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Technologies for Printing Sensors and Electronics over Large Flexible Substrates: A Review

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***Abstract*— Printing sensors and electronics over flexible substrates is an area of significant interest due to low-cost fabrication and possibility of obtaining multifunctional electronics over large areas. Over the years, a number of printing technologies have been developed to pattern a wide range of electronic materials on diverse substrates. As further expansion of printed technologies is expected in future for sensors and electronics, it is opportune to review the common features, complementarities and the challenges associated with various printing technologies. This paper presents a comprehensive review of various printing technologies, commonly used substrates and electronic materials. Various solution/dry printing and contact/non-contact printing technologies have been assessed on the basis of technological, materials and process related developments in the field. Critical challenges in various printing techniques and potential research directions have been highlighted. Possibilities of merging various printing methodologies have been explored to extend the lab developed standalone systems to high-speed roll-to-roll (R2R) production lines for system level integration.**

***Index Terms*—Printed Sensors, Printed Electronics, Flexible Electronics, Large Area Electronics, Roll-to-Roll, Dispersion Solutions**

I. INTRODUCTION

PRINNTING technologies are aiding and revolutionizing the burgeoning field of flexible/bendable sensors and electronics by providing cost-effective routes for processing diverse electronic materials at temperatures that are compatible with plastic substrates. Simplified processing steps, reduced materials wastage, low fabrication costs and simple patterning techniques make printing technologies very attractive for the cost-effective manufacturing [16-18]. These features of printed electronics have allowed researchers to explore new avenues for materials processing and to develop sensors and systems on even non-planar surfaces, which otherwise are difficult to realize with the conventional wafer-based fabrication techniques. The printed electronics on flexible substrates will enable conformable sensitive electronic systems such as electronic skin that can be wrapped around the body of a robot or prosthetic hands [20-25]. Printed electronics on polymer substrates has also opened new avenues for low-cost fabrication of electronics on areas larger than the standard wafers available commercially. In accordance with the electronics industry roadmap, the research in this field is slowly inching towards a merge of well-established microelectronics and the age-old printing technologies [26]. This is evidenced by development of devices such as, large area printed pressure sensors [5, 27-29], radio frequency identification tags (RFID) [11, 12], solar cells [30], light emitting diodes (LED) [13] and transistors [14].

Traditional approaches for printing electronics and sensors involve bringing pre-patterned parts of a module in contact with the flexible (or non-flexible) substrates and transferring the functional inks or solutions onto them [6-9, 11, 13]. The two major approaches usually followed for development of printing/coating system are contact and non-contact printing, as shown in Fig. 1 and described later in Section IV. In contact printing process, the patterned structures with inked surfaces are brought in physical contact with the substrate. In a non-contact process, the solution is dispensed through via openings or nozzles and structures are defined by moving the stage (substrate holder) in a pre-programmed pattern. The contact-based printing technologies comprise of gravure printing, gravure-offset printing, flexographic printing and R2R printing. The prominent non-contact printing techniques include screen-printing, slot-die coating and inkjet printing. The non-contact printing techniques have received greater attractions due to their distinct capabilities such as simplicity, affordability, speed, adaptability to the fabrication process, reduced material wastage, high resolution of patterns and easy control by adjusting few process parameters. [17, 18, 31-34]. Recently, the newly emerging polymeric stamp based printing methods such as nanoimprint, micro-contact printing and transfer printing have also attracted significant interest, especially for inorganic monocrystalline semiconductors based flexible electronics [19, 30, 33-37].

This paper presents a survey of various printed electronics technologies. A few review articles reported in this field previously have reviewed some of the printing technologies individually [17-19, 30-37]. For example, the review paper by Singh *et al*. [38] focusses on inkjet printing, the one Schift *et al.* [39] focusses on nanoimprinting, Carlson *et al.* [40] have discussed transfer printing, Perl *et al.* [2] have presented review on microcontact printing and Søndergaard *et al.* [41] have discussed R2R fabrication. Differently from previous reviews, this survey paper brings together various printing techniques and provides a detailed discussion by also involving the key electronic and substrate materials, and the systems. Critical limitations of each technology have been highlighted and potential solutions or alternatives have been

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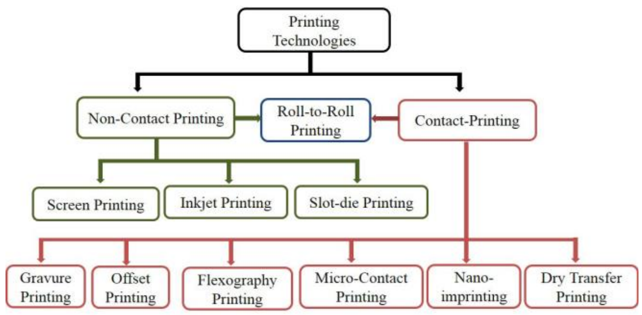


Fig. 1. The classification of common printing technologies.

explored. This paper also evaluates printed electronics on the basis of electrical characteristics of the resulting devices or sensors and materials (organic/inorganic) they are made of. Often, the advantages of printing technologies eclipse the challenges associated with them. As most of the printing technologies share common processing techniques, the development of a common platform is also explored assuming that the limitations of one method could be overcome by the advantages of the others.

This paper is organized as follows: The Section II presents various printable electronic materials and gives an overview of solution processable conductors, semiconductors and dielectric materials (organic/inorganic) along with dry transferrable inorganic monocrystalline materials. The substrate materials are presented in Section III, where their physical and chemical behavior to different classes of materials and the constraints in using them for different applications are also described. The Section IV gives a thorough description of various non-contact and contact printing technologies utilized for deposition and patterning of solution based materials. A comprehensive survey of various process parameters, system and materials related requirements with illustrative examples of manufactured devices and circuits is also presented. This section also discusses some of the non-conventional stamp printing techniques usually utilized for microcontact printing, nanoimprinting and dry transfer printing of monocrystalline inorganic semiconductors. Finally, the Section V summarizes the key observations and presents the future research directions.

II. Printable Materials

The selection of colloidal solution with specific rheological properties for printed electronics is at the core of developing a consistent manufacturing process. In order to achieve the goal of low-cost and lightweight printed electronics, a large variety of materials (organic and inorganic) have been explored. These materials could be divided into three categories: (a) conductors; (b) semiconductors; and (c) dielectrics [19, 31, 35-37]. Beside these, some composite materials, having dual nature as insulator, ferroelectric, piezoelectric, piezoresistive, and photosensitive properties are also used in thin film printed devices. Hybrid organic/inorganic materials have also been used to compensate for the slow speed organic based electronic devices [34, 37]. Majority of printable materials are in the form of solutions, which require specific properties to allow proper printing on variety of plastic or paper substrates. For example, proper dispersion of nanoparticles is essential to avoid agglomeration. Further, an acceptable level of chemical and physical stabilities is needed to maintain a balance between Brownian and gravitational motion of the particles. In the following sections, we discuss the common organic/inorganic materials that are suitable and easily processable through printing technologies.

A. Conducting Materials

Conducting materials are the main structural blocks of all electronic devices as they form the fundamental part of the device layers or interconnections. Deposition techniques for patterned metal structures and interconnects are now at mature stage with possibility of obtaining structures with controlled thickness and resolution. Various printing technologies require a different set of parameters such as viscosity, surface tension, conductivity and compatibility of the solvents with the underlying materials (in multilayer structures) (see Table II) [30]. Therefore, a careful selection of appropriate conductive material is needed by also taking into account the work function of the neighboring materials. Some of these metals have already secured their place in printing technologies by showing good dispersing properties in the form of colloidal solutions. Properties of these solutions are adjusted according to the desired printing technology by using surfactants and volatile additives. Amongst the list of metals practiced for printed electronics, silver (Ag) based pastes and solutions are the choice of most of the researchers due to its good physical and electrical performance on plastic substrates [31, 33, 35]. Being counted in the category of precious metals, it cannot serve the purpose of low cost flexible electronic devices, which is the true essence of printing technology. Besides silver solutions, the carbon and copper based inks are also used. But oxidation of copper based inks after printing do not serve the purpose very well [30, 42].

Mimicking metallic conductivity, crystalline organic conducting materials such as polyacetylene films combined with p-dopants were first reported by Shirakawa *et al.* Soon after this discovery, the n-type materials were also investigated [43]. All these organic conducting materials are categorized as intrinsic conducting polymers. Chemical structure of intrinsically conductive polymers can be tailored to get desired electronic and mechanical properties [44].The metallic conductors having work functions closely matching with the energy levels of p- and n-type semiconductors have already been developed. In contrast, for organic conductors, the materials having compatible work function with p-type semiconductors are mostly reported [36, 44, 45]. Polyacetylene, polypyrrol, polyphenylene, poly (p-phenylene vinylene), polythiophene polyaniline, polyaniline doped with camphor sulfonic acid and PEDOT:PSS (3, 4-polyethylenedioxythiopene-polystyrene sulfonic acid) are some of the most commonly reported hole-injecting polymers used with organic semiconductors. The PEDOT:PSS has been widely studied for transparent conducting polymer anodes as it exhibits a very high conductivity of about 300 S/cm. A

TABLE I
COMPARISON OF POLYMER SUBSTRATES **[36, 56-60]**

Property	**PET**	**PAcr**	**PEN**	**PC**	**PS**	**PI**
Tg (°C)	70	105	120	145	203	270
Upper Tm (°C)	115	175	268	115-160	180-220	250-320
CTE (ppm/°C)	33	70	20	75	54	8-20
% Transparency	90	>90	88	92	89	35-60
Water Absorption (%)	0.6	0.2	0.4	0.25	1.4	2-3
Y. Modulus (10^9N/m^{2}Gpa)	2-2.7	2.4-3.4	0.1-0.5	2.6	-	2.5
Solvent Resistance	Good	Good	Good	Poor	Poor	Good
Surface Roughness	Poor	Fair	Poor	Good	Good	Good
Dimensional Stability	Good	Good	Good	Fair	Fair	Fair

detailed description of these materials is given in [36]. Despite the attractions of low cost and easy solution processing techniques the organic conductors have far less conductivities than the conventional metals such as Ag, which has conductivity of 6.30×10^7 S/m.

Another class of conducting polymers developed for printing and flexible electronics is based on nanocomposites, made by mixing of metallic nanoparticles with organic elastomers such as (Poly (dimthylsiloxane) PDMS) [46, 47]. Conductivity of such composites is based on the percolation threshold of the fillers. With different ratio of elastomer and nanofillers the flexible and stretchable conductive sheets and patterns have been investigated in [48]. Besides metallic nanoparticles fillers, the multi wall carbon nanotubes (MWCNT)/PDMS composites, reported recently, offer a very attractive mechanical and electrical properties for large area flexible electronics [46-50] and touch sensors [27-29]. A challenging issue with nanocomposite materials is the proper dispersion of nanofillers in the base polymer, which greatly affects the rheological properties of the mixture to be printed. The particle agglomeration in such nanocomposites affects the printability and uniformity of the layer after printing as compared to other printable solutions. To overcome this problem and to enhance the final dispersion the nanofillers are first added to a dispersant solution and then mixed with the base polymer [48]. Due to unique electrical, mechanical and optical properties, the printable solutions of graphene (conductivity of ~1.00×10^8 S/m) and carbon nanotube (CNT) are also in vogue nowadays for flexible electronics [51-55]. Due to their distinctive properties, graphene and CNTs have also garnered recently a significant attention as potential candidate for the replacement of ITO [31, 35, 36, 44].

B. Semiconductor Materials

Semiconductor materials are critical components for developing active electronic and sensing devices. The transduction of free carriers within the semiconductor dielectric interface is usually the focal points in development of electronic devices and systems. Like conducting materials discussed above for printing technologies, the organic/inorganic semiconducting materials are also used for printable sensors and electronics. Inorganic materials have superior properties in terms of performance and stability while solution processable organic semiconductors are attractive due to low cost processing at ambient environment and flexibility. Examples of inorganic semiconductors commonly used for flexible electronics are Si [61-65], oxides of transition metals [66, 67] and chalcogenides [68]. Apart from chemical vapor deposition (CVD) of amorphous silicon for large area flexible electronics, crystalline Si is also used in flexible electronics by employing dry printing technique [62, 69]. Si can be solution processed by using precursors or nanoparticles, but typically requires high annealing temperature (550–750°C) in inert atmosphere makes it incompatible with almost all plastic and paper substrates. This is evident from Table I, where glass transition temperature (Tg) of various plastic substrates are given along with other properties. Oxides of transition metals are sometimes used in flexible electronics but through vacuum deposition techniques other than printing. ZnO and GIZO can be solution processed and even printed but sintering temperature of 300–500°C is necessary to achieve optimum mobility [19, 70]. Very few works have been reported on solution processed inorganic semiconductors and their compatibility with the usual printing techniques [19, 31, 71].

