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Fabrication of nano-sized resist patterns on flexible plastic film using thermal curing nano-imprint lithography

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Abstract

Due to polymer's excellent flexibility, transparency, reliability and light weight, it is a good candidate material for substrate of devices including organic electronic devices, biomedical devices, and flexible displays (LCD and OLED). In order to build such devices on polymer, nano- to micron-sized patterning must be accomplished. Since polymer materials reacts with organic solvents or developer solutions which are inevitably used in photolithography and cannot bear high temperature (~140 °C) process for photoresist baking, conventional photolithography cannot be used to polymer substrate. In this research, monomer based thermal curing imprinting lithography was used to make as small as 100 nm dense line and space patterns on flexible PET (polyethylene-terephthalate) film. Compared to hot embossing lithography, monomer based thermal curing imprint lithography uses monomer based imprint resin which consists of base monomer and thermal initiator. Since it is liquid phase at room temperature and polymerization can be initiated at 85 °C, which is much lower than glass temperature of polymer resin, the pattern transfer can be done at much lower temperature and pressure. Hence, patterns as small as 100 nm were successfully fabricated on flexible PET film substrate by monomer based thermal curing imprinting lithography at 85 °C and 5 atm without any noticeable degradation of PET substrate.

Keywords: Flexible substrate; Transparent polymer film; PET (polyethylene-terephthalate) film; Hot embossing nano-imprinting lithography; Monomer based imprinting lithography; Benzyl-methacrylate monomer; Thermal initiator; Polymerization

1. Introduction

Due to polymer's excellent flexibility, transparency, reliability and light weight, it is a good candidate material for substrate of devices [1] including all polymer electronic devices, biomedical devices, chip for wearable PC, and flexible (roll-up) displays (LCD and OLED). In order to build devices on polymer substrate, a patterning technology [2,3], which enables to fabricate patterns as small as a few tens of nanometers to a few microns on a large area must be developed.

Reliable, low cost and high throughput lithography process is key step for successful mass production of integrated circuits. Using conventional deep UV photolithography, sub 100 nm size patterning can be done. However, such a photolithography technique becomes more complicated, its throughput is being decreased, and it requires more costly fabrication equipment. Patterning by photolithography can only be done on rigid semiconductor wafers. Thus, new patterning technology, which can make nano-sized patterns economically and can handle non-wafer-type substrates such as flexible polymer films, needs to be developed.

Significant amount of effort is being placed in developing alternative patterning technology for nano-sized patterns. In case of extreme UV photolithography [4] and immersion lithography [5,6], it is being developed to make sub 70 nm patterns on Si wafer substrate for microelectronic devices and has to use very costly equipment. Contrary to photolithography, nano-imprinting lithography

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[7–10] can fabricate the same or smaller sized patterns with high throughput and lower cost. Furthermore, this process does not require precise focusing, non-planar and flexible substrate can be used.

In case of hot embossing imprint lithography, sub 100 nm patterns can be made on a large substrate, however, the process often requires the imprinting step at temperature as high as 180 °C and pressure as high as 60 atm. Thus, flexible polymer films cannot be used as the substrate. Recently developed monomer based imprinting lithography [11,12] can produce the patterns at much lower pressure and temperature.

Monomer based thermal curing imprint lithography is similar to hot embossing imprint lithography. While the hot embossing process uses the thermoplastic polymer resin, such as PMMA (polymethylmethacrylate), monomer based thermal curing imprint lithography uses a mixture of pre-polymer (monomer) and thermal radical generator, which is triggered by heating and initiate the polymerization reaction. Since the imprinting resin for thermal curing imprint lithography is in liquid phase at room temperature and thermal curing of pre-polymer is initiated at much lower temperature than the glass transition temperature of thermoplastic polymers, imprinting temperature and pressure of thermal curing imprint is much lower than those of hot embossing imprint lithography. Thus, 100 nm sized pattern fabrication can be done on flexible polymer substrate by nano-imprinting lithography.

