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Low-temperature fabrication of HfAlO alloy dielectric using atomic-layer deposition and its application in a low-power device

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ABSTRACT

This paper presents a systematic study of HfAlO alloy films, which were prepared by doping HfO₂ with different amounts of Al using atomic layer deposition at low growth temperature (150 °C). The chemical constituents of the various HfAlO films were determined by X-ray photoelectron spectroscopy. Carrier transport through the HfAlO layer presented a good fit with the Poole-Frenkel emission and Fowler-Nordheim tunnelling mechanisms in different voltage ranges. Barrier heights (conduction band offset) between the HfAlO films and Si substrate, which were derived from the Fowler-Nordheim fittings, increased with increasing Al content in the alloy films, and the leakage current was suppressed well owing to the higher barrier. The 10 nm HfAlO dielectric layer, which was grown by the repeated deposition of 1 cycle each of HfO₂ and Al₂O₃ (defined as H1A1), showed a high gate capacitance of 723 nF/cm², a high breakdown voltage of 8.0 V, a small leakage current, and a smooth surface. HfAlO films doped with different amounts of Al₂O₃ in HfO₂ were employed to study their effects on the operation of amorphous indium-gallium-zinc oxide (a-IGZO) thin-film transistors (TFTs). The a-IGZO TFTs based on

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the HfAlO₃ gate dielectric showed desirable properties including a high $I_{\text{on/off}}$ ratio of 1.7×10^7 , a small subthreshold swing of 176 mV/decade, and low operating voltage of 2.0 V.

Keywords:

HfAlO₃, High- k gate insulator, a-InGaZnO, Thin-film transistor, Atomic layer deposition, Low-power device

1. Introduction

The material HfO₂ has been widely used and investigated in transistors [1-3], ferroelectric memory [4], and resistive random memory [5, 6], owing to its high dielectric constant ($k = 20\text{--}25$), relatively large band gap (~ 5.8 eV), and compatibility with current semiconductor technologies. However, HfO₂ has some unavoidable disadvantages, i.e., HfO₂ film contains a high concentration of oxygen vacancies and oxygen interstitials, which cause many shallow traps in the film, and it has a low crystallisation temperature [7]. To overcome these shortcomings, additional dopants, such as, Si, La, and Al are introduced into HfO₂ to form Hf-based hybrid materials, which have been employed in microelectronic devices [8-10]. Researchers have devoted considerable attention to HfAlO films (fabricated by incorporating Al₂O₃ in HfO₂ films), which effectively suppressed the interface trap density (D_{it}) and increased the band gap and the breakdown field [11]. Band alignment between HfAlO and the semiconductor is of great importance for MOSFET applications, which could improve the barrier height for carriers and reduce the leakage currents [12, 13]. The properties of the interface between HfAlO and different films, for instance, graphene [14], InP [12], Ga₂O₃ [11], and Ge [13], have also been widely studied. Furthermore, researchers have explored the influence of the chemical composition of

HfAlO films [15], and investigated the effects of the thermal stability on annealing temperature [16, 17]. HfAlO films have been grown by diverse methods such as solution-process [18], sputtering [19, 20], and atomic layer deposition (ALD) [12, 21, 22]. Among these alternative technologies, ALD is a technique to fabricate films by way of chemical vapour deposition and is based on ordered and self-saturated surface reactions. ALD is an effective and important method that allows dense, smooth, and highly conformal films to be fabricated with angstrom-precision thickness control [23]. HfAlO films grown by ALD play important roles in memory devices, especially in NAND flash memories [24], and resistive random memory devices [25]. Furthermore, HfO₂-based materials have been used as the gate dielectric as a replacement for conventional SiO₂ to improve the device performance and realise the low-voltage operation of transistors [26, 27].

