

# Soft Microcontact Printing with Force Control using Microrobotic Assembly based Templates

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**Abstract**—In this study, soft microcontact printing with force control using microrobotic assembly based templates is investigated. Polystyrene microparticles are assembled automatically in a 2-D desired pattern on a glass substrate using an Atomic Force Microscope nanoprobe installed on a nanopositioning stage. A force-controlled printing process of the patterned stamp is conducted after making a template and stamp from the assembled microparticles. Aluminum sputtering of the pattern on the glass and ultrasonically removing the microparticles is used to make a template. Soft lithography method is used to mold elastomeric polymers on the template to make a stamp. The stamp is inked and printed with a force-controlled system on a Petri dish substrate. Depending on the particle size and contact force, a smaller micro or nanometer size pattern can be formed. Since the spherical patterns on the stamp collapse due to the interfacial contact force, force-controlled microcontact printing is crucial. Using a fluorescent protein for inking the stamps enables the fluorescent imaging of the imprints. Preliminary experiments using  $5\ \mu\text{m}$  and  $10\ \mu\text{m}$  diameter polystyrene particles showed the feasibility of our technique. Thus, it is possible to get nanopatterns using assembled microparticle based stamps in high volumes.

## I. INTRODUCTION

High volume fabrication of micro/nanoscale patterns has been a significant challenge in micro/nanorobotics and assembly area. Micro/nanorobotic precision assembly and manipulation systems are mostly use a single [1] or an array [2] of manipulators which results in a low volume and low speed manufacturing capability. As a possible solution to this substantial issue for industrial high throughput applications, micro/nanorobotic approach can be used to fabricate master templates or masks which could be replicated large number of times using high volume micro/nanofabrication techniques such as molding, contact printing, embossing, optical lithography, etc.

Microrobotic assembly of fine particles enables to produce a series of complex and precise patterns, hence a primary structure for printing templates. The fabrication of complex patterns of aligned microstructures has required the use of multiple applications of lithography [3]. However, the use of a single stamp as the patterning element removes the difficulty of aligning separate elastomeric stamps. Stamps may collapse due to interfacial adhesion. The low modulus and low surface energy of the elastomers allow atomic-scale

conformal contacts to be established without the application of an external force [4]. On the other hand, pressing the stamp against a surface collapses the topography of the spherical patterns on the stamp such that each recessed layer contacts the surface in stepwise sequence; the greater the applied force, the larger the area of the spheres that contacts the surface.

Therefore, a force-controlled printing is advantageous to get nanoimprints from micro features by controlling the contact force. This paper proposes a force-controlled contact printing of molded elastomer stamps using microassembled particles as a template. The organization of the paper is as follows: First, the microcontact printing steps are presented. Second, the microrobotic assembly of microparticles is explained. Then, the soft lithography and molding process are introduced. Next, the polymer inking and force-controlled printing are defined. Finally, the experimental results are discussed, and concluded with the summary of the current and future works.

## II. METHOD

The following schematic resembles the microrobotic assembly based templates and soft lithography steps of a desired pattern (Fig. 1). In Fig. 1a microparticles are pushed and assembled to form a pattern. Here four  $10\ \mu\text{m}$  and a  $25\ \mu\text{m}$  diameter particles are positioned in a row with given distance of  $30\ \mu\text{m}$  each. To make a patterned template,  $3\ \mu\text{m}$  uniform layer of aluminum is sputtered to fill the gaps around/between microparticles. Microparticles are then removed using the ultrasonic bath at high frequency and the holes remain under the microparticles where the aluminum is not exposed. The template can be used several times to make stamps by soft lithography. In Fig. 1b Poly(dimethylsiloxane) (PDMS) polymers fill the holes and after curing can be peeled off and used for stamping. The last step is to grow a layer of ink (Green Fluorescent Protein-GFP) on the patterned stamp. By controlling the time and contact force, microcontact printing is possible. In the following sections, these steps are explained in detail.

