AEROSOL JET PRINTING OF ELECTRONICS: AN ENABLING TECHNOLOGY FOR WEARABLE DEVICES

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ABSTRACT

Additive manufacturing has revolutionized the way products are designed and fabricated to include the field of printed electronics. Direct write (DW) technologies used to print three-dimensional (3D) electronic and sensor devices have experienced spectacular growth due to their capability to offer rapid prototyping of high-performance devices for a broad range of applications. This growth is driven by many factors to include significantly reduced design-to-product lead time and fabrication of complex geometries on conformal and flexible substrates. Originally developed by the Defense Advanced Research Projects Agency (DARPA) Mesoscopic Integrated Conformal Electronics (MICE) Program for the fabrication of mesoscale electronics, DW technologies have been explored for a range of applications including active and passive components, sensors, 3D structures, as well as applications in biology.

This paper focuses on one emerging DW approach, Aerosol Jet Printing (AJP), as a non-contact method to print fine features using different types of materials over various surfaces. Aerosol Jet systems are able to print a wide variety of electronically, optically, and biologically functional materials on geometrically complex substrates that can be conformal, flexible, and stretchable. The Aerosol Jet process utilizes printable inks based on solutions or nanoparticle suspensions and can include metals, alloys, ceramics, polymers, adhesives, and/or biomaterials. A wide variety of substrates, to include silicon, polyimide, glass, FR-4 and aluminum oxide can be used to print these materials provided the ink is compatible with the substrate.

Like other DW technologies, the AJP process offers the distinct benefit of fabrication without conventional masks, with a reduction in material consumption due to selective deposition of inks at digitally defined locations on the substrate. Use of this additive process eliminates the waste of hazardous materials used in the etching processes employed by subtractive methods. AJP systems use an atomizer to create a dense aerosol of micro-droplets that are focused into an aerosol stream, resulting in deposits that can be one tenth the size of the nozzle opening at a standoff height of up to 5 millimeters. These capabilities enable the fabrication of highly integrated devices expanding from the originally targeted mesoscale application to micro- and nano-scale applications.

Design and innovative fabrication of more connected and "smart" products can be realized using AJP to meet the miniaturized, flexible, and conformal form factors desired in today's Internet of Things (IoT) global marketplace. AJP technology has opened up new avenues for bio-integrated electronics to include electronic textiles, wearable electrochemical systems, electronic epidermal tattoos, and permanent and dissolvable implantable devices. While it has been demonstrated that AJP is an enabling technology in the growing field of wearable devices, there are major challenges in widespread adoption of this innovative approach. This paper provides an overview of AJP technology and summarizes the historical underpinning of its development, underlying principles of its technique, and challenges presented in widening its adoption with industry.

Key words: Aerosol Jet Printing, Direct Write, Printed Electronics, Internet of Things, mesoscale electronics, flexible hybrid electronics, bio-integrated electronics, electronic textiles, wearable electrochemical systems.

INTRODUCTION

The electronics industry is continuously searching for advancements in materials and processes to lower cost, reduce lead times, and increase complexity within shrinking form factors. Today's conventional material sets and processes are becoming limited in enabling achievement of these objectives for electronic and sensor devices. Additive Manufacturing (AM) is revolutionizing the way that products, including electronics, are being designed, manufactured, and used today. The introduction of AM technology in the late 1980s has led to the development of new materials and processes that can be applied to the electronics sector [1]. The ability to perform rapid prototyping of electronics provides an edge on time-tomarket for new product development.

Utilizing AM processes for three dimensional (3D) printed electronics eliminates the time involved to generate new designs and design iterations, to include the time to send designs for outsourcing and back to perform concept validation and rapid prototyping. Utilizing in-house AM processes also enables companies to minimize valuable intellectual property (IP) from falling into the wrong hands. This paper will explore AM processes commonly used in the 3D printing of electronic and sensor devices and will focus on one specific emerging approach used in the applications of highly integrated devices. A summary will be provided of the historical underpinning of this technology's development, underlying principles of its technique, and challenges presented in widening its adoption with industry.

