REPLICATION OF NANO/MICRO QUARTZ MOLD BY HOT EMBOS싱
AND ITS APPLICATION TO BOROSILICATE GLASS EMBOS싱

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Glass hot-embossing is one of essential techniques for the development of high-performance optical, bio, and chemical micro electromechanical system (MEMS) devices. This method is convenient, does not require routine access to clean rooms and photolithographic equipment, and can be used to produce multiple copies of a quartz mold as well as a MEMS component. In this study, quartz molds were prepared by hot-embossing with the glassy carbon (GC) masters, and they were applied to the hot-emboss of borosilicate glasses. The GC masters were prepared by dicing and focused ion beam (FIB) milling techniques. Additionally, the surfaces of the embossed quartz molds were coated with molybdenum barrier layers before embossing borosilicate glasses. As a result, micro-hot-embossed structures could be developed in borosilicate glasses with high fidelity by hot embossing with quartz molds.

Keywords: Hot embossing; glass-to-glass embossing; glassy carbon; quartz glass; borosilicate glass; focused ion beam; dicing.

1. Introduction

There have been widespread demands for the high-throughput, high-resolution and cost-effective patterning techniques for glasses in the application fields of MEMS such as photonic crystals1,2 and biochips3 because glasses have excellent optical properties (e.g., high refractive index, low UV absorption level) and chemical/thermal stability, which are essential for high-performance optical and bio MEMS applications.1-5 Conventional microstructuring methods of glasses include wet/dry etching, laser machining, powder blasting, and mold replication technique.1-7 Among these, glass hot-embossing techniques are of interest to fabricate high-precision glass components because they are convenient, do not require routine access to clean rooms and photolithographic equipment, and can be used to produce multiple copies of a quartz mold.1-10

As mold materials for glass production, carbide alloys, such as silicon carbide and tungsten carbide, have been widely used in the precision glass molding process (GMP) due to their hardness and dimensional stability at high temperature. In the previous

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studies, we have proved that a GC is also adequate mold material for high-temperature-embossing of quartz and borosilicate glasses due to its excellent properties (e.g., high operating temperature up to 2000°C, chemical stability, high hardness, and wear resistance). Excellent chemical stability of GC enables for the ease of demolding, further its amorphous structures allows for the nanoscale processing. Although GC is one of promising mold materials for glass embossing, it is but very expensive.

The motivation of the work is that replicated quartz glasses can be used as mold as well as MEMS components. Due to their excellent UV transmission, quartz molds are generally used for the UV-imprint process which can fabricate nano-/microstructures on permanent use UV curable resist at room temperature under low pressure (<0.5 bar). Because quartz mold is technically more difficult to be prepared and much more expensive as compared to a conventional silicon or nickel molds, a fabrication process for a quartz mold is one of the critical issues that need to be studied for the acceptance of UV-imprint technologies in industrial applications. Quartz molds can also be used for hot-embossing of glasses (those have lower softening temperature than quartz). For the process to be successful, the quartz surface has to be coated with an adequate barrier layer to prevent the glass-to-glass bonding phenomena.

In this study, quartz molds were prepared by hot-embossing with glassy carbon masters and applied to the hot-emboss of borosilicate. Glassy carbon molds for the replication of quartz molds were prepared by two different machining processes, including dicing and FIB milling techniques. Prior to borosilicate embossing with quartz molds, the surfaces of quartz molds were sputtered with molybdenum.

2. Experimental Procedure

As a master material for quartz embossing, 3 mm-thick glassy carbon (GS-20, Tokai Carbon, Japan) plate with porosities of 2–3 vol.% was used. The initial surface roughness (Rₐ) for the GC plate measured using an optical interferometer was less than 5 nm. Focused Ga⁺ ion beam milling tests were performed using computer-controlled FIB system (FIB2000A, Hitachi, Japan), and dicing tests were performed using a dicing saw machine (model DAD 522, Disco) with the 20 µm-wide blade.

Glass embossing and heat treatment tests were performed in vacuum using a hot-embossing equipment. The maximum specification of this equipment is 10 kN of load, 1400°C of heating temperature and 0.07 Pa in vacuum. Alignment accuracy between the upper mold and the lower mold was below 10 µm. A position of the upper mold at vertical axis is controlled with ball screw. A GC sample milled by focused ion beam was heated at 1400°C for 10 min to prevent the surface contamination, which generates by the precipitation of gallium ions during heating in the emboss process. Subsequently, the GC sample was cooled to below 200°C without any coolant in vacuum. Borosilicate glass (IWAKI CODE 7740 Pyrex Glass, Asahi technoglass) and quartz glass (PXST, Asahi technoglass) were used as embossing materials. In hot-emboss process, a master structure on a mold surface was pressed into a substrate at an elevated temperature, and then the
applied pressure and the temperature were kept constant for certain time. After completing embossing step, temperature dropped to below 200°C naturally, the glassy carbon mold was removed from the glass substrate. Embossing system provides forced demolding function, and both embossing and demolding was conducted with the upper mold moving speed of 0.3 mm/min. The applied pressure is defined $P_a = L_n/A$, where $L_n$ is the normal load and $A$ is the area of specimen. Residual surface features and the residual surface roughness were determined by interferometric microscopy (ZYGO surface profilometer) and scanning electron microscopy (SEM). For the SEM, sample surfaces were over-coated with a thin Au layer using an auto fine coater (JFC-1300, Jeol).

