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Effect of Oxygen Contents on the Property of Hydrophobic Thin Films Deposited on Flexible Substrates Using Plasma-enhanced CVD

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Abstract--Silicon-containing thin films were deposited on flexible substrates via plasma-enhance CVD (PECVD) using a raw material of tetramethylsilane (TMS) and an additive oxygen precursor. The existence of hydroxyl group prepared with an additive oxygen gas was responsible for the decrease of hydrophobic properties. However, the associated film hardness and surface roughness were apparently improved at an adequate mixing TMS and oxygen precursor.

1. Introduction

Recently, flexible substrates had been comprehensive used in optoelectronic devices [1-2]. However, they were poor in the hardness and permeation of gases or vapors. Silicon-containing thin films deposited on flexible substrates were good candidates to solve these weaknesses [3]. Plasma-enhanced chemical vapor deposition (PECVD) was a promising technology to provide these functional films. In our previous report, a hard coating with hydrophilic property was achieved using novel TMS raw material and oxygen precursors [4]. Except for the approach for the improved hardness on flexible substrates, the effort to prevent from vapor permeation was also demanded for the consideration of the related optoelectronic device reliability.

In this paper, TMS monomer was employed as the precursor to prepare a water repellent thin film onto flexible substrates. The effect of the additive oxygen precursor on the fabricated films was evaluated from properties of the hydrophobic, surface uniformity, and hardness.

2. Experiment

Silicon-containing thin films were deposited using a PECVD system as shown in Fig. 1. The organosilicon compound of TMS was liquid at room temperature and sealed in a steel vessel as shown in the photograph in Fig. 1. To investigate the effect of the additive oxygen precursor on the fabricated thin films, the gas flow ratios of TMS/O₂ were varying from 60/0 to 60/120 sccm. Cleaned polymethylmethacrylate (PMMA) and polycarbonate (PC) plates were used as substrates.

Film thickness was measured using a surface profile system (Dektak 6M). Hydrophobic properties were evaluated with a contact angle meter (FTA125 Contact Angle Analyzer). The chemical bonding states were investigated using a Fourier transformation infrared spectrometer (FTIR, JASCO-410). The related surface morphologies and hardness were conducted from an atomic force microscopy (AFM, DI3100) and a NanoTest 550 (Micro Materials Ltd., UK) instrument.



Fig. 1 Configuration of PECVD system

3. Results and discussions

In this study, the optimal deposition conditions concerned about the water contact angle performance using TMS monomer was firstly developed. A superior water repellent film was obtained at a gas flow rate of 60 sccm, rf power of 30 W, and deposition pressure of 27 Pa, respectively. Sequentially, the evolutions of film properties depended on the additive oxygen gas flow rate varied from 15 to 120 sccm was incorporated with TMS monomer. Figure 2 shows evolutions of the water contact angles of the thin films prepared at various TMS/O₂ gas flow ratios. The water contact angle prepared with TMS monomer was higher than 103° while that of PMMA and PC substrates were about 65° and 73°, respectively. The water contact angle was decreased with increasing oxygen precursor. The photographs of water drops on films at TMS/O₂ gas flow ratios of 60/0 and 60/120 sccm as well as the uncoated PC substrate are shown in Figs. 3(a)-(c). They were found to evolve from hydrophobic to hydrophilic property. This indicated that chemical compositions and structures had been transformed as introduced oxygen precursor. The associated chemical bonds on the film surface prepared at various TMS/O2 gas flow ratios observed from FTIR



Fig. 2 Evolutions of the water contact angles prepared at various TMS/O₂ gas flow ratios.



Fig. 3 Photographs of water drops of the films prepared at TMS/O₂ gas flow ratios of (a) 60/0, (b) 60/120 sccm, and (c) uncoated PC substrate.

measurements are shown in Figs. 4(a) and 4(b). In the FTIR spectrum of the film prepared from TMS monomer, the absorbance band due to Si-C deformation vibration in the Si-CH₃ group was detected at 1270 cm⁻¹. In addition, the absorbance bands appeared at 2900 and 2925 cm⁻¹ were identified as C-H related bond in the asymmetric stretching of -CH₃ group. These bonds were reported to present the hydrophobic properties and resulted in water repellency [5]. However, they were disappeared as the films prepared using additive oxygen precursor. This revealed that -CH₃ groups originated from TMS raw material had been decomposed due to the appearance of the additive oxygen precursor and replaced by the stretching bond of Si-OH appeared at around 2800 - 3700 cm⁻¹ and 940 cm⁻¹. Since the hydroxyl group performed hydrophilic properties [6], the associated water contact angles were thus obviously decreased.



Fig. 4 FTIR spectra of the film surface prepared at (a) TMS monomer and (b) various TMS/O₂ gas flow ratios .

The hardness and surface roughness of these films deposited on PC and PMMA substrates are shown in Figs. 5(a) and 5(b). As incorporated with oxygen precursor, the surface roughness was decreased with increasing oxygen contents. Therefore, the additive oxygen precursor was able

to introduce the oxygen atoms in the films and result in a flattened surface. In terms of the film hardness, the film hardness showed an opposite trend to the surface roughness. The smoothest surface roughness corresponded to a hardest one. The superior hardness was originated from the fabricated films being gradually close to stoichiometric SiO_2 derived from the chemical bond position of Si-O-Si shown in Fig. 4(b). In addition, the surface uniformity and hardness was degraded again at higher oxygen precursor due to the damage originated from excess oxygen gas.



Fig. 5 Surface roughness and hardness evolutions for the films deposited on (a) PC and (b) PMMA substrates at various TMS/O₂ gas flow ratios.

4. Conclusions

A water repellent film possessed a contact angle higher than 100° was achieved using TMS monomer. The obvious -CH₃ group was responsible for the significantly hydrophobic property. In addition, the hydrophilic performance was a consequence of the appearance of hydroxyl group as introduced the additive oxygen precursor. The surface roughness and hardness was improved due to the increasing oxygen contents in these fabricated films. As a result, by altering the deposited TMS/O₂ gas flow ratios, a multi-functional thin film structure such as water repellent films, hydrophilic films, and hard coatings is promising for the package of the optoelectronic devices employed flexible substrates.

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