheath gas flows. The focused aerosol flow, which is a multi-phase stream of solid nanoparticles and ink solvents in a gas, is passed through a nozzle (~100-300 µm in diameter) and deposited on a target surface/substrate (1-5 mm standoff distance). Because there is a continuous flow of aerosol, a mechanical shutter is used to control the deposition of material on the substrate. The ratio of the sheath gas flow rate (ShGFR) to the carrier gas flow rate (CGFR) is termed as the focusing ratio (Fr) which is a consequential factor in determining line morphology [18]. Among other process factors (shown in Figure 4), nozzle geometry is also important as it influences the fluid flow dynamics and, thereby, the morphology of printed geometries [4, 19].

1.5. Literature Review - Research Challenges and Gaps in the State-of-the-Art

We now identify the main impediments in the context of process control, monitoring and modeling in AJP.

1.5.1. Process Complexity: Despite the advantages and novel applications engendered by AJP, complex material, machine, and process interactions influence line morphology as shown in Figure 4. A review of literature reveals that the ShGFR and CGFR are the most consequential factors [3, 4, 18].
Figure 4: A list of material, machine, and process factors influencing the morphology and functional integrity of an AJP-printed line.

Further process complexity stems from the effect of material formulations and properties. Ink temperature instability, as well as solvent evaporation are noted as the leading causes of material-related fluctuations, such as ink pre-drying and particulate accumulation within the nozzle [17]. As mentioned earlier, depending on link viscosity, ultrasonic or pneumatic atomization can be used [12, 18, 20-27]. For instance, Figure 5 exemplifies different situations where the aerosol flow deviates from its optimal target conditions.

![Image](image_url)

Figure 5: Different situations where line morphology deviates (drifts) from the target, stemming from complex materials, process, and machine interactions. (The images are thresholded to binary equivalents.)
Figure 5(a) and (b): Improper process conditions, such as low carrier gas flow rate and inadequate ink atomization, lead to insufficient flow density and discontinuous flow.

Figure 5(c): Overspray occurs as the focusing ratio (F_R), defined as the ratio of ShGFR to CGFR, exceeds the critical limit necessary to maintain flow collimation.

Figure 5(d): Excess flow results from increased ink temperature and, thus, excessive ink atomization (under fixed process parameters).

Figure 5(e): Inconsistent flow collimation usually occurs when the F_R is exceedingly high.

In other words, the AJP process has a tendency to drift given the complex interactions amongst variables. Hence, in situ monitoring and closed-loop control in AJP process is a burgeoning need.

1.5.2. Lack of Quantifiers of Line Morphology: Several researchers have investigated the effect of AJP process parameters on print quality, employing offline characterization techniques [18, 28-33]. For example, in a study by Mahajan et al. [18], the effects of sheath gas flow rate (ShGFR), carrier gas flow rate (CGFR), and print speed (P_S) on the geometrical and electrical properties of aerosol jet printed lines were investigated. Verheecke et al. [32] utilized image processing and profile analysis for offline characterization of line morphology. For process control, more response variables capturing different aspects of print quality, such as overspray, continuity, density, etc., should be defined and quantified in a real-time fashion. There is a gap in the literature in the context of online characterization of line morphology.

1.5.3. Lack of Integrated Process Modeling: Several researchers have worked on discrete-phase modeling, as well as aerosol transport and deposition mechanisms in AJP.
Schulz et al. [34] presented mathematical models for discrete phase in a micro-capillary with a focus on a new deposition technique called collimated aerosol beam direct-write (CAB-DW). Their simulation results showed that the degree of collimation at the nozzle exit is around 20 times larger than that at the nozzle entrance. Schulz et al. also showed that at high velocities of the aerosol flow, the Saffman force becomes significant causing the particles to migrate radially inward (toward the center line) [34]. This phenomenon was further corroborated by Akhatov et al. [35, 36] where they showed that not only the particle inertia and Stokes drag force, but also the Saffman force should be considered when modeling the aerosol flow through long micro-capillaries. The authors demonstrated that the magnitude of the Saffman force, hence the degree of collimation is influenced by the size as well as the position of particles in a shear flow. Akhatov et al. [37] also presented a new solution for gas flow in a nozzle of slowly-converging diameter.

Maximum spread factor was one of the main aerodynamic criteria employed by Feng [38] in modeling of aerosol deposition on free surfaces. Feng studied the dynamics of micro-droplet impact in the phases of spreading, receding-relaxation, and wetting equilibrium. Feng’s experimental results showed that when the effects of viscosity are small compared to those of inertia and surface tension, there is a clear distinction between periodic oscillation and aperiodic creeping-to-capillary equilibrium. Feng also investigated the phenomena of droplet bounce after deposition which may be significantly influenced by the contact angles and droplet viscosity.

In another research work by Feng [39] concerning deformations of a sessile drop under an impinging jet, it was shown that unsteady droplet deformation occurs when the capillary number (representative of the influence of viscous forces versus surface tension across an
interface) exceeds a critical value of shape instability. This phenomenon was captured by an empirical model developed for a practical range of the Reynolds number and droplet sizes.

The aforementioned CFD-related research in the literature predominantly focusses on the behavior of the discrete phase, while more holistic physical models are necessary to establish a relationship between process parameters and line morphology attributes. The objective of the present work (from a CFD perspective) is to forward a 2D model to explain the overall interactions between the gas flows (namely, sheath and carrier) and to simulate the influence of main process parameters on the aerodynamics of aerosol flow in the print head, as well as during the deposition on a free surface substrate. A 3D-CFD model will be presented as part of our future work with consideration of more parameters, such as contact angle, surface tension, Weber number (We), etc.

The rest of this chapter is structured as follows: the experimental AJP setup, integrated with a CCD camera is described in Sec. 2. A 2D-CFD model simulating aerosol flow in the deposition head is presented in Sec. 3. A novel digital image processing method devised to quantify line morphology and, ultimately, capture process drifts is delineated in Sec. 4. The experimental results and conclusions are discussed in Sec. 5.

2. Experimental Setup

2.1. Machine and Materials

All the experiments discussed in this chapter were carried out using an Optomec model A-300 aerosol jet printer. Pure (99.998%) and dry nitrogen (at 20 °C) was the primary source of material used for both sheath and carrier gas flows. The carrier gas flow was
saturated, initially, on passing through a bubbler containing deionized water. Conductive
lines were printed on a commercial glossy photo paper (HP, 4×6 in) using an aqueous ink
(Clariant PRELECT® TPS-50) loaded with silver nanoparticles (50 wt%, particle size in
the range of 30 nm to 50 nm). The ink density, viscosity, and surface tension were 1.8
(g/ml), 15 ± 2 (mPa s), and 35 ± 3 (mN/m), respectively. The ink solvent system was made
up of water and iso-propyl alcohol (1:1 by volume). The ultrasonic bath stabilizer was set
at 21°C.

2.2. CCD Camera Integrated AJP Setup

A picture and a schematic diagram of the sensor-integrated Optomec AJ-300 AJP
printer are shown in Figure 6. A five megapixel (5 MP) CCD color camera (model: Point
Grey-Grasshopper) with a 2.5X – 10X variable magnification lens was the primary source
of images in this study. Illumination was provided by a LED ring light which has a
brightness in the range of 30,000 – 40,000 Lux and a color temperature of 6000 K. The
images taken have dimensions of 2448 × 2048 pixels, and each pixel has dimensions of
0.36 × 0.36 μm at the set field of view.
Figure 6: (a)-(b) Pictures and (c) a schematic diagram of the experiential setup showing the imaging components installed on the Optomec AJ-300 system.

As schematically shown in Figure 6(c), immediately after printing a set of lines, the platen is automatically translated under the high-resolution CCD camera (installed perpendicular to the substrate, coaxial to the nozzle), and digital images are acquired. The whole process of translation and imaging is completed within 30 seconds (see Sec. 4.2 for a note on processing time delay). In other words, our experimental setup enables monitoring of the AJP process in an online, near real-time manner. Subsequently, as delineated in Sec. 4, six morphological features, i.e., (i) Line Width ($L_W$), (ii) Line Density ($L_D$), (iii) Edge Quality/Smoothness ($L_{EQ}$), (iv) Overspray Index ($L_OS$), (v) Line Discontinuity ($L_{Disc}$), and (vi) Internal Connectivity (measured using the Fiedler Number...
(λ2)), are quantified using the image processing method developed in this study. Next, we use a computational fluid dynamics (CFD) model to explain the various aerodynamic phenomena influencing print quality in AJP.

We also attempted to incorporate a high power lens to the existing alignment camera on the AJ-300 printer. However, this caused significant jitter and instability (attributed to the added weight). The other challenges with using the alignment camera are as follows: (1) the alignment camera is not coaxial, hence it is not tractable to capture the topology of a line; (2) only a small portion of the line was captured in absence of a high magnification, wide angle lens on the alignment camera; (3) consistent illumination cannot be provided due to the glancing-angle nature of the alignment camera.

2.3. Test Artifact

In this work, we focus on simple line-type geometries as shown in Figure 1. This is because, simple geometries allow more accurate modeling and analysis of process phenomena. Furthermore, the overlapping lines used in making complex structures may occlude the changes in print quality. In contrast, a single line can provide greater sensitivity and clarity for analysis. From a practical vista, real-life electronic devices are replete with simple lines; indeed, simple line-type features are printed on electronic devices and wafers as part of the layout for quality assurances purposes. This placement of simple line-type features is analogous to the process monitoring coupons or structures used in electronics fabrication. Simple structures, such as Van der Pauw, transfer length measurement, or capacitor structures, are routinely built on PCBs and wafers to verify the quality of the
deposited material and processes [40]. Accordingly, the use of simple line-type structures for process monitoring is reasonable from a practical perspective.

3. A 2D-CFD Model to Study the Aerosol Flow Dynamics

A 2D-CFD model is proposed in this study to explain the underlying aerodynamic phenomena that govern the experimentally observed trends in line morphology. The intent is to quantify line morphology as a function of nozzle geometry, gas flow rates, as well as ink characteristics (such as viscosity, density, surface tension, etc. [12]).

CFD modeling and simulation of the AJP process under identical experimental conditions will facilitate a priori anticipation of the effects of the process parameters, such as ShGFR and CGFR, on line morphology. Moreover, in case of process drift, the CFD model will provide a baseline for understanding how the process may have changed, and thus facilitate bringing the process back in control. The technical terms used in this section are listed in Table 2.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>( C_v )</td>
<td>Heat Capacity at Constant Volume</td>
<td>( u )</td>
<td>x-Component of the Fluid Flow/Mixture Velocity</td>
</tr>
<tr>
<td>( d )</td>
<td>Displacement Vector</td>
<td>( u_N )</td>
<td>x-Component of the Individual Phase Velocity</td>
</tr>
<tr>
<td>( D )</td>
<td>Droplet Diameter(^1)</td>
<td>( \mathbf{v} )</td>
<td>Droplet Velocity Vector</td>
</tr>
<tr>
<td>( D_v )</td>
<td>Volume-Equivalent Sphere Diameter</td>
<td>( v )</td>
<td>y-Component of the Fluid Flow/Mixture Velocity</td>
</tr>
<tr>
<td>( e )</td>
<td>Specific Internal Energy</td>
<td>( v_N )</td>
<td>y-Component of the Individual Phase Velocity</td>
</tr>
<tr>
<td>( e_N )</td>
<td>Specific Internal Energy of the Individual Phase</td>
<td>( V_d )</td>
<td>Droplet Volume</td>
</tr>
<tr>
<td>( f )</td>
<td>Drag Factor(^2)</td>
<td>( \dot{w} )</td>
<td>Rate of Work Done per Unit Mass</td>
</tr>
<tr>
<td>( F )</td>
<td>Inviscid Flux Vector</td>
<td><strong>Greek Letters</strong></td>
<td></td>
</tr>
<tr>
<td>( g )</td>
<td>Gravitational Acceleration Vector</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( J )</td>
<td>Inviscid Flux Vector</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \mathbf{f} )</td>
<td>Inviscid Flux Jacobian</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( k )</td>
<td>Fluid Thermal Conductivity(^3)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^1\) \( D_v \) is also known as Volume-Equivalent Sphere Diameter.
\(^2\) Drag Factor can be calculated using the formula: \( f = \frac{F}{D \cdot \rho \cdot u^2} \), where \( F \) is the drag force, \( D \) is the diameter, \( \rho \) is the density, and \( u \) is the velocity.
\(^3\) Fluid Thermal Conductivity can be expressed as: \( k = \frac{\text{Heat Capacity}}{\text{Density} \cdot \text{Temperature}} \).
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m$</td>
<td>Mass</td>
</tr>
<tr>
<td>$p$</td>
<td>Fluid Flow/Mixture Pressure(^4)</td>
</tr>
<tr>
<td>$Q$</td>
<td>Solution Vector</td>
</tr>
<tr>
<td>$\dot{q}$</td>
<td>Rate of Heat Transfer per Unit Mass</td>
</tr>
<tr>
<td>$R$</td>
<td>Universal Gas Constant</td>
</tr>
<tr>
<td>$S_M$</td>
<td>Momentum Source Term</td>
</tr>
<tr>
<td>$S_E$</td>
<td>Energy Source Term</td>
</tr>
<tr>
<td>$t$</td>
<td>Time</td>
</tr>
<tr>
<td>$T$</td>
<td>Fluid Flow Temperature</td>
</tr>
<tr>
<td>$u$</td>
<td>Velocity Vector</td>
</tr>
<tr>
<td>$\mu_c$</td>
<td>Dynamic Viscosity of the Continuous Phase</td>
</tr>
<tr>
<td>$\rho$</td>
<td>Fluid Flow/Mixture Density(^6)</td>
</tr>
<tr>
<td>$\rho_i$</td>
<td>Density of the Individual Phase</td>
</tr>
<tr>
<td>$\rho_c$</td>
<td>Density of the Continuous Phase</td>
</tr>
<tr>
<td>$\rho_d$</td>
<td>Density of the Discrete Phase(^7)</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>Deviatoric Stress Tensor</td>
</tr>
<tr>
<td>$\phi$</td>
<td>Scalar of Interest</td>
</tr>
<tr>
<td>$\Phi$</td>
<td>Dissipation Function</td>
</tr>
<tr>
<td>$\omega_d$</td>
<td>Angular Velocity of a Droplet</td>
</tr>
<tr>
<td>$\Omega_c$</td>
<td>Vorticity of the Continuous Phase</td>
</tr>
</tbody>
</table>

1 The uniform model was selected to represent the droplet size distribution. The mean value is 3 (µm).
2 The drag coefficient was calculated using the spherical drag law assuming smooth aerosol particles.
3 The preset value of the fluid thermal conductivity is 0.0242 (W/(m. K)).
4 The fluid pressure was obtained using the ideal-gas law, being in the range of 424-1734 (Pa).
5 The fluid viscosity was defined using Sutherland’s law.
6 The ideal-gas law was employed to define the fluid density.
7 The preset value of the density of the discrete phase is 1800 (kg/ m\(^3\)).

* In this section, vectors are represented as bold, non-italic symbols; scalars are represented as italic, non-bold symbols. Subscripts/superscripts \(c\), \(d\), \(E\), \(f\), \(i\), \(L\), \(M\), \(N\), \(r\), \(R\), \(v\), \(x\), and \(y\) are respectively representative of continuous phase, discrete phase, energy, face value, the \(i\)th element, left, momentum, individual phase, reference state, right, volume, x-direction, and y-direction.

3.1. Geometry Modeling and Meshing

3.1.1. Determining the Deposition Head Geometry: As the first step towards constructing a CFD model, the deposition head assembly including the nozzle (Figure 7(a)-(b)) was modeled in the ANSYS-Fluent environment. The nozzle has a diameter of 150 µm. The veracity of the constructed model was corroborated based on a patented design by King [41] (of Optomec, Inc.), shown in Figure 7(d). This patented design is identical to the nozzle used in our machine. Furthermore, to observe the internal structure and measure the dimensions more accurately, we X-ray CT scanned the deposition head as shown in Figure 7(c). Figure 7(a) depicts the main components of the deposition head including:

1. an outer shell on which both ShGF and CGF inlet ports are embedded;
2. ShGF-guiding upper and lower plenum chambers (UPC and LPC, respectively);
3. a converging combination chamber (CC) where the aerosol flow is ShGF-collimated, and then accelerated; and
(4) a deposition nozzle which further increases the linear momentum of the flow required for stable and coherent aerosol deposition.

3.1.2. **Meshing:** Having determined the geometry, the closed surface was divided into discrete facets which allow finer meshes to be generated, and as a result, the geometric domain to be approximated more accurately. Both the horizontal and vertical edges were further divided into 10 sections with no bias allowing for mesh refinement. Soft, quadrilateral dominant meshes with no suppression were generated and refined three times with consideration of a medium degree of smoothing to ensure accuracy of simulation results.

![Figure 7](image)

Figure 7: (a) The main components of the deposition head. (b) The deposition head assembly. A cross-sectional view of the deposition head (c) obtained using X-ray CT and (d) found in literature [41] used to model the geometry.

### 3.2. Governing Equations

The aerosol flow in AJP is essentially a multiphase flow composed of three components: (i) solid nanoparticles suspended in a (ii) liquid environment forming the ink droplets conveyed by a flow of (iii) carrier gas (usually Nitrogen) [42–44]. The density-based Navier-Stokes algorithm was employed to solve the multi-phase problem, which is a high-speed compressible flow in a nozzle with complex geometry. The velocity, pressure, and density fields are obtained from the momentum equations (Eq. (2)-(3)), the equation of state (Eq. (5)), and the continuity equation (Eq. (1)), respectively.
For the ShGF-collimated aerosol deposition through a long, converging micro-capillary nozzle, the equations of continuity, momentum, and energy are solved simultaneously. For each simulation under steady-state conditions, a converged solution was obtained. Eq. (1)-(5) show the governing equations for a 2D compressible, Newtonian fluid flow [45, 46],

Continuity Eq.: \[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0 \tag{1}
\]

x-Momentum Eq.: \[
\frac{\partial (\rho u)}{\partial t} + \nabla \cdot (\rho u \mathbf{u}) = -\frac{\partial p}{\partial x} + \nabla \cdot (\mu \nabla u) + S_{Mx} \tag{2}
\]

y-Momentum Eq.: \[
\frac{\partial (\rho v)}{\partial t} + \nabla \cdot (\rho v \mathbf{u}) = -\frac{\partial p}{\partial y} + \nabla \cdot (\mu \nabla v) + S_{My} \tag{3}
\]

Energy Eq.: \[
\frac{\partial (\rho e)}{\partial t} + \nabla \cdot (\rho e \mathbf{u}) = -p \nabla \cdot \mathbf{u} + \nabla \cdot (k \nabla T) + \Phi + S_E \tag{4}
\]

Eq. of State (for Perfect Gas): \[
p = \rho RT \quad \text{and} \quad e = C_v T \tag{5}
\]

where \(\rho\) is the fluid density (kg/m\(^3\)), \(\mathbf{u}\) is the velocity vector (m/s), \(u\) is the x-component of velocity (m/s), \(v\) is the y-component of velocity (m/s), \(p\) is the fluid pressure (Pa), \(\mu\) is the fluid dynamic viscosity (mPa.s), \(e\) is the specific internal energy (J/kg), \(k\) is the fluid thermal conductivity (W/(m.K)), \(T\) is the fluid temperature (K), \(R\) is the universal gas constant (m\(^3\).Pa/(K.mol)), and \(C_v\) is the heat capacity at constant volume (J/K). In addition, \(S_M\) and \(S_E\) are the momentum and energy source terms, respectively; \(\Phi\) represents the dissipation function [46].

We note that under steady-state conditions, the time-dependent term, \(\frac{\partial (\cdot)}{\partial t}\), in Eq. (1)-(4) vanishes. With consideration of the nozzle outlet diameter as well as the average diameter of the combination chamber (both as the characteristic length) together with the mean velocity, density, and dynamic viscosity of the fluid flow, the Reynolds number (Re) was computed under identical experimental conditions as a function of ShGFR. It was observed that the flow regime would remain laminar in the specified range of the ShGFR (40-140 sccm). Hence, a laminar viscous model was chosen. Figure 8 shows the Reynolds number
plotted against ShGFR for the internal flow in the combination chamber as well as at the
nozzle exit, demonstrating that the flow regime remains laminar as the ShGFR increases.

Figure 8: The Reynolds number as a function of ShGFR showing that the internal flow
remains laminar both in the combination chamber and at the nozzle exit. A laminar
viscous model was chosen, as a result.

3.3. Discrete-Phase Modeling

The aforementioned continuity, momentum, and energy equations must be modified
when the fluid flow is composed of multiple phases. Eq. (6)-(9) [47] express the combined
phase continuity, momentum, and energy equations.

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot \left( \sum_{N} \rho_{N} \alpha_{N} u_{N} \right) = 0 \tag{6}
\]

\[
\frac{\partial}{\partial t} \left( \sum_{N} \rho_{N} \alpha_{N} u_{N} \right) + \nabla \cdot \left( \sum_{N} \rho_{N} \alpha_{N} \nabla u_{N} \right) = -\frac{\partial p}{\partial x} + \rho \mathbf{g} \cdot \nabla + \nabla \cdot (\sigma_{c}^{d} \mathbf{e}) \tag{7}
\]

\[
\frac{\partial}{\partial t} \left( \sum_{N} \rho_{N} \alpha_{N} v_{N} \right) + \nabla \cdot \left( \sum_{N} \rho_{N} \alpha_{N} v_{N} u_{N} \right) = -\frac{\partial p}{\partial y} + \rho \mathbf{g} \cdot \nabla + \nabla \cdot (\sigma_{c}^{d} \mathbf{e}) \tag{8}
\]

\[
\frac{\partial}{\partial t} \left( \sum_{N} \rho_{N} \alpha_{N} e_{N} \right) + \nabla \cdot \left( -u_{c} \sigma_{c} \mathbf{c} - v_{c} \sigma_{c} \mathbf{c} + \sum_{N} \rho_{N} \alpha_{N} u_{N} e_{N} \right) = \dot{q} + \dot{w} \tag{9}
\]

where \( N \), in general, represents the individual phases or components constituting the
multi-phase system, \( \rho_{N} \) is the density of the individual phase, \( \alpha_{N} \) is the volume fraction of
the individual phase, \( u_{N} \) and \( v_{N} \) are, respectively, the x- and y- components of the velocity.
of the individual phase, \( \mathbf{u}_N \) is the velocity vector of the individual phase, \( p \) is the mixture pressure, \( \mathbf{g} \) is the gravitational acceleration vector, \( \sigma_c^d \) is the deviatoric stress tensor, and \( e_N \) is the specific internal energy of the individual phase. In addition, \( \dot{q} \) and \( \dot{w} \) are the rate of heat transfer and work done per unit mass, respectively.

Interacting with the continuous phase, the discrete phase is primarily composed of the ink droplets having a mean diameter of 3 \( \mu \text{m} \). It was assumed that the discrete phase has the same temperature as the ink vial bath (21 °C). In addition, the droplets are injected from the CGF surface (inlet) in the normal direction (shown in Figure 10) with an initial velocity which is the same as the carrier gas flow velocity.

Newton’s second law of motion, Eq. (10), is employed to determine the movement of droplets,

\[
\sum \mathbf{F} = V_d \rho_d \frac{d\mathbf{v}}{dt}
\]  

where \( V_d \) is the droplet volume, \( \rho_d \) is the droplet density, and \( \mathbf{v} \) is the droplet velocity vector. Schematically shown in Figure 9, the sum of hydrodynamic forces, \( \sum \mathbf{F} \), acting on a particle is mathematically expressed by Eq. (11).

\[
\sum \mathbf{F} = \mathbf{F}_D + \mathbf{F}_{\text{Basset}} + \mathbf{F}_{VM} + \mathbf{F}_{PG} + \mathbf{F}_g + \mathbf{F}_{Bu} + \mathbf{F}_{Saff} + \mathbf{F}_{Mag}
\]  

where \( \mathbf{F}_D \) is the drag force, \( \mathbf{F}_{\text{Basset}} \) is the Basset force, \( \mathbf{F}_{VM} \) is the virtual mass force, \( \mathbf{F}_{PG} \) is the pressure gradient force, \( \mathbf{F}_g \) is the gravitational force, \( \mathbf{F}_{Bu} \) is the buoyancy force, \( \mathbf{F}_{Saff} \) is the Saffman lift force, and \( \mathbf{F}_{Mag} \) is the Magnus lift force [2, 35, 36].
Figure 9: A free body diagram showing the forces acting on a particle in a shear flow. \( \mathbf{u} \) is the carrier flow velocity vector; \( \mathbf{v} \) is the particle velocity vector; \( \mathbf{\omega}_d \) is the particle rotation vector.