From printability viewpoint, the solubility and proper dispersion of organic semiconductors are the important parameters. Commonly used solution processed organic semiconductors, having acceptable charge transport and mobility include regioregular poly(3-hexylthiophene) (P3HT), poly(triarylamine), poly(3,3-didodecyl quaterthiophene) (PQT), poly(2,5-bis(3-tetradecyllthiophen-2-yl) and thieno[3,2-b] thiophene) (PBTTT) [36, 44]. Fullerenes and solution processable derivatives such as phenyl-C61-butyric acid methyl ester (PCBM) blended with P3HT are some of the commonly used electron donor and acceptors in the bulk heterojunction devices [54, 72]. Additionally carbon nanotubes and graphene are also currently under investigation due to their high-mobility [73]. Stability and reliability of organic materials for long time processing is very challenging especially the low ionization energies are prone to oxidation which results in slow responses and degradability of the devices. Further, developing printable n-type organic semiconductor is challenging due to instability which is becoming one of the serious obstacles in development of organic CMOS devices [74, 75].

C. Dielectrics

For applications requiring high capacitance in multilayered printed structures, thin layers of dielectric materials are essential for proper insulation to prevent leakage currents and sometimes to obtain low voltage operation for field effect devices. A uniform layer of dielectric is needed to promote the activation of the medium caused by electric field or other transduction phenomena. Inorganic materials, such as silica, alumina, and other high permittivity oxides often used in electronics on flexible substrates are usually not printable [17-19, 31-33]. Low cost organic dielectric materials that are available in large quantities and can be dissolved in various solvents and solution can be printed easily as compared to inorganic counterparts. In most of the printed electronics, semiconductor/dielectric interface is of prime importance for the high performance and stability of the devices. Self-assembled monolayers are sometimes used for modification of the dielectric surface. Some of the commonly used organic dielectric materials in printed electronics are poly (4-vinylphenol) (PVP), poly (methyl methacrylate), Polyethylene Terephthalate, Polyimide, Polyvinyl alcohol and Polystyrene. [17, 18, 31]. Besides dielectric layer in electronic devices, solution processed organic dielectric materials are also used for final encapsulation of printed devices.

III. Substrates for Flexible Sensors and Electronics

It is the flexibility of the polymer substrates, which is providing grounds for low cost high-speed manufacturing of devices over large areas using various printing technologies in a R2R production line. To replace planar rigid substrates, the flexible substrates are required to possess properties such as dimensional stability, thermal stability, low coefficient of thermal expansion (CTE), excellent solvent resistance and good barrier properties for moisture and gases. There are three types of substrates that could be employed for flexible electronic devices: thin glass, metal foils and plastics [56, 57] [76]. Thin glass is bendable but the intrinsic brittle property limits its utility in flexible electronics. Metal foils on the other hand can sustain very high temperatures and provide a window for inorganic materials to be deposited on it but the surface roughness and high cost of the materials hinder its use for flexible electronics.

Plastic materials are the potential candidates for applications requiring high degree of bendability, transparency and emissive properties. Plastic materials provide a reasonable tradeoff between physical, chemical, mechanical and optical performance as described in Table I. In addition, the central idea of the low cost flexible electronics (e.g. R2R manufacturing) is feasible with plastic substrates. The main issue in use of plastic substrates is the lower glass transition temperatures (T_g) (Fig. 2), which limits its utility to organic materials. Polymer substrates are divided into three main groups [56, 57] i.e. semi-crystalline, amorphous and solution cast amorphous. Semi-crystalline polymers used in flexible electronics include polyethylene terephthalate (PET), heat stabilized PET, polyethylene napthalate (PEN), and heat stabilized PEN and polyetheretherketone (PEEK). Amorphous polymer substrates include polycarbonate (PC) and Polyethersulphone (PES), which are non-crystalline thermoplastics that can be melt-extruded or solvent casted [77]. Some of the amorphous group that cannot be melt processed include such as modified polycarbonate (PC), Polyethersulphone (PES), polyarylate (PAR), polycyclic olefin (PCO) or polynorbonene (PNB) and polyimide (PI). These substrate materials are discussed in detail in [36, 56, 57].
The semi-crystalline polymer substrates with T_g higher than $140°C$ (e.g. heat stabilized PET and PEN) generally tend to have high melting points, which allows the polymers to be melt processed without significant degradation [56]. Except polyimide, which can be yellow as well as transparent, all other polymeric substrates given in Table I meet the optical clarity requirements. The effect of thermal stress and mismatch between the CTE of substrates and the deposited

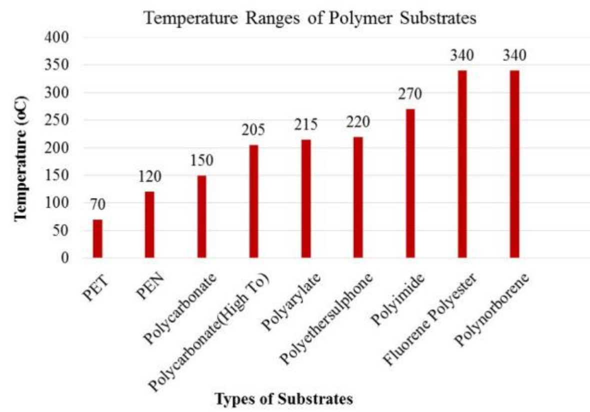


Fig.2. Glass transition temperatures of commonly used plastic substrates in printed sensors and electronics.

material are critical for efficient performance of the electronic devices. Due to this CTE mismatch, the deposited layers become strained and crack under thermal cycling. For example, in the temperature range from room temperature up to T_g, the CTE for flexible substrates such as PEN and PET, the typical is 18-20 ppm and 20-25 ppm respectively. This means if material with different CTE (e.g. amorphous polymers have CTE 50ppm/^{o}C below T_g) deposited on top of these substrates can expand 3 times, above the Tg value of the substrates, ultimately causing undesirable mismatch in the fabricated structures vis-à-vis original layout [56, 78]. A low CTE (typically < 20ppm/^{o}C) is desirable to match the thermal expansion of the substrate to the subsequent layers which are deposited on top of it.

Applications such as RFID, sensors, active matrix backplane, OTFTs and OLEDs etc. also affect the choice of substrate. While substrate related tolerance is acceptable in some applications but the requirements are stringent for others. For display applications, optical clarity is important where a total light transmission (TLT) of > 85% over a wavelength range of 400-800nm are required [10, 36, 56]. This is only required for light emission through substrates in bottom-emission and electrophoretic displays, whereas for top-surface emission of light, optical clarity of the substrate is not essential. The physical forms of the substrates i.e. flatness, light weight, ruggedness, conformable, rollable/foldable and ease of handling are some of the features that affect their selection. Similarly, the T_g is of paramount importance as it limits the use of materials (organic/inorganic) due to incompatibility in terms of processing temperature.

Upper processing temperature (Tm) is another important parameter that should be considered in addition to dimensional stability due to thermal stresses. As in traditional paper printing, a wide range of chemicals and surfactants are used to adjust the properties of the solution for efficient transfer of the ink from system to substrate [56, 57]. Some of these polymer substrates (amorphous) are poor resistant to solvent absorption as compared to semi-crystalline polymers. This issue is very critical for sensors application where a slight modulation in transducer values can affect the whole process. Humidity has a major effect on the polymer material used for flexible

TABLE II
COMPARISON OF VARIOUS PRINTING TECHNIQUES

Parameter	Gravure	Offset	Flexographic	Slot-die	Screen	Inkjet	Microcontact & Nanoimprint	Transfer
Print Resolution (μm)	50-200	20-50	30-80	200	30-100	15-100	1-20	4-50
Print Thickness (μm)	0.02-12	0.6-2	0.17-8	0.15-60	3-30	0.01-0.5	0.18-0.7	0.23-2.5
Printing Speed (m/min)	8-100	0.6-15	5-180	0.6-5	0.6-100	0.02-5	0.006-0.6	NA
Req. Solution Viscosity (Pa. S)	0.01-1.1	5 – 2	0.010-0.500	0.002-5	0.500–5	0.001-0.10	~ 0.10	NA
Solution Surface Tension (mN/m)	41- 44		13.9 -23	65-70	38-47	15-25	22-80	NA
Material Wastage	Yes	Yes	Yes	Yes	Yes	No	Yes	No
Controlled Environment	Yes	Yes	Yes	No	No	No	No	Yes
Experimental Approach	Contact	Contact	Contact	Contact-less	Contact	Contact-less	Contact	Contact
Process Mode (sample pattern line)	Multi-steps	Multi-Steps	Multi-steps	Single-Step	Multi-Steps	Single-Step	Multi-steps	Multi-Steps
R2R Compatibility	Yes	Yes	Yes	Yes	Yes	Inter-mediate	Yes	No
Hard Mask Requirement for each Printing Step	No	No	No	No	Yes	No	No	Yes
Printing area	Large	Large	Large	Large	Medium	Large	Medium	Medium
References	[4, 31, 34, 73, 75, 79]	[31, 34, 80-82]	[31, 34, 75] [8, 83-85]	[86-90]	[31, 34, 91-94]	[31, 35, 95-97][74][38, 98, 99]	[2, 7, 100-106]	[37, 62, 107-111]

TABLE III
COMPARISON OF ELECTRICAL CHARACTERIZATION RESULTS OF SAMPLE DEVICES DEVELOPED WITH DIFFERENT PRINTING TECHNOLOGIES AND MATERIALS

Printing Technology	Material (Organic/Inorganic)	Mobility (cm^2/V.Sec)	On/Off ratio	Threshold Voltage (V)	Reference
Flexography	P(NDI2OD-T2) (Organic)	0.1 - 0.3	10^5–10^6	10.20	[83]
Flexography	GSID 104031-1 (Organic)	1.8-4.1×10^{-3}	N/A	6	[75]
Flexography	6,13-bis triisopropylsilylethynyl) (Organic)	2.0-5.8 ×10^{-3}	N/A	5.1	[75]
Gravure	SW-CNTs (Organic)	0.04-0.17	10^3-10^5	-4.5	[73]
Gravure	P(NDI2OD-T2) (Organic)	0.1 - .2	10^5-10^6	5	[83]
Gravure	SW-CNTs (Organic)	0.07-.15	10^3-10^4	-20	[83]
Inkjet	TIPS–Pentacene (Organic)	0.15-.53	10^6	2	[99]
Inkjet	P(NDI2OD-T2) (Organic)	~ 0.1	~10^5	~ 10	[83]
Inkjet	C60, fullerene (Organic)	2.2-2.4	10^7-10^8	5	[95]
Inkjet	P3HT/PS (Organic)	0.02	10^6	>30	[112]
Nanoimprint Lithography	Poly(3-Hexylthiophene) P3HT (Organic)	0.86 - 0.98	7.02 × 10^3	15.7	[113]
Screen	Dinaphtho-Thieno-Thiophene (DNTT) (Organic)	0.33	10^4	N/A	[114]
Transfer	μs-Si (Inorganic)	105	10^2	~1.5	[115]
Transfer	Poly-Si (Inorganic)	30	N/A	10	[116]
Transfer	Single-Si (Inorganic)	230	>10^5	~1	[117]
Transfer	Single-Si (Inorganic)	300	N/A	N/A	[116]

substrates affecting the resistivity to a significant value. Absorption of water adds to the increase in weight of the substrate and also alters the dielectric constant for the capacitive sensors. To overcome this issue, a thin barrier coating of transparent oxides is applied on the surfaces of polymer substrates, especially for sensors used in food and medical packaging. In nutshell, to replace planar substrates, the polymer substrates should mimic their properties such as dimensional stability, thermal stability, low CTE, excellent solvent resistance and good barrier properties for moisture, air and gases [56, 57, 76].

