

Epitaxial ALD BeO: Efficient Oxygen Diffusion Barrier for EOT Scaling and Reliability Improvement

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Abstract—In a previous study, we demonstrated that the BeO film grown by atomic layer deposition (ALD) on Si and III–V metal–oxide–semiconductor devices has excellent electrical and physical characteristics. In this paper, we discuss the physical and electrical properties of ALD BeO as an oxygen diffusion barrier on scaled 4-nm HfO₂/BeO gate stacks. Thin BeO layers are deposited onto (100) p-Si substrates as an alternative to SiO₂ as an interfacial passivation layer (IPL). X-ray photoelectron spectroscopy and transmission electron microscopy show that the BeO IPL acts as an effective oxygen barrier against SiO_x native oxide formation during postdeposition annealing (PDA). The use of ALD BeO as an oxygen diffusion barrier results in lower equivalent oxide thickness, more competitive leakage current, and better reliability characteristics after PDA than Al₂O₃ and HfO₂ gate stacks.

Index Terms—Atomic-layer-deposited (ALD) beryllium oxide (BeO), beryllium oxide (BeO) interfacial layer passivation (IPL), oxygen diffusion barrier.

I. INTRODUCTION

TO SCALE silicon-based metal–oxide–semiconductor field-effect transistors (MOSFETs), equivalent oxide thicknesses (EOTs) below 1 nm are required for high-performance technology nodes beyond 22 nm [1]. Therefore, scaling the interfacial passivation layer (IPL) between the high-*k* dielectric and Si channel plays an important role. As a possible solution, SiO₂ IPL thinning or scavenging was induced during source/drain dopant activation annealing by introducing an oxygen absorption layer within the gate stack [2], [3], and molecular beam epitaxy (MBE) grown SrO₂ was studied as

an IPL [4]. Many researchers have focused on amorphous high-dielectric-constant gate dielectrics such as ZrO₂ and HfO₂ for scaling THE EOT; however, sustaining their amorphous structure during high-temperature postdeposition annealing (PDA) is difficult, as evidenced in many investigations [5]–[8]. After thermal treatment, most ultrathin high-*k* dielectrics tend to become polycrystalline with significant grain boundary formation, thereby increasing the leakage current [8]. Crystalline oxides on Si and III–V have attracted interest due to their advantages over amorphous oxides, such as no recrystallization during high temperature treatments and a defined stoichiometry at the interface. There are several epitaxial oxides, such as LaLuO₃ [9], LaHfO₂ [10], CeO₂ [11], SrO₂ [12], and SrTiO₃ [13], but these are not thermally stable. They are also not efficient oxygen diffusion barriers and must be grown by MBE, which requires ultrahigh vacuum (UHV). These oxides, furthermore, have no self-cleaning reaction on III–V substrates [14].

In this paper, we present an alternative solution that consists of replacing SiO₂ with a thin and epitaxial high-*k* IPL. With *k* = 6.8, 10.6-eV band gap, 2.3-eV conduction band (CB) offset, and high thermal stability [15], atomic layer deposition (ALD) BeO is a promising candidate. ALD BeO layers are epitaxially grown on Si at low temperatures (200 °C ~ 250 °C) and pressures (0.2 ~ 0.3 torr) using dimethylberyllium and water precursors. This allows good control of the physical thickness and causes minimal oxidation of the Si surface. We will show that BeO IPL suppresses not only Si oxidation but also leakage current after PDA. The physical and electrical properties of devices fabricated with HfO₂/BeO (IPL) gate stacks will be discussed and benchmarked against HfO₂ without an IPL and HfO₂/Al₂O₃ (IPL) gate stacks.

II. FABRICATION PROCEDURE

MOS capacitors (MOSCAPs) were fabricated on p-type Si (100) with doping concentrations of approximately $5 \times 10^{17}/\text{cm}^3$. After 1% high-frequency dip surface cleaning, 5 ~ 10 Å BeO was deposited as an IPL at 250 °C using dimethylberyllium and H₂O as the precursors. As a reference, Al₂O₃ of a similar thickness was deposited by ALD using trimethylaluminum and H₂O as the precursors, and ALD HfO₂ (40 Å) was deposited on top of the IPL. PDAs from 600 °C to 900 °C were performed by rapid thermal annealing under N₂ ambient for 30–60 s. In these devices, physical-vapor-deposited TaN was used for the gate electrode.