2. Experiment

Si templates used in this study were made using conventional photolithography and reactive ion etching. A photoresist pattern was generated on a Si by conventional DUV lithography and then SI substrate was etched by reactive ion etching with fluorocarbon gas. Then, ashing of remaining photoresist is done by oxygen plasma. The surface of Si template was coated with the monolayer of releasing material (heptadecafluoro-1,1,2,2-tetra-hydrodecyl) trichlorosilane [11,12] to prevent stiction between template and resin after imprinting. Almost all the surface area was covered with surface protrusions, including as small as 100 nm line and space patterns. Cross-sectional SEM micrograph of Si template, which has the dense array of 100 nm lines and spaces, is shown in Fig. 1. A 125 μm thin, flexible and transparent PET (polyethylene-terephthalate) film, coated with ITO (indium tin oxide) film was used as the substrate.

Schematic diagram of an imprinting system used in this study is shown in Fig. 2. For the delivery of uniform pressing force over large area substrate, imprinting system [11,13,14] is based on pressurized chamber. This design is also effective to handle flexible polymer substrate. A stack of Si template and PET film coated with a resin is placed within the loading chamber, consisting of heated copper plate and elastomer sheet. Then the loading chamber is pumped to vacuum in order to remove any air bubble,

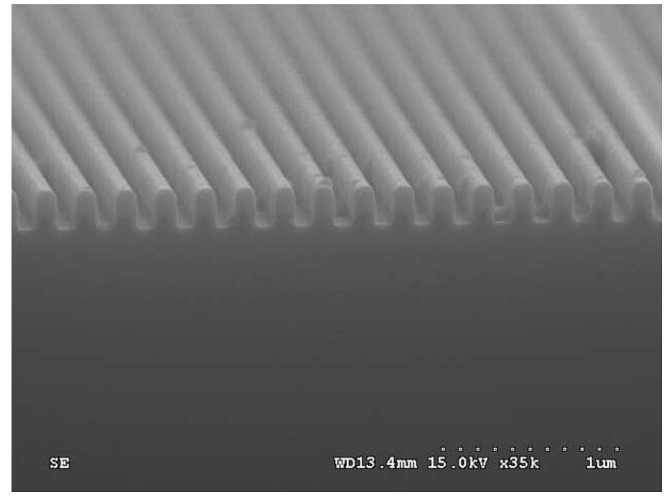

Fig. 1. Cross-sectional SEM micrograph of Si template, which has the dense array of 100 nm lines and spaces.

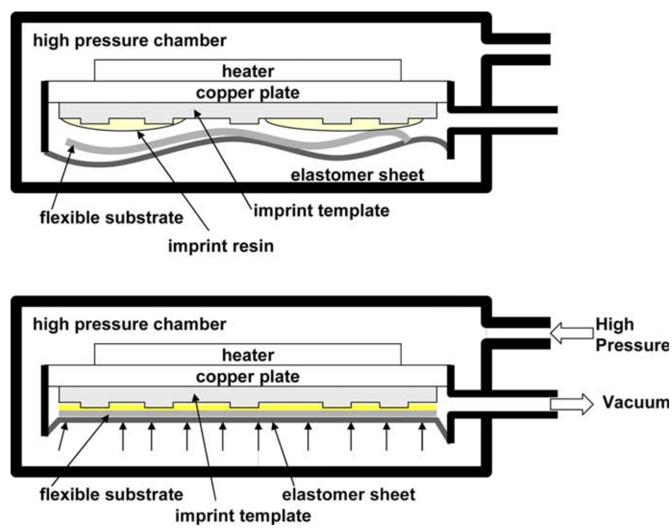

Fig. 2. Schematic diagram of an imprinting system. For the delivery of uniform pressing force over large area substrate, imprinting system is based on pressurized chamber.

trapped between template and PET film. A flexible PET film is also straightened up at this stage. Imprinting is done by applying high pressure into main chamber, combined with pumping of load chamber to vacuum. During imprinting, the resin becomes a thin layer by squeezing out any extra resin. Then the copper plate is heated and the polymerization of monomer resin is initiated. After reaching the setting temperature (85 °C at elastomer surface), a stack of template and substrate is held for 5 min before heating is off. The imprinting pressure was maintained until the substrate temperature reached 50 °C. Since the pressing force is uniformly applied through elastomer side from the isotropic pressure of main chamber, the deliver of the pressing force is very effective and highly uniform.

A thermally curable monomer based resin contains 90% of liquid phase monomer (benzyl-methacrylate, $C_{11}H_{12}O_2$), 8% of vaporization inhibitor (its polymer, poly benzyl-