### A. Microrobotic Assembly

Using a XYZ positioning stage with few nanometer precision and an atomic force microscope nanoprobe as a manipulator for pushing particles enables precise microrobotic assembly

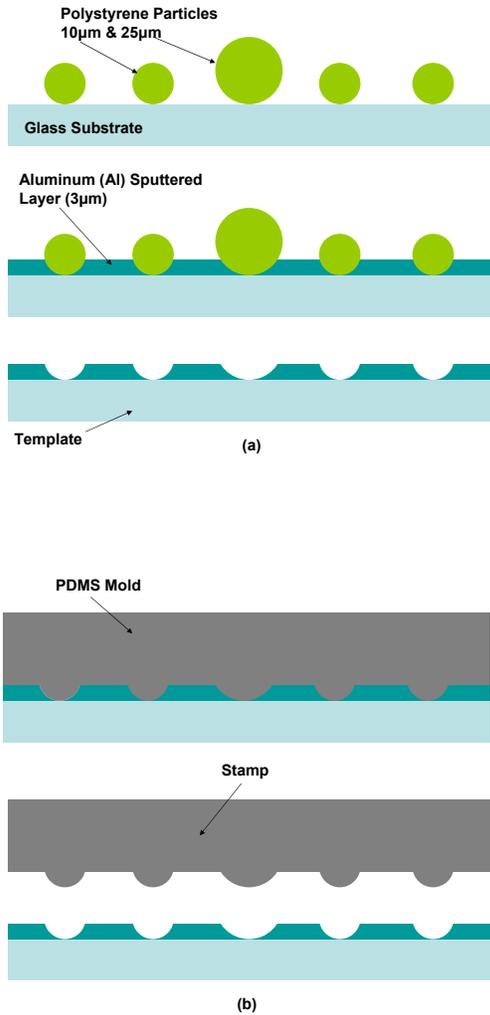


Fig. 1. Microrobotic assembly based templates and soft lithography steps of a desired pattern: (a) Making the template by microrobotic assembly method; (b) Molding process and making the stamps

in 2-D [5]. Advances in vision based autonomous assembly is beneficial for making any desired patterns of microparticles in few minutes. For smaller scale where the visual feedback is not sufficient, developed virtual reality environment provides a physical user interface for advanced telemanipulation [6] and assembly of nanoparticles with force feedback [7].

In the current vision based micromanipulation system (Fig. 2) [8], an image processing tool detects the microparticles in real time using the generalized Hough transform. Initial positions are defined as circles and the goals as pluses to determine the final desired position. There is a Wavefront expansion motion planner to avoid obstacles and to find the closest path and particles to the goal (Fig. 3). It takes few minutes to assemble a simple pattern by several pushings of the polystyrene microparticles.

Sample patterns are assembled and depicted by the vi-

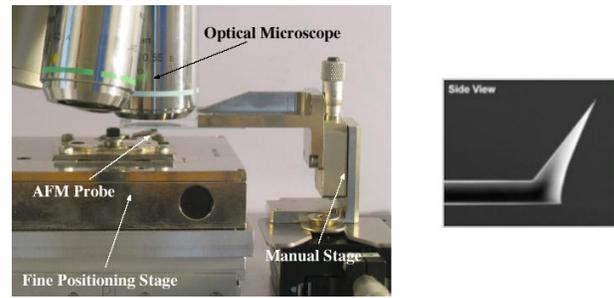


Fig. 2. Micromanipulation system: (left) Actual system under microscope; (right) AFM probe side view