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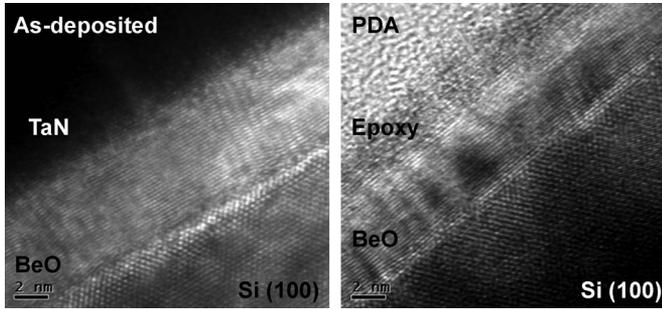


Fig. 1. High-resolution cross-sectional TEM analysis of TaN (or epoxy)/BeO/Si (a) without and (b) with 600 °C PDA. ALD BeO shows a layer-by-layer single crystalline structure and no significant interfacial native oxide growth after PDA.

III. RESULTS AND DISCUSSION

A. TEM Characterization

The transmission electron microscopy (TEM) image in Fig. 1 displays almost layer-by-layer single crystallinity and distinct interface for ALD BeO before and after PDA.

The crystallization of a nearly amorphous BeO film was shown to occur at 300 °C [16]. There is no significant interfacial native oxide growth after a 600 °C PDA. The etch rate of the buffered oxide etchant is much slower for BeO (14 ~ 15 Å/min) than for Al₂O₃ (> 100 Å/min). The high crystallinity of ALD BeO slows the permeation of etchant and also the diffusion of oxygen (or oxygen exchange between Si and the gate dielectric) during PDA.

B. Optical Characterization

Oxygen vacancies are the dominant origin of the CB offset ΔE_c and the energy band gap E_g lowering in bulk and thin films, and they may facilitate the diffusion of oxygen [17]. Therefore, a study of ΔE_c and E_g may offer an insight into the oxygen diffusion [17], [18]. ΔE_c of ALD BeO is still not explicitly known, although a second-harmonic generation (SHG) analysis was performed in our previous study [15]. Internal photoemission (IPE) analysis was therefore performed to provide further information about the interface characteristics (including ΔE_c) of the ALD BeO gate stack. ALD BeO displays a lower photocurrent than Al₂O₃ [see Fig. 2(a)]. The amorphous nature of the insulator results in a significant subthreshold IPE, indicative of oxide CB tailing [18]. However, the fundamental IPE threshold is essentially the same in both the crystalline and amorphous structures since the momentum conservation condition has no significant influence [18]. Therefore, extracting the IPE threshold from $Y^{1/p} - h\nu$ plots of crystalline BeO is a valid way of determining the interface barrier. From the IPE analysis shown in Fig. 2(a), one can infer that ALD BeO has a 0.4-eV-higher ΔE_c than ALD Al₂O₃, which is reported to be around 1.9 eV [19]. Thus, ALD BeO ΔE_c may be around 2.3 eV. Fig. 2(b) shows the SHG results for the 5-nm BeO (Al₂O₃) film on a p-Si substrate. Since the transition from three- to two-photon processes takes place at around 1.67 and 1.71 eV before and after PDA, respectively, BeO ΔE_c with a Si substrate is estimated to be 2×1.67 eV (1.71 eV)–1.1 eV (Si energy band gap) = 2.24 and 2.32 eV

before and after PDA, respectively. These results agree with the IPE results ($\Delta E_c = 2.3$ eV) in Fig. 2(a). More details about the SHG analysis can be found in [15]. Fig. 2(c) is the vacuum ultraviolet (VUV) spectroscopic ellipsometry results for the BeO (Al₂O₃) film on a p-Si substrate for a full energy band-gap analysis. For the data in Fig. 2(c), BeO E_g is estimated to be 7.87~7.91 eV. However, determining E_g is based on the linear extrapolation of the absorption energy $\gg E_g$, which we cannot access in that spectral region, causing uncertainty about the exact E_g . (The analysis in Fig. 2 requires an intensive investigation.) Fig. 3 illustrates the band diagram of HfO₂, HfO₂/Al₂O₃, and HfO₂/BeO IPL MOSCAPs. The BeO band structure was estimated from the analysis in Fig. 2. Because we have no capacity to measure the valence band spectrum, the valence band offset ΔE_v was calculated from E_g and ΔE_c . The band offsets ΔE_c and ΔE_v of BeO with Si are much greater than those of Al₂O₃ or HfO₂.