IV. PRINTING TECHNOLOGIES

The development of thin film devices either by the use of printing or coating of hybrid organic/inorganic materials is one of the many ways explored to simplify processing steps, facilitate location specific deposition and enhance the production speed. The chemical solution or nanoparticles of functional materials are used in the form of colloidal solution in most of printing technologies. These solutions are deposited directly on rollable substrates using controlled dispensing processes or coated on substrates using controlled pressures and speeds [34, 71, 95, 96, 118]. The key benefit of printing techniques is the reduction in material wastages, as the solution is printed on the defined location in single step and the residual solution is collected back for subsequent use. These dispensing and coating processes have led to promising results especially with organic materials, as organic thin film transistors (TFT), OLEDs, sensors, solar cells, RFID tags, printed batteries and capacitors have been demonstrated, summarized in Table III [16-18, 31, 32]. An important benefit of printing technologies is that they enable production of large area electronics and sensors by R2R manufacturing in a cost effective way.

Printing technologies are divided into two broad categories, as shown in Fig. 1. The non-contact and contact-based patterning discussed in this section follow the classification given in Fig 1 and the state of the art, the pros/cons and the challenges of these printing techniques are discussed below.

A. Non-Contact Printing Technologies

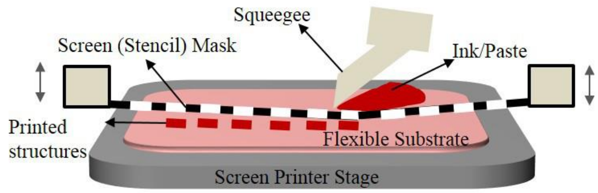
1) Screen Printing

Screen-printing is the most popular and matured technology for printed electronics as it has been practiced in electronics industry for quite some time now to print metallic interconnects on printed circuit boards. It is faster and more versatile in comparison to other printing tools, as it adds simplicity, affordability, speed and adaptability to the fabrication process. The results from screen-printing can be reproduced by repeating a few steps and an optimum operating envelope can be developed quickly [18, 31, 72, 118-120].

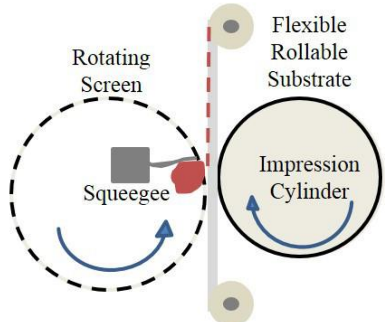
Two different assemblies of screen printers i.e. flatbed and rotary are used for R2R manufacturing described in Fig. 3(a-b) respectively [18]. Screen printer has simple setup comprising of screen, squeegee, press bed, and substrate, as shown in Fig. 3. In flatbed, the ink poured on the screen is squeegeed to move across the screen resulting in its transfer through the stencil openings to the substrate beneath it. For optimization of the materials and processing steps, flat bed screen-printing is a powerful tool for small laboratory systems. Flatbed screens can be substituted by rotary screen for continuous processing in which the web of the screen is folded while the squeegee and ink are placed inside the tube.

Relatively high speeds can be achieved by rotary screen as compared to flatbed, but the screens for rotary setup are expensive and very difficult to clean [18, 121]. Although a very simple process, the print quality and characteristics are affected by various factors such as solution viscosity, printing speed, angle and geometry of the squeegee, snap off between screen and substrate, mesh size and material [91, 118, 122]. The paste viscosity and surface tension of the substrate are important for complete dispensing of the paste through the screen mask. Screen printing technique is usually compatible with the high-viscosity inks as the lower viscosity inks will simply run through the mesh rather than dispensing out of the mesh [34, 92]. Without giving any consideration to proper tuning of the ink properties and mesh count, the nominal values of 50-100µm are common print resolutions and wet thicknesses of a few microns. The possibility of printing relatively thick layers could enable printing of low-resistance structures, also with conducting polymers, by compensating the high volume resistivity with a thicker layer [31].

In addition, a compromise between surface energies of substrates and surface energies of the inks is important for high-resolution line widths [92, 93, 97]. The reduced surface energies of the substrates reduce the wettability of the solution, which results in improved line resolution. If the critical surface tension of a substrate is lower than the surface energies of inks, good resolution can be achieved even with low viscosity inks. Although high viscous inks are required to minimize ink flow on the substrate, the low viscosity is desirable to dispense the solution through the mesh to realize structures with fine edges and resolution. In this scenario, the low viscosity inks are preferred as the wettability of the substrate can be controlled by adjusting the surface energies of

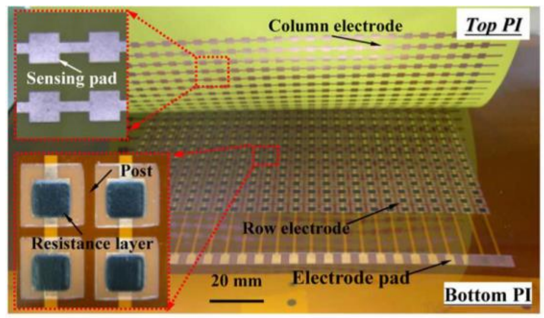


(a)

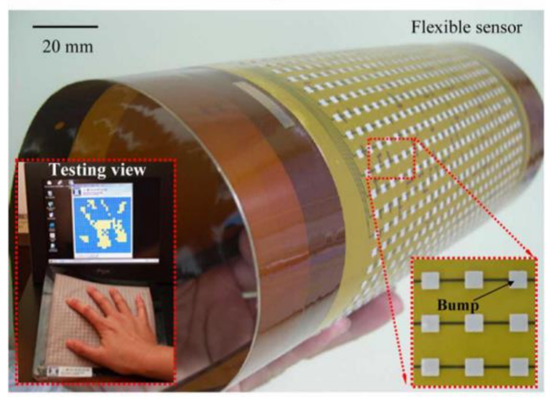


(b)

Fig.3. (a) The flatbed Screen printing with planar substrates under screen and squeegee for solution dispensing, (b) Rotary screen printer with moving substrate (web) between cylindrical mask and impression cylinder.



(a)



(b)

Fig. 4. Screen printed pressure sensors on large flexible polyimide films, (a) Inside view between two PI films, including a resistance layer, posts, and electrodes. (b) Flexible sensor with bump structures on the top film [5]

the substrate. The low viscosity inks possess high degree of flowability, which reduces the chances of mask blockage, and leads to even edges of printed lines and, smooth surface of the printing films [92, 93].

Material, strength and number of meshes in screen also play a major role in high-resolution patterning, as screen is developed by using different sizes of mesh openings and several materials ranging from polyester to stainless steel. The technological development in the screen mesh is made by

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modifying the silk strength by using materials such as nylon, polyester and stainless steel. The increase in the strength of the mesh wire material used in the screen mask and the mesh count result in improved printing quality. For printing stability during mass production, a screen made of stainless steel mesh with three times more in strength than conventional stainless steel mesh has also been developed [122, 123].

The feasibility of screen-printing for flexible electronics has been demonstrated through a number of printed sensors, electronics devices and circuits. For example, all screen printed TFTs have been demonstrated in [119, 124, 125]. Screen printing was claimed to be used for the first time to develop OLEDs by investigation the process and solution parameters i.e. viscosity of the solution and mesh count of the screen [122]. Multilayer high-density flexible electronic circuits, connected through micro via holes with embedded passive and optical devices, have been realized by using advanced screen printing processes [118]. Screen-printing is also used for patterning to develop shadow masks for fabrication of organic TFT. Screen printed electrical interconnects for temperature sensor on PET substrate are reported in [93]. The large area flexible pressure sensor shown in Fig. 4 is fabricated by utilizing two polyimide films as top and bottom films and connecting the electronic circuits through micro via holes. Fig. 5 shows all screen printed pressure sensors developed by using piezoelectric Poly(vinylidene-fluoride-trifluoroethylene) (PVDF-TrFE) and piezoresistive multi wall carbon nanotubes (CNT) in Poly(dimethylsiloxane) (MWCNT/PDMS) nanocomposite materials [27-29]. All structures of metal plates, interconnects and sensitive materials are deposited by using screen-printing technology. All structural features of a humidity sensor including the interconnect patterns and protective polymer layers are also screen-printed [91, 94, 120]. Screen printing of cobalt hydroxide has been reported for obtaining supercapacitors [126]. The Spectral-Domain Optical Coherence Tomography is used in a simulated R2R process for monitoring the structural properties of moving screen printed interdigitated electrodes [127].

Unlike many other manufacturing techniques, the screen-printing does not require high capital investment. Accompanied by some supplemental methods such as inkjet technology, vapor deposition and laser processing, screen-printing is employed in most of the production lines of printed electronics. Using the supplemental technologies often results in cost reduction [123, 128]. Despite these attractions, screen-printing also poses a few challenges for developing all layers of a flexible device. These include, high wet thickness of the film, exposure of the ink to atmosphere and the dry out of the ink on the mask that deteriorates the mask designs of the screen [72]. However, the advantages such as high definition and high precision of multilayer structures add to the figure of merits of the screen printing techniques as compared to other deposition techniques for large-scale production.

2) Inkjet Printing

Inkjet printing is the rapidly emerging technique for direct patterning of solution based materials deposition. Materials in the form of colloidal or chemical solution are deposited through a micrometer sized inkjet nozzle head. A number of mechanisms for actuation of inkjet nozzle head have been developed. Among these, the most prominent techniques are thermal, piezoelectric and electrohydrodynamic inkjet systems. Droplets (often called Drop-on-Demand (DoD)) of very small dimensions are ejected at the corresponding pulse generated by either thermal or piezoelectric actuators used in the inkjet nozzle head [38, 98, 112, 129]. The Fig. 6(a-b) shows the mechanism of droplets actuation through piezoelectric and electrohydrodynamic setups. In electrohydrodynamic printing, solution is ejected by generating a high electric field between the nozzle and a counter electrode. A stable cone jet is the primary requirement of electrohydrodynamic inkjet system. The type of applied voltage defines the mode of ink ejection from the nozzle. DC voltage results in an intact jet while AC voltage at different frequencies and functions define the drop-on-demand mode of the system. An intact jet can be utilized for continuous patterning of solution as well as drop-on-demand similar to thermal and piezoelectric nozzle heads [130].

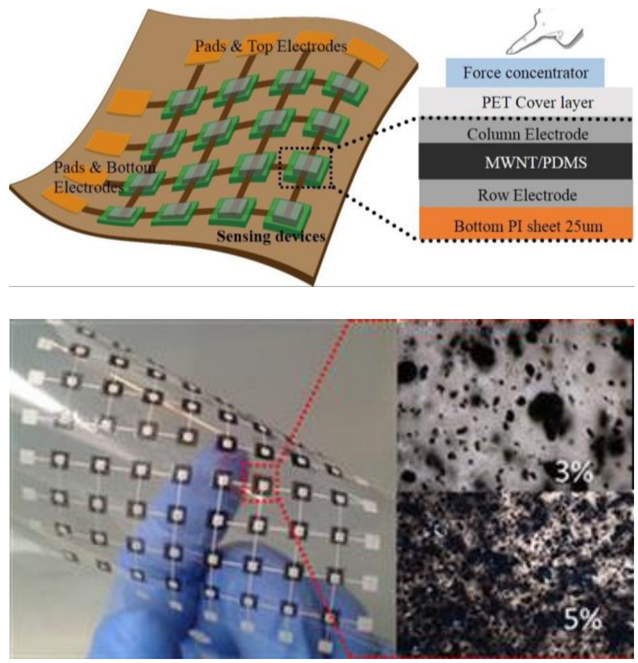

Fig.5. (*top*) Scheme of pressure sensors array. (*bottom*) Screen-printed piezoresistive sensors array using MWCNT/PDMS nanocomposites.

Another interesting feature of electrohydrodynamic printing is the spray coating of colloidal solutions shown in Fig. 6(c). Thickness in the range of nanometers can easily be achieved just by increasing the electric field value along with the distance between nozzle and substrate. A very fine layer of conductor, semiconductor and insulator can easily be deposited by adjusting the conductivity and viscosity of the solutions to obtain a stable cone jet. This technique has been successfully utilized in fabrication of a range of electronic devices and in biological systems [71, 131-133]. Besides electrospray deposition, some researchers are also exploring “Aerospray” for thin film deposition and patterning of electronic materials [134, 135].

