XII International Conference on Computational Plasticity. Fundamentals and Applications
COMPLAS XII
E. Oñate, D.R.J. Owen, D. Peric and B. Suárez (Eds)

SIMULATION OF DEFORMATION OF LAYERED SHEET DURING MICRO POWDER IMPRINTING PROCESS

FUJIO TSUMORI*, YANG XU, HYUNGOO KANG, TOSHIKO OSADA, HIDESHI MIURA

* Department of Mechanical Engineering
Kyushu University
Motooka 744, Nishi-ku, Fukuoka City, Japan
e-mail: tsumori@mech.kyushu-u.ac.jp

Key words: FEM, Micro Powder Imprinting Process, Layered Sheet, Interface.

Summary. This paper reports simulation of deformation during micro powder imprinting process, which is a newly developed process to form micro patterned surface on ceramic sheets. The process is proposed as a combined process of traditional hot-embossing and powder metallurgy. A compound sheet of powder material and polymer binder is pressed by a mold to be transcribed a micro-pattern on its surface. After pressing, the binder is removed by heating, and the sheet is sintered. Finally dense ceramic sheet with fine pattern can be obtained. This process can be used also for layered sheet of two different materials. By using this layered sheet, we can make a pattern not only on the surface of the upper layer but also along the interface between each layer. Of course, the same pattern with the mold's shape can be transcribed on the surface, while, there can be found another micro pattern along the interface. These two patterns of the surface and the interface are useful to fabricate ceramic sheet with patterns on its both surfaces. For example, if a compound sheet was used as an upper layer and a pure organic sheet as a lower, the lower organic layer can act as a sacrificed layer. After debinding and sintering, only the upper layer remains, which has micro-patterns on the both sides.

The shape of interface between layers can be controlled by changing properties of layers. It could be also influenced by the imprinting conditions, such as temperature and pressure. In order to design the shape of the interface, finite element analysis was employed in the present paper. Mooney-Rivlin's deformation model was used to express deformation of materials during imprinting. In this paper, we compared the simulated results with experimental data to show effectiveness of the present simulation method.

1 INTRODUCTION

Nano imprint lithography (NIL) has been proposed for the semi-conductor fabrication field, and much research has been performed since the 1990s [1]. The NIL process is a simple hot-embossing process which employs very fine patterned mold to transcribe the pattern onto polymer thin film. The resolution of this NIL process is extremely fine. However, the process material has been limited to only polymer materials. In order to enlarge variety of materials for this imprint process, authors focused on powder metallurgy process, in which metal or ceramic product can be formed from powder material. A mixture of powder and polymer binder material may be used as a starting material for this powder metallurgy field. This mixture can flow into a mold to make a product's shape. The mixed polymer binder can be removed from the formed powder compacts, and the compacts will be sintered to obtain solid product. This is called powder injection molding process [2], which is one of the typical powder metallurgy processes. We employed the same kind of compound material, which is a mixture of powder and polymer, for the imprint process. The process was named as micro powder imprint (μPI) process [3-6]. Prepared compound sheet was applied to the imprint process, and the patterned sheet was debound and sintered subsequently. We have employed this technique to enhance performance of solid oxide fuel cell by fabricating micro-patterned ceramic sheet for electrolyte layer of the fuel cell [4-6].

Next, the above micro powder imprint process was improved to enlarge shape variation of products [7, 8]. In this improved process, layered sheet was prepared for work material as shown in Fig. 1. When the layered sheet was imprinted, we can obtain not only the pattern on the surface, which is same with the mold pattern, but also the pattern along interface between the layers. If the compound sheet, which consists of powder and polymer, was used for the upper layer and pure polymer material was for the lower layer, the lower layer can be easily removed during the debinding process. After sintering, we can obtain a thin sheet that has micro patterns on the both sides. The imprint process for layered material has been already reported [9, 10]; however, it has not been used effectively. As the present process is combined with powder metallurgy process, the interface structure can be used directly as a pattern on the rear surface.