C. C-AFM Characterization

The nanostructure of the BeO thin film was not experimentally studied due to the absence of an application for the BeO thin film and the difficulties inherent in the evaporation method (the only method to grow BeO before ALD), which requires high thermal energy [20]. BeO possesses a unique combination of thermal, dielectric, and mechanical properties that are highly desirable in electronic applications [21]. Conductive atomic force microscopy (C-AFM) is particularly helpful for investigating semiconductor nanostructures, as well characterizing dielectric layers locally [22], [23]. Unlike traditional techniques that provide spatially averaged information of the microscopic phenomenon, C-AFM allows the dielectric film to be both topographically and electrically characterized with nanometer resolution. Using constant bias and contact modes, we have mapped a few cursory current images.

Fig. 4 shows C-AFM results on all 5-Å BeO, Al₂O₃, and HfO₂ samples. The physical thickness is calculated from the deposition rate. A backside (or substrate) voltage bias of 1 V was applied, and leakage current was monitored with a sensitivity of 10 nA/V. Fig. 4(a) shows that the topography of the image is smooth without any bumps or hollow surfaces. However, the current images in Fig. 4(b) display some isolated dark spots, suggesting current conduction through leakage paths [24]. The dark spots in the current image may be due to the low-level leakage current under a high electric field, generally known as stress-induced leakage current (SILC) [25]. High crystallinity and band offsets in Figs. 1 and 3 may suppress the leakage current and defect generation, resulting in few ALD BeO conduction paths (dark spots). Similar results, displaying many dark spots, were seen in the Al₂O₃ current image. (To explain this phenomenon, an intensive investigation for C-AFM is required.)

D. XPS Characterization

Ex situ X-ray photoelectron spectroscopy (XPS) analysis was performed in a UHV chamber equipped with a monochromatic Al K_α X-ray source.

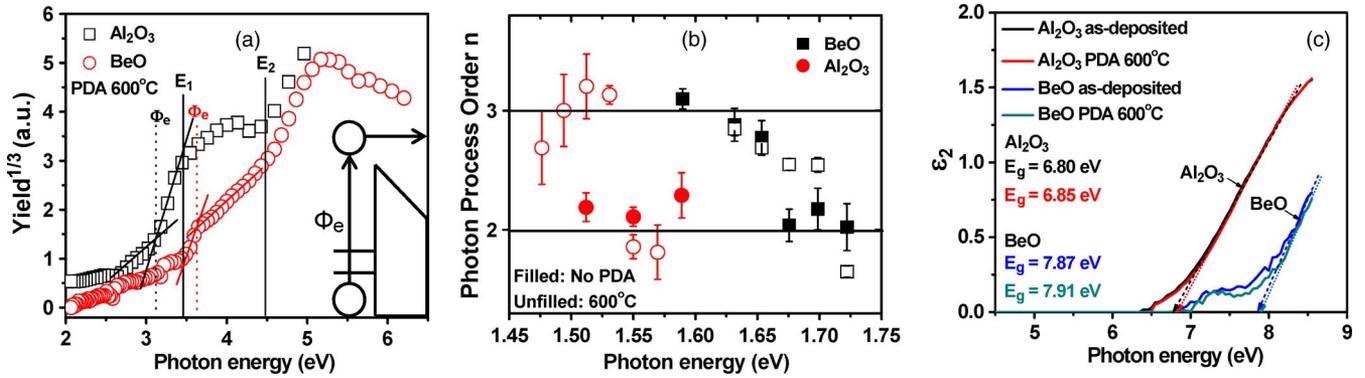


Fig. 2. (a) Spectra of electron IPE from the VB of silicon to the CB (cf. the inset) of crystalline BeO epitaxially grown on (100) Si as compared with amorphous film of Al₂O₃ on (100) Si, linearized in $Y^{1/3} - hv$ coordinates. (Lines) Linear interpolation procedure used to infer the electron energy barrier Φ_e . (b) SHG results of BeO (47.8 Å) and Al₂O₃ (48.5 Å) on p-Si(100). (c) VUV spectroscopic ellipsometry results for full-energy band-gap analysis.

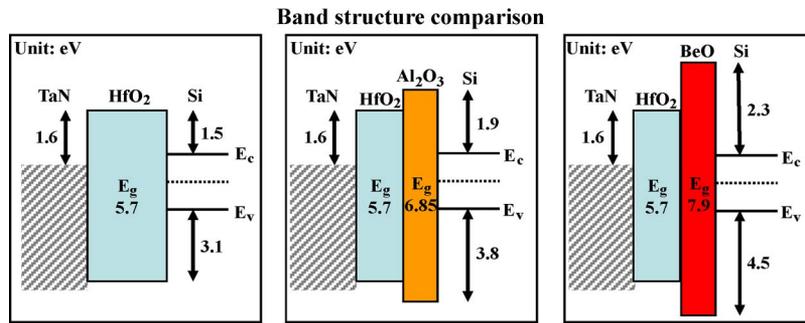


Fig. 3. Schematic band structures of HfO₂, HfO₂/Al₂O₃, and HfO₂/BeO gate stacks on Si substrates. The ALD BeO band structure was estimated from the analysis in Fig. 2.