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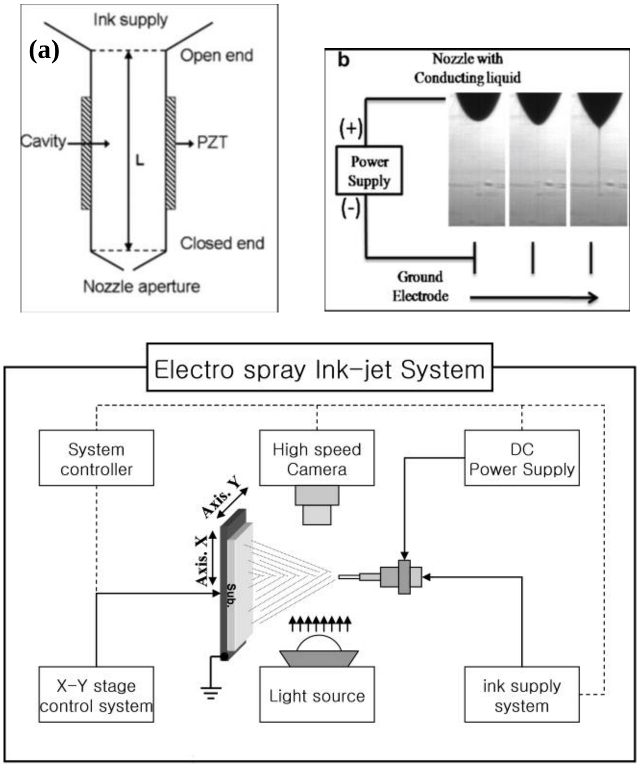

Fig.6. (a) Schematic diagram of the piezoelectric inkjet head, with PZT actuator [1]. (b) Schematic description of Electrohydrodynamic inkjet system. Electric field is generated between nozzle and counter electrode [15]. (c) Description of an Electrospray system with complete setup [19-21].

Inkjet printing has been used to fabricate TFT consisting of ZnO and PVP as the active device region and gate dielectric respectively [136]. A high performance n-channel transistor with uniform amorphous C60 Fullerene is developed by using inkjet printing and vacuum drying process [95]. Complementary circuits composed of pentacene and a xylene carboxylic diimide derivative of p- and n-channel TFTs are also fabricated on flexible foils. Staggered configuration of TFTs is followed for development of a flexible CMOS device, by printing both the n- and p-type organic materials separately using inkjet technology. The misalignment between energy levels due to the wide band gap of organic materials results in large barriers for charge injection, causing in reduced performance of the circuit [99]. Resistors, capacitors and inductors are developed using inkjet printing on polyimide substrate with various functional inks [96]. Inkjet in comparison to flexography, spin coated and gravure printing generally results in rougher and far less uniform morphologies with only partial uniform coverage of the channel region [83]. Fig. 7 shows pictures of different patterned structures and devices realized by using inkjet printer on a paper substrate. Chemical stability, solubility in common solvents, inexpensive solution and low temperature processing are some of the key requirements of inkjet printable materials alongside excellent charge transport properties in ambient conditions.

Development of colloidal solution for proper ejection of droplets on a targeted area by keeping an acceptable quality of the printed circuits is challenging due to the influence of evaporation rate of the solvents and orientation of the active particles. Slow speed due to limited number of nozzles and

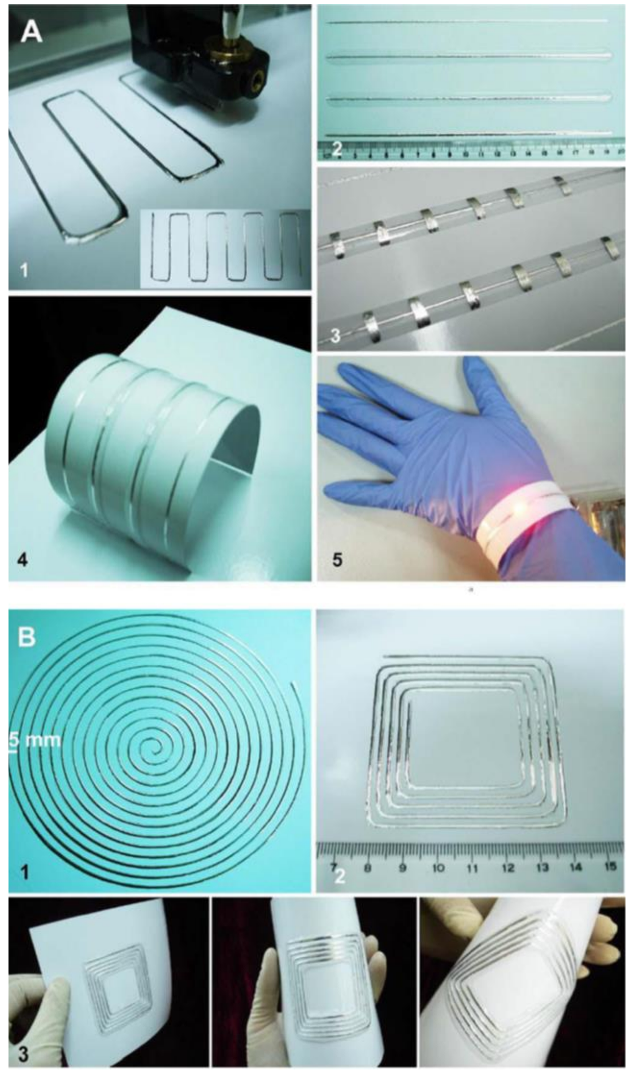

Fig. 7. (A) $GaIn_{24.5}$ based liquid alloy directly printed on coated paper by a dispenser printer and optical images for various conductive wires on coated paper: (1) Manufacturing process of printed electronics. The inset shows the regular bending circuit; (2) Metal conductor lines covered with silicone rubber; (3) Multi-layer structure or electrical nodes; (4) Three dimensional structure of printed conductor on paper; (5) Galvanical annulus wire attached with LED. (B) Optical images for printed functional components on coated paper: (1) Inductance coil; (2) RFID antenna; (3) Demonstration of flexibility of printed electronics [12].

possible clogging renders to the complexities of the inkjet system. Low throughput due to slow speed of inkjet printing process is a challenge for becoming an industrial production technique for printed electronics instead of its very promising results on laboratory scale. Low pattern resolution in the range of 20-50 µm and more, adds to the issues of inkjet system due to the spreading of solution on target substrate and chaotic behavior of droplets during the time of flight. Necessary modifications to the viscosity, concentration and solvent system are needed for proper ejection of the droplets without blocking the nozzle. Spreading of droplets, bulging out of the ink after sintering due to hydrophobic substrates, shape, thickness and morphology of the dried droplets has to be controlled [34, 121]. Different techniques for controlling wetting/dewetting of printed patterns on flexible substrates are

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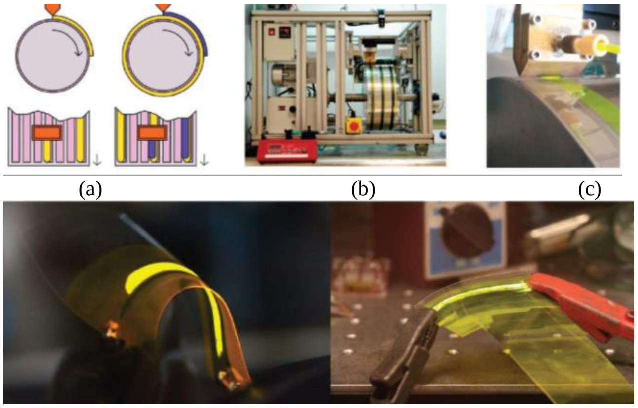


Fig.8. (a) Schematic view of the slot-die roll coating of the active layer (yellow) and the semitransparent anode (blue) on top of a flexible cathode-coated substrate (pink). (b) Photograph of a roll coater depositing the active layer. (c) Close-up photograph of the slot-die head during coating of an active layer stripe. (d) Photograph of a slot-die–coated LEC, illustrating the bidirectional light emission and the device conformability. (e) Light emission from a semitransparent slot-die–coated LEC following > 6 months storage in a glove box [13].

already under investigation and techniques like modifying surface properties of substrates by plasma treatment [137], localized micro-plasma treatment [138], tailoring adhesion and cohesion of ink particles within and with substrates [139], and adding gelating polymers [140] have been developed.

3) Slot Die Coating

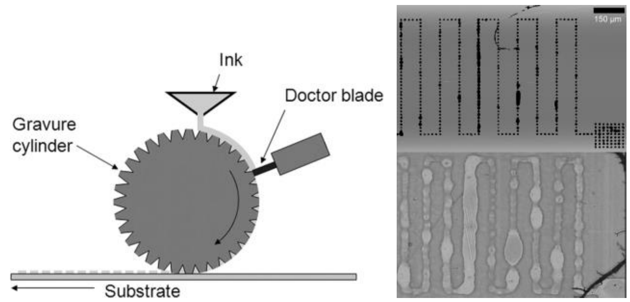
Slot die coating is a direct way of developing R2R process whereby solution is coated on the substrate by dispensing as shown in Fig. 8. Slot die coating process can be divided into two steps, a uniform flow of the coating solution achieved (Fig. 8 (a)) in the first step is followed by adjusting the operating variables like the stand-off between slot die and moving substrate, and speed of the substrate in the second step [86, 87]. The solution is poured from top through a via-opening shown in Fig. 8(a), and substrate mounted on the rotating cylinder Fig. 8(b). This type of coating is favorable for large areas, but pattering of high resolution structures is difficult to obtain. That is why this technique is usually practiced for large area devices i.e. light emitting diodes and solar cells. Fig. 8(d) shows image of a slot-die-coated light emitting electrochemical cell.

An operating envelope is usually developed for an optimized process by using the maximum and minimum coating speeds. The operation is affected by various coating defects such as dripping, air entrainment, ribbing, start-up and shut-down periods of coating cycle. Inefficient control of the printing process results in wastage of the coating solution and also affects the shape of the patterns on the substrate by introducing the edge effects [88-90]. Increased fluid viscosity, slot gap, coating gap and decreased dip lip length reduce the size of the coating bead, which consequently shortens the time required to reach steady state condition [89]. The stability issues related with this process makes it challenging to adapt this method for printing electronics on flexible substrates.

B. Contact Printing Technologies

1) Gravure Printing

Gravure printing utilizes direct transfer of functional inks



(a) (b)

Fig.9. (a) Schematic of a Gravure printing system, (b) Gravure Printed structures on flexible substrate [4].

through physical contact of the engraved structures with the substrate. It is capable of producing high quality patterns in a cost-effective manner typical of a R2R process. The gravure printing tools consist of a large cylinder electroplated with copper and engraved with micro cells, as shown in Fig. 9. The microcells are engraved either by using electromechanical means or using laser [18, 33, 121, 141]. Engraved cylinder is electroplated with chrome to protect it from wear and tear during the ink transfer and contact with the substrate. Engraved cells are filled with the ink either by using a reservoir beneath rotating gravure cylinder or through a nozzle dispenser from top, as shown in Fig. 9 (a). A doctor blade is used for removing extra ink from the rotating cylinder. Ink is transferred through capillary action onto a rollable substrate when it comes in between the engraved and impression cylinders. Surface properties of the substrate are also modified to facilitate the transfer of ink from the cells. Solution properties and cell width/depth ratio play major role in gravure printing along with other system parameters [121]. The low viscosity inks (Table II) are often used to prevent ink bleed out from the gravure cells. The low viscosity ink also speeds up the process, and allows emptying of the cells achieving better line resolution [142].

