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Microcontact Printing of Polymeric Devices: Fabrication Techniques, Applications, and Challenges †

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Abstract: Microcontact printing (μCP) has become an emerging method for creating exact patterns on a range of substrates. This short review paper seeks to give a brief summary of the developments, fabrication techniques, applications, and difficulties in the microcontact printing of polymeric devices. In this systematic review, 20 papers from various fields were chosen for study. This review begins by introducing the basics of microcontact printing and discussing its capacity to transfer predetermined patterns with submicron resolution from an elastomeric stamp to substrates. Then, various microcontact printing production techniques for polymeric devices are reviewed. Furthermore, this review explores the broad range of applications enabled by microcontact printing, including electronics, biotechnology, nanotechnology, and surface engineering. Additionally, the potential difficulties and challenges associated with using microcontact printing processes are discussed. This literature review is to give researchers and practitioners a thorough understanding of microcontact printing by integrating the results from a few chosen studies. It promotes additional study and innovation in this promising sector by highlighting the most recent advancements, manufacturing techniques, and difficulties related to the manufacture of polymeric devices.

Keywords: microcontact printing; polymeric devices; applications; challenges

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1. Introduction

Microfabrication, the method of creating objects on a micrometer scale, has revolutionized several fields of science and technology [1]. It includes a range of methods that enable the precise manipulation of materials and designs [2]. Some common microfabrication methods include photolithography, deposition process, and soft lithography [3]. Among the soft lithography methods, microcontact printing (μCP) has emerged as a versatile and efficient technique to create patterns onto substrates with submicron resolution [4].

Microcontact printing was originally developed in 1996 [5]. It is based on a printing principle that uses an elastomeric stamp, ink, and a solid substrate on which the printed material is deposited. The stamp, typically made of polydimethylsiloxane (PDMS), acts as an ink pad, carrying the patterned molecules or materials [6]. Upon contact with the substrate, the pattern is transferred through selective adhesion [7] or transfer of functional molecules [8]. Various microcontact printing methods have been developed to cater to specific requirements. A wide variety of polymers [9,10], biomolecules [11–14], and nanoparticles [6,15] can be used to create micro- and nano-patterns using this relatively easy and affordable method. Although the effectiveness of μCP and its suitability for a wide variety of inks have been demonstrated on tiny surface areas, it has also been successfully experimented on large surface areas of 6×2 cm^2 [5] and even in large-scale production [16].

The versatility of microcontact printing finds application in a broad range of disciplines. In electronics, it facilitates the fabrication of micro- and nanoscale electronic devices, sensors,

and integrated circuits [17]. In biotechnology, microcontact printing enables the precise patterning of biomolecules, cells, and tissues, thereby enhancing the understanding of cellular interactions and the development of bioactive surfaces [18,19]. Furthermore, the technique has found utility in the creation of optical devices, microfluidics, and surface engineering [20–22], opening doors for innovations across multiple fields.

In this review article, 20 (twenty) published papers [3–22] on microcontact printing have been reviewed. The fabrication techniques described in those papers, characterization of the printed parts, their applications, and printing challenges have been discussed. In this paper, fabrication methods for three types of devices based on their purposes are discussed: mechanical, electronic, and biological devices. Ten papers about fabricating the devices and supports used for mechanical purposes were 3D microstructure channels by Borowiec et al., 2018 [18]; conductive structures by Hizir et al., 2020 [7]; 3D patches with self-assembly capabilities by Zimmermann et al., 2018 [9,10], and Sperling et al., 2019 [20]; variable height structures by Chang et al., 2022 [3]; SiOx/(Cyclo-olefin polymer) COP stamps for TiO2 patterning by Wu et al., 2020 [15]; the precision printing tool for rough surfaces by Akarsu et al., 2021 [8], covalent modification tool for 2D carbon nanomaterial by Zhu et al., 2018 [21]; and the embedded gold nanoparticle stripe into optical devices by Wang et al., 2020 [6]. Three papers about the electrical purposes were hybrid inverter circuits by Yu et al., 2021 [16], and soft and stretchable electronics by Yalcintas et al., 2019 [17], and Kumar et al., 2018 [14]. Seven papers about creating devices and tools for biological analysis were microarray and multiplex devices for cell culture and biological screening by Foncy et al., 2018 [5], Jin et al., 2023 [19], Hu et al., 2018 [11], Wu et al., 2018 [22], Qiu et al., 2021 [12], Gimenez et al., 2022 [13], and Ruan et al., 2019 [4].

Table S1 in the supplementary section provides a summary of the objective of each paper, the stamp, ink, and substrate used, and a brief description of the manufacturing methods.

