

Development of nanostructured foil mold for roller nano imprint process

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Abstract:

Roller nano imprint (RNI) process is expected as a key technology that realizes a cheap and high through-put nano fabrication process. RNI is a kind of a rolling process that replicates nano structures of a mold to a work material. Development of nano structure mold is therefore essential for development of RNI technology. In this paper, development of a nano-structure foil mold made of TiN and Ni is reported. The foil mold is applied for the RNI experiments. It is shown that RNI modifies optical reflectance of work material. Application of RNI to nano dot array fabrication is studied also.

Keywords: Nano manufacturing, Roller imprint, Nano structured mold, Sputter coating

1. Introduction

Patterned nano/micro structures have various applications in many engineering areas, such as bioengineering, opto-electrical engineering, material engineering and nano-mechanical engineering [1]. They have capability to address environmental problems, energy problems, and medical problems. Especially, patterned nano structures on the surface of a material induce various surface functions [2, 3, 4, 5]. It is known that a submicron pillar array fabricated on the surface of optical materials reduces reflection of light [3, 4]. A metallic line array on a transparent material causes polarization [5]. Furthermore, nano-dot array induces localized Surface Plasmon Resonance, which are useful for optical sensors.

Such nano/micro structures are usually fabricated by Photo Lithography (PL) or Electron Beam Lithography (EBL). These lithography methods however are disadvantageous in terms of cost of facility, stringency in method and through-put. In order to overcome these disadvantages, challenges to develop cheap nano fabrication technologies are desired. In these challenges, process should be simple and robust as principle, usage of expensive equipments and hazardous chemicals should be minimized, and at the same time, quality of product and throughput should be superior to the conventional lithography methods, which the authors call “Economical Nano-Technology”

Roller nano imprint (RNI) process is a promising technology to achieve the Economical Nano-Technology. RNI process is a kind of rolling technology that replicates nano structures of a mold to a work material. Development of a nano structure mold is therefore essential for development of RNI technology. In this paper, a nano-structure foil mold made of TiN and Ni is developed. Also, the foil molds are applied for RNI experiments. Through these experiments, possibility of RNI is studied.

2. Experimental method and setup

Figure 1 illustrates procedure of the RNI process

examined in this paper [6]. Firstly, Nano Plastic Forming (NPF) is used to fabricate a master mold. Secondly, the master mold is coated with hard materials. The coated layer is then peeled off to acquire a foil mold. Thirdly, the foil mold is used in RNI to transfer the patterns onto the surface of rolling work materials. It is noted that EB lithography is not utilized in this process. NPF is capable of fabricating nano/micro structures directly on a work material by plastic forming, and its facility cost is much cheaper than that of EB lithography.

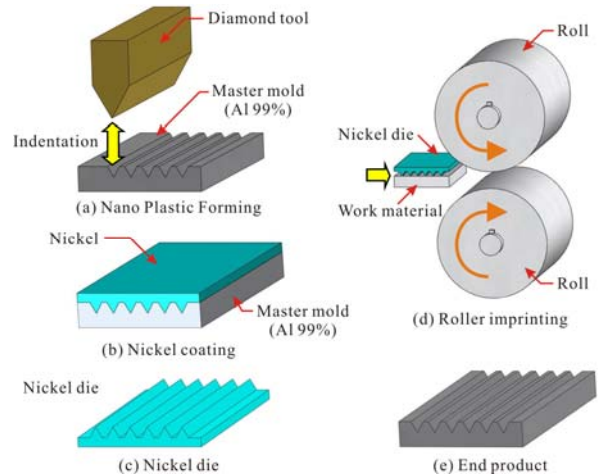


Figure 1: RNI process examined in this project.

Figure 2 shows the NPF device. It consists of high precision X-Y stages, a Z stage, linear motion controllers, a control computer, a load cell, a diamond tool and data acquisition system. The computer and the linear motion controller control the XY-stages and the Z-stage. Resolution of those stages is 10nm. A load cell is mounted on the Z-stage for measurement of the forming load. A knife edge tool made of a single crystal diamond is used in the experiment. Its edge angle is 60degrees, and its width is 0.6mm. The tool edge is polished very sharp, and its edge radius is smaller than 50nm.