Smoothness, compressibility, porosity, ink receptivity, wettability, viscosity, solvent evaporation rate, drying, doctor blade angle and pressure, impression pressure, printing speed and uniformity of the gravure cylinder diameter are few of the parameters defining the printed results on flexible substrates. Experimental results show that the rate of ink transfer increases with increase in the surface energy and contact angle of the lower plate due to the strong adherence of ink to the substrate [143]. For efficient transfer of the ink, proper ratio of the cells width and depth (usually 7-8) is required [4]. This in turn facilitates uniformity and critically avoiding the crossover of neighboring pattern lines. Keeping the cell spacing at a proper ratio (usually between 1.06-1.4) [4], results in a fairly uniform lines otherwise increasing the ratio results in scalloped lines on the targeted substrates, which greatly degrade the print quality. The maximum acceptable inconsistency in the cell size and spacing is less than 1 µm. Optimum dimensions of the features on gravure cylinder are required which not only promises of less drop spacing but also has an adequate cell emptying capability [4]. The effect of

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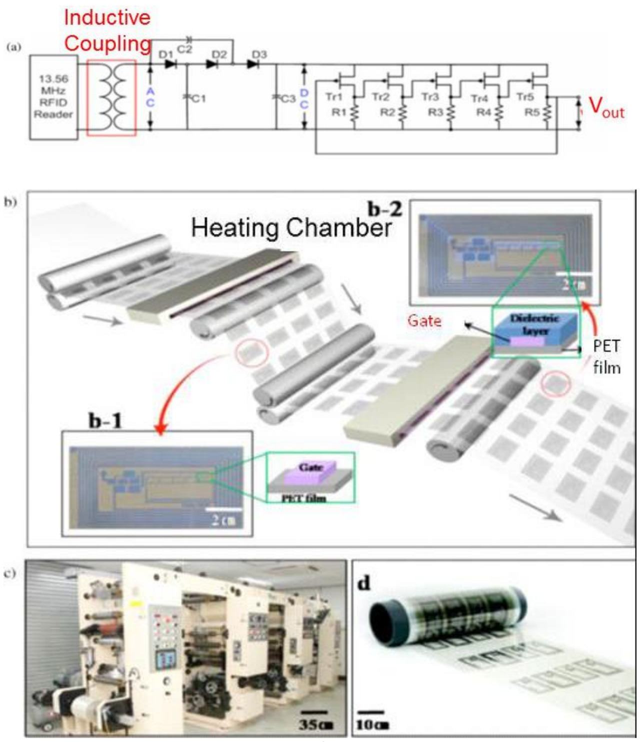

Fig.10. (a) Schematic circuit diagram for 13.56-MHz-operated 1-bit RF tag, (b-1) R2R gravure-printed antennas, electrodes and wires using silver- ink, (b-2) Printed layers of dielectric ink at selected spots, (c) Gravure printer with two printing units, (d) Image of printed 13.56-MHz-operated 1-bit RF tags [11].

shear force in the printing mechanism also has significant importance in gravure printing. The amount of transferred ink is enhanced as a result of decrement in the internal angle between the printing direction and the pattern-line direction, which increases the shear force. Experiments have shown magnifying dependence of the transferred ink on the angle between printing direction and the pattern-line direction [80]. If the line width is very narrow and the effect of the sidewalls is not negligible, it can be assumed that the sidewalls disturb the transfer ink more significantly along the cross direction than in the pattern-line direction [80].

Fig. 10 shows scheme of a fully gravure printed 13.56-MHz 1-bit RF tag obtained by printing conductive patterns and dielectric layers as precursors at different stages of the gravure system. Gravure printing based bottom gated TFTs, ring oscillators, solar cells, LEDs and sensors have been reported in literature [4, 33, 75, 121, 142-144]. Printed lines showed overlay printing registration accuracy (OPRA) of ±10 µm. Single wall carbon nanotubes (SWCNTs) are used as semiconducting layer in all gravure-printed TFTs reducing electrical fluctuations by controlling the OPRA and edge waviness of conductive patterns [4, 73, 75, 80, 144, 145].

Major obstacle that gravure printer is facing, is of printing lines with high resolution i.e. less than 20 µm [121]. The inability to produce uniform structures with sharp edge pattern lines [Fig. 9(b)] restricts the utility of gravure printing to fabrication of such layers in electronic devices, which do not require patterns of very high resolution. For this reason, gravure printing has been used for fabrication of organic flexible LEDs and photovoltaic devices. The frequent

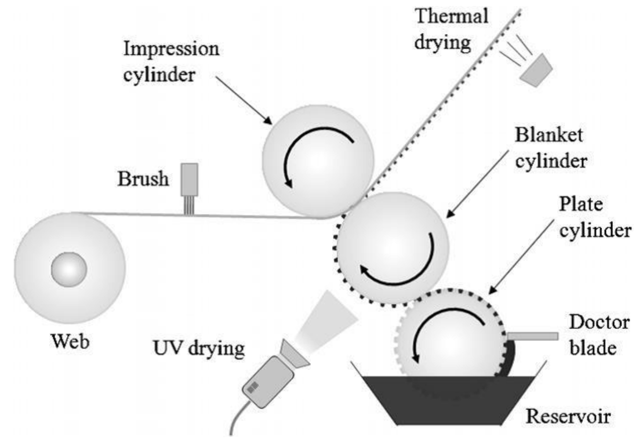

Fig.11. Schematic of a typical micro-gravure-offset printing process. Plate cylinder picks up the solution and transfers it to the blanket cylinder, which ultimately transfers the solution onto the substrate moving between impression and blanket cylinders [9].

replacement of gravure cylinders after continuous use in roll-to-roll (R2R) system adds to the maintenance cost of printing technology. Achieving the goals of getting small channel lengths, enhancing printing resolution with proper circuits design in order to avoid parasitic capacitances and gate overlaps are restricted by the limitations of the existing technology [33, 75, 146].

2) Gravure-Offset Printing

Gravure-offset printing is an advanced version of gravure printing in which an extra elastic blanket (Fig. 11) is used to avoid damages to the cylinder due to direct contact with the substrate [18, 147]. This elastic blanket picks up the ink from the grooves of the cylinder and transfers it to the targeted surface. Printing velocity, pressure and blanket's thickness are some of the process parameters that affect consistency of the printing results. Dependence on the printing speed and blanket's thickness are more dominant parameters due to the minimal contact time between ink and the blanket [80, 121, 143].

Electronic devices like TFT, resistors, RFID, sensors and solar cells have been developed with gravure-offset printing [73, 144, 145]. An overlay accuracy of ±50um is maintained during the entire printing process. Non-availability of proper printed inks for high frequency operations (>13.56MHz) diodes, adds to the complexities of the development of gravure-offset printed electronic systems [11, 79, 81-83].

Reliability of the gravure-offset printer is very critical for its application in printed electronics [82]. Several forces are involved in transfer of the ink including adhesive force between the blanket and the ink, cohesive force within the ink, adhesive force between the ink and gravure and adhesive force between ink and substrate. Surface energies of the blanket along with roll speed and pressure play important role in the strong adhesion between solution and the substrate. Proper manipulation of all these forces is important to pick and dispense the solvent on polymer substrate. Nearly 100% transfer of ink is desirable as any open holes or missing of dispense could possibly result in broken patterns and hence result in failure of the printed features [143]. Fast rolling speed

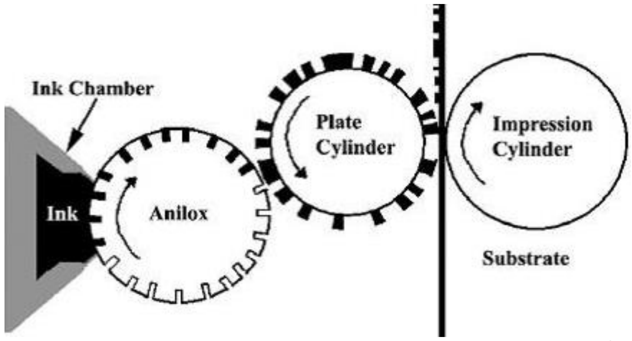

Fig.12. Flexographic Printing. Annilox cylinder picks up the ink from solution chamber, transfers it to the plate cylinder having raised structures, which ultimately transfers the ink onto the substrates running between the plate and the impression cylinders [8].

and non-uniform pressure may affect the print quality and reliability of the printing process [81]. The wave-like edges of printed patterns and blank areas are attributed to fast rolling speed and non-uniform pressure. Another issue related to this is the lifespan of the blanket, which is shortened by the continuous absorbance of the solvent. This results in reduced ink viscosity and affects the final pattern widths on the substrates by spread out during the set process. A successful printed device needs all the subsequent layers to be smooth and evenly deposited with defect-free layers [83].

3) Flexographic Printing

Flexographic printing is used for high speed runs of printed electronics and is more attractive than gravure and offset for high resolution patterns [18]. A wide variety of ink (solvent-based, water-based, electron-beam curing inks, UV curing inks and two part chemically curing inks etc.) can be printed by flexographic printing with a rubber or polymer plate having raised patterns that are developed by photolithography and are attached to a cylinder as shown in Fig. 12. On contact with the inked areas of the annilox cylinder, these raised patterns on plate cylinder (Fig. 12) serve to print on the substrate running between print/plate and impression cylinders. This results in uniform thin layers and offers improved pattern reliability and sharper edges than gravure printing. Annilox roll primary controls the quantity of ink to be transferred to the plate and to the substrate subsequently. The annilox volume i.e. size and frequency of engraved cells strongly affects the printed network tracks and sheet resistance. Filling the engraved annilox requires a delicate balance between mixture of nanoparticles and carrier fluid.

High concentrations of solutions are desired for good conductivities (in case of metallic inks), however this leads to high viscosities which do not fall into the operating envelope of typical flexographic printing [8, 84, 148]. Patterns with resolution between 50-100μm are reported in literature and with proper control of process parameters and substrate surface properties, this could be reduced to around 20μm [8, 84, 148]. Flexographically printed films are reported to be uniform and slightly less smooth than the spin-coated/gravure printed films [83]. However, flexographic printing is susceptible to film instability and dewetting, which facilitate many defects such as open lines, overlapped lines and edge waviness effects. Controlling the waviness of the print edges is very important especially in the case of antennas and RF performance. These issues can be resolved by controlling load pressure and cells aspect ratio.

Being dependent on the picked-up ink by the printing cylinder with engraved trenches (like in the gravure printing), flexography is an inconvenient way of getting continuous printing patterns in case if any of the cells is blocked or eroded by continuous operations. Also due to the inclusion of flexible plate for transferring ink onto substrate, the actual pattern lines tend to diverge from the targeted resolutions as the patterns on the flexible or polymer plate deform due to applied pressure.

This is a bottleneck in the way of creating narrower pattern lines on flexible substrates for high-resolution devices and structures [8, 10, 34, 84, 148]. An optimum range of width and thickness is needed for the printed patterns to decrease the ohmic losses and also increase efficiency of the printed devices [97]. For thick film deposition through flexographic, several printing passes with similar parameters are required, which also minimizes sheet resistance. Repeating the same procedures need proper alignment of the equipment for subsequent layers which adds to the complexities of the system [85]. The current technology limits the highly desirable features such as high switching speed and reduced supply voltage that are needed for many applications. These limitations result in degraded device parameters like charge-carrier mobility, parasitic capacitances and overlay precision registration accuracy [75]. Challenges to be overcome for fine patterning are surface irregularities and pores, nonuniform films, ragged lines and non-availability of suitable functional materials [149].

4) Micro-Contact (μCP)

For microcontact printing (μCP), a conformal contact of patterned elastomeric stamp with target surface is the key requirement for successful transfer of structures. Proper control and alignment of the stamps on micrometer scale is required. Microcontact printing has the ability to produce multiple copies of 2-dimensional patterns by using patterned stamp developed through master mold [7, 102-104]. Poly (dimthylsiloxane) (PDMS) is the frequently used elastomer due to its extraordinary properties as compared to other elastomers such as Polyurethanes, Polyimides, and cross-linked Novolac resin. Properties which distinguish PDMS from rest of the elastomers include conformability to large area, deformable to conform onto nonplanar surfaces, elasticity for easy release, low surface free energy, chemical inertness, homogeneous, isotropic, optically transparent and durability for multiple uses [7, 104]. Microcontact printing is an effective technique for preparation of substrates and patterning a wide range of materials, which are sensitive to light, and etchants. The master mold on a silicon wafer is often prepared with standard photolithography as shown in Fig. 13. Features are then replicated by pouring in the elastomer, which takes the shape of patterned mold Fig. 13(b-c). Photolithography is used for high resolutions and complex structures by incorporating microfluidic channels. Computer controlled milling machine is also used to develop master

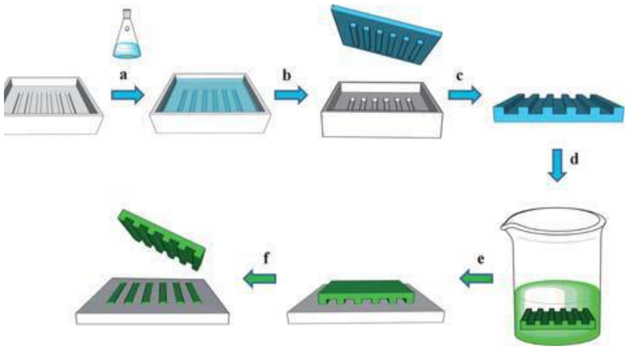

Fig.13. Microcontact printing (µCP) steps: (a) prepolymer poured on a photo-lithographically structured master, (b) curing of prepolymer and peeling off the elastomer stamp, (c) the stamp is cut in smaller pieces, (d) the stamp is inked by soaking it in an ink solution, (e) printing ink by contacting the stamp with a suitable surface, (f) patterned substrate [7].

