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Electron beam direct writing of nanodot patterns on roll mold surfaces by electron beam on–off chopping control

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ABSTRACT

Roll-to-roll (RTR) technology is a high-throughput production method for nanoimprint lithography (NIL). However, the fabrication of a roll mold for RTR-NIL is difficult because of the cylindrical shape of the mold. We previously developed a technique for direct writing with an electron beam (EB) on a rotating cylindrical substrate that permits the production of seamless molds. However, the only patterns that could be written by this technique consisted of lines and spaces. Because nanodot patterns would be very useful for producing novel devices, we developed a direct EB method for writing patterns of nanodots onto the surface of a rotating roll mold by EB on–off chopping control by a beam-blanking system. However, because the EB resist is a soft polymer material, it has insufficient toughness for use in RTR-NIL. Therefore, after the development of the EB resist on the roll mold surface, a pattern-transfer process, such as dry etching or metal deposition and lift-off, is necessary. We chose the metal lift-off process, and we developed a technique for vacuum evaporation with a rotating cylindrical substrate, which we used to produce metal roll molds with various nanodot patterns.

1. Introduction

In the next generation of manufacturing techniques, there is a marked need for a nanoscale patterning method that has high throughput and high resolution, and which is also cost-effective. Roll-to-roll nanoimprint lithography (RTR-NIL) [1–3] has received a great deal of attention as a technique for the fabrication of devices of the next generation, including solar cells [4], transparent conductive sheets [5], and wire grid polarizers [6–8], among others [9].

We have previously developed a technique for direct writing with an electron beam (EB) on a rotating cylindrical substrate to produce seamless nanoscale molds [3,10–12]. Recently, an EB writer with a stencil mask has been used to achieve high-throughput writing [13].

Nanoscale patterns of dots are now very useful for novel devices, such as plasmonic devices and antireflection structures [14,15]. However, no methods for fabricating nanodot patterns by direct EB writing have been established. We have therefore developed a method for writing patterns of nanodots onto the surfaces of roll molds, and we examined the effects of the speed of rotation of the mold, the EB current, and the EB dose.

Because they are made of soft polymer materials, EB resists are insufficiently tough for use in RTR-NIL. Therefore, after the development of EB resist, it is necessary to use a pattern-transfer process such as dry etching or a metal deposition and lift-off process. In the present case, dry etching of roll substrate is difficult because it would be necessary to rotate the substrate in a plasma atmosphere. We therefore chose to use a metal lift-off process in which the roll mold is mounted on a rotating stage in a conventional evaporation system, and we examined the deposition characteristics of this system. Our reason for choosing vacuum evaporation to apply the metal in the lift-off process is that this is a dry process. An electroforming process would also be a candidate as a suitable process, and might have considerable potential in NIL [16]; however, electroforming processes are wet processes, so sometimes the resulting patterned EB resists suffers from problems associated with dissolution or expansion. Dissolution of the EB resist results in a loss of pattern, and expansion causes unintended changes in the pattern size. The vacuum evaporation system, on the other hand, is not a wet process and is therefore free from such problems of dissolution or expansion. We believe that with careful selection of the solution, the electroforming process, and the EB resist material, electroforming could be used to fabricate lift-off patterns, but in this study, we chose the vacuum evaporation system.

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2. Experimental apparatus and procedures

Fig. 1 illustrates the process of EB direct writing with a rotating roll mold. A 34-mm-long aluminum cylinder with an outer diameter of 34 mm was used as the substrate for the roll mold. ZEP520A (Zeon Corp., Tokyo), a positive EB resist with a dedicated ZED-N50 developer, was used as the EB resist [17]. To obtain a thin coating, ZEP520A was diluted to 50% by adding ZEP-A thinner. The roll mold substrate was dipped in the diluted ZEP520A resist at a speed of 3 mm/s, kept in the solution for 5 s, and then pulled out at a speed of 0.2 mm/s [Fig. 1(1)]. Next, the sample was baked at 180 °C for 20 min [Fig. 1(2)]. The resulting ZEP520A film had a thickness of approximately 120 nm.

Next, we mounted the roll substrate on rotating equipment and we installed this in the EB writing system. A scanning electron microscope (ERA-8800FE; Elionix Inc., Tokyo) with a blanking system was used for EB writing [Fig. 1(3)].

To fabricate the nanodot pattern, the roll mold was rotated in the circumferential direction and, simultaneously, the EB was switched on and off (chopping) by controlling the blanking signal. The EB was focused at a single point on the surface of the roll mold and the nanodot pattern was produced by revolving the roll while the EB was switched on and off. In this study, one EB drawing field was equal to one circumference on the roll mold. To prevent overwriting of the nanodot pattern, for example, by duplicate EB writing, the chopping signal was switched off after one EB drawing field. The total chopping time was therefore equal to that required by the roll mold to complete one revolution. We therefore needed to calculate the relationship between the rotation speed, the chopping speed, and the EB dose.