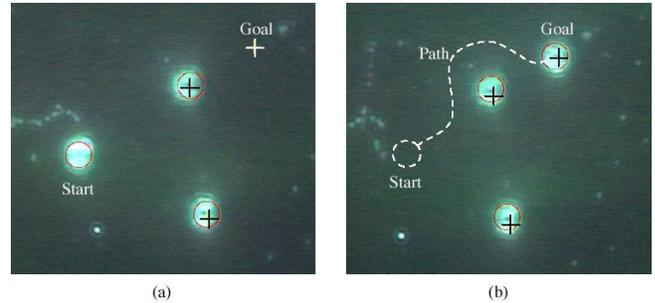
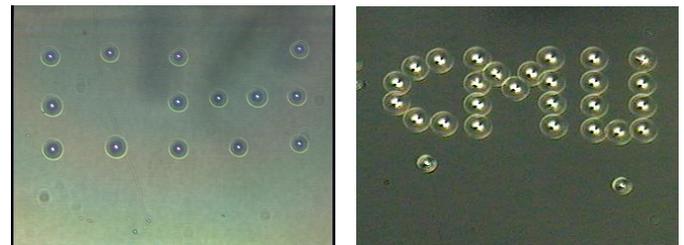


Fig. 3. (a) Initial configuration of particles, goal position marked by user; (b) particle pushed to goal by motion planning

sion based manipulation as in Fig. 4. A high powered light microscope with up to 1000 $\times$  magnification (Nikon Eclipse L200) is capable of detecting micro-scale particles down to 1 micron. Templates can be designed as simple geometries of repeated letters, squares or a class of aligned patterns using different size particles. Particles in a single letter should be close enough, so that the resulting prints look more of a continuous pattern while the contact time and force are being changed.



(a) 5  $\mu\text{m}$  diameter polystyrene microparticles for writing letters-CM, zoom: 1000 $\times$   
 (b) 10  $\mu\text{m}$  diameter polystyrene microparticles for writing 'CMU' pattern, zoom: 500 $\times$

Fig. 4. Sample patterns made using the vision based microassembly technique with a nanoprobe

First step for making stamp is designing a patterned template that can be molded several times. The templates are produced by aluminum sputtering of the manipulated particles on the glass substrate followed by the high frequency sonica-

tion bath to remove microparticles. After the manipulation task which basically produces the pattern, aluminum sputtering of the substrate is realized using Perkin Elmer 2400 8L sputtering system. For every one micron aluminum layer, it takes about an hour of sputtering time. The pattern depths can be varied depending on the sputtered aluminum layer. It is like a half sphere that gets smaller in radius as it goes deeper and flattens at the tip. The sputtering power is kept low so that the chamber temperature does not rise to the melting temperature of the polystyrene particles. It is proposed to have a ten minute break after every one micron aluminum sputtering to cool down the chamber. During sputtering the aluminum layer fills the gaps between particles and form a smooth thin layer with remaining holes on the unexposed surfaces covered by microparticles as shown in Fig. 5. The microparticles are then released from the holes using an ultrasonic bath system.

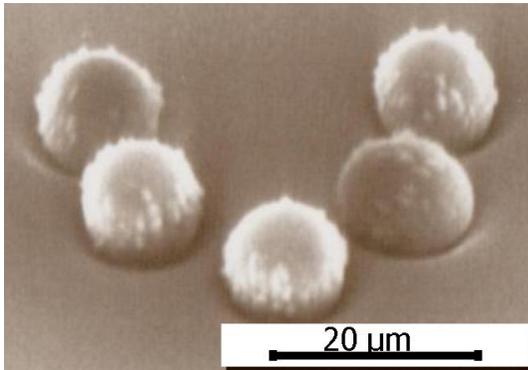


Fig. 5. SEM image of the 10  $\mu\text{m}$  diameter polystyrene template after aluminum sputtering and before ultrasonic removal of particles

### B. Soft Lithography

Soft lithography is a method for transferring a surface pattern that makes use of an elastomeric stamp. Poly(dimethylsiloxane) (PDMS), a soft polymer, is used to make the stamp from the template. The PDMS base (Sylgard 184) and the curing agent are mixed 10:1 weight ratio. Mixing with the curing agent produces bubbles inside the solution that might later form a cavity in the stamp which is undesirable. To avoid that, the template and the PDMS solution are both placed inside the vacuum chamber for about 30 minutes to de-bubble. Then, the chamber is tilted so that the PDMS covers the empty holes of the pattern gradually. PDMS is cured on the hot plate in 100  $^{\circ}\text{C}$  temperature for 45 minutes and can be peeled off from the template.