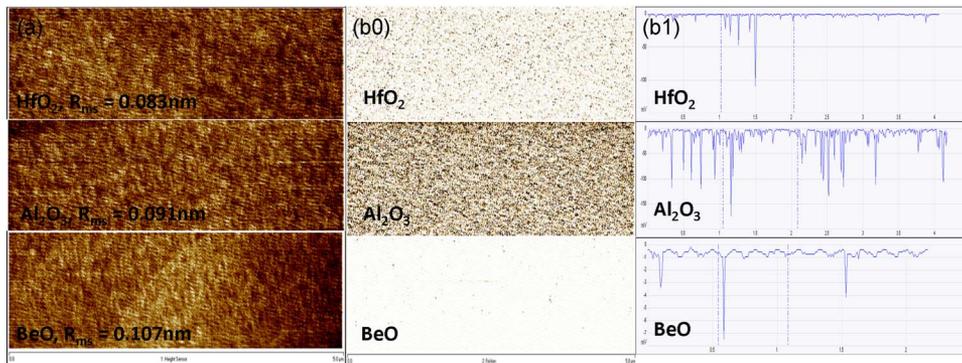


Fig. 4. (a) Surface roughness and (b0) current spots and (b1) line scan images at a 1-V backside bias for HfO₂, Al₂O₃, and BeO samples (all 5-Å physical thickness with a scan area of $5 \times 2.5 \mu\text{m}^2$) (b0) BeO displays fewer current leakage spots than the other oxides.

Fig. 5 shows the Si 2p XPS analysis for 15-Å BeO and Al₂O₃ samples, respectively. ALD Al₂O₃ is a more efficient and better known diffusion barrier layer than HfO₂ [26]. Accordingly, Al₂O₃ was studied to compare the performance of BeO and Al₂O₃ oxygen diffusion barriers. The binding energies of Al₂O₃ 2p and BeO 1s are 74~76 eV and 112~116 eV, respectively (see Fig. 5). The outermost valence band electrons, related to atomic cohesion, are Al 3p and Be 2s for Al₂O₃ and BeO, respectively. Therefore, one can infer that the binding energy of Be-O is higher than that of Al-O. As the bond distance decreases, binding energy increases, leading to a higher melting point for BeO (2507 °C) than for Al₂O₃ (2072 °C) [27]. Both BeO and Al₂O₃ bonds are almost covalent bonding [15]. The short bond distance causes more orbital splitting and results in a high-energy band gap, which indicates the suppression of

oxygen vacancies [28]. Hence, one can infer that BeO may be an efficient oxygen diffusion barrier, although many factors such as atomic networks must be still investigated. After a 900 °C PDA, as shown in Fig. 5, the Si-O peak in ALD BeO is 32% lower than the Al-O peak. These experimental results support the theoretical basis.

E. Electrical Characterization

Figs. 6–8 display the capacitance–voltage ($C-V$) and current–voltage ($I-V$) characteristics of HfO₂ (40 Å), HfO₂ (40 Å)/Al₂O₃ (5 Å), and HfO₂ (40 Å)/BeO (5 Å) gate stacks on p-Si substrates. In Fig. 6(a), ALD BeO has a good interface quality compared with the other gate stacks. In Fig. 6(b), the Al₂O₃ flat-band voltage V_{fb} increases as the IPL

