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Microfabrication by hot embossing and injection molding at LASTI

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Abstract LIGA process includes three processes as X-ray lithography, electroforming to fabricate metalic molds and replication, and can be fabricated nano and micro parts for various devices that it is difficult to product by conventional machining methods. A key technology which gathers mass-production efficiency in the LIGA process is micro-replication technology.We choiced hot embossing and injection molding methods for replication. For a demonstration, two kinds of Ni molds, a mesh pattern within a line width of 100 μm, and an aspect ratio of 1.0 and a mesh pattern within a line width of 40 μm, and an aspect ratio of 2.5, were prepared. These were produced with X-ray lithography and nickel electrofoming technique. In hot embossing, an experiment of micro-replication using polymethyl methacrylate (PMMA) and polycarbonate (PC) sheets succeeded. At injection molding, it could not transfer well with PMMA and PC, but injection temperature was set up highly, and it succeeded by cycloolefin polymer. Furthermore, we measured sidewall's surface roughness of microstructures produced at each steppes of the LIGA process, and it checked that the LIGA process had processing accuracy higher than a conventional machining method.

1
Introduction

In recent years, microfabrication technologies have been promoted with development devices in various fields such as micro-mechanics, micro-optics, sensor and actuator. Furthermore, the demand of cheap micro parts made from a plastic is becoming large by chemical, medical and biological worlds. On the other hand, precise processing of several 10-micrometers size cannot do the conventional machining technology. The LIGA processes [1] advocated by the Karlsruhe Nuclear Center (KfK) in Germany in the 1980 s are the high precise microfabrication technology using an X-ray lithography, and mass production technologies which can be carried out by an electroformed mold and replication methods. We conducted basic experiments of hot embossing and injection molding methords as a replication process in the LIGA process. The pattern of fabricated microstructures chose a mesh pattern with which all the patterns are connected in 2-dimensions. Furthermore, sidewall' s surface roughness of the microstructure produced in each steppes of the LIGA process. In this paper, these results are reported.

2
Replication of microstructures

We chose hot embossing and injection molding methods from the following reasons in various replication methods. We are considered to development of an optical device and a biochip. The hot embossing method can transfer a pattern in a large area by comparatively low load. On the other hand, the injection molding method can be manufactured arrays of several 10 microparts per an injection molding cycle within automatically driven. Furthermore, it seems to be easy to fabricate structures with high aspect ratio by injection molding compared with hot embossing. At injection molding, melted resin flow into metallic mold, so this methord represents an advantageous for 3-dimensional microstructures with high aspect ratio.

If a microstructure with a comparatively low aspect ratio is produced, the structure within a pattern size of nanometer-order can be fabricated by hot embossing and injection molding as put in practical use in fabrication of a compact disk (CD) and a digital video disc (DVD). However, fabrication of precise structures with high aspect ratio is difficult. When the deployment to an actual microstructure is considered, we must be optimized the replication condition in a pattern connected in 2-dimensions. There are many reports in which fabrication of structures with isolated patterns like a square pillar, or a simplified pattern like a line and space, was successful. However, there are few reports in which fabrication of structures with all patterns are connected like a mesh pattern, was successful.

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3
Experiments and results

3.1
Preparation of Ni molds with a mesh pattern

We prepared two types of Ni molds for a demonstration, a large size pattern of a Ni mold was named mold type I (Ni mold with the pattern of a mesh structure within a line width of 200 μm, a space width of 100 μm, and a depth of 200 μm), and a more precise pattern size was named mold type II (Ni mold with the pattern of a mesh structure within a line width of 40 μm, a space width of 40 μm, and a depth of 100 μm).

The Ni mold was manufactured as follows. The output beam from the beam line BL11 for deep X-rays lithography [2] of New SUBARU SR facility [3] was irradiated on 1mm-thick polymethyl methacrylate (PMMA) sheet through an X-ray mask with a mesh pattern. The total irradiated energy of dose is 3000 mA-min. After exposure, the PMMA resist was developed in GG developer, and coated with a nickel sputter layer as an electrode for nickel electroforming. In the electroforming process, initial cathode current density was set to 1 A/dm^2, and for approximately 480 h. It ground by having made the external surface of a electroforming structure. The current density was gradually raised to 5 A/dm^2. In the last, a Ni electrofoming object were polished, and was made to complete as a Ni mold.