DIRECT WRITE PROCESSES

Multiple AM processes have been developed for printing electronics. Many of these processes fall into the Direct Write (DW) category with the actual method or technique used varying based on the complexity and end requirements of the electronic product being printed. The most common techniques used in industry today include aerosol jet (AJ) technology, piezo drop on demand (DOD) inkjet printing, and precision micro-dispensing. For applications specific to printing non-planar, conformal, and flexible electronics, the AJ technology process provides many advantages over other AM techniques due to its ability to selectively deposit ultrafine print feature line widths at 30 microns and below . This unique capability offers rapid prototyping for 3D printing of size-sensitive, high performance, highly integrated electronics applications such as those in the bio-integrated electronics market where device density is significantly increasing.

Direct Write Background

Much of the early work in DW technologies was accomplished through funding from Government agencies such as the Defense Advanced Research Projects Agency (DARPA), Office of Naval Research (ONR), and National Science Foundation (NSF). DARPA's first AM program, the Solid Freeform Fabrication (SFF) program, was launched in 1990 to support "tool-less" fabrication and tightly coupled design and fabrication tools.

In 2002, the DARPA Mesoscopic Integrated Conformal Electronics (MICE) program was started to develop technologies capable of rapidly prototyping customized electronic parts with a single piece of equipment from digital computer aided design (CAD) data through deposition of a wide variety of materials on any type of substrate to include conformal substrates and low temperature substrates. This enabled the printing of miniaturized and rugged mesoscale electronics without the use of masks on any surface, conformal and non-conformal, through the 3D integration of active and passive components [2]. It was from this work that multiple DW techniques were developed and patented.

Direct Write Overview

DW technologies have found a wide application over the past two decades to additively manufacture printed electronics, microelectronic devices, micro-engineered components, and potential for a number of other applications as well [3]. The use of DW processes has many advantages in electronics applications over the use of conventional subtractive manufacturing processes. A conventional lithographic-based subtractive process is followed to manufacture electronics as illustrated in Figure 1 and starts with a base substrate (step a). A conductive layer, such as copper (Cu), is deposited onto the substrate (step b) followed by deposition of a photoresist layer (step c). Ultraviolet (UV) light and a mask, created uniquely for the specific design, are used to transfer the conductor pattern on top of the photoresist layer (steps d and e). Lastly, an etching process is used to remove the conductive layer (step f) under the undeveloped photoresist material leaving behind the conductive pattern on the substrates (step g). This multi-step process, involving customized masks and removal of various layers, i.e. waste, adds time and cost to the manufacturing lifecycle in addition to the deposition of harmful residual chemicals into the environment [4].



Figure 1. Conventional subtractive manufacturing process for electronics [4].

The DW process is an attractive alternative to conventional subtractive manufacturing processes used for electronics, microelectronic devices, and micro-engineered components such as microelectromechanical systems (MEMS) and microfluidic devices. While there are differing opinions on the definition of DW, it is defined for purposes of this paper as "additive techniques enabling the deposition of electronic components and functional or structural patterns, out of different kinds of materials, directly following a preset layout in a data driven way without utilizing masks or subsequent etching processes" [4]. With the structure of the product being achieved during the DW deposition process, the material properties are developed through a post-processing step which typically involves curing or sintering [5]. While a variety of DW methods and processing techniques have been developed over the last two decades, most of these methods and techniques can be grouped into one of four techniques: (1) droplet-based direct writing; (2) flow-based direct writing; (3) tip-based direct writing; and (4) laser-based direct writing.

Droplet-based direct writing consists of two main subcategories, inkjet printing and AJ printing, both of which rely on ejecting droplets of liquid material from a nozzle(s). The inkjet printing sub-category can be further broken down into two key technologies, continuous jetting and DOD. Next, the DOD technology can be categorized into two types of actuation methods, piezoelectric inkjet nozzles and thermal inkjet nozzles. Microfab and NanoDimension are two commercial manufacturers of piezoelectric DOD inkjet printing methods using droplet-based direct writing.

The AJ technology sub-category, commonly known as the Aerosol Jet Printing (AJP), consists of an atomizer and deposition head for depositing material at a relatively large

standoff height on a substrate [4]. Optomec and Integrated Deposition Systems (IDS) are two manufacturers that have commercialized this technology into their Aerosol Jet© and Nanojet systems, respectively [28]. AJP will be discussed in further detail in another section of this paper.

