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Annealing effect on physical and electrical characteristics of thin HfO_2, $HfSi_xO_y$ and HfO_yN_z films on Si

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ABSTRACT

We report the effect of annealing on electrical and physical characteristics of HfO_2, $HfSi_xO_y$ and HfO_yN_z gate oxide films on Si. Having the largest thickness change of 0.3 nm after post deposition annealing (PDA), HfO_yN_z shows the lowest leakage current. It was found for both as-grown and annealed structures that Poole–Frenkel conduction is dominant at low field while Fowler–Nordheim tunneling in high field. Spectroscopic ellipsometry measurement revealed that the PDA process decreases the bandgap of the dielectric layers. We found that a decreasing of peak intensity in the middle HfO_yN_z layer as measured by Tof-SIMS may suggest the movement of N toward the interface region between the HfO_yN_z layer and the Si substrate during the annealing process.

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1. Introduction

Aggressive downscaling of CMOS circuits produces high leakage current due to direct tunneling of electrons through the gate SiO_2 layer (thickness <1.5 nm). According to the International Technology Roadmap for Semiconductors (ITRS), to keep the equivalent oxide thickness with low leakage current in the gate oxide, alternative high dielectric constant (high-*k*) oxides such as ZrO_2, HfO_2 and Ta_2O_5 have been considered as possible candidates for further downscaling [1–4]. As the best candidate for gate oxide application, HfO_2 has a high dielectric constant (ranging from 20 to 49 for the monoclinic, cubic and tetragonal phase [5–7], a large band gap with sufficient band offset (>1.5 eV) and is thermally stable in contact with silicon substrates [8,9].

The main drawback of HfO_2 is the low crystallization temperature ($T < 400$ °C) which can promote crystal boundaries within the layers to act as leakage paths and impurity getters during processing. Therefore it is an important issue to increase the kinetic stability while keeping the advantages of HfO_2 for its adoption into semiconductor manufacturing. The first method to increase the crystallization temperature of HfO_2 is to form a silicate of HfO_2, but then the dielectric constant decreases [10]. The other option is to form an aluminate layer of HfO_2 which has a rather high dielectric constant due to Al_2O_3 itself being a high-*k* material [11].

Up to date, however, few publications show the annealing effect on the electrical performance of the dielectric layer and especially the elemental depth profile in the layer. This report presents the effect of annealing on the electrical and physical characteristics of Hf-based gated oxides.

2. Experimental procedure

HfO_2, $HfSi_xO_y$ and HfO_yN_z films with thicknesses around 4 nm were deposited on HF cleaned 12″ n-type Si substrates by atomic layer deposition (ALD). Tetrakis(ethylmethylamino)-hafnium (TEMA-Hf) and Tetrakis(ethylmethylamino)-silicon (TEMA-Si) plus ozone were used as precursors. Ion implantation, reported to be successfully integrated into a traditional CMOS process of N_2^+ in ALD was introduced to form the HfO_yN_z layer. After deposition, in order to investigate the annealing effect on Hf-based gate oxides, one group of samples was subsequently annealed at 1000 °C, 10 s in N_2 ambient. In order to facilitate electrical characterization, top electrodes made up of 400 nm thick Al dots with different diameters – 350, 600 and 1300 μm – were deposited by sputtering on both the as-grown and annealed samples. The electrical properties were characterized by a semiconductor parameter analyser (HP 4156 A). Capacitance–voltage measurement was performed

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by a precision LCR meter (HP 4284A) with parallel mode (Cp) at 1 kHz. Structural investigation was done by Transmission Electron Microscopy (TEM, FEI TECNAI F30). To obtain detailed information about the annealing effect on the dielectric layer as well as the interfaces between layers, we used time-of-flight secondary ion mass spectrometry (ToF-SIMS, ION-TOF-V) with a 1 keV Cs^+ primary ion beam at incident angle of 45° for elemental depth distribution and the interface composition.

3. Results and discussion

The leakage current plot as a function of applied voltage of as-grown samples is shown in Fig. 1. The leakage current was obtained under electron injection from the Si substrate. For all as-grown samples the leakage current level is similar up to an applied voltage of −2.5 V while it becomes clearly different as the applied field is higher than −2.5 V. The highest leakage current was observed for the HfO_2 sample in the whole range of applied fields. The $HfSi_xO_y$ and HfO_yN_z samples show a lower leakage current, which indicates Si and N incorporation into HfO_2 has a role to decrease leakage current due to their high conduction band (CB) offset of 3.2 and 2.4 eV for SiO_2 and Si_3N_4, respectively. Robertson [12] reports that the CB offset of $HfSiO_4$ is 1.8 eV, which is between that of HfO_2 (1.4 eV) and SiO_2. The inset in Fig. 1 shows the two regions of different leakage current mechanisms of the HfO_yN_z sample: Trap assisted tunneling (TAT) and Poole–Frenkel (P–F) conduction in low electric field were observed [13] while Fowler–Nordheim (F–N) tunneling was dominant at high field. From the P–F plots in ln (J/E) vs. $E^{1/2}$, the trap depth ϕ_{P-F} of as-grown HfO_2, $HfSi_xO_y$ and HfO_yN_z samples was 0.57 eV, 0.61 eV and 0.59 eV, respectively. It known that Si or N incorporation into HfO_2 increases the trap depth of dielectric layers [5].

Fig. 2 presents the annealing effect on leakage current of Hf-based oxide samples. The leakage current of all annealed samples at −1 V is more than first order of magnitude higher than that of the as-grown samples. The leakage current mechanism is similar to the as-grown samples in Fig. 1, while the transition point from P–F to F–N occurs at higher field.

