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High Performance Printed Electronics on Large Area Flexible Substrates

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Abstract—**Printed electronics has attracted significant interest in recent years due to simple and cost effective fabrication of flexible devices with reduced e-waste. Further, it could potentially lead to development of multifunctional devices over large areas. Over the years, various printing technologies have been developed to pattern diverse flexible surfaces to develop wide range of electronic devices. A large part of the research so far has focussed on organic semiconductors based devices, even if the modest performance they offer is insufficient for several emerging applications (e. g. internet of things (IoTs), smart cities, robotics, etc.) where fast computation and communication are required. The high-performance requirements could be addressed with printed devices made from high-mobility materials based on single crystal silicon (Si) and graphene. This paper presents the key printing methodologies (i.e. contact and transfer printing) that are being explored for high-performance devices and circuits using nano to macro scale structures such as semiconductor nanowires (NWs), nanoribbon (NR), and ultra-thin chips (UTCs) as well as graphene. Few examples of high-performance devices obtained using contact and transfer printing are also presented.**

Keywords— Contact printing, Transfer printing, Flexible Electronics, Nanostructures, Ultra-thin Chips, Large Area Electronics

I. INTRODUCTION

Printed electronics is revolutionizing the next generation flexible electronics through simplified processing steps, location specific deposition, reduced materials wastage, low cost of fabrication, and low temperature processing and novel patterning techniques [1-4]. These attributes have motivated the development of various technologies for printing electronic devices and circuits on large area flexible, resorbable and biocompatible substrates. For example, the printed technologies have been explored for the development of devices such as printed Field effect Transistors (FETs) [5-8], optical [9], electrochemical [10] and pressure [11-16] sensors, radio frequency identification tags (RFID) [17, 18], supercapacitors [19, 20], solar cells [21], stretchable interconnects [3, 22] and light emitting diodes (LED) [23] etc. for applications such as electron skin (e-skin) and flexible displays [1, 24-27].

The initial advances in the field were driven by organic semiconductors [5]. However, their poor mobility limits the use of devices made from them to the low-end applications. The advent of printed electronics with organic semiconductors marked a positive development in the field, but the advances using these materials are not in sync with application requirements. For example, the switching frquency of transistors made from organic semiconductors is not good enough for fast communication and computation needed for applications such as internet of things (IoT) [16]. In this regard, the single crystal silicon (Si) is the natural choice as most of the planar electronics today uses this material and a mature CMOS technology exists. The challenge is to tailor this mature technology and make it compatible with flexible substrates and this is where methods such as transfer, and contact printing of nano/micro/macro structures come into picture. To this end, materials such as carbon nanotubes (CNTs), graphene, and Si nanostructures have been explored [16, 28, 29]. This paper presents few examples of high perfromance devices developed using transfer and contact printing. The discussion on transfer printing covers a range of nano to macro scale flexible devices such as nanoribbons (NRs) based transistors, ultrathin chips (UTCs) [5, 30] and graphene based touch sensors [28, 29, 31]. Subsequently, the Si and Zinc Oxide nanowire (NWs) based devices developed using contact printing are discussed [32].

The paper is organized as: Section II explains the difference between contact and transfer printing technologies. Section III presents examples of devices and circuits developed using transfer printing and Section IV presents the examples based on contact printing. Finally, the key outcomes and future research directions are summarised in Section V.

II. TRANSFER AND CONTACT PRINTING TECHNOLOGIES

A. *Transfer Printing:*

Using transfer printing technique, the micro/nano structures (Si, GaAs NWs or NRs) obtained using top-down or bottom-up approach are transferred over the desired flexible/rigid substrate using a PDMS stamp [33-37]. In an alternative transfer printing approach, the standard CMOS processed Si wafers were subjected to post-thinning of Si wafer and transferring [38-40]. CMOS compatibility opens up new avenues as the approach could be extended to obtain bendable chips with complex integrated circuits and eventually complete bendable sensing systems. Both the transfer printing approaches for large area flexible electronic devices is discussed below in details.

B. *Contact printing:*

Contact printing is a dry transfer technique which is suitable for any type of bottom-up (oxides, nitrides, Arsenide etc.) and top-down fabricated NWs. In contact printing process, the patterned NWs, NRs, in their respective donor substrates are brought in physical contact with the flexible receiver substrate. The contact-based printing technologies comprise of gravure printing, gravure-offset printing, flexographic printing and roll-to-roll (R2R) printing. The contact printing approach for the high-performance devices on large area is discussed in section III.