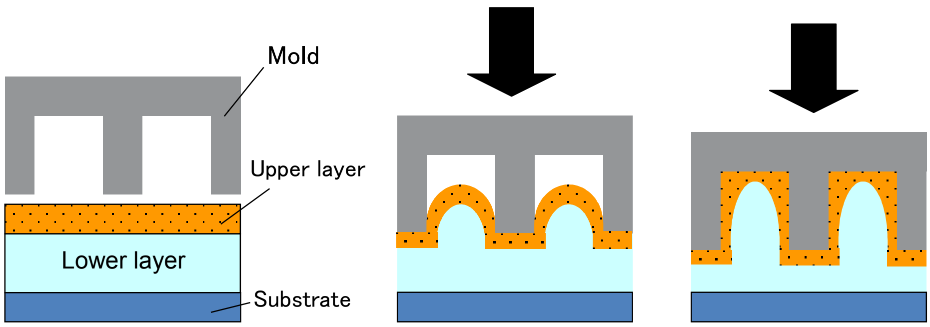


Figure 1: Schematic of imprint process with multi-layered material.

The structure along the interface should be well controlled; however, it is not easy to expect the shape of interface before forming. The structure can be changed, if we employed another material for each layer [7]. In the present paper, FEM simulation was performed to predict the interface shape of the layered imprinting process. We also applied FEM for the simulation of multi-step imprinting process [11], which will be described later.

2 IMPRINTING PROCESS FOR LAYERED SHEET

As described in the previous section, the imprinting process for layered sheet material is a powerful tool to obtain a ceramic or metal sheet with patterns on the both sides. Figure 2 shows a typical experimental result of imprinted layered sheet. In this experiment, poly (methyl methacrylate), or PMMA, was used as a polymer material. Magnetite nano particles were

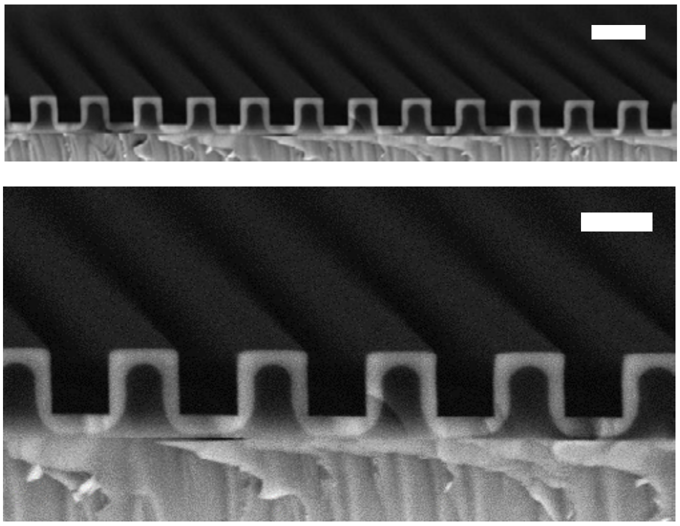


Figure 2: SEM images of an imprinted sample to layered material. The upper layer contains magnetite nano particles. Scale bars in upper and lower figures are 10 μm and 5 μm, respectively.

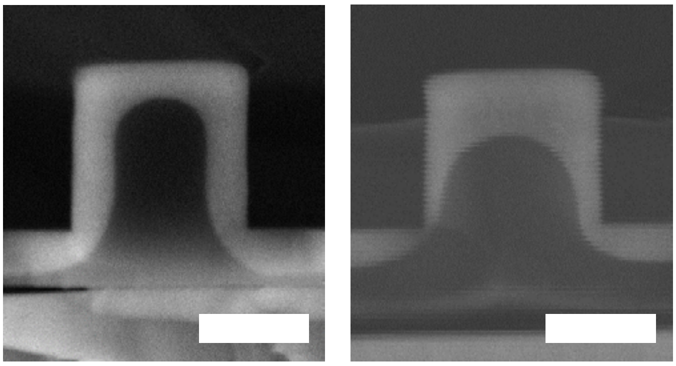
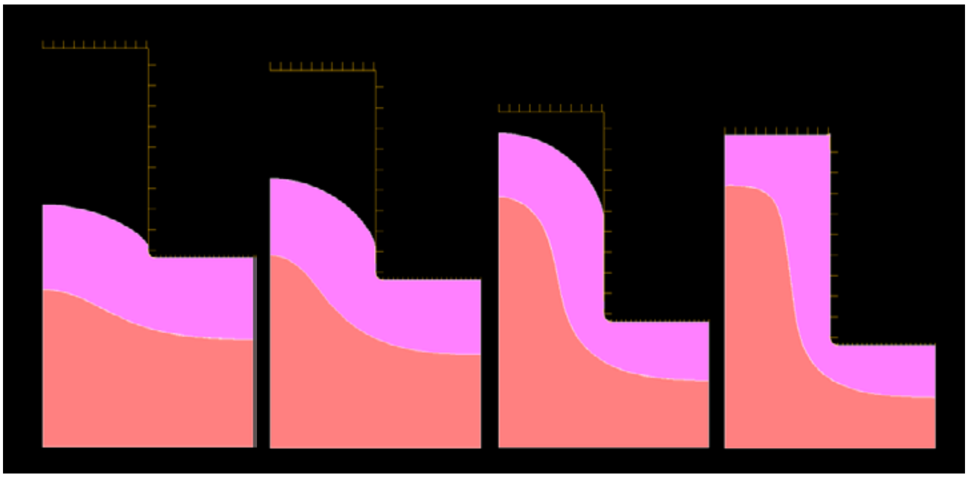