Thin-film transistors (TFTs) are regarded as important components in electronic devices, and amorphous indium-gallium-zinc oxide (a-IGZO) has been widely studied for the fabrication of TFTs owing to its excellent performance in terms of stability, high electron mobility, low off current, surface smoothness, and low processing temperature [28, 29]. Emerging applications of a-IGZO TFTs, such as sensors [30], wearable electronics [31], and thin-film circuits [32, 33] have attracted considerable attention for use in flexible devices, because of the ease of deposition of a-IGZO on a flexible substrate. When developing flexible devices and circuits, the identification of a befitting dielectric that can be deposited at low temperature is crucial, owing to the temperature-sensitive nature of flexible substrates [34, 35]. The use of solution-induced HfAlO film has also been used in low-voltage-operating InZnO TFTs, while, high-temperature annealing was needed to realise a low leakage current [18]. HfAlO alloy could be a promising dielectric for use in low operating voltage a-IGZO TFTs, which may be powered by lithium ion

batteries. [36-39] It is worthwhile to fabricate high-quality HfAlO alloy films grown by ALD at low temperature and to study its applications in low-power a-IGZO TFTs.

In this study, three HfAlO films, consisting of HfO₂ doped with different amounts of Al, were grown using ALD at 150 °C. The chemical bonding states of the HfAlO films have been investigated by using X-ray photoelectron spectroscopy (XPS). Metal-insulator-semiconductor (MIS) capacitors were fabricated to study the electron transport mechanism and measure the robustness of the HfAlO films. Current-voltage (I - V) and capacitance-voltage (C - V) measurements were conducted to explore the electrical characteristics of HfAlO films. The a-IGZO TFTs, based on a HfAlO gate dielectric layer, exhibit excellent properties, and show promise for use in future electronic devices with low power consumption requirements.

2. Experimental

In this work, we fabricated various HfAlO alloy dielectric films by using ALD at a growth temperature of 150 °C. The HfAlO films were grown using a sequence of x HfO₂ cycles followed by y Al₂O₃ cycles, and this process was repeated n times to achieve a thickness of 10 nm. The three types of HfAlO films are denoted as H_xA_y, i.e., H₂A₁, H₁A₁, H₁A₂. Two control samples only including either HfO₂ or Al₂O₃ were also used for comparison purposes. For the HfO₂ cycle, tetrakis(dimethylamino)hafnium (TDMAH) and deionised H₂O were selected as the metal precursor and oxygen source, respectively. Similarly, trimethylaluminium (TMA) and deionised H₂O were used for the Al₂O₃ deposition. During film growth, high purity N₂ was used as the carrier gas with which to introduce the precursors to the reactor chamber and the pressure in the chamber was kept at 0.2 Torr. The dose time of TDMAH, TMA, and H₂O was 0.3 s, 0.03 s and 0.015 s, respectively. Every dose operation was followed by a 30 s purging process. The chemical

bonding states of the HfAlO films were examined by XPS with ThermoFisher ESCALAB 250 equipment. The different HfAlO alloy films were employed in a-IGZO TFTs as the gate dielectric layers, which were first grown by ALD on heavily doped n-type Si substrates. Following the deposition of HfAlO film, a 24-nm a-IGZO ($\text{In}_2\text{O}_3:\text{Ga}_2\text{O}_3:\text{ZnO} = 1:1:1$ mol. %) channel layer was deposited by radio-frequency (RF) sputtering at room temperature. During the sputtering, the RF power and Ar pressure were maintained at 90 W and 3.65 mTorr, respectively. Finally, 100-nm Al source and drain contact electrodes were deposited by thermal evaporation. The channel length (60 μm) and width (2000 μm) of the a-IGZO TFTs were defined by a shadow mask. Before the electrical measurements, the TFTs were annealed in air at 150 °C for 1 h. The capacitance of the dielectrics and carrier transport mechanisms were explored by fabricating an MIS capacitor with different dielectrics using a shadow mask and thermal evaporation. I - V and C - V measurements were conducted to explore the performance of the devices using Agilent B2900 equipment and an E4980A Precision LCR Meter. The HfAlO film morphology and surface roughness was determined by atomic-force microscopy (AFM) and reported for an area of $10 \times 10 \mu\text{m}^2$.