The drag force, \( \mathbf{F}_D \), exerted on a particle moving in a laminar, viscous flow is given by Eq. (12) [48],

\[
\mathbf{F}_D = 3\pi \mu_c D_v f (\mathbf{u} - \mathbf{v})
\]  

(12)

where \( \mu_c \) is the dynamic viscosity of the continuous phase, \( D_v \) is the volume-equivalent sphere diameter, \( f \) is the drag factor, and \( (\mathbf{u} - \mathbf{v}) \) is the relative velocity. Depending on the conditions and physics of a problem, the Stokes equation, i.e., Eq. (12), should be corrected; a new term referred to as the drag coefficient, \( C_d \), is added to the Stokes equation, as follows [2].

\[
\mathbf{F}_D = \frac{\pi}{8} \rho C_d D_v^2 f (\mathbf{u} - \mathbf{v})^2
\]  

(13)

In this study, aerosol particles are assumed smooth spheres. Hence, the following form of the drag coefficient called the Spherical Law [49, 50], was chosen as part of modeling the discrete phase in ANSYS,
\[ C_d = b_1 + \frac{b_2}{Re} + \frac{b_3}{Re^2} \]  

(14)

where, \( b_1, b_2, \) and \( b_3 \) are constants which are selected over ranges of the Reynolds number.

The Basset force, \( \mathbf{F}_{Basset} \), is expressed as [2, 48],

\[
\mathbf{F}_{Basset} = \frac{3}{2} D^2 \sqrt{\pi \rho_c \mu_c} \left[ \int_0^t \frac{d}{dt} \left( \mathbf{v} - \mathbf{u} \right) \frac{\mathbf{u} - \mathbf{u}_0}{\sqrt{t - t'}} dt' + \frac{(\mathbf{v} - \mathbf{u}_0)}{\sqrt{t}} \right]
\]  

(15)

where \( D \) is the droplet diameter, \( \rho_c \) is the density of the continuous phase, \( \mathbf{u} \) is the velocity vector of the continuous phase, \( \mathbf{v} \) is the droplet velocity vector, and \( (\mathbf{v} - \mathbf{u}_0) \) is the initial velocity difference or relative velocity.

The virtual mass force, \( \mathbf{F}_{VM} \), also known as the apparent mass force, is defined as follows [48],

\[
\mathbf{F}_{VM} = \rho_c V_d \left( \frac{\mathbf{d} \mathbf{u}}{\mathbf{d} t} - \frac{d \mathbf{v}}{d t} \right)
\]  

(16)

where, \( V_d \) is the droplet volume. Assuming the droplets are smooth spheres, a virtual mass factor (added mass coefficient) of 0.5 was used [2].

The force resulting from the difference between the pressure of the continuous phase and that of the discrete phase is represented by the pressure gradient force, \( \mathbf{F}_{PG} \), given by Eq. (17) [2, 35, 36],

\[
\mathbf{F}_{PG} = m \frac{\rho_c}{\rho_d} \left( \frac{d \mathbf{u}}{d t} - [(\mathbf{v} - \mathbf{u}) \cdot \nabla] \mathbf{u} \right)
\]  

(17)

where \( m \) and \( \rho_d \) are the droplet mass and the density of the discrete phase, respectively.

The buoyancy force, \( \mathbf{F}_{Bu} \), is equal to [2, 35, 36],

\[
\mathbf{F}_{Bu} = m \left( 1 - \frac{\rho_c}{\rho_d} \right) \mathbf{g}
\]  

(18)

where \( \mathbf{g} \) is the gravitational acceleration vector.
The Saffman lift force, $F_{Saff}$, which is due to the pressure distribution on a droplet in a velocity gradient [48], is expressed as,

$$F_{Saff} = 1.61 \mu_c D |\mathbf{u} - \mathbf{v}| \sqrt{\frac{D^2 d\mathbf{u}}{\gamma_c dy}}$$  \hspace{1cm} (19)

where $\gamma_c$ is the kinematic viscosity of the continuous phase.

The Magnus lift force, $F_{Mag}$, which is due to rotation of particles is expressed as follows [2, 48],

$$F_{Mag} = -\frac{3}{4} \frac{m}{\rho_d} \left( \frac{1}{2} \Omega_c - \omega_d \right) \times (\mathbf{v} - \mathbf{u})$$  \hspace{1cm} (20)

where, $\Omega_c$ is the vorticity of the continuous phase surrounding the droplet and $\omega_d$ is the angular velocity of the droplet.

Because the size of aerosol particles is typically in the range of $0.5$–$5 \mu m$ [2] and the continuous phase is a gaseous medium assumed to be nonrotational, only the drag force, as well as the Saffman lift force are significant, and accordingly, included in the discrete phase modeling [2, 36, 48, 51].

### 3.4. Boundary Conditions

As shown in Figure 10, the following four types of boundary conditions were defined.
(1) Velocity-inlet was the type of boundary set for both sheath gas flow (ShGF) and carrier gas flow (CGF) with consideration of the initial velocity ($u_0$) and pressure ($p_0$). Flowing normal to the boundary surface at ambient temperature (300 K), these two gas streams consist of nitrogen with a mass density and dynamic viscosity modeled by the ideal-gas law and Sutherland’s law, respectively. Eq. (21) gives Sutherland’s formula developed for the shear viscosity of gasses [52].

$$\frac{\mu}{\mu_r} = \frac{T_r + A_1}{T + A_1} \left(\frac{T}{T_r}\right)^{1.5}$$

In this equation, $\mu_r$ represents the gas viscosity at the reference temperature of $T_r$; $A_1$ is a constant.

(2) Stationary-wall boundary condition was set for both the deposition head and the nozzle interior surface with no-slip condition for the continuous phase and reflect condition
for the discrete phase. It was assumed that all of the deposition head surfaces are made from stainless steel.

(3) Pressure-outlet with escape condition for the discrete phase was the boundary type defined for the jet flow leaving the nozzle in the direction normal to the surface at atmospheric pressure.

(4) Stationary wall with no-slip condition for the continuous phase and trap condition for the discrete phase was the type of boundary set for the substrate operating at 40°C. A moving wall condition allows simulation of print speed. We will employ the moving wall scenario in our future work for 3D-CFD modeling.

3.5. A Review of the Numerical Schemes Used for Solving the CFD Model

The CGF inlet is the surface where from the computation of the flow field variables starts. The Implicit, Least Square Cell Based (i.e., Eq. (22) which is based on the Green-Gauss theorem), and Second Order Upwind (Eq. (23) schemes [49] were employed, respectively, for discretization of time, gradient, and flow to obtain the face fluxes for all cells [49].

\[
(\nabla \phi)_0 \cdot \Delta d_i = (\phi_i - \phi_0)
\]
\[
\phi_f = \phi + \nabla \phi \cdot d
\]

In Eq. (22)-(23), $\nabla \phi_0$ is the gradient of the scalar of interest at the central cell (i.e., Cell 0), $\Delta d_i$ is the displacement vector between the centroids of Cell 0 and the $i$th cell, and $\phi_f$ is the face value of the scalar of interest. The Roe Flux-Difference Splitting (Roe-FDS) scheme was used for evaluation of the inviscid flux vector, $J$, at each face as shown by Eq. (24) [49],

\[
J = 0.5 \left[ (J_R + J_L) - \Gamma |J| \delta Q \right]
\]
where, $\mathbf{J}_R$ is the right flux vector, $\mathbf{J}_L$ is the left flux vector, $\hat{\mathbf{J}}$ is the inviscid flux Jacobian composed of a diagonal matrix of eigenvalues as well as a modal matrix, $\mathbf{P}$ is the preconditioning matrix, and $\delta \mathbf{Q}$ is the spatial difference between the right and left solution vectors. Based on the eigenvalues of the system, the flux vector contains characteristic information as advancing through the domain.

The drag coefficient on the interior wall of the deposition head was selected to monitor the convergence of solutions. All simulations presented in this study converged to a steady-state solution.

### 3.6. Limitations of the 2D CFD Model

We herewith note the following limitations of the 2D-CFD model of AJP presented in this work, vis-à-vis a 3D model:

The velocity and pressure fields obtained from a 2D and a 3D flow may differ due to distinctive definitions of the boundaries of a geometry. A 2D model assumes that the geometry of the print head assembly is infinitely wide in the z-direction, while a 3D model considers the wall effects.

Hence, from the line morphology perspective, unlike the 2D model, almost all the morphological features observed experimentally such as overspray, line density, line discontinuity, edge smoothness, etc., can be verified by a 3D model.

In general, a 3D model might furnish tighter model agreement with experimental observations of influence of AJP process parameters, such as print speed, standoff distance, etc., on line morphology which are not captured with a simplified 2D model of the process. We will extend the present 2D model to a 3D-CFD model for AJP in our future work. In
the subsequent section (Sec. 4), we detail the image processing approach devised to quantify line morphology obtained from online images.


The aim of this section is to propose morphology quantifiers from image data to capture different attributes of line quality. These quantifiers are: (1) line width ($L_w$), (2) line density ($L_ρ$), (3) Edge Quality/Smoothness ($L_{EQ}$), (4) Overspray Index ($L_{OS}$), (5) Line Discontinuity ($L_{Disc}$), and (6) Internal Connectivity measured via the Fiedler Number ($λ_2$).

Unlike image-based transforms, such as 2D-FFT, Hugh transform, and Radon transform, which may only capture the overall morphology of an image, these quantifiers are specifically formulated to capture specific (local) aspects of the morphology [53]. In case of drift in print quality, the presented approach can be used to identify the specific characteristics of line morphology that are affected. This allows an appropriate corrective action to be taken based on the CFD model. Furthermore, because elementary matrix operations are used to obtain these quantifiers, they are more computationally efficient compared to involved image analysis techniques.

4.1. Initial Processing of Online Images

As an initial step in the quantification of line morphology, an online RGB (true color) image is converted to its grayscale equivalent. Then, two spatial threshold parameters, called Line Threshold and Overspray Threshold, are determined systematically using a threshold estimator (proposed in this study). It calculates the global extrema of the first derivative of the image’s intensity profile as exemplified in Figure 11(a)-(c). Based on the
values of two threshold parameters, the image is segmented into three separate zones of background (BG), overspray (OS), and line width (LW). The threshold parameters are determined once and remain fixed thereafter provided that illumination conditions and camera settings are not changed. The zone detection and segmentation paves the way for the main morphology-quantifying algorithm (delineated in Sec. 4.3) to quantify the morphological features of the line.

Figure 11: (a) The zones of background (BG), overspray (OS), and line width (LW) detected based on two spatial threshold parameters (obtained using a threshold estimator, proposed in this study); (b) The one-dimensionalized intensity profile of the image shown in Part (a); (c) The first derivative of the intensity profile indicating the approximate range of each zone based on which the threshold parameters (as reference intensities) as well as the coordinates of the threshold lines (shown in Figure 12) are determined.

To further illustrate the threshold estimation approach proposed for initial processing of online images, we consider the line shown in Figure 11(a). We note that the line is printed horizontally and hence, the image should be processed row-wise. If the average intensity of all pixels located in a row are considered and plotted against row number, the intensity profile shown in Figure 11(b) is obtained.

The intensity profile in Figure 11(b) indicates the approximate range of the line, overspray, and background. To find the coordinate and intensity of each zone more accurately, the first derivative of the intensity profile is estimated as shown in Figure 11(c). Based on this intensity profile, by definition, the line width zone (LW) is composed of those pixels whose intensity derivatives fall between the two global extrema. The
corresponding intensities of the extrema are used as reference values in the quantification of line width (Sec. 4.3.1). Also, their corresponding coordinates are referred to as the threshold lines (as shown in Figure 12).

Similarly, the overspray zone (OS) is composed of those pixels whose intensity derivatives fall between a smaller reference value near zero (referred to as the overspray threshold) and the negative global extremum (for the upper side), as well as between the positive global extremum and the overspray threshold (for the lower side). Finally, the background zone (BG) consists of those pixels whose intensity derivatives are almost zero on average.

### 4.2. Note on Processing Time Delay due to Image-based Quality Monitoring

We concede that there is a certain amount of delay associated with the proposed image-based line morphology monitoring approach. For processing the true-color images (from the CCD camera) the processing time is ~1 second. However, the time needed to translate the stage under the camera should be added to the overall processing time. The stage on the Optomec AJ-300 printer used in this study has a translation rate that can be adjusted as high as 100 mm/s, while the CCD camera is approximately 200 mm away from the center of the platen. Accordingly, such rapid translation would make the change in position from printing to inspecting be a few of seconds, at most.

In general, the image processing and acquisition delay is intrinsic to any putative quality control scheme, and is often characterized with a term called response time (and others, such as overshoot, jitter, nonlinearity, etc. in control theory [54, 55]). This response time can only be minimized but not eliminated. Nonetheless, the tradeoff between delay
time and quality control is inherent to the electronics manufacturing industry. We give the following practical examples from electronics packaging:

- The latest component placement machines are capable of working with rates exceeding 100,000 per hour. However, for precision components, automated vision-based inspection steps are initiated, which slow down the placement rate to 5,000 per hour.

- In placement of precision components, local fiducials are often used as guides in addition to global fiducials to minimize errors. The placement of components relative to local fiducials requires high fidelity imaging, which invariably leads to delay.

- In flux dipping process, handling precision components are inspected using image/vision acquisition systems, both before and after dipping. This inspection step adds a delay to the typical flux dipping process, but is done nonetheless to ensure component quality.

Likewise, the emphasis in AJP is on low volume-high, complexity-high value exotic devices (e.g., conformal antennae, flexible electronics etc.). Hence, in this work, accurate assessment of line quality is given precedence over delay.

4.3. Extracting the Line Morphology Features from an Image

4.3.1. Line Width (Lw): This quantifier is intended to ascertain the average width of a printed line. Once the optimal values of the threshold parameters have been calculated, their corresponding image row coordinates are obtained from the intensity profile and used as reference lines (called the threshold lines) in detection of the upper and lower edges of a printed line. Then, for each column array of pixels, the intensity of the individual pixels
is compared to that of the threshold lines. The line edge is found when the intensity of a pixel matches the threshold intensity.

Figure 12: Visual representation of the morphology quantifiers proposed in this study. 

- **Line Width (L\textsubscript{W})**: The average distance between the upper and lower edges (shown in green).
- **Line Density (L\textsubscript{\rho})**: The average intensity of all pixels constituting the line.
- **Edge Quality (LEQ)**: The inverse, average distance between each edge and its corresponding threshold line (shown as white dashed).
- **Overspray (LOS)**: The weighted, average distance between each overspray pixel and its corresponding line edge multiplied by the intensity.
- **Line Discontinuity (LDisc)**: The average number of failures in the edge detection. The image is read from top to bottom.

The algorithm is also applied to the other edge of the line. As shown in Figure 12, Line Width (L\textsubscript{W}) is defined as the difference between the detected upper and lower edges of the line (shown in green) for each column array of pixels averaged over the entire column space, mathematically expressed by Eq. (25),

$$L_{W} = \frac{\sum_{j=1}^{n} (L_{E(U,j)} - L_{E(L,j)})}{n}$$  \hspace{1cm} (25)

where, \(L_{E(U,j)}\) and \(L_{E(L,j)}\) are the upper and lower edges of the \(j\)th column, respectively; \(n\) is the total number of columns (representative of the line length).

**4.3.2. Line Density (L\textsubscript{\rho})**: This quantifier indicates how dense/close/packed deposited aerosol particles are. Mathematically, line density estimates the sparsity aspect of a line. It
can be also reflective of the degree of material coalescence after deposition. The density of a line is defined as the average of the grayscale intensity values of all pixels located between the detected upper and lower edges (i.e., located within the line area) as mathematically shown by Eq. (26). In order to offset the effect of illumination on the intensity of the pixels (depending on experimental conditions, camera settings, etc.), the background intensity is subtracted from the calculated line density.

$$L_p = \frac{\sum_{j=1}^{n} \sum_{i=L_{E(U,j)}}^{L_{E(L,j)}} I_{i,j}}{\sum_{j=1}^{n} (L_{E(U,j)} - L_{E(L,j)} + 1)} - \overline{T}_{Background}$$ \hspace{1cm} (26)

In the above equation, $L_{E(U,j)}$ and $L_{E(L,j)}$ are the upper and lower edges of the $j^{th}$ column, respectively; $I_{i,j}$ is the intensity of the pixel belonging to the $j^{th}$ column and $i^{th}$ row; $n$ is the total number of columns; $\overline{T}_{Background}$ is the background intensity (calculated based on the intensity of the pixels located outside the overspray zone).

**4.3.3. Edge Quality or Line Smoothness (LEQ):** This quantifier is representative of the evenness or smoothness of the edges of a line; it captures the degree of uniformity of material spread subsequent to deposition. Depending on the process parameters, as well as the interaction between the ink droplets and the substrate influenced by the surface energy, surface tension, and contact angle, the edges of a printed line may vary significantly. Although not pursued in this study, the smoothness of an edge can be further quantified using the wavelength and amplitude of the sinusoidal shape of the edge.

As mentioned above, based on the optimal values of the threshold parameters, two reference lines, i.e., the upper and lower threshold lines, are obtained. Referring to Figure 12, edge quality ($L_{EQ}$) is defined as the inverse of the average distance between the detected
line edges (green solid lines) and their corresponding threshold lines (white dashed lines). This quality quantifier is mathematically expressed by Eq. (27),

\[
L_{\text{EQ}} = \left( \frac{\sum_{j=1}^{n} \left( \left( L_{E_{(U,j)}} - Y_{UT} \right)^{2} + \left( L_{E_{(L,j)}} - Y_{LT} \right)^{2} \right)}{2n - 1} \right)^{-1}
\]

(27)

where, \( L_{E_{(U,j)}} \) and \( L_{E_{(L,j)}} \) are the upper and lower edges of the \( j \)th column, respectively; \( Y_{UT} \) and \( Y_{LT} \) are the row coordinates of the upper and lower threshold lines, respectively; \( n \) is the total number of columns. Relatively large values of Edge Quality (\( L_{\text{EQ}} \)) indicate that the edges of a line have a high degree of smoothness.

**4.3.4. Overspray Index (L\(_{\text{OS}}\))**: This quantifier measures overspray or spread of the aerosols deposited beyond the edges of a line. Overspray implies how efficiently aerosols are deposited during printing. In general, overspray is always unfavorable and should be minimized. Overspray is critical to the functional integrity of a printed device as large overspray may lead to short circuits and higher resistance \([18, 56, 57]\). Overspray Index (\( L_{\text{OS}} \)) is defined as the logarithmically-weighted distance between each overspray pixel and its corresponding line edge multiplied by the pixel intensity averaged over the entire overspray space shown in Figure 12, as mathematically expressed by Eq. (28),

\[
L_{\text{OS}} = \frac{\sum_{j=1}^{n} \left[ \sum_{i=1}^{L_{E_{(L,j)}} - 1} \left( l_{i,j} \times \ln \left( L_{E_{(L,j)}} - Y_{i,j} \right) \right) + \sum_{i=L_{E_{(U,j)}} + 1}^{m} \left( l_{i,j} \times \ln \left( Y_{i,j} - L_{E_{(U,j)}} \right) \right) \right]}{\sum_{j=1}^{n} \left[ L_{E_{(L,j)}} + \left( m - L_{E_{(U,j)}} + 1 \right) \right]}
\]

(28)

where, \( L_{E_{(U,j)}} \) and \( L_{E_{(L,j)}} \) are the upper and lower edges of the \( j \)th column, respectively; \( l_{i,j} \) and \( Y_{i,j} \) are the intensity and the coordinate of the pixel belonging to the \( j \)th column and \( i \)th row, respectively; the denominator terms define the entire overspray space. The
logarithmic function was employed to accentuate the adverse effect of distance for an overspray particle located far from the edge.

4.3.5. Line Discontinuity ($L_{\text{Disc}}$): This quantifier captures the condition of a printed line not being continuous. Generally, it is desirable to minimize line discontinuity, as it may result in an increase in line resistance. With reference to the approach used to detect the upper and lower edges of a line (in Line Width ($L_w$); Sec. 4.3.1), Line Discontinuity ($L_{\text{Disc}}$) is defined as the number of times that no edge is detected by the algorithm normalized by the total number of defect opportunities (i.e., the total number of image columns multiplied by 2) when scanning through the entire column space. This can be mathematically shown by Eq. (29),

$$L_{\text{Disc}} = \frac{\|F\|_0}{2 \times n}$$  \hspace{1cm} (29)

where, $\|F\|_0$ is the number of unsuccessful attempts (failures) in edge detection; $n$ is the total number of columns.

This quantifier assumes that the discontinuities of a line influence the edges. $L_{\text{Disc}}$ cannot capture a crack, for example, if the edges of a line are not affected. In such cases, other quantifiers, such as Line Density ($L_\rho$) and Fiedler Number ($\lambda_2$), should also be used as part of the characterization of line morphology.

4.3.6. Fiedler Number ($\lambda_2$): We use a graph-theoretic image quantifier called Fiedler number ($\lambda_2$) as a lumped measure of line morphology. Fiedler Number is representative of the algebraic connectivity of the graph resulting from a line. It can be a measure of not only surface morphology but also the internal structure and/or structural connectivity of a printed line [58]. Our previous publications give more details [58, 59] on how the Fiedler number for an image can be estimated.
We now detail experiments conducted to map the effect of three factors, i.e., ShGFR (Sec. 5.1), CGFR (Sec. 5.2), and $P_S$ (mentioned in Sec. 5.3) on line morphology using the aforementioned six quantifiers. The experimental line morphology trends observed by varying ShGFR and CGFR will be juxtaposed with CFD simulation results.

We have relegated the experimental results from varying $P_S$ to Sec. 5.3 owing to the inability to verify the (experimental) trends with the CFD model. This is because, the current CFD model is restricted due to the moving wall boundary condition.

5. Experimental Results and Verification with the CFD model

In this section, the effects of the sheath gas flow rate (ShGFR) and the carrier gas flow rate (CGFR) on line morphology is quantified using the six quantifiers defined in Sec. 4. Besides, we validate the 2D-CFD model with experimental data described in Sec. 3. The current work will be further verified in the future with a 3D-CFD model and multi-factor design of experiments as part of our future investigations.

5.1. Effect of the Sheath Gas Flow Rate (ShGFR) on Line Morphology

5.1.1. Experimental Observations: A randomized single-factor experiment was conducted with the sheath gas flow rate (ShGFR) varying from 40 to 140 sccm (in 20 unit increments). The carrier gas flow rate (CGFR) was kept fixed at 30 sccm. Silver nanoparticle lines (3 mm long each) were printed ten times in a single pass for each treatment combination on glossy photo paper at a speed of 1 mm/s, utilizing the ultrasonic atomizer (set at 550 mA) and a 150 µm nozzle. Also, the platen temperature and working distance (print standoff) were set at 40 °C and 3 mm, respectively. Using CCD camera,
digital images were captured from each line and, subsequently, the six morphological features (described in Sec. 4) were quantified (for each line).

As shown in Figure 13, when the ShGFR increases from 40 sccm to 80 sccm, the line width decreases. The line density as well as edge quality remain approximately unchanged, and the amount of overspray decreases slightly. However, as the ShGFR increases further from 80 sccm to 140 sccm, not only do both line density and edge quality decrease dramatically, but also the amount of overspray increases significantly. Hence, the ShGFR of 100 sccm can be considered as the critical point of flow instability.

![Figure 13](image)

**Figure 13**: The effect of the sheath gas flow rate (ShGFR) on line morphology. The carrier gas flow rate (CGFR) and print speed (Ps) were fixed at 30 sccm and 1 mm/s, respectively.

5.1.2. Characterization of the Line Morphology: Referring to Figure 14, the following inferences can be made regarding the line morphology characterized using the six quantifiers developed in Sec. 4. Line width and overspray thresholds of 0.30 and 0.60 (in the range of [0-1]) were set, respectively, as part of the algorithm employed for image processing.

1. The Line Width (Lw) decreases almost linearly as the ShGFR increases, confirming the trend observed visually (Figure 13).
(2) The Line Density ($L_\rho$) remains almost unchanged as the ShGFR is in the range of 40-100 sccm. However, as the ShGFR is increased further, the line density decreases dramatically to a minimum due to the transition in the stability of the flow regime resulting in overspray formation. This experiment also indicated the process condition where the highest amount of density is achieved, i.e., 60 sccm. Lines with high density are desirable because they typically have lower electrical resistance.

(3) Likewise, the Edge Quality/Smoothness ($L_{EQ}$) indicates that the optimal operation for the ShGFR is in the range of 40-100 sccm where the edge smoothness quantifier approximately remains unchanged. However, once the ShGFR exceeds the critical point of 100 sccm, the edge quality drops significantly.

(4) Overspray ($L_{OS}$) decreases to a minimum as the ShGFR is increased from 40-80 sccm. This is because, the aerosol flow is collimated. An optimally collimated flow also leads to well-defined edges. However, increasing the ShGFR beyond the stability threshold results in overspray.

(5) Line Discontinuity ($L_{Disc}$) signalizes out-of-the-window conditions (i.e., when the ShGFR is 120 and 140 sccm) where either no edge is detected or edges are detected sporadically along the length implying the line is disconnected. Such a line has a large degree of resistance attributed to the poor density and high overspray.