Figure 3: SEM images of imprinted samples. Left figure shows the sample with lower molecular weight PMMA for the lower layer. Right figure shows the sample used PMMA with higher Mw as the lower layer. Scale bar in each figure shows 3 μm.

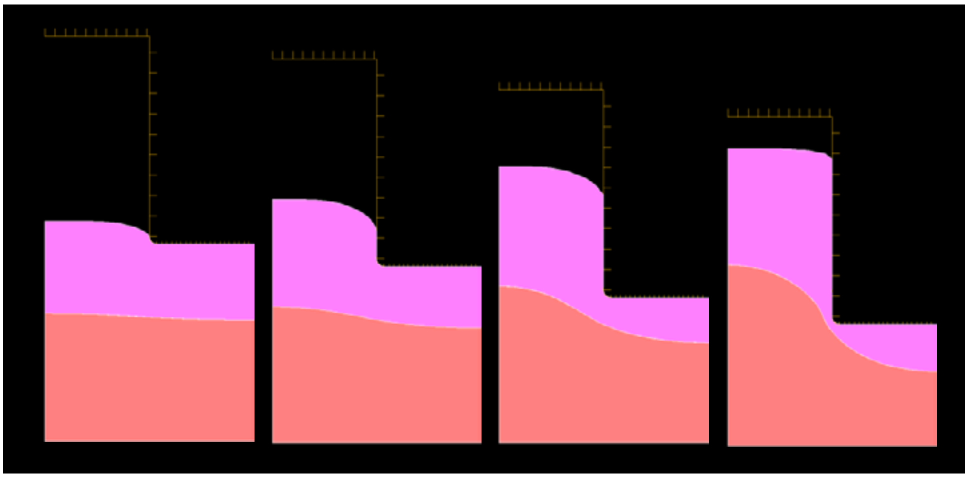
dispersed in the upper layer which looks white. The line and space pattern was well-printed, and the pattern contained clear interface curves inside.

The interface was changed when the molecular weight (Mw) of polymer material was changed. Figure 3 is an example of changing the molecular weight [7]. The upper layer material of each sample was the same one, PMMA with 120 k Mw dispersed with magnetite nano particles. The lower layer of the left sample was PMMA with 15k Mw, while the right sample's one was 350 k Mw. Molecular weight of polymer changes mechanical properties, such as elasticity, viscosity, hardness and so on. Generally, as the molecular weight increases, higher elasticity and higher viscosity were obtained.

To express deforming behavior of polymer during the imprint process, Mooney-Rivlin's



(a)



(b)

Figure 4: Simulated results of imprint for layered sheets : (a) The upper layer was higher Mw, and (b) The lower layer was higher.

deformation model was employed in the present study. The Mooney-Rivlin parameters of C_{10} and C_{01} for higher Mw material were set 13.3 MPa and 3.33 MPa, respectively. While, for lower Mw material, 2.00 MPa and 0.50 MPa were set. We employed a commercial FEM software (MSC-Marc) for the following simulation. The initial geometry was set to make the same situation with the above experiment. During imprint process, deformation is usually large so that we employed adaptive remeshing option for the present problems. To simulate deformation of cross section in the line and space structure. Two-dimensional plane strain problem was solved in the simulation.

Figure 4 shows the simulated results. Figures 4(a) and 4(b) represent high Mw upper layer and high Mw lower layer, respectively. In the latter case, the simulation could not finish because of large strain, so that the cavity has not been filled in the last figure. The obtained interface lines were different each other. The polymer with lower Mw can flow so easily that it pushed up in Fig. 4(a), while it flowed into the mold cavity earlier in Fig. 4(b). The shapes are similar with the experimentally obtained ones as shown in Fig. 3. From these results, it is found that mechanical properties of each layer is one of key factors to determine the interface structure.

