

# ELECTROSTAMPING THROUGH SAM LAYER FOR 1:1 TRANSFER OF 40-NM-WIDE PATTERNS OVER MM<sup>2</sup> AREA

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## ABSTRACT

The paper reports a novel approach to improve transfer resolution and transfer uniformity of electrostamping, based on thin-film edge electrode lithography (TEEL). TEEL transfers nanopatterns collectively corresponding to thin-film edge electrodes on a mold according to an electrochemical reaction in the water meniscus formed between the mold and the substrate. High relative humidity can ensure the meniscus forms stably for uniform transfer, while resolution can be degraded due to meniscus extension. A hydrophobic SAM layer is used for the first time on the contact surfaces of the electrostamping mold to control meniscus size and thereby to achieve high-resolution; 1:1 transfer of 40-nm-wide patterns over several mm<sup>2</sup> area was achieved.

## INTRODUCTION

Nanoimprint lithography (NIL) is a promising candidate for next generation lithography due to its inherent simplicity and low cost of operation [1]-[5]. However, conventional NIL suffers from problems such as resist adhesion during demolding and non-uniformity of residual film between the mold and the substrate (Figure 1a). As a solution, a resistless electrochemical NIL called nanoelectrode lithography (Figure 1b) is proposed [6]. Nanoelectrode lithography is a method of pattern transfer that uses a conductive mold to generate oxide patterns on a target substrate by an electrochemical reaction (ER) in the water meniscus formed between them. Oxide patterns with a line width ranging from microscale to nanoscale can be transferred directly on the surface of a semiconductor or metal layer via the ER between conductive patterns of the mold and the target material. Compared with conventional NIL, nanoelectrode lithography is not only able to reduce the number of process steps and thus fabrication cost, but also to avoid pattern defects, thereby improving accuracy because of the resistless process [7]-[9]. However, both NIL methods are 1:1 transfer of convex patterns of mold. Therefore the resolution of transfer is limited by the resolution of conventional nano fabrication. Therefore, a fine mold is needed for high transfer resolution, resulting in increasing of mold fabrication cost.

To realize high resolution while maintaining the advantage of resistless process, we proposed a novel electrostamping method that utilizes thin-film edge electrodes (TEEs) to transfer nanopatterns in our previous work [10]-[11]. As shown in Figure 1c, thin-film electrodes were made on the side walls of protrusions of a NIL mold to generate oxide patterns on a target substrate by the electrochemical reaction (ER) in the water meniscus formed between the edge of electrodes and the substrate. Compared to conventional NIL, thin-film edge electrode

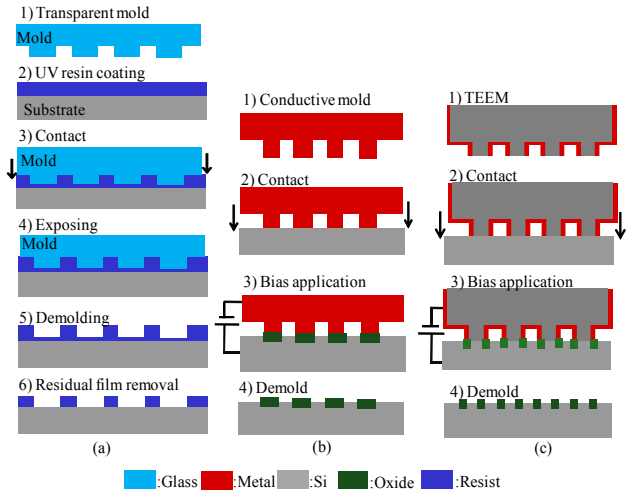


Figure 1: (a) Process chart of conventional UV-type NIL. (b) Process chart of mold-based TEEL.

lithography (TEEL) is able to reduce the number of process steps thus fabrication cost as low as the nanoelectrode lithography. On the other hand, the resolution of TEEL is only dependent on the lateral thickness of TEEs but not on convex size of mold, which can be easily thinned for higher resolution. In comparison with nanoelectrode lithography, TEEL can realize high resolution beyond conventional lithography at a low mold cost.