Flow-based direct writing (FBDW) differs from dropletbased direct writing methods in that the material continuously flows instead of being jetted as droplets. This allows the material viscosity to vary over a greater range. A tightly controlled air pressure system and dispensing/writing tip, such as those commercially available from MicroPen and nScrypt, are used to precisely deposit material onto a variety of substrate types to include flexible and non-planar types [4].

Tip-based direct writing uses a dip-pen nanolithography (DPN) technique. Using capillary effects, an atomic force microscope (AFM) tip can transport material onto a substrate, having an affinity for the material, after the AFM tip having being dipped into the material. This technique is particularly useful in nanoscale applications having the ability to create features as fine as 12nm in line width. A modified version of DPN, thermal dip-pen nanolithography (tDPN), uses an AFM tip compatible with heat which melts the material previously deposited on the AFM tip [4].

Laser-based direct writing methods rely on the use of a laser in the processing of material to that it can be deposited on the substrate. While there are many laser-based processes, only those that are used in an additive technique are discussed. These can be broken into three sub-categories: laser-guided direct write (LGDW) system, flow-guided direct write (FGDW) system, and laser induced forward transfer (LIFT) technique [4], to include its further development referred to as matrix-assisted pulsed laser evaluation – direct write (MAPLE-DW) [5].

| Table 1. | Comparison | of Direct-Write | techniques and |
|----------|------------|-----------------|----------------|
| methods | [4]. | | |

| DW techniques | | Resolution | Material viscosity range (Pa·s) or types | | Writing speed | 3D capability* | |
|----------------------|---------------|------------|--|---|---------------------|------------------------------|-----|
| Inki | Inkjet | Continuous | Droplet size 20µm- 1mm typically 150 µm | <0.01 | | 60mm3/s | •• |
| Droplet- based DW | | DOD | Droplet size 15 - 200µm | <0.04 | | 0.3mm3/s | |
| | Aerosol Jet | | Line width 10- 150µm thickness 10nm-5µm | <2.5 | | 0.25mm3/s (single nozzle) | ••• |
| FBDW | | 50µm | <1,000 | | 50mm/s typically | ••• | |
| Tip-based DW | Tip-based DPN | | 12nm line width and 5nm spatial resolution | Thiol molecules, macromolecules, nanoparticles | | 0.2-5 µm/s | • |
| Laser | 1 | GDW | 2 µm | Non-absorbent droplets and solid particulates | Close to that of | 10-4mm3/s | •• |
| beam DW | FGDW | | 25µm | Atomizable fluids and colloids | Jet | 0.25mm3/s | •• |
| | | LIFT | 10-200um | Solids | | 3 -50mm/s | •• |

* • little 3D capability, •• moderate 3D capability, ••• excellent 3D capability

AEROSOL JET PRINTING (AJP) PROCESS

Like other DW technologies, the AJP process was developed during the MICE project in the early 2000s that was funded by DARPA [6]. The most notable of the AJP systems developed included the Aerosol Jet[®] and Nanojet systems that were commercialized by Optomec and IDS, respectively [28]. For the purposes of this paper, the Optomec Aerosol Jet[©] system will be further explored in detail.

AJP Background

Dr. Michael Renn developed the AJP technology through work supported by DARPA [12]. Dr. Renn joined Optomec in 1999 as the Principal Investigator on the DARPA MICE project that launched the AJ technology [11]. Dr. Renn initially applied in 1998 for the patent of a "DIRECT WRITETM SYSTEM" with award of Pat. No 7,270,844 in 2007 [7]. Dr. Renn and the Optomec team also received a patent in 2006 for "Apparatuses and Method for Maskless Mesoscale Material Deposition" with award of Pat. No. 7,045,015 [8]. Optomec commercialized the direct-write aerosol printing technique with their Maskless Mesoscale Materials Deposition (M3D) system in 2004 and later trademarked their system as Aerosol Jet® [9] [13].

Today, a number of Aerosol Jet[®] systems area available to develop, fabricate, enhance and repair high performance electronic and biologic devices. Optomec's Aerosol Jet[®] systems are utilized in a number of different industries to include semiconductor packaging, displays, consumer electronics, aerospace products, defense applications, automotive, and life sciences [19].