3. Results and Discussion

3.1. Preparation glassy carbon negative master by FIB milling and dicing

Glassy carbon negative masters were prepared by two different types of fabrication techniques including focused ion beam milling and dicing techniques. Figure 1 represents a glassy carbon mold for micro-chamber arrays fabricated by dicing technique under the feed speed of 50 mm/min. The glassy carbon master has array of pyramids with height of 200 µm, bottom width of 400 µm and side wall angle of 45° on its surface. Prior to the hot-emboss tests, the glassy carbon mold was annealed at 1000°C in vacuum. Subsequently, a 100 µm line-and-spacing pattern was fabricated in 1-mm-thick glassy carbon plate by FIB milling. As a machining process, FIB milling offers the high flexibility in the working shapes, the dimensions (a scale ranging from a few tens of nanometers to hundreds of micro-meters), and the material selectivity. These characteristics allow that the mold with nano/microstructures can be milled directly on metal, silicon, glass, carbon substrate, diamond without any pattern transfer or electroplating. The milled glassy carbon surface was then heat-treated at 1400°C for 10 min in vacuum to prevent the Ga contamination that occurs during high temperature imprinting (>250°C) due to the precipitation of the implanted gallium ions.

Fig. 1. SEM image of the glassy carbon surface machined by dicing saw: array of pyramids with height of 200 µm, bottom width of 400 µm and side wall angle of 45°
3.2. Fabrication of quartz mold by hot embossing with glassy carbon master

Quartz positive molds were replicated from the glassy carbon negative master by hot-embossing. Based on the hot-emboss conditions of borosilicate glass given in Ref. 7, complete filling conditions were investigated with different time at the same temperature and pressure, and could be obtained at a press-head temperature of about 1305°C with a press pressure of 0.22 MPa and an embossing time of 400 s (an emboss velocity of 50/min and a heating rate of 0.5°C s⁻¹). The SEM images of the replicated quartz patterns are represented in Fig. 2. The obtained results were well in agreement with our previous study.

In Fig. 3, by adapting 35°C higher embossing temperature, the embossing time for complete filling could be reduced to 200 s under the same imprint pressure.

![Fig. 2. SEM images of quartz glass surfaces embossed under the embossing temperature of 1305°C and the applied pressure of 0.22 MPa and different embossing time; (a) 100, (b) 300, and (c) 400 s.](image)

![Fig. 3. (a) A SEM image of quartz glass surface embossed under the embossing temperature of 1340°C, the applied pressure of 0.22 MPa, and the embossing time of 200 s. (b) A high magnification SEM image of (a).](image)

3.3. Hot embossing of borosilicate glass with molybdenum sputtered quartz mold

The final process step is glass-to-glass embossing. For the process to be successful, there are some major concerns that need to be overcome. One of them is that a permanent bonding between the quartz glass and the borosilicate glass may occur because embossing temperature is 100°C higher than annealing temperature of borosilicate. It has been reported that permanent glass bonding occurs above 550°C, which is approximately equal to the glass annealing temperature for borosilicate glass.¹⁵
A solution to this problem is to coat the surface of quartz mold with a barrier layer. Under the high imprinting temperature, the barrier material should have the following characteristics: (a) good chemical stability to prevent the adhesion to glasses, (b) high hardness and toughness to prevent deformation or breaking, (c) superior resistance to heat shock, and (d) excellent durability to be used repetitively, thereby allowing for the reduction of overall production cost of glass elements.

The barrier material chosen first was sputtered aurum layer with 200 nm in thicknesses. However, the Au layer did not have a role as protecting layer due to its insufficient durability. As shown in Fig. 4, the Au layer was partially delaminated during demolding after the first imprinting test.

Second, molybdenum (having melting point of 2600°C) was chosen as a barrier material. Molybdenum film with 100 nm in thickness deposited by sputtering on glass provides an inexpensive, inert and mechanically durable layer. In Fig. 5, embossing conditions required for complete filling were investigated under different embossing time using a Mo-deposited quartz glass mold. As shown in Fig 5(c), a complete filling could be obtained under the following conditions: the temperature, 650°C, the pressure, 0.22 MPa, and the embossing time, 1200 s. Expected application of the embossed borosilicate sample is a pyramid microlens array for controlling light diffusion. Figure 6 (a) shows the SEM images of quartz mold that was obtained by hot-embossing with the GC milled using a focused ion beam. In Fig. 6(b), the micro patterns were replicated on borosilicate surface with good fidelity under the temperature of 650°C, the pressure of 0.22 MPa and the embossing time of 600 s.

Fig. 4. SEM image of borosilicate glass surface embossed using the Au-coated quartz mold under the temperature of 650°C, the pressure of 0.22 MPa and the embossing time of 300 s.

Fig. 5. SEM images of borosilicate glass surfaces embossed using the molybdenum-coated quartz mold under the embossing temperature of 650°C and the applied pressure of 0.22 MPa and different embossing time; (a) 600 s, (b) 900 s, (c) 1200 s.
4. Summary

In this study, quartz molds were prepared by hot embossing with the glassy carbon masters, and they were applied to the hot-emboss of borosilicate. Glassy carbon masters were machined by dicing and focused ion beam milling techniques. The achieved glassy carbon masters then applied to the hot-emboss process to produce the quartz molds. Finally, micro-hot-embossed structures were developed in borosilicate glasses with high fidelity by hot embossing with the Mo-sputtered quartz molds. This method showed great potential to fabricate multiple quartz molds efficiently and at a very low cost.

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