Our goal is to establish a mold-based TEEL as a candidate of next generation NIL technology for mass production. Therefore, TEEL are required to have high resolution, high throughput and high uniformity. Collective transfer of nanopatterns in high throughput has been demonstrated using mold-based TEEL [10]-[11]. However, the trade-off between uniformity and resolution over larger area becomes a problem, because the transfer resolution is heavily influenced by meniscus size in addition to the thickness of TEEs. Meniscus extension arose at high relative humidity (RH) helps uniform electrical contact between hard surfaces over large area but the ER occurs in wider portion than TEEs, resulting in resolution degradation.

## CONCEPT AND PRINCIPLE

In this study, a hydrophobic SAM layer is used for the first time on the contact surfaces of thin-film edge electrode mode (TEEM) and the target substrate to control meniscus size and thereby to achieve high-resolution even under an atmospheric condition of high relative humidity.

The structure of TEEM and the principle of transfer when a SAM layer is introduced to both the surface of the mold and the substrate are shown in Figure 2. The proposed TEEM is composed of four parts, insulating

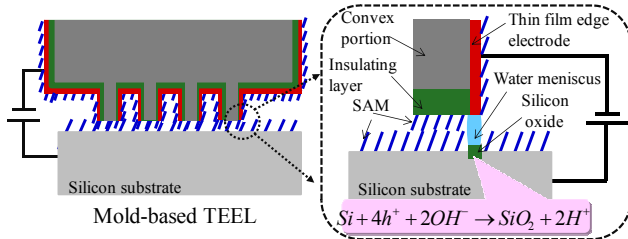


Figure 2: Concept schematic of controlling water meniscus in thin film edge electrode lithography (TEEL)

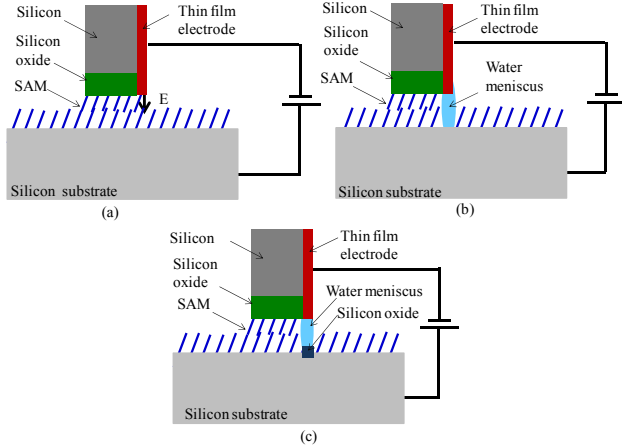


Figure 3: Principle Transfer of TEEL with SAM layer

stamping structures, nanometric-thick single-layer TEEs formed on the sidewall of the stamping structures, a base, and a SAM layer coated on the surface of those portions. Silicon oxide is formed on the surface of the silicon-made convex structures to act as insulating layer to avoid electrical convex-portion transfer of TEEM. The TEEs are mechanically and electrically connected to the base via the insulating patterns on the base. The TEEs enables not only to generate nano-patterns smaller than the convex patterns on TEEM via an ER but it also enables collective transfer and thus high throughput. On the other hand, application of the hydrophobic SAM layer to both the surface of TEEM and the target substrate not only can prevent meniscus extension for realization of high resolution, but also can improve contact uniformity for high transfer uniformity.

As shown in Figure 3, SAM layers can be decomposed on both surfaces when a voltage is applied between TEEs and the substrate, in which these surfaces become hydrophobic from hydrophilic and confine the meniscus for localized ER to generate oxide patterns. Therefore, meniscus extension between the TEEM and the substrate can be prevented even at high RH because other surfaces remain hydrophobic, enabling high resolution.

The SAM layer is required to be highly hydrophobic, flexible and electrically decomposable. Here, HMDS is used as the SAM layer because it meets the above-mentioned requirements. In addition, Ru is adopted as the TEEs material for its good electrical conductivity even after being oxidized.