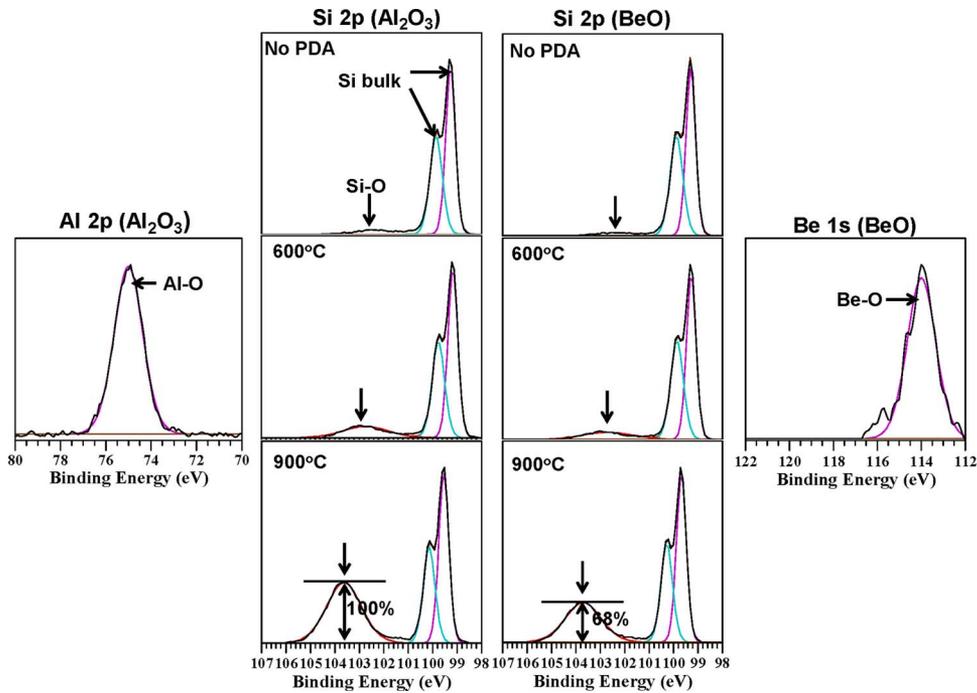


Fig. 5. XPS spectra for Si 2p of 15-Å (a) Al₂O₃ and (b) BeO gate stacks. The interfacial layer (Si–O peak) increases with increased PDA temperatures. However, BeO displays 32% lower interfacial native oxide signal than the Al₂O₃ case.

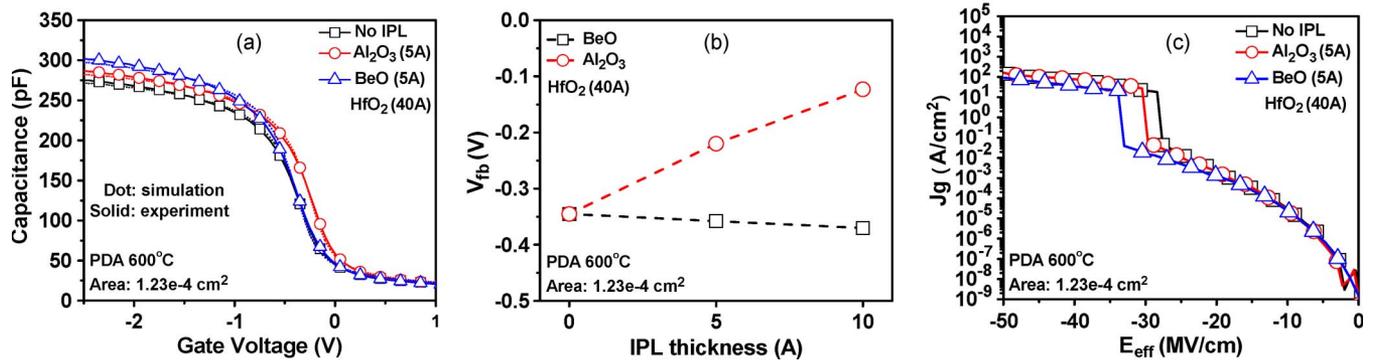


Fig. 6. (a) $C-V$, (b) V_{fb} , and (c) $I-V$ characteristics of HfO₂, HfO₂/Al₂O₃, and HfO₂/BeO gate stacks on p-Si substrates.

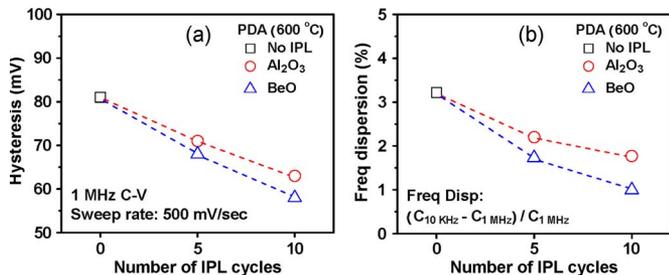


Fig. 7. (a) $C-V$ hysteresis and (b) $C-V$ frequency dispersion characteristics of HfO₂, HfO₂/Al₂O₃, and HfO₂/BeO gate stacks on p-Si substrates.

thickness increases, but the reverse happens with BeO. ALD Al₂O₃ is known to have negative fixed charges [30]; therefore, ALD BeO may have positive fixed charges. BeO V_{fb} shifts less than Al₂O₃, as the IPL increases. Consequently, BeO may exhibit fewer total fixed charges. In Fig. 6(c), all gate stacks have similar leakage currents, but ALD BeO clearly shows a higher breakdown field than the other gate stacks.