3. Results and discussion

The chemical constituents and element binding energy of various HfAlO films were first determined by XPS. Fig. 1 shows the X-ray photoelectron spectra of the Al 2p, Hf 4f, and O 1s core levels of the Al_2O_3 and HfO_2 films. The binding energy was calibrated with the position of the C1s peak at 284.8 eV. For the Al_2O_3 film, Fig. 1(a) shows the Al 2p peak, which is fitted as a single peak at 74.9 eV. The O 1s peak is located at 531.6 eV, as shown in Fig. 1(b), which is fitted well as a symmetrical single Al-O peak. The peak positions of Hf 4f_{5/2}, Hf 4f_{7/2}, and O 1s

for HfO₂ films were fitted at 18.5 eV, 16.8 eV and 530.0 eV, respectively, as shown in Figs. 1(c) and 1(d). The binding energy of the Al–O bond (531.6 eV) is obviously larger than that of the Hf–O bond (530.0 eV), which means that Hf is a more ionic cation than Al[40]. The Al 2p, Hf 4f, and O 1s peak positions in the various HfAlO films are summarised in Figs. 2(a), 2(b), and 2(c). All peaks of Al 2p, Hf 4f, and O 1s were observed to shift to a higher binding energy as the Al₂O₃ concentration in HfAlO films increased, indicating enhanced incorporation of the Al₂O₃ and HfO₂ layers and the formation of Hf–O–Al bonding. The position of the Al 2p peak shifted from 74.1 eV in H2A1 to 74.6 eV in H1A2, while the Hf 4f peaks shifted from 17.2 eV and 18.7 eV in H2A1 to 17.8 eV and 19.3 eV in H1A2. The chemical compositions of various HfAlO films were determined by the elemental peak intensity in the XPS spectra. The detailed statistical compositions of the five films are provided in Table 1. The HfO₂ mole fraction values in the H2A1, H1A1, and H1A2 dielectric layers, 0.60, 0.45, and 0.31, respectively, were obtained by calculation [40].

Fig. 1. XPS results of the (a) Al 2p and (b) O 1s core levels for Al₂O₃ film, and (c) Hf 4f and (d) O 1s core levels

for HfO₂ film.

Table 1 Elemental composition of HfAlO films estimated by XPS and the mole fraction of HfO₂ in HfAlO.

	HfO ₂	H2A1	H1A1	H1A2	Al ₂ O ₃
HfO ₂ mole fraction value in HfAlO	1	~0.60	~0.45	~0.31	0
Hafnium at. %	32.8%	15.8%	11.0%	7.0%	0
Aluminium at. %	0	18.4%	26.1%	31.7%	40.1%
Oxygen at. %	67.2%	65.8%	62.9%	61.3%	59.9%

The ALD process we developed was designed with the aim of achieving Hf–O–Al bonding.

HfO₂ film growth is represented by two reaction equations:

Similarly, Al₂O₃ film growth can be expressed by two reaction equations:

Hf–O–Al bonding may be formed by combining Hf–OH and Al(CH₃)₃ between Eqs. (2) and (3) or Al–OH and Hf[N(CH₃)₂]₄ between Eqs. (4) and (1). The position of the O 1s peak for Hf–O–Al bonding was fitted at 530.9 eV and the peak intensity increases as the amount of incorporated Al increases, as shown in Fig. 2(c). The oxygen vacancies may be reduced with the formation of Hf–O–Al bonding, which could decrease the number of defects and suppress the leakage current through the HfAlO alloy film.

Fig. 2. XPS spectra for (a) Al 2p, (b) Hf 4f, and (c) O 1s core levels from HfAlO films.

To study the electron transport mechanism and measure the robustness of the HfAlO films, MIS capacitors were fabricated and relatively large voltages were applied to the devices. The I - V characteristics of the capacitors are shown in Fig. 3(a) and a schematic diagram is shown as the inset. The leakage currents can be seen to decrease with increasing Al content of the HfAlO films and the breakdown voltages for various HfAlO films are all greater than that of HfO_2 film, which may be caused by the decreasing number of defects. We carried out an in-depth analysis of the leakage mechanism by fitting the I - V curves by different models. Possible mechanisms include direct tunnelling, Schottky emission, Poole-Frenkel (P-F) emission, and Fowler-Nordheim (F-N) tunnelling.

P-F emission occurs because of the emission of trapped electrons into the conduction band. The I - V curve is given by

$$I \propto V \exp \left[\frac{q}{kT} (2a\sqrt{V} - \phi_0) \right], \quad (5)$$

where a is a constant, q is the electron charge, and ϕ_0 is the barrier height [41].