Thicker stamps are better for easier pattern transfer, but it would be hard to peel off. For very thick stamps there is a chance of destroying the aluminum layer of the template during peel off and losing the chance of using them several times. It is also a mirrored image that produces the right image after printing.

Since the PDMS bumps replicating the holes have low aspect ratio, it is challenging to transfer the patterns. Therefore, harder stamps can be also made by using other polymers like

SU-8. However, since PDMS surface chemistry and stamping process are well-defined, and soft PDMS has more robustness against alignment errors, PDMS is used in the experiments as the stamp material.

### C. Polymer inking

Since the process is analogous to stamping with liquid ink, this process is named polymer inking. First insert the polymer ink on the molded stamp patterns and wait to form a chemical bond to the surface. An alternative is to spin-coat a polymer ink layer on a flat substrate [9]. The stamp with predefined patterns is pressed against the polymer layer on the ink pad at a temperature close to the glass transition temperature ( $T_g$ ) of the polymer. Pressing with the stamp causes deformation of the polymer layer. When the stamp is removed subsequently from the ink pad, it will take with it the polymer ink that is adhered on top of the protrusion patterns.

Here green fluorescent protein (GFP) is used on the patterned PDMS stamp to transfer the pattern. To prepare and use the GFP solution, it is diluted in PBS. Successful printing of GFP requires inking stamps with a solution of GFP having a concentration greater than 50  $\mu\text{g mL}^{-1}$ . The prepared solution is put on the stamp which has been cleaned and rinsed in the water-ethanol (80:20) mixture. The stamp is placed in a polystyrene Petri dish containing droplets of water to prevent the instant evaporation of the ink solution for one hour. The stamp and the GFP should be covered so the light does not change the fluorescence characteristics of the protein. The goal is to have the GFP form a chemical bond with the stamp after it settles down in one hour. It is required to have a protein layer forming on the pattern so when the solution is sucked out from the stamp the bonded layer stays there. The protein layer is observed under the fluorescent inverted microscope as a green layer before and after stamping to make sure the GFP is transferred to the substrate. It is critical not to wait very long after sucking the solution for the GFP gets dried out so that the printing stage is unsuccessful. A key is to keep the time short between drying of the stamp (after its inking) and printing of the proteins to less than 1 minute [10]. We can still try stamping several times with higher contact force to get several prints from the same inked pattern. Stamps can be reused for 50 times if they are sonicated in water-ethanol mixture for 5 minutes after each inking and printing cycle [11].

### D. Force-controlled printing

1) *Theory*: To calculate for the deformations of the spherical stamp patterns pressed on the Petri dish substrate, Johnson-Kendall-Roberts (JKR) contact mechanics model [12] is used. It predicts high elastic deformation of soft and highly adhering materials correctly. Using this model, contact radius  $a$  of a particle on a flat surface with a normal load of  $P$  is given as

$$a = \left( \frac{R}{K} \left( P + 3\pi R\omega + \sqrt{6\pi R\omega P + (3\pi R\omega)^2} \right) \right)^{1/3} \quad (1)$$

where  $K$  is the equivalent modulus of elasticity of the materials in contact,  $R$  is the radius of the spherical patterns on the stamp, and  $\omega$  is the interfacial adhesion energy between the stamp and the Petri dish substrate. For sphere on a flat surface,  $K$  is derived as

$$K = \frac{4}{3} \left( \frac{1 - \nu_1^2}{E_1} + \frac{1 - \nu_2^2}{E_2} \right)^{-1} \quad (2)$$

