

Charge trapping characteristics of atomic-layer-deposited HfO₂ films with Al₂O₃ as a blocking oxide for high-density non-volatile memory device applications

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Abstract

Charge trapping characteristics of high-relative permittivity (high- κ) HfO₂ films with Al₂O₃ as a blocking oxide in p-Si/SiO₂/HfO₂/Al₂O₃/metal memory structures have been investigated. All high- κ films have been grown by atomic layer deposition. A transmission electron microscope image shows that the HfO₂ film is polycrystalline, while the Al₂O₃ film is partially crystalline after a high temperature annealing treatment at 1000 °C for 10 s in N₂ ambient. A well-behaved counter-clockwise capacitance–voltage hysteresis has been observed for all memory capacitors. A large memory window of ~7.4 V and a high charge trapping density of ~1.1 × 10¹³ cm⁻² have been observed for high- κ HfO₂ charge trapping memory capacitors. The memory window and charge trapping density can be increased with increasing thickness of the HfO₂ film. The charge loss can be decreased using a thick trapping layer or thick tunnelling oxide. A high work function metal gate electrode shows low charge loss and large memory window after 10 years of retention. High- κ HfO₂ memory devices with high- κ Al₂O₃ as a blocking oxide and a high work function metal gate can be used in future high-density non-volatile memory device applications.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

High-relative permittivity (high- κ) charge trapping layers in poly-Si/SiO₂/high- κ /SiO₂/Si (SOHOS) memory structure are of increasing interest for charge storage non-volatile memory (NVM) device applications [1–8]. Memory devices with nano-crystals have been reported by several groups [9–17], but they have a uniformity problem. A single quantum well (HfO₂/TiO₂/Al₂O₃) memory capacitor with a small memory window (~1.6 V) has also been reported

[18]. Silicon-nitride (Si₃N₄) charge trapping layers in a poly-Si/SiO₂/Si₃N₄/SiO₂/Si (SONOS) memory structure have been studied extensively with poor retention and scaling issue [19, 20]. The high- κ film as a charge trapping layer should be used in the SONOS structure to improve the program/erase speed, vertical scaling and charge retention characteristics [21–23]. To further improve the vertical scaling and charge retention characteristics of non-volatile memory devices, the high- κ materials with a large barrier height, such

Table 1. The growth and capacitance–voltage (*C–V*) hysteresis window of all memory capacitors with an Al gate electrode.

Memory capacitors	SiO ₂ (nm)	HfO ₂ (nm)	Al ₂ O ₃ (nm)	Memory window $V_g - V_{FBN} = \pm 15$ V
S1	3	5	10	~7.0 V
S2	3	10	10	~7.4 V
S3	4	5	10	~6.0 V

as Al₂O₃ films, are interesting alternatives as a blocking oxide [21–24]. To obtain high-performance non-volatile memory devices, a high work function metal gate electrode in a metal/Al₂O₃/HfO₂/SiO₂/Si (MAHOS) memory structure has been studied for a new generation of memory technology. In this paper, charge trapping characteristics of atomic-layer-deposited high- κ HfO₂ films with high- κ Al₂O₃ as a blocking oxide and a high work function metal gate (Pt) on SiO₂/p-Si substrates in the MAHOS memory structure have been investigated.

2. Experimental details

High- κ HfO₂ and Al₂O₃ films were deposited on p-type silicon (1 0 0) substrates with a resistivity of 15–25 Ω cm by atomic layer deposition (ALD) using hafnium tetra-chloride (HfCl₄) and tri-methylaluminium (Al(CH₃)₃) precursors at a substrate temperature of 300 °C. The precursor temperatures were 185 °C for HfCl₄ and 23 °C for Al(CH₃)₃. Prior to deposition of high- κ HfO₂ and Al₂O₃ films, the p-Si substrate was cleaned by dipping in dilute HF for 1 min using the Radio Corporation of America (RCA) process to remove native oxide from the surface. Then, tunnelling oxides (SiO₂) with thicknesses of 3 nm–4 nm were grown by a rapid thermal oxide (RTO) system with a temperature of 1000 °C for 20–60 s. High- κ Al₂O₃ films as a blocking oxide were used for all memory capacitors. After deposition of high- κ films, a post-deposition annealing (PDA) was carried out at 1000 °C for 10 s in N₂ ambient. Table 1 shows all high- κ memory capacitor structures. After deposition of an aluminium (Al) or platinum (Pt) gate electrode (gate area: 3.14×10^{-4} cm²), post-metal annealing with a temperature of 400 °C for 5 min was done using forming gas [N₂(90%) + H₂(10%)]. To probe the thickness and microstructure of high- κ HfO₂ memory capacitors, high-resolution transmission electron microscopy (HRTEM) was performed using a JEM 3000F field emission system with an operating voltage of 300 kV and a resolution of 0.17 nm. Electrical properties such as high-frequency (100 kHz) capacitance–voltage, current–voltage, charge loss and retention of all memory capacitors were carried out using HP 4284A LCR meter and HP4156B semiconductor analyser systems.