(6) The trend in Fielder number ($\lambda_2$) indicates that the line printed at the ShGFR of 60 sccm has the highest internal connectivity and low overspray with density and edge quality being in the optimum range. Once the flow exceeds the critical point of flow stability (ShGFR > 100 sccm, corresponding to a focusing ratio, $F_R \sim 3$), the least amount of internal connectivity is obtained where the edges are disconnected.
These visually corroborated experimental observations (Figure 14) lead to the following two inferences: (i) the six morphology quantifiers are indicative of line quality; and (ii) an optimal process window can be established with the ShGFR being in the range of $60 < \text{ShGFR} < 100$ sccm, the CGFR being at 30 sccm, and $P_S$ being at 1 mm/s. This corresponds to a focusing ratio ($F_R$) in the region of 2 to 3. This range of $F_R$ corroborates with the work of Mahajan et al. [18].

**5.1.3. Validation with Offline Measurements:** In order to validate the results and the trends observed in the online experiment (Figure 14), an identical and separate single-factor experiment was repeated where the printed lines were examined off the experimental testbed using an optical microscope. The same characterization method (delineated in Sec. 4) was employed to process the images and quantify the morphological features.

For brevity, Figure 15 demonstrates a comparison between the online and offline results for three quantifiers of line morphology, i.e., Line Density ($L_\rho$), Overspray ($L_{OS}$), and
Fielder number ($\lambda_2$). The morphological features extracted from the online and offline images show identical trends when the ShGFR is varied from 40 sccm to 140 sccm. In other words, compared to the traditional offline method, the proposed online characterization approach is capable of quantifying and capturing the variation in line quality. This is verified by the Pearson’s correlation coefficient of approximately 95% between the offline and online results.

![Figure 15: A comparison between the online and offline experimental results vs. ShGFR, signifying the consistency between the two methods. The error bars are ($\pm 1\sigma / \sqrt{n}$) long where $n$ equals the number of replications (10). Capturing the same trend for each morphology attribute, the online and offline quantifiers were plotted on two axis to offset the difference arising from different image properties. The online measurements are mapped to the primary ordinate axis, and the offline measurements to the secondary ordinate axis.](image)

5.1.4. Validation with Pneumatic Atomization: A randomized, single-factor experiment using the pneumatic atomizer was conducted. This was done to test the hypothesis that the trends in the line morphology features vs. ShGFR are independent of the type of atomizer used. In this experiment, the atomization and exhaust flow rates were set at 550 and 540 sccm, respectively. The results are shown in Figure 16. The rest of the experimental conditions (except the type of ink and substrate) are identical to the tests hitherto described using the ultrasonic atomizer.
Figure 16: The changes in line morphology as a function of ShGFR, employing the pneumatic atomizer. The lines are of Paru PG-007 Ag ink printed on a Ube UPLEX-75S polyimide film.

Juxtaposing Figure 13 and Figure 16, it is evident that the pneumatic- and ultrasonic-based morphology features are visually similar; note overspray beyond the ShGFR of 100 sccm in both cases. Demonstrated in Figure 17, a quantitative comparison between the lines indicates the similarity of trends vs. ShGFR for both atomization techniques. Pearson’s correlation coefficient was assessed to be in the range of 70% to 95% for the trends shown in Figure 17. The slight difference probably stems from the type of ink and substrate.

Figure 17: A comparison between the ultrasonic and pneumatic experimental results of line morphology vs. ShGFR, corroborating the consistency of trends between the two atomization techniques. The error bars are \((\pm 1\sigma/\sqrt{n})\) long where \(n (=10)\) equals the number of replications.

5.1.5. Validation with CFD Simulations: The experimental data (in Sec. 5.1.2) showed that as the ShGFR increases, the line width, which is hypothesized to be proportional to the width of the aerosol jet flow, decreases almost linearly to a minimum. Additionally, it is hypothesized that the sheath gas flow interacts initially with the carrier gas flow and as a result influences the aerosol deposition. Proper material deposition results in lines
showing a large degree of internal connectivity, represented by Fiedler number. We also hypothesize that beyond the critical point of 100 sccm, the aerosol jet becomes hydrodynamically instable leading to formation of overspray. This hypotheses are tested via simulation of the deposition process using the 2D-CFD model described in Sec. 3. The results are shown in Figure 18.

Figure 18: (a) The influence of ShGFR on line width (left axis) and on the aerosol jet width (right axis); (b) The influence of ShGFR on the carrier flow deposition spread (right axis) and on the internal connectivity of the lines, represented by Fiedler number (left axis). (c) Plot of the Reynolds number vs. ShGFR indicating that the aerosol flow in the combination chamber becomes hydrodynamically instable when the ShGFR ≥ 100 sccm (i.e., Re ~ 950) [60]. The error bars are (±1σ/√n) long where n equals the number of replications (10). An empirical geometry factor of 6 was used to scale the 2D-CFD simulation to the 3D experimental data.

Figure 18(a) reveals that both line width (left axis) and aerosol jet width (right axis) are influenced by the ShGFR almost similarly, corroborating the hypothesis that the line width is proportional to the width of the aerosol jet flow. The difference between the two trends stems from the 2D modeling of the process. A 3D-CFD model could help alleviate this discrepancy. An empirical geometry factor of 6 was used to scale the 2D simulation in accordance with the experimental data. Figure 18(b) shows that the ShGF significantly influences the CGF deposition spread (right axis) and, consequently, the internal connectivity of the resulting line represented by the complement of Fiedler number (left axis). In other words, the interaction between the two gas flows influences the particle deposition distribution and, as a result, the final morphology of the line.
A stable flow regime in the combination chamber, where the sheath gas is introduced as a sheet-like flow, prevents formation of overspray and allows unperturbed deposition of aerosols. As shown in Figure 19, at high ShGFRs (≥ 100 sccm), due to the maximum pressure limit, which is a function of the nozzle geometry, pressure builds up noticeably leading to uneven aerosol deposition and, as a result, poor line morphology. This model-derived observation is corroborated by the empirical results delineated in Sec. 5.1.2, where it was shown that stable and collimated aerosol deposition can be obtained when the ShGFR is less than 100 sccm with CGFR at 30 sccm and $P_S = 1$ mm/s. Furthermore, corroborated by the experimental observations, the degree of flow collimation (represented by the particle deposition profile in Figure 19) increases with the ShGFR, as observed experimentally in Figure 13 and Figure 14.
Although not investigated in this study, aerosol deposition distribution is affected not only by the fluid’s inertia as well as surface tension represented by Weber number, but also by the contact angle and wetting mechanism. For example, an improper contact angle leads to excessive ink dispersion (after deposition) and distorted line edges [38]. Furthermore, the jet length and the nozzle diameter both are consequential to the flow stability. The current 2D model is primarily concerned with the aerodynamics of the aerosol flow in the deposition head.

The pressure profiles of the aerosol flow at different levels of ShGFR (40 sccm - 140 sccm) are shown in Figure 20(a-f). The CGFR was fixed at 30 sccm. As the ShGFR increases, the flow pressure in the combination chamber increases proportionately from
around 286 Pa (for the ShGFR of 40 sccm) to 1155 Pa (for the ShGFR of 140 sccm). It is evident that pressure buildup in the chamber starts at the ShGFR of 80 sccm onwards. Therefore, it can be concluded that the deposition head has a maximum pressure limit of approximately 622 Pa.

Figure 20: The influence of ShGFR on the flow pressure profile as well as on the trajectory of particles in the combination chamber. The ShGFR of 80 sccm seems to be the onset of pressure buildup in the chamber. Hence, the maximum pressure limit can be considered approximately 622 Pa. In this simulation, the carrier gas flow rate (CGFR) was set at 30 sccm.

5.2. Effect of the Carrier Gas Flow Rate (CGFR) on Line Morphology

5.2.1. Experimental Observations: A single-factor experiment was designed to investigate the influence of the CGFR (representative of the rate of aerosol transfer) on the main components of line morphology. The CGFR was varied in a randomized fashion from 20 to 45 sccm (with 5 sccm increments). In this experiment, both the sheath gas flow rate (ShGFR) and print speed (Ps) were maintained at 60 sccm and 1 mm/s, respectively. These
two values were selected based on the results obtained from the experiments of ShGFR (Sec. 5.1) and $P_s$ (see Sec. 5.3). Similar to these experiments, silver nanoparticle lines (3 mm long each) were printed 10 times in a single pass for each CGFR level.

The effect of the CGFR on line morphology is visually shown in Figure 21. As the CGFR increases, the edge quality/smoothness improves, remains steady between 30-35 sccm, and deteriorates at 40 sccm onwards. The highest level of edge regularity was observed at the CGFR of 30 sccm. No line discontinuities were observed in this experiment.

![Figure 21](image)

**Figure 21**: (a-f) The effect of the carrier gas flow rate (CGFR) on line morphology, both the sheath gas flow rate (ShGFR) and print speed ($P_s$) were fixed at 60 sccm and 1 mm/s, respectively.

The line width increases proportionately with the CGFR. For the CGFRs of 30, 35, and 40 sccm, the line width remains approximately unchanged because the aerosol flow reaches its maximum collimation (focusing) capacity. Comparatively, in the range of 20-25 sccm, the carrier flow is lean in aerosol, and the flow collimation remains undisturbed. This corresponds to a focusing ratio ($F_R$) of $\sim 2$ to $3$. In contrast, at the CGFR of 45 sccm onwards no more material can be accommodated in the flow without perturbation, as a result both line width and overspray increase dramatically.
**Collimation Capacity** is defined as the ratio of the original width (or diameter) of an unfocused aerosol flow to the minimum width (or diameter) achievable via aerodynamic focusing. Collimation capacity is a function of nozzle geometry as well as focusing ratio ($F_R$). At the CGFRs of 20 and 25 sccm (where the $F_R$ is 3 and 2.4, respectively), the sheath flow is dominant over the aerosol flow. In this region, any increase in the rate of material transfer (i.e., the CGFR) leads to an increase in the line width. However, if the CGFR is increased further, the flow reaches the maximum collimation capacity where any increase in the flow rate no longer results in an increase in the line width. Nevertheless, based on the law of conservation of mass, this leads to an increase in the line thickness. In this region where the sheath flow is still dominant, the focusing ratio is in the range of 1.5-2. Ultimately, when the $F_R$ drops below 1.5, the aerosol flow momentum disturbs the collimation boundary layer. This phenomenon results in unfocused material deposition and, consequently, overspray. **Collimation Boundary Layer** is defined as the shear layer created due to the velocity gradient between the sheath and aerosol flows.

### 5.2.2. Characterization of the Line Morphology:
Referring to Figure 22, the following inferences can be made regarding the line morphology characterized using the six quantifiers developed in Sec. 4. Updated line width and overspray thresholds of 0.40 and 0.80 (in the range of [0-1]) were set, respectively, as part of the algorithm employed for image processing.

1. Line Width ($L_W$) increases with the CGFR as observed visually (in Sec. 5.2.1).
2. The Line Density ($L_{\rho}$) stabilizes when the aerosol flow reaches the maximum collimation capacity, implying that any further increase in the flow rate results in an increase in the line thickness and, as a result, the line density remains unaffected.
(3) The Edge Quality (LEQ) shows edges with the highest level of smoothness are obtained when the CGFR is between 25-30 sccm.

(4) Overspray (LOS) increases almost monotonously with the CGFR, depending on the flow collimation regime, represented by the focusing ratio (FR).

(5) Line Discontinuity (L_Disc) showed that all lines are printed with zero degrees of discontinuity. Hence, this morphology quantifier is not shown in Figure 22.

(6) Fielder number ($\lambda_2$) is a graph-theoretic morphology quantifier, utilized to assess the structural connectivity of the printed lines. The maximum structural connectivity is achieved at the CGFR of 30 sccm ($F_R \sim 2$).

![Graphs showing line morphology features as functions of CGFR.]

Figure 22: The main features of the line morphology as a function of the CGFR captured using the quantifiers developed in this study. The error bars are ($\pm 1 \sigma/\sqrt{n}$) long where $n$ equals the number of replications (10). The secondary abscissa tracks the corresponding carrier gas flow pressure (CGFP, Pa)

5.2.3. Validation with Offline Measurements: Following the previous procedure for offline validation, Figure 23 is a comparison between the online and offline
characterization of line morphology. The features extracted from the online and offline images show similar trends when the CGFR is varied from 20 sccm to 45 sccm.

![Graph showing line width and CGFR comparison](image)

Figure 23: A comparison between the online and offline experimental results vs. the CGFR, signifying the consistency between the two methods. The error bars are \(\pm \frac{1}{\sqrt{n}}\) long where \(n\) equals the number of replications (10). Capturing the same trend for each morphology attribute, the online and offline quantifiers were plotted on two axis to offset the difference arising from different image properties.

### 5.2.4. Validation with CFD Simulations:

The experimental data (in Sec. 5.2.2) showed that as the CGFR increases, the flow gains more momentum diminishing the degree of collimation (exerted by the sheath flow) and, as a result, the line width increases. In general, the degree of collimation can be defined as the ratio of the width (or diameter) of an unfocused aerosol flow (e.g., at zero ShGFR) to the width (or diameter) when the flow is focused. This quantity relatively indicates the effectiveness of the sheath gas flow vis-à-vis the focusing of the aerosol flow. In this section, to corroborate these observations, we first test the hypothesis that the line width is proportional to the width of the aerosol jet flow via simulation of the deposition process using the 2D-CFD model described in Sec. 3.

As shown in Figure 24, there is close agreement between the experimental and CFD-derived trends substantiating the hypothesis that the line width (left axis) is proportional to the width of the aerosol jet flow (right axis).
Figure 24: The influence of the CGFR on the line width (left axis) and on the aerosol jet width (right axis). The error bars are $(\pm 1\sigma/\sqrt{n})$ long where $n$ equals the number of replications (10). The objective was to show the model could capture the same trend as the experiment; a geometry factor of 6 was used for scaling the CFD simulation results to the experimental observations.

The flow velocity profile obtained from the CFD model is shown in Figure 25, which corroborates that the (central) aerosol flow gains larger momentum when the CGFR is increased. The increase of momentum enables the particles to overcome the collimation force exerted by the sheath flow. As a result, the focusing ratio ($F_R$) decreases, and less collimated, wider lines (still smooth and good in quality) are obtained, as observed experimentally in Figure 22(a). In other words, the degree of collimation (represented by the particle deposition profile) decreases with increasing the CGFR. However, if the CGFR exceeds the critical point of stability, overspray forms as observed experimentally in Sec. 5.2.2.
Figure 25: The influence of increasing CGFR on the flow velocity profile and on the particle trajectory (a)-(b): in the combination chamber; (c)-(d) during the deposition process. In this simulation, the sheath gas flow rate (ShGFR) was set at 60 sccm.

5.3. Effect of Print Speed (Ps) on Line Morphology

5.3.1. Experimental Observations: As visually shown in Figure 26, a single-factor experiment was designed to investigate the influence of Print Speed (Ps), also known as translation speed or stage speed (as one of the machine parameters compared to the flow parameters delineated in Sec. 5.1 and 5.2), on line morphology. The P_s was varied in a randomized fashion from 0.5 mm/s to 5 mm/s (with 0.5 unit increments). In this experiment, both sheath and carrier gas flow rates were kept at 60 sccm and 30 sccm, respectively. These two values were selected based on the results of the experiments
conducted on the ShGFR (Sec. 5.1) and CGFR (Sec. 5.2). Silver nanoparticle lines (3 mm long each) were printed 10 times in a single pass for each $P_S$ level.

![Figure 26: The influence of Print Speed ($P_S$) on line morphology. The edge quality and line width are more significantly affected than the other morphology features (developed in Sec. 4).](image)

An initial, visual investigation of the line morphology (shown in Figure 26) reveals the fact that the edge quality/smoothness is influenced by print speed most significantly (compared to the other features of line morphology developed in Sec. 4). As the print speed increases, the edge quality deteriorates markedly; similarly but less prominently, the line width decreases.

The physics behind the aerosol flow deposition under the influence of print speed can be captured by linear mass density (mass per unit length) as expressed by Eq. (30),

$$\lambda_m = \frac{\omega_p \bar{\rho}_s \phi_s \dot{Q} \eta}{60 \nu}$$

(30)

where $\lambda_m$ is the linear mass density of a printed line (g/mm), $\omega_p$ is the mass fraction of the solid particles loaded in the ink, $\bar{\rho}_s$ is the average density of the solvent system (g/mL), $\phi_s$ is the volume fraction of the solvent in the aerosol mixture, $\dot{Q}$ is the volumetric flow rate of the aerosol or carrier gas flow (sccm), $\eta$ is the aerosol transport and deposition efficiency, and $\nu$ is the print speed (mm/s).

The above equation connects the aerosol volumetric flow rate with print speed. At a constant volumetric flow rate ($\dot{Q}$), there is a nonlinear relationship between the linear mass
density ($\lambda_m$) and print speed ($n$) implying that lines with poor quality are printed in terms of density and discontinuity as the print speed increases inordinately. This phenomenon is visually observed in Figure 26. The linear mass density is representative of the ratio of the material deposition rate to the translation rate. Once the print speed exceeds a critical value, the deposition process is no longer material rate-dominated but print speed-dominated. In the print speed-dominated region, there is no proper material coalescence after deposition which adversely influences the edge quality, line density, as well as line discontinuity.

5.3.2. Characterization of the Line Morphology: According to Eq. (30) an increase in the print speed results in less material deposition per unit length. Referring to Figure 27, the following inferences can be made regarding the line morphology.

1. Line Width (LW) decreases sharply in the material-dominated region ($P_S \leq 1.5$ mm/s) and, then, approximately levels off in the print speed-dominated region. With regard to the ink properties as well as the ink-substrate interaction, it seems the high rate of translation in the latter region prevents the deposited aerosol from proper coalescence. Hence, the print speed of 1.5 mm/s can be considered as a critical point of translation beyond which the line linear density decreases significantly.

2. Edge Quality ($L_{EQ}$) is optimum when $P_S \leq 1.5$ mm/s and, then, deteriorates significantly once the rate of translation becomes dominant.

3. The highest level of Line Density ($L_\rho$) is obtained in the material rate-dominated region where the phenomena of aerosol coalescence and spread determine the final morphology of the line governed by surface tension and surface energy, respectively.

4. Overspray ($L_{OS}$) decreases exponentially, remaining approximately unchanged in the print speed-dominated region.
It can be concluded that the Print Speed (Ps) parameter should be maintained around the critical value of 1.5 mm/s for the process to remain within the optimal operability window.

![Graphs showing line morphology features](image)

**Figure 27:** The line morphology features captured using the quantifiers developed in this study. The error bars are $(\pm 1\sigma/\sqrt{n})$ long where $n$ equals the number of replications ($n = 10$).

### 6. Conclusions and Future Work

An Optomec AJ-300 aerosol jet printer was instrumented with a CCD camera for online monitoring of line quality. Various image-based quantifiers were subsequently defined to capture various attributes of line morphology. They include: line width ($L_w$), line density ($L_\rho$), Edge Quality/Smoothness ($L_{EQ}$), Overspray Index ($L_{OS}$), Line Discontinuity ($L_{Disc}$), and Fiedler Number ($\lambda_2$).

Experiments were conducted by varying main process parameters, i.e., sheath gas flow rate (ShGFR), carrier gas flow rate (CGFR), and print speed (PS). Line morphology was
characterized using the six quantifiers, based on which an optimal process window was ascertained. From our experimental investigation, the optimum focusing ratio ($F_R$) and print speed ($P_S$) are 2.7 and 1 mm/s, respectively. The accuracy of these quantifiers was verified with offline characterization results. The correlation between the online and offline measurements was assessed to be approximately 95%. The quantifiers were able to capture the experimental trends of line morphology irrespective of the type of atomizer (pneumatic or ultrasonic); the method is therefore applicable to both ultrasonic and pneumatic atomization (nebulization) techniques.

In addition, a 2D computational fluid dynamics (CFD) model was forwarded to explain the underlying aerodynamics of the AJP process. The CFD model explains the underlying reason for the trends observed in line morphology contingent on the carrier and sheath gas flow rates. The image data and subsequent analysis can serve to identify process drifts, and the CFD model provides a basis for adjustment to correct the process drift.

As part of our future work, the effects of other process parameters (such as atomization current, nozzle diameter, and platen temperature) will be investigated through a multifactor design of experiment. The CFD model will be augmented with more variables. This will allow closer agreement between experimental and theoretical results. Besides, the proposed monitoring scheme will be extended to be used for closed-loop control.

To quantify line thickness, we will use approaches, based on the concept of shape-from-shading (see Chapter 3) as well as stereomicroscopy. Regarding the latter approach, a setup is shown in Figure 28. In this technique, two images are taken at slightly different perspectives (Figure 28(b) and (c)) using calibrated stereo cameras (located off the testbed), and analyzed to extract the height of the structure based on the differences in perspective.
Stereo-photometry has been implemented in literature [61, 62]. This method has advantages of a large standoff distance and is more robust to rough or irregular depositions. The shape and thickness of the structures can be determined accurately offline, using methods such as stylus profilometry and/or white light interferometric microscopy, and used to calibrate and verify the measurement system.

Figure 28: The proposed stereomicroscopy-based approach which will be used to quantify line thickness in our future work. (a) A picture of the experimental setup equipped with a stereomicroscope; (b) and (c) the left and right views of a printed line, respectively. The line thickness is quantified based on the two perspective views.
References


1. Introduction

1.1 Objective

The goal of this research is the online monitoring of functional electrical properties, e.g., resistance and capacitance in printed flexible electronic devices made using Aerosol Jet Printing (AJP) process [1] [2]. In pursuit of this goal, the objective of this work is to recover the cross-sectional profile of AJP-deposited electronic traces (lines) using Shape-from-Shading (SfS) analysis of online images acquired by an in-process CCD camera integrated into an Optomec AJ-300 machine. An accurate characterization of the cross-section is essential for monitoring device resistance and other functional properties. For instance, as per Ohm’s law, the electrical resistance of a conductor is inversely proportional to its cross-sectional area. The central hypothesis is that the electrical resistance of an AJP-deposited line estimated online and in situ from its SfS-derived cross-sectional area is within 20% of its offline measurement.

This research is the first to report the application of SfS techniques for in situ, online estimation of line resistance; it addresses the following open questions and thus paves the way for certify-as-you build quality assurance in AJP [3]:

- What is the effect of process parameters, such as sheath gas flow rate, on line cross-section?

- What approach is required for in situ, non-contact measurement of line cross-sectional area, and subsequently, online prediction of the electrical resistance?
1.2 Motivation

Aerosol jet printing (AJP) is a droplet-based, direct write additive manufacturing (AM) technique that has emerged as the process of choice for making flexible hybrid electronic devices, conformal antennae, organic photovoltaic devices, among others [1] [2]. Figure 1(a)-(b) exemplify flexible electronic devices made using AJP at Binghamton University (SUNY). Despite their far-reaching potential, AJP processed devices are currently relegated to prototype-demonstrator roles due to poor repeatability and reliability of material deposition as exemplified in Figure 1(c-f). Poor deposition consistency has a deleterious impact on the functional electrical properties, e.g., resistance, of a printed device [4]. Besides, the AJP process tends to drift over time due to changes in material properties. Hence, an empirically established and stationary process window may not suffice for functional quality assurance [5]. Consequently, online monitoring of electrical properties is urgently needed to ensure commercial viability of AJP-processed devices.

However, the online monitoring of functional electrical properties of AJP-printed devices is not possible without instantaneous estimation of the cross-sectional area. Characterization of line cross-section with offline approaches, such as white light interferometry and stylus profilometry, introduces an inordinate amount of delay. Furthermore, contact-based techniques are liable to damage the soft, un-sintered deposits. Consequently, in situ, non-contact measurement techniques is required for online estimation of the line cross-sectional area in AJP.
Figure 1: (a-b) Examples of AJP-printed electronics fabricated at Binghamton University (SUNY); (a) An antenna printed on a flexible glass substrate; (b) Silver interdigitated electrodes (IDEs) printed on flexible polyimide. (c-h) AJP lines from the authors experiments exemplifying various line characteristics (Source: [5]).

From a broader perceptive, this work fits seamlessly within the context of certify-as-you-build quality assurance paradigm in AM, which was described by Huang et al. in a recent NSF-sponsored workshop [3]. The aim of the certify-as-you-build paradigm is to guarantee the functional performance of AM parts by integrating physical models, in-process sensing, and data analytics. This work is focused on the last two aspects. Precedents are evident in several recent publications; for instance, Rao et al. [6] proposed a heterogeneous sensor data fusion approach to identify potential failure modes as well as detect the onset of anomalies in a real-time manner in fused filament fabrication (FFF) process. In addition, they introduced a spectral graph theory (SGT) approach for classifying the dimensional integrity of AM components [7]. Similarly, two image-based control paradigms were introduced by Arciniegas et al. [8] for closed-loop control of surface defects in electrophotographic (EP3D) AM process. Specific to AJP, the authors have
recently published results combining computational fluid dynamics modeling and image-based monitoring of deposit quality [5, 9]. In a similar vein, Sun et al. presented statistical models to predict line quality in AJP using offline microscopy images [10].