Assuming  $\nu_1 = 0.5$  and  $E_1 = 750 \text{ KPa}$  for the PDMS stamp and  $\nu_2 = 0.27$  and  $E_2 = 169 \text{ GPa}$  for the polystyrene Petri dish substrate,  $K = 1326 \text{ KPa}$ . Depending on surfaces in contact and also a polymer ink used, adhesion energy is varied from 25 to  $1000 \text{ mJ/m}^2$  with a nominal value of  $100 \text{ mJ/m}^2$ . Nominal spherical radius of the stamp pattern is taken as  $R = 5 \text{ }\mu\text{m}$ . The contact area of the pattern features increases by applying higher contact forces (Fig. 6 and 7). Fig. 6 predicts the contact radius of the spherical patterns as a function of contact load. Applying  $30 \text{ mN}$  contact force on  $5 \text{ }\mu\text{m}$  patterns can fully collapse them to get a full print of the pattern. It requires a less contact force to collapse smaller spherical patterns. In order to get nanoprints from the  $5 \text{ }\mu\text{m}$  pattern, small contact force of about  $0.1 \text{ mN}$  should be applied (Fig. 7). However, even with zero force contact, minimum print size is around  $1 \text{ }\mu\text{m}$  for  $R = 5 \text{ }\mu\text{m}$ . For  $R = 0.5 \text{ }\mu\text{m}$ , smallest features can be as small as around  $100 \text{ nm}$  diameter. The contact radius is also sensitive to the adhesion energies between surfaces in contact. The protein layer changes the adhesion energy and smaller adhesion enables smaller patterns although risking the transfer of the ink to the substrate.

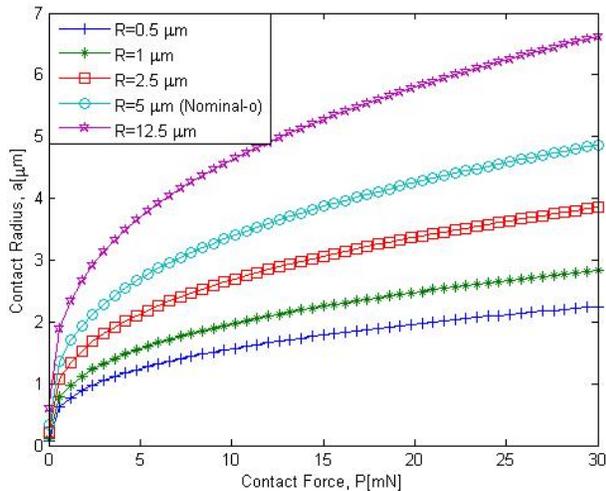


Fig. 6. Contact radius between different spherical patterns ( $0.5\text{--}12.5 \text{ }\mu\text{m}$ ) on the stamp and the substrate ( $\omega = 100 \text{ mJ/m}^2$ ) as a function of contact force

Most of the polymers like PDMS are hydrophobic so it is better to stamp them on very hydrophilic substrates. It is possible to modify the surface chemistry by an appropriate material.  $O_2$  plasma treatment of the PDMS or a layer of Silane is often necessary to create a hydrophilic surface, which

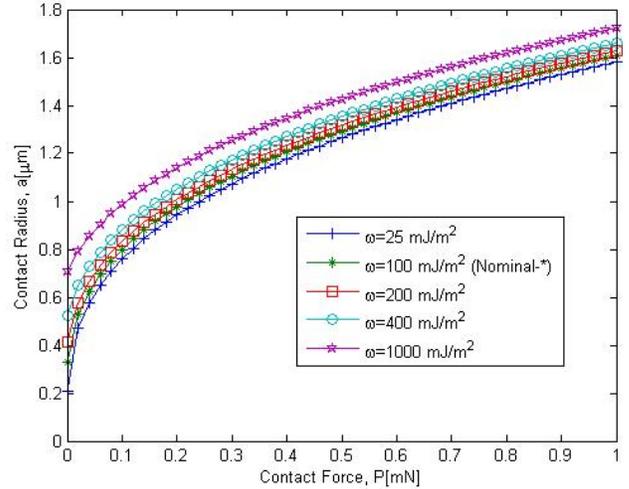


Fig. 7. Contact radius between  $5 \text{ }\mu\text{m}$  radii spherical patterns on the stamp and the substrate as a function of contact force when adhesion energy between surfaces are varied from  $25\text{--}1000 \text{ mJ/m}^2$

ensure a uniform coating of a polar polymer material. After some trial and error printing on different materials like glass and alumina, Petri dishes found to be a very good substrate for microcontact printing.