In Fig. 7, the BeO IPL exhibits slightly less hysteresis and frequency dispersion. Low hysteresis and frequency dispersion may indicate a low interface defect density. In Fig. 8, the PDA in N₂ atmosphere increases the EOT for HfO₂ and HfO₂/Al₂O₃ gate stacks due to the native oxide (SiO_x) growth, although the PDA increases the dielectric constant of HfO₂ due to carbon reduction or polycrystallization.

From Fig. 8, HfO₂ and HfO₂/Al₂O₃ (5 Å) gate stacks show 3 ~ 4 Å and 2 Å SiO_x growth after a 600 °C PDA, respectively. However, in contrast, the HfO₂/BeO gate stack has a drop in EOT and both the lowest EOT and leakage current density after the 600 °C PDA. The BeO IPL efficiently suppresses the oxygen diffusion from HfO₂ to Si substrates SiO₂ and results in thinner native oxide (SiO_x) and the smaller EOT for HfO₂/BeO gate stack after 600 °C PDA. After a 900 °C PDA, the EOT of the HfO₂/BeO gate stack increases, but it is still lower than the other gate stacks. It is generally known that gas (including oxygen) diffusing into the oxide is proportional to the number of pinholes and the pinhole size in the oxide.

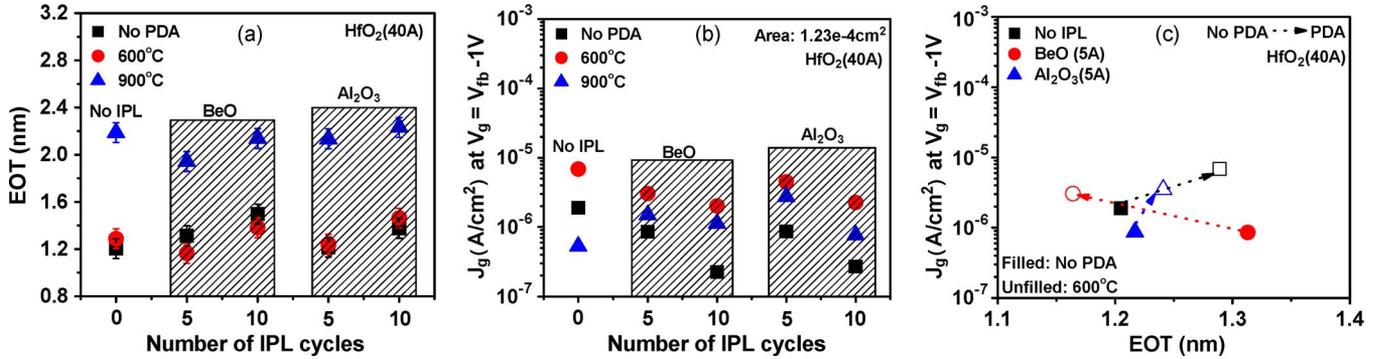


Fig. 8. (a) EOT and (b) J_g versus the number of IPL cycles, and (c) J_g versus EOT for HfO_2 , HfO_2/Al_2O_3 , and HfO_2/BeO gate stacks under various PDA conditions.

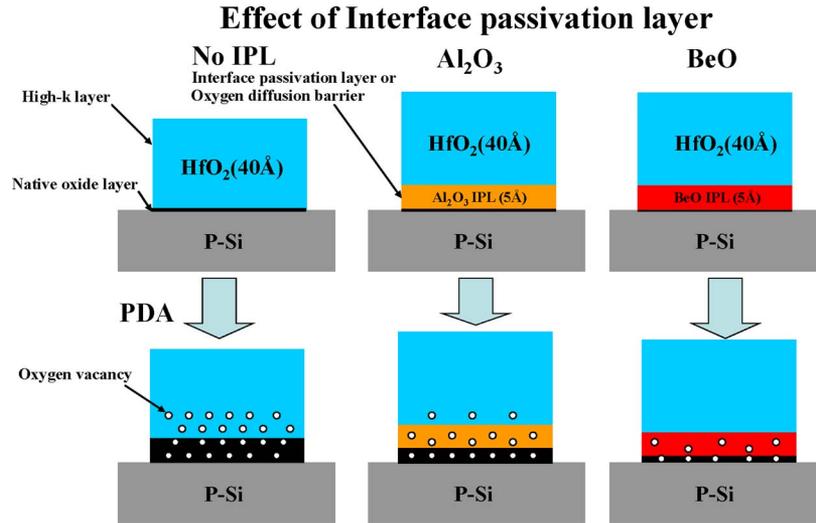


Fig. 9. Schematic MOS structures showing the growth of interfacial native oxide and oxygen vacancy generation on HfO_2 , HfO_2/Al_2O_3 , and HfO_2/BeO gate stacks before and after PDA. Interfacial layer growth and oxygen vacancy generation were estimated from Figs. 5, 6, and 8 results.

