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N-doped Al_2O_3 thin films deposited by atomic layer deposition

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ABSTRACT

The present study focused on nitrogen doped Al_2O_3 thin films using atomic layer deposition, varying the deposition temperature from 55 to 170 °C. Al_2O_3 thin film growth rate and electrical properties were mostly dependent on deposition temperature. Nitrogen concentration decreased from 2.7 to 2.4% with increasing deposition temperature. X-ray photoelectron spectroscopic analysis confirmed that nitrogen doping in Al_2O_3 decreased formation of oxygen related defects, including non-lattice oxygen. Surface morphology analyses also showed that N-doping reduced Al_2O_3 film surface roughness. Reduced oxygen related defects significantly reduced leakage current by 1000 times when comparing with as-deposited films. Minimum leakage current (5×10^{-10} A/cm^2) was observed for N-doped Al_2O_3 film deposited at 170 °C and post-annealed at 400 °C, including a decrease by 10 times through N-doping.

1. Introduction

Metal oxide thin films have been employed in many different fields, including optoelectronics, microelectronics, optical sensors, and integrated circuits [1–4]. Silicon oxide (SiO_2) has been the most widely used dielectric material in semiconductor integrated circuits. However, SiO_2 thin films exhibit high leakage current due to capacitance problems arising from SiO_2 low dielectric constant according. On the other hand, if high *k* material is used as the gate dielectric, even if it has the same capacitance it can be applied as a thick film to solve the leakage problem. Hence, a variety of high *k* metal oxides, including Al_2O_3, Ta_2O_5, Nb_2O_5, and HfO_2, have been investigated to replace SiO_2. High *k* oxide materials in integrated circuits also require low leakage current density, high dielectric breakdown voltage, and high capacitance density [5,6].

Al_2O_3 is one of the most promising SiO_2 alternatives due to its high permittivity (~9) compared with SiO_2 (~4), excellent thermal stability, and wide energy bandgap (8.3 eV) [7–10]. Various deposition techniques have been used to prepare Al_2O_3 thin films, including atomic layer deposition (ALD), vacuum evaporation, pulsed laser deposition, chemical vapor deposition, sol–gel processing, magnetron sputtering, and molecular beam epitaxy, etc. [11–14]. Among these, ALD easily controls film thickness and composition because it uses an independently pulsed precursor and the film surface has limited reaction. Thus, many studies have reported ALD synthesized Al_2O_3 thin films using trimethyl-aluminum (TMA) and H_2O with 2–3 fold lower leakage current than thermal SiO_2 films with the same equivalent oxide thickness [15–23]. However, the interface between Si and Al_2O_3 generates diffusion and electron trapping, which increases leakage current compared with thermal oxide films [24,25]. Anion doping to Al_2O_3 using ALD is a potential solution to overcome this problem, decreasing defects and reducing the number of traps. Oxygen diffusion probability is also reduced and disturbance created at the interface.

Recent studies have compared electrical properties of plasma enhanced ALD (PEALD)-Al_2O_3 film with nitrogen gas to films grown without nitrogen gas, and leakage current of Al_2O_3 films grown by PEALD has been compared with films grown by remote plasma ALD [26–28].

The current study synthesized N-doped Al_2O_3 thin films using ALD. Low growth temperature is an important factor in ALD to prepare temperature sensitive thin films compared with other methods. We also investigated the impact of nitrogen doping on ALD deposited Al_2O_3 thin film electrical characteristics.

2. Experimental procedures

Pristine and N-doped Al_2O_3 thin films were deposited on p-type Si (100) wafers at various deposition temperatures (55, 70, 90, 110, 130,

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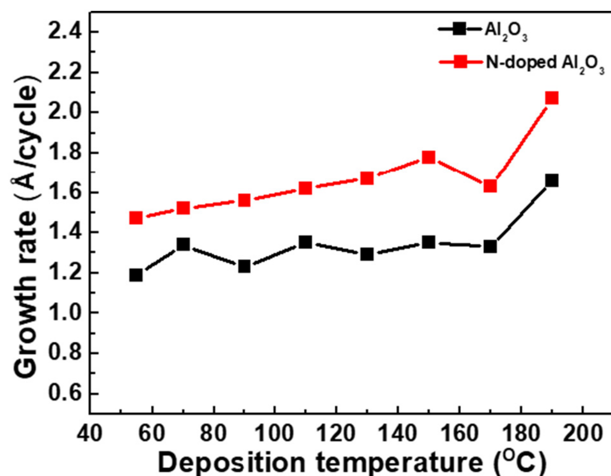

Fig. 1. Al_2O_3 film growth rate after 100 reaction cycles on Si (100) substrates.

150, 170, and 190 °C) with working pressure approximately 0.1 kPa via ALD using a traveling wave type Lucida D100 system (NCD Tech, Inc. Korea) [18–22]. TMA (Ezchem Co., Ltd., Korea) and deionized water were used as precursors for Al and O reactants, respectively [29,30], with nitrogen carrier gas. Ammonia water (28%) was used to dope nitrogen in the Al_2O_3 thin films.