Three-dimensional diagram measurement result by the scanning electron microscope (SEM) (S-800, HITACHI Co.,Ltd.) photograph and laser beam microscope (VK-8500, 8510, KEYENCE Co.,Ltd.) of each Ni molds are shown in Fig. 1 (a) and (b), respectively. The size of Ni molds are17 × 17 mm × height of 3 mm, and the size of a pattern area are 17 × 3 mm.

3.2
Hot embossing

The experiment of hot embossing was conducted using the equipment developed by Maeda Group of the Institute of Mechanical Systems Engineering of the AIST (National Institute of Advanced Industrial Science and Technology) in Japan. The experiment procedure was shown in Fig. 3. The Ni mold was fixed on the upper stage of the equipment (Fig. 2 (a)) and a resin sheet is arranged on the bottom stage of the equipment. (Fig. 2 (b)) Next, after heating the resin sheet and the Ni mold more than the glass transition temperature of the resin. The Ni mold contacted on the surface of the resin sheet by a servomotor drive. The contacting force was applied until it became a setting value, and was hold to press in the resin sheet for several miniutes. (Fig. 2 (c)) Then, after removing load and cooling in air below to glass transition temperature, dembossing was done. The pattern of the Ni mold was transferred on the resin sheet. (Fig. 2 (d))

The experiment of the replication by hot embossing carried out the following parameters, and investigated the optimized condition. First, polycarbonate (PC) and PMMA were chosen as a material of a resin sheet. The glass transition temperature of PC and PMMA are 150 and 110 °C, respectively. PC and PMMA are easy to get as the shape of a sheet. Therefore, we thought PC and PMMA are suitable for this investigation research. Moreover, in order to do easy the dembossing of the Ni mold and the resin sheet, a mold release agent is applied to the surface of the Ni mold pattern. Fluoride and phlorocarbon were chosen as mold release agents. Next, the contacting force which suppresses the Ni mold on the resin sheet, holding time and the heating temperature at the embossing pocess, and the cooling temperature at the dembossing process, were adjusted. Form evaluation of the fabricated pattern was