AJP Methodology

AJ systems utilize an aerosol stream to focus, deposit, and pattern the deposition material onto the substrate. The AJ system consists of the following subsystems: (1) an atomizer that creates a dense mist of "material (ink)" droplets, (2) a carrier gas is introduced to transport the aerosol, (3) an inflight transport conditioning channel for refinement, (4) a flow guidance deposition head for focusing the aerosol stream, (5) deposition of ink droplets through a nozzle, and (6) computer-controlled translation of the substrate for printing on to non-planar surfaces. The mist of ink droplets is aerodynamically focused in the system and collimated with a co-axial flowing sheath gas stream resulting in a mist jet that can become up to 10 times smaller than the diameter of the nozzle orifice [6] [9]. This high-speed stream of ink material uses inertial impaction of the ink droplets onto the substrate after the impinging jet leaves the deposition nozzle.

A schematic of the AJ process is shown in Figure 3 below. The co-flowing sheath gas stream also serves as a method of preventing direct contact of the ink droplet stream with the nozzle channel wall. This is significant in preventing clogging of the nozzle. The 3D patterning deposition of the AJ system is accomplished with coordinated movement of a motion control system with data input by the toolpath generated with a CAD software interface. This coordinated movement controls relative motion of the substrate with respect to the deposition nozzle. Lastly, patterning is accomplished with the use of a mist-diverting device (not shown in Figure 2) that acts as a "shutter" to interrupt the deposition of ink onto the substrate from the continuous mist stream [6].



Figure 2. Schematic of the Aerosol Jet process [6].

One of the advantages of the AJ process is its unique ability to deposit materials on planar and non-planar substrates. This unique capability is attributed largely to the relatively high standoff (3-5 mm) of the deposition nozzle above the substrate and the long focal length of the deposition stream exiting the nozzle. This enables the AJ process to build 3D features onto non-planar (shaped) substrates, print material into trenches, or print material over steps and contours [3].

AJP Materials

The AJ systems are designed to dispense a wide variety of organic and inorganic materials onto a number of different substrates. Materials such as metals, conductors, insulators, ferrites, oxides, polymers, adhesives and biological materials are but a few examples of materials that can be deposited using the AJ process. These materials can be deposited on nearly any surface to include silicon, glass, plastics, metals, ceramics, polyimides and polyesters [9]. AJ technology equipment manufacturer, Optomec, provides a list of known materials (see Table 2) that can be used with their Aerosol Jet[®] systems. A number of post treatment methods ranging from traditional oven sintering to UV curing, and even laser sintering, enable these materials to become functional [17].

 Table 2. Commercially-available materials supported by

 Aerosol Jet© technology [17].

| | 0/L'] | |
|--|--|---|
| METAL INKS | RESISTOR INKS | NON-METALLIC CONDUCTORS |
| Gold Platinum Silver Nickel Copper Aluminum | Carbon Ruthenate | Single Wall Carbon Nanotubes Multi Wall Carbon Nanotubes PEDOT:PSS |
| DIELECTRICS AND ADHESIVE | SEMICONDUCTORS | OTHER |
| Polyimide Polyvinylpyrrolidone (PVP) Teon AF SU-8 Adhesives Opaque coatings UV adhesives | Organic semiconductors Single Wall Carbon Nanotubes Organic semiconductors | General solvents, acids & bases Photo and etch resists DNA, Proteins, Enzymes |

Materials to be dispensed must be formulated such that they can be adequately atomized into a dense mist with a large quantity of fine microdroplets [6]. This translates to a material that must have a viscosity lower than 1000 centipoise (cP). For conductors, this equates to material selection of inks with a viscosity less than 1000 cP and capable of resulting in a resistivity equal to or greater than that of the bulk material [10]. There is a balance to tailoring ink formulations, through dilution, in order to accomplish these two goals [6]. Dilution includes adding a prescribed volume of solvents and/or polymer additives to the ink formulation. Variables of this tailoring process include filler particle size (small), distribution of filler particle sizes (wide), and amount of solvents and additives (reduced viscosity) [10]. Figure 3 below illustrates the key components of printable ink formulations for electronics [14].



Figure 3. Key components of printable inks for electronic devices [14].

