Excellent interface properties of pentacene based metal-oxide-semiconductor diodes utilizing HfON high-k gate insulator

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1. Introduction
In addition to the carrier mobility, the threshold voltage ($V_{th}$) which is related to the flat-band voltage ($V_{fb}$) is another important parameter that must be controlled to ensure proper operation of the OFETs. However, due to trap states generated at the interface between gate insulator and organic semiconductor, such as pentacene, by air exposure and fabrication process, the $V_{fb}$ of the pentacene based OFETs usually show a range of shifts [1, 2]. In this paper, we report that pentacene based MOS diodes with HfON gate insulators show a small $V_{fb}$ shift and a negligible frequency dispersion in C-V characteristics.

2. Experimental Procedure
Pentacene based MOS diodes were fabricated on n+-Si(100) substrates with a top-contact bottom-gate device geometry. 8.8 nm-thick HfON (CET: 2.7 nm) and 9.0 nm-thick HfO2 (CET: 5.6 nm) gate insulators were fabricated on n+-Si(100) substrates by ECR sputtering [3]. SiO2 (CET: 9.5 nm) films were grown on chemically cleaned n+-Si(100) substrates by wet thermal oxidation at 850°C. Pentacene films (20 nm) were deposited on the three different gate insulators by thermal evaporation at room temperature (2.0×10^{-6} Torr and 0.1 nm/s). For C-V and J-V measurements, Au top electrodes and Al bottom electrodes were deposited by an ex-situ evaporation process. AFM and XRD measurements were carried out to characterize the microstructures of pentacene films. The measurements were carried out in air.

3. Results and Discussion
Figure 1 shows the C-V characteristics for the pentacene based MOS diodes with different gate insulators. It is found that pentacene based MOS diodes with HfO2 and HfON gate insulators show a small $V_{fb}$ shift, while Au/pentacene/SiO2/n+-Si(100) MOS diodes show a large positive $V_{fb}$ shift. And C-V characteristics for pentacene based MOS diodes with SiO2 and HfO2 gate insulators are obviously frequency dependent. Interestingly, the C-V characteristics of the pentacene based MOS diodes with HfON gate insulators almost do not show frequency dependence. Moreover, the accumulation capacitances of the pentacene based MOS diodes with SiO2 and HfO2 gate insulators are larger than the capacitances of gate insulators, which might be attributed to the effects of peripheral region [4]. Figure 2 shows the J-V characteristics for the MOS diodes. It is found that the leakage current ratios $J_{on}$/ $J_{off}$ for the pentacene based MOS diodes with SiO2 and HfO2 gate insulators are very high, even in the depletion condition, which further proves the effects of peripheral region. However, the accumulation capacitances of pentacene based MOS diodes with HfON gate insulators are lower than the capacitances of HfON insulators, and the $J_{off}$ of the pentacene based MOS diodes with HfON gate insulators is lower compared to other two types of pentacene based MOS diodes. It seems that the electrical properties of the pentacene based MOS diodes with HfON gate insulators are not strongly affected by the effects of peripheral region.

Figure 3 shows the AFM images of 1.6 nm-thick pentacene films on different gate insulators. Clearly, the nucleation density for pentacene films on HfO2 is lower compared to the pentacene films on SiO2 and HfON. From Fig. 4, one can see that the grain sizes of 20 nm-thick pentacene films on HfO2 and HfON are larger than that of 20 nm-thick pentacene films on SiO2. It is known that the surface energy of HfO2 (34 mJ/m²) matches well with the surface energy of orthorhombic pentacene (38 mJ/m²) [5], which leads to low nucleation density of pentacene films. Because the surface energy of SiO2 (61 mJ/m²) is not compatible with that of pentacene, the nucleation density for pentacene films grown on SiO2 is high. And high nucleation density usually leads to small grain size. Therefore, the grain size of pentacene films grown on HfO2 is larger than that of pentacene films grown on SiO2. For pentacene films grown on HfON, the high RMS roughness of HfON causes high nucleation density [6]. However, due to the low surface energy of the HfON [7], pentacene islands grow up as film thickness increases. As a result, 20 nm-thick pentacene films grown on HfON also have large grain sizes. From XRD spectra (not shown), it is found that the pentacene films on the three different types of gate insulators have different crystalline qualities.

4. Conclusion
Pentacene based MOS diodes with HfON gate insulators were investigated. It was found that the MOS diodes show a small $V_{fb}$ shift and negligible frequency dispersion in C-V characteristics, which might be attributed to the excellent interface properties and microstructures of pentacene films.

Acknowledgements
This work was partially supported by a Grant-in-Aid for Scientific Research on Priority Areas (A) (No. 19206039) from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

References
Fig. 1  C-V characteristics for the pentacene based MOS diodes with (a) SiO2, (b) HfO2, and (c) HfON gate insulators; the insets show the schematic diagrams of the MOS diodes.

Fig. 2  J-V characteristics for Au/pentacene/insulator/n^-Si(100) and Au/insulator/n^-Si(100) MOS diodes.

Fig. 3  AFM images of 1.6 nm-thick pentacene films grown on (a) SiO2 with an RMS roughness of 0.15 nm, (b) HfO2 with an RMS roughness of 0.17 nm, and (d) HfON with an RMS roughness of 0.24 nm.

Fig. 4