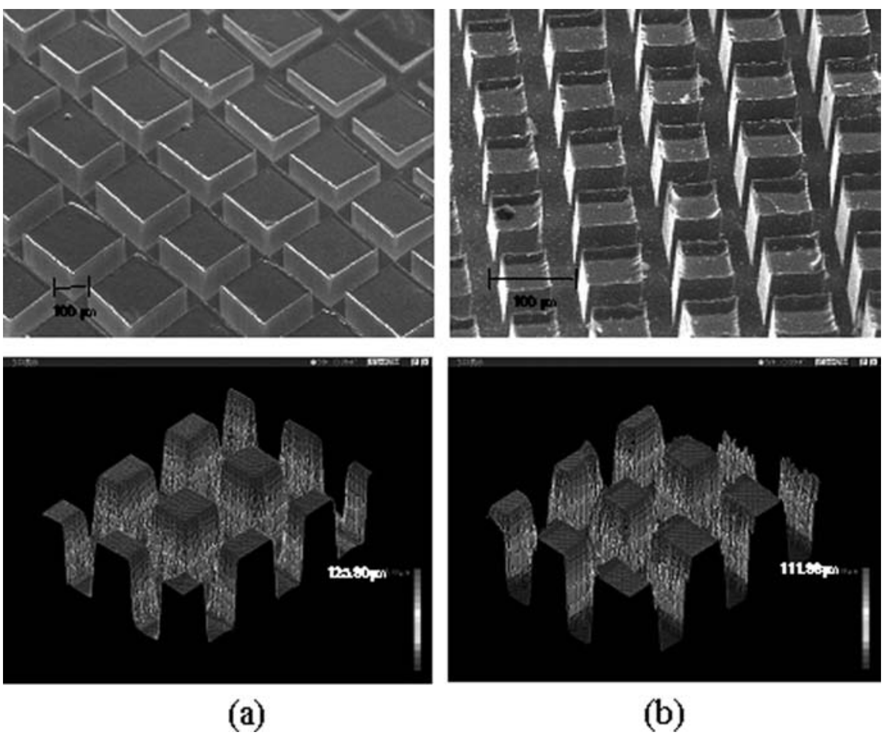

Fig. 1. SEM images and 3D-curvatures measured by Laser Microscope of Ni mold microstructures **a** the mold type I (Ni mold with the pattern of mesh structure within a line width of 200 μm, a space width of 100 μm, and a depth of 200 μm), **b** the mold type II (Ni mold with the pattern of mesh structure within a line width of 40 μm, a space width of 40 μm, and a depth of 100 μm)

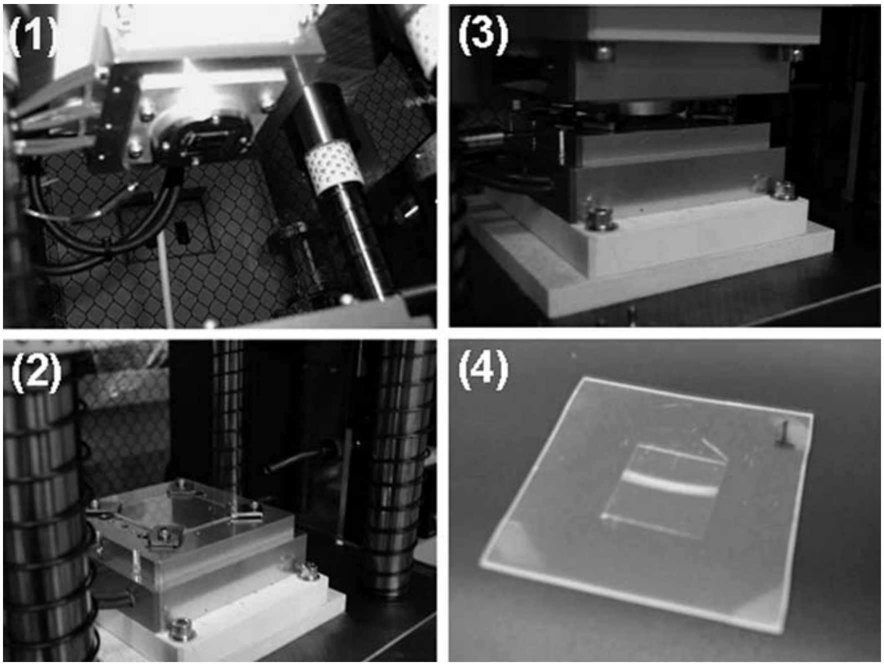

Fig. 2. The experiment procedure of hot embossing, **1** the Ni mold fixed to the upper stage, **2** a resin sheet fixed to the bottom stage, **3** the Ni mold press on the resin sheet, **4** a sample of the fabricated replication