2. Methodology

A comprehensive evaluation was conducted on the scholarly articles published between 2018 and 2023 about the manufacturing techniques of microcontact for polymeric devices, focusing on their applications and associated challenges. The literature search was performed on ScienceDirect, PubMed, and Google Scholar between 2018 and 2023. Various combinations of keywords such as “microcontact printing”, “polymeric device”, “application”, and “challenges” were utilized. Only English peer-reviewed studies that focused on the microcontact manufacturing of polymeric devices were chosen for the review.

3. Stamp, Ink and Substrate

In 18 (eighteen) papers out of 20 (twenty), PDMS was used for the stamp. The usual way of creating the stamp is by exposing a silicon wafer with SU8 polymer resist through a chromium mask. Once Sylgard 184 PDMS (a product by Dow Corning, Midland, MI, USA; mixed in a 1:10 ratio of curing agent to prepolymer) is poured onto the patterned wafer, it is subjected to a curing process at a temperature of 60 °C for a duration of four hours. Following this, the stamp is removed from the wafer [7]. Other materials used for the stamp are silicon and SiOx/(Cyclo-olefin polymer) COP. As for the substrate, glass was the most common material. However, PDMS, Quartz sheet, and polymer-coated glass were also used. With regard to ink, a wide variety of materials were used including polymers, metallic nanoparticles, and protein cells. Table S1 provides a detailed outline for those materials.

4. Fabrication

To perform microcontact printing through microthermoforming, Borowiec et al. inked the stamp and positioned it on the polycarbonate (PC) membrane as shown in Figure S1. It was then placed into a microthermoforming machine and heated to 100 °C. Later, 4-bar pressure was applied to wrap the membrane around the stamp pattern. Finally, the

machine was cooled down and the PC membrane was released from the mold. The final microchannel is shown in Figure S2a [18]. Hizir et al. used a roll based contact lithography method, utilizing equipment which was developed at the lab [7]. A printing roller was set up above a linear stage that had a vacuum chuck for holding substrates. A PDMS stamp was fitted with the roller with an ink pad placed on the chuck, and the roller was rolled over the inkpad. Later, the inkpad was replaced by the substrate and the roller with the stamp was rolled back in the opposite direction to transfer the ink onto the substrate. After that, the substrate was annealed at 150 °C for twenty minutes. They were able to print 5 µm hexagonal pattern printed on polymer substrate [7]. Zimmermann et al. [9,10] coated the PDMS stamp with polyethyleneimine (PEI) solution, pressed the loaded stamp on a silica particle monolayer and lifted the particles off the glass substrate as shown in Figure S3. The particles were released from the stamp using ultrasonic treatment in either ethanol or acetone creating 3D structure patches as shown in Figure S2b. In another study, using the same method, they produced single and double silica patches with different thicknesses and diameters. Foncy et al. [5] first fabricated three PDMS microfluidic devices that had chambers with inlets and outlet channels. Then, the microfluidic device and stamps were aligned, and ink was pumped into the chamber as shown in Figure S4. Later on, the stamp was separated from the device and pressed against the glass slide of area 6 cm × 2 cm for one minute. Finally, the stamp was taken off. Using the microfluidic devices, they were also able to perform multiplex printing as shown in Figure S2c.

As shown in Figure S5, Jin et al. [19] deposited antibody solution droplets onto the tips of PDMS stamp micropillars. The printed droplets rapidly evaporated and created a precipitate on the tips of the micropillars. After incubating, washing, and blow-drying the stamps, µCP was carried out on surface-activated coverslips to create antibody arrays with well-defined geometries as small as 20 µm as shown in Figure S2d. Yu et al. [16] used a low boiling point fluorinated ink solvent that facilitated the rapid formation of a hydrophobic ink layer along the intricate roll stamp pattern. As a result of the poor bonding between the hydrophobic siloxane polymer stamp and the fluorinated ink, the dried ink could easily be transferred to many substrates in the subsequent stage. In the process developed by Chang et al. [3], the use of octafluorocyclobutane (C4F8) plasma treatment replaced liquid ink solutions by depositing a fluorocarbon (FC) ink layer onto the stamp. As shown in Figure S6, by lightly positioning the stamp on the substrate and subjecting it to high-energy plasma-containing ions, thermal evaporation of FC ink could occur in the space between the stamp and substrate, creating inverted patterns that were transferred through plasma etching. This process also allowed multipatterning, as shown in Figure S2e. Figure S7 depicts the printing process utilizing microparticles coated with low molecular weight inks, as described by Sperling et al. [20]. The functional molecule rhodamine B isothiocyanate (RITC) was applied onto a PDMS stamp with a potentially functionalized surface, and then two surfaces were pressed together. Upon exposure to sonication, the patchy particles got detached from their substrate. Akarsu et al. [8] used poly-N-[tris(hydroxymethyl)-methyl]acrylamide (PTrisAAm) polymer brushes to alter the PDMS stamps. This modification allowed for the covalent immobilization of organosilanes. These immobilized organosilanes were subsequently transferred to oxide surfaces in a later step. This method effectively regulated the ink flow of organosilanes on capillary-active substrates with rough surfaces, resulting in high printing precisions. Figure S8 illustrates the printing process.