In this study, the fixed EB writing conditions were as follows: acceleration voltage: 10 kV, rotation speed: 0.5 rpm. The speed of 0.5 rpm is the optimum value that provides a sufficient EB dose while retaining a fine dot pattern (unit drawing length). The blanking system was capable of dividing each EB drawing field into 520,000 dots.

The minimum size of each dot was therefore 206 nm (34 mm × π/520000 = 206 nm). This 206 nm value is the unit drawing length. The drawing pattern consisted of patterns representing 0 and 1 (0 = beam off, 1 = beam on), and the maximum length of each sequence was 520,000 characters. When the EB had completed writing one circumference on the roll mold, it was moved arbitrarily in the longitudinal direction by the EB blanking system to permit drawing over the whole surface of the roll.

In our setup, EB drawing required 120 s per revolution, because the rotational speed was 0.5 rpm. The EB drawing time per dot (the unit drawing time) was therefore 230 μs (120 s/520,000).

The EB dose can be calculated by the following expression:

$$\text{EB dose} = (\text{EB current} \times \text{unit drawing time})/(\text{beam diameter} \times \text{unit drawing length})$$

The beam diameter was about 20 nm. Therefore, if the EB current was 50 pA, the EB dose was:

$$(50\ \text{pA} \times 230\ \mu\text{s})/(20\ \text{nm} \times 206\ \text{nm}) = 280\ \mu\text{C/cm}^2$$

First, we examined the effect of controlling the EB dose on the properties of the nanodot pattern. The conditions for producing nanodots patterns by direct EB writing in this experiment were as follows: EB current: 50, 60, 80, 100, or 120 pA; EB dose: 280, 336, 448, 560, or 672 $\mu C/cm^2$.

Next, we examined the effects of changing the unit drawing length on the properties of the nanodot pattern. For example, a single ‘1’ design and the appropriate space sequence gave a 206-nm dot size and space pattern. A double ‘1’ character, that is a ‘11’ sequence, produced a 412-nm dot size. Furthermore, the single ‘1’ design also determines the unit drawing time (230 μs). In other words, the ‘1’ design determines both the unit drawing length and the unit drawing time.

The conditions necessary to produce a nanodot pattern by direct EB writing were as follows: EB current: 60 pA; EB dose: 336 $\mu C/cm^2$.

After the EB direct writing process, the ZEP520A layer was developed by treatment with ZED-N50 at 20 °C for 1 min [Fig. 1(4)]. At this stage, the nanodot resist pattern on roll substrate had been fabricated [Fig. 1(5)].

Next, we deposited a layer of chromium on the aluminum roll substrate by vacuum evaporation (VPC-260F; Ulvac Kiko, Inc., Kanagawa), as shown in Fig. 2. The speed of revolution of the deposition stage was 6 rpm and the evaporation time was 4 min [Fig. 2(a)].

Subsequently, the ZEP520A layer was removed by using the appropriate remover (ZEDMAC) to leave metal patterns on the roll

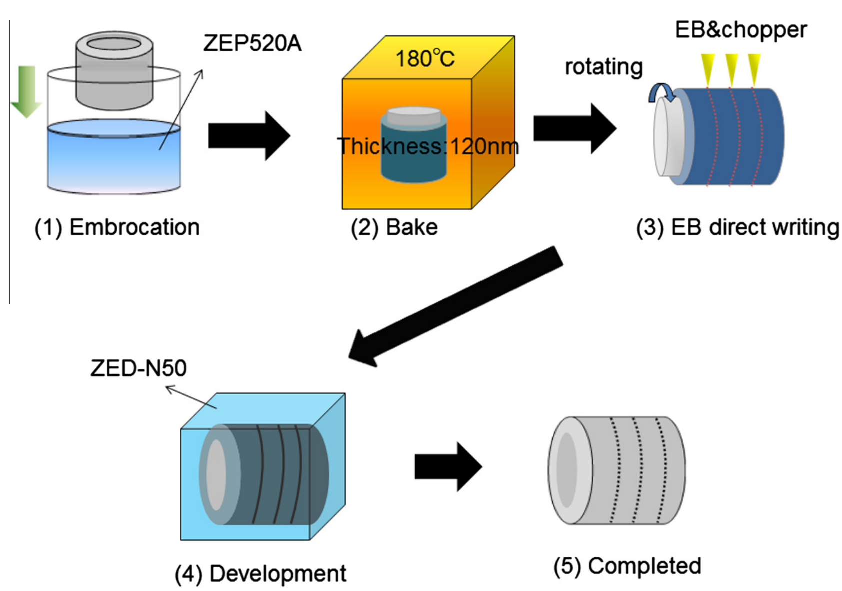

Fig. 1. Process for fabrication of the roll mold by EB direct writing.