This work is a direct continuation of the authors’ recent research in AJP [5, 9]. The prior work defined six image-based quantifiers of line quality. These quantifiers were primarily concerned with 2D characteristics of a line including: (1) line width, (2) line density, (3) edge quality, (4) overspray, (5) line discontinuity, and (6) line connectivity. However, line cross-sectional area was not estimated in our previous work. This gap is addressed in this research using SfS image processing [11-13]. To reiterate: online monitoring of line resistance is not possible without estimating line cross-sectional area. Future extensions of this work will integrate process modeling, 2D quantifiers, and cross-sectional area estimates for more accurate prediction of line electrical properties given AJP process parameters [5, 9].

The rest of the chapter is organized as follows. In Sec. 2, the image-based reconstruction of line topology is discussed. This includes a review of shape-from-shading (SfS) techniques, i.e., Horn’s [11, 13], Pentland’s [14], and Shah’s [15] methods, followed by case studies with synthetically generated basic geometries to contrast the performance of each technique. Shah’s method was qualitatively evaluated to have the best performance. In Sec. 3, the AJP process and the experimental procedure are discussed. Subsequently, in Sec. 4, the Shah’s method is used for online recovery of the topology of AJP-printed lines deposited on a silicon substrate by varying one process parameter - sheath gas flow rate (ShGFR). This section also quantifies the effect of the ShGFR on line cross-section and
hence line resistance, and further verifies the SfS estimates of cross-sectional area with offline measurements.

2. Image-Based Reconstruction of Line Topology

2.1 Overview of Shape-from-Shading Principles (SfS)

The aim of this section is to explain the concept of Shape-from-Shading (SfS) analysis [13]; and elucidate its use in reconstructing the cross-sectional profile of AJP-printed lines given a gray-scale image captured using a coaxial CCD camera installed on an Optomec AJ-300 setup (shown in Sec. 3, Figure 10). Image intensity captured by a camera from a surface depends on: (i) illumination, characterized by direction, position, as well as spectral energy distribution; (ii) surface reflectivity (albedo), which is a measure of the interaction of the surface with incident light; (iii) surface geometry (which is the unknown variable in SfS); and (iv) camera direction.

Figure 2 encapsulates the application of SfS to AJP, as summarized therein. The SfS problem is the inverse of acquiring an image with a camera – SfS attempts to recover the 3D geometry of a subject given an image, its illumination direction, and surface reflectivity. The SfS problem is an intrinsically underdetermined problem, i.e., there are more parameters than equations [11, 16, 17]. Consequently, certain assumptions are needed to simplify the SfS equations to a well-posed problem so that a unique solution can be obtained. These assumptions are discussed in Sec. 2.2.

![Figure 2: The shape-from-shading (SfS) problem: reconstruction of the 3D topology of an AJP-deposited electronic trace based on the intensity gradient information captured from the surface by an image [45].](image-url)
The topology of a surface can be recovered if the normal vector at each point on a surface is known [11]. With reference to Figure 3, the normal vector, \( \mathbf{n} \), of a surface, \( Z(x,y) \), in Cartesian coordinates is defined as follows [11, 13]:

\[
\mathbf{n} = (n_x, n_y, n_z) = [\sin \alpha_n \cos \beta_n, \sin \alpha_n \sin \beta_n \cos \alpha_n]^T
\]

(1)

where, \( \alpha_n \) and \( \beta_n \) are the tilt and slant angles, respectively. We note that \( \alpha_n \in \left[0, \frac{\pi}{2}\right] \) and \( \beta_n \in [0, 2\pi] \). As shown in Figure 3, the tilt angle \( (\alpha_n) \) is the angle between the \( x \)-axis and the projection of the normal vector \( \mathbf{n} \) on the \( xy \) plane; the slant angle \( (\beta_n) \) is the angle between the \( z \)-axis and the normal vector \( \mathbf{n} \). The surface gradient, \( \nabla Z = (p, q)^T \), representing the orientation of the surface, is given by Eq. (2) and Eq. (3), respectively. The surface normal vector, \( \mathbf{n} \), is written in terms of the surface gradient components, i.e., \( p \) and \( q \), in Eq. (4) [11, 13].

\[
p = \frac{\partial Z(x,y)}{\partial x}
\]

(2)

\[
q = \frac{\partial Z(x,y)}{\partial y}
\]

(3)

\[
\mathbf{n} = \frac{[-p, -q, 1]^T}{\sqrt{1 + p^2 + q^2}}
\]

(4)

The illumination direction, \( \mathbf{l} \), of a light source (for example, the LED ring light in the experimental setup, shown in Figure 10) is defined based on the tilt and slant angles of illumination, i.e., \( \alpha_1 \) and \( \beta_1 \), respectively, as mathematically shown by Eq. (5). It is noted that \( \alpha_1 \in \left[0, \frac{\pi}{2}\right] \) and \( \beta_1 \in [0, 2\pi] \).

\[
\mathbf{l} = [\sin \alpha_1 \cos \beta_1, \sin \alpha_1 \sin \beta_1, \cos \alpha_1]^T
\]

(5)

Based on the assumptions delineated in Sec. 2.2, the reflectance or intensity \( (R) \) of a surface in the gradient space is given by Eq. (6), where \( \rho \) is the surface reflectivity (albedo) [11, 13]. By definition, the surface reflectance, \( R(p,q) \), and image irradiance, \( E(x,y) \), correspond mathematically and, hence, have the same form.
The surface reflectance equation (i.e., Eq. (6)), which is essentially a nonlinear partial differential equation (PDE), constitutes the basis for all SfS methods used in this work, namely, Horn’s [11, 13], Pentland’s [14], and Shah’s [15]). Given boundary conditions, solving this equation requires estimation of illumination direction, \( I \), as well as surface reflectivity (albedo), \( \rho \).

\[
E(x, y) = R(p, q) = \rho I^T \mathbf{n} = \frac{\rho I^T [-p, -q, 1]^T}{\sqrt{1 + p^2 + q^2}} \tag{6}
\]

The surface reflectance equation (i.e., Eq. (6)), which is essentially a nonlinear partial differential equation (PDE), constitutes the basis for all SfS methods used in this work, namely, Horn’s [11, 13], Pentland’s [14], and Shah’s [15]). Given boundary conditions, solving this equation requires estimation of illumination direction, \( I \), as well as surface reflectivity (albedo), \( \rho \).

Figure 3: The captured topology of a surface in form of an image depends on: (i) illumination direction \( I \), (ii) surface reflectivity \( \rho \), and (iii) camera direction \( V \) [11, 16, 17]. The surface at each point is represented by its normal vector \( \mathbf{n} \). \( \alpha \) and \( \beta \) are the tilt and slant angles, respectively.

2.2 Assumptions

In SfS, the fundamental equations are the image irradiance equation (Eq. (6)) and the surface recovery equation (see Eq. (22)). The key parameters are surface reflectivity or albedo \( \rho \), illumination direction \( I \), surface normals \( \mathbf{n} \) expressed in terms of surface gradients \( p \) and \( q \), and surface profile \( Z \). As mentioned in Sec. 2.1, the SfS problem is inherently ill-posed. To address this challenge, the following assumptions are made:

1. The camera has orthographic projection (vs. perspective), meaning a point in 3D space can be projected to a new point in 2D space with consideration of a magnification.
factor. We note that in the perspective projection, a point in 3D space is projected to a new point in 2D space as a function of its z-coordinate.

(2) The z-axis of the camera represents the optical axis.

(3) The surface is diffuse or Lambertian; Lambertian surfaces reflect light uniformly in all directions vs. specular surfaces where reflectively is non-zero only at the reflected angle [11, 13].

(4) The surface is not self-shadowing. Hence, image intensity or surface reflectance at a point is a function of the angle between illumination direction (I) and the surface normal (n), as well as the surface reflectivity (ρ).

(5) Both illumination direction and surface reflectivity (albedo) are assumed to be constant.

2.3 Estimation of Illumination Direction (I) and Surface Albedo (ρ)

The first step to any SfS approach is the estimation of illumination direction, I, and surface reflectivity (albedo), ρ. To estimate these, it is assumed that the normal vectors of a 3D surface are all uniformly distributed [11]. Referring to Eq. (1), the distribution of the surface normals with regard to the tilt (αn) and slant (βn) angles can be expressed, as follows.

\[ f(\beta_n, \alpha_n) = \frac{\cos \alpha_n}{2\pi} \]  \hspace{1cm} (7)

Subsequently, to find illumination direction and surface albedo, the image irradiance equation (already given by Eq. (6)) are rewritten in terms of the tilt and slant angles (i.e., Eq. (8)). Eq. (8) is obtained by substituting Eq. (1) and Eq. (5) in Eq. (6). Then, the first and second moments are taken, as shown by Eq. (9) and Eq. (10), respectively [11, 13].

\[ E(\beta_n, \alpha_n) = \rho (\sin \alpha_I \cos \beta_I \sin \alpha_n \cos \beta_n + \sin \alpha_I \sin \beta_I \sin \alpha_n \sin \beta_n + \cos \alpha_I \cos \alpha_n) \]  \hspace{1cm} (8)
Given a gray-scale image, with consideration of the assumptions mentioned in Sec. 2.2, estimates of surface albedo, $\hat{\rho}$, as well as illumination direction, $\hat{I}$, are then obtained using Eq. (11) and (12), respectively.

$$\mu_1 = \int_0^{2\pi} \int_0^{\pi/2} E(\beta_n, \alpha_n) f(\beta_n, \alpha_n) \, d\alpha_n \, d\beta_n = \frac{\pi}{4} \rho \cos \alpha_1 \quad (9)$$

$$\mu_2 = \int_0^{2\pi} \int_0^{\pi/2} (E(\beta_n, \alpha_n))^2 f(\beta_n, \alpha_n) \, d\alpha_n \, d\beta_n = \frac{1}{6} \rho^2 (1 + 3 \cos^2 \alpha_1) \quad (10)$$

An analytical proof of the mathematical expression for the slant angle, i.e., $\beta_t = \tan^{-1}\left(\frac{\partial E}{\partial y} / \frac{\partial E}{\partial x}\right)$, is given by Zheng, et al. [18]. Figure 4 summarizes the mathematical formulation and provides a roadmap for numerical estimation of the surface albedo ($\rho$) and illumination direction ($I$).
2.4 Review Shape-from-Shading (SfS) Methods

In this section, frequently used SfS approaches are reviewed. There are four broad classes of SfS approaches: (i) minimization, (ii) propagation, (iii) local, and (vi) linear [11, 19]. This categorization is based on the method used to approximate the image irradiance equation, i.e., Eq. (6).

(1) The minimization approach recovers the 3D topology of a structure by iteratively minimizing an energy function over the entire space of an image. The energy function accounts for the error between image irradiance, \( E(x, y) \), and reflectance map, \( R(p, q) \), i.e., the two sides of Eq. (6). Based on this approach, there are several
methods/algorithms available in literature, for example, Horn [12, 20, 21], Chellappa [18, 22], Szeliski [23], Maydan [24], Kuo [25], Bobick [26], and Yang [27].

(2) In the **propagation** approach, the solution is propagated omnidirectionally from known points (e.g., boundaries) to the entire domain. Tourin [28], Oliensis [29-31], and Bruckstein [32] are examples of SfS algorithms proposed based on the propagation approach.

(3) The **local** approach recovers the local, pixel-by-pixel geometry of a structure based on the local intensity gradients between a pixel and its (immediate) neighbors. Subsequently, the locally recovered geometries are attached together piece-by-piece to form the final 3D topology of the structure. Rosenfeld *et al.* [33] proposed a surface recovery algorithm based on this approach.

(4) In the **linear** approach, the image irradiance equation, which is a nonlinear partial differential equation (PDE) is linearized using the Taylor series expansion and then solved. Pentland [14] and Shah [15] are examples of algorithms developed based on the linear approach.

In this section, three SfS methods are delineated, namely the methods proposed by Horn, Pentland, and Shah. The performance of these methods has been consistently tested in literature by recovery of the 3D profile of regular and complex geometries such as human faces [11, 13, 19, 34]. In addition, computationally efficient algorithms have recently been advanced for these methods. In this work, all of the three methods were adopted from literature and then modified to handle AJP-printed electronic structures. The local and propagation SfS approaches are not used due to their underlying computational complexity and resulting delay, which makes them ill-suited for online monitoring purposes. Each of
the three methods, i.e., Horn, Pentland, Shah used in this work are compared and contrasted in Table 1, and explained in detail herewith.

Table 1: A comparison of the methods used in this study for *in situ* recovery of the 3D topology of AJP-printed electronic structures. (Source: [14, 15, 19].)

<table>
<thead>
<tr>
<th>Method</th>
<th>Class</th>
<th>Solution Scheme</th>
<th>Properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>Horn</td>
<td>Min.</td>
<td>Iterative</td>
<td>• Slow convergence; intermediate stability.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>• Based on variation calculus</td>
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<td></td>
<td>• Does not consider interreflection</td>
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<td></td>
<td></td>
<td></td>
<td>• Sensitive to light source direction</td>
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<td></td>
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<td></td>
<td>• Over-smooth surface recovery</td>
</tr>
<tr>
<td>Pentland</td>
<td>Linear</td>
<td>Non-iterative</td>
<td>• Fast convergence</td>
</tr>
<tr>
<td></td>
<td></td>
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<td>• Does not consider interreflection</td>
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<td></td>
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<td>• Susceptible to frequency doubling (i.e., inconsistency between shape and illumination condition)</td>
</tr>
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<td>• Sensitive to nonlinear changes of reflectance</td>
</tr>
<tr>
<td>Shah</td>
<td>Linear</td>
<td>Iterative</td>
<td>• Fast convergence, good surface recovery when light source direction is close to viewing direction (as in this study)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>• Does not consider interreflection</td>
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<td></td>
<td></td>
<td></td>
<td>• Simple yet efficient</td>
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<tr>
<td></td>
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<td>• Sensitive to noise; sensitive to <em>self-shadow</em>.</td>
</tr>
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</table>

(a) *Horn’s Method (Minimization)*

Horn’s Method (a minimization method) formulates an error function [11] composed of two constraints, and then tries to minimize it iteratively. The first constraint, $\varepsilon_1$, is called the brightness constraint. It accounts for the difference ($\Delta$) between the reflectance of a recovered surface, $R(p,q)$, and the irradiance of the original image, $E(x,y)$. It is mathematically expressed in Eq. (13),

$$\varepsilon_1 = \iint (\Delta(x,y))^2 \, dx \, dy$$  \hspace{1cm} (13)

where $\Delta(x,y) = E(x,y) - R(p,q)$; we note that $p = \partial Z_{(x,y)}/\partial x$ and $q = \partial Z_{(x,y)}/\partial y$.

The second constraint, $\varepsilon_2$, called the smoothness constraint, ensures that a smooth surface is recovered. The second constraint is mathematically expressed as follows,
\[ \varepsilon_2 = \iint (p_x^2 + q_y^2 + p_y^2 + q_x^2) \, dx \, dy \]  
where, \( p_x^2 = \partial^2 Z / \partial x^2 \), \( p_y^2 = \partial^2 Z / \partial y^2 \), \( p_x^2 = \partial^2 Z / \partial x \, \partial y \), and \( p_y^2 = \partial^2 Z / \partial y \, \partial x \).

Therefore, the error function of minimization, \( \varepsilon \), which incorporates both the constraints of brightness and smoothness is as follows [11],

\[ \varepsilon = \iint (\Delta(x, y))^2 + \gamma (p_x^2 + q_y^2 + p_y^2 + q_x^2) \, dx \, dy \]  

In Eq. (15) \( \gamma \) is a positive constant which controls the relative contribution of the two constraints in the error function.

The error function, \( \varepsilon \), is minimized using the Euler-Lagrange equations, resulting in a system of second-order partial differential equations (PDEs). They are discretized and solved using numerical schemes [19]. Eqs. (16) and (17) show the Euler-Lagrange equations written based on the error function (Eq. (15)) [11].

\[ \frac{\partial \varepsilon}{\partial p} - \frac{\partial}{\partial x} \left( \frac{\partial \varepsilon}{\partial p_x} \right) - \frac{\partial}{\partial y} \left( \frac{\partial \varepsilon}{\partial p_y} \right) = 0 \]  \hspace{1cm} (16)
\[ \frac{\partial \varepsilon}{\partial q} - \frac{\partial}{\partial x} \left( \frac{\partial \varepsilon}{\partial q_x} \right) - \frac{\partial}{\partial y} \left( \frac{\partial \varepsilon}{\partial q_y} \right) = 0 \]  \hspace{1cm} (17)

If the partial derivatives of Eqs. (16) and (17) are taken for each term with respect to the error function (\( \varepsilon \)), the following set of PDEs (i.e., Eqs. (18) and (19) called Laplace’s equations) are obtained after simplification.

\[ \nabla^2 p = \frac{1}{\gamma} \left( R(p, q) - E(x, y) \right) \frac{\partial R(p, q)}{\partial p} \]  \hspace{1cm} (18)
\[ \nabla^2 q = \frac{1}{\gamma} \left( R(p, q) - E(x, y) \right) \frac{\partial R(p, q)}{\partial q} \]  \hspace{1cm} (19)

Note that \( \nabla^2 p = \frac{\partial^2 p}{\partial x^2} + \frac{\partial^2 p}{\partial y^2} \) and \( \nabla^2 q = \frac{\partial^2 q}{\partial x^2} + \frac{\partial^2 q}{\partial y^2} \). Eqs. (18) and (19) are discretized and solved iteratively using numerical methods. Eqs. (20) and (21), show an explicit, iterative scheme (forward difference in step, \( k \), and central difference in space \((m, n)\)) to solve for the surface gradients \((p \text{ and } q)\). Based on the Taylor series expansion of the Laplace
equations (Eqs. (18) and (19)), the central difference scheme has an accuracy of second order in the gradient space, i.e., \(O(\Delta p^2, \Delta q^2)\).

\[
p_{m,n}^{k+1} = 0.25 \left[ \left( p_{m+1,n}^k + p_{m-1,n}^k + p_{m,n+1}^k + p_{m,n-1}^k \right) + \frac{1}{\gamma} \left( E(m,n) - R(p_{m,n}^k, q_{m,n}^k) \partial R \frac{\partial}{\partial p} \right) \right] \\
q_{m,n}^{k+1} = 0.25 \left[ \left( q_{m+1,n}^k + q_{m-1,n}^k + q_{m,n+1}^k + q_{m,n-1}^k \right) + \frac{1}{\gamma} \left( E(m,n) - R(p_{m,n}^k, q_{m,n}^k) \partial R \frac{\partial}{\partial q} \right) \right] 
\]

In Eqs. (20) and (21), \(m\) and \(n\) are defined based on the 2D pixel grid of an image.

Finally, Eq. (22) [11] is used to obtain the 3D profile of a surface, \(Z\),

\[Z = \sum \left( \frac{-i \left( \omega_x F_p(\omega_x, \omega_y) + \omega_y F_q(\omega_x, \omega_y) \right)}{\omega_x^2 + \omega_y^2} \right) e^{i(\omega_x x + \omega_y y)}\]

where \(F_p\) and \(F_q\) represent the Fourier transform of the surface gradients, i.e., \(p\) and \(q\), respectively; \(\omega_x\) and \(\omega_y\) are the angular frequency in the \(x\) and \(y\) direction, respectively.

One further step is required to make the computed surface gradients \((p\) and \(q)\) integrable at each iteration [11]. This is done by taking the partial derivatives of the recovered surface, i.e., \(Z\), followed by taking the Fourier transform of the resulting surface gradients, and finally updating Eq. (22). The minimization problem is thus a boundary value problem solved iteratively; both initial and boundary conditions need to be specified. The boundary conditions in this study were defined by assigning large values to the height of bright pixels typically located in the middle of an AJP line (see Figure 11, for example). The aforementioned mathematical formulation, as well as an outline for numerical implementation of the Horn’s method are given in Figure 5.
(b) Pentland’s Method (Linear)

In this method, the surface normal vector, \( \mathbf{n} \), is defined in an inverse manner, compared to the initial definition where \( \mathbf{n} = [-p, -q, 1]^T / \sqrt{1 + p^2 + q^2} \); as a result, the surface reflectance, \( R(p, q) \), which is mathematically corresponds to image irradiance, \( E(x, y) \), is expressed [11, 14] as follows,
\[ E(x,y) = R(p,q) = \rho \mathbf{1}^T \mathbf{n} = \frac{\rho \mathbf{1}^T [p,q,-1]^T}{\sqrt{1 + p^2 + q^2}} \]

\[ = \rho \frac{p \sin \alpha \cos \beta + q \sin \alpha \sin \beta + \cos \alpha}{\sqrt{1 + p^2 + q^2}} \]

Subsequently, with consideration of only the first-order term of the Taylor series expansion about the point \((p_0, q_0) = (0,0)\) (assuming a diffuse surface), the surface reflectance equation is linearized and reduced to Eq. (24).

\[ E(x,y) = R(p,q) \approx p \sin \alpha \cos \beta + q \sin \alpha \sin \beta + \cos \alpha \]  

(24)

Taking the Fourier transform of Eq. (24) followed by rearrangement results in,

\[ F_Z(\omega_x, \omega_y) = \frac{F_E}{-i \omega_x \sin \alpha \cos \beta - i \omega_y \sin \alpha \sin \beta + \cos \alpha} \]

(25)

where \(F_Z\) and \(F_E\) are the Fourier transform of the surface profile, \(Z(x,y)\), and image irradiance, \(E(x,y)\); \(\alpha\) and \(\beta\) are the tilt and slant angles of the illumination direction vector \(\mathbf{1}\); \(\omega_x\) and \(\omega_y\) are the angular frequency in the \(x\) and \(y\) direction, respectively.

Finally, the surface profile is obtained by taking the inverse Fourier transform of \(F_Z(\omega_x, \omega_y)\) [11, 14]. The aforementioned mathematical formulation and an outline for numerical implementation of the Pentland’s method are shown in Figure 6.
(c) **Shah’s Method (Linear)**

In Shah’s method, which is an iterative linear approach, an error function ($\varepsilon$) is defined based on the difference between image irradiance, $E(x,y)$, and surface reflectance, $R(p,q)$, as mathematically expressed by Eq. (26). The error function is subsequently linearized by ignoring the second- and higher-order terms of the Taylor series expansion about the point, $Z_{(x,y)}^{k-1}$, i.e., the surface profile recovered at iteration $k - 1$. After rearrangement and simplification, the surface is recovered iteratively assuming $Z_{(x,y)}^{0} = 0$, using Eq. (27). For an image of size $(m, n)$, there is a system of $m \times n$ linear equations that is solved using the Jacobi iterative method [11, 15].
In Eq. (27), \( i_x \) and \( i_y \) are the \( x \) and \( y \) component of the illumination direction vector, \( \mathbf{I} \), respectively. The surface gradients, i.e., \( p \) and \( q \), are updated in each iteration using a finite difference scheme where \( p = Z_{(x,y)} - Z_{(x-1,y)} \) and \( q = Z_{(x,y)} - Z_{(x,y-1)} \). The aforementioned mathematical formulation as well as a pseudo-algorithm for numerical implementation of the Shah’s method are given in Figure 7.

**Mathematical Formulation**

**Step 1:**
Defining an error function, \( \varepsilon \), based on the difference between image irradiance, \( E_{(x,y)} \), and surface reflectance, \( R_{(p,q)} \), (Eq. (26))

**Step 2:**
Recovery of 3D surface profile via linearization of the error function using Taylor series expansion. (Eq. (27))

**Numerical Implementation**

**Phase I: Pre-calculation**
If not known, following the algorithm shown in Figure 4, estimate:
1. Surface reflectivity/albedo (\( \rho \)).
2. Illumination direction (\( \mathbf{I} \)).

**Phase II: Initialization**
1. Surface gradients (\( p^0 \) and \( q^0 \)).
2. Surface profile, \( Z^0_{(x,y)} \).
3. Maximum number of iterations.

**Phase III: Computation**
For \( k=1 \): the max number of iterations,
1. Compute the reflectance map (\( R^k_{(p,q)} \)), using Eq. (6).
2. Compute the error function, \( \varepsilon^k \), using Eq. (26).
3. Recover the 3D surface profile, \( Z^k_{(x,y)} \), using Eq. (27).
4. Update the surface gradients (\( p^k \) and \( q^k \)).

Figure 7: A summary of the mathematical formulation of the Shah’s method together with a pseudo-algorithm proposed for numerical implementation [11].

### 2.5 Case Study – Recovery of the Surface Profile of Synthetic and Real Images

The aim of this section is to ascertain which of the three SfS methods, i.e., Horn, Pentland, and Shah, is most suitable for recovering the topology of AJP lines. For this purpose, two 2D synthetic shaded images including a sphere and a cylinder are produced.
The latter approximates the geometry of real AJP-printed lines. These simple geometries are used to compare the performance of the aforementioned SfS methods. Additionally, a real AJP printed line is used as a crosscheck. For the generation of the synthetic images, a surface albedo ($\rho$) of 0.5 and illumination direction, $\mathbf{I}$, of $[0.2, 0, 0.98]^T$ was considered.