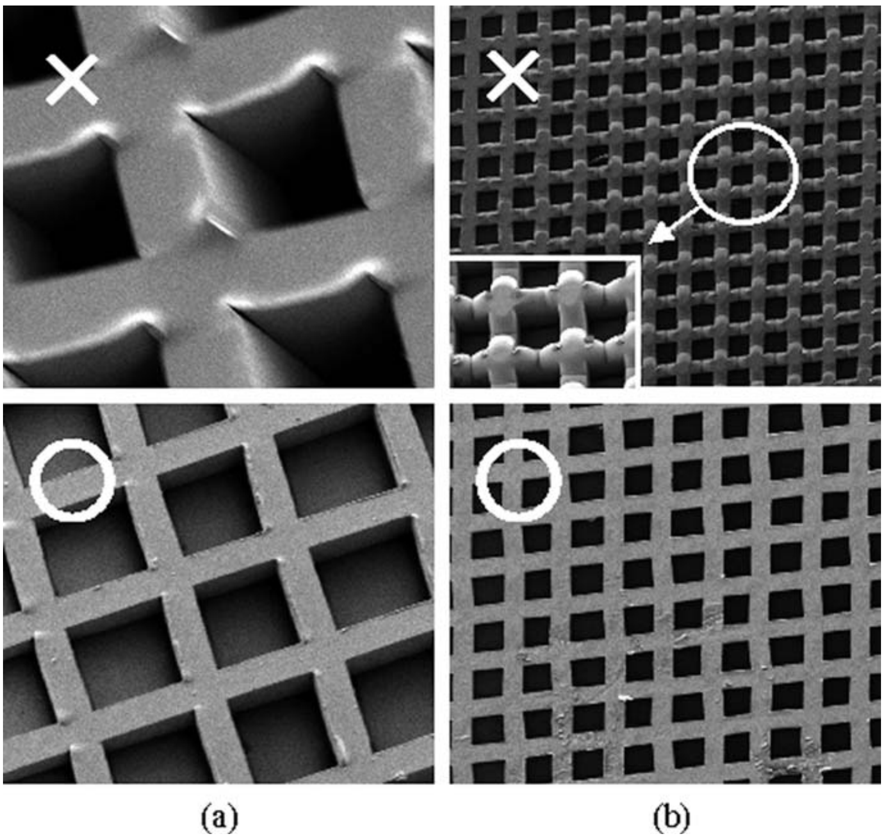

Fig. 3. Estimation of fabricated replications, **a** modification of the edge shape of a pattern (upper: good sample, bottom: bad sample), **b** transferability of a pattern (upper: good sample, bottom: bad sample)

performed mainly by SEM observation. An example of the evaluation was shown in Fig. 3. Fig. 3 (a) shows modification of the pattern edge portion generated at the dembossing process. Modification arises that the temperature at the dembossing process was unsuitable. Moreover, Fig. 3 (b) is an example to which filling of the resin is not carried out to the whole pattern surface from the reason of which the contacting force and the heating temperature at the embossing process were unsuitable. Based on such form evaluation, the result of optimized replication conditions in hot embossing were summarized in Tables 1 and 2. Conditions in Table 1 are in case of using the Ni mold type I, and that in Table 2 are in case of using the Ni mold type II. The numerical values which have drawn the underline are optimized conditions. Photographs of SEM image, and 3D diagram by the laser microscope of the pattern fabricated by the optimized condition using the Ni mold type I and II were shown in Fig. 4 (a) and (b), respectively. The result of these experiments show which microfabrication by hot embossing succeeded in Ni molds of pattern size like type I and II. When the optimized replication conditions in each Ni mold are compared, it turns out clearly that the conditions of the mold release agent and the contacting force differ. The more precise mold pattern in the case of the Ni mold type II, has been required larger contacting force than the Ni mold type I in order to stuff the resin into the pattern.

Table 1. Replication condition in hot embossing using the Ni mold type I

Polymer	Polycarbonate (PC)	Polymethylmethacrylate (PMMA)
Mold release agent	F	F, **Phlorocarbo**
Contacting force (kN)	1, 5	**1**
Holding time (sec)	20, 60	**60**
Embossing temperature (°C)	180	**140**
Dembossing temperature (°C)	130	**95**

Table 2. Replication condition in hot embossing using the Ni mold type II

Polymer	Polycarbonate (PC)	Polymethylmethacrylate (PMMA)
Mold release agent	F	F
Contacting force (kN)	1, 5	1, **5**
Holding time (sec)	20, 60	**60**
Embossing temperature (°C)	180	**140**
Dembossing temperature (°C)	130	**95**

Although phlorocarbon used for the Ni mold type I as a mold release agent was able to carry out coating to the Ni mold by photo-chemical vapor deposition (CVD), coating of a mold release agent will become difficult in case of the the Ni mold with precise patterns like mold type II. Therefore, we have not tried to coat the phlorocarbon on the pattern of the Ni mold type II. Fluoride coating was sprayed on the pattern surface of the Ni mold, and was heated to dry in the furnace.