Pure aluminum and soda glass are used as the master mold material. Size of a pure aluminum master mold is

15mm x 15mm x 1mm, and that of a soda glass master mold is 25mm x 30mm x 1mm. Parallel grooves are fabricated on the surface of the master mold plate by indenting the knife edge tool in the NPF process. Pitch setting of grooves is adjusted in between 250nm to 8 μ m. The indentation load is 0.3N, 0.5N or 0.7N. Depth of a groove depends on hardness of the master mold material; depth of the groove on the aluminum master mold is from 1 μ m to 5 μ m, but that on the soda glass master mold is from 60nm to 160nm. Each stroke of indentation takes around 5 second. It took about a week to fabricate a master mold.

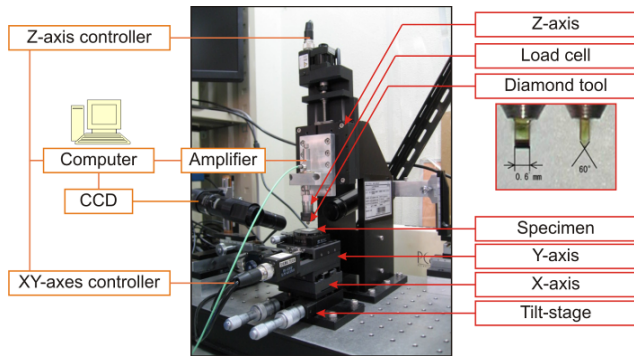


Figure 2: Nano Plastic Forming equipment.

After the NPF, the master mold is utilized for fabrication of a foil mold by coating technique. Three kinds of foil molds are examined.

(1) **Ni plating foil mold:** As for the aluminum master mold, a thin gold (Au) layer is coated on the surface of the master mold by using a DC sputter coater. Then, A thick Ni layer is coated by electro plating. The thickness of the Ni layer is 200 μ m. Finally, the Ni layer is peeled off by hands to acquire a Ni foil mold.

(2) **Ni sputtering foil mold:** As for the soda glass master mold, Ni is first coated on the surface of the mold by using an RF magnetron sputter coater. Thickness of the coated layer is 300nm. Then electro plating is applied on the sputtered Ni layer till the thickness becomes 30 μ m.. Then the Ni layer is peeled off from the master mold to acquire a foil mold.

(3) **TiN composite mold:** Remover compound is coated on the soda glass master mold by spin coating. Then, TiN is coated on it by sputter coating. On the TiN layer, a TiN/Ni composite layer is coated by sputter coating. The composition of a target used in this coating process is 70%TiN+30%Ni. Its composition is determined according to the sputtering rate of each material so that the composition of the coated layer becomes 50%TiN+50%Ni. On the composite layer, Ni is coated. Finally, a thick Ni layer is coated by electro plating. Figure 3 illustrates construction of the coated layers made by this method. After coating, the layer is peeled off from the master mold.

Those acquired foil molds are applied for RNI experiments. Figure 4 shows the rolling mill used for the RNI experiment. Diameter of flat rolls is 27mm, and width of rolls is 30mm. The die and work material are

butted and rolled together. Rolling speed was 850 mm/sec. Rolling load was controlled by adjusting rolling gap. All of RIN process was conducted under room temperature. No lubricant or separating compound was used in the RIN experiments.

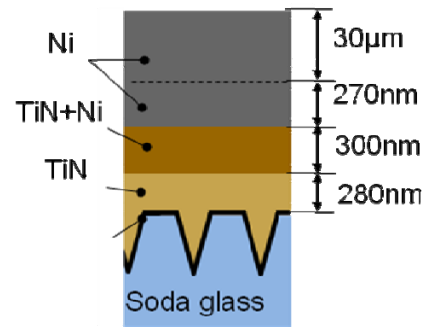


Figure 3: Construction of the composite mold

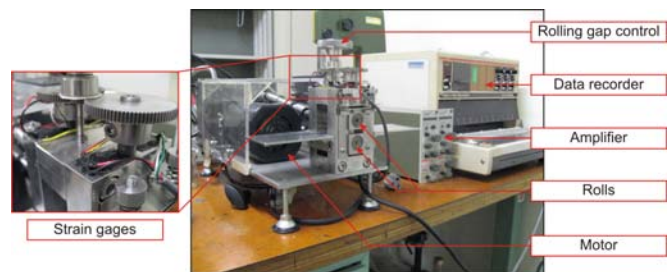


Figure 4: Rolling mill used for roller imprinting experiment.

