Fabrication of Flexible Fine Through-hole Electrodes for Printed Devices using UV-NIL

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Demand for printed electronic devices is increasing, likewise the attention being paid to through-hole electrodes as an integration approach. In comparison with wire bonding integration methods, through-hole electrodes occupy smaller surface areas and have lower power consumption rates. Generally, the diameters of through-holes fabricated in flexible substrates are larger than those developed in rigid substrates. In this study, ultraviolet nanoimprint lithography was used to fabricate microscale through-holes in a flexible substrate, and these holes were subsequently filled with silver ink to form through-hole electrodes.

Keywords: Through-hole electrode, Silver ink, Three-layer structure, UV-NIL

1. Introduction

There is a growing demand for printed devices such as sensors [1], solar cells [2], and batteries [3]. Unlike conventional approaches, which are based on rigid substrates, printed devices can be fabricated at lower costs through the use of flexible substrates [4]. Printed devices can combine multiple functions within a single substrate to enable integration of multiple printed devices [5]. Integration in two-dimension increases the surface area of the device, while three-dimensional integration is also desirable [6]. Through-hole electrodes offer an approach to three-dimensional integration of devices. Through-silicon via (TSV) is a type of through-hole electrode, which occupies a smaller area and has lower power consumption than the wire bonding integration methods [7]. Generally, the diameters of through-holes fabricated in flexible substrates are larger than those produced in rigid substrates such as silicon. Fabrication of small diameter through-holes encourages further integration of flexible devices. Nanoimprint and UV nanoimprint lithography are suitable techniques used for fabricating through-holes on flexible substrates [8-12] and UV-NIL can be used to fabricate patterns using an inverted mold in which a UV curable resin is dropped onto a substrate, allowing the mold to set, exposing to UV light under pressure, and finally curing the resin. Merits of UV-NIL include its high transparency, high throughput, and low cost of equipment [13]. However, a residual layer of thickness [14-17] typically accumulates at the bottom of the holes formed by UV-NIL. In this study, the amount of the dripping of the low-viscosity resin as well as pressurization were controlled, thereby allowing for a residual film-free transfer. Using silver ink, microscale fine wiring was fabricated on a through-hole electrode [18]. A three-layer structure was fabricated by forming fine wiring [19] above and below the through-hole electrode in perfect alignment [20].

2. Experimental

2.1. Fabrication of master mold and replica mold

Three types of the silicon master molds were fabricated in order to obtain the three-layer stacked structure. Master molds were made using photolithography. The photomask described a pattern of the lower layer wiring (Fig. 1a), the through-hole (Fig. 1b), and the higher layer wiring (Fig. 1c), as shown below (in Fig. 1), while the processes involved in the fabrication of the master mold are shown in Fig. 2.

Prepared Si, washed with acetone (GODO Co.,
A 30-nm-thick Cr layer was deposited on the master mold (Fig. 3a). The master mold was immersed in release agent Optool DSX 1.0% (Dakin Industries Ltd.) for 3 h (Fig. 3b). It was then baked for 5 min (Fig. 3c) and later immersed in Novec 7300 (3M Japan Ltd.) for 5 min as well (Fig. 3d).

The fabrication process of the replica mold is presented in Fig. 4.

SU-8 3025 was dropped on the master mold and polyester film (Cosmoshine A4300; Toyobo Co., Ltd.) was placed on it (Fig. 4a). The SU-8 3025 was filled by applying a pressure of 0.5 MPa (Fig. 4b). The SU-8 3025 was cured by UV irradiation at 400 mJ/cm² while simultaneously applying a pressure of 0.5 MPa (Fig. 4c). Baking was performed at 95 °C.
for 10 min (Fig. 4d). Thereafter, the pattern of the master mold was transferred to the UV curable resin by releasing the film (Fig. 4e). The release treatment was performed under the same conditions as those of the master mold.

2.2. Fabrication of work mold

The process of a work mold is shown in Fig. 5. The UV curable resin (PARQIT OEX-028-X433-3, AUTEX Co., Ltd.) was dropped onto Cosmo Shine A-4300, and a replica mold was placed after evacuation (Fig. 5a). Approximately 6.0 MPa of pressure was applied for 5 min to fill the holes in the replica mold with the UV curable resin (Fig. 5b). The UV curable resin was cured by UV irradiation at 400 mJ/cm² while applying a pressure of 6.0 MPa (Fig. 5c). The work mold was then transferred to the UV curable resin by releasing the film (Fig. 5d). The release treatment was performed by changing the metal in Fig. 3a to platinum.