As per the method developed by Yilcintas et al. [17], the stamp was fastened to a 3-axis motion system and coated with eutectic gallium–indium (EGaIn) with the help of the roller. The stamp was then pressed onto the PDMS substrate under a continuous load. To finish the manufacturing of the flexible device, a coating of PDMS was applied over the printed circuitry to act as a seal. Figure S2f illustrates a printed electronic using this method. Hu et al. [11] performed the µCP using two methods: stamping and covalent [11]. For the first method, they had a stamp with an array of circular slots, and they treated it with O2 plasma. They coated their PDMS substrate with the fibronectin and added protein cells on top of

it. Afterward, they pressed the stamp onto the protein layer. As the stamp surface was highly reactive from the plasma treatment, the protein cells that came in contact with the stamp (except the circular slot areas) were peeled off when the stamp was removed, leaving behind a circular array of cells. For the second method, the researchers added the protein cells onto the stamp with cylindrical micropillar, treated the substrate with plasma, and used Ester-based protein immobilization method to bind the proteins onto the substrate. When the stamp and substrate were pressed together, the protein cells were transferred from the micropillar to the substrate. Using similar methods, Qiu et al. discussed μCP to construct platforms for cell cultures and biosensors [12]. They explained the process of creating arrays of protein cells, antibodies, and bacterial strains onto the substrates for further research.

Wu et al. (2020) [15] used vacuum ultraviolet (VUV) lithography to create a new cyclo-olefin polymer (COP) stamp. A hydrophilic SiOx/COP μCP stamp was created by forming a patterned relief on the COP plates, resulting in punch heights of around 180 nm. Subsequently, the titania precursor gel was transferred, which allowed the formation of TiO2 micropatterns on flexible polymer substrates. Zhu et al. [21] developed a simple yet powerful method for modifying sp2 hybridized carbon surfaces. They utilized the inverse electron demand Diels–Alder (IEDDA) reaction and microcontact printing to generate a patterned-modified surface by covalent modification. Initially, the PDMS stamp was filled with ink containing ferrocene-tetrazine (Fc-O-Tz), and then, the solvent was allowed to evaporate. Then, they pressed the stamp onto a highly ordered pyrolytic graphite (HOPG) surface, allowed it to react for five minutes, and then rinsed the surface off. Ruan et al. [4] started by spin-coating the PDMS with polyamic acid (PAA). Later, they printed the PAA on the quartz and then converted the PAA into the polyimide (PI) through the imidization process. After that, they used laser direct writing carborization (LDWc) to form a carbon disk array (CDA) of nanometer thickness. To make the holey array, the researchers dipped the PDMS with the PAA from the previous process into water and transferred the film onto the quartz substrate. Then, through the imidization and the LDWc process, holey carbon films were created as shown in Figure S2g. Wu et al. (2018) [22] initially coated the hydrophobic surface with a thin coating of homogeneous polydopamine (PDA). It was then brought into contact with an activated PDMS stamp. The elimination of PDA from the contact zone occurred when the stamp was released, leveraging the disparity in surface energy between the hydrophobic surface and the stamp. Consequently, the hydrophobic surface obtained a PDA pattern that matched the stamp. Negative microcontact printing enabled the effective creation of arrays of liquid droplets and single cells on perfluorinated surfaces.

Wang et al. [6] used wrinkled stamps to create hydrophobic stripe patterns on the substrate. The gap between the stripes was backfilled by hydroxyl-functional poly(2-vinyl pyridine) (P2VP), and the substrate was immersed in the gold-citrate solution and then washed to deposit the gold nanoparticles onto the P2VP stripe. In a subsequent microcontact printing stage, poly(ethylene imine) surface-decorated wrinkled stamps were used to create nanolattices, which is shown in Figure S2h [6]. Kumar et al. [14] coated the silicon substrate with silk [14]. Later, they loaded the stamp with either lithium bromide for positive resist or methanol for negative resist and stamped the silk film with it. After they were developed in water, the pattern was created. To make the split ring resonator (SRR), they coated the pattern with gold and immersed it in lithium bromide solution, which resulted in an SRR on the substrate. Gimenez et al. [13] presented a new lithography method that utilized grayscale patterns produced on a flexographic photopolymer mold. These patterns were then transferred to epoxy resin and a single PDMS stamp to create a range of microprint designs. Human-induced pluripotent stem cell (hiPSC) designs were created to test the functionality of this contact printing technique [13].