3.3 Injection molding

In the experiment of injection molding, JMW-013S-10t (Juken Machine Works Co.,Ltd., Japan) injection molding machine was used. A photograph and specifications of this machine are shown in Fig. 5 (a). The pattern surface of the Ni mold have no coating by a mold release agent using the Ni mold type I and II. The Ni mold has been fixed in the mold base shown in Fig. 5 (b), and was included in the injection molding machine. In manufacture of this mold base, it was especially cautious of the following points. If the volatilization gas which occurs from internal air and an internal resin material which dissolved is not discharged out of the Ni mold, it will become the cause of defective fabrications. Heat insulation compression is performed quickly. The gas which remained will become as extraordinary high temperature, and the resin having referred to this high temperature gas will be disassembled. Moreover, a resin may be burned. In consideration of such things, when carrying out the dembossing not only of the

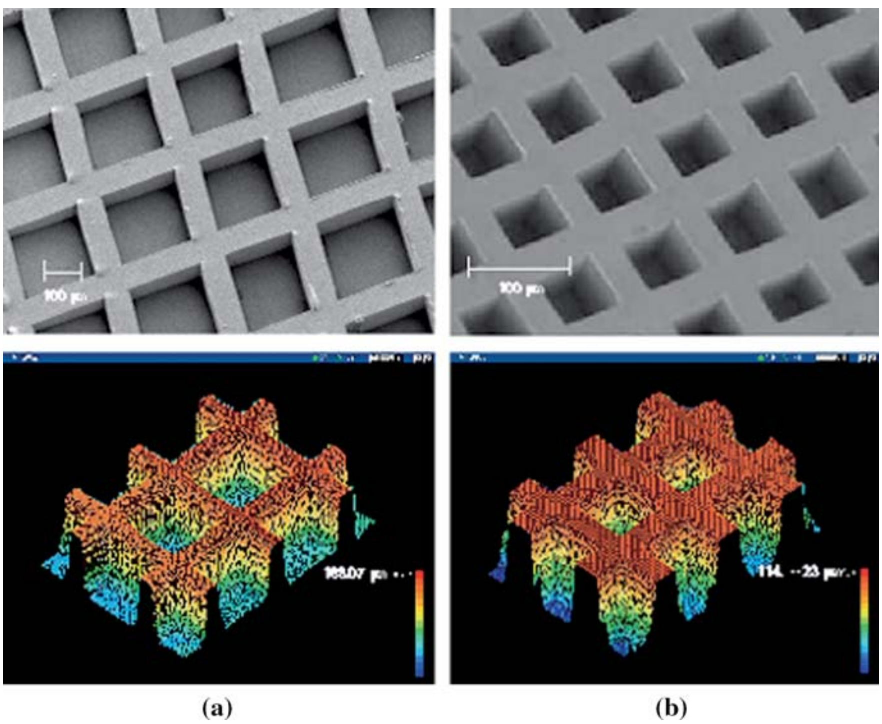

Fig. 4. SEM images and 3D-curvatures measured by Laser Microscope of PMMA hot embossing replicated microstructures, **a** the Ni mold type I (Ni mold with the pattern of mesh structure within a line width of 200 μm, a space width of 100 μm, and a depth of 200 μm), **b** the mold type II (Ni mold with the pattern of mesh structure within a line width of 40 μm, a space width of 40 μm, and a depth of 100 μm)