3. Results and discussion

Figure 1 shows the typical HRTEM image of a p-Si/SiO₂/HfO₂/Al₂O₃/Al memory structure (S3). Excellent interfaces of p-Si/SiO₂, SiO₂/HfO₂, HfO₂/Al₂O₃ and Al₂O₃/Al have been observed. Thicknesses of tunnelling oxide (SiO₂), high- κ trapping layer (HfO₂) and high- κ blocking oxide (Al₂O₃)

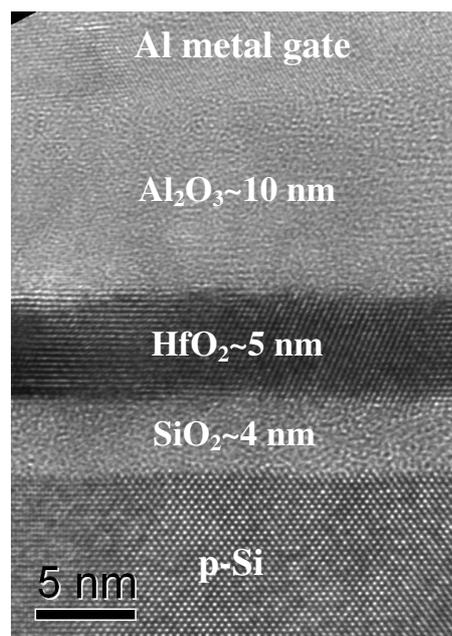


Figure 1. A typical high-resolution transmission electron microscope (HRTEM) image of a high- κ HfO₂ charge trapping layer with high- κ Al₂O₃ as a blocking oxide for the memory capacitor (S3).

are found to be ~4 nm, ~5 nm and ~10 nm, respectively. It is observed that the HfO₂ film shows polycrystalline, while the Al₂O₃ film as a blocking oxide shows almost amorphous after a high temperature annealing treatment (~1000 °C, 10 s) in N₂ ambient. It is also investigated that the high- κ Al₂O₃ film as a blocking oxide has a great impact to enhance the electron injection from the Si substrate under the program condition and to enhance the hole injection under the erase condition of memory capacitors, and it has been explained by a schematic energy band diagram as shown in figure 2. The program and erase conditions are used positive and negative gate voltages, respectively. Under positive gate voltage, the electrons can be tunnelled from the Si substrate and captured by the traps in the HfO₂ films. The trapped electrons in the HfO₂ films will create an electric field distribution to inhibit electron injection. Under the programming condition, the flat-band voltage shift (ΔV_{FB}) can be related to the charge transfer through the corresponding charge centroids [25] in the HfO₂ films by the following relation

$$\Delta V_{FB} = \frac{Q_{HfO_2}}{\epsilon_0} \left[\frac{x_{BO}}{\epsilon_{BO}} + \frac{x_{HfO_2}}{\epsilon_{HfO_2}} \right], \quad (1)$$

where Q_{HfO_2} is the amount of trapped charges per unit area in HfO₂ films. The ϵ_{HfO_2} and ϵ_{BO} are relative permittivities of the trapping oxide (HfO₂) and the blocking oxide (Al₂O₃), respectively. The x_{HfO_2} and x_{BO} are thicknesses of the trapping oxide (HfO₂) and the blocking oxide (Al₂O₃), respectively. The ϵ_0 is the permittivity of free space. The charge centroids can be measured from the tunnelling oxide (SiO₂)/high- κ HfO₂ interface. Assuming a uniform trap distribution in the high- κ HfO₂ films, the charge centroid will be approached half the HfO₂ film thickness and the flat band voltage shift can be