The synthetic images were created based on the equation of surface for each geometry, i.e., Eq. (28) for the sphere and Eq. (29) for the cylinder. The positive (or negative) root of Eq. (28) and Eq. (29) was considered for image generation because only a hemisphere is visible when an image is captured. The sphere and cylinder had a radius, $r$, of 50 pixels as well as an $xy$ domain of (75 pixels × 75 pixels) and (150 pixels × 150 pixels), respectively.

\begin{align}
Z_{\text{sph}} &= \pm \sqrt{r^2 - (x^2 + y^2)} \tag{28} \\
Z_{\text{cyl}} &= \pm \sqrt{r^2 - y^2} \tag{29}
\end{align}

Subsequently, to obtain the reflectance map of each surface (based on Eq. (6)), the two components of the surface gradient, i.e., $p$ and $q$, given by Eqs. (30)-(33) for the sphere and cylinder, were computed numerically. We note that a gray-scale image is ultimately formed once the negative values of the surface reflectance are set to zero followed by normalization.

\begin{align}
p_{\text{sph}} &= \frac{\partial Z_{(x,y)}}{\partial x} = -x(r^2 - (x^2 + y^2))^{-1/2} \tag{30} \\
q_{\text{sph}} &= \frac{\partial Z_{(x,y)}}{\partial y} = -y(r^2 - (x^2 + y^2))^{-1/2} \tag{31} \\
p_{\text{cyl}} &= \frac{\partial Z_{(x,y)}}{\partial x} = 0 \tag{32} \\
q_{\text{cyl}} &= \frac{\partial Z_{(x,y)}}{\partial y} = -y(r^2 - y^2)^{-1/2} \tag{33}
\end{align}

Image quality significantly influences the quality of surface reconstruction. Finally, a high-resolution image captured by an optical microscope (Zeiss Axio Imager M1M) of an AJP-printed line was used. The results of surface recovery for each of the geometries discussed in this section using the Horn’s (minimization), Pentland’s (linear), and Shah’s
(linear) methods are shown in Figure 8. An examination of the quality of the recovered surfaces shows that the Shah’s method has the highest degree of accuracy (see Figure 8(d), (h), and (l)). Hence, Shah’s method is applied to our experimental results (discussed in the forthcoming Sec. 4) to recover the topology of AJP-printed lines.

![Figure 8: The 3D surfaces of a synthetic sphere, a synthetic cylinder, and a real AJP-printed line recovered using Horn’s method [11, 13], Pentland’s method [14], and Shah’s method [15]. It is evident that Shah’s method has the highest fidelity in surface reconstruction (see (d), (h), and (l)).](image)

3. Experimental Studies

3.1 Background of Aerosol Jet Printing (AJP) Process

In AJP process (Figure 9) a high-pressure flow of a carrier gas, typically N₂, is passed through a container, called bubbler which is filled with ink solvents. As a result, the carrier gas is saturated. This saturated carrier gas flow (CGF) is then injected into the ink reservoir where due to the Venturi effect, the ink is drawn upward via a capillary tube. Consequently,
a fine spray of micro-droplets is generated. The heavier droplets fall back into the reservoir, while the lighter ones are carried upward toward the virtual impactor. The virtual impactor aerodynamically separates the droplets having lower momentum (via an exhaust gas flow, EGF) and allows the rest to flow toward the deposition head. In the deposition head, a secondary gas flow, called sheath gas flow (ShGF), is introduced to collimate or focus the aerosol flow into a narrow beam. The collimated aerosol flow accelerates through a converging nozzle and is deposited on a substrate.

Figure 9: A schematic diagram of aerosol jet printing (AJP) process utilizing a pneumatic atomizer. The setup has been instrumented with a CCD camera to capture online images.

The unique capabilities of AJP stem from the ability for high-resolution printing (≤ 10 μm) on non-planer and flexible surfaces; printing materials with a wide spectrum of viscosities (1 to 1000 cP); aerodynamic (non-contact) control of feature size [2, 35-37]. A review of literature shows that AJP has a wide range of applications including: conformal circuits and metallic traces deposited on 3D surfaces [36]; silver electrodes with a high degree of conductivity [38]; complex intermediate structures [39]; organic light emitting diodes (OLEDs) [40]; solar cell current-collecting grids [41, 42]; etc. With the aid of direct etching of dielectric patterns, Rodriguez et al. [43] showed that AJP would be a potential substitute for conventional electronics manufacturing techniques. In an effort toward
complete additive manufacture of printed circuit boards (PCBs), Christenson *et al.* [2] demonstrated AJP-based deposition of passive components, e.g., resistors, capacitors, strain gauges, interdigital sensors, etc., as well as interconnects.

### 3.2 Experimental Setup and Materials

* (a) Sensor-Instrumented Testbed

All samples in this study are printed using an Optomec AJ-300 machine. A high-resolution charge-coupled device (CCD) camera (Edmund Optics, Grasshopper, GS3-U3-50S5C-C) supported by a 2.5X to 10X variable magnification lens (Edmund Optics, VZM 1000i) is used to capture online images. The online images have dimensions of 2448 × 2048 pixels, and each pixel has dimensions of 3.45 × 3.45 μm at the set field of view. The imaging system is illuminated by a LED ring light having a brightness of 30000 - 40000 Lux and a color temperature of 6000 K. The main components of our experimental testbed are shown in Figure 10.

* (b) Materials

The sheath and atomization (carrier) gas flows are dry and pure streams of N2 flowing at ambient temperature (21 °C). The ink used in all printing tasks is composed of a mixture of two silver nanoparticle inks of PARU (MicroPEPG007MOP and MicroPEPG007EG) mixed with a ratio of 5:1 by weight, respectively. The former is comprised of silver nanoparticles (~ 66 wt%) and 1-methoxy-2-propanol (MOP) as solvent having a bulk density and viscosity of 1.5-2.5 g/ml and 50 cP, respectively. The latter is comprised of silver nanoparticles (~33 wt%) and ethylene glycol (EG) as solvent, approximately having a bulk density and viscosity of 1.2-3.3 g/ml and 39 cP, respectively.
Figure 10: Pictures of the aerosol jet printing (AJP) setup instrumented with image-based and temporal sensors. Reconstruction and quantification of line topology are based on online images captured using the CCD camera.

(c) Design of Experiments

A randomized single-factor experiment was conducted to investigate the influence of sheath gas flow rate (ShGFR) on line topology. The ShGFR was varied from 40-140 sccm with 20 sccm increments. This will be expanded to other factors in forthcoming publications. A pneumatic atomizer (PA) was used with an atomization gas flow rate (AGFR) and exhaust gas flow rate (EGFR) of 580 sccm and 560 sccm, respectively. The difference between these two flow rates is equivalent to a carrier gas flow rate of 20 sccm (used with ultrasonic atomization). The print speed (PS) was set at 5 mm/s. As exemplified in Figure 14, conductive structures composed of silver nanoparticle lines were deposited in a single pass on a silicon wafer using a 150 µm nozzle tip at a working distance of 3 mm. The silicon substrate was heated via the platen already set at 40 °C. Each print was replicated 5 times. Table 2 summarizes the range of process parameters used in this study.
Two sets of experiments are conducted in this work. In the first experimental set, as discussed above, an $n$-type silicon wafer, approximately 500 µm thick is used as a substrate. It has a surface roughness of approximately a few tens of angstroms coated with no thermal oxide layers. This substrate was used to ascertain the accuracy of the SfS approach, and narrow the ShGFR range to an acceptable range. The second set of experiments is conducted on a custom-engineered flexible Kapton (Polyimide) substrate supplied by our industrial collaborator.

Table 2: Process parameters and materials used in this study for AJP-printing of electronic devices.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Experiment 1</th>
<th>Experiment 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>ShGFR</td>
<td>40-140 sccm</td>
<td>60-100 sccm</td>
</tr>
<tr>
<td>AGFR</td>
<td>580 sccm</td>
<td>500 sccm</td>
</tr>
<tr>
<td>EGFR</td>
<td>560 sccm</td>
<td>450 sccm</td>
</tr>
<tr>
<td>PS</td>
<td>5 mm/s</td>
<td>4 mm/s</td>
</tr>
<tr>
<td>Nozzle Size</td>
<td>150 µm</td>
<td>300 µm</td>
</tr>
<tr>
<td>Working Distance</td>
<td>3 mm</td>
<td></td>
</tr>
<tr>
<td>Ink</td>
<td>PARU (Solvent Ratio=5:1 by Weight)</td>
<td></td>
</tr>
<tr>
<td>Substrate</td>
<td>$n$-Type Silicon Wafer</td>
<td>Flexible Kapton (Polyimide)</td>
</tr>
<tr>
<td>Sintering</td>
<td>Oven-sintering (2 hours at 200 °C)</td>
<td></td>
</tr>
</tbody>
</table>

Furthermore, the bubbler which delivers saturated gas flow and prevents the ink from drying out was filled with 2-methyl-2-propanol, one of the ink solvents. The ink was sonicated prior to use. A five-minute equilibration time was considered after changing each ShGFR set point to ensure constant (steady-state) delivery of aerosols to the print head. Online images were captured of the centerline of each printed structure using the high-resolution CCD camera. Figure 11 exemplifies online images captured of the first replicate of each factor level.
Figure 11: The influence of ShGFR on the 2D characteristics of AJP-printed lines captured online using a high-resolution CCD camera. (The atomization and exhaust gas flow rates are 580 sccm and 560 sccm, respectively.)

It is noted that an inordinate increase in ShGFR (ShGFR > 100 sccm) results in pressure buildup in the nozzle and, consequently, uneven material deposition followed by nozzle clogging. This phenomenon was shown using computational fluid dynamics (CFD) simulations and empirical studies in our prior work [5, 9]. In addition to formation of overspray, the linear momentum of aerosols increases with ShGFR resulting in more post-deposition spreading, and as a result, less line thickness as evident in Figure 11(d-f)).

4. Results

4.1 Reconstruction of Line Topology

Representative 3D profiles of the AJP lines recovered using Shah’s method are shown in Figure 12. An initial examination of the recovered profiles shows that line thickness increases with ShGFR. The fidelity of profile recovery is influenced by image resolution, illumination conditions, and surface reflectivity including deposited material and substrate reflectivity. The influence of image resolution/quality on the accuracy of surface recovery
can be observed, for example, by comparing the surfaces shown in Figure 12 (based on camera images) with that shown in Figure 8(l) (based on a microscope image). The practical hurdles for implementing this approach are further delineated in Sec. 4.3.

As observed from Figure 11, line intensity increases with the ShGFR. In other words, the lines having lower thickness profiles unexpectedly appear brighter in the images (see Figure 11(e)-(f), for example). This may adversely influence the thickness of the recovered topologies as evident in Figure 12 where the lines printed at higher ShGFRs have correspondingly taller profiles (compare Figure 11(e)-(f) with Figure 12(e)-(f)). Such an error in surface recovery can be corrected by assigning the complement of intensity values to the line regions. Another disadvantage of SfS algorithms is that they cannot discriminate between convex and concave surfaces [11].

As demonstrated in Figure 13, to verify the accuracy of the online recovery, the cross-sectional profile of the lines estimated from the Shah’s method were compared with those measured offline using an optical profilometer (Wyko NT 1100 - Veeco Instruments) as the ground truth. Almost the whole profile of a line can be truly recovered when print quality is within the optimal process window (see Figure 13(a)). Less accurate profiles are recovered when the print quality drops at the ShGFR of 120 and 140 sccm; compare Figure 11(e) and (f), and Figure 13(e) and (f)). There are two reasons for the difference between the offline-observed and online-predicted profiles at the ShGFR of 120 sccm and 140 sccm: (i) In our previous work [5, 9], it was demonstrated using CFD modeling and empirical tests that inordinately high ShGFR values (> 100 sccm) leads to aerodynamic instability at the nozzle exit which disturbs the flow and often causes nozzle clogs. Aerodynamically instable deposition is the root of poor line quality. As a result, the
overall intensity gradient between the lines and substrate decreases, and therefore the line topology is recovered less accurately.

(ii) It was observed that the sets of lines pertaining to the ShGFR of 120 and 140 sccm are more spread out, which in turn leads to solvent evaporation from the surface; and creates a less reflective surface. This is evident on comparing Figure 11(c)-(f) with Figure 11 (a)-(d).

Figure 12: The profile of AJP-printed lines recovered using Shah’s method [15] (shown only for the first replicate). The recovered profiles are normalized to be in the range of [0 1].
Figure 13: A representative comparison between the online and offline profile recovery. The offline recovery (as the ground truth) is based on an optical profilometer.

4.2 In Situ Prediction of Line Resistance

The aim of this section is to predict the resistance of a line as a function of its cross-sectional area estimated using the Shah’s method and the AJP process conditions. Based on the results of the first set of experiments described in Sec. 4.1, the ShGFR is narrowed to the range of 60 to 100 sccm, treated as the second set of experiments under the conditions summarized in Table 2.

The second experimental set was conducted on a flexible, unheated polyimide (Kapton) substrate. The second experiment is more in lockstep with the authors’ long-term research
focus on flexible electronics manufacturing (Binghamton University (SUNY) is the NY State node and founding member of the Flexible Hybrid Electronics Manufacturing Innovation Institute, FHEMII). Furthermore, instead of a simple line, a test artifact with four-terminal structures was printed. Real images and a schematic of the printed structures are shown in Figure 14. The 4-point probes method (also known as Kelvin sensing) was used for offline resistance measurements [44]. This combination of ink and substrate allows industry-standard measurement of line resistance and thus helps to establish a baseline reference for in situ SfS-based predictions of resistance.

The atomization and exhaust gas flow rates (AGFR and EGFR) were set at 500 and 450 sccm, respectively. Aerosol generation and deposition were carried out utilizing a pneumatic atomizer and a 300 µm nozzle. To be consistent with the previous experiment, working distance was kept at 3 mm. The silver nanoparticle structures were printed in a single pass with a speed (PS) of 4 mm/s and then oven-sintered at 200 °C for 1 hour. Figure 15 exemplifies CCD images captured in situ from the printed structures before sintering.
Figure 14: (Top) The dimensions of the printed test artifact with four-terminal structures for measurement of the line resistance. The four $1 \text{ mm} \times 1 \text{ mm}$ pads are the probe points. (Bottom) Silver nanoparticle devices printed on flexible Kapton.

The line profile is estimated \textit{in situ} using the Shah’s SfS method. This is quantitatively shown in Figure 16(a) where the cross-sectional area of each line is quantified by numerical integration of the thickness profile averaged over the entire length (of the line). Per Ohm’s law, the resistance of a line ($R$, Ω) is inversely proportional to its cross-sectional area ($m^2$), and directly proportional to its length ($L$, m), with a constant, $\rho$ (resistivity, Ω-m). Once the cross-sectional area of a line is quantified, Ohm’s law is used to estimate the resistance. The resistivity, $\rho = 3.18 \times 10^{-8}$ Ω.m, is considered to be twice the bulk resistivity of silver. Figure 16(b) maps line resistance predicted online using Ohm’s law based on the SfS-derived cross-sectional area as a function of ShGFR. The mean error between the actual and predicted line resistance is less than 20%. The error can be further decreased with consideration of multiple-input models taking into account not only the cross-sectional area but also 2D line quality quantifiers from our previous work [5, 9]. This will be forwarded
as part of the future work; the practical challenges of implementing this work are delineated in Sec. 4.3.

![Figure 15: High-resolution CCD images captured in situ before sintering from four-terminal conductive structures.](image)

4.3 Notes on Practical Implementation

**Computational Load.** The computational load of the SfS algorithm depends on: (i) surface recovery accuracy (as a function of the number of iterations); (ii) image size. It is approximately in the range of 10-20 seconds (implemented in the MATLAB environment). Programming in other environments (e.g., C++) or use of parallel computing may significantly reduce the run time to less than 10 seconds.

**Sampling Frequency.** Sampling frequency depends on the magnitude of a shift (drift or error) in the process mean. To detect small process drifts, higher sampling frequencies are needed. To capture the process dynamics accurately, online images are acquired with a
frame rate being in the range of 5-10 frames per second (fps). The sampling frequency may be increased to the limits of our imaging hardware’s resolution to detect very small shifts. Currently, our imaging system has a maximum frame rate of 15 fps. This allows for video- graphic feedback monitoring and control of the process in the future.

**Latency of the Calculation.** This is a function of:

(1) The time needed to translate the stage under the high-resolution camera (programmed to be a fraction of a second). The stage on the Optomec AJ-300 printer used in this study has a maximum translation speed of 100 mm/s. Accordingly, such rapid translation would make the change in position from printing to inspection be a few of seconds, at most.

(2) The time needed to capture and store an image is less than 1 sec (5-10 fps).

(3) The time needed to process the image and compute line cross-sectional area (less than 20 second).

(4) For closed-loop control, the amount of time needed to find optimal solutions for the corrective action plan and actuation (part of our future work).

5. Conclusions and Future Work

In this work, the cross-sectional profile of AJP-deposited electronic traces (lines) was estimated from online images using shape-from-shading (SfS) analysis. Three SfS approaches were tested, namely, Horn’s, Pentland’s, and Shah’s methods. Tests with synthetic and experimentally obtained images showed that the Shah’s method was most suitable. Specific inferences are as follows:

(1) Silver nanoparticle test electronic devices in the form of four-terminal structures were
printed, and their electrical resistance was measured offline using the Kelvin sensing method. AJP experiments were conducted under varying sheath gas flow rate (ShGFR). The cross-sectional profile at each of these conditions was estimated using the Shah’s method. The recovered topologies of the printed electrical structures were further processed, and line cross-sectional area was quantified.

(2) The cross-sectional area versus ShGFR trends are verified using optical profilometry. Comparison of the offline-measured and online-estimated profiles reveals that the most accurate recovery of line cross-sectional profiles is obtained when the ShGFR is set between 60 and 100 sccm. This is because of aerodynamic instability in the aerosol flow at extreme ShGFR values. At low ShGFRs (< 40 sccm) the aerosol flow is not sufficiently collimated, while at high ShGFRs (> 100 sccm) the pressure in the nozzle increases leading to overspray. The line cross-sectional area increases with ShGFR and hence the resistance decreases. The error between the SfS-estimated line resistance and offline measurement is within 20%.

In the forthcoming research, the aim is to augment the online quantification of line cross-sectional profile with 2D line quality aspects from the previous work, and expand the experimental conditions to other factors, such as carrier gas flow rate, print speed, etc. [5, 9].
References


Chapter 4: A Novel Sparse Representation-Based Classification (SRC) Approach for Real-time Monitoring of Device Functional Properties in AJP Process

1. Introduction

1.1. Research Goal and Objectives

The goal of this work is to establish a platform for in situ monitoring of the functional properties – including electrical and mechanical properties, e.g., resistance, fatigue life, etc. – of printed electronic devices, fabricated using aerosol jet printing (AJP). In pursuit of this goal, the objectives are: (i) in situ image acquisition of a printed device/line right after deposition; (ii) in situ image processing and quantification of the device/line morphology; and (iii) development of a machine learning model to estimate the device functional properties in near real-time. We have already addressed the first and second objectives, as delineated in our previous publications [1-4]. This study primarily pertains to the development of a supervised multiple-input, single-output (MISO) classification model for in situ estimation of printed line resistance as a function of process parameters as well as quantified characteristics of line morphology. The research hypothesis is that the resistance of a printed line is estimated in near real-time with an accuracy of ≥ 90%. This work addresses the following research question: Could a MISO machine learning model, based on sparse representation for classification (SRC), accommodate the multivariate and heterogenous nature of sensor data in AJP, and thus be used for in situ monitoring of device functional properties?

1.2. Motivation and Significance

Categorized as a direct-write (DW) additive manufacturing (AM) technique [5], AJP has emerged as the process of choice for the fabrication of a broad spectrum of electronic
devices, such as: transistors [6-8]; fine-pitch electronics [9]; sensors [10, 11]; organic light emitting diodes [12]; and optoelectronic devices [13]. This is mainly due to the unique capabilities and advantages of AJP, including: (i) deposition of fine microstructures with feature sizes as small as 10 μm; (ii) high placement accuracy; (iii) accommodation of a wide range of ink viscosity, ranging from 0.7-2500 cP; (iv) having a variable, large standoff distance, which allows for material deposition on non-planer surfaces; and (v) room-temperature operation [5, 14, 15]. Figure 1 exemplifies some AJP electronic devices fabricated at State University of New York (SUNY) at Binghamton [3].

![Figure 1](image_url)  
**Figure 1.** Examples of electronic devices, AJ-printed at SUNY-Binghamton: (a) an antenna printed on a flexible glass substrate; (b) reduced graphene oxide supercapacitors (rGO-SC) and silver interdigitated electrodes (IDE), printed on a slim glass substrate. (Source: [3])

Despite such advantages and the host of critical applications, the AJP process is highly unstable and complex [16], prone to gradual drifts and changes in machine behavior and deposited material. Therefore, realization of real-time process monitoring and control seems to be inevitable. This work is a direct continuation of the authors’ recent research in AJP [1-4]. The prior works paved the way for *in situ* monitoring of device functional properties by:

(i) introducing image-based quantifying algorithms of line morphology, i.e., (1) line width; (2) line density; (3) edge quality/smoothness; (4) overspray; (5) line discontinuity; and (6) Fiedler number, which is a graph-theoretic quantifier reflecting
surface morphology [17]. Figure 2(a) illustrates the concept behind each of the quantifiers used in this study to extract line morphology information from an image.

(ii) forwarding an image-based approach for reconstruction/recovery of the 3D profile of AJP lines based on shape-from-shading (SfS) analysis [1], which further allows for quantification of the 3D characteristics of the line morphology, such as thickness, cross-sectional area (CSA), and surface roughness. Figure 2(b) schematically depicts the recovery of the cross-sectional profile of an AJP line using the SfS method.

<table>
<thead>
<tr>
<th>a. 2D Quantification of Line Topology</th>
<th>b. 3D Quantification</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Line Width ($l_w$)</td>
<td>AJP-Printed Device</td>
</tr>
<tr>
<td>Upper Edge</td>
<td>Online Image Acquisition</td>
</tr>
<tr>
<td>Width</td>
<td>50 μm</td>
</tr>
<tr>
<td>2. Line Density ($l_p$)</td>
<td>3SFS</td>
</tr>
<tr>
<td>Average Intensity within a Line</td>
<td>3D Recovery of Topology</td>
</tr>
<tr>
<td>3. Edge Quality ($l_{eq}$)</td>
<td></td>
</tr>
<tr>
<td>Edge Profile</td>
<td></td>
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<tr>
<td>4. Overspray ($l_{os}$)</td>
<td></td>
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<tr>
<td>Overspray</td>
<td></td>
</tr>
<tr>
<td>Distance and intensity</td>
<td></td>
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<tr>
<td>5. Line Discontinuity ($l_{md}$)</td>
<td></td>
</tr>
<tr>
<td>Discontinuous Edge</td>
<td></td>
</tr>
<tr>
<td>6. Fiedler Number ($\lambda_2$)</td>
<td></td>
</tr>
<tr>
<td>Network Graph</td>
<td></td>
</tr>
</tbody>
</table>

Figure 2. (a): The concept behind the image-based algorithms, concerning the 2D characteristics of line morphology; (b) Reconstruction of the cross-sectional profile of an AJP-printed line, using the SfS image analysis. The 2D and 3D quantified features are fed as input sensor data to a supervised machine learning model to estimate line resistance in near real-time. (Source: [1, 3, 4, 17])

In the previous chapter, it was demonstrated that if the length, CSA, and resistivity of a line were known, the resistance could be directly estimated according to Ohm’s law. However, the direct use of the Ohm’s law may result in inaccurate estimation of the resistance, particularly in cases where sampling frequency is low. In other words, tighter
estimation of the functional properties is desired in practice. This gap is addressed in this work via forwarding a machine learning model, developed based on the concept of sparse representation for classification.

The rest of the paper is presented as follows: in Sec. 2, the AJP process is delineated, followed by a description of our sensor-instrumented experimental setup in Sec. 0. This setup has been equipped with a high-resolution charge-coupled device (CCD) camera in addition to a standard process monitor camera, allowing for \textit{in situ} as well as real-time image acquisition. Sec. 4 details the development of the MISO machine learning model. Experimental results – including materials, design of experiments, experimental observations, and assessment of the SRC model’s performance – are discussed in Sec. 5.