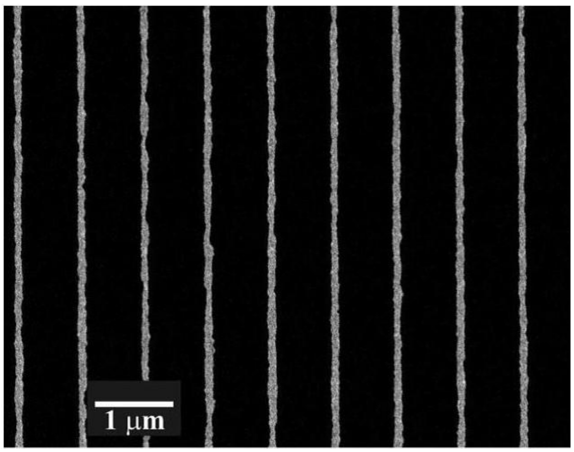

Fig.14. SEM image of 100nm wide Au lines by µCP using heavy-weight dendritic thioethers as inks and octadecanethiol as backfilling agent [2].

molds for microcontact printing in order to make route for researcher having no facilities of clean room environments [7, 150]. Surfaces like silicon, thick layers of SiO_2, PMMA and polypropylene are reported to be scribed by milling machine of which glass and PMMA could be scribed more satisfactorily than other materials [150]. Review of the evolution of microcontact printing over past few years categorically discussing the improvements made to the technique, new variations and new applications are discussed in detail [151]. The low surface energy due to flexibility of siloxane chain and low intermolecular forces between the methyl groups enhances the peeling and printing of the materials by PDMS stamp. The targeted substrate's surface energy must be high enough in order to release the inked material from the stamp. Therefore, the surface chemistries of the stamp and substrate are very important for efficient transfer. A proper ratio of height to width of the feature is required to avoid collapsing of stamp during peeling or capillary action during inking [106].

In order to make conformal contact with the substrate, the stamp must be flexible enough and also must have sufficient mechanical strength to maintain the topographical features during the printing process. The interaction of stamp, ink and substrate needs to be optimal to guarantee efficient delivery of ink only in the areas of contact. Fig. 14 shows SEM image of 100nm wide gold (Au) patterns developed by using µCP [2]. The flexible nature of stamp prohibits for going down for sub-micron features as the small features tend to collapse and larger noncontact areas tend to sag upon contact with the substrate. Polar molecules are difficult to print due to the hydrophobic nature of PDMS stamp, as a result insufficient ink is picked up by the stamp to transfer onto substrate [7]. Challenges in µCP include diffusion of SAM-forming molecules to areas not contacted by the stamp, broadening of features and blurring of feature edge. Furthermore, µCP requires precise adjustment of the surface energies for efficient transfer either by chemical modification or topography for the direct transfer from mold to the first or subsequent substrates [2, 34, 151]. Swelling of the soft polymer used for transferring micrometer-scaled patterns is another issue encountered in µCP, which often results in increased feature sizes. Excess ink can also enhance diffusion of the imprinted molecules on the surface accompanied by diffusion of non-covalently-bound molecules after printing. Deformation of the PDMS stamps due to their elastomeric nature, such as pairing, buckling or roof collapse of structures during contact with the surfaces is a problem that results in distorted patterns. Contamination of the patterns, influence of reproduction of the pattern upon force application, peeling the stamp from the master are also some of the main obstacles in stamp development, which gets complicated with nanometer-scale corrugations [2, 106, 151].

5) Nano-Imprinting (NI)

NI is used to pattern materials by mechanical and physical deformation of wet layer using hard or soft mold followed by different temperature processes. The principle of nanoimprinting is simple shown in Fig. 15. The system consist of a mold with nano-scale patterned structures on the surface that is pressed into a solution cast (Fig. 15(2)) on a substrate at a controlled temperature and pressure. As a result, thickness contrast into the casted layer is created. A thin residual layer of polymeric material is intentionally left underneath the mold protrusions, and acts as a soft cushioning layer that prevents direct impact of the hard mold on the substrate and effectively protects the delicate nanoscale features on the mold surface [152]. The two most crucial process steps that influence the pattern quality and throughput are resist filling and demold characteristics. A controlled pressure is needed not to destroy the imprint patterns [6]. Diverse NI approaches are developed i.e. thermal, photo, ultraviolet (UV), step-and-flash and roller nanoimprinting. In UV-NI transparent polymers are considered to be the best alternatives for developing reliable mold. Common mold materials are quartz or silica molded by electron beam lithography (EBL) method [105, 152-154].

Molding of polymers have some advantages as, they can replicate nanostructures over large area, materials are inexpensive and compatibility with low cost processes. Due to mechanical molding of the polymer material, NI is very challenging and entirely different from traditional fabrication techniques, embarking into new challenges and obstacles which are difficult to overcome [39]. The effect of spatially

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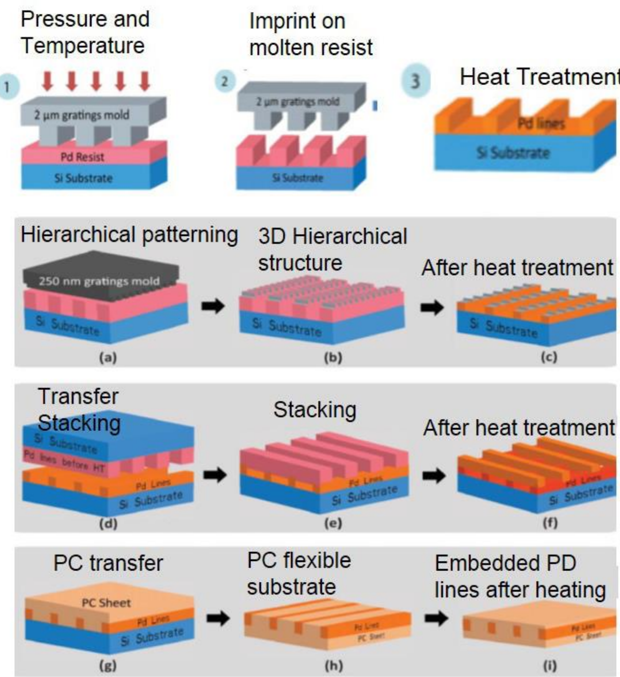

Fig.15. Scheme of noimprinting. (1) The spin coated Pd benzylthiolate film imprinted using a Si mold (2) The molten precursor flow into and fill the channels in the Si mold, (3) After demolding, Pd benzylthiolate patterns on the substrate are heat treated at 250°C for 1 hour to obtain Pd patterns [6].

confining materials to nanoscale dimension give rise to physical, electrical and chemical changes in properties that differ significantly from those of micro and macrostructures. Advantages claimed for developing NI technologies include high patterning resolution, high pattern transfer fidelity, 3-D patterning, large area, ability to reduce the fabrication steps, high throughput and low cost. But the challenges for NI technology overshadow these advantages. The critical challenges for NI include overlay alignment, template fabrication, defect control, high yield, and seeking especially suitable application fields [155]. Defect density and mask damage due to contact and low wafer throughput are few of the major complications in overcoming the challenges of this technology [6, 17, 152, 154]. Defect densities are very high and due to mechanical instabilities of the polymers leading to vertical and lateral collapse of features, this replication technique has very limited commercial applications. Coefficient of thermal expansion of the master and polymer replica has influence while removing the replica from master damaging the fragile nanostructures. Time required i.e. 10-15 min per replication for heating and cooling cycles, is longer than other soft printing techniques [153]. The thin layer of residue left on the mold features for soft cushioning to prevent the nano features from deteriorating upon contact with the soft polymeric material. In order to support alignment, fidelity and throughput with low defect densities in the printed patterns, a uniform thick residual layer is preferred but that renders the technology less critical for precise dimension control. The temperature budget of about 125°C is challenging for some of the plastic substrates with low glass transition temperature, which can create dimensional instabilities.

The residual layer to be removed via etching is very

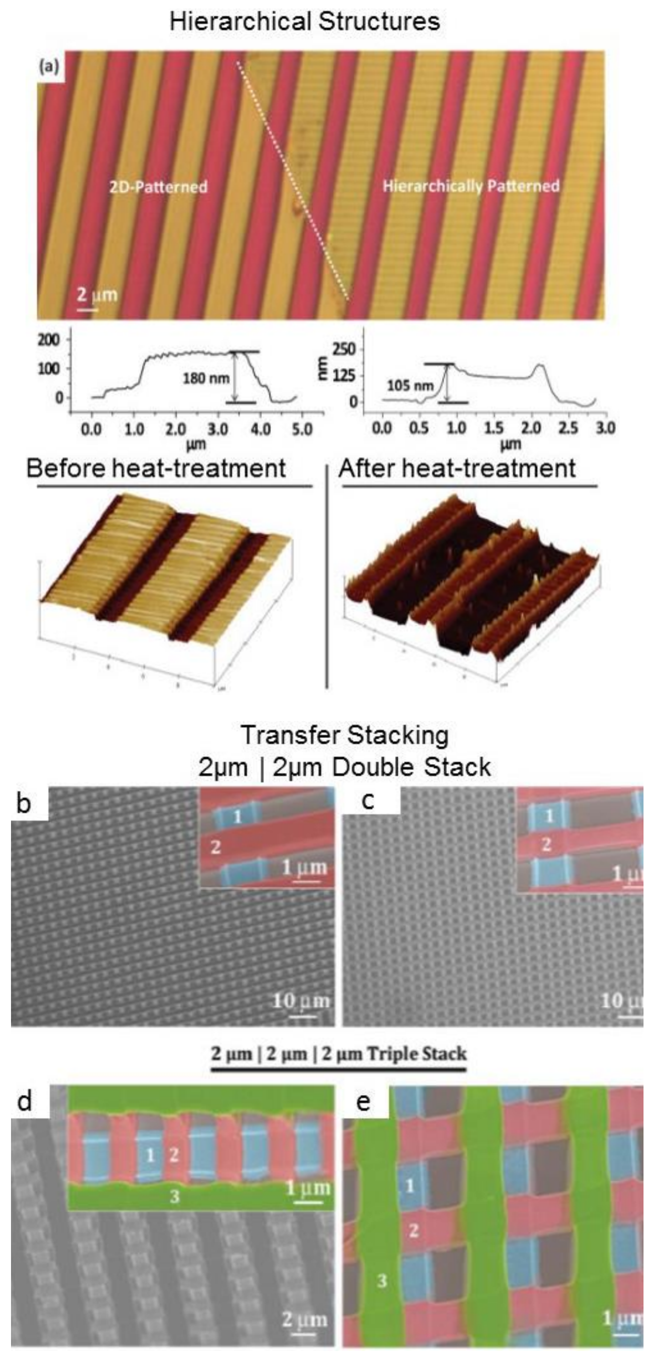

Fig.16. Optical image of large area Pd structures with a primary mold of 2mm gratings and secondary mold of 250nm gratings. SEM and AFM images of Pd hierarchical structures - before heat treatment (left). Widths of primary lines are 2mm and secondary lines are 160nm with corresponding heights of 180nm and 40nm, respectively [6].

undesirable as it is costly, hard-to-control, complicated to integrate into mass production line and might be harmful for an underlying organic layer. Several issues i.e. introducing soft gate dielectrics and misalignment of the stamp during the imprint process need to be taken care off before transferring NI process to a R2R manufacturing unit [156]. Particle related defects are one of the key concerns for nanoimprinting, since the particle can amplify the defect to become much larger than the particle itself [100]. Fig. 16 shows large area three stacks hierarchical structures with a primary mold of 2 mm gratings and secondary mold of 250 nm gratings successfully

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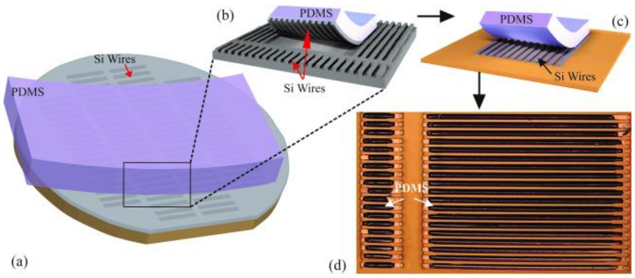

Fig.17. (a) Conformal contact of polymeric stamp with the Si wires, (b) Pick-up of wires by peeling-off the stamp, (c-d) Wires transferred to final substrate.

developed by using nanoimprinting [6]. TFT having channel lengths ranging from 5 µm to 250 nm are reported to be fabricated on Si and PEN foils. Designing on flexible and wavy foils, showing in-plane uncertainties and a high sensitivity to thermal as well as pressure changes, remains a big challenge [2, 100, 101, 106, 156].