## FABRICATION OF TEEM

The fabrication process flow of the TEEM before the application of the SAM layer is shown in Figure 4. The process began with a p-type (100)-oriented Si substrate

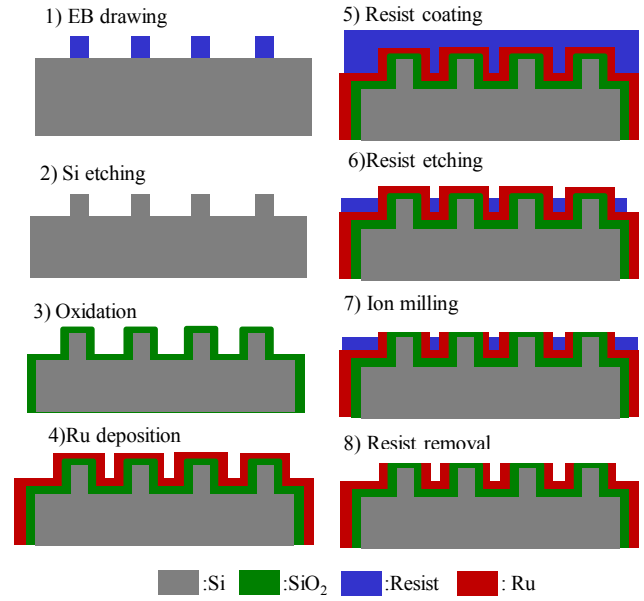


Figure 4: Process flow of TEEM without SAM layer

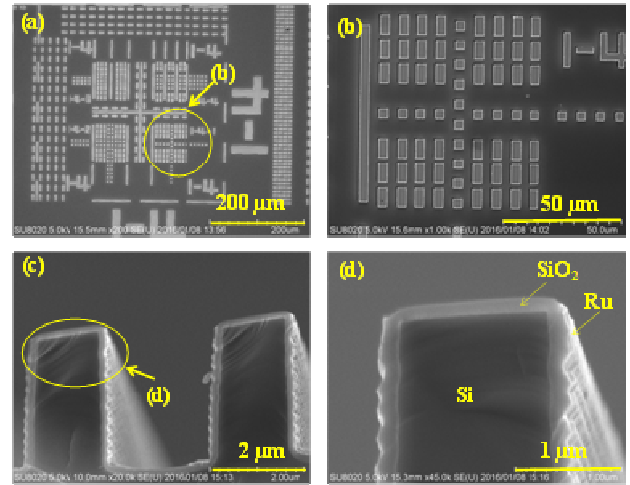


Figure 5: (a) SEM image of the fabricated thin film edge electrode mold (TEEM); (b) Close-up SEM image of the fabricated TEEM in (a); (c) Sectional image of the fabricated TEEM; (d) Close-up sectional image of the fabricated TEEM in (c)

with a thickness of 525  $\mu\text{m}$  and an electrical resistivity of 1-10  $\Omega\cdot\text{cm}$ . First, electron-beam lithography process was conducted to form a photoresist pattern on Si substrate for shaping convex patterns of TEEM. Then, these photoresist patterns were transferred on the silicon substrate by deep reactive ion etching process. After that, a 160 nm-thick silicon oxide film was formed as an insulating layer in a thermal oxidation process. Then, a 40 nm-thick Ru film was deposited through an electron-beam evaporation process for creating the TEEs. Next, photoresist was filled on the stamping structure for protecting the Ru film on the recess portion of the TEEM. After that, photoresist was removed partly to expose the top surface of stamping structure. Then, an ion-milling process with high verticality was conducted to etching the Ru film on the top surface of the TEEM to form the TEEs. At last, the residual photoresist was removed, and the fabrication of TEEM was finished.

The fabricated TEEM shown in Figure 5 had a pattern area size of 4 mm×4 mm, a 40 nm-thick TEEs and a 3 μm-height convex patterns with a pitch of 4 μm.

## TEEL

The setup for performing TEEL shown in Figure 6 was composed of a pressing unit, an electrical unit and a temperature controlling unit. The pressing unit enabled application of force ranging from 0 to 500 N, and the electrical unit made it possible to apply a bias voltage with arbitrary waveform. The temperature unit was able to adjust relative humidity (RH) between the mold and the substrate from 0% to 100% by adjusting temperature of the sample stage.