5. Application

Borowiec et al. suggested that creating small-scale surface patterns and transferring extracellular matrix (ECM) proteins can be utilized to mimic and produce intricate shapes with various applications in biotechnology [18]. Hizir et al. believed that using a printing roll to transfer ink onto the substrate could be helpful for high-volume production, especially in the fields of flexible electronics, optics, surface patterning, and photovoltaics [7]. 3D patches with self-assembly capacity developed by Zimmermann et al. will open up new possibilities for surfaces to be modified chemically and physically by incorporating the corresponding nanoadditives into their bulk [9,10]. Sperling et al. believed that by introducing patchy particles, anisotropic structures could be fabricated [20]. Chang et al. printed letters with three different pixel lines (colors) and are hopeful that FC structures can be used for biological implants [3]. Wu et al. (2020) mentioned that their tool can be used on a variety of substrates, particularly flexible polymer substrates, and it could help in the creation of stretchable and wearable gadgets [15]. Akarsu et al. opined that their method can be applied to create patterns on complex geometrical structures [8]. Zhu et al. found that tetrazine derivatives can efficiently react with an HOPG surface and with microcontact printing techniques, producing surfaces with micrometer-scale resolution. Their method can be used to alter graphene in a fast and catalysis-free environment [21]. Wang et al. created gold nanoparticle stripes to be embedded into optical and sensing devices [6].

Yu et al. experimented with printing fluoropolymer on organic (pentacene) and oxide (IGZO) Thin-Film Transistors (TFTs) and found that they worked well together to create hybrid inverter circuits [16]. Yalcintas et al. manufactured the stretchy microcapacitor and the microresistor, and the μCP developed by them is an efficient method for the scalable production of soft and flexible microelectronics [17]. Kumar et al. believe that new processes can be further developed for fabricating optics, surface engineering, MEMS, and integrated circuits [14]. Foncy et al. fabricated a cell microarray by successfully patterning prostate cancer cells on glass slides, which can be used to conduct research in biology and pharmacology such as drug screening, tissue engineering, etc. [5]. Jin et al. produced microarrays of antibodies with great resolution and multiplexity [19]. The analyses of Hu et al. can be used to understand how cell–biomaterial interactions at biointerfaces can be patterned at the microscopic level [11]. Wu et al. (2018) anticipated that the negative microcontact printing technique would find extensive use in high-throughput chemical and biological screening and analysis [22]. The electrically conductive CDAs and HCFs developed by Ruan et al. have a wide range of applications in electronics, photonics, energy storage, catalysis, tissue engineering, and physical and chemical sensing [4]. In their study, Qui et al. suggested that the patterned surfaces generated by μCP might be utilized to construct novel platforms for various practical applications such as biosensors, bacterial control, etc. [12]. Gimenez et al. believe that their method's adaptability makes it possible to quickly prototype micromolds and relief structures for use in microfluidic and microelectromechanical devices, and devices with chemical and biological applications [13].

6. Challenges

In the study of long-term cell culture on patterned structure, Borowiec et al. [18] reported that protein cells started to detach from the pattern after 10 days as shown in Figure S9a. Hizir et al. [7] observed two issues with their eighteen experimental results. Four of the printed models had clumping issues where, in a few places, materials got aggregated instead of depositing evenly. While pressing the PDMS stamp against the silicon particles and later on releasing the particles, some low molecular weight PDMS got transferred, which contaminated the whole substrate. Zimmermann et al. [9,10] solved the problem by using oxygen plasma treatment on the surface. Sperling et al. [20] reported that future research into the effects of various printing parameters, such as holding time, printing pressure, and printing temperature, as well as the effect of the polymer's chain length on printing performance, or the application of this method to other binding chemistries, represents a challenge. Chang et al. [3] used plasma etching to transfer the pattern onto

the substrate. High energetic ions can corrode the FC structures. Figure S9b shows the impact of etch selectivity on the layer depth. Therefore, selecting and maintaining the etch selectivity is important to obtain the desired pattern. The polymer brush-grafted μCP method introduced by Akarsu et al. [8] was able to transfer ink on uneven surfaces. However, the fluorescence image showed that there was a difference in printing precision in different areas.