Negative effects of an improperly formulated ink include: difficulty of controlling print quality and undesired spreading of printed feature edges (too much solvent), clogged nozzle (too wide distribution of particle sizes), and/or increased resistivity after cure [10]. It is key to create a balance of solvent and cosolvents in the ink such that it allows the material to be atomized, removal of a portion of the volatile solvent during the in-flight transport conditioning (evaporation), and the remaining nonvolatile solvent to result in a cohesive printed feature. This key formulation of an ink material is what facilitates the printing of high aspect-ratio features and sophisticated 3D microstructures [6]. AJ technology equipment manufacturer, Optomec, provides a design for manufacturability guideline (see Figure 4) for selecting material formulations to use with their aerosol jetting systems [20].



Figure 4. Optomec design for manufacturability guidelines for ink materials used with aerosol jetting systems [20].

AJP Applications

A multitude of applications have been recognized as printable using AJP technology. A significant amount of work has been performed to date in electronics applications, particularly given the underpinning foundation and funding for AJ technology, the DARPA MICE program, was to develop a method to print mesoscale electronics using a maskless process. While most work has been focused on printing electronics, work has also been performed in the medical industry for biological applications and the promising application of printing 3D microstructures [15].

With industrial developments such as the Internet of Things (IoT), 3D printing of electronics devices has become an attractive alternative to traditional methods. AJP technology offers a viable method to enable the creation of more connected and "smart" products in the miniaturized, flexible, and conformal form factors desired [15]. The ultimate goal is create a single printed system solution for electronics of the future. However, individual capabilities must be developed first. Electronics components can typically be categorized into three types: passives, actives, and sensors. Examples of printable passive components include resistors (including high-power [10]), capacitors (including supercapacitors [14]), inductors, interconnects, antennas, waveguides, and other types of transmission lines. Given the relative simplicity of these types of components, they are attractive for 3D printing and are relatively achievable for printing with AJP processes [15]. For example, 3D printed antenna systems have been extensively studied with the attractiveness of printing directly onto structures, to include planar, non-planar (curved), and stretchable surfaces to create a highly integrated system [10].

Active electronic components tend to be more complex than passive components due to being comprised of multi-material structures. Examples of printable active components include transistors, switches, organic light-emitting diodes (LEDs), photovoltaics, batteries, and fuel cells (including biofuel cells). Currently, the 3D printing of active components is implemented with a hybrid process of both additive and traditional subtractive processes. AJP technology provides the advantage of active component implementation on nonstandard substrates such as paper, fabric, and lowtemperature polymers [15]. For example, research is actively being performed in the area of energy harvesting systems and power sources for wearable end applications thus driving the need for smaller, lighter weight, and more compliant for body-integration applications [14].

The ability to embed sensors for highly integrated systems is driving considerable development of printed sensor electronic components. Examples of sensor components include strain gauges, capacitive sensors, temperature sensors, dielectric elastomer devices, photodetectors, and chemical sensors (including biosensing). The high demand for IoT devices and sensors is a key driver for taking advantage of the benefits of rapid prototyping, highly integrated, and substrate-agnostic printing capability using AJP technologies [15]. For example the field of personalized IoT devices includes wearables that must be intimately mated with the human body for various applications such as healthcare, sports, fitness, and wellness [14].

AJP Strengths and Weaknesses

AJP technology offers the potential to provide advantages over traditional subtractive processes for manufacturing electronics. A list of advantages and benefits of using this specific DW technique is provided below [10] [16] [19].

- Design flexibility with direct output from CAD/CAM
- Rapid product development with quick-turn of new or modified design features
- Eliminates additional tooling, i.e. no masks, resists, or custom stencil tooling required
- Selective deposition reduces use and disposal of hazardous and toxic substances
- Reduced overall number of processing steps, i.e. no plating or etching required
- Reduced energy consumption (potentially) from simplified manufacturing process
- Fine print line width feature sizes from 30 microns to millimeters
- Print thickness from 100 nanometers to 10's of microns
- Large nozzle standoff distance for conformal printing on non-planar surfaces
- Print on plastic, ceramic, and metallic substrates
- Print active, passive, and sensor components
- Print bio-materials
- Print using commercially-available materials, i.e. up to 1000 cP in viscosity
- Scalable (potentially) to support high-volume production requirements

While there are numerous advantages to using AJP technology, there are still challenges to be overcome before further breakthrough can be achieved in terms of building confidence in AJP technology and facilitating widespread adoption. This DW technique's materials and processes must continue to be refined in order to address shortcomings and disadvantages. A list of these known issues and opportunities for improvement are provided below [10] [15] [16].