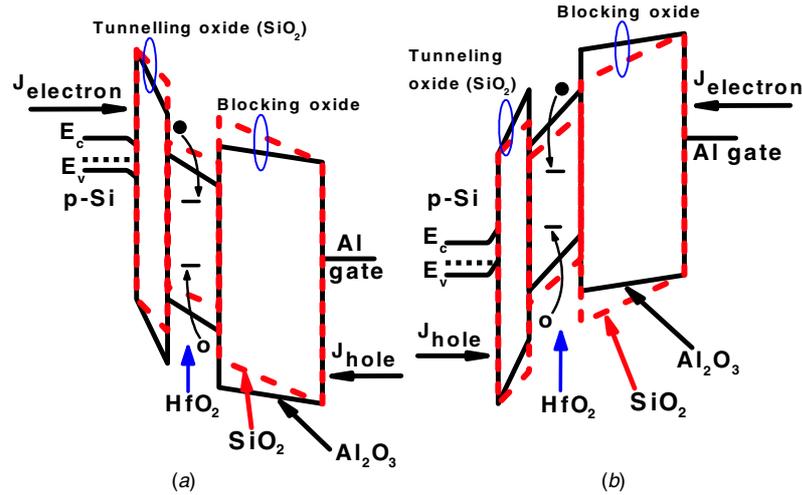


Figure 2. Schematic energy band diagrams of p-Si/SiO₂/HfO₂/Al₂O₃/Al (solid line) and p-Si/SiO₂/HfO₂/SiO₂/Al (dash line) memory structures under (a) program and (b) erase modes.

simplified as

$$\Delta V_{FB} = \frac{Q_{HfO_2}}{\epsilon_0} \left[\frac{x_{BO}}{\epsilon_{BO}} + \frac{x_{HfO_2}}{2\epsilon_{HfO_2}} \right]. \quad (2)$$

The capacitance-equivalent thickness (CET) of SiO₂/HfO₂/Al₂O₃ stack dielectrics can be written as

$$x_{eff} = x_{TO} + \frac{\epsilon_{TO}}{\epsilon_{HfO_2}} x_{HfO_2} + \frac{\epsilon_{TO}}{\epsilon_{BO}} x_{BO}, \quad (3)$$

where ϵ_{TO} is the relative permittivity and x_{TO} is the thickness of tunnelling oxide (SiO₂). The electric fields across tunnelling oxide (E_{TO}) and blocking oxide (E_{BO}) under an erase mode [24] can be explained by the following equations:

$$E_{TO} = \frac{|V_g| - \phi_{ms} - \phi_s - \Delta V_{FB}}{x_{eff}}, \quad (4)$$

$$E_{BO} = \left[E_{TO} + \frac{Q_{HfO_2}}{\epsilon_0 \cdot \epsilon_{TO}} \right] \cdot \frac{\epsilon_{TO}}{\epsilon_{BO}}, \quad (5)$$

where V_g is the applied gate voltage, ϕ_{ms} is the work function difference between the gate electrode and p-Si substrate, and ϕ_s is the surface potential. Relative permittivities of HfO₂ and Al₂O₃ films are found to be ~ 17 and ~ 8 , respectively, after a high temperature annealing treatment at ~ 1000 °C for 10 s in N₂ ambient. Assuming the permittivity of ~ 3.9 of SiO₂, relative permittivities of pure HfO₂ and Al₂O₃ films are calculated using p-Si/SiO₂(3 nm)/HfO₂(10 nm)/Al and p-Si/SiO₂(3 nm)/Al₂O₃(10 nm)/Al stack capacitor structures, respectively, with the series capacitance method [26]. The high-frequency (100 kHz) capacitance is used to calculate the relative permittivity. Using those relative permittivity values in the above equations and applying the $V_g = 15$ V, the electric field across tunnelling oxide (E_{TO}) is ~ 14 MV cm⁻¹ and the electric field across blocking oxide (E_{BO}) is ~ 7.7 MV cm⁻¹ for the high- κ Al₂O₃ blocking oxide, while the E_{TO} is ~ 8.3 MV cm⁻¹ and the E_{BO} is ~ 10 MV cm⁻¹ for the SiO₂ blocking oxide. It is noted that the electric field across tunnelling oxide is very high and electric field across blocking oxide is very small if one can use the Al₂O₃ as a blocking oxide instead of SiO₂ as a blocking oxide. The larger relative permittivity of a trapping layer (HfO₂) and