The F-N tunnelling current is given by the following formula

$$\frac{J}{E^2} = C_1 e^{-C_2/E}, \quad (6)$$

Where C_1 and C_2 are given by

$$C_1 = q^3 m / 16\pi^2 \hbar m_{\text{ox}} \phi_0, \quad (7)$$

$$C_2 = \frac{4}{3} \frac{(2m_{\text{ox}})^{1/2}}{q\hbar} \phi_0^{\frac{3}{2}}, \quad (8)$$

where J is the current density, E is the electric field, $2\pi\hbar$ is Planck's constant, and m and m_{ox} are the electron mass in free space and in the oxide, respectively [42]. Further, m_{ox} is $0.23m$ for Al_2O_3 and $0.17m$ for HfO_2 , respectively [43, 44]. As for HfAlO , m_{ox} is $0.20m$.

Fig. 3. (a) I - V measurements of Al/HfAlO/ n^+ -Si devices with different Hf/Al ratios, and a compliance current of

10^{-3} A was set during the electrical measurements. (b) Statistics of the barrier heights and leakage currents at 3.0 V

bias. Linear fittings of the leakage current by the (c) P–F emission and (d) F–N tunnelling models. The dotted lines are the experimental results and the solid lines are the fittings. The insets in Fig. 3c and Fig. 3d are the energy–band diagrams of the P–F emission and F–N tunnelling conduction mechanisms, respectively.

The leakage currents of the 10–nm thin films of HfAlO with different Hf/Al ratios are fitted by Eqs. (5) and (6), and the linear fittings are shown in Figs. 3(c) and 3(d), respectively. The fitted results indicate that, under a lower E , the carriers can traverse the films via shallow traps, and when E is sufficiently strong, the energy of the carriers is such that they are able to partly tunnel through the barrier. The barrier height can be deduced from the F–N fitting results. Details of the barrier heights and leakage currents at 3.0 V bias are plotted in Fig. 3(b). The barrier heights are 1.33, 1.46, 1.47, 1.45, and 1.95 eV for HfO₂, H2A1, H1A1, H1A2, and Al₂O₃, respectively. The barrier heights of the H2A1, H1A1, and H1A2 films are larger than that of the HfO₂ film, which could also be responsible for suppressing the leakage current.

Fig. 4. (a) C – V characteristics (main image) and structural diagram (inset) of Al/HfAlO/p–Si capacitors with the

five different dielectrics. (b) AFM image of 10 nm H1A1 film in a scan area 10 × 10 μm².

Subsequently, Al/HfAlO/p–Si capacitors were fabricated to explore the capacitance of the HfAlO dielectrics (C_{ox}). Fig. 4(a) and the inset show the C – V characteristics and structural diagram of capacitors, respectively. The C_{ox} values of the HfAlO films ordered from large to

small are 1010, 791, 723, 680, and 550 nF/cm² for HfO₂, H2A1, H1A1, H1A2, and Al₂O₃, respectively. The capacitance decreases as the percentage of Al in the dielectric film increases. The surface morphologies of the HfAlO dielectric films were also studied by AFM. Fig. 4(b) shows the AFM image of the 10-nm H1A1 film. HfAlO films were all prepared by ALD, which is based on ordered and self-saturated surface reactions and allows highly conformal films to be fabricated layer-by-layer with angstrom-precision thickness control. The root-mean-square (RMS) values of the roughness are 0.28, 0.34, 0.31, 0.33, and 0.25 nm for HfO₂, H2A1, H1A1, H1A2, and Al₂O₃, respectively, which confirm the smooth surface morphology of our ALD films.

Fig. 5. (a) Structural diagram of a-IGZO TFTs. (b) Transfer characteristics of a-IGZO TFTs with different HfAlO gate dielectrics. (c) Output characteristics of the a-IGZO TFT with 10-nm H1A1.