The electrostamping process of TEEL consists of 6 steps: 1)set the TEEM and Si substrate on the sample stage of pressing unit; 2)adjusted the stage temperature for target RH; 3)applied a force to ensure the contact between the TEEM and the substrate; 4)maintained the contact condition for several minutes to make sure the mold and substrate are at the same temperature as the sample stage; 5)applied a bias voltage between the TEEM and the substrate to induce electrochemical reaction for generating oxide pattern corresponding to TEEs; 6)released the TEEM and the substrate from sample stage by unloading the force.

A p-type (100) oriented Si substrate with an electrical resistivity of 5-10 Ωcm was used for transfer. A HMDS layer was evaporated on the surface of both TEEM and the substrate at a temperature of 150°C for 5 min as the SAM layer before conducting TEEL.

To confirm the meniscus control effect of SAM, TEEL was conducted at a high RH of 80% when SAM film was introduced to both the surface of TEEM and the Si substrate. A100N force and 17V bias voltage was applied between the TEEM and Si substrate in air with a high RH of 80% and a temperature of 10.1 °C. To prevent the target surface from Joule heating, the bias voltage with a pulse duration of 1 s and a separation period of 1 s was used, and the total time of bias application was 30s. As shown in Figure 7, oxide patterns of 40 nm in width were collectively transferred in an area over several mm<sup>2</sup>. These results indicated that 1:1 transfer of 40 nm-wide nanopatterns was possible even under a high RH of 80%. Therefore, the application of SAM layer to both the surface of the mold and the substrate made it possible to realize of high resolution and uniform transfer in large area required for mass production by performing TEEL at high RH.

As a comparison, TEEL when the SAM layer was only introduced to the surface of TEEM (Figure 8a) was performed under the same condition. From the results shown in Figure 8 (c)-(d), it can be seen that line-width of transferred pattern was extended to 140 nm from 40 nm (Figure 7-8). This result showed that control of meniscus extension only on the TEEM side was not enough for high transfer resolution at a high RH of 80%.

Furthermore, TEEL was also conducted under the above-mentioned conditions except no introduction of SAM layer (Figure 9a) and the results were shown in Figure 9 (b)-(c). These results showed that the line-width of transferred pattern was about 10 μm, which was

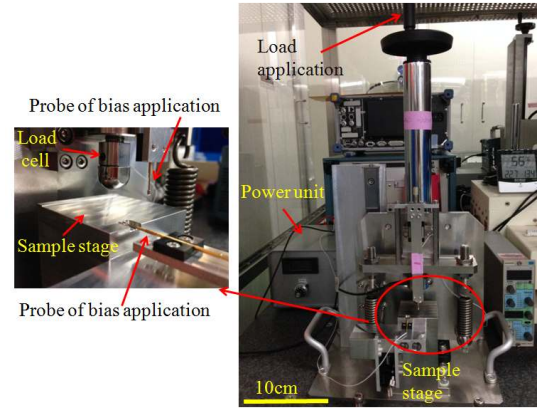


Figure 6: Picture of the setup for performing TEEL

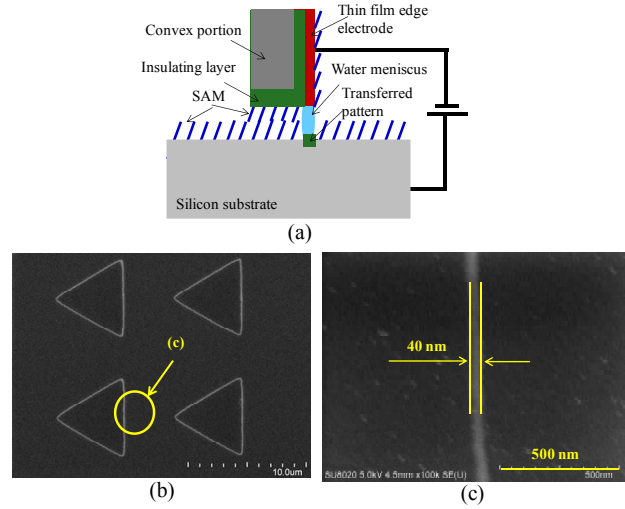


Figure 7: (a) Schematic illustration of TEEL when SAM layer is introduced to both the surface of the mold and the substrate; (b) SEM image of transferred oxide patterns using 40 nm-thick TEEs when SAM layer is introduced to both the surface of the mold and the substrate; (c) Close-up of the transferred pattern in (b).

