Title	ナノインプリント法による機能性材料の微細パターニングに関する研究
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Citation	
Issue Date	2016-03
Type	Thesis or Dissertation
Text version	ETD
URL	http://hdl.handle.net/10119/13530
Rights	
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Study on fine patterning of functional materials by nanoimprint method

Abstract

A great attention has been paid to printing technologies to fabricate various precise devices. Since the printing method patterns devices directly from functional inks, the process becomes very simple with maximum efficiency in using of raw materials. Therefore, it has advantages over conventional processes including vacuum deposition and photolithography in terms of production cost and environmental burden. However, the printing technologies developed until now have serious issues in resolution. Namely the smallest pattern size by printing is limited more than 10 μm.

Nanoimprint method, including nanoimprint lithography (NIL) and nano-rheology printing (nRP), is kind of printing methods and is an approach for solving the problem. NIL method can form the resist pattern for the patterns of functional materials, while nRP can form the patterns of them directly. Recently in Shimoda Lab., it was succeeded to pattern the RuO electrodes by nRP. RuO exhibited high conductivity. It is tried to make a fine pattern of RuO by the same method but only confirmed that breaking of the patterns happened during sintering because of its crystallization and grain growth. To avoid the breaking due to crystallization, an amorphous material is desirable to be use. A candidate of the amorphous material which has also a high conductivity would be Lanthanum Ruthenium Oxide (LRO).

In this study we reported that plastic deformation ability was greatly improved by adding La to Ru so as to allow forming a nano-sized gel pattern by n-RP. The gel pattern was successfully converted to an amorphous LaRuO without breaking during sintering. Moreover, we investigated the structures of solutions, gels and solids and conversion process from solution to solid via gel to understand why LRO has a high plastic deformation ability in nRP.

It became apparent the developed La-Ru solution was a solution in which La solvents and Ru ones independently dispersed by the mass spectrum analysis. Therefore a gel ought to have a same kind of structure: a mechanical mixture of La clusters and Ru clusters. One of the most impressing feature among these gels was that the La-gel showed a strong molecular crystallinity in the temperature range ca. from 100 to 200°C, which was made clear by the XRD analysis. That means La cluster in a solution can be stabilized by a cohesion energy derived from the molecular crystallinity. That in turn rises the desorption temperature of residual organic elements from a gel.

The gel thus analyzed was thermally imprinted in the nRP process. In imprinting at 175°C, the imprinting ability of Ru-gel was greatly decreased while the other LRO and La gels showed clear imprinted patterns. This result indicated that addition of La element to Ru one was able to improve the imprinting property. That was also confirmed by the viscoelastic measurement: the tan δ of LRO-gel was greatly enhanced compared with that of the Ru-gel.

After sintering the gel, the structure and conductivity of a solid film were investigated. It was confirmed that both the films of La-O and Ru-O were crystals and those of LaRuOs were amorphous. It was found that the more La element was added, the higher the resistivity. But the decrease of conductivity is relatively small up to 33% of the La addition.

Based on the result above, we succeeded to make a fine line pattern of the LRO-gel at the ratio of La/Ru = 25/75 by the nRP, which was able to be sintered without any breakings. The resultant line had a width of 30 nm with a good conductivity. This fine patterning was caused by the following two reasons: retaining organic elements to higher temperature in a La containing gel because of its nature of molecular crystallinity and amorphous structure of LaRu solid which prevents it from spontaneous breaking during sintering.

Keywords: nanoimprint, direct nanoimprint, printed electronics, solution process, lanthanum ruthenium oxide