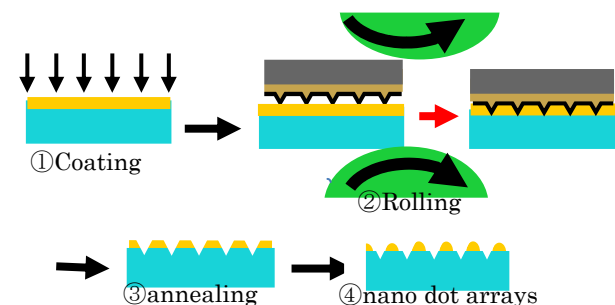


Figure 5: nano dot array fabrication process by roller imprinting.

Two kinds of RNI experiments are carried out. First experiment is the imprinting test on an aluminum plate. Mirror finished pure aluminum plates are used as a work material. The size of the work material is 15mm x 15mm x 1mm. The Ni plating foil mold (1) and the work material are butted, and rolled together. Gap of rollers is adjusted so that the rolling load is maintained within 100N-120N. Fabricated nano structure is analyzed by AFM and FE-SEM observation. Optical surface function is evaluated by a spectrometer (SEC2000-VIS/NIR).

The second experiment is application for a nano dot array fabrication process by combination of coating, patterning by RNI, and annealing. A gold (Au) coated soda glass plate is used as a work material. The size of

the soda glass plate is 15mm x 15mm x 1mm, and thickness of the coated Au layer is 10nm. The coated soda glass is rolled together with the mold (2) or (3). Parallel grooves are fabricated on the coated gold layer. The specimen is then annealed in a electric furnace under the ambient atmosphere. Nano dots are evaluated by FE-SEM observation.

3. Experimental results and Discussion

3.1 Ni plating foil mold

Figure 6 shows FE-SEM micrographs of structures fabricated on the surface of (a) a master mold, (b) a foil mold, and (c) a roller nano imprinted aluminum plate. A variety of line structures with different sizes were fabricated within the 10 mm x 10 mm area of the master mold. It is confirmed from these figures that the master mold's structure is successfully replicated on the foil mold and the rolled material. The structure on the rolled material is uniform and no apparent defects are found.

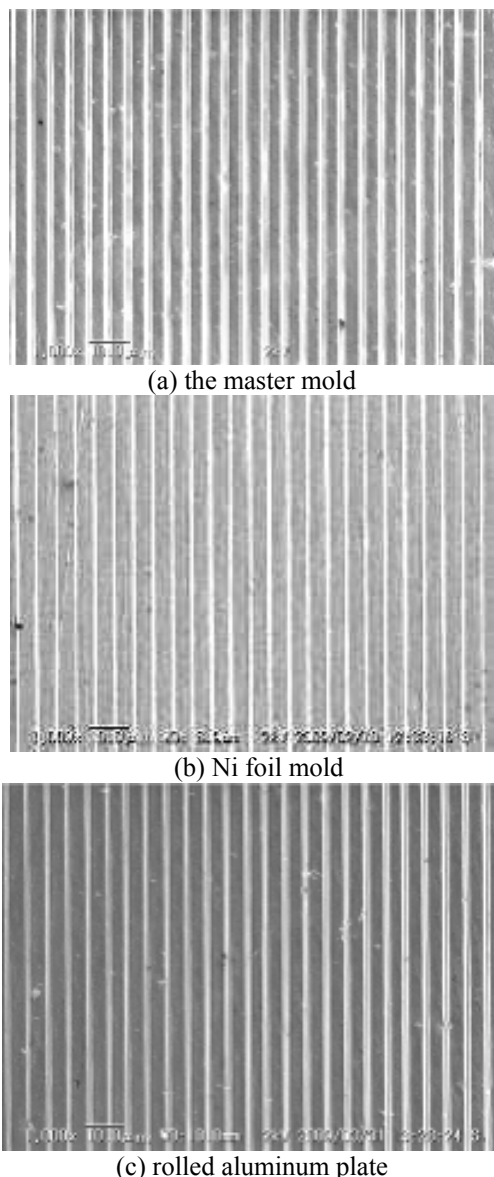


Figure 6: FE-SEM micrographs of line structures on the surface of the molds and a rolled aluminum plate.

