**Lifetime Prolongation of Release Agent on Antireflection Structure Molds by Means of Partial-filling Ultraviolet Nanoimprint Lithography**

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**Abstract**—Release agent becomes an imperative element in ultraviolet nanoimprint lithography (UV-NIL) for preventing the adhesive resin from adhering to the surface of antireflection structures (ARS) mold. However, complete filling of the resin of a high-aspect-ratio ARS mold during UV-NIL generates a strong release force (RF) that deteriorates the release agent and shortens the lifetime of the ARS mold. In this paper, we proposed a technique of partial-filling UV-NIL in order to reduce the RF and consequently, prolong the lifetime of the release agent on ARS mold. The release and optical properties of the ARS were measured to determine the lifetime of the release agent on the mold, and complete-filling UV-NIL was also executed for comparison. By means of partial-filling UV-NIL, we successfully fabricated ARS films with excellent performance up to 75th imprint compared to complete-filling UV-NIL up to the 40th imprint.

**Keywords**—ultraviolet nanoimprint (NIL); lifetime; partial filling; release force; contact angle

**I. INTRODUCTION**

Mass fabrication of antireflection structure (ARS) films by ultraviolet nanoimprint lithography (UV-NIL) [1] is highly demanded for the rapid growth of flexible electronic fields. Examples of such application include flat-panel displays (FPDs), wherein antireflection (AR) structures are used to improve the visibility of the screen, and solar cells, where AR structures are used to enhance the energy absorption efficiency. In UV-NIL, release agent becomes an imperative element to impede the adhesion of resin on top of ARS mold. However, prolonging the lifetime of release agent on ARS mold surfaces is a challenge for their mass fabrication. This is owing to the degradation of release agent by mechanical and chemical factors. Tada et al. reported that during repeated UV-NIL, the release agent is mechanically removed [2]. Moreover, Truffier-Boutry et al. reported that the strikes of radicals to the release agent decreased its density. The radicals are generated from UV curable resin during ultraviolet light exposure of UV-NIL [3].

Our previous investigation [4] showed that complete filling of the resin for a high-aspect-ratio mold results in a strong release force (RF), which degrades the release agent gradually and consequently shortens the lifetime of the ARS mold. We also discussed the correlation between the resin filling behavior and the durability of the release agent on the mold at various filling pressures [5]. Then, by considering the concept of partial filling in rheology polymer [6, 7], we assumed that partial filling the resin during UV-NIL can reduce the RF and lower the aspect ratio of the replicated ARS film. Thus, prolongs the lifetime of the release agent on ARS mold.

**II. RESIN FILLING CONCEPT**

The idea of partial-filling UV-NIL is associated with the presence of capillary force ($P_c$) that acts on the substrate owing to the difference of surface energy and the formation of fine nanostructures [8, 9] that affects the filling behavior. The needle-like ARS shape in our study is identical to the shape of the capillary, thus, the equation of $P_c$ that we used in explaining the phenomenon of resin fillings as following [10];

$$P_c = 2\gamma\cos\theta/a$$  \hspace{1cm} (1)

Here, $a$ is pitches of the needle-like AR structures, $\gamma$ is the surface tension of the resin, and $\theta$ is contact angle (CA) in the capillary. The illustration of resin filling behaviors of ARS mold is explained in Fig. 1.

**Fig. 1.** Resin filling behaviors of uncoated and coated molds with release agent: (a) positive value of capillary force that attracts resin filling in hydrophilic ARS mold denotes as $P_c$; (b) negative value of $P_c$ that against the resin filling in hydrophobic ARS mold denotes as $P_c$.  

In normal phenomena, with an uncoated hydrophilic ARS mold (Fig. 1(a)), the positive values of $\cos\theta$ and the narrow pitches of the needle-like ARS structures generates a positive value of $P_c$ for pulling the resin downward. Otherwise, when