blocking oxide (Al₂O₃) leads to larger difference of E_{TO} and E_{BO} . The calculated energy band diagram on p-Si/SiO₂/HfO₂/Al₂O₃/Al (solid line) and p-Si/SiO₂/HfO₂/SiO₂/Al (dash line) structures under the program/erase mode is shown in figure 2. Under the program mode, electron current ($J_{electron}$) from the Si conduction layer can be enhanced and hole current (J_{hole}) from the gate electrode can be suppressed using high- κ Al₂O₃ as a blocking oxide. Under the erase mode, the J_{hole} from the Si conduction layer can be enhanced and the electron current from the gate electrode can be suppressed using high- κ Al₂O₃ as a blocking oxide. To get a high program/erase speed, the difference $J_{electron} - J_{hole}$ should be increased. It is important to note that the back tunnelling currents (J_{hole} under program mode and $J_{electron}$ under erase mode) can be suppressed using a large barrier height and large relative permittivity of the high- κ blocking oxide. It is also observed that the high- κ HfO₂ layer can not only improve charge trapping properties but also can improve the program/erase speed and device scaling. Due to the advantage of the high- κ Al₂O₃ as a blocking oxide, memory characteristics of the p-Si/SiO₂/HfO₂/Al₂O₃/Al (or Pt gate electrode) structures have been investigated in this study.

A well-behaved high-frequency (100 kHz) counter-clockwise capacitance–voltage ($C-V$) hysteresis of a memory capacitor (S2) has been observed with different sweeping gate voltages (figure 3). The counter-clockwise $C-V$ hysteresis of p-Si/SiO₂/HfO₂/Al₂O₃/Al memory capacitors confirms that the electrons can be injected from the Si substrate to the HfO₂ trapping layer under positive gate voltage and the holes can be injected from the Si substrate to the HfO₂ trapping layer under negative gate voltage. A quasi-neutral capacitance–voltage ($C-V$) curve (i.e., no charging up or discharging the memory capacitor) has been obtained with the application of a small sweeping gate voltage ($V_g \sim \pm 3$ V). The sweeping gate voltage step was 0.1 V s⁻¹ during the $C-V$ measurement for all memory capacitors. The quasi-neutral flat-band voltage (V_{FBN}) is shown in figure 3. A large memory window of $\Delta V \approx 7.4$ V has been observed under a sweeping gate voltage of $V_g \approx \pm 15$ V, and it can be used in multi-level charge

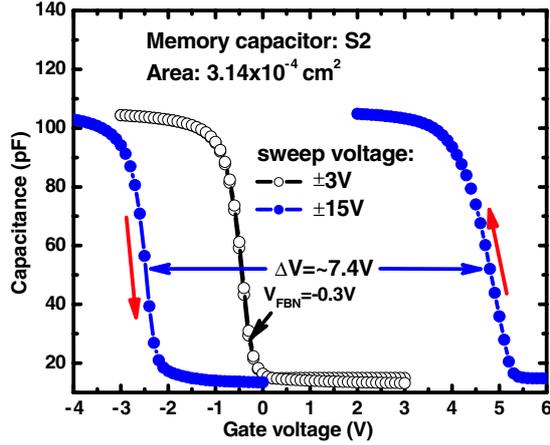


Figure 3. Capacitance versus gate voltage characteristic of the high- κ HfO₂ memory capacitor (S2).