III. TRANSFER PRINTED DEVICES

(i) Nano-Ribbons (NRs) based devices

Transfer printing of high aspect ratio NWs and NRs using polymeric stamp involves a top down approach (as shown in Fig. 1(a)) [33-37, 41]. This technique allows to print NWs, NRs with uniform dimensions and high control over the nanostructures geometry (thickness, width and length) which results in NWs or NRs based large scale FETs. The selective doping of the NRs can be carried out using spin-on dopant (SOD) or ion implantation while the Si-NRs are still on the SOI substrate (Fig. 1(a)). A flat PDMS stamp is used to pick up the released Si-NRs after etching the buried oxide from the source wafer. The NR array subsequently transferred with high yield and controlled alignment to the destination flexible substrate [37] (Fig 1(b)). This process is advantages in term of low temperature processing of engineered array, CMOS

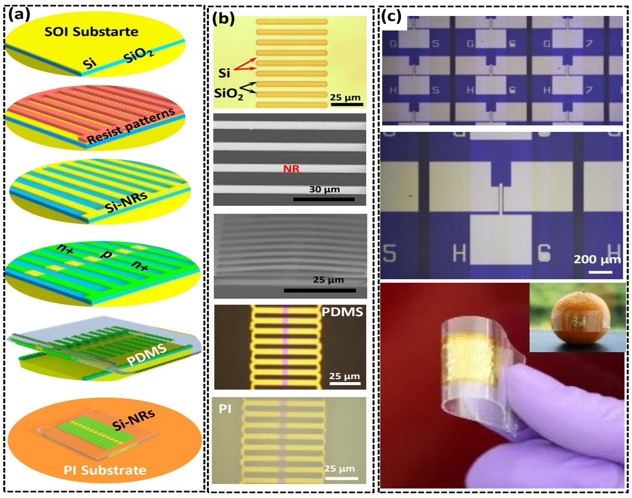

Fig. 1 (a) Steps for the fabrication and transfer printing of Si NRs. (b) Transferred NRs on desired flexible substrates. (c) Optical Images for the array of NRs – FETs.

compatibility and overall simplicity. After printing the active NR array to the flexible substrates (Fig 1(c)), the fabrication of NRFETs is completed by deposition of gate dielectric and contact metallization. The fabricated NRFETs demonstrated high performances, mobility > 600 cm^2/Vs with a high I_{ON}/I_{OFF} ratio $>10^6$. The Flexible NR – FETs enable the transfer printing process to be very robust for the development of large area high performance devices.

(ii) Ultrathin chips (UTC)

The ability to develop flexible devices and integrated circuits (ICs) based on aforementioned nano-materials remains an elusive task especially in regards with the reliability and performance over large areas. Ultrathin and flexible chips are expected to greatly enhance the emerging thin-film and organic semiconductor technologies by combining the well-known high performance of Si chip technology with the large area and system-in foil applications. In this direction, a cost effective method for obtaining ultra-thin chips on foil has been developed (Fig. 2). The chips have been obtained through post-processing steps, which means the method is compatible with standard CMOS process. CMOS compatibility opens up new avenues as the approach could be extended to obtain bendable chips with complex integrated circuits and eventually lead to the development of complete bendable sensing systems. The presented approach will possibly lead to new methods for heterogenous integration of organic and inorganic semiconductor based electronic components on foil -each complementing the other.

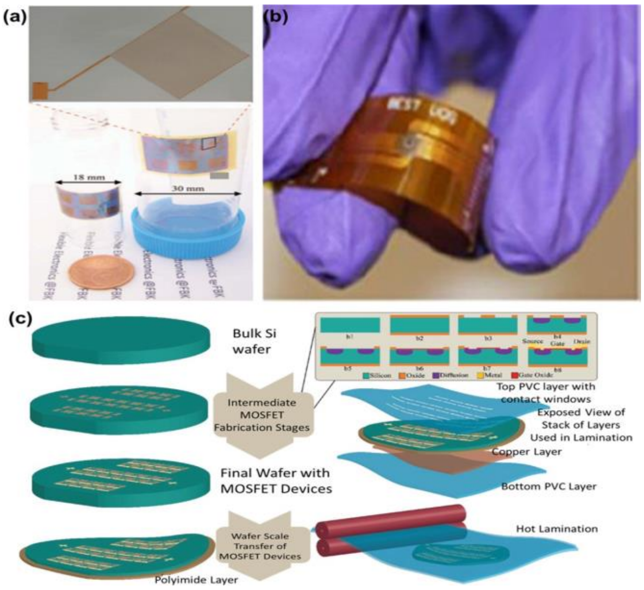

Fig. 2 (a) Passive line (Metallic line) on a flexible silicon [38]. (b) MOSFET on UTC bonded to a PCB [42]. (c) Schematic illustration of wafer scale transfer and packaging of ultra-thin chips [5].