6) Transfer Printing

In order to get fast flexible electronics, a new technique recently developed, i.e. Transfer Printing of silicon microwires and structures is getting great interest for the future trend of successful, fully flexible electronic devices and sensors. In this printing process the conventional photolithography technique is used to get micro and nano wires of Si on the wafers itself and then transferring them onto flexible substrates through a PDMS stamp [109-111, 157, 158]. Very well developed manufacturing and processing technologies for electronic grade silicon constituting high levels of purity, surface smoothness, control over crystallinity, doping concentration and type, and the resulting high carrier mobilities make it a distinguished candidate in the current scenario of large area electronics as well.

A conformable PDMS stamp is used to pick up the free standing microstructured silicon from top of Si wafers after etching and transferred with controlled orientation to flexible substrate. Fig. 17 shows the processing steps involved in realizing Si microwires and their transfer to secondary substrate. The PDMS stamp is peeled back retrieving the Si ribbons with fast speed enhancing the kinetic control of adhesion [159]. Rate dependent adhesion and printing of the solid structures with high peel velocity (typically 10 cm/sec) and low stamping velocity (~1 mm/sec) respectively has been investigated [40]. The mechanics of kinetic dependence of switching of adhesion has its origin in the viscoelastic response of the elastomeric stamp. Adhesiveless stamping like this is very valuable for wafer-based microstructures printing, to operate it from moderate to high temperatures [109].

Nanostructures through bottom-up approach are also being explored. The main challenges in the bottom-up approach of fabricating microstructures relate to control of dimensions, uniformity, the doping levels, crystallographic orientation and purity of the material. Also for scalable integration over large areas, producing well-arranged arrays of these structures are challenging [160]. Besides deploying nano/microstructures onto secondary substrate by the usual dry transfer printing through PDMS stamp, solution based printing by casting the

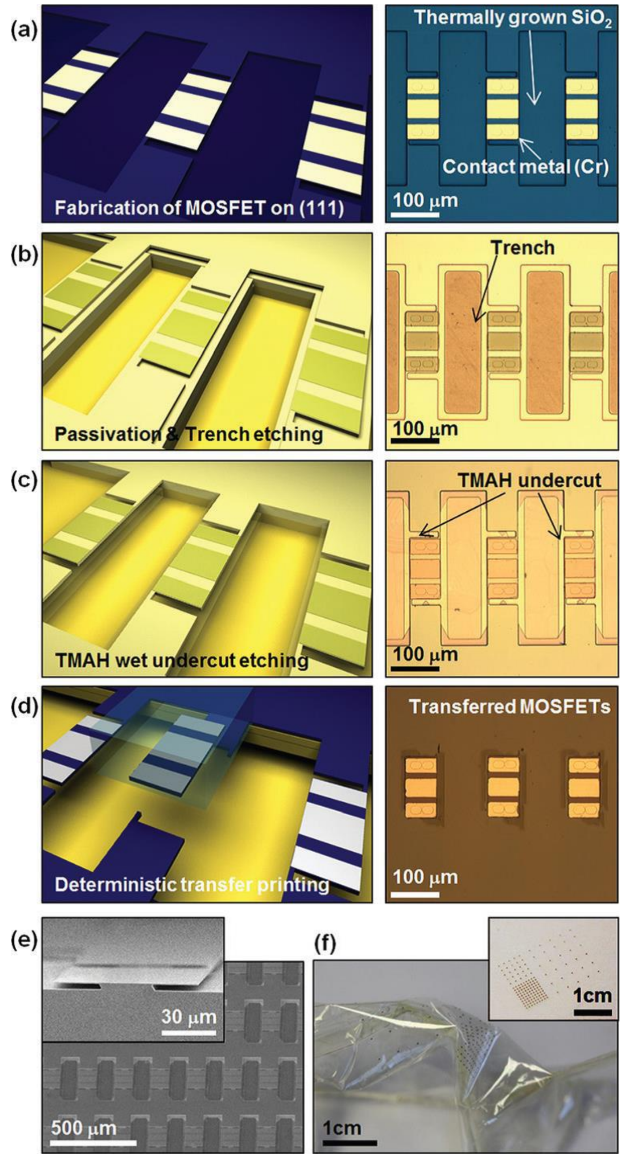

Fig.18. Schematic illustrations and optical microscope images of steps for forming releasable single crystal silicon MOSFETs on bulk wafers and their deterministic assembly on foreign substrates by transfer printing (a) fabrication of single crystal silicon MOSFETs with thermal gate oxide and metallization for source/drain and gate electrodes, (b) uniform deposition of a layer of SiNx followed by etching of trenches between the devices (c) wet anisotropic undercut etching using TMAH (d) manipulation of MOSFETs by transfer printing (e) SEM images of devices completely and partially undercut (inset). (f) 150 MOSFETs fabricated in this manner after transfer printing onto a substrate of ultrathin PET with thickness of 2.5µm [14].

microstructures in a solution are also practiced. Although manufacturability of wet transfer (through fluidic transfer) of structures is unclear as the doping levels and uniform surfaces are not well recognized [161].

Based on the active area needed and subsequent alignment defines the methodology for either using strips or membranes of Si in the transfer printing process for flexible electronic devices. Although both the approaches have pros and cons in relation to alignment and ease in undercutting of the structures due to the exposed area for etchants both for Si and BOX. The backside surface quality is acceptable in the flipped transferred NMs but the non-uniformity of the doping profile on the backside is the main limitation. Solid source diffusion has the issues of large feature sizes and driving the dopants in larger depth in NMs increasing sheet resistance. Instead ion implantation is employed which gives good results [110]. Simple integrated circuits like NMOSFET, CMOS inverters, sensors, three and five stage ring oscillators and differential amplifiers are reported to be developed on a flexible polyimide substrate using transfer printing of Si micro ribbons [111, 157, 162, 163]. Thermoelectric energy harvesters by transfer printing of arrays of alternately doped Si wires have also been reported in literature [158]. Flexible TFT with 1.5µm channel length was developed on plastic substrate showing very high frequency ranges in GHz. Radio frequency (RF) characterization under bending conditions showed slight performance enhancement with larger bending strains [116, 164]. Device performance can further be enhanced by using strained silicon channel [117]. A single-pole through switch containing two PIN diodes were realized and transferred to polymer substrate, by doing selective doping of 200-nm thick and 30um wide SiNM on SOI wafer. Fig. 18(a-f) shows images of printed multilayer of Si platelets, large area negative index metamaterial, epidermal electronics, LEDs, LED display and flexible integrated circuits transferred to secondary substrates through dry transfer technique respectively.

The rigid microstructures on flexible electronics experience tensile and compressive strains during bend into convex and concave shapes. The employment of these structures onto flexible substrates strongly depends on the failure mechanisms like interfacial slippage and delamination. The dimensions and mechanical properties of micro/nanoscale semiconductor wires, ribbons, bars, or membranes determine their bending mechanics. A practical design rule might be that the silicon strain must remain below 0.1%, which leads to a degree of bendability of r~2.5 cm for polymer substrate, which is still sufficient for many devices and applications [37, 107, 108]. Skinniness size of the Si nano membranes (SiNM) permit the planar-type structure to have very high level of mechanical bendability [165]. Rigorous necessities of active circuitry for large-area RF systems that could operate in L-band and even higher are required for flexible electronics applications [166]. Successful development of technology protocol for transferring Si based structures can complement the slow speeds of organic materials which could balance the total cost of manufacture of flexible devices. Challenges of misalignment of neighboring strips movement during undercutting, registration of pre-doped regions, gate dielectric materials at low temperatures and surface related issues for the stamps are very critical to be controlled [40].

C. Roll to Roll (R2R) Printing

The ultimate goal of above technologies is to obtain fast and efficient production line by merging various printing schemes. In highly optimized laboratory processes, better performance is achieved by developing a close relationship between the processing methods, materials, solvents, substrates and drying conditions. Printing processes matured at lab level, need to be transferred to large scale fast production lines with the same level of performance. Merging different printing techniques into a single production line is rather a challenging task, as very precise control of the process and material parameters need to be tuned especially when the substrate is moving with very high speeds as 5-50m/min (see Table II). Therefore, investigation of the optimal and matching processing conditions is desired for the new arrangement which is not at all obvious, especially when considering that there are boundary conditions (i.e. materials, solvents, multilayer processing, overlay registration accuracy, drying temperature, speed, etc.) involved in fast R2R processing [3, 11, 18, 128].

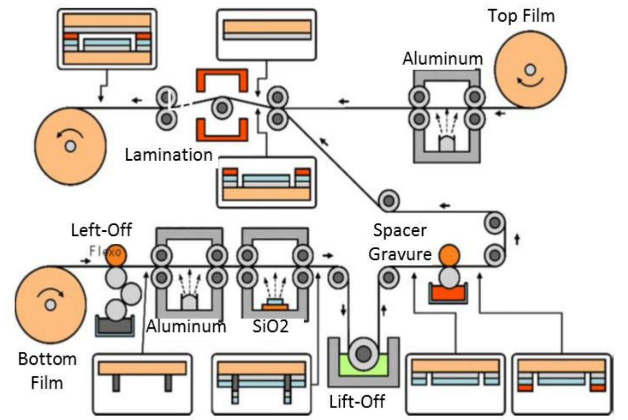

Fig.19. Schematic Illustration of Typical Roll-to-Roll system where different deposition, patterning and sintering modules are installed [10].

Fig.20. Image of a Roll-to-Roll (R2R) setup in a clean room environment with different printing process units installed in the production line [3]

R2R as a commonly shared platform has the potential for a continuous and high throughput process for deposition of diverse materials on large substrate rolls (often called "web") [3, 128]. Beside instrumentation and hardware for control system, R2R line is equipped with several rollers over which the web (flexible substrates) passes with controlled tension. As described in Section III, these webs are the backbone of a R2R system, and should be accurately controlled during passage through different rollers and processing sections. Two main rollers unwind/rewind installed at the ends, are dedicated for the release and collection of the web from the processing section of the R2R manufacture line [3, 10]. Fig. 19 shows schematic of a typical R2R system. Processing sections installed on a typical R2R line include tools for deposition, patterning and packaging based on the structural requirements of the device. Gravure, offset, flexography, rotary screen-

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printing and nanoimprint techniques are among the favorable candidates for R2R configuration and have been explored in detail [18, 34, 167-169]. Fig. 20 shows image of a R2R system in clean room environment.

R2R fabrication is more attractive for organic/polymer based thin film devices and have been explored extensively for solar cells, organic/polymer light emitting diodes, and display devices [10, 18, 167]. Beside these applications, focus is also towards development of sensory devices and patterned structures, which has become possible due to the rapid development of stable R2R systems with more patterning tools. Rectenna, solar cells and RFID vapor sensors developed through R2R fabrication on a PET substrate are reported [11, 79, 167, 170]. The combination of slot-die coating and laser direct writing on a R2R setup is reported very recently for development of piezoresistive strain sensors [171]. Recent advances in the large-scale integration of arrays for electronic and sensor applications involve the contact printing of single crystalline inorganic-nanowire (NW) at defined locations in R2R fashion.[169]. High-quality graphene film by R2R CVD and transfer process is also reported [172].