The structural diagram and transfer characteristics of the a-IGZO TFTs with different HfAlO films as gate dielectrics are shown in Figs. 5(a) and 5(b), respectively. The interface between the channel layer and gate dielectric layer would be expected to influence the performance of the

TFTs, especially the surface roughness of the gate dielectric material. As shown in Fig. 5(b), very little hysteresis is observed, which could be a benefit of the smooth surface of the dielectric layers and low density of slow interface traps [28]. However, a-IGZO TFTs with 10-nm HfO₂ film as the gate dielectric layer experience a strong leakage current, resulting in that TFTs cannot be turned off successfully. This indicates that doping HfO₂ with Al₂O₃ could suppress the leakage current effectively. In addition, large capacitance is necessary to operate these TFTs at low power. Therefore, H1A1 was selected as the optimal gate dielectric layer to maintain a high carrier mobility and large on/off ratio. As shown in Fig. 5(c), clear pinch-off behaviour can be found in the output curves of the a-IGZO TFT with the 10-nm H1A1 dielectric. The *SS* value is determined by the gate dielectric interface trap density D_{it} , the bulk trap density in the semiconductor channel N_{bulk} , and in the case with the non-encapsulated channel, also the top surface trap density of the semiconductor channel D_{top} :

$$SS = \frac{k_B T \ln 10}{q} \left[1 + \frac{q^2}{C_{ox}} D_{total} \right]$$

where $D_{total} = D_{it} + tN_{bulk} + D_{top}$, and t is the thickness of the channel [29, 45]. The D_{total} values and other main characteristics of TFTs gated by different HfAlO layers are also compared in Table 2. The linear (μ_{lin}) and saturation (μ_{sat}) mobility were extracted from the transfer curves at V_{DS} of 0.1 V and 2.0 V, respectively. The TFTs that contain 10-nm H1A1 exhibit desirable properties for low-power operations, including low threshold (or turn on) voltage ($V_T = 0.83$ V), high saturation mobility ($\mu_{sat} = 5.52 \pm 0.07$ cm²/Vs) and linear mobility ($\mu_{lin} = 4.52 \pm 0.06$ cm²/Vs), a small subthreshold swing ($SS = 176$ mV/dec), and a high on/off current ratio ($I_{on}/I_{off} = 1.7 \times 10^7$).

Table 2 Comparison of a-IGZO TFTs gated by different HfAlO dielectric layers.

Gate	μ_{sat}	μ_{lin}	V_{T}	SS	$I_{\text{on}}/I_{\text{off}}$	$N_{\text{SS}}^{\text{max}}$
dielectrics	(cm^2/Vs)	(cm^2/Vs)	(V)	(mV/dec)		($\text{eV}^{-1}\text{cm}^{-2}$)
HfO ₂	8.83±0.12	7.58±0.10	0.95	... ^a	7.6×10 ⁶	... ^a
H2A1	5.97±0.08	4.94±0.07	0.88	168	2.2×10 ⁶	8.91×10 ¹²
H1A1	5.52±0.07	4.52±0.06	0.83	176	1.7×10 ⁷	8.75×10 ¹²
H1A2	3.93±0.05	2.98±0.04	0.87	185	1.1×10 ⁷	8.89×10 ¹²
Al ₂ O ₃	3.68±0.05	2.58±0.04	0.91	195	7.3×10 ⁶	7.75×10 ¹²

^aParameter is difficult to extract because of poor device performance.

4. Conclusions

In summary, films of HfAlO alloys grown by ALD at low temperature with different Al content were studied for implementation as a gate dielectric for a-IGZO TFTs. The chemical constituents of various HfAlO films were determined by XPS. As the doping Al content of the HfAlO increased, the number of defects in the films decreased, the barrier height was enlarged, and the leakage current was suppressed to a satisfactory extent. The 10-nm H1A1 dielectric layer showed low surface roughness and a high breakdown voltage. Importantly, the a-IGZO TFTs based on H1A1 film exhibited desirable properties, including a steep subthreshold slope, negligible hysteresis, a high $I_{\text{on/off}}$ ratio, and a low operating voltage. In terms of carrier transport, the P-F emission and F-N tunnelling mechanisms were found to play important roles in different voltage ranges. The results confirm that our a-IGZO TFTs gated by an HfAlO alloy dielectric material show great promise for use in future low-power electronics applications.

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