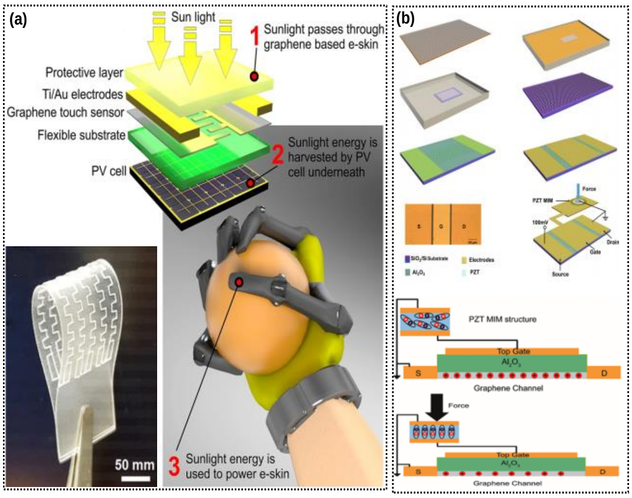

Fig. 3 Single layer graphene based touch sensor (a) using dry transfer [24], (b) wet transfer [29].

Circuits developed on thin Si would offer an alternative route to achieve the required performance for the devices [42, 43]. A number of technological advances have been developed to achieve a ultra-thin Si, an in-depth review on this can be found in [30]. Several different techniques have been developed showing the ultra-thin chip (UTC) fabrications (as shown in Fig. 2) [5, 38-40, 42, 43]. Post-thinning of the Si could be transferred to the destination substrate for the development of the flexible electronics. In this regard, we have explored the PDMS-assisted transfer printing which is widely used in transfer of micro/nano structures of Si [37] to achieve a wafer scale transfer of UTC [5], thereby, showing the promise of UTC for flexible electronics.

(iii) Transfer printed Graphene Based sensors

Graphene is considered to be one of the most promising material in the post-Si era owing to its intrinsic electrical, mechanical and optical properties. Especially, CVD grown graphene have been popular choice owing to its large area commercial viability. Dry and wet transfer process has been two widely adopted techniques for realisation of graphene based flexible devices [24, 28, 29]. We have explored the dry transfer process for realisation of energy autonomous graphene based capacitive touch sensors showing an excellent performance. The dry method enabled rapid and large-area transfer of graphene without affecting the intrinsic properties of graphene and utilized for the development of energy autonomous e-skin (shown in Fig. 3 (a))[24]. In another approach wet transfer process was adopted for the realisation of high performance graphene FET (GFET) [28, 29]. The developed GFET has also been explored for low voltage piezo potential based pressure sensors. Such sensors could pave away for the development of low power active matrix e-skin [28, 29] (shown in Fig. 3 (b)).

IV. CONTACT PRINTED DEVICES

Here we present the printed semiconducting NWs grown at high temperatures followed by contact printing.