Since the leakage current is related to the physical thickness and the band gap (BG) of a layer, we measured the thickness change and bandgap of as-grown and annealed samples by variable angle spectroscopic ellipsometry (Woollan VASE M-2000, spectral range 200–1000 nm). The measured data is summarized in Table 1. It is noteworthy that the biggest annealing effect on thickness

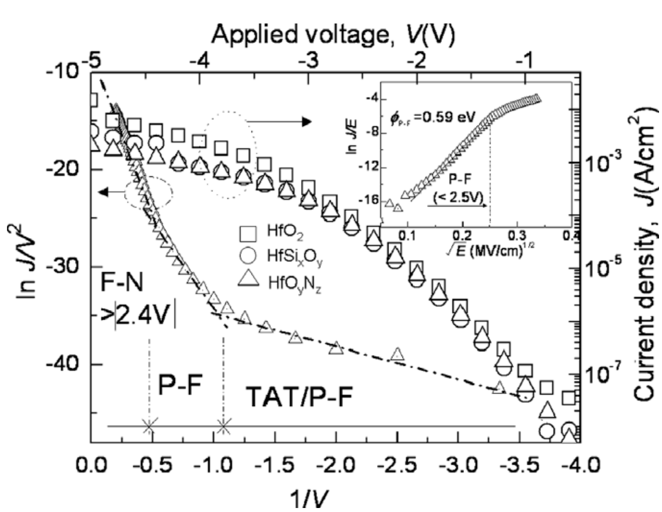

Fig. 1. Leakage current plot of as-grown HfO_2 based metal–insulator–semiconductor (MIS) structures. The inset presents the P–F plot of the HfO_yN_z sample.

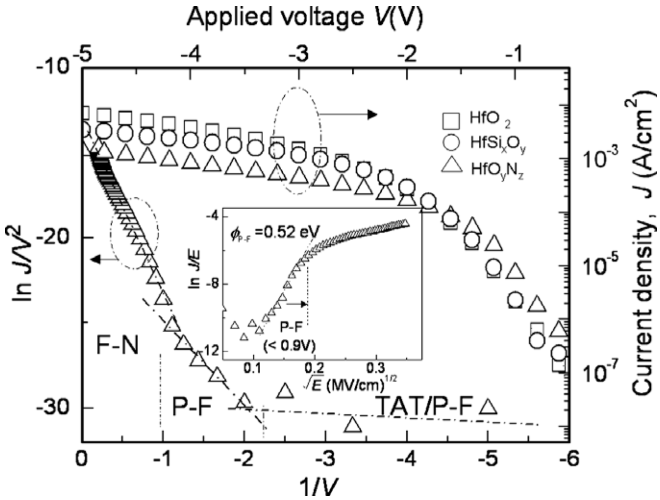

Fig. 2. Annealing effect on leakage current of HfO_2 based MIS capacitors. The inset shows the region of Poole–Frenkel conduction (<1.0 V) and Fowler–Nordheim tunneling in high field range (>1.1 V). The crossover point from P–F to F–N is lower than for as-grown samples.

Table 1
Annealing effect on physical thickness of Hf-based gate oxides measured by spectroscopic ellipsometry.

	HfO_2 (nm)	$HfSi_xO_y$ (nm)	HfO_yN_z (nm)
As-grown	4.35 ± 0.02	4.36 ± 0.02	4.45 ± 0.02
Annealed	4.25 ± 0.03	4.17 ± 0.03	4.15 ± 0.03

change was found in the HfO_yN_z sample. The dielectric functions of the films, which show a steep rise of absorption in the UV part of the spectrum, were parameterized by a Tauc-Lorentz-function. The bandgaps obtained from a fit of this parameterization to the ellipsometric data are 5.35, 5.40 and 5.38 eV for the as-grown HfO_2, $HfSi_xO_y$ and HfO_yN_z, respectively. Regarding the annealing effect on the bandgap, shown in Fig. 3, it is clearly revealed that the annealing process promotes the decrease of the bandgap for all samples in this study. Wang et al reported that the formation of Si-rich Hf-silicate stems from the linkage between SiO_2 and Hf precursor during the first HfO_2 monolayer formation [14]. Since Hf-silicate is thermally stable in contact with Si and HfO_2, silicate formation may occur during the deposition of HfO_2 and HfO_yN_z. Additional annealing would promote silicate formation in the interface layer. To elucidate this point further, we characterized

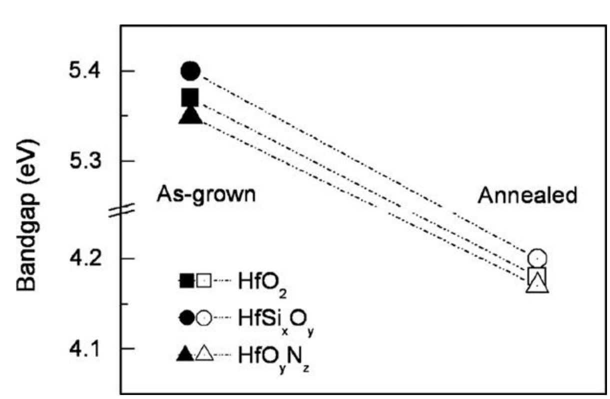

Fig. 3. Bandgap changes of as-grown and annealed HfO_2, $HfSi_xO_y$ and HfO_yN_z samples using variable angle spectroscopic ellipsometry (VASE).

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