2.3. Fabrication of the lower layer wiring

First, the resin body was fabricated by dropping the UV curable resin (PAK-01-CL; Toyo Gosei Co., Ltd) onto a polyester film and placing the work mold on it (Fig. 6a). The UV curable resin was cured by UV irradiation at 400 mJ/cm² while applying 0.5 MPa of pressure (Fig. 6b). This was followed by the transfer of the pattern of the work mold to the UV curable resin by releasing the film (Fig. 6c). The resin body was subjected to the same release treatment as that of the master mold (Fig. 6d). Silver ink (Smart Screen F, Techno Alpha Co., Ltd) was then filled to the bottom by evacuation (Figs. 6e and 6f) and the excess ink was removed by wiping the top of the pattern with a Kim Towel (Nippon Paper Crecia Co., Ltd.) (Fig. 6g). Next, the silver ink was sintered (Fig. 6h), UV curable resin (ETAX-003XC, AUTEX Co., Ltd) dropped onto the silver pattern on the resin body (Fig. 6i), and then cured by UV irradiation at 3000 mJ/cm² while applying a pressure of 1.0 MPa (Fig. 6j). The lower layer wiring was fabricated by releasing the film (Fig. 6k). Because ETAX-003XC retains its adhesiveness even after curing, the silver pattern in the replica mold was taken out with UV-NIL.

2.4. Fabrication of the through-hole electrodes and upper layer wiring

A 0.5-μL portion of PAK-01-CL was dropped, and the work mold of the through-hole was placed on the lower layer wiring (Fig. 7a). UV curing resin between the lower layer wiring and work mold was removed by applying 2.0 MPa pressure for 5 min (Fig. 7b). The UV curable resin was cured by UV irradiation at 400 mJ/cm² while applying a 2.0 MPa pressure (Fig. 7c). The through-hole was fabricated on the lower layer wiring by removing the work mold (Fig. 7d), and subsequently, the through-hole electrodes were fabricated by filling the through-hole with silver ink (Fig. 7e). The filling method adopted was the same as that of the lower layer wiring (Figs. 6e–6h), while the higher layer wiring was fabricated on the through-hole by the same method.
process utilized in fabricating the through-hole electrodes.

![Fabrication process of the through-hole electrodes.](image1)

3. Results and discussion

Figure 8 shows the width and height of the molds. The height of the master mold corresponds to the thickness of SU-8 3025. Curing shrinkage is responsible for the inverse relationship that exists between the height and the number of transfers. Depths and heights were measured the cross-sectional view of spare mold samples using SEM. Hereafter, depths and heights were measured by the same manner.

|               | Wired pattern | Through hole |
|---------------|---------------|--------------|
| **Master mold** |               |              |
| Width [μm]    | 5.3           | 10.2         |
| Height [μm]   | 3.0           | 8.4          |
| **Replica mold** |               |              |
| Width [μm]    | 5.0           | 10.0         |
| Depth [μm]    | 2.9           | 7.7          |
| **Work mold**     |               |              |
| Width [μm]    | 4.7           | 9.7          |
| Height [μm]   | 2.9           | 7.3          |

Fig. 8. Optical microscope images of fabrication of each mold and its dimensions.

Figure 9 shows SEM images of the lower layer wiring.

![SEM images of the fabricated lower layer wiring.](image2)

Figure 10 shows optical microscope images of the resin body of the lower layer wiring (see Fig. 6k).

![Optical microscope images of the fabricated lower layer wiring.](image3)

The resin body has a width of 4.6 μm and a depth of 2.6 μm, while the lower layer wiring has a width of 4.2 μm and a height of 2.6 μm. The difference in measurements between the resin body and the work mold is attributed to curing shrinkage. Heating
evaporates the solvent, which consequently results in obtaining a lower layer wiring of reduced width.

Figure 11 shows the optical microscope images of the resin body of the through-holes and the lower layer wiring (see Figs. 7d,e).

The resin body of the through-holes have a width of 9.7 μm and a depth of 7.0 μm, while the through-hole electrodes have a width of 9.5 μm and a height of 7.0 μm.

Figure 12 shows the optical microscope images of the resin body and wiring of the higher layer by silver filling.

The calculation method of resistance of connected silver wiring and measurement set-up are shown in Fig. 13. The measurements of resistances were contacted on higher layer wiring areas by electric tester. Measurements were carried out for several points. The actual resistance value of the combined lower layer wiring-through-hole electrode-upper layer wiring was 55–180 kΩ which was much larger than the calculation value of 1.74–8.69 Ω. This reason is considered as follows. One is oxidation of silver ink surface during fabrication process. Other is residual layer was partially remined at connecting interface. This causes conduction loss of silver ink wiring. To avoid these issues, further improvement is necessary and this is future work.