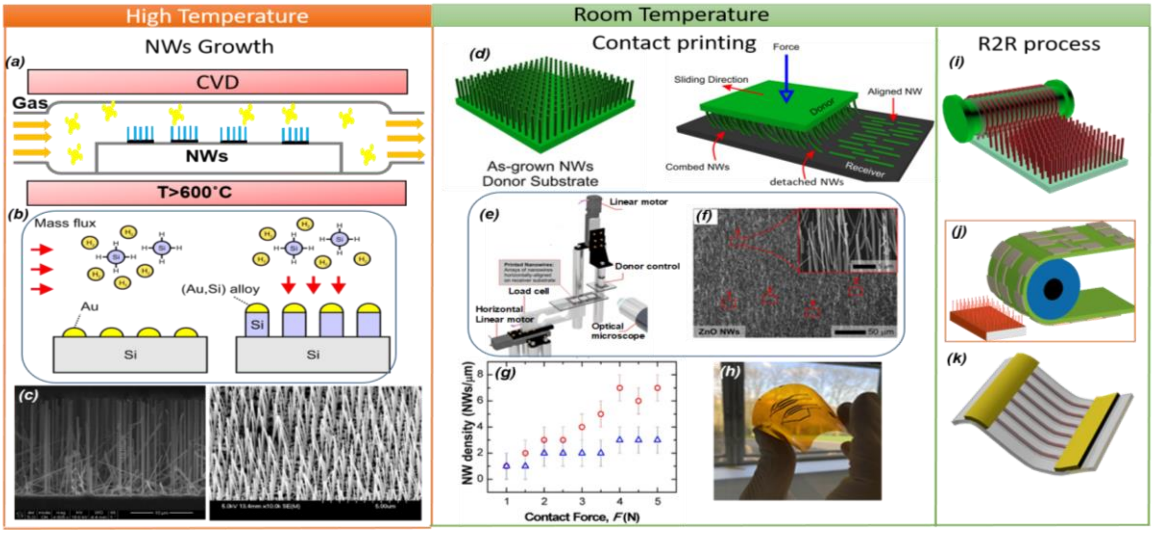

Fig. 4 Overview of the steps involved in a contact printing process. (a&b) High temperature CVD assisted growth of NWs by VLS mechanism. (c) Cross sectional SEM images of Si NWs. (d) Schematic illustration of contact printing (CP) process. (e) 3D illustration on in-house developed CP set up. (f) SEM image of contact printed highly dense ZnO NWs. (g) Statistical analysis of printed ZnO NWs, (h) Photograph of flexible UV photodetector developed using printed Si/ZnO NWs. (i-k) Schematic illustration of the concept of Roll to roll process based on contact printing.

Semiconducting NWs in the sub-100 nm diameter range have been conveniently grown using high temperature vapour phase techniques [44-46]. Chemical vapour deposition (CVD) based high temperature techniques offers unique advantages such as compositional control, wide materials system (Si, Ge, ZnO, III-Vs etc.), single crystalline structure etc., CVD assisted Vapour-Liquid-Sold (VLS) growth method is the popular technique for the growth of NWs in the diameter range of 10-100 nm. This high temperature process uses Si, sapphire, metals (steel) as substrates for the growth of these NWs (Fig. 4 (a-c)).

The printing system consists of a vertical motion-controlled stage where the rigid substrate (donor) is firmly attached. The flexible receiver substrate is fixed over a stage with horizontal movement. The directional sliding of donor substrate with controlled shear force over flexible receiver substrate plays central role in the printing process. The alignment of printed NWs is dictated by the controlled shear force acting the donor-receiver interface. Various factors governing the contact printing process are presented in Fig. 4 (d&e). The aim of the printing process is to obtain high yield, alignment, uniform interspacing and high density of NWs. Additionally, the process includes organic functionalization to improve the printing efficiency. These functionalization agents anchor the NWs during the transfer from donor to receiver and improve the surface selectivity and alignment. Contact printing is a promising technique for transferring these high temperature grown NWs over flexible polymer substrates such as PI, PET, PVC etc [47-50].

Printing of planar NWs heterostructures of Si/NWs over flexible PI has been demonstrated using an indigenously developed contact printing system (Fig. 4 (e-h)). Printing routines have been established using this system to obtain selective alternative layers of Si and ZnO NWs. A transparent flexible large area circuit UV photodetector has been shown using this printed NWs (Fig. 4h). The potential of the contact printed process is currently being extended towards large area R2R printing (Fig. 4 (i-k)). For example, bottom-up techniques have the ability to grow high quality semiconducting NWs over the surface of any geometrical shape such as cylinder, rods, rollable foils etc, which provides enormous freedom to design and develop such R2R NW printing systems. These developments will be a good platform for future nano-manufacturing tools and systems.

V. CONCLUSIONS

Printed flexible devices over large area demonstrating high performances have attracted greater interest for enabling low cost fabrication. In this paper, we have presented a comprehensive overview of contact and transfer printing technologies employed for the development of high performance and flexible FETs and sensors over large areas from nano to macro scale. With the help of transfer printing approach, large area alignment of NWs and NRs can be achieved. However, with the transfer printing approach, the complexity increases for building 3D stacks as well as integration with R2R printing. In contrast, contact printing is better suited for R2R printing and producing 3D stacks with quasi aligned nanostructures. As a future direction, the transfer printing approach can be modified to make it compatible with the R2R process, while the alignment of printed structures can be improved using contact printing. 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 cost effectiveness of printing technologies and employing them for flexible electronics will enable new classes of applications and is projected to dramatically change the electronics industry landscape. Printed electronics and sensing will also have a major societal and economic impact with skilled labour from print industry gradually developing printed electronics.

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