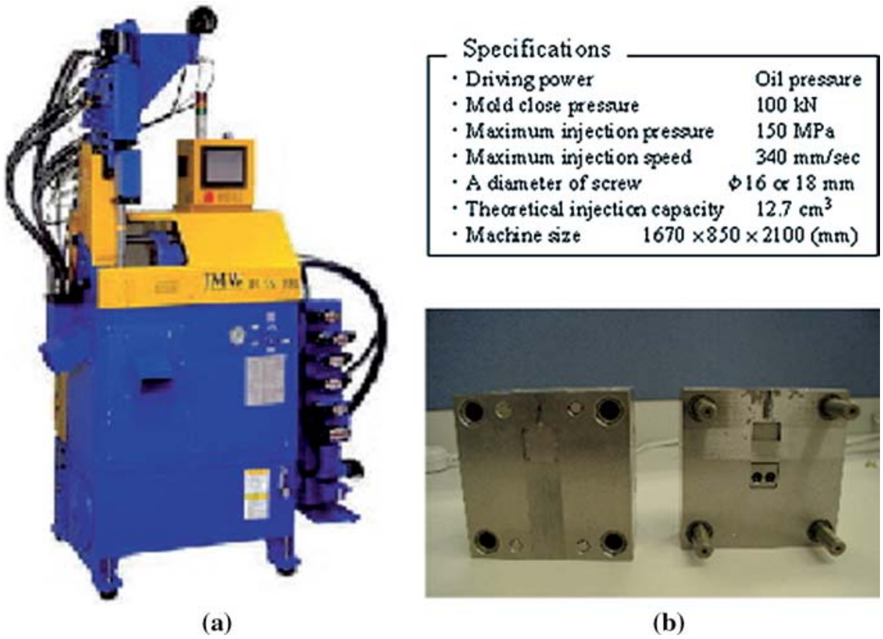

Fig. 5. **a** Injection molding machine and specifications, **b** mold base of the injection molding machine

Table 3. Replication condition in injection modling using the Ni mold type I

Polymer	Polycarbonate (PC)	Polymethylmethacrylate (PMMA)	Cycloolefin polymer (COP)
Injection temperature(°C)	310, 320, 330, 340	288, 300, 310	258, 268, **273**
Molding temperature (°C)	140, 150, 155, 160	95, 140	100, **105**
Injection pressure (MPa)	7.0, 8.0	7.0	**7.0**
Holding pressure (MPa)	6.0, 7.2, 8.0	6.0	**7.0**
Injection speed (mm/sec)	88, 106, 128	106	**106**
Injection time (sec)	2.0	2.0	**2.0**
Cooling time (sec)	10.5	8.5	**10.5**

usual gas vent slot but the cast to the mold base, it used and projected and the gas omission mechanism was added also to the crevice between a pin or a nest.

Experimental results are shown in Tables 3 and 4. Important parameters of injection molding are as follows. resin material, injection temperature, molding temperature, injection pressure, holding pressure, injection speed, injection time, and cooling time. Table 3 shows the result of this experiment in the case of using the Ni mold type I. Table 4 is as a result of using the Ni mold type II. Values with underline in tables are optimized conditions and showed photographs of the SEM images, and 3D diagram measured by the laser microscope of the pattern fabricated on this condition in Fig. 6 (a) and (b), respectively. Materials for injection molding are chosen PC and PMMA, in order to compare with results of replication experiments of hot embossing, and cycloolefin polymer (COP) added in consideration of application to an optical device. Furthermore, the grade of each resin material chose the class which was best in mobility. However, as using PC and PMMA, the Ni mold type I with comparatively large pattern size was not able to succeed in perfect fabrication. However, COP was able to be optimized so that clearly from Tables 3 and 4. Since such a situation, the injection molding replication by PC and PMMA was not tried by the Ni mold type II. When the optimal condition in the mold type I and II are compared, the injection temperature