The material of a small molecular size has relatively fewer and smaller pinholes [31].

BeO is a well-known gas diffusion barrier because its molecular size is very small and comparable with that of oxygen gas [15]. While thermally grown native oxide (SiO_x) on Si during PDA may be acceptable for traditional complementary MOS technology. PDA-induced native oxides such as Ga_xO_y and As_xO_y on III–V substrates may negatively influence the device performance [32]. (For all practical purposes, the EOTs of BeO and Al_2O_3 samples are essentially the same, i.e., 0.1-nm difference.) Fig. 9 is a schematic comparison of the IPL effect before and after PDA. The native oxide growth and the oxygen vacancy generation shown in Fig. 9 are based on the results in Figs. 5, 6, and 8. From Fig. 9, ALD BeO effectively prevents oxygen diffusion from HfO_2 and interfacial reactions, and suppresses the oxygen vacancies in HfO_2 during annealing up to 600 °C. On the other hand, the 0.5-nm-thick Al_2O_3 film could not prevent oxygen diffusion, and consequently, the EOT increased.

Oxygen vacancies within gate dielectrics are known to be the origin of charge trapping, which degrades reliability characteristics [33]–[35]. To investigate the interface quality of the three gate stacks in Fig. 9, typical methods, such as stress-induced flat-band shift ΔV_{fb} , SILC, and time-dependent dielectric breakdown (TDDDB), were used. Fig. 10 displays

the ΔV_{fb} and SILC under the same effective stress field ($E_{eff} = (V_g - V_{fb})/EOT$). The ALD BeO IPL in Fig. 10 is more stable in terms of ΔV_{fb} and exhibits better immunity to Fowler–Nordheim stress under the same E_{eff} . Fig. 11 shows the charge-trapping characteristics of the HfO_2 , HfO_2/Al_2O_3 , and HfO_2/BeO gate stacks, which display the start of breakdown at $E_{eff} = -21$, -24 , and -26 MV/cm, respectively. Unlike the HfO_2/BeO gate stack, the HfO_2 and HfO_2/Al_2O_3 gate stacks show significant soft breakdown. The results in Figs. 10 and 11 suggest that ALD BeO may suppress native oxide growth at the interface and the oxygen vacancy generation in HfO_2 gate dielectric during PDA, thereby improving reliability.

IV. CONCLUSION

In summary, BeO and HfO_2 have been deposited using dimethylberyllium and tetrakis(diethylamino)hafnium as the metal oxide precursors, respectively, and water vapor as the oxidizer to study the behavior of ALD BeO as an interface passivation layer after annealing. BeO deposited on Si displays a layer-by-layer single crystal, a distinct interface, and no significant interfacial native oxide. The ALD BeO IPL (or oxide barrier layer) has been seen to suppress interfacial native oxide (SiO_x) growth. A 0.4 ~ 0.5-nm-thick ALD BeO effectively prevents oxygen diffusion from HfO_2 and interfacial reactions