In Figure 7, the cross sections of the line structure on the surface of the master mold, the foil mold, and roller imprinted plate are depicted. As for observation on the cross section, the specimens were cured in the epoxy resins. Then, the specimen was cut and polished until the middle part of the line patterns. From Figure 6, it can be confirmed that the foil mold is able to replicate the master mold's structures with a good consistency in terms of shape and geometry. The cross section shows that the resulted structures on foil mold have no apparent change on the tip radius and angle as compared to the master mold's structures. Using Ni foil mold, the structures are uniformly replicated onto the surface of an aluminum plate by RNI process.

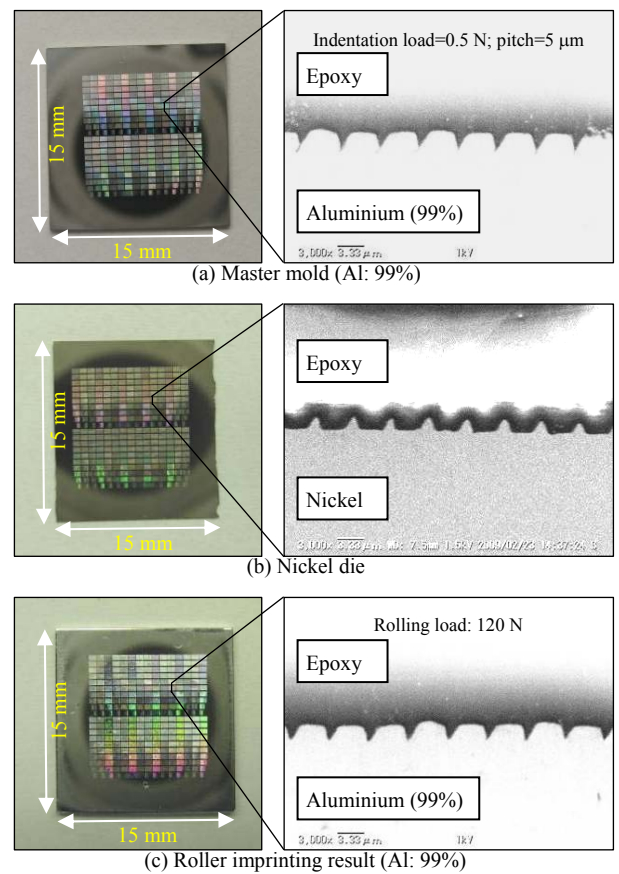
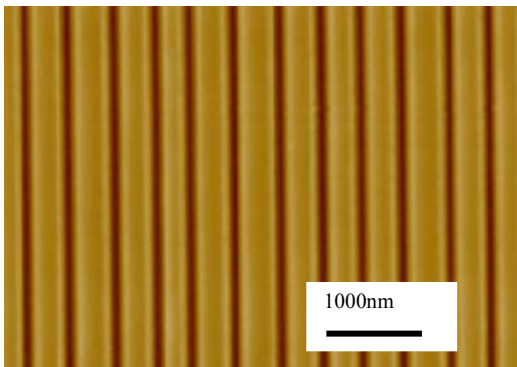


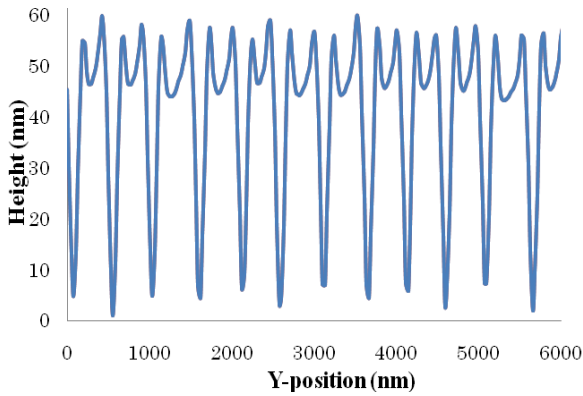
Figure 7: Fabricated line pattern with cross sections.