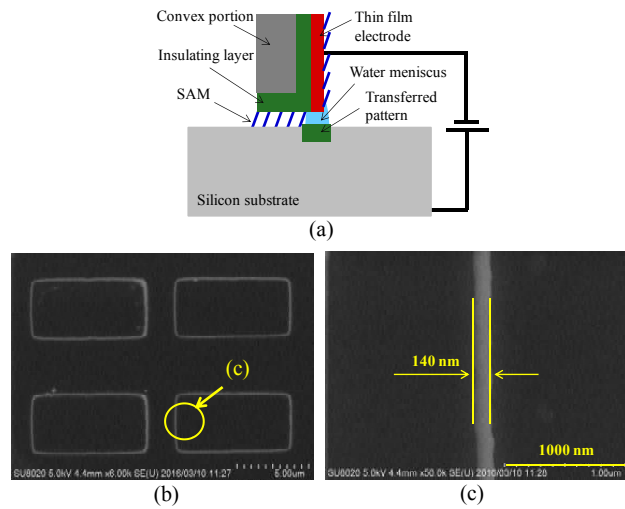


Figure 8: (a) Schematic illustration of TEEL when the SAM layer was only introduced to the mold; (b) SEM image of transferred oxide pattern using 40 nm-thick TEEs when the SAM layer was introduced to the mold only; (c) Close-up of the transferred oxide pattern in (b).



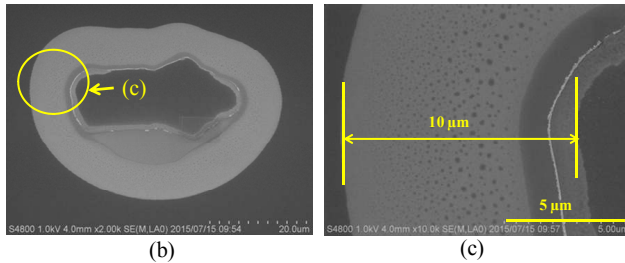
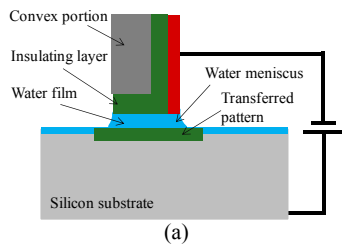


Figure 9: (a) Schematic illustration of TEEL without application of SAM; (b) SEM image of transferred oxide pattern using 40 nm-thick TEEs without application of SAM; (c) Close-up of the transferred oxide pattern in (b).

considerably larger than the thickness of TEEs. This is because the meniscus under the mold and the water film on the substrate surface were connected to each other at RH of 80%, resulting in drastic degradation of resolution.

From these results, it can be seen that it is important to control meniscus extension both on the side of TEEM and the substrate for high resolution even under an atmospheric condition of high RH.

In addition, the SAM layer on the surface of the convex portion also may be destroyed by the bias voltage when performing TEEL. Continuous use of TEEM may cause degradation of transcription resolution due to SAM layer deterioration on the convex portion of TEEM. Therefore, it is better to remove the SAM layer after each TEEL and deposit again before next TEEL to ensure the effect of SAM layer.

## CONCLUSION

We aimed to develop a new-type-resistless NIL to realize transfer resolution beyond conventional lithography in high throughput and low cost so that this technique would be available for industrial use. In order to solve the trade-off between uniformity and resolution of TEEL, a hydrophobic SAM layer was introduced to both the mold and the substrate for preventing meniscus extension. As a result, a 1:1 transfer of 40 nm-wide TEEs was succeeded even at a high RH of 80% over several mm<sup>2</sup> area. These result indicated that 1:1 transfer of TEEs was possible even at high RH, showing the realization possibility of high resolution and uniform transfer in large area required for mass production.

We are currently reducing the thickness of TEEs for high resolution and improving the TEEM and the transfer equipment for uniform transfer of patterns in large scale.

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