- Assessing print quality of deposited ink material
- Minimum print feature size is 10 um but unreliably until >100 um
- Maximum print feature size is 150 um
- Wetting characteristics may require substrate surface preparation
- Only utilize inks with viscosity in range between 1 and 1000 cP
- CAD design tools need maturing for AJP interoperability
- Conductive inks require sintering to maximum conductance resulting in substrate and/or other component damage in addition to conductor cracking for substrates with a coefficient of thermal expansion mismatch

- Proprietary equipment resulting in lack of material and process modeling
- Lack of standards and quality metrics for assessing print quality and reliability

AJP Challenges

Even with all the recent advancements in the printed electronics technology arena, there are still challenges facing the widespread commercial adoption of this technology. A number of these challenges are discussed below. While some of these challenges can be overcome with innovative approaches and materials, others are examples of how this technology is still in its early stage with more research and development to be performed [23].

- 1. Manufacturing, Assembly and Integration: Improve pick and place tooling compatibility with foundry-based thinned devices in a hybrid integration with flexible substrates and interconnects.
- 2. System Integration: Demonstrate thermal management capability and packaging concepts.
- 3. Innovative Printing Processes: Continue reduction in feature sizes with emerging printing processes and multi-layer print capability.
- 4. Materials Manufacturing and Scale-up: Challenges with materials scale-up, through layer vias, and reliability due to CTE mismatch and delamination.
- 5. Modeling and Design Tools: Need development of comprehensive tool sets and modeling algorithms.
- 6. Education and Training: Curriculum development partnering with existing STEM programs and access to design tools and software.
- 7. Standards, Testing and Metrology: Need defined standards on inspection and test for low and high production (i.e. in-line high speed and automated quality control tools).

One of the greatest challenges with AJP technology involves understanding, predicting, and selecting the multiple parameters that affect print quality of deposited ink material. There are a number of factors of which print quality using this technique is reliant upon as shown below in the fishbone diagram in Figure 5. Material and ink formulation present the largest challenge to 3D printing electronics using AJP technology [15].



Figure 5. Factors affecting print quality using AJP technology [15].

FUTURE OUTLOOK

Additive Manufacturing (AM), particularly Aerosol Jet (AJ) printing and similar advanced micro dispensing developments, are key technologies for enabling previously impossible designs to become realities. Furthermore, when these 3D print technologies are combined with flexible and stretchable substrate materials, the potential applications become more broad and fascinating with the capabilities they enable. When engineers can design and fabricate structures that are truly conformal in three dimensions, i.e. soft, flexible and even stretchable, it's not much of a leap to begin to imagine that "human like" structures can be created. These can include wearable apparel, external on the surface of skin devices and even devices implanted within the body. Historically, biomedical applications have not been able to fully take advantage of the latest state of the art electronics and senor technologies due largely to the traditional rigid and bulky form factors they are packaged within and the inherent incompatibility with the human physiology.

Daily inroads are being made in the printed electronics arena for printing on flexible and stretchable substrates. These advancements are due largely to the collaborative work across industry, government, and academia such as the Department of Defense's (DoD) Manufacturing USA Institute for Flexible Hybrid Electronics (FHE) known as NextFlex. NextFlex was formed in 2015 to bring together large and small companies for teaming up with academic institutions, nonprofits and governments with one mission to advance the flexible hybrid electronics' manufacturing ecosystem in the United States [22]. There are additional consortiums that have been established to help further development of FHE technology and printed electronics. While NextFlex serves as the manufacturing consortium, two other consortiums exist with a focus in their respective research and development areas (see Figure 6). One of the long standing consortiums is FlexTech whose mission is to provide end-to-end support from materials providers to applications users. The other is the Nano-Bio Materials Consortium (nbmc) whose mission is to create an integrated suite of nano-bio materials technologies and transition them to production [21].



Figure 6. FHE consortiums [21].