2. Aerosol Jet Printing (AJP) Process

Figure 3 schematically illustrates a flow diagram of the AJP process, utilizing a pneumatic atomizer (PA) as the mechanism of choice for aerosol generation. A high-pressure flow of an inert gas – typically pure and dry N\textsubscript{2} referred to as atomization gas flow (AGF) – is injected into the ink reservoir via a constricted tube positioned over a capillary. Based on the Venturi effect, the ink is drawn upward in the capillary and then sheared by the atomization jet passing through an orifice; consequently, a fine spray of micro-droplets is generated. Larger droplets fall back into the reservoir, while smaller ones form a semi-uniform multi-phase flow toward the virtual impactor (VI). We note that a bubbler filled with the ink solvent(s) is used to provide a saturated flow for atomization, and thus to minimize the loss of solvent from the ink (causing the ink viscosity to change). This process of saturating the atomization gas utilizes a sparger, allowing for an efficient gas-liquid
contact. The virtual impactor aerodynamically separates those droplets being of low linear momentum and small size (using an exhaust gas flow, EGF), while allowing the rest to flow as a uniform stream of aerosols toward the deposition head. In the deposition head, a second gas flow, called the sheath gas flow (ShGF), is concentrically introduced to collimate/focus the aerosol flow into a narrow beam. The rate ratio of the ShGF to the aerosol flow is termed the “focusing ratio” [18]. The collimated aerosol flow ultimately accelerates (while its pressure decreases) passing through a fine, converging nozzle (made up of ceramic), and impinges on a free surface/substrate. The aerosols additively constitute a free-form structure, having experienced post-deposition phenomena of spreading, receding, relaxation, and coalescence [19].

![Figure 3. A schematic flow diagram of the aerosol jet printing process. In this AJP configuration, pneumatic atomization is the mechanism of choice for aerosol generation. A uniform and collimated aerosol flow is obtained with the aid of the virtual impactor and the deposition head, respectively.](image)

3. **Sensor-Instrumented Setup**

   **I. In situ Monitoring Using a CCD Camera:**

   As illustrated in Figure 4, an Optomec AJ-300 aerosol jet printer (Albuquerque, NM, USA) is instrumented with a high-resolution (5 MP) CCD camera (Edmund Optics,
Grasshopper, GS3-U3-50S5C-C, Barrington, NJ, USA), allowing for independent, *in situ* image acquisition. The images acquired by this camera have a maximum window size of 2448 × 2048 pixels. Co-axial to the nozzle, the camera is mounted on a 2.5X-10X variable magnification lens (Edmund Optics, VZM 1000i, Barrington, NJ, USA). The resolution of the imaging system is approximately 0.36 × 0.36 μm/pixel in the x- and y-direction. Illumination is provided by an LED light (AmScope, Irvine, CA, USA) which has a maximum brightness of 30,000 – 40,000 Lux and a color temperature of 6,000 K.

Figure 4. Pictures of the AJP experimental setup, instrumented with a high-resolution CCD camera and a variable magnification lens, allowing for *in situ* image acquisition. In addition, the standard process camera is used for real-time image acquisition and process monitoring.
II. Real-time Monitoring Using the Process Monitor Camera:

Positioned inclined to the platen at about 45°, the process monitor camera is a 1.3 MP GigE color camera (Point Grey Flea3, Richmond, BC, Canada) mounted on an optical 1.0X lens and a 4.0X magnification module (Infinity Photo-Optical InfiniStix™, Boulder, CO, USA). The lens has a working distance of 94 mm. Illumination for this camera was provided by a fiber light (Dolan-Jenner PL-800, Boxborough, MA, USA), mounted opposite to the camera. The illumination position was adjusted such that noise-free, saturated background images were obtained, aiding in accurate detection of printed line edges (see Figure 12, for example). Being of raw12 data format, the images acquired by the process monitor camera were saved as .tif file format.

4. Development of a MISO Machine Learning Model

4.1. Review of the Related Research

Sparse representation for classification (SRC) [20, 21] has emerged as an accurate and computationally-fast supervised classification approach [22, 23], and been utilized in a broad range of applications, such as computer vision [24], healthcare [25], damage detection [26], and real-time monitoring of advanced manufacturing processes [23]. Using SRC, Tootooni et al. demonstrated: (i) classification of global neurophysiological states from electroencephalography (EEG) signals [27]; and (ii) classification of the dimensional variation of additively-manufactured parts [28]. In addition, they showed that the SRC could be successfully implemented for classification of nonstationary and nonlinear data, similar to AJP image-based sensor data, discussed in this study [27]. Furthermore, it has been demonstrated that the SRC can handle data with autocorrelation, intermittency, periodicity, and low signal-to-noise ratio [27, 29, 30]. These all justify the suitability and
flexibility of the SRC for *in situ* functional monitoring of AJ-printed electronics that is inherently comprised of complex multivariate sensor signals. There are several methods of SRC, reviewed in detail in [22, 23]. However, they are discussed briefly here to preserve continuity.

The SRC has been established, based on the fact that sensor data streams, belonging to the same state of a system, have stronger correlation than those collected from different states of the system. It formulates a supervised classification problem as an underdetermined system of linear equations [21, 22] where new sensor data also known as an incoming vector of features, \( Y \in \mathbb{R}^{m \times 1} \), is expressed as a linear combination of design/training data, \( A \in \mathbb{R}^{m \times N} \), as mathematically expressed by Eq. (1), where \( N \) stands for unknowns, i.e., the total number of samples, and \( m \) stands for equations, i.e., the total number of features. Note that the design matrix, also known as the measurement matrix, reflects the history (i.e., the previous states) of a system.

\[
Y = A\beta + \varepsilon
\]  

(1)

In Eq. (1), \( \varepsilon \) represents the approximation error [20, 22]. The aim is to perform classification by estimating the unknown vector of membership coefficients, \( \beta \in \mathbb{R}^{N \times 1} \). However, since the classification problem is ill-posed \((m \ll N)\), it is converted to a minimization problem, assuming \( \beta \) is sparse – i.e., being of the smallest number of nonzero elements [22] – as shown in Eq. (2),

\[
\left\{ \begin{array}{l}
\min \| \beta \|_0 \\
\text{s.t.: } f(Y - A\beta) \leq \delta
\end{array} \right.
\]  

(2)

where, \( \| \beta \|_0 \) represents the \( \ell_0 \)-norm of the unknown vector of membership coefficients to be minimized subject to a function of the fit/residual error, \( Y - A\beta \), being smaller than or equal to a set threshold, \( \delta \) [22]. There are several research works in literature, dedicated to solving this NP-hard optimization problem [31-33].
Basically, there are three categories of methods in literature [22, 23], developed to solve
the sparse estimation problem: (i) \( \ell_1 \)-minimization or convex optimization methods [34-
37]; (ii) greedy methods [38-40]; (iii) Bayesian methods [41-43].

The optimization methods convert the sparse estimation solution (represented via Eq.
(2) [22]) to a convex optimization problem, as mathematically expressed in Eq. (3).

\[
\begin{align*}
\min_{\beta} & \left\{ \|\beta\|_1 \right\} \\
\text{s. t. :} & \|Y - A\beta\|_2^2 \leq \delta \\
\end{align*}
\]

In Eq. (3), \( \|\beta\|_1 \) and \( \|Y - A\beta\|_2^2 \) respectively represent the \( \ell_1 \)-norm of the unknown
vector of membership coefficients and the \( \ell_2 \)-norm of the fit error. The least absolute
shrinkage and selection operator (LASSO) [37] – one of the main pillars of the SRC method
forwarded in this study – is an optimization method, which can be implemented based on
the shooting algorithm [44], coordinate descent algorithm [45], and/or alternating direction
method of multipliers (ADMM) [46], among others. It is worth mentioning that ADMM is
the algorithm of choice in this work. Total variation regularization [36], Dantzig selector
[34], and basis pursuit de-noising [35], are other convex optimization methods.

Heuristic/greedy methods – such as thresholding [40], regularized and stage-wise
orthogonal matching pursuit (OMP) [38, 40], subspace pursuit [47], least angle regression
[39], and compressive sampling matching pursuit [48] – are less computationally
expensive. However, they may fail to reach the global minimum. Similar to the
optimization methods, the greedy methods use the \( \ell_2 \)-norm of the fit error, i.e., \( \|Y - A\beta\|_2^2 \)[22].

Relevance vector machine (RVM) [41, 42] as well as sparse Bayesian learning [43] are
examples of Bayesian methods. Unlike the convex optimization and greedy methods that
provide point estimates of the classification membership coefficient vector (\( \beta \)), the
Bayesian methods enforce sparsity, assuming a posterior distribution or Gaussian likelihood function on the coefficients [22, 23].

Bastani et al. [23] proposed a combined greedy-Bayesian sparse estimation approach, utilizing the advantages of RVM (a Bayesian method) and OMP (a greedy method). Barazandeh et al. [22], developed a robust, hybrid SRC algorithm, capable of classifying sensor data in the presence of outliers and non-Gaussian noise.

4.2. Data Structure

As listed in Table 1, the input features fed to the SRC model include: (i) three AJP process parameters; (ii) 2D and 3D quantified characteristics of line morphology; (iii) the first and last few graph-theoretic Laplacian eigenvalues [17]; and (iv) image-based texture measures [49]. We note that the Laplacian eigenvalues reflect various aspects of the algebraic connectivity of the graph resulting from an image. Tootooni et al. [27], showed that the first and last few Laplacian eigenvalues could capture a significant portion of the variability of sensor data in complex systems.

Table 1. Input features, fed to the SRC-based machine learning model for the in situ estimation of line resistance.

<table>
<thead>
<tr>
<th>Feature Type</th>
<th>Component</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Process Parameter</td>
<td>ShGFR, PS, and EGFR.</td>
</tr>
<tr>
<td>2 2D Characteristics of Line Morphology [3, 4]</td>
<td>(L_W, L_p, L_{EQ}, L_{OS}, \text{and } L_{Disc}.)</td>
</tr>
<tr>
<td>3 3D Characteristics of Line Morphology [1, 2]</td>
<td>(L_T) and (L_{CSA}.)</td>
</tr>
<tr>
<td>4 Spectral Graph-Theoretic Laplacian Eigenvalues [17]</td>
<td>(\lambda_2: \lambda_6 - \lambda_{end-4}: \lambda_{end}.)</td>
</tr>
</tbody>
</table>
4.3. Sparse Estimation

As a supervised learning method [28], the SRC requires a priori labels for classification. In this study, classes of line resistance, \( C \in \{2,3,4, \ldots \} \), are defined based on four-point probe measurements (as discussed in Sec. 5.2.3 and 5.3.3). Envisioned in Figure 5, the design matrix takes the following form: \( A = [A_{1,m,n}^1 A_{m,n}^2 \ldots A_{m,n}^C] \). In the design matrix, the input features (listed in Table 1) are placed in the rows \( (i = 1 \ldots m) \), and observations or samples per class, i.e., images and/or segmented parts of the images, are placed in the columns \( (j = 1 \ldots n) \). Also, note that the total number of samples equals \( N = n \times C \). The objective of building the SRC model is two-fold: (i) to estimate the unknown vector of membership coefficients, \( \beta \); (ii) determine the class of new incoming image data, \( Y \), which indicates the range of the current resistance of a deposited line. This objective is realized by formulating a novel, combined minimization problem based on the Least Absolute Shrinkage and Selection Operator (LASSO) [37], Elastic Net (EN) [50], and Ridge Regression (RR) [51]. Mathematically expressed by Eq. (4), the minimization problem simultaneously minimizes:

(i) the sum of squared errors of estimation, i.e., the \( \ell_2 \)-norm of the fit error, \( \| Y - A\beta \|_2^2 \);
(ii) the variance, i.e., the \( \ell_2 \)-norm, of the vector of membership coefficients, \( \| \beta \|_2 \);
(iii) the number of non-zero elements, i.e., the \( \ell_1 \)-norm of the vector of coefficients, \( \| \beta \|_1 \).

\[
\hat{\beta} = \arg\min_{\beta} \left( \| Y - A\beta \|_2^2 + \lambda \left( \frac{1-\alpha}{2} \| \beta \|_2^2 + \alpha \| \beta \|_1 \right) \right)
\tag{4}
\]

In Eq. (4), \( \lambda \in \mathbb{R} \) is a regularization parameter, which controls the overall contribution of the penalty term \( \left( \frac{1-\alpha}{2} \| \beta \|_2^2 + \alpha \| \beta \|_1 \right) \). We note that the \( \hat{\beta} \in \mathbb{R}^{N \times 1} \), i.e., the solution to the minimization problem, is a sparse vector of coefficients, concatenated over all classes.
(see Figure 5), which can be mathematically defined as: $\hat{\beta}_{N,1} \equiv \begin{bmatrix} \hat{\beta}_{n,1}^1 & \hat{\beta}_{n,1}^2 & \ldots & \hat{\beta}_{n,1}^C \end{bmatrix}^T$. 

$\alpha \in \mathbb{R}$ is a weighting parameter between (0 1]; if $\alpha \approx 0$, the problem is formulated based on Ridge regression; if $\alpha = 1$, the problem is formulated based on the LASSO; and if $0 < \alpha < 1$, it is based on Elastic Net.

Finally, as mathematically conveyed by Eq. (5), the class of printed line resistance is estimated to be $\hat{C}$, if the corresponding membership coefficients of that class, i.e., $\hat{\beta}^c \in \mathbb{R}^{n \times 1}$, yield the minimum of the fit error among other classes. In Eq. (5), as discussed, $Y$ is a vector representing new morphology data given by an incoming image. Besides, $\delta_c(\hat{\beta})$ is an operator which creates a sparse vector of $\hat{\beta}$ where all elements (coefficients) are zero except those of Class $c$, i.e., $\delta_c(\hat{\beta}) = [0 \ldots \hat{\beta}_c^c \ldots 0]^T$ for $c = 1, 2, \ldots, C$.

$$\hat{C} = \arg\min_c \left(A\delta_c(\hat{\beta}) - Y\right) \text{ for } c = 1, 2, \ldots, C$$

(5)
Figure 5. The structure of Design, Validation, and Testing matrices as well as of the vector of membership coefficients, $\beta$. Each matrix/vector is a concatenation of submatrices/subvectors of all classes. The morphology features of interest are placed in the rows and samples in the columns according to percentages defined for training, validation, and testing (60%, 30%, and 10%, respectively, in this study).

### 4.4. Numerical Implantation

Figure 6 illustrates a pseudo-algorithm for the numerical implantation of the SRC method. It is comprised of four phases: (i) initialization; (ii) data preparation; (iii) validation (parameter tuning); and (iv) testing, where the last two phases run a common classification subroutine.

#### I. Initialization

The results of *in situ* image processing and quantification of line morphology are stored in a matrix, called the *Grand Matrix*, where morphology features are placed in the rows.
and samples/images in the columns. Depending on the tradeoff between the depth of learning vs. processing time, a combination of the 2D and 3D morphology features are chosen and stored as a new matrix (called the Feature Matrix). This matrix is used as the primary source of data for classification. A vector of \textit{a priori} classification labels (also known as gold standards) is defined, according to the objectives as well as resolution of classification. The partitioning percentages are specified prior to the analysis for the construction of the training, validation, and testing matrices.

\textbf{II. Data Preparation}

The proposed SRC method requires allocation of an equal number of samples per class. Therefore, if $n^C$ is the number of samples in class $C$, the overall number of samples used for the classification is: $\min(n^1, n^2, ..., n^C)$. Subsequently, according to the partitioning percentages (defined in Phase I), the training/design ($A$), validation ($V$), and testing ($T$) matrices are randomly constructed out of the Feature Matrix (as visualized in Figure 5).

\textbf{III. Validation (Parameter Tuning/Optimization)}

In the validation process, two ranges of values are initially defined for the regularization and weighting parameters, i.e., $\lambda$ and $\alpha$ respectively. To estimate the optimums of these two parameters, we utilized a grid spacing (heuristic) approach in which a 2D grid space was formed (based on the divisions of each range), and then the performance of the classification algorithm was assessed at each grid point, employing the design ($A$) and validation ($V$) matrices. The estimated values of the regularization and weighting parameters ($\lambda_{opt}$ and $\alpha_{opt}$) are subsequently used to classify the testing dataset. We note that the range of the classification parameters (i.e., the grid dimensions) should be broad enough to ensure reaching the global optimum. On the other hand, the grid spacing
influences not only the accuracy of the validation process, but also the processing time (see Figure 10 in Sec. 5.2 as an example of parameter tuning). Hence, an optimal combination of the grid size and spacing should be adopted to obtain efficient parameter estimation.

**IV. Testing**

In the testing phase, the *Classification Subroutine* is similarly employed to independently assess the performance of the trained classifier, using the untouched matrix of testing \((T)\). The *Classification Subroutine* ultimately forms a confusion matrix and calculates several measures of the classification performance, such as F-score, the area under the receiver operating curve (AUROC), overall accuracy, recall, precision, specificity, informedness, markedness, and likelihood ratios.

**V. Classification Subroutine**

The right pane of Figure 6 exhibits a pseudo-algorithm for the numerical implementation of the *Classification Subroutine*. It is composed of three parts: (i) estimation of the vector of membership coefficients, \(\hat{\beta}^c\); (ii) sparsification of the \(\hat{\beta}^c\); and (iii) classification. For each sample, which is mathematically expressed as a vector of features \((Y_{(c,j)}^c \in \mathbb{R}^{m \times 1})\) for \(c = 1,2, \ldots, C\) and \(j = 1,2, \ldots, n\) where \(c\) is a counter of classes and \(j\) is a counter of samples per class, the vector of membership coefficients, \(\hat{\beta}_{(j)}^c \in \mathbb{R}^{N \times 1}\), is correspondingly calculated. Based on the range of samples per class, \(n\), the \(\hat{\beta}_{(j)}^c\) is subsequently sparsified and saved as a new vector \((J \in \mathbb{R}^{N \times 1})\), which has nonzero elements only for the called class/label, \(c\). Then the approximation/fit error (called the *norm* mathematically), i.e., the error between the sample vector, \(Y_{(c,j)}^c\), and the product of \(A.J\) \((\in \mathbb{R}^{m \times 1})\), is calculated against all classes. The class, leading to the lowest amount of error, is marked as a predicted class. Finally, based on the actual, \(c\), and predicted, \(p\), classes, a
confusion matrix is constructed, and the main measures of classification performance are calculated.

### Figure 6

A pseudo-code for the numerical implementation of the proposed SRC method, including: initialization, data preparation, validation, and testing along with a classification subroutine. The subroutine is further composed of estimation and sparsification of the vector of membership coefficients, $\vec{\beta}$, as well as of classification and computation of performance measures.

#### 5. Experimental Results

The objective of this section is to discuss experiments, designed to systematically assess the performance of the proposed SRC method for both in situ and real-time estimation of line resistance, as delineated in Sec. 5.2 and Sec. 5.3, respectively.
5.1. Materials

A dry and pure (4.8 Grade) stream of N₂, flowing at ambient temperature (21 °C), is the primary medium of transport for both sheath and atomization gas flows. It is supplied in house.

A two-component silver nanoparticle ink, i.e., MicroPEPG007MOP and MicroPEPG007EG (Paru Co., Seoul, Korea), was used to print electronic structures (discussed in Sec. 5.2.1) for the in situ monitoring experiment. The two components were mixed with a ratio of 5:1 by weight, respectively. The first component is composed of silver nanoparticles (~66 wt%) and 1-methoxy-2-propanol (MOP) as solvent; it has a bulk density and viscosity of 1.5-2.5 g/ml and 50 cP, respectively. The second component is composed of silver nanoparticles (~33 wt%) and ethylene glycol (EG) as solvent; it has a bulk density and viscosity of 1.2-3.3 g/ml and 39 cP, respectively. We note that both of the ink components have a particle size of 80 ±10 nm.

A single-component ink (Paru Co., Seoul, Korea), i.e., PG007-AP, was used to print the electronic structures of the real-time monitoring experiment. Composed of silver nanoparticles as well as 1-methoxy-2-propanol (MOP) as solvent, this ink has an average size, particle loading, and viscosity of 100 nm, ~60 wt%, and 100 cP, respectively.

The bubbler was filled with MOP to compensate for solvent evaporation during atomization, and thus to stabilize the ink viscosity. Prior to use, the ink was ultrasonically sonicated for approximately two minutes to obtain uniform dispersion of particles.

A sheet of polyimide (125 μm Kapton, DuPont Electronic Technologies, Circleville, OH, USA) was used as a flexible substrate, recommended for the manufacturing of flexible and hybrid electronics (FHE).
5.2. *In situ* Monitoring of Line Resistance

5.2.1. **Design and Analysis of Experiments:** As detailed in Table 2, three single-factor factorial experiments were initially conducted under same experimental conditions to observe the effects of: (i) sheath gas flow rate (ShGFR), (ii) exhaust gas flow rate (EGFR), and (iii) print speed (PS) on printed line morphology and resistance. The ShGFR, EGFR, and PS were varied randomly in the range of 40-100 sccm, 400-480 sccm, and 1-4 mm/s, respectively; atomization gas flow rate (AGFR) was fixed at 500 sccm. An equilibration time of five minutes was held between each set point change of ShGFR or EGFR to ensure steady-state transport and deposition of aerosols. Utilizing the pneumatic atomizer and a 300 μm nozzle (representing the exit diameter), electronic devices composed of silver nanoparticle lines (illustrated in Figure 7) were printed (repeated three times for each treatment combination of the experimental design) in a single pass on an unheated polyimide substrate. The choice of nozzle size was based on previous studies [3, 4, 18] where pressure buildup and hence, flow stagnation were identified as phenomena adversely influencing the stability of aerosol deposition. The working distance/print standoff, i.e., the distance between the nozzle and the substrate, was kept at 3 mm in all experiments. The printed structures were oven-sintered (Binder, Inc., Bohemia, NY, USA) for one hour at 200 °C. The line resistance was measured using the four-point probe method via a source meter (Keithley-2614B, Tektronix, Inc., Beaverton, OR, USA).

Table 2. Process parameters, materials, and experimental methods used to systematically investigate the influence of each factor on line morphology and resistance.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Part I (ShGFR Study)</th>
<th>Part II (EGFR Study)</th>
<th>Part III (PS Study)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ShGFR (sccm)</td>
<td>40, 60, 80, 100</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>EGFR (sccm)</td>
<td>450</td>
<td>400, 430, 450, 480</td>
<td>450</td>
</tr>
<tr>
<td>PS (mm/s)</td>
<td>4</td>
<td>4</td>
<td>1, 2, 3, 4</td>
</tr>
<tr>
<td>AGFR (sccm)</td>
<td>500</td>
<td>500</td>
<td>500</td>
</tr>
<tr>
<td>------------</td>
<td>------</td>
<td>------</td>
<td>------</td>
</tr>
<tr>
<td>Atomizer Type</td>
<td>Pneumatic</td>
<td>Pneumatic</td>
<td>Pneumatic</td>
</tr>
<tr>
<td>Nozzle Size (µm)</td>
<td>300</td>
<td>300</td>
<td>300</td>
</tr>
<tr>
<td>Working Distance (mm)</td>
<td>3</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>Ink</td>
<td>Silver NP</td>
<td>Silver NP</td>
<td>Silver NP</td>
</tr>
<tr>
<td>Substrate</td>
<td>Polyimide</td>
<td>Polyimide</td>
<td>Polyimide</td>
</tr>
<tr>
<td>Sintering Type</td>
<td>Oven (1 h at 200 °C)</td>
<td>Oven (1 h at 200 °C)</td>
<td>Oven (1 h at 200 °C)</td>
</tr>
<tr>
<td>Resistance Measurement</td>
<td>4-Point Probe</td>
<td>4-Point Probe</td>
<td>4-Point Probe</td>
</tr>
</tbody>
</table>

Figure 7. (a) The dimensions of the AJ-printed structures; (b-d) Silver nanoparticle electronic structures, deposited on a flexible polyimide substrate at four levels of ShGFR, EGFR, and PS.

5.2.2. **Experimental Observations:** The high-resolution imaging system (introduced in Sec. 0) allows for *in situ* image acquisition from the deposited electronic structures. Once a line or a set of lines/structures has been printed, the platen is rapidly translated under the CCD camera, and then images are captured from some or all parts of the printed structures as exemplified in Figure 8.

Subsequently, the acquired images are processed using image processing algorithms [3], developed in MATLAB, Python, and C++ environments (utilized depending on the platform of choice). As categorized in Table 1 and visualized in Figure 2, five characteristics of printed line morphology (including: line width, line density, edge
quality/smoothness, overspray, and line discontinuity) in addition to eleven traits of image texture (including: mean, standard deviation, smoothness, skewness, uniformity, entropy, contrast, correlation, energy, homogeneity, and max probability [49]) are extracted from the online images. Besides, having transferred an image to a network graph, ten spectral graph-theoretic Laplacian eigenvalues, i.e., $\lambda_2: \lambda_6 - \lambda_{\text{end-4}}: \lambda_{\text{end}}$ (proposed by Rao et al. [17]), are quantified. We note that all of the aforementioned traits relate to the 2D characteristics of the line morphology.