Table 4. Replication condition in injection molding using the Ni mold type II

Polymer	Cycloolefin polymer (COP)
Injection temperature(°C)	273, 288, 298, 308, 318, 328, **338**, 348
Molding temperature (°C)	**105**, 110
Injection pressure (MPa)	6.0, 6.5, **7.0**, 8.0
Holding pressure (MPa)	6.0, 6.5, **7.0**, 7.5, 8.0
Injection speed (min/sec)	75, 88, **106**, 113, 118, 128
Injection time (sec)	**2.0**
Cooling time (sec)	**10.5**

differs only. Injection temperature meant the temperature of the melted resin material in a heating cylinder, and it was made to represent with the setting temperature of a heating cylinder for convenience. Although the optimized injection temperature changes with kinds of a resin, as temperature is high, it is better in the mobility of a resin material. However, if temperature is too high, a resin will cause decomposition by heat and the problem which discoloring will occur. As the pattern of a mold becomes precise, the work which separates a replication from a mold becomes more difficult. In the case of the Ni mold type II, it will not succeed in fabrication, if the injection temperature of COP hardly raises by heating below the critical temperature.

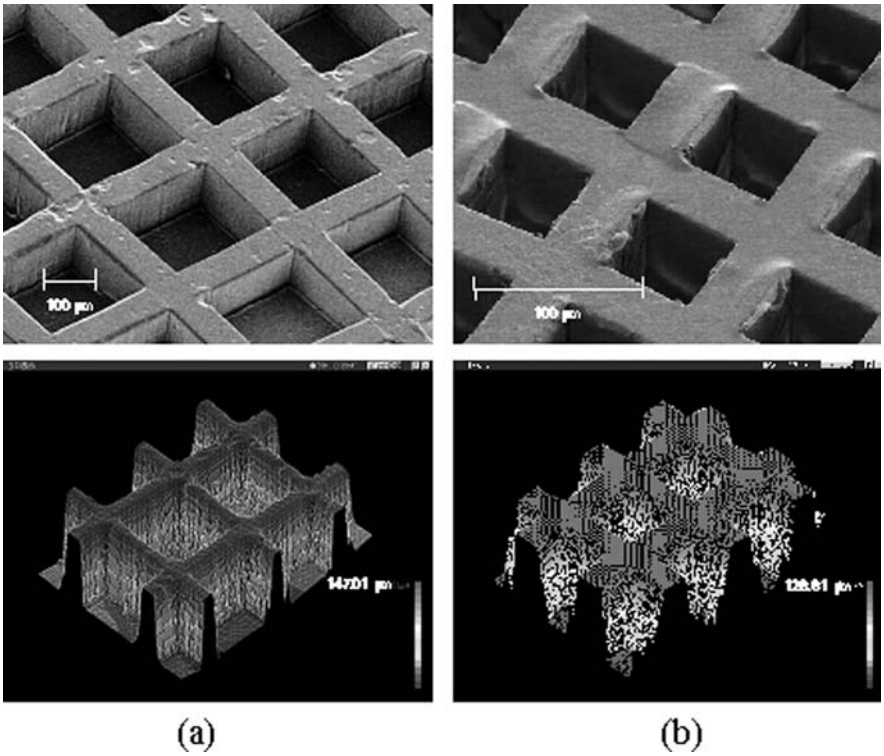

Fig. 6. SEM images and 3D-curvatures measured by Laser Microscope of COP injection molding replicated micro-structures, **a** the Ni mold type I (Ni mold with the pattern of mesh structure within a line width of 200 μm, a space width of 100 μm, and a depth of 200 μm), **b** the mold type II (Ni mold with the pattern of mesh structure within a line width of 40 μm, a space width of 40 μm, and a depth of 100 μm)

As far as we know, the experiment which succeeded in injection molding fabrication by precise mesh pattern like the Ni mold type II have not been reported. Since the resin sheet heated more than the glass transition temperature is perpendicularly pushed all over a pattern to a mesh pattern in hot embossing, if even residents gas can be eliminated, it can be imagined that pattern transfer can be carried out easily. However, at injection molding, the resin which dissolved completely enters horizontally to the injection gate prepared near the one side of a Ni mold to a mesh pattern, and resin flows into the mesh pattern. When it has solidified before the resin which entered into the pattern at the first spreads round the whole pattern, a resin cannot be poured into a pattern from a gate any more. Filling of a resin progresses as follows. At first, a resin into which it injected from the injection gate is filled up with the parallel slot of the injection direction. Next, another slot of a mesh pattern, a slot perpendicular to the injection direction, is buried. Therefore, in selection of the resin used for injection molding, mobility becomes most important factor. It is expected that injection molding with mesh pattern that spread in 2-dimensions is quite difficult.

