Nanoimprinted Deposition Masks for Area Selective Atomic Layer Deposition of Aluminum Oxide

C. Miyajima, S. Ito, M. Nakagawa
IMRAM, Tohoku University, Sendai, Miyagi 980-8577, Japan
masaru.nakagawa.c5@tohoku.ac.jp

Area selective deposition (ASD) has received much attention as a bottom-up micro- and nano-fabrication method for functional materials. Atomic layer deposition (ALD) is one of the promising technologies used for ASD to form inorganic oxide layers on substrates by mutual doses of an organometallic precursor and oxidant. ALD process for polymer resist thin films caused an organometallic precursor of trimethylaluminum (TMA) to infiltrate into the thin film near the surface.[1] We took a look of the difference in formation of aluminum oxide in an ALD manner between bare inorganic substrate and organic resist surfaces. In this study, we investigated whether ALD-modified UV-cured resin thin films could be removed completely by respective physical and chemical dry etching procedures or their combination.

A UV-curable liquid of NL-SU1[2] composed of two-kind dimethacrylate monomers was used for UV nanoimprinting. The UV-cured films on Si substrates were modified by oxygen reactive ion etching (O_2 RIE) and 5-cycle ALD with TMA and H_2O.[3] Substrate surfaces after physical Ar ion milling and chemical O_2 RIE[4] were analyzed by atomic force microscopy (AFM), time-of-flight secondary ion mass spectrometry (TOF-SIMS), and FE-SEM. A UV-cured film without ALD was used as reference for surface analysis.

AFM revealed the presence of some residues after either physical or chemical dry etching of ALD-modified UV-cured films. Total counts of secondary ions detected for Si surfaces after dry etching procedures were summarized in Table 1. Organic C_4H^- and inorganic AlO_2^- ions were detected after physical Ar ion milling, while only inorganic AlO_2^- ions were detected after chemical O_2 RIE. Interestingly, these organic and inorganic ions were hardly detected after Ar ion milling and subsequent O_2 RIE. No residues were observed on substrate surfaces by AFM after the sequential etching procedures. These strongly suggested that the sequential etching procedures completely ALD-modified UV-cured films. It was considered that an inorganic-rich layer near the surface could be etched by Ar ion milling and then an organic layer was removed by O_2 RIE. Taking the result into consideration, we propose the area selective-atomic layer deposition using nanoimprint deposition masks as illustrated in Fig.1. FE-SEM images (Fig. 2) supported that an alumina layer formed by 5-cycle ALD were selectively deposited on Si substrate surfaces unmasked with nanoimprinted patterns.

---

[1] M. Nakagawa et al., *J. Vac. Sci. Technol. B*. **36**, 06JF02 (2018).
[2] T. Uehara et al., *J. Photopolym. Sci. Technol*. **29**, 201 (2016).
[3] Y. Ozaki et al., *Jpn. J. Appl. Phys.* **57**, 06HG01 (2018).
[4] T. Uehara et al., *Bull. Chem. Soc. Jpn*. **91**, 178 (2018).

Table 1. Total counts of secondary ions detected for Si substrate surfaces after each dry etching by TOF-SIMS. $SiHO_3^-$, AlO_2^-, and C_4H^- were derived from Si substrate, aluminum oxide, and resist, respectively.

ALD cycle	dry etching	total count		
		$SiHO_3^-$	AlO_2^-	C_4H^-
5	O_2 RIE	8.3×10^5	3.2×10^4	2.2×10^4
5	Ar ion milling	4.6×10^5	1.6×10^4	1.2×10^5
5	Ar ion milling, subsequent O_2 RIE	8.6×10^5	9.0×10^2	2.1×10^4
0 (ref.)	O_2 RIE	6.8×10^5	9.4×10^2	1.2×10^4

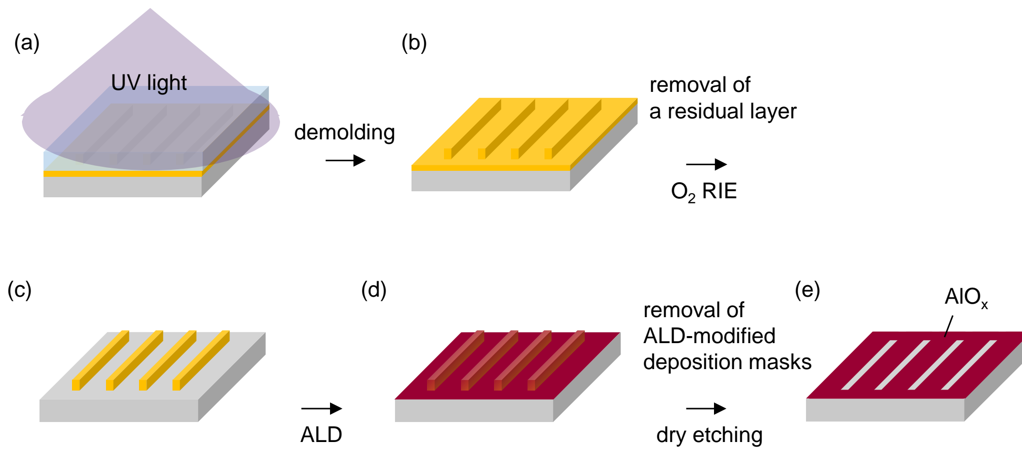

Figure 1: Schematic illustrations of area selective atomic layer deposition of aluminum oxide using UV-nanoimprinted deposition masks after removal of a residual layer.

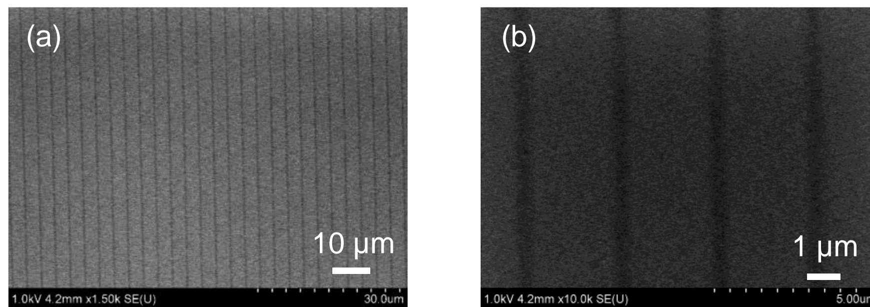

Figure 2: (a) Wide (1.5k) and (b) magnified (10k) FE-SEM images of selectively deposited aluminum oxide by 5-cycle ALD using 500 nm-linewidth imprint deposition masks.