3 MULTI-STEP IMPRINTING PROCESS FOR LAYERED MATERIAL

In the previous section, we showed some experimental and computational examples of the imprint process for layered materials. The present process has further possibility to enhance variety of deformation. In this section, multi-step imprinting process is proposed and demonstrated. Schematic of the process is shown in Fig. 5.

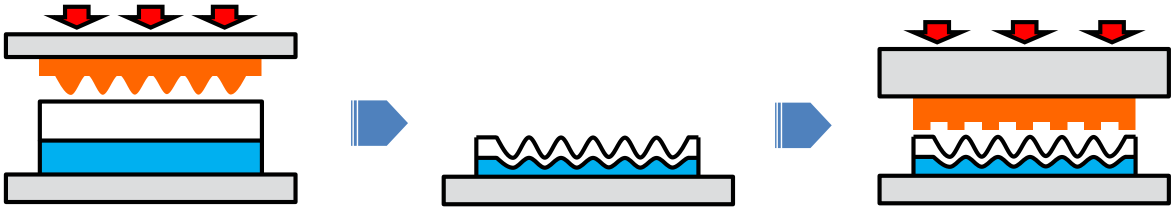
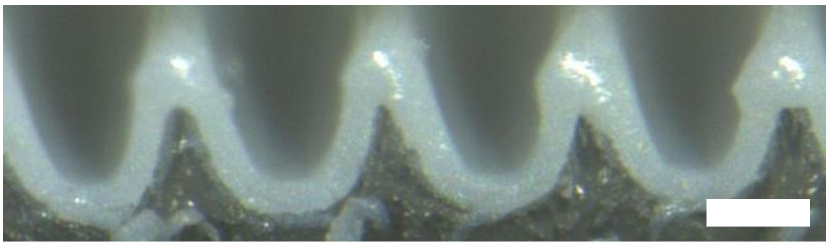
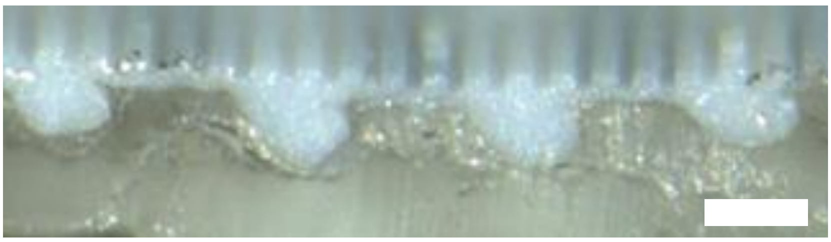


Figure 5: Schematic of multi-step imprint process with multi-layered material. The imprinted layered sheet is imprinted again by another mold.

At first, layered sheet was imprinted by a mold. This is the same process with the previous section's one. Next, the patterned sheet is imprinted again by another mold. After the second imprinting, the same pattern with the second mold is obtained on the surface. At the same time,



(a)



(b)

Figure 6: Cross-sectional micro view of imprinted layered sample. The upper figure shows after the first imprint (a), and the lower one shows after the second imprint (b). Scale bar in each figure shows 50 µm.

shape of the interface between upper and lower layers will be changed.

Figure 6 shows an example by this multi-step imprinting process. In this experiment, polyvinyl alcohol (PVA) was used as polymer material with glycerin as a plasticizer. Upper white layer contained yttria stabilized zirconia (YSZ). The detailed information about these materials can be found in ref. [5]. Wavy patterned mold was used for the first imprint, and subsequently, fine rectangular wave pattern was imprinted. Wavy surface and also wavy interface were observed after the first imprinting as shown in Fig. 6(a). After the second imprinting, the second mold pattern was also successfully transcribed, and it is noted that shape of interface had been changed dramatically during the process, as shown in Fig. 6(b).

We can design different patterns on the surface and the interface by this technique; however, it is found that controlling of the interface shape is not easy to estimate in advance. Also in this case, FEM simulation could be a useful tool. Figure 7 shows a simulated result, which resembles the above experiment. In this simulation, a flat plate was used as the second mold to simplify the problem; however, the change of the interface can be well-analyzed by the simulation. Although the process could not be finished because of large strain, it is found that the shape of the interface shows the same tendency with the result shown in Fig. 6.