Yu et al. [16] pointed out that the interfacial surface energy at the air-water interface and the contact angle of the liquid affects the adhesive force. Optimizing those two parameters for printing is important. According to Yalcintas et al. [17], in order for electrical devices to function properly after transferring LM to the elastomeric substrate, it is crucial to avoid any bridging, which refers to the presence of a residual EGaIn layer between the features. In addition, the analysis of six samples revealed that the EGaIn layer on the loaded stamps had a thickness of 2.4 $\pm$ 0.58 μm, as determined by examining 20 printed lines with line widths ranging from 15 to 100 μm. This considerable fluctuation may alter final EGaIn trace thicknesses. Jin et al. discussed two opposing effects of the hydrophobicity of PDMS on printing. Hydrophilic surfaces moistened the antibody droplet on the micropillar tip, making droplet printing easier, but their high surface energy made it difficult to transfer the antibody from the stamp to the coverslip in the μCP process [19]. Ruan et al. [4] opined that despite DLWc's success, creating homogenous and ultrathin carbon films with it remains difficult. This is largely due to the quick pyrolysis/carbonization reaction that the concentrated laser beam's extreme heating impact causes in the polymer precursor, which produces the porous structure that is commonly seen.

7. Discussion and Conclusions

This short review provides an overview of microcontact printing, a versatile technique for transferring predefined patterns onto substrates. Twenty relevant papers were selected for review, highlighting the recent advancements in this field. Different manufacturing methods for fabricating polymeric devices using microcontact printing have been explored in this review. The most common form of fabrication process is to coat the substrate with the desired material, load the stamp with the ink, and press it onto the substrate to obtain the desired geometry. The future applications of microcontact printing span across diverse fields, including electronics, tissue engineering, biotechnology, surface engineering, etc. However, challenges such as poor coverage, clumping, delamination, and contamination still need to be addressed. Future research should focus on overcoming these obstacles and investigating fresh applications to fully realize the potential of microcontact printing in expanding the boundaries of science and technology.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/engproc2024076042/s1, Table S1: Summary table of twenty paper; Figure S1: Microstructuring and microcontact printing of PC membrane by Borowiec et al. [18] © 2018, American Chemical Society. Used with permission; Figure S2: (a) Surface topography of thermoformed patterns by Borowiec et al. [18] © 2018, American Chemical Society. Used with permission. (b) Three-dimensional patches by Zimmermann et al. [9] © The Royal Society of Chemistry 2018; used under CC BY-NC (c) fluorescence image of multiplex printing by Foncy et al. [5] © 2018 Foncy et al. Used under CC BY (d) Fluorescence micrograph showing the printed antibody array on the coverslip (Scale bar, 50 μm) by Jin et al. [19] © 2023 Wiley-VCH GmbH; used with permission. (e) Multipatterning by plasma printing by Chang et al. [3] © 2022, American Chemical Society; used with permission. (f) Stretchable electronics developed by Sperling et al. [20] © The Royal Society of Chemistry 2019; used under CC BY-NA. (g) Holey arrays developed by Ruan et al. [4] © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. Used with permission. (h) Gold nanopartilce stripe (scale bar: 2 μm). printed by Wang et al. [6] © 2020 The Authors. Published by WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim; Used under CC; Figure S3: Silica particle printing by Zimmermann et al. [9,10] © The Royal Society of Chemistry 2018; used under CC BY-NC; Figure S4: Microcontact printing with microfluidic device by Foncy et al. [5] © 2018 Foncy et al. Used under CC BY; Figure S5: Microcontact printing by Jin et al. [19] © 2023 Wiley-VCH GmbH; used with permission; Figure S6: Plasma-assisted

microprinting by Chang et al. [3] © 2022, American Chemical Society; used with permission; Figure S7: Printing by Sperling et al. [20] © The Royal Society of Chemistry 2019; used under CC BY-NA; Figure S8: Printing by Akarsu et al. [8] © 2021 The Authors. Published by American Chemical Society; used under CC-BY-NC-ND 4.0; (a) Detaching cells after 10 days from Borowiec et al. [18] © 2018, American Chemical Society; used with permission. (b) Effect of etch selectivity on pattern depth from Chang et al. [3] © 2022, American Chemical Society; used with permission. (c) Fluorescence image of polymer brush grafted μCP from Akarsu et al. [8] © 2021 The Authors. Published by American Chemical Society; used under CC-BY-NC-ND 4.0.

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