FHE take advantage of the soft flexible properties enabled by printing, e.g. AJ technology or micro dispensing, onto soft stretchable substrates to resolve many of the challenges associated with these materials. Noteworthy applications exist within both the commercial and military sectors for these type of innovative manufacturing techniques. Much overlap or dual use between the commercial and military sectors exist for leveraging lessons learned in materials and processes, however, the end use environmental requirements, ruggedness, and reliability typically differ. Some examples of current work being performed in each of these sectors is provided below.

Bio-Integrated Epidermal Tattoos

One recent revolutionary development in bio-integrated electronics is the fabrication of 'epidermal' electronics that enable wireless data transfer from biosensors. The incredible work being performed by Dr. John Rogers and his team at Northwestern University has led to the development of a dual wireless sensor (see Figure 7) that can replace the rat's nest of wires and aggressive adhesive used with traditional monitoring systems with the same range of measurement, precision, and accuracy of vital signs data [24]. Dr. Rogers is a true pioneer in the industry of bioelectronics and FHE. Prior to his current role at Northwestern, Dr. Rogers career includes work at MIT, Bell Labs and the University of Illinois at Urbana/Champaign just to name a few. He, and his team, have led numerous efforts that resulted in many breakthroughs in the industry regarding biomedical applications.



Figure 7. Skin-like devices for wireless monitoring of vital signs in neonatal intensive care [24].

These battery-free, wireless, epidermal devices have many benefits for patients, particularly for premature babies whose skin is very fragile. The wireless sensors eliminate the physical barrier created by a baby being tethered to its crib for continuous vital sign monitoring to enable freedom of movement for healthcare personnel to perform imaging studies and for parents to have skin-to-skin contact as early as possible. The antenna system and data recording electronics are printed onto a thin, soft, flexible substrate that measures 5 cm by 2.5 cm (see Figure 8). Side-by-side studies of traditional wired sensors to wireless sensors were performed on over 70 premature babies in the neonatal intensive care units (NICU) at Prentice Women's Hospital and the Ann & Robert H. Lurie Children's Hospital in Chicago with successful data collection results in precision and accuracy [25].



Figure 8. Dual wireless sensors (chest and foot) for premature babies [25].

Wearable Electrochemical Systems

Wearable devices are an enabling technology for the healthcare industry with direct applications in sports, fitness, and wellness. Multi-parameter wearable sensors offer pain free real-time monitoring capability without the need for invasive methods to collect samples for biomarker monitoring. Another development in bio-integrated electronics by Dr. Rogers and his team at Northwestern University is a soft, flexible and stretchable microfluidic system (see Figure 9) that can be bonded to the skin without chemical or mechanical irritation and measures sweat and sweat biomarkers accurately and in real time. Dr. Rogers teamed with the Gatorade Sports Science Institute, the Seattle Mariners, the US Air Force, and the Shirley Ryan AbilityLab to further develop, test, and validate the device.



Figure 9. Skin-like microfluidic systems for real-time analysis of sweat [24] [26].

The soft, flexible, microfluidic device has microscopic channels that route sweat to reservoirs where colorimetric analysis is performed on key bio-markers. Initial prototypes (see Figure 9, *left*) measured chloride loss, glucose, lactate, and pH levels with newer prototypes (see Figure 9, *right*) also quantifying concentrations of heavy metals like lead and arsenic, and urea and creatinine levels (kidney health). The coloration of the sensors is the result of the response to

biomarkers in the sweat collected in the reservoirs and are quantitatively assessed by an app on a smartphone.

Additional examples of the wide scale commercial potential for these "lab on a chip" wearable technologies include Gatorade's Hydration Patch and GE's Sweat patch. Both were recently developed and aimed at monitoring hydration levels of athletes for enhanced performance. The military is actively performing research and development in the areas of environmental health and protection for soldiers to include the U.S. Army's Military Operational Medicine Research Program (MOMRP) integrated Real-Time Physiological Status Monitoring (RT-PSM) systems and exposure-related health effects monitoring platforms [27]. The potential market for these types of innovations in commercial, industry, and military sectors is significant and growing rapidly.

Flexible Meta-Skin

The application of flexible electronics technology includes expanding its use from localized bio-integrated areas on the human skin to large scale use as a meta-skin (see Figure 10) to form an adaptive system for camouflage and display [28]. The development of this visual appearance modulation capability was inspired by nature, namely cephalopods (e.g. octopus, squid, cuttlefish), given their unique ability to generate displays that can change in response to changes in background scenes and lighting conditions. This visual appearance modulation capability has been replicated using flexible electronics to mimic biologic color tuning through autonomous sensing and adaptation to the surroundings with potential relevance to consumer, industrial, and military applications [29].