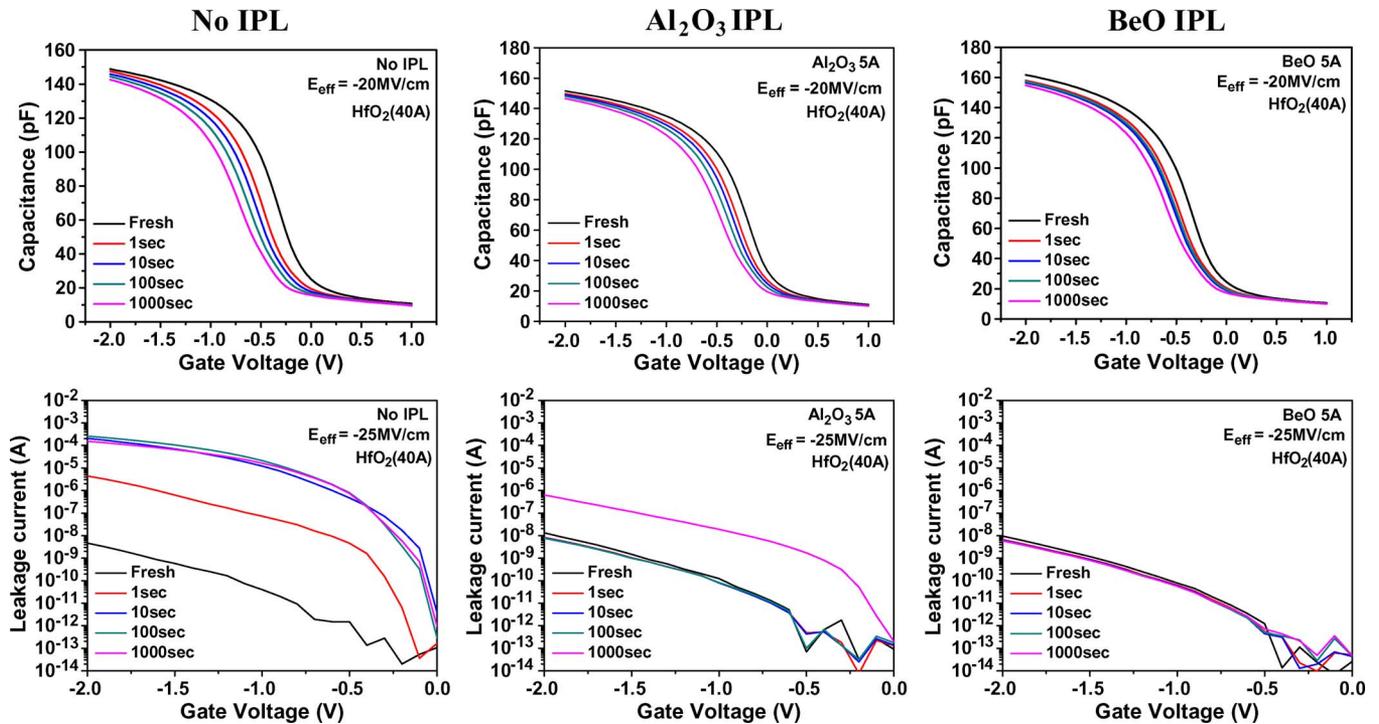


Fig. 10. Stress-induced V_{fb} shift (ΔV_{fb}) and SILC characteristics on HfO_2 , HfO_2/Al_2O_3 , and HfO_2/BeO gate stacks at 600 °C.

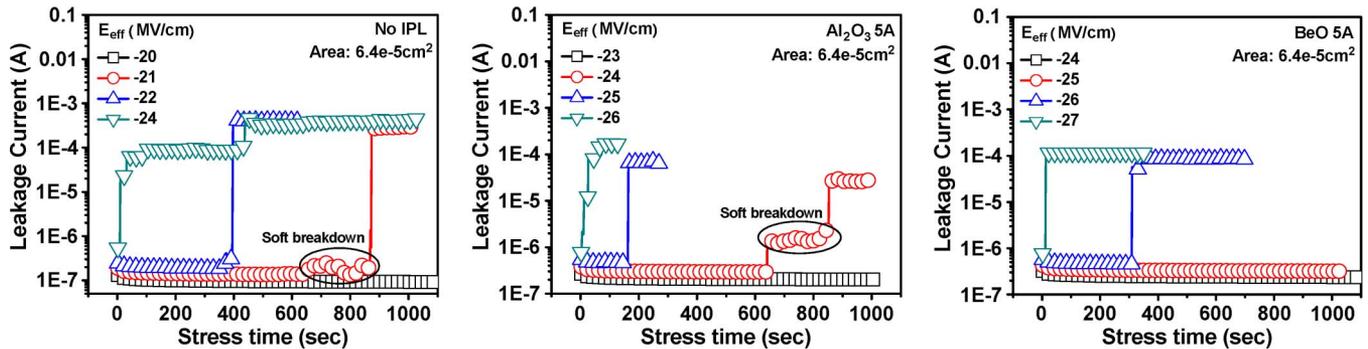


Fig. 11. Charge-trapping characteristics of TDDB for the three gate stacks.

during annealing up to 600 °C, whereas a 0.5-nm-thick Al_2O_3 film could not prevent oxygen, which consequently increased the EOT and degraded the reliability characteristics. The insertion of the 0.5-nm BeO IPL between HfO_2 and Si substrate improves the electrical characteristics and reliability. Moreover, the BeO IPL has a merit in terms of the EOT scaling due to the suppression of oxygen diffusion during high-temperature annealing, as compared with HfO_2 and HfO_2/Al_2O_3 gate stacks.

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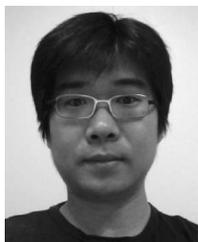
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