3.2 Ni spattering foil mold

Figure 8(a) shows an AFM image of the line structure fabricated on the surface of the soda glass master mold. Forming load was 0.7N, and pitch setting of the grooves was 500nm. Figure 8(b) is the profile of the surface structure scanned to the horizontal direction in (a). No cracks or chippings are found in the figure. In (b), embossments are found at the both sides of indented grooves. These results indicate that the soda glass plate exhibits ductile manner deformation when the depth of indentation is small enough. Under such a condition, the shape of knife edge tool is transferred onto the soda glass plate without causing defects.



(a) AFM image



(b) Profile of the line structure

Figure 8: Line structure fabricated on the surface of the soda glass master mold.

than 1 μ m inverted from the soda glass mold is fabricated on the Ni foil. However the lines are not uniform along longitudinal, and some defects are seen. Figure 9(b) is the profile of the structure. Height of the structure is not uniform, but dispersion of the height is less than 10nm.

3.3 TiN composite mold

Cross section of TiN composite mold is depicted in figure 10. The cross section was machined by FIB etching, and was subsequently observed with SIM (Scanning Ion Micrograph). The top layer is TiN, and second layer is the TiN+Ni mixture layer. The bottom layer is Ni layer. These three layers bonded well, and they did not separate during the peeling operation. TiN+Ni mixture layer was efficient to bond the TiN layer and Ni layer.

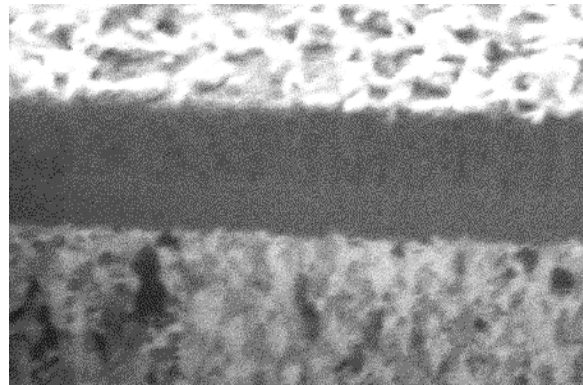
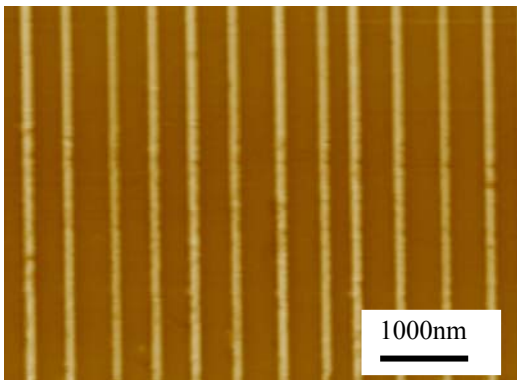
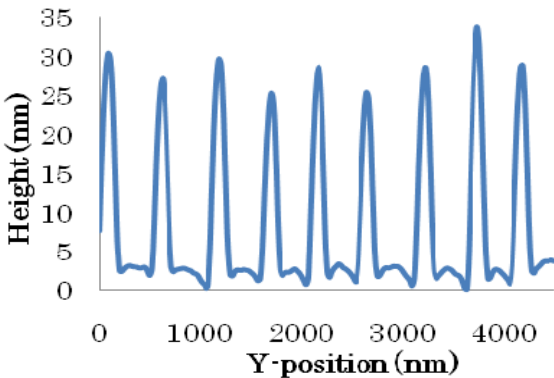


Figure 10: Cross section of TiN composite mold.