As visually observed from Figure 8, the line width decreases significantly as the ShGFR and the EGFR increase. The rate of decrease in the line width is less prominent with regard to the PS. Also, no significant amount of overspray is observed under the set printing conditions (discussed in Table 2). The changes in the line width can be attributed to the following transport phenomena, governing aerosol transport and deposition in AJP:

(i) the ShGFR directly influences the collimation forces acting externally on the aerosol flow; (ii) the EGFR is the rate of material removal from the aerosol flow at the virtual impactor; the higher the EGFR, the lower the rate of aerosol transfer to the deposition head; (iii) PS inversely affects the linear mass density (mass per unit length) of the material deposition. These phenomena were discussed in detail in one of the authors’ recent publications [3]. Although the line width is influenced similarly (with respect to the changes in the ShGFR, EGFR, and PS), both the line thickness and the cross-sectional area (CSA) are affected differently. This stems from the fact that unlike the EGFR and the PS, the ShGFR does not change the rate of material deposition per unit length but the amount of the deposition collimation and thus density. As a result, more focused and thicker lines are deposited as the ShGFR increases; this implies that less resistive lines are obtained. The
trends of the line width vs. the ShGFR and PS are consistent with our previous observations [3, 4], indicating the repeatability of the results.

It is expected that under the optimal, experimental conditions (discussed in Sec. 5.2.1), the edge quality does not change significantly with the ShGFR/EGFR but with the PS [3]. However, Figure 8(b-ii) and (b-iv) indicate formation of rough edges after the aerosol deposition. An assignable cause was detected, revealing that such an anomaly in the edge smoothness stemmed from the presence of impurities on the surface.

Shown in Figure 8(c-i), the lines printed at the speed of 1 mm/s are of the highest quality characterized by smooth and uniform edges. The noticeable drop in the edge quality at the high PS’s is due to the fact that the deposited aerosols do not completely reach receding, relaxation, and wetting equilibrium [19].

In addition to the aforementioned traits and experimental trends, two 3D morphology characteristics, i.e., line thickness and cross-sectional area (CSA), were quantified in situ, having recovered the 3D profile of the printed lines using shape-from-shading (SfS) analysis [2], as exemplified in Figure 9.

We note that the formation of highly reflective (bright) edges at the print speeds of 1 mm/s and 2 mm/s (see Figure 8(c-i) and (c-ii)) results in the appearance of narrow valleys in their corresponding recovered surfaces (see Figure 9(c-i) and (c-ii), respectively). However, these bright edges are not significantly wide compared to the entire width of the lines. Therefore, the accuracy of the profile recovery is not affected significantly. In general, the adverse influence of the bright edges can be removed by, for example, high dynamic range imaging or diffuse illumination. We removed the background of the images.
before the analysis to obtain a uniform ground reference for the quantification of the thickness and CSA.

Figure 8. A series of images captured *in situ* using a high-resolution CCD camera, demonstrating the effects of ShGFR, EGFR, and PS on the printed line morphology.

Figure 9. *In situ* reconstruction of the 3D profile of the printed lines, using shape-from-shading analysis [2], allowing for quantification of the line thickness and cross-sectional area.
The line CSA is quantified by numerical integration of each 2D cross-section, averaged over the entire length of the line. The authors’ previous work regarding SfS [1] showed that Shah’s method [52] would perform well particularly when the direction of light source is comparable to that of camera (like our imaging system). Consequently, the Shah’s method was used in this study as the method of choice for the in situ recovery of the line cross-section. Although not used in this study, several other traits – such as surface roughness parameters (Ra, Rq, and Rt) – were additionally quantified. They could, on the one hand, aid in obtaining more accurate classification, but on the other hand, add more complexity and increased processing time to the model.

5.2.3. Classification of Line Resistance: The design matrix \((A \in \mathbb{R}^{34 \times 720})\) is composed of 34 rows, i.e., \(m = 34\) (representing the quantified morphology features detailed in Table 1), by 720 columns, i.e., \(N = 720\) (representing observations or samples). This is constructed as follows: there are four levels for each of the three factors (ShGFR, CGFR, and PS), experimentally designed to be repeated three times. From each of the 36 resulting experimental runs, an image was acquired. Each image was, subsequently, divided to 20 segments to populate enough samples for the training, validation, and testing datasets. Consequently, 720 samples were generated in total. The full field of view of the imaging system allows for capturing approximately 880 \(\mu\text{m}\) of the printed lines. Similarly, the vector of new morphology data, extracted from an incoming image, is composed of 34 rows, i.e., \(Y \in \mathbb{R}^{34 \times 1}\). Three classes of line resistance, \(c \in \{1,2,3\}\), were defined as follows: \textbf{Class 1}: \(\text{LR} < 1.2\\Omega\); \textbf{Class 2}: \(1.2\\Omega \leq \text{LR} \leq 1.6\\Omega\); and \textbf{Class 3}: \(\text{LR} > 1.6\\Omega\). 60% of the data was randomly dedicated to training, 30% to validation, and 10% to training. As discussed in Sec. 4.4, the grid spacing approach was employed to map
a 2D space and thus to find the optimal values of $\lambda$ and $\alpha$, using the validation dataset, as exemplified in Figure 10.

The results of the in situ sparse representation-based classification (SRC) of the line resistance are illustrated in Table 3 for a simulation run. The performance of the SRC classifier was initially evaluated based on F-Score, defined as the harmonic mean of precision and recall (sensitivity), as mathematically expressed by Eq. (6) [53, 54],

$$F_{\text{Score}} = 2 \times \frac{(P \cdot R)}{(P + R)}$$  \hspace{1cm} (6)

In Eq. (6), $P$ and $R$ stand for the precision and the recall, respectively.

![Figure 10. An example of the 2D space, mapped using a grid spacing (heuristic) approach, which locates the optimal values of the regularization and weighting parameters, i.e., $\lambda$ and $\alpha$.](image)

Figure 10. An example of the 2D space, mapped using a grid spacing (heuristic) approach, which locates the optimal values of the regularization and weighting parameters, i.e., $\lambda$ and $\alpha$. 
Table 3. The results of the *in situ* classification of the line resistance (for a single run). In this simulation, the LASSO was automatically chosen by the background algorithm as an optimization method, based on the value of $\alpha = 1$. The Elastic Net and Ridge Regression methods would have been alternatively selected if $0 < \alpha < 1$ and $\alpha \approx 0$, respectively.

<table>
<thead>
<tr>
<th>Sparse Estimation Classification</th>
<th>Estimated Condition</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Optimal Method: LASSO</strong></td>
<td><strong>Class 1</strong></td>
</tr>
<tr>
<td><strong>True Condition</strong></td>
<td><strong>Class 2</strong></td>
</tr>
<tr>
<td><strong>Class 1</strong></td>
<td><strong>Class 3</strong></td>
</tr>
<tr>
<td><strong>Class 2</strong></td>
<td>1.00</td>
</tr>
<tr>
<td><strong>Class 3</strong></td>
<td>0.00</td>
</tr>
<tr>
<td><strong>Recall</strong></td>
<td>1.00</td>
</tr>
<tr>
<td><strong>Precision</strong></td>
<td>1.00</td>
</tr>
<tr>
<td><strong>False Alarm</strong></td>
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</tr>
<tr>
<td><strong>Specificity</strong></td>
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</tr>
<tr>
<td><strong>Optimization</strong></td>
<td>$\lambda$ (Opt)</td>
</tr>
<tr>
<td><strong>$\alpha$ (Opt)</strong></td>
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</tr>
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<td><strong>Performance Evaluation</strong></td>
<td>F-Score</td>
</tr>
<tr>
<td><strong>F-Score</strong></td>
<td>0.99</td>
</tr>
</tbody>
</table>

To avoid the intrinsic bias of classification, resulting from the random partitioning of the training, validation, and testing datasets, the classification algorithm was run 100 times. As a result, an average F-Score of $0.95 \pm 0.005$ was obtained with consideration of a significance level of 0.05. This implies that the line resistance can be estimated *in situ* with an accuracy of approximately 95%. The performance can be further improved by optimal selection of the line morphology features. While selection of a larger number of features results in improved classification performance, it increases computation time (particularly in the training and validation phases).

As illustrated in Figure 11, the performance of the proposed SRC method was compared and contrasted systematically against that of seven well-known classifiers – including: artificial neural network (ANN) [55]; decision tree (DT) [56]; discernment analysis (DA) [57]; Naïve Bayes (NB) [58, 59]; k-nearest neighbors (kNN) [60]; support vector machine (SVM) [61-64]; and ensemble of learners (Ens) [65, 66] – vs. two, three,
four, six, and nine classes of the line resistance. This consisted of a combination of 40 simulations (8 classification methods by 5 class numbers), each repeated 100 times.

![Graph showing F-Score vs. Number of Classes](image)

Figure 11. A comparison between the performance of the SRC method and that of other well-known classification methods including: artificial neural network (ANN); decision tree (DT); discernment analysis (DA); Naïve Bayes (NB); k-nearest neighbors (kNN); support vector machine (SVM); and ensemble of learners (Ens). The error bars represent 95% confidence intervals around the F-Score means.

Figure 11 implies that as the number of classes increases, the classification performance initially increases to a maximum (where the number of classes is 3), and then decreases sharply to a minimum (where the number of classes is 6). Beyond this point, the F-Scores overall show inconsistent trends; for most of the classifiers, an increasing trend is observed. Besides, ANN was not able to classify the line resistance beyond that point; this is because both the precision and the recall were zero, resulting in an undefined value (NaN) in the calculation of the F-Score (see Eq. (6)). The accuracy of the SRC classifier is comparable to that of kNN, both following the accuracy of the Ens classifier, as expected. In addition, the SRC, kNN, and Ens classifiers exhibit a relatively low amount of variation across all classes. This is an indication of the fact that the proposed SRC classifier can be used as a robust and high-performance classifier for the *in situ* estimation of line resistance in AJP.
In addition to the comparative assessment of the performance, all the classifiers were relatively compared with respect to their computation times (including the three phases of training, validation, and testing). kNN, DT, and DA had the smallest computation times (being in the order of a few tens of seconds). Both SVM and NB exhibited intermediate computation times, i.e., approximately a few hundreds of seconds, which were one order of magnitude smaller than those of SRC and Ens classifiers. We note that such observations are merely forwarded here to provide rough estimates about the computation time of each classifier. Since their background algorithms are not optimized at the same level, no strict conclusions can be further deduced from the observed trends.

5.3. Real-time Monitoring of Line Resistance

5.3.1. Design and Analysis of Experiments: As discussed in Sec. 0, the new experimental setup configuration – utilizing the standard process monitor camera instead of the integrated CCD camera – allows for image acquisition, processing, and ultimately estimation of device functional properties in near real-time. In order to assess the robustness of the SRC method in real-time classification of line resistance, a two-factor factorial experiment was designed where the print speed (PS) was changed randomly at three levels of 2, 4, and 6 mm/s, each at two ShGFR levels of 40 and 60 sccm. In addition, each treatment combination of the experimental design was repeated five times (resulting in 30 runs/samples, in total). The two pneumatic atomization parameters, i.e., AGFR and EGFR, were set at 640 and 610 sccm, respectively. A 200 μm nozzle tip was used to print enhanced four-point probe structures (illustrated in Figure 12(c)) where all of the four pads
were post-printed at the same horizontal level with a speed of 4 mm/s to reduce the printing time and thus to minimize the adverse and uncontrolled effects of the AJP system drift.

Under constant illumination conditions, online images were acquired from the central line of the printed structures (15 mm long) while the deposition of material, with a frame rate of 50 fps and exposure time of approximately 1 ms (as exemplified in Figure 12). The field of view of the camera was narrowed to a window of 100×300 px (which approximately corresponds to 100×300 μm using a calibration factor of 1.01 μm/px) to: (i) restrict the image acquisition window only to the depth of field; and (ii) obtain an optimal image size for online processing and quantification of the line morphology. This means that only 100 μm of the line length can be observed per frame. However, 40, 80, and 120 μm are traveled between each image with respect to the set frame rate of 50 fps as well as the PS of 2, 4, and 6 mm/s, respectively. In other words, at the PS of 2 and 4 mm/s, over-sampling is occurring, while at the PS of 6 mm/s, image acquisition is influenced by unavoidable under-sampling (i.e., loss of information in areas not imaged). Hence, to prevent oversampling between images, the over-sampled areas were cropped away before the analysis, adjusted according to their corresponding PS’s. The same image processing logic and routines detailed in Sec. 5.2.2 – were applied to the cropped images to extract a wide range of morphological features.
Figure 12. (a-b) Examples of online images captured using the process monitor camera with a frame rate of 50 fps and exposure time of 1 ms, windowed to a field of view of 100×300 μm. (c) The dimensions of the printed structures.

5.3.2. Experimental Observations: After printing, the samples were baked in a convection oven at 200 °C for 1 hour (plus additional 20 minutes dedicated to temperature ramp-up). Subsequently, the resistance of the printed structures was measured using the four-point probe method, plotted vs. PS at the two levels of the ShGFR in Figure 13(a). The plotted trends indicate that the PS has much more significant influence on the line resistance (LR) at the ShGFR of 40 sccm compared to 60 sccm (almost a 30% increase in the line resistance from 2-6 mm/s at 40 sccm vs. only a 7% increase at 60 sccm). This is an implication of the fact that there is a strong interaction between the PS and the ShGFR. In other words, there exists a nonlinear relationship between the line resistance and the factors. This hypothesis was statistically tested using analysis of variance (ANOVA) as shown in Table 4, where the small p-values indicate that not only the two main factors, i.e., the ShGFR and PS, but also the interaction between them are significantly influential on
the line resistance. The contour plot shown in Figure 13(b) aids in locating the optimal values of the line resistance. For example, printing at low levels of the PS and high levels of the ShGFR results in the most conductive structures. This physical phenomenon stems from the fact that an optimal combination of the ShGFR and the PS allows for formation of a collimated flow of aerosols in a stable fashion [3, 19]. Overall, the ShGFR of 60 sccm results in deposition of narrower and thicker lines which are less sensitive to the effects of PS.

![Graph](image)

Figure 13. (a) Line resistance (LR) vs. print speed (PS), plotted at two levels of ShGFR (40 and 60 sccm). The error bars are ($\pm 1\sigma$) long where $\sigma$ is the standard deviation of the measurements; (b) A contour plot of the LR, mapped over various levels of the PS and the ShGFR. Optimal LR values can be obtained by setting the PS at a low level and the ShGFR at a high level.

Table 4. The ANOVA table for the line resistance, demonstrating that the ShGFR, the PS, as well as their interaction are statistically significant.

<table>
<thead>
<tr>
<th>Source</th>
<th>DF</th>
<th>Adj SS</th>
<th>Adj MS</th>
<th>F-Value</th>
<th>P-Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Model</td>
<td>5</td>
<td>1074.11</td>
<td>214.823</td>
<td>786.10</td>
<td>0.000</td>
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<tr>
<td>Linear</td>
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<td>914.49</td>
<td>304.829</td>
<td>1115.45</td>
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<tr>
<td>ShGFR</td>
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<td>510.72</td>
<td>510.716</td>
<td>1868.85</td>
<td>0.000</td>
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<tr>
<td>PS</td>
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<td>403.77</td>
<td>201.886</td>
<td>738.76</td>
<td>0.000</td>
</tr>
<tr>
<td>ShGFR*PS</td>
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<td>159.63</td>
<td>79.813</td>
<td>292.06</td>
<td>0.000</td>
</tr>
<tr>
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<td>24</td>
<td>6.56</td>
<td>0.273</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>29</td>
<td>1080.67</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**5.3.3. Classification of Line Resistance:** Based on the measurements of the line resistance (falling in a wide range between approximately 44-62Ω), five distinct
classification labels were defined, as follows: **Class 1:** $44.0 \Omega \leq L_R < 45.5 \Omega$; **Class 2:** $45.5 \Omega \leq L_R < 47.0 \Omega$; **Class 3:** $47.0 \Omega \leq L_R < 48.5 \Omega$; **Class 4:** $50.0 \Omega \leq L_R < 60.0 \Omega$; and **Class 5:** $L_R \geq 60.0 \Omega$. The SRC model was run 25 times (employing random data allocation per simulation) which resulted in an F-Score of $0.92 \pm 0.013$. Furthermore, a 10-fold cross-validation scheme was implemented to check the robustness of the method; an F-Score of $0.96 \pm 0.017$ was obtained. In addition to the F-Score, the AUROC (extended to multi-label classification by calculating for all combinations of labels) was used as a second measure of classification accuracy. Similarly, having run the classification model 25 times, an average AUROC of $0.96 \pm 0.006$ was obtained. Both the F-Score and the AUROC imply that using the SRC method, the line resistance can be estimated in near real-time with an accuracy of $\geq 0.90$.

6. **Conclusions and Future Work**

In this work, a new MISO machine learning model was implemented, based on the concept of sparse representation for classification (SRC). The proposed model minimizes not only the sum of squared errors of estimation, but also the variance as well as the number of membership coefficients. As a result, a combined $\ell_1$ and $\ell_2$ optimization problem is formulated, based on Least Absolute Shrinkage and Selection Operator (LASSO), Elastic Net, and Ridge regression. The robustness of the model was tested in both *in situ* and real-time monitoring cases, where in the former case, a high-resolution CCD camera was the primary means of image acquisition, and in the latter one, the AJP standard process monitor camera was utilized. Correspondingly, experiments were designed to print conductive test structures with different morphology characteristics via varying three process parameters.
(ShGFR, EGFR, and PS). The four-point probe method was used to measure the resistance of the AJ-printed structures, and define \textit{a priori} classification labels. In all classification experiments, 60\%, 30\%, and 10\% of the data was randomly allocated to training, validation, and testing, respectively. F-Score and the AUROC were two measures of classification accuracy. The performance of the SRC model was contrasted against that of seven well-known classifiers as well as tested vs. a wide range of class numbers. In addition, a 10-fold cross-validation scheme was implemented to check the robustness of the model. The results of this study showed that line resistance could be estimated both \textit{in situ} and in real-time with an accuracy of $\geq 90\%$. They also imply that the proposed model can accommodate the multivariate and heterogenous nature of AJP sensor data and thus, be used for online estimation of device functional properties.

As part of the future work:

1. The computer algorithms and routines developed in this work both for image processing and for classification will be optimized and integrated. This not only reduces the processing time, but also paves the way for implementation of closed-loop control in AJP, allowing for conformal printing of electronics with uniform functional properties.

2. Currently, about sixty 2D and 3D morphology traits can be extracted from an image. The performance of the SRC classifier can be further improved by optimal selection of the morphology features, for example, with consideration of an accuracy threshold to avoid unnecessary data processing.

3. Via computational fluid dynamics (CFD) modeling, the underlying phenomena behind experimental observations and trends will be further investigated. The development of
a CFD model also allows for mapping of velocity and pressure fields as well as particle trajectories, and thus simulation of line morphology particularly in critical conditions where conducting an experiment is time-consuming and expensive (such as at high ShGFR’s).

4. Finally, this study will be augmented to include other influential factors and process parameters (e.g., ink type, substrate type, nozzle size, platen temperature, among others).
References


Chapter 5: A Computational Fluid Dynamics (CFD) Study of Material Transport and Deposition in Aerosol Jet Printing (AJP) Process

1. Introduction

1.1. Research Goal and Objectives

The overarching goal of this work is to establish a physics-based framework for in situ monitoring and control of AJP process. In pursuit of this goal, the objective is to forward a CFD model to understand the underlying physics behind aerosol transport and deposition in AJP. The aim is to explain the causal aerodynamic phenomena behind experimental observations, using a compressible, turbulent multi-phase model developed for the flow of aerosols within the deposition head and also after the deposition on a free surface. The research hypothesis is: the aerodynamics of aerosol jet printing is captured using a compressible, turbulent multi-phase flow model.

1.2. Motivation and Significance

AJP – a droplet-based direct-write additive manufacturing technique – has emerged as a high-resolution method for the fabrication of a broad spectrum of electronic devices, e.g., optoelectronic devices [1], fine-pitch electronics [2], sensors [3, 4], and transistors [5-7]. AJP allows for fine deposition of material on non-planer surfaces (with feature sizes as small as 10 μm), and accommodates a wide range of ink viscosity (approximately 1-2500 cP). This implies that inks based on novel and advanced materials, such as graphene oxide, PEDOT:PSS, carbon nanotube, and noble metal nanoparticles, can be deposited for device fabrication. Figure 1 exemplifies electronic devices AJ-printed at the State University of New York (SUNY) at Binghamton, including: (a) an antenna printed on a flexible glass
substrate; (b) reduced graphene oxide supercapacitors (rGO-SCs) and silver interdigitated electrodes (IDEs) printed on a slim glass substrate; and (c) silver IDEs printed on a polyimide substrate [8].

Despite the unique advantages and host of applications engendered by AJP, the process is highly unstable, complex, and prone to gradual drifts. Hence, implementation of process monitoring and control in AJP seems to be inevitable. To realize this objective, the authors already forwarded an image-based framework for real-time monitoring as well as closed-loop control of the AJP process, as delineated in the prior publications [8-13]. In this regard, a wide range of experiments was carried out to explore the AJP system dynamics. This study, which is a direct continuation of the previous works, is dedicated to the development of a 3D compressible, turbulent multi-phase flow CFD model to explain the underlying physical phenomena behind the experimental observations and trends.

1.3. Review of the Related Research

There are several research works in literature, investigating various aspects of the material deposition in the AJP process. Chen et al. [14] studied the effect of droplet size on overspray, an important characteristic of line morphology. In this regard, they developed a
CFD model to investigate the principles of overspray formation as a function of the droplet size distribution and ShGFR.

Feng [15] studied the deformation of a sessile drop under an impinging jet. It was demonstrated that above a critical value, when viscous forces become dominant (compared to surface tension forces), unsteady droplet deformation occurs. Besides, Feng [16] studied the dynamics of micro-droplet impingement on a surface in regard to post-deposition phenomena of spreading, receding-relaxation, and wetting equilibrium. The results showed that when the influence of viscosity is negligible, periodic oscillation and aperiodic creeping-to-capillary equilibrium are discernable. In addition, contact angles as well as droplet viscosity were identified as two critical factors, influencing droplet bouncing after impingement remarkably. Feng, furthermore, investigated [17] the nature of negative pressure distribution during pneumatic aerosol generation with respect to geometric parameters, using a CFD model.

Discrete phase in a micro-capillary was studied in a research work by Schulz et al. [18]. They showed that as aerosol flow velocity increases, the Saffman force becomes significant, leading to radially inward movement of the particles within the capillary. Similarly, Akhatov et al. [19, 20] demonstrated that the Saffman force, drag force, as well as particle inertia are the main factors in the modeling of aerosol flow through long micro-capillaries. In another research work, Akhatov et al. [21] also presented solutions for the problem of gas flow in a slowly-converging micro-capillary.
2. **Development of a CFD Model**

In this section, we forward a 3D computational fluid dynamics (CFD) model to understand the underlying dynamics behind aerosol transport and deposition in AJP, and simulate formation of line topology under various process conditions. Building a CFD model involves: (1) geometry modeling; (2) meshing; (3) setting up the governing equations; (4) specifying the boundary conditions as well as material properties; and (5) selecting numerical schemes to solve a system of coupled, linearized algebraic equations relating cell center values (having discretized the governing partial differential equations (PDEs) of the problem). We note that the discretization of the governing equations is based on finite volume method. The simulation results are verified with the physical conditions of the problem, and then validated with offline experimental measurements. The modeling and simulations presented in this chapter were carried out in the ANSYS-Fluent environment (V. 16.2). The objective is to develop the numerical solution to a compressible and turbulent multi-phase flow problem: (i) in the deposition head (as an internal flow problem); (ii) during deposition on a rigid and movable substrate (as an external flow problem). In general, there are two types of errors, namely, discretization and linearization. The former reduces with refining the mesh, while the latter decreases by iteration. In our simulations, a linearization tolerance of $10^{-6}$ was set for all conservation imbalances. This ensures obtaining an approximate yet reliable solution to the mathematical model (consisting of the governing equations and boundary conditions).
2.1. Geometry Modeling and Meshing

As illustrated in Figure 2, the 3D geometry of the deposition head and its surrounding environment was modeled based on dimensions obtained from X-ray computed tomography (CT) imaging [8]. The modeled geometry is composed of the following components: (1) ShGF inlet; (2) upper plenum chamber; (3) lower plenum chamber; (4) combination chamber; (5) nozzle; and (6) a relatively-wide cylindrical volume (embedded beneath the nozzle to freely observe the effects of aerosol deposition). The entire volume of the geometry is then meshed using a mixture of smooth and soft quadrilateral elements compiled and refined with respect to both curvature and proximity. To obtain an accurate solution near the walls, three layers of inflation were considered on all surfaces (with a geometric growth rate of 1.2). This is to ensure that equilibrium turbulent boundary layers are covered with a sufficient number of cells [22].

2.2. Governing Equations (Mathematical Model)

2.2.1. Mass, Momentum, and Energy: A combination of density-based and pressure-based Navier-Stokes solution algorithms was used, respectively, to obtain a steady-state solution to the coupled multi-phase problem, and to bring the conservation imbalances below the specified linearization tolerance ($10^{-6}$). Once a steady-state solution has been achieved, unsteady discrete-phase models are used to track a large number of injected particles in the system, released from the CGF inlet. The conservation of energy (in addition to the conservation of mass and momentum) is added to the model as a fundamental law of fluid flow to account for the effects of compressibility at high flow velocities. Eqs. (1)-(4) illustrate the basic governing equations (in conservative
integral form) which constitute the basis for the flow of a compressible and Newtonian fluid.