2) *Experiments:* The printing step can be realized both manually or automatically. Using a force-controlled automatic system (Fig. 8) [13] it is possible to control the time, force and also velocity of the approach for the printing phase. Here, a ten gram load cell with  $0.1 \text{ mN}$  force resolution is used. It is a self-aligning system for repeatable and robust microcontact printing.



Fig. 8. Photo of the force-controlled system setup and its components

In this setup, either the stamp or the substrate can be stationary and the other is moving. The stationary part is fixed

on the face with a spherical joint for self-alignment. The force-controlled system is programmable and contact forces can be recorded by the load cell. The stage is automatically retracted after the contact force reaches the required force for printing.

### III. RESULTS AND DISCUSSIONS

Using a force-controlled system, microcontact printing of the 'C' pattern is conducted and examined on the substrate. Five micron polystyrene particles are initially assembled to form a 'C' pattern shown in Fig. 9. Three micron layer of aluminum is sputtered and particles are removed to form a template. A thick-backing PDMS stamp is molded and used for stamping on a Petri dish substrate. After inking with GFP, the pattern is printed using different contact forces and observed under the fluorescent microscope. According to the effective contact radius, the imprints can be similar or smaller than the microparticle size (Fig. 10). Contact radius of the spherical patterns on the stamp is increased for higher contact forces as expected and predicted from the theory.

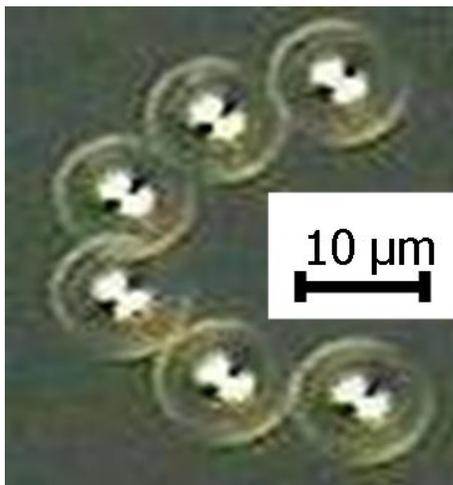


Fig. 9. Microrobotic assembly of  $5 \mu\text{m}$  radii particles to form a 'C' pattern

Final imprints might have some defects due to inking and printing imperfections. Inking defects occur when the proteins are not formed a uniform layer on the spherical pattern tips or are either dried or washed out after suction of the inking polymer. Alignment and levelling of the stamp is very important which makes a big difference in the micro/nano-contact printing of proteins. Like in most conventional contact print applications, it would be much simpler to print larger micro patterns with high aspect ratio stamps.

The resolution of the force-controlled system is limited to  $0.1 \text{ mN}$  and applying microforces for sub- $100\text{nm}$  nanoprinting is not feasible with the current setup. AFM as friction force microscopy (FFM) imaging mode can be used to image patterns on the substrate after printing. Since the surface friction changes on the spots with the printed proteins, lateral friction force and voltage are increased and the printed pattern can be verified in nanometer precision.