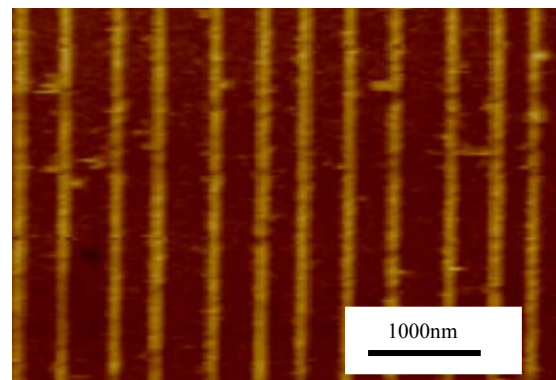
(a) AFM image



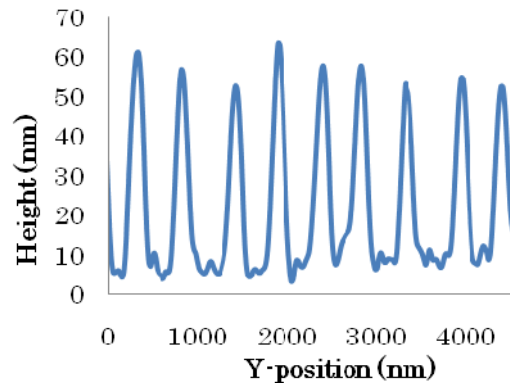
(b) Profile of the line structure

Figure 9: Line structure on the Ni spattering foil mold.

Figure 9(a) shows an AFM image of the line structure of the Ni spattering foil mold. The line structure smaller



(a) AFM image



(b) Profile of the line structure

Figure 11: Line structure on the TiN composite mold.

Figure 11(a) shows an AFM image of the line structure of a TiN composite mold. The line structure is replicated

on the TiN composite mold from the soda glass master mold. Also many defects are seen in the figure. Figure 11(b) is the profile of the structure. Dispersion of height is due to dispersion of the depth of grooves fabricated on the master mold by NPF process.

Spatter coating technique must be developed in order to acquire a finer and more accurate nano structure foil mold. Also, remover compound is important to surface quality of the foil mold because the line structure and foil mold itself are often damaged in peeling operation. Remover compound is also needed to be studied in the future work.

3.4 Modification of optical property by roller imprinting

Figure 12 shows the reflectance measurement results of the fabricated structures. It is found from the figure that reflectance of aluminum plates is greatly affected by the pitch setting of the grooves on its surface. The reflectance values are significantly decreased to approximately 60% relative to the mirror finished aluminum when 8 μm pitch structures are fabricated on the aluminum surface. This is because its structures are inclined 30° with respect to the normal direction. Hence, the light is dispersed to the side and only part of it which strikes the flat surface is reflected. These values keep decreasing with smaller pitch settings. At 1 μm and 2 μm pitch settings, peculiar optical property is observed between 500-700 nm wavelengths, which correspond to green, yellow, and red colors. The maximum reflectance peak of 70 % is recorded at 610 nm wavelength. However, since the ratio between flat area and overall surface area is 70% for 8 μm pitch setting, the reflectance value is further reduced to less than 40%. In addition, the peculiar optical property is shifted to 570-800 nm with a peak of 47% at 680 nm wavelength.

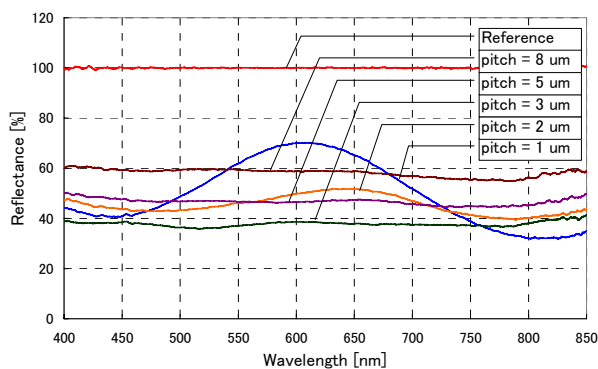


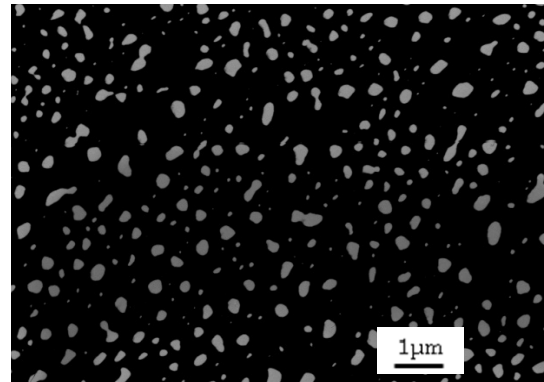
Figure 12: Reflectance measurement results.