Figure 14 shows X-ray CT (XWES-SH160, XMS Co., Ltd.) images of the three-layer structure. The resolution of this device is 0.6 μm. It can be seen that the alignment of the lower layer wiring, the through-hole electrode, and the upper layer wiring are accurate and that they are in close contact three-dimensionally.

Figure 15 shows the TEM (Transmission Electron Microscope) images of the lower layer for two magnifications: 17500 times and 88000 times.

The silver ink was sintered and converted into silver crystals. These small particles have a thin and almost uniform contrast on their surfaces, thus lending credence to the assumption that the particles are amorphous.
4. Conclusion

The through-hole electrode for wiring was successfully fabricated using silver ink. The resistance value of the integral lower layer wiring to through-hole electrode to upper layer wiring was measured as 55–180 kΩ. X-ray CT showed that the three-layer structure was integrated three-dimensionally. Lastly, the TEM image indicates the probability of the formation of silver crystals.

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References

1. S. Khan, S. Tinku, L. Lorenzelli, and R. S. Dahiya, IEEE Sens. J., 15 (2015) 3146.
2. H. Antoniadis, F. Jiang, W. Shan, and Y. Liu, Proc. 35th IEEE Photovoltaic Specialists Conf., (2010) 001193.
3. B. Choi, J. Nho, H. Cha, T. Ahn, and S. Choi, IEEE Trans. Ind. Electron., 51 (2004) 140.
4. V. Subramanian, P. C. Chang, D. Huang, J. B. Lee, S. E. Molesa, D. R. Redinger, and S. K. Volkman, Proc. 19th Int. Conf. VLSI Des., (2006) 1.
5. I. Jeerapan and S. Poorahong, J. Electrochem. Soc., 167 (2020) 037573.
6. G. Van der Plas, P. Limaye, I. Loi, A. Mercha, H. Oprins, C. Torregiani, S. Thijs, D. Linten, M. Stucchi, G. Katti, D. Velenis, V. Cherman, B. Vandevelde, V. Simons, I. De Wolf, R. Labie, D. Perry, S. Bronckers, N. Minas, M. Cupac, W. Ruythooren, J. Van Olmen, A. Phommahaxay, M. de Potter de ten Broeck, A. Opdebeeck, M. Rakowski, B. De Wachter, M. Dehan, M. Nelis, R. Agarwal, A. Pullini, F. Angiolini, L. Benini, W. Dehaene, Y. Travaly, E. Beyne, and P. Marchal, IEEE J. Solid-State Circuits, 46 (2011) 293.
7. M. Motoyoshi, Proc. IEEE, 97 (2009) 43.
8. S. Y. Chou, P. R. Krauss, and P. J. Renstrom, J. Vac. Sci. Technol. B, 14 (1996) 4129.
9. J. Haisma, M. Verheijen, K. Van Den Heuvel, and J. Van Den Berg, J. Vac. Sci. Technol. B, 14 (1996) 4124.
10. J. Taniguchi, Y. Kamiya, T. Ohsaki, and N. Sakai, J. Photopolym. Sci. Technol., 22 (2009) 175.
11. J. Taniguchi, Y. Kamiya, and N. Unno, J. Photopolym. Sci. Technol., 24 (2011) 105.
12. N. Unno and J. Taniguchi, J. Photopolym. Sci. Technol., 24 (2011) 57.
13. N. B. T. Yusof and J. Taniguchi, Microelectron. Eng., 110 (2013) 163.
14. S. Gillesac, M. Meier, M. Prömpers, A. van der Hart, C. Kügeler, A. Offenhäusser, and D. Mayerac, Microelectron. Eng., 86, (2009) 661.
15. N. Koo, J. W. Kim, M. Otto, C. Moormann, and H. Kurz, J. Vac. Sci. Technol. B, 29 (2011) 06FC12.
16. J. Taniguchi and N. Unno, J. Photopolym. Sci. Technol., 28 (2015) 547.
17. H. Ueda and J. Taniguchi, J. Photopolym. Sci. Technol., 31 (2018) 283.
18. R. Ji, M. Hornug, M. A. Verschuuren, R. van de Laar, J. van Eekelen, U. Plachetka, M. Moeller, and C. Moormann, Microelectron. Eng., 87 (2010) 963.
19. N. Sato and J. Taniguchi, Microelectron. Eng., 193 (2018) 79.
20. H. Wadayama, T. Okabe, and J. Taniguchi, Microelectron. Eng., 193 (2018) 47.