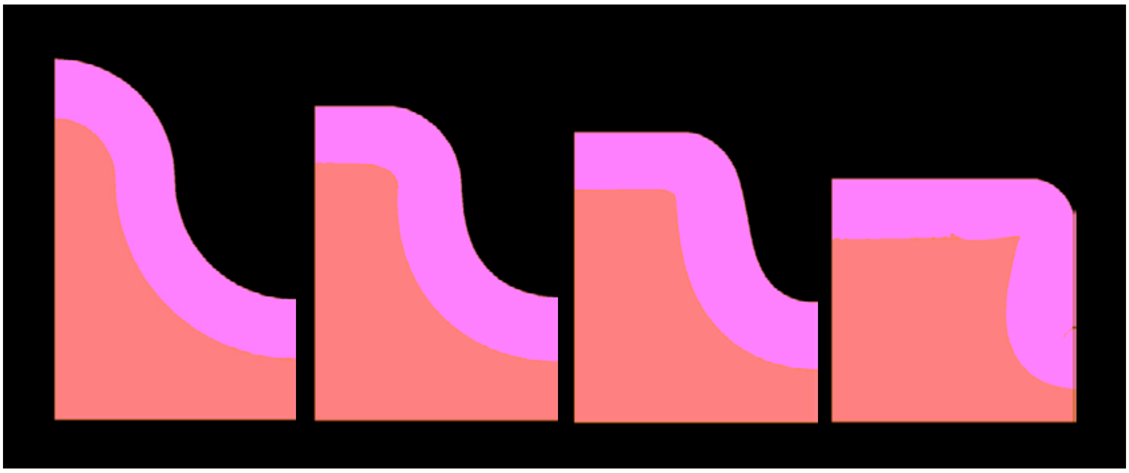


Figure 7: Simulated result of multi-step imprinting process for layered material.

4 CONCLUSIONS

- FEM simulation was performed to describe deformation of layered sheet material during the micro powder imprinting process. The simulated results showed good agreement with the experimental ones.
- Multi-step imprinting process for layered sheet was proposed to enhance deformation mode of the interface between upper and lower layers.
- FEM was also employed to simulate the multi-step imprinting process. The result showed that the simulation is effective to design the product shape for the process.

5 ACKNOWLEDGEMENT

This work was supported by JSPS KAKENHI Grant Number 25630028. We are also grateful for funding from the International Research Center for Hydrogen Energy of Kyushu University.

REFERENCES

[1] Chou, Y.S. et al., Imprint Lithography with 25-Nanometer Resolution, *Science* (1996), **272**: 85-87.

[2] German, M.R., *Powder Injection Molding*, Metal Powder Industries Federation (1990).

[3] Xu, Y., Tsumori, F. et al., Fabrication of Micro Patterned Ceramic Structure by Imprinting Process, *J. Jpn. Soc. Powder Powder Met.* (2011) **58**-11: 673-678.

[4] Xu, Y., Tsumori, F. et al., Study on Fabrication of Micrometer Patterned Ceramic Sheet with Micro Powder Imprinting (μPI) Method, *Advanced Science Letters* (2012), **12**: 170-173.

[5] Tsumori, F. et al., Micro Imprinting Process for Surface Patterning of Ceramic Sheet, *Proc. Powder Mettallurgy World Congress* (2013), P-T6-72.

[6] Xu, Y. et al., Improvement of Solid Oxide Fuel Cell by Imprinted Patterns on Electrolyte, *Proc. IEEE-NEMS* (2013) 887-890.

[7] Xu, Y., Tsumori, F. et al., Effects of Resist Thickness and Viscoelasticity on the Cavity Filling Capability in Bilayer Thermal Embossing, *Jpn. J. Apl. Phys.* (2011), **50**: 06GK11.

[8] Xu, Y., Tsumori, F et al., Improvement of Solid Oxide Fuel Cell by Imprinted Patterns on Electrolyte, *Proc. IEEE-NEMS* (2013) 887-890.

[9] Faircloth, B. et al., Bilayer, nanoimprint lithography, *J. Vac. Sci. Technol. B* (2000), **18-4**: 1866-1873.

[10] Konishi, T. et al., Multi-layered resist process in nanoimprint lithography for high aspect ratio pattern, *Microelectronic Engineering* (2006), **83-4-9**: 869-872.

[11] Xu, Y., Tsumori, F et al., Improvement of Solid Oxide Fuel Cell by Imprinted Patterns on Electrolyte, *Micro & Nano Letters*, (submitted).