3.5 Application to fabrication of nano dot array

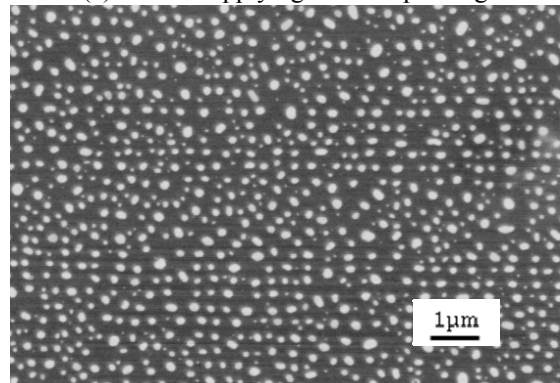
Figure 13 shows FE-SEM micrographs of Au nano dot arrays fabricated on soda glass plates by the combination of coating, RNI and annealing process. (a) is nano dots fabricated only by Au coating and annealing process without applying RNI. (b) is nano dots fabricated by combination of coating, RNI by Ni spattering foil mold and subsequent annealing processes. (c) is those fabricated by similar combination process, but the TiN composite mold is used in RNI.

In (a), many random nano dots are observed. Their

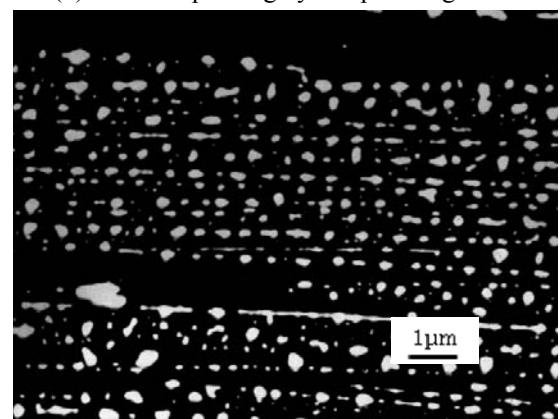
diameter is smaller than $1\mu\text{m}$. It is confirmed that nano dots can be fabricated by simple annealing process. However, shape of these dots is random, and size of dots is not uniform. On the other hand, in (b), many dots are aligned in the horizontal direction, and their sizes are almost uniform. It is apparent that regularity was given by RNI in nano dot array fabrication mechanism. Thus, The Ni spattering foil mold is useful for fabrication of ordered nano dot arrays. However, durability of the mold is not sufficient for practical use, for its nano structure is distorted after several imprinting tests.



(a) Without applying roller imprinting



(b) Roller imprinting by Ni spattering mold



(c) Roller imprinting by TiN composite mold

Figure 13: FE-SEM micrographs of gold nano dot arrays fabricated on soda glass plates.

In (c), nano dots are almost aligned in horizontal direction, but size and shape of nano dots are not uniform when compared to nano dots in (b). TiN composite mold developed in this experiment is not efficient for nano dot array fabrication. This is due to roughness of the mold's

surface and low accuracy in shape discussed in section 3.3. Since TiN mold is harder than Ni mold, much better durability can be expected. The TiN mold should be improved in the future work.

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4. Conclusion

Three kinds of foil molds were developed, and they were applied to RNI experiments. It was shown that the Ni plating foil mold is able to replicate master mold's structure to an aluminum work material by RNI with a good consistency in terms of shape and geometry. Also, optical reflection characteristic of aluminum plate can be modified by RNI with the Ni plating foil mold.

Ni sputtering foil mold was developed, and it was applied to patterning of coated Au layer in the nano dot array fabrication process. Fabricated nano dots were uniform in size and they were aligned in lines. Regularity on nano dots was apparently improved by RNI with the Ni sputtering foil mold.

TiN composite mold was successfully fabricated by adopting a mixture layer between TiN and Ni sputtering layer. The composite mold was also applied to the nano dot array fabrication process. Although fabricated nano dots were aligned in lines, size and shape of nano dots were not uniform. This is due to rough surface and low accuracy in shape of the TiN composite mold.

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