Figure 10. Multi-layer meta-skin (left) with flexibility demonstrated through wrapping around the human hand/wrist [28].

Multiple approaches have been used to engineer a flexible system that adopts similar principles to cephalopods to perform visual appearance modulation. These include concepts for employing tunable frequency selective surface technology to create a scattering suppression effect [28] to systems that combine high-performance, multiplexed arrays of actuators and photodetectors in multilayer configurations on flexible substrates, with overlaid arrangements of pixelated, colorchanging elements [29]. A schematic of the latter approach described is provided below in Figure 11 for a 16x16 array meta-skin device. The top most layers shown in Figure 11(A) correspond to the leucodye composite (replicates a cephalopod's chromatophore) and the silver white reflective background (replicates leucodye). The next layer supports an ultrathin silicon diode for actuation to modulate the leucodye layer (replicates muscle). The bottom most layer, separated from the third layer by a PDMS material, provides distributed, multiplexed photodetection, (replicates opsin) [29].



Figure 11. Exploded view of the layered components of a 16×16 array of interconnected unit cells in a full, adaptive flexible meta-skin [29].

With the emergence of the Internet of Things (IoT) in the 21st century, the demand for wearable devices, to include biointegrated electronics, is expected to grow into a \$46B business by 2024 [18]. Wearable devices are an enabling technology for the healthcare industry with direct application in sports, fitness, and wellness as proven by the examples described above. The future outlook for wearable devices within the military community is substantiated by publications such as the Army's 2019 Modernization Strategy, which cites the "Science of Additive Manufacturing" as one of its Priority Research Areas for the development of next generation munitions for increased range and lethality [30]. Future adoption of 3D additive printed electronics technologies are also exemplified by the efforts of the U.S. Air Force and the wearable performance monitoring systems that are being developed at the Air Force Research Lab (AFRL). One of the most notable examples of the DoD's commitment for military adoption of these technologies was the conception and establishment of NextFlex, the DoD Manufacturing USA Institute for FHE. DoD made an initial \$75M dollar investment to help develop, maintain, and retain critical flexible electronics manufacturing technology within the United States for the dual purpose of commercial and military applications [22].

CONCLUSION

A broad-brush review has been presented of manufacturing techniques developed to print electronics in an additive approach without the use of masks and harmful etching chemicals typical of subtractive processes. The DW methods described vary in their capabilities to address different facets of printed electronics to include 1D to 3D printing as well as writing on conformal, flexible and stretchable substrates. Among the group of DW methods, the flow-based direct write approach using AJP technology shows great potential for printing fine feature conformal, non-planar features for highly integrated devices.

Much work has been performed to date using AJP technology for the fabrication of 3D printed electronics. Materials formulations and the processes used to deposit the ink on the substrate are key to achieving the functional performance and reliability needed across system application domains. Continued work is still needed in developing new materials and refining processes to support maturation of these methods thus enabling these techniques to become more readily adopted.

DW methods have a wide application window from prototyping through low-to-high level production. The various electronic devices printed to date using DW techniques are evidence these methods will be a key driver in accomplishing rapid prototyping and manufacturing of electronics and sensors, to include bio-integrated electronics. New applications are being announced regularly in this emerging technology area. AJP technology, along with other DW techniques, will enable the additive manufacturing of next-generations miniaturized and rugged electronics on nonplanar, conformal, and flexible substrates.

The high demand for IoT electronics and sensors is a key driver for taking advantage of the benefits of rapid prototyping, highly integrated, and substrate-agnostic printing capability using DW methods. The field of personalized IoT includes wearable devices in small, lightweight, and conformable form factors and is expected to grow into a \$46B business by 2024. The emergence of FHE leveraging DW techniques is a primary enabler of bringing electronics off rigid printed circuit boards and fitting them to the contours and movement of the human form. The work samples presented in this paper are but a few of the many biointegrated electronics devices with end applications in consumer electronics, healthcare (sports, fitness, and wellness), defense, aerospace, and automotive.

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