4
Sidewall's surface roughness of microstructures

The sidwall's surface roughness of precise structures in each process of the LIGA process was measured using the laser beam microscope. In order to prevent loss by the penetration of laser light in the case of resin materials, the resin surface was coated by Au-Pd Chemical Vacuum Deposition (CVD). In compared with measured PMMA surface roughness before and after CVD. The surface roughness before CVD wa s 35.6 nm, and after that was 33.8 nm. Therefore, we judged that deference of the surface roughness by Au-Pd CVD could be disregarded. Measured surface roughness of the PMMA resist, the Ni molds, molded products were 34 to 120 of nm shown in Fig. 7, which was a sufficient transferability for practical application using the LIGA process. As compared with the surface roughness (about 500 nm) of an ordinary mold fabricated by electrical discharge machine, the surface roughness of the mold fabricated by LIGA process proves very smooth. At the X-ray lithography process, it is especially less than 35 nm with RMS. Yoshimura et al [4] reported that they fabricated Ni mold and molded products which the surface roughness was several tens of nm,

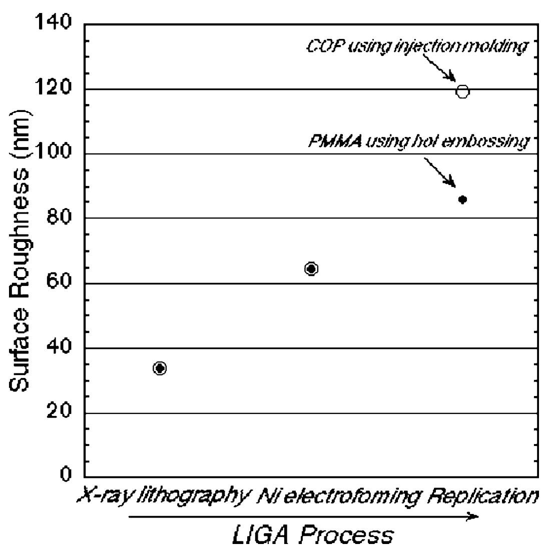

Fig. 7. Measured surface roughness of product's sidewall in the LIGA process

and if the conditions of a regist development process, and an electroforming and a replication process are optimized, it will be thought that it has suggested that the mass-production accuracy of dozens of nano-order becomes possible.

5 Summary

The LIGA process is an integrated process to fabricate micro parts for various devices. We produced the Ni mold type I (Ni mold with the pattern of mesh structure within a line width of 200 μm, a space width of 100 μm, and a depth of 200 μm) and the Ni mold type II with more aggressive pattern size (Ni mold with the pattern of mesh structure within a line width of 40 μm, a space width of 40 μm, and a depth of 100 μm) by X-ray lithography and electroforming, and carried out the basic experiment by two replication methods as hot embossing and injection molding. Usual pattern selected for replication experiments in the LIGA process are isolated patterns as dots or line & space patterns. However, a mesh pattern which spread in 2-dimensions has not been reported. We selected a mesh pattern to apply to device developments. Experiments using both of Ni mold types succeeded, and larger contacting force need to fabricate structures with more precise pattern by hot embossing. At injection molding, fabrication only by COP succeeded, and it turns out that it is necessary to raise the injection temperature in connection with the mobility of a resin for downsizing of a pattern. In addition, it is the first report to have succeeded in replication by injection molding with the precise mesh pattern within a line width of 40 μm, a space width of 40 μm, and a depth of 100 μm.

Moreover, sidewall's surface roughness of micro-structures produced by LIGA process was measured, the surface roughness of the replication product as the last step of the LIGA process was about 100 nm, and has checked that the very smooth sidewall's was acquired as compared with a metallic mold produced by a conventional electrical discharge machining technology.

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