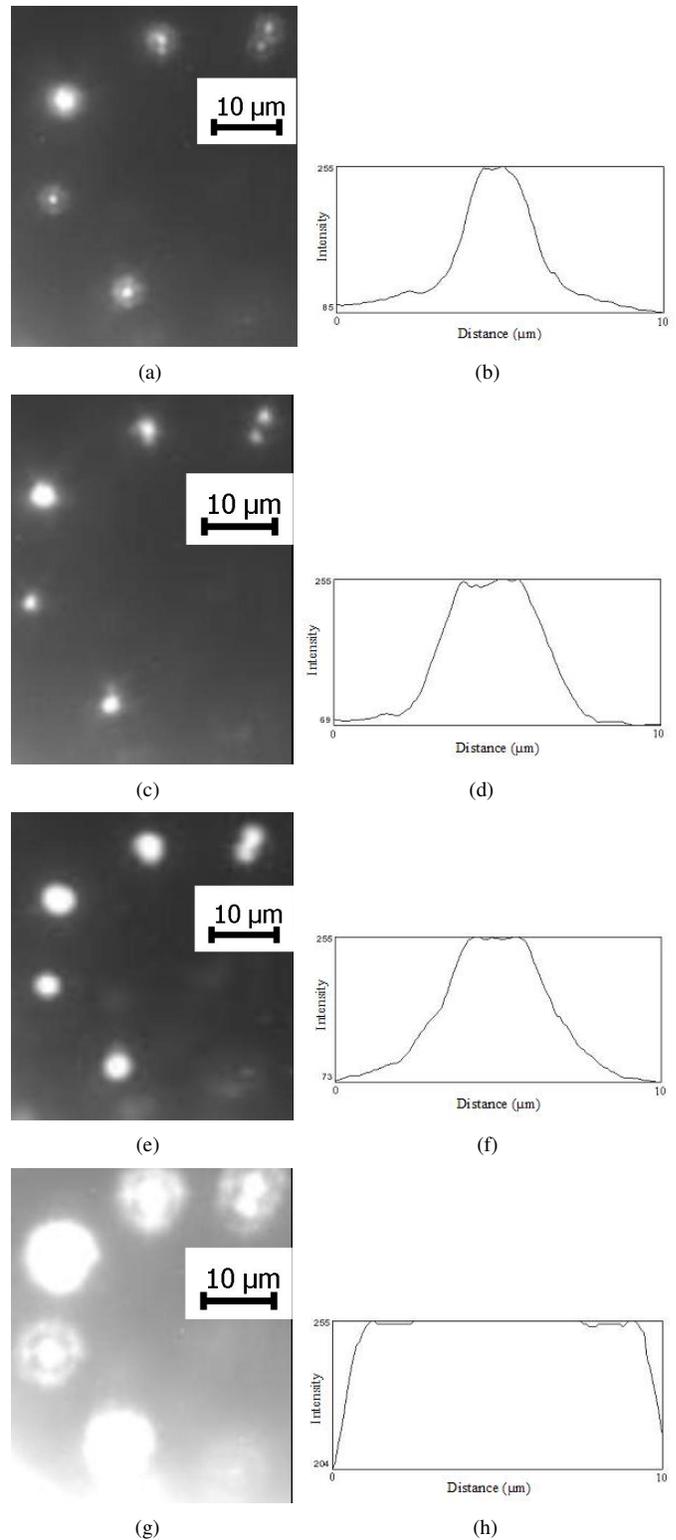


Fig. 10. Force-controlled microcontact printing of the 'C' pattern on the Petri dish substrate by increasing contact force; right diagrams are 2D fluorescent intensities

#### IV. CONCLUSION

In this study, microrobotic assembly based templates are used for microcontact printing of fluorescent proteins. It is shown how to create patterns of proteins having a length scale smaller than the assembled micro-particles using controlled-force microcontact printing. The spherical geometry of the microparticles contributes to reduce the printed areas below the effective size of the molded features. The transfer of the proteins from the stamp to the substrate surface occurs during the printing step.

The advantage of our microcontact printing method is high volume template fabrication using automatic microrobotic assembly that helps to get smaller imprints from micro features using force control. Further research goals are using harder polymers for stamping in order to reduce the chance of pattern collapse, improving the inking process to form a very uniform protein monolayer, advancing force-controlled setup for precise force control, and verifying and characterizing imprinted patterns on the substrate using frictional force imaging in an Atomic Force Microscope.

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