ALD growth cycles for pristine Al_2O_3 thin films progressed as: TMA pulse (0.1 s) → N_2 purge (8 s) → H_2O pulse (0.1 s) → N_2 purge (8 s). Few studies have considered nitrogen doping in Al_2O_3. Therefore, we develop a strategy to optimize the number of ALD steps to input nitrogen by controlling deposition temperature, hence minimizing oxygen deficiencies.

ALD growth cycles for pristine N-Al_2O_3 thin films progressed as: TMA pulse (0.1 s) → N_2 purge (8 s) → NH_4OH (0.1 s) → N_2 purge (8 s) → H_2O pulse (0.1 s) → N_2 purge (8 s). After finalizing the number of nitrogen steps to maximize nitrogen concentration, deposition temperatures were optimized to compare pristine Al_2O_3 to N-doped Al_2O_3 thin films. Nitrogen concentration was 2.7 at.% in N-doped Al_2O_3 thin films. Three samples were chosen with 55, 130, and 170 °C deposition temperatures, annealed for 30 min at 400 °C, and used for further analysis.

Prepared film thickness was measured by ellipsometer (Gatan, L117C, 632.8 nm He-Ne lasers), and film density calculated using the Lorentz-Lorenz equation. Surface morphologies were investigated using atomic force microscopy (AFM, MultiMode 8, Bruker, USA). X-ray photoelectron spectroscopic (XPS) analysis was performed using an Al Kα monochromator (1486.6 eV) with variable spot size (30–400 µm in 5 µm steps) to determine the elements present in the prepared thin films (XPS, K-alpha, Thermo UK) at the 8A2 beamline of the Pohang Accelerator Laboratory. Electrical performance was obtained via current-voltage measurement at room temperature.

3. Results and discussion

Fig. 1 shows Al_2O_3 growth rate in ALD as measured by spectroscopic ellipsometry. Pristine Al_2O_3 thin films showed similar thickness (approximately 11–12 nm) as deposition temperature increased from 55 to 170 °C after 100 reacting cycles, and approximately 16 nm at 190 °C deposition temperature. This implies Al_2O_3 growth rates of 1.2–1.3 Å/cycle up to 170 °C and 1.6 Å/cycle at 190 °C [31]. N-doped Al_2O_3 thin

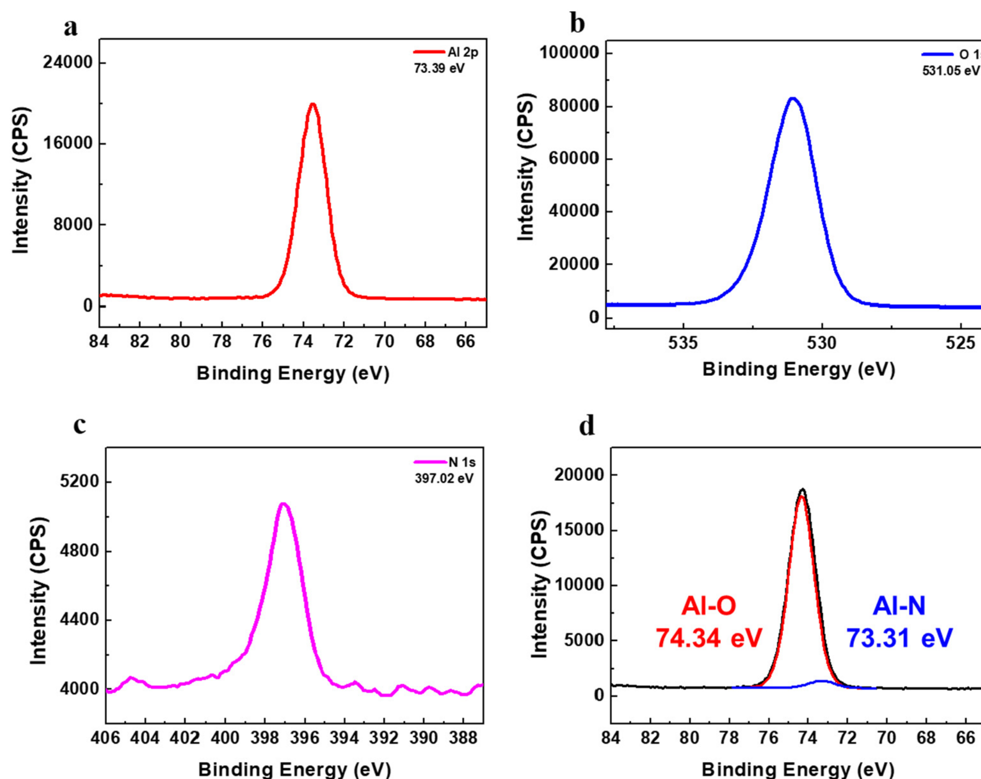

Fig. 2. XPS narrow scan spectra of N-doped Al_2O_3 thin films deposited at 55 °C and annealed at 400 °C: (a) Al 2p, (b) O 1s, (c) N 1s, and (d) deconvolution of Al 2p spectrum.

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