Conservation of Mass:

\[
\frac{\partial}{\partial t} \iiint_V \rho \, dV + \iint_{\partial V} [\mathbf{n} \cdot (\rho \mathbf{v})] \, d\mathcal{A} = 0 \tag{1}
\]

Conservation of Momentum:

\[
\frac{\partial}{\partial t} \iiint_V [\rho \mathbf{v}] \, dV + \iint_{\partial V} [\mathbf{n} \cdot (\rho \mathbf{v} \cdot \mathbf{v})] \, d\mathcal{A} = - \iiint_V \nabla \mathbf{p} \, dV + \iint_{\partial V} [\mathbf{n} \cdot (\mu \mathbf{v} \cdot \mathbf{v})] \, d\mathcal{A} + \iiint_V S_M \, dV \tag{2}
\]

Conservation of Energy:

\[
\frac{\partial}{\partial t} \iiint_V [\rho C_v T] \, dV + \iint_{\partial V} [\mathbf{n} \cdot (\rho C_v T \mathbf{v})] \, d\mathcal{A} + \iiint_V [\rho \nabla \cdot \mathbf{v}] \, dV + \iint_{\partial V} [\mathbf{n} \cdot (\kappa \nabla T)] \, d\mathcal{A} = - \iiint_V \nabla \cdot \mathbf{p} \, dV + \iint_{\partial V} [\mathbf{n} \cdot (\mu \nabla \mathbf{v})] \, d\mathcal{A} + \iiint_V S_E \, dV \tag{3}
\]

Eq. of State:

\[ p = \rho RT \text{ (for perfect gas)} \tag{4} \]

In Eqs. (1)-(4), \( \rho \) is the fluid density [kg/m\(^3\)], \( \mathbf{v} \) is the velocity vector [m/s], \( \mathbf{n} \) is surface normal vector, \( p \) is the fluid pressure [Pa], \( \mu \) is the fluid dynamic viscosity [mPa.s], \( C_v \) is the heat capacity at constant volume [J/K], \( T \) is the fluid temperature [K], \( \kappa \) is the fluid thermal conductivity [W/(m.K)], \( R \) is the universal gas constant [m\(^3\).Pa/(K.mol)].

Besides, \( S_M \) and \( S_E \) are the momentum and energy source terms, respectively. Furthermore,
\( t, \mathcal{V}, \) and \( \mathcal{A} \), are used to respectively represent: time, volume, and area in the integrals and partial derivatives. \( \Phi \) is the dissipation function, defined as follows [23]:

\[
\Phi = \mu \left( \frac{2}{\rho} \left( \left( \frac{\partial u}{\partial x} \right)^2 + \left( \frac{\partial v}{\partial y} \right)^2 + \left( \frac{\partial w}{\partial z} \right)^2 \right) + \left( \frac{\partial v}{\partial z} + \frac{\partial w}{\partial y} \right)^2 \right) + \frac{\mu}{\rho} (\nabla \cdot \mathbf{v})^2
\]

(5)

In Eq. (5), \( u, v, \) and \( w \) are the \( x-, y-, \) and \( z- \)-component of the velocity vector, \( \mathbf{v} \), respectively.

### 2.2.2. Turbulence:

a. Model: The realizable \( k-\varepsilon \) viscose model [24] utilizing scalable wall functions was used to account for the effects of turbulence – together with viscose heating/dissipation (accounted for in the energy equation, Eq. (3)); streamline curvature; and compressibility (for more accurate prediction of free shear layers) – in aerosol flow. This is because the interaction between the central aerosol flow and the focusing sheath gas flow in the deposition head results in a combined, canonical flow characterized with secondary complex features, such as streamline curvature, rotation, and vortices. The advantage of using the realizable model is that it – unlike the standard and Re-Normalisation Group (RNG) models – allows for satisfaction of some mathematical constraints on the Reynolds stresses (which, by definition, reflects turbulent fluctuations in fluid momentum) [22, 24].

The transport equations for the realizable \( k-\varepsilon \) model is shown in Eq. (6)-(7):

\[
\frac{\partial}{\partial t} (\rho k) + \nabla \cdot (\rho k \mathbf{v}) = \nabla \left[ (\mu + \frac{\mu_t}{Pr_k}) \nabla k \right] - [\rho \varepsilon + \varepsilon_D] + \left[ \mu_t S^2 - \frac{\mu_t}{\rho Pr_k} \mathbf{g} \cdot \nabla \rho \right] + S_k
\]

(6)
where, \( k \) is the fluid turbulence kinetic energy \([\text{J/kg}]\); \( \mu_t \) is the turbulent/eddy viscosity \([\text{kg/(s.m)}]\); \( \text{Pr}_k \) and \( \text{Pr}_\varepsilon \) are the turbulent Prandtl numbers for the turbulence kinetic energy and dissipation rate, respectively [Prandtl number is a dimensionless number, representing the ratio of viscose diffusion to thermal diffusion]; \( \varepsilon \) is the turbulence dissipation rate \([\text{J/(kg.s)}]\); \( \varepsilon_D \) is the rate of dissipation resulting from the fluctuating dilatation of compressible turbulence \([\text{J/(kg.s)}]\); \( S_k \) and \( S_\varepsilon \) are the turbulence kinetic energy and dissipation rate source terms, respectively. We note that the rate of strain of fluid elements is defined as: \( S = \sqrt{2S_{ij}S_{ij}} \) where \( S_{ij} = 0.5(\nabla \mathbf{v} + (\nabla \mathbf{v})^T) \). Furthermore, \( C_{1\varepsilon} \), \( C_{2\varepsilon} \), and \( C_{3\varepsilon} \) are constants. In Eq. (6) and (7), starting from the left-hand side, the first term – also known as the unsteady term – represents the rate of change of turbulence kinetic energy (\( k \)) and dissipation rate (\( \varepsilon \)), respectively; similarly, the second and third terms reflect the convection and diffusion transport, correspondingly; the fourth term is the rate of destruction; and the other remaining terms indicate the rate of production as well as sources. Compared to other \( k-\varepsilon \) models, the realizable model reflects the spectral energy transfer more properly; this can be implicitly realized by looking at the fifth term of Eq. (7), i.e., \( \rho \left\{ \max \left( 0.43, \frac{S_k}{S_\varepsilon + 5} \right) \right\} S_\varepsilon \), where there is no dependency on the eddy viscosity (\( \mu_t \))
and thus, on the generation of turbulence kinetic energy. Besides, the destruction term in Eq. (7), i.e., \( \rho C_2 e \frac{\varepsilon^2}{k + \sqrt{\rho \varepsilon}} \), is not of singularity (because of \( \sqrt{\frac{\mu}{\rho \varepsilon}} \) in the denominator).

Once both \( k \) and \( \varepsilon \) have been obtained, the eddy viscosity – assumed to be an isotropic scalar quantity and work well for jet flows where only one of the turbulent shear stresses dominates [22] – is updated using Eq. (8), as follows:

\[
\mu_t = \rho \frac{k^2}{\varepsilon}
\]

\[
4.04 + \left( 0.33 \cos^{-1} \left( 2.45 \frac{S_{ij}S_{jk}S_{ki}}{(S_{ij}S_{ij})^{3/2}} \right) \right)^{-1}
\]

\[
\left[ k \left( \frac{S_{ij}S_{ij} + (\Omega_{ij} - \varepsilon_{ijk}\omega_k - 2\varepsilon_{ijk}\omega_k)}{(\Omega_{ij} - \varepsilon_{ijk}\omega_k - 2\varepsilon_{ijk}\omega_k)} \right) \right]^{1/2}
\]

We note that the whole term inside the outermost parentheses (called \( F_\mu \) hereafter) is constant in the standard \( k-\varepsilon \) viscous model, while here is a complex function of \( \Omega_{ij} \) as well as \( \omega_k \) – in addition to \( k \) and \( \varepsilon \) known as the turbulence fields – which are the mean rate of rotation tensor and angular velocity of the system of rotation (moving reference frame), respectively.

b. Near-Wall Treatment: In this study, we used scalable wall functions [22] to more accurately account for near-wall effects on aerosol deposition in the presence of turbulence. Based on the law-of-the-wall [25], the flow and turbulence parameters are expressed as a logarithmic function of the distance from the wall, \( y \) – or \( y^* \) in non-dimensionalized form – as shown in Eq. (9). Subsequently, a constraint is introduced, which prevents the wall
functions from deterioration in situations where $y^*$ falls below a certain threshold as a result of grid refinement (i.e., when $y^* \leq 11.225$) [22], as mathematically stated by Eq. (10). We note that the imposed constraint on $y^*$ (shown in this section for momentum transport) is similarly applied to standard energy and turbulence wall functions.

\[
\frac{\bar{v}F_\mu^{0.25}k^{0.5}}{\tau_w/\rho} = 2.39\ln(9.79y^*) \tag{9}
\]

\[
y^* = \max \left( \left( \frac{\rho F_\mu^{0.25}k^{0.5}y}{\mu} \right), 11.225 \right) \tag{10}
\]

In Eq. (9)-(10), $\bar{v}$ is the mean velocity at the wall-adjacent cell [m/s]; $\tau_w$ is the wall shear stress [kg/(m.s²)] defined based on Newton’s law of viscosity as $\tau_w = \mu \left( \frac{\partial u}{\partial y} \right)_{y=0}$ where $u$ represents the $x$-component of velocity; and $y$ is the distance from the wall [m]. The term in the left hand-side of Eq. (9), i.e., $\frac{\bar{v}F_\mu^{0.25}k^{0.5}}{\tau_w/\rho}$, represents non-dimensionalized velocity as a lumped combination of the flow and turbulence parameters.

\textbf{2.2.3. Discrete-Phase Modeling:} A coupled two-phase flow model (allowing for interphase exchanges of mass, momentum, and energy between the continuous and discrete phases) was established with the aim for: (i) steady/unsteady particle tracking in the system; and (ii) simulation and characterization of particle deposition under various process conditions. All of our discrete-phase simulations and calculations of particle trajectories were performed, having obtained a fully converged continuous-phase flow field.

Particle trajectories are calculated by time integration of the force balance equation – i.e., Eq. (11), based on Newton’s second law of motion written in a Lagrangian frame of
reference – where the forces acting on the particle are equated with the particle acceleration.

\[
\frac{\partial \mathbf{v}_D}{\partial t} = [\mathbf{f}_g + \mathbf{f}_{Dg} + \mathbf{f}_{SL} + \mathbf{f}_{VM} + \mathbf{f}_{PG} + \mathbf{f}_{Others}] 
\]  (11)

In Eq. (11), \( \mathbf{v}_D \) is the discrete-phase/particle velocity vector [m/s]. In addition, \( \mathbf{f}_g + \mathbf{f}_{Dg} + \mathbf{f}_{SL} + \mathbf{f}_{VM} + \mathbf{f}_{PG} \) represent the gravitational, drag, Saffman lift, virtual mass, and pressure gradient forces per unit particle mass, correspondingly. The gravitational force (per unit mass), \( \mathbf{f}_g \), equals \( g(\rho_D - \rho)/\rho_D \) where \( \rho_D \) represents the particle density [kg/m\(^2\)]. Similarly, the drag force, \( \mathbf{f}_{Dg} \), can be further defined as [22],

\[
\mathbf{f}_{Dg} = \frac{\mathbf{v} - \mathbf{v}_D}{24\rho_D d_D^2/(18\mu C_{Dg} Re)} 
\]  (12)

where \( d_D \) is the particle diameter [m], \( C_{Dg} \) is the drag coefficient, and \( Re \) is Reynolds number. The denominator in the right-hand side of Eq. (12) is termed as the particle relaxation time [26]. The Saffman lift force, \( \mathbf{f}_{SL} \), reflecting the lift acting on a small spherical particle in a slow shear flow [27, 28], is given by Eq. (13). Akhatov et al. [18-20, 29] demonstrated the significance of this force in modeling of high-velocity transport and deposition of droplets through long microcapillaries.

\[
\mathbf{f}_{SL} = \frac{5.2\rho \sqrt{\mu D (S_{ij})(\mathbf{v} - \mathbf{v}_D)}}{\rho_D d_D (S_{nmn}S_{mmm})^{0.25}} 
\]  (13)

Mathematically expressed by Eq. (14)-(15), the virtual mass, \( \mathbf{f}_{VM} \), and pressure gradient, \( \mathbf{f}_{PG} \), forces can be ignored in situations where the density of the continuous phase is remarkably lower than that of the discrete phase (i.e., \( \rho/\rho_D \ll 1 \)) [22]. In Eq. (14), \( C_{VM} \)
represents the virtual mass factor, set at 0.5 in this study. A more detailed review of forces acting on a microdroplet is given as part of one of our previous publications on AJP [8].

$$\vec{F}_{VM} = C_{VM} \frac{\rho}{\rho_D} \left( \vec{v}_D \vec{v} \vec{v} - \frac{d\vec{v}_D}{dt} \right)$$ (14)

$$\vec{F}_{PG} = \frac{\rho}{\rho_D} \vec{v}_D \vec{v}$$ (15)

2.2.4. Boundary Conditions: Illustrated in Figure 2, the following boundary conditions were defined for the transport and deposition of aerosols in the AJP process, modeled as a compressible, turbulent multi-phase flow problem:

(1) **Mass flow inlet** was the type of boundary set for the sheath gas flow (ShGF), being in absolute frame of reference. Based upon experimental sensor data (directly obtained from the Optomec AJ printer’s data logger), both mass flow rate and gauge pressure were set as a function of the ShGFR. In addition, two turbulence parameters, i.e., turbulent intensity and turbulent viscosity ratio, were set at 1.5% and 10, respectively. The thermal condition of the inlet ShGF imposed a temperature of 300K.

(2) **Mass flow inlet** was similarly chosen for the aerosol gas flow (AGF). The mass flow rate and gauge pressure were set, according to the experimental data. Besides, the same turbulence parameters and thermal condition were employed.

(3) **Stationary wall** with no-slip condition using the standard roughness model (having a roughness constant of 0.5) was the type of boundary set for the main body of the deposition head, which also includes the combination chamber as well as the nozzle (all being at 300K). Furthermore, “reflect” condition was the type of boundary set for the discrete phase model (DPM), utilizing the generic erosion model with a friction coefficient of 0.2.
(4) **Pressure outlet** was the boundary of choice for the atmospheric environment surrounding the jet of aerosols depositing on the substrate, with “escape” condition for the DPM. The turbulence component of the boundary was defined using backflow turbulent intensity and viscosity ratio, set at 1.5% and 10, respectively (similar to the inlet conditions).

(5) **Moving wall** with no-slip condition (utilizing the standard roughness model) was chosen to reflect the substrate conditions. In addition, “wall-film” condition was the type of boundary set for the discrete phase where the particle-substrate interactions – including four regimes of sticking, rebounding, spreading, and splashing [22] – were estimated using Stanton-Rutland model.

Figure 2 – A cross-section of the deposition head, showing various types of boundary conditions defined for the problem of aerosol transport and deposition in the AJP process.
3. Experimental Results

3.1. Materials

Supplied in house, both sheath and atomization gas flows are composed of a pure (4.8 Grade) and dry stream of Nitrogen, flowing at ambient temperature (21 °C).

A silver nanoparticle ink (Paru Co., Seoul, Korea), having a particle size of 80 ±10 nm, was used for printing four-point probe test structures (delineated in Sec. 3.2). The ink consists of the following components, mixed with a ratio of 5:1 by weight, respectively:

(i) MicroPEPG007MOP: This part is composed of silver nanoparticles (~66 wt%) and 1-methoxy-2-propanol (MOP) as solvent, and has a bulk density and viscosity of 1.5-2.5 g/ml and 50 cP, respectively.

(ii) MicroPEPG007EG: This part is composed of silver nanoparticles (~33 wt%) and ethylene glycol (EG) as solvent, and has a bulk density and viscosity of 1.2-3.3 g/ml and 39 cP, respectively.

The electronic structures were deposited on a flexible substrate, made up of polyimide (UPILEX 125S, Ube Plastics, Tokyo, Japan).

3.2. Design and Analysis of Experiments

Detailed in Table 1, a single-factor factorial experiment was designed and conducted to investigate the influence of ShGFR on line morphology. In this experiment, the ShGFR was randomly changed at four levels of 40, 60, 80, and 100 sccm. Each treatment combination of the experimental design was repeated 3 times. AGFR and EGFR were set at 500 sccm and 450 sccm, respectively; as a result, an aerosol flow rate of 20 sccm was obtained. Utilizing the pneumatic atomizer and a 300 μm nozzle, silver nanoparticle-based
devices (illustrated in Figure 3(b)) were printed on a polyimide substrate (Nitrogen-dried, having rinsed with Isopropyl Alcohol (IPA) and Acetone) at a speed of 4 mm/s. Working distance/standoff, i.e., the distance between the nozzle and the platen, was set at 3 mm. Subsequent to printing, the devices were sintered in a convection oven (Binder, Inc., Bohemia, NY, USA) at 200 °C for 1 hour. A 5-minutes equilibration time was taken between each set point change to obtain a steady-state flow of material. Prior to printing, the ink was ultrasonically sonicated for approximately 2 minutes to ensure uniform dispersion of particles in the ink medium.

Table 1. Details of the designed experiment, conducted to systematically investigate the influence of ShGFR on line morphology.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>ShGFR (sccm)</td>
<td>40, 60, 80, and 100</td>
</tr>
<tr>
<td>EGFR (sccm)</td>
<td>450</td>
</tr>
<tr>
<td>PS (mm/s)</td>
<td>4</td>
</tr>
<tr>
<td>AGFR (sccm)</td>
<td>500</td>
</tr>
<tr>
<td>Atomizer Type</td>
<td>Pneumatic</td>
</tr>
<tr>
<td>Nozzle Size (µm)</td>
<td>300</td>
</tr>
<tr>
<td>Working Distance (mm)</td>
<td>3</td>
</tr>
<tr>
<td>Ink</td>
<td>Silver NP</td>
</tr>
<tr>
<td>Substrate</td>
<td>Polyimide</td>
</tr>
<tr>
<td>Sintering Type</td>
<td>Oven (1 h at 200 °C)</td>
</tr>
</tbody>
</table>
3.3. **Experimental Observations**

Using the high-resolution imaging system (delineated in Chapter 1), online images were acquired from the central trace/line of each printed device, as exemplified in Figure 4. It is evident that as the ShGFR increases from 40 sccm to 100 sccm, the line width decreases. This implies that under steady aerosol generation and transfer, thicker lines are deposited as a result of the increase in the ShGFR. Besides, no significant amount of overspray was observed in the range of 40-100 sccm. This is an indication of stable and collimated material deposition.
Figure 4 – Influence of the ShGFR on the printed line morphology. An increase in the ShGFR leads to deposition of narrower and thicker lines. Insignificant overspray formation is indicative of stable, collimated aerosol deposition.

3.4. Validation of the CFD Model

Using the 3D-CFD model forwarded in this study, the collimated deposition of aerosols on a free surface was simulated at the four levels of the ShGFR, as demonstrated in Figure 5. The changes in the line width were contrasted against both online and offline experimental observations, plotted in Figure 6. The online measurements were based on the images captured using the CCD camera (discussed in Sec. 3.3), quantified using algorithms already developed in one of the authors’ prior research works [8]. The offline measurements were obtained by a white light interferometer (Wyko NT1100, Veeco Corp., Plainview, NY, USA).
Figure 5 – A CFD simulation of the collimated deposition of aerosols on a free surface at four levels of the ShGFR. Consistent with experimental observations, narrower lines are obtained as the ShGFR increases.

Figure 6 – Validation of the CFD simulation of line width vs. ShGFR with both online and offline experimental measurements.

The trend estimated by the CFD model closely mimics that of the offline measurements. In other words, there is not statistically significant difference between the two trends ($\rho=0.93$ where $\rho$ is the Pearson correlation coefficient). We note that the trend captured by the online images is slightly different, stemming from the image-based measurements of the line width. Generally, image acquisition at higher magnification results in more accurate quantification of line width.
4. Simulation Results

Using the CFD model, several simulations were carried out to understand the underlying phenomena behind experimental observations. They were focused on the flow of material: (i) within the print head and nozzle; (ii) during the deposition on a free surface; and (iii) after the deposition (where the deposited aerosols experience the phenomena of coalescence, spreading, receding, relaxation, and wetting equilibrium [16]).

Figure 7 depicts the influence of the ShGFR on the aerosol flow streamlines as well as velocity profile within the combination chamber. As the ShGFR increases, the flow of aerosols gets more collimated in the form of a narrow beam, which is further accelerated when passing through the nozzle. Based on the aerosol flow diameter at the inlet and outlet of the combination chamber, a dimensionless variable is defined, called the \textit{collimation ratio}, representing the ratio of the inlet flow diameter to the outlet flow diameter. An increase in the collimation ratio is observed from approximately 4 (at the ShGFR of 40 sccm) to 5 (at the ShGFR of 100 sccm).
Figure 7 – Simulation of the collimation of aerosols within the deposition head with the aid of the sheath gas flow (ShGF). The aerosol flow is further accelerated by the nozzle before deposition.

To assess the stability of the aerosol flow, Reynolds number (Re) was monitored at the combination chamber exit as well as at the nozzle exit, plotted in Figure 8 as a function of the ShGFR. It turned out that the collimated flow of aerosols would remain laminar even at the ShGFR of 100 sccm. This implies that stable material transfer and deposition happen with respect to the set experimental conditions (discussed in Table 1).
A comparison of the jet of aerosols (exiting from the nozzle) at the four levels of the ShGFR is demonstrated in Figure 9. The jet width decreases as the ShGFR increases. This is an indication of narrower and thicker deposition of the material on the substrate (as observed experimentally in Sec. 3.3). We note that the streamlines ultimately diverge near the surface due to the surface barrier and pressure accumulation. This phenomenon gets less prominent as the collimation power exerted by the sheath flow increases (See Figure 9(d)).
Figure 9 – Simulation of the deposition of aerosols on a free surface under the influence of the sheath flow. As the ShGFR increases, the jet width decreases, indicative of formation of narrower and thicker lines.

The impact of the aerosols on the substrate was investigated, utilizing the Weber number (We) – defined as the ratio of aerodynamic forces to surface tension forces – as illustrated in Figure 10. We note that there exists a wall film due to the continuous deposition of the aerosols. Since the substrate is not heated, the cold-wall-with-film model (also known as cold-wetting [22]) can properly reflect the physics behind the aerosol impingement. Under the presence of the wall film, when the Weber number is low (We ≤
2, see Figure 10(a-b)) for example), the impinging aerosols are reflected. On the other hand, at high Weber numbers, the aerosols stick to the substrate and contribute to the wall film formation (see Figure 10(c-d)).

Figure 10 – Impingement of aerosols on a cold surface with film under various levels of the ShGFR. At large Weber numbers ($\geq 2$), the incoming aerosols stick to the substrate.
Figure 11 – Aerosol distortion from a spherical shape upon impingement on a surface vs. ShGFR, captured using normalized displacement, based on the Taylor analogy breakup model. The edge aerosols experience less distortion.

The Taylor analogy breakup (TAB) model [22] – where the droplet surface tension forces, drag force, and viscosity forces are analogous to the restoring force, external force, and damping force of a spring mass system, respectively – was employed for post-deposition analysis of the aerosol oscillation and distortion. Visualized in Figure 11, the normalized displacement represents the displacement of the aerosol equator from its undisturbed position (normalized with respect to the initial, undisturbed equator). Note that those aerosols, deposited along the center of the line, are distorted much more significantly than the edge aerosols, remaining almost undistorted upon impingement. The magnitude of the distortion increases with the ShGFR.
5. Conclusions and Future Work

In this work, a 3D compressible, turbulent multi-phase flow CFD model was forwarded to explain the aerodynamics behind aerosol transport and deposition in the AJP process. A single-factor factorial experiment was conducted to not only investigate the influence of ShGFR on line morphology, but also validate the model. There was a good agreement between the CFD estimation of line width and the experimental measurements. The following conclusions were deduced from the results of this study:

- As the ShGFR increases, the flow of aerosols gets more collimated in the form of a narrow beam, which is further accelerated when passing through the nozzle. As a result, narrower and thicker structures are deposited on a substrate.
- The Reynolds numbers, calculated at the combination chamber exit as well as at the nozzle exit, showed that stable material transfer and deposition would happen with respect to the set experimental conditions.
- Under the presence of wall film, at low Weber numbers, the impinging aerosols are reflected, while at high Weber numbers, they stick to the substrate and contribute to the wall film formation.
- The edge aerosols remain almost undistorted upon impingement, while those deposited along the line center, are distorted much more remarkably.

As part of the future work, this CFD study will be expanded with incorporation of more process parameters, such as print speed, atomization gas flow rate, exhaust gas flow rate, and nozzle size as well as ink material properties. This will pave the way for the realization of physics-based monitoring and control of the AJP process.
References