Significant progress has been made in design, technical and process capabilities of printing technologies in recent years. Much more work needs to be done before the field is ready to be scaled up for R2R process technology [128, 168]. Organization of the different film forming techniques according to the distinct categories of coating and printing is not straightforward. It is critical to develop a mechanics model to eliminate the gap between the conceptual design, materials and the process parameters [3]. One of the main challenges is how to model the effects of material, structure, and process together and optimize them to make reliable multilayered flexible sensors and devices with acceptable performance. Some of the other challenges are those that relate to the cost and performance of flexible circuits, panel size, process throughput, substrate distortion, barrier layer technology and yield of the process on which R2R technology is based [18, 128, 167, 169, 171]. Despite the vigorous attractions of large area flexible sensors and electronics, this new technology must overcome significant technical and process challenges in order to gain ground for practical high volume applications [128, 169].

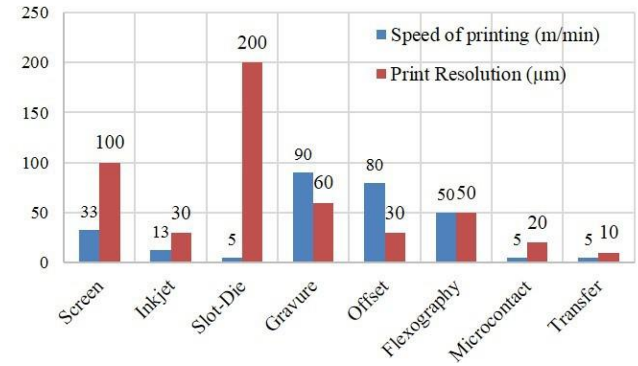

Fig. 21. Graph showing maximum values reported for Speed of printing and Print Resolution based on the data from Table II

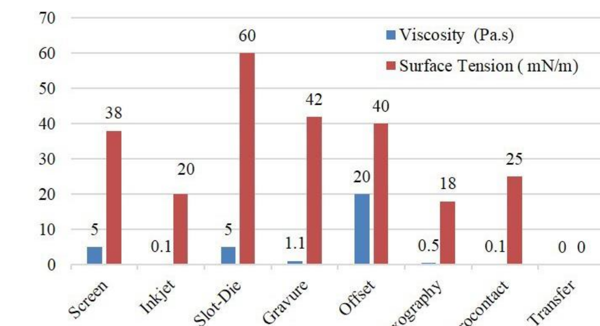

Fig. 22. Graph showing maximum values reported for viscosity and surface tension required for printing techniques based on the data from Table II.

Fig. 21 and Fig. 22 summarize the common features of various printing technologies. Fig. 21 shows the comparison of printing speeds and capability of print resolution that could be achieved by each printing technology. Whereas Fig. 22 shows the common material's properties, i.e. viscosity and surface tension of the solutions used in printing technologies. Table IV and Table V summarizes qualitatively the mechanism, process requirements, materials and critical limitations of non-contact and contact printing technologies reported in literature. Main features of all the printing technologies are highlighted to explore the possibilities of

TABLE IV
SUMMARY OF NON- CONTACT PRINTING TECHNIQUES AND CHALLENGES

Print Type	Mechanism and Features	Challenges	References
Screen	1. Most mature conventional printing technique. 2. Controlled deposition with fast speeds and versatile. 3. Desired pattern defined by open area of mesh. 4. Paste, printing process, substrate. 5. Squeegee, pressure and screens.	1. Drying of solvents in mask and deteriorated printed patterns. 2. High-resolution uniform line patterns not possible < 30 µm. 3. High viscosity required to prevent spreading and bleed out. 4. Dry out of the ink on the mask when kept for long time. 5. Large wet thickness of printed films and high surface roughness.	[18, 31, 34, 72, 91, 93, 94, 118-120, 123]
Inkjet	1. Low viscosity needed as compared to other techniques. 2. Ejection of droplets by using different actuation phenomena. 3. Thermal, Piezoelectric, and Electrostatic driven printer. 4. Position specific deposition of droplets. 5. Low material wastage.	1. Periodic bulging of lines with high contact angle in droplets. 2. Coffee-ring effect due to unequal distribution of dried solute. 3. Slow speed, multinozzle in parallel, misfiring, clogging. 4. Pixilation-related issues due to drop-on-demand. 5. Chaotic droplets at high frequencies and splashes with printed lines.	[38, 71, 95-99, 129-133]
Slot-Die	1. Uniform film thickness can be determined by controlling the feed rate and coating speed. 2. The first step establishes steady and uniform coating flow. 3. Second step adjusts gap between slot die and moving web. 4. Defect-free in long-time steady operation after adjustments. 5. Prewetting on die surface, fluid viscosity, slot gap, coating gap, and die lip length.	1. Operating window of operation is bound by a minimum and a maximum coating speed for uniform coatings. 2. Various coating defects, such as dripping, air entrainment, ribbing, etc. are observed outside the region. 3. Wastage of solution and unnecessary edge effects due to inefficient transient operation. 4. Proper control of start-up and shut-down periods are important.	[86, 88-90]

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TABLE V
SUMMARY OF CONTACT- PRINTING TECHNIQUES AND CHALLENGES

Print Type	Mechanism and Features	Challenges	References
Gravure	1. Ink pick-up by the engraved cells of the cylinder. 2. Substrate properties of interest - smoothness, compressibility porosity, ink receptivity and wettability. 3. Ink properties of interest - ink chemistry, viscosity, solvent evaporation rate and drying. 4. Doctor blade, angle and impression pressure, printing speed and uniformity of the gravure cylinder diameter.	1. Cylinder life and higher cost. 2. Proper ratio of cell spacing to cell width. 3. Consistent straight printed lines with fine edges. 4. Defects related challenges due to contact printing techniques. 5. Pick out effect due to direction of trenches to printing patterns 6. Enhancement of reliability for printed electronics. 7. Large degree of control for size and shape of conductive lines.	[4, 18, 31, 33, 34, 73, 75, 79, 121]
Offset	1. Elastic blanket roll picks up the ink from the grooves of the gravure plate and transfers it to the target surface. 2. The goal of the set process is 100% transference. 3. Enables the printing of ink onto hard surfaces. 4. Main process parameters are roll speed and pressure.	1. Width of the printed line increases with solvent absorption. 2. Lifespan of blanket expires quickly with absorbing solvent. 3. Viscosity thickening decreases blanket's absorbing power. 4. Spreading of line during set process. 5. High rolling resistance due to the fast rolling speed. 6. Wave like pattern's edge with the vertical direction.	[11, 18, 31, 34, 79-83, 121, 143]
Flexographic	1. Patterns are raised on low-cost flexible plate (attached to a cylinder) using photolithography. 2. High flexibility and low pressure imposed on substrate allows using this process for fragile and stiff substrates. 3. Better pattern quality and integrity in both vertical and horizontal compared to gravure.	1. Divergence from nominal specified values with squeezing. 2. Marbling effect, a typical printing problem. 3. Alignment of multi layers transfer appears cumbersome. 4. Tensile stresses occur with solvent evaporation or temperature. 5. Surface roughness of printed patterns approximately 6-8μm. 6. Layer cracks and non-uniform films.	[8, 10, 18, 31, 34, 75, 83-85, 97, 148]
Micro-Contact	1. Masters developed by photolithography and CNC. 2. Stamp is ''inked'' and put in contact with the substrate surface and transfer occurs at point of contact. 3. Straightforward method for the preparation of micro and nanostructured surfaces. 4. Mostly employed by the biological sciences	1. Peeling stamp from master with nano-structures corrugations. 2. Hydrophobicity of PDMS is a problem with polar inks. 3. Swelling of the stamp during inking increase the pattern sizes. 4. Diffusion of imprinted molecules on patterned surface. 5. Pattern reproduction challenge due to forces exerted on stamp. 6. Pairing, buckling or roof collapse of structures during contact.	[2, 7, 100-106]
Nano-Imprint	1. A hard mold developed by Photolithography that contains nanoscale surface-relief features. 2. Master pressed into a polymeric material cast at a controlled temperature and pressure. 3. Creating a thickness contrast in the polymeric material.	1. Damage to the fragile nanostructures when removing. 2. Coefficient of thermal expansion of master and polymer 3. Defect density control is the stringent requirement.. 4. More time required per replication for heating and cooling. 5. Vertical and lateral collapse of the replica features. 6. Poor fidelity of replication of nanostructures less than 50 nm.	[2, 7, 100-104, 106, 155, 156]
Transfer	1. Wires and membranes realized on SOI or bulk wafers using standard photolithography. 2. Transferring the structures using PDMS stamp onto the flexible substrates. 3. Transferring and printing through solution casting. 4. Photolithography + Micro Contact printing.	1. Misalignment of neighboring strips during undercutting. 2. Registration of pre-doped regions of transferred nanomembranes. 3. Active area needed for device & using strip or mesh NM. 4. Undercutting of strips and meshes based on exposed area. 5. Gate dielectric at low temperatures needs to be explored. 6. Stamp related issues as described in micro-contact printing. 7. Surface manipulation of the stamp for efficient transfer. 8. Backside surface quality of flipped transferred structures. 9. Doping profiles of flipped & non-flipped structures.	[37, 107-111, 116, 157, 159-164]

merging different techniques to develop a common manufacturing platform where limitations of one technology can be overcome by using another. Attractive features of high-resolution patterning and deposition of diverse materials by nanoimprint techniques can be harnessed by integrating them with the fast printing and coating tools for advancement of a R2R manufacturing system. Similarly, the development of soft polymeric stamps can provide a route to common platform for developing an optimized transfer printing protocol with photolithography, which could finally be implemented in fast R2R manufacturing track to achieve the real goal of low cost large area flexible sensors and electronics.

V. CONCLUSION

Printed sensors and electronics have attracted greater interest as printing enables low cost fabrication. The increased number of research articles and demonstration of printed sensors and electronics in a number of applications reflects the keen interest of the researchers in their quest to fulfill the promise of large area electronics on flexible substrates through cost-effective printing technologies. In this paper, we have presented a comprehensive overview of various technologies that have been employed so far for the printed devices such as TFTs, LEDs, sensors, displays, solar cells, RFID tags, printed batteries, energy harvesters and capacitors.

Material solutions with adjusted rheological properties and optimum processing parameters are the major paradigms for current research on printed electronics. Most of the existing printing technologies use solution based organic materials, which often result in transistors with modest performance, which is suitable for low end applications such as RFID and displays. The performance of printed devices is also affected by the resolution limits of current printing technologies, which is much poorer than possible with current micro/nanofabrication tools. For fast communication and computations in emerging areas such as internet of things

(IoT) will require cost-effective electronics with high-performance. Recent progresses with printing of high-mobility materials holds a great promise for the high-performance printed electronic systems. Advances in dry transfer printing of inorganic materials could complement the organic materials based solutions. A possible approach is to employ stamp printing techniques for high-mobility semiconductor material (both solution and solid state) deposition and exploiting conventional printing technique for interconnects and metallization. However, due to resolution limits of current printing tools the full potential of printing has not been realized. Printing of high-mobility materials with resolutions comparable with the current micro/nanofabrication tools will be a significant step towards cost-effective high-performance electronic systems. The hetero-integration, involving devices based on both organic and inorganic materials, is another interesting area that could lead to stable electronic systems with good mechanical, chemical and electrical properties.

The printed devices and circuits demonstrated in labs often use standalone printing technologies. For large scale production, there is a need to scale or merge these printing technologies on R2R production lines without sacrificing the chemical, physical or electrical characteristics of the device. The large area electronics through R2R production lines is foreseen to play a major role in the cost-effective manufacturing of nonconventional electronic devices and systems. Various mechanisms and challenges summarized in Table IV for each of the printing technology highlight the possible alternatives for developing a universal printing platform where limitations of one can be overcome by another while maintaining the optimum process parameters and solution properties. Development of an efficient platform by assembling different coating, printing and patterning tools to develop a very robust process protocol will result in high throughput and low cost devices.

Following the trends of paper printing industry, the manufacturing cost of plastic electronics is expected to reduce [128, 173, 174] by the fast speed printing of electronic components at defined locations [33]. The cost-effectiveness of printing technologies and employing them for flexible electronics will enable new classes of applications, and dramatically change the electronics industry landscape. Printed electronics and sensing will also have a major societal and economic impact with skilled labor from print industry gradually developing printed electronics.

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