(MLC) storage memory device applications. The capacitance-equivalent thicknesses are found to be ~ 9.2 nm for S1, ~ 10.3 nm for S2 and ~ 11.2 nm for S3 memory capacitors. The capacitance density of > 3.0 fF μm^{-2} is high enough to scaling the device. According to industrial technology roadmap on semiconductor (ITRS), the thicknesses are needed to be 3 nm for tunnelling oxide (SiO₂), 4 nm for the silicon-nitride (Si₃N₄) trapping layer and 4 nm for blocking oxide (SiO₂) for 45 nm technology node in the SONOS flash memory device. So, the total thickness of the SONOS flash memory is 11 nm. Furthermore, the SONOS memory device cannot be scaled below the 45 nm technology node, and one of the reasons is the high leakage current. Our memory device shows the small CET of 9.2 nm with a low leakage current because of thick high- κ HfO₂ as a trapping layer and the high- κ Al₂O₃ as a blocking oxide. It is believed that our memory device can be used below the 45 nm technology node.

Figure 4 shows the flat-band voltage shift (V_{FB}) versus drive gate voltage ($V_{\text{g}} - V_{\text{FBN}}$). The V_{FB} shift has been measured with respect to the V_{FB} of quasi-neutral $C-V$ curve (V_{FBN}). The V_{FB} shift increases with increasing the drive gate voltage. The V_{FB} shifts at $V_{\text{g}} - V_{\text{FBN}} \approx +15$ V are found to be ~ 4.4 V for S1, ~ 5.1 V for S2, ~ 4.2 V for S3 and ~ 3.0 V for pure Al₂O₃ charge trapping layers. A pure Al₂O₃ charge trapping layer in the p-Si/SiO₂ (3 nm thick)/Al₂O₃ (15 nm thick)/Al structure has been used for comparison (figure 4). The V_{FB} shift of all HfO₂ charge trapping layers is higher than that of the pure Al₂O₃ charge trapping layer and it can be explained as follows. Conduction band offset [$\Delta E_{\text{c}} \approx (E_{\text{c}})_{\text{HfO}_2} - (E_{\text{c}})_{\text{Si}}$] and valence band offset [$\Delta E_{\text{v}} \approx (E_{\text{v}})_{\text{Si}} - (E_{\text{v}})_{\text{HfO}_2}$] of the HfO₂ films with respect to the Si substrate are found to be ~ 1.7 eV and ~ 3.1 eV, respectively, and those values of the pure Al₂O₃ films are found to be ~ 2.8 eV and ~ 4.9 eV, respectively. It indicates that more electrons or holes from the Si substrate can be injected into the HfO₂ films rather than the Al₂O₃ films because Fowler–Nordheim (FN) tunnelling current has been dominated at the same gate voltage. The V_{FB} shifts are found to be ~ 1.2 V for the HfO₂ film (S1) and ~ 0.5 V for the pure Al₂O₃ films under the same drive gate voltage (+5 V), suggesting that the FN tunnelling current from the Si substrate to the HfO₂

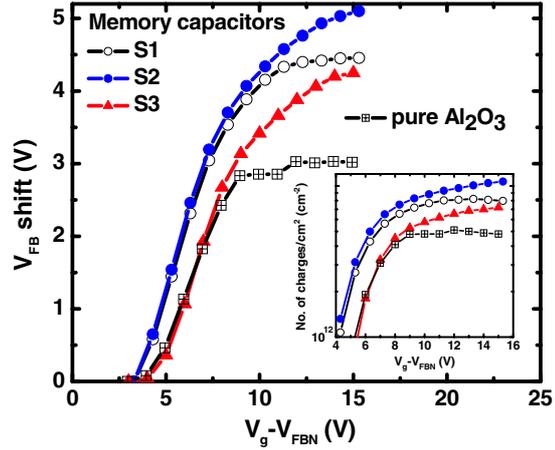


Figure 4. Flat-band voltage (V_{FB}) shift versus gate drive voltage ($V_{\text{g}} - V_{\text{FBN}}$) for all high- κ HfO₂ charge trapping memory capacitors. The charge trapping density of all memory capacitors has been shown in the inset. The V_{FB} shift and charge trapping density of an Al₂O₃ charge trapping memory capacitor have been plotted for comparison.

trapping layer is higher than that of the pure Al₂O₃ films. The valence band offset and conduction band offset of all memory structures have been measured by ultra-violet photoelectron spectroscopy (not shown here). The memory window and the V_{FB} shift of all HfO₂ memory capacitors are higher than that of the pure Al₂O₃ film, due to higher charge trapping sites and higher charge trapping density in the HfO₂ film [21]. A thicker HfO₂ film (~ 10 nm) has a larger memory window than that of a thin (~ 5 nm) HfO₂ film (table 1), due to high charge trapping density of the thicker HfO₂ film (inset of figure 4). The charge trapping density for all memory capacitors is calculated using this equation,

$$N_{\text{charge}} = \Delta V_{\text{FB}} \cdot C_{\text{ox}}, \quad (6)$$

where C_{ox} is the total oxide capacitance per unit area. A high charge trapping density of $\sim 1.1 \times 10^{13}$ cm⁻² has been observed for a high- κ memory capacitor, indicating that the high- κ HfO₂ charge trapping layer shows promise for application.

Erasing characteristics for all memory capacitors are shown in figure 5. Erasing drive voltages ($V_{\text{g}} - V_{\text{FBN}}$) with an Al gate electrode are found to be -10.0 V for S1, -10.5 V for S2 and -11.7 V for S3 memory capacitors. A large erasing voltage is necessary for all memory capacitors, due to large valence band offset (~ 3.1 eV) and small amount of hole injection from the Si substrate. The memory capacitor (S3) has a large erase voltage (-11.7 V), due to the thicker tunnelling oxide (~ 4 nm) and lower tunnelling probability of holes from the Si substrate into the high- κ charge trapping layers. The erasing voltage of the S1 memory capacitor with a Pt gate is -8.3 V. It is observed that the work function is ~ 4.1 eV for the Al gate and ~ 5.2 eV for the Pt gate. The erasing voltage can be drastically decreased with a high work function metal gate (Pt), due to lower leakage current from the gate electrode. Figure 6 shows the leakage current of the high- κ HfO₂ memory capacitor (S1). The gate leakage current is lower for a high work function metal gate and the memory capacitor can be operated up to the gate voltage of

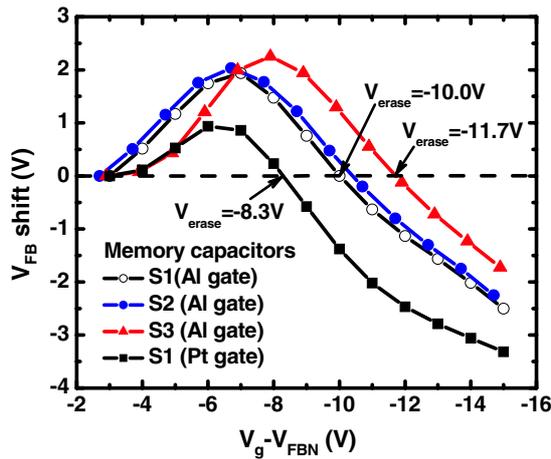


Figure 5. Erasing characteristics of all high- κ HfO₂ memory capacitors with Al and Pt metal gates.

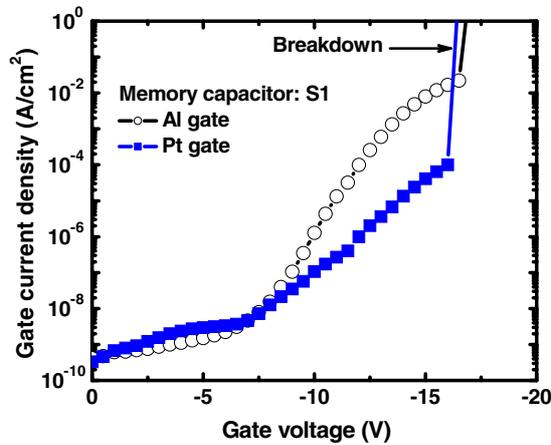


Figure 6. Leakage current characteristics of the memory capacitor (S1) with Al and Pt gate electrodes.

~16 V because of the high breakdown voltage of < -16 V. It is noted that a low backward tunnelling current, thin tunnelling oxide and small valence band offset between the trapping layer and Si substrate are necessary for easy erasing of memory devices.

Normalized capacitances versus time have been shown in figure 7. To investigate the charge loss, all memory devices are programmed using $V_g - V_{FBN} = +6$ V for 60 s. Then, the capacitance has been measured at a gate voltage of $V_g - V_{FBN} = -1.5$ V. A constant gate voltage of $V_g - V_{FBN} = -1.5$ V has been applied to the gate electrode at all times during the capacitance measurement. The memory capacitor (S1) with a thin (~5 nm) HfO₂ charge trapping layer shows a large charge loss as compared with a thick (~10 nm) charge trapping layer (S2). The memory capacitor (S3) shows a negligible charge loss as compared with those of other memory capacitors, due to low probability of charge loss through thick tunnelling oxide (~4 nm). The impact of the metal gate electrode on the memory window after 10 years of retention has been shown in figure 8. At first, the memory capacitors (S2) are programmed under a drive gate voltage of +15 V for 10 s. Then, the V_{FB} has been

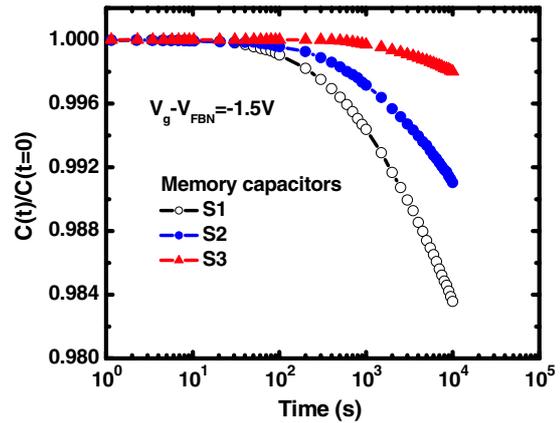


Figure 7. Capacitance versus elapsed time of all high- κ HfO₂ charge trapping memory capacitors.

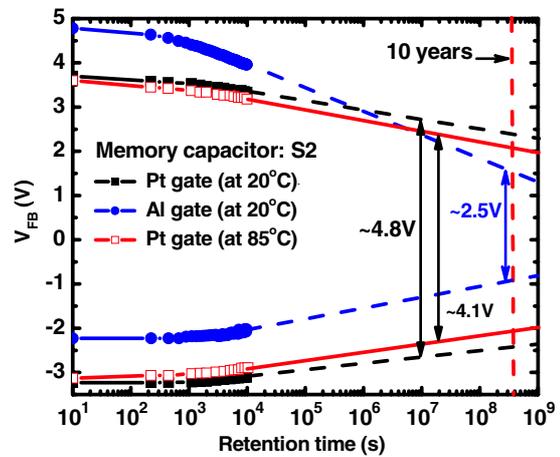


Figure 8. Retention characteristics of the high- κ HfO₂ memory capacitor (S2) with Al and Pt gate electrodes.

measured with time. The memory capacitor has been grounded during V_{FB} measurement. Similarly, the memory capacitors (S2) are erased under a drive gate voltage of -15 V for 10 s and the V_{FB} is measured with time. Assuming the logarithmic behaviour for retention, the extrapolation of V_{FB} shift for all memory capacitors has been performed up to 10 years. Memory windows of the Al and Pt gate electrodes are estimated to be ~2.5 V and ~4.8 V at 20 °C after 10 years of retention. The memory window of the Pt gate is ~4.1 V at 85 °C after 10 years of retention. An excellent retention has been observed by the Pt gate electrode, due to the low leakage current of the memory capacitors (figure 6). Charge losses of the Al and Pt gate electrodes are estimated to be ~60% and ~31%, respectively, after 10 years of retention. The charge loss of the memory capacitor (S2) with a Pt gate electrode increases (slightly) ~39% at 85 °C after 10 years of retention. She *et al* [27] reported that the charge loss of HfO₂ charge trapping memory devices is higher ~56% at 85 °C after 10 years of retention. Due to the large memory window and low charge loss of the high- κ HfO₂ charge trapping memory device with high- κ Al₂O₃ as a blocking oxide and a high work function metal gate, it can be used in future high-density and scaled flash memory device applications.

4. Conclusion

Charge trapping characteristics of the high- κ HfO₂ charge trapping memory capacitors with high- κ Al₂O₃ as a blocking oxide have been investigated. They show a high charge trapping density, large memory window, low charge loss and small equivalent oxide thickness. The high- κ memory devices with a high work function metal gate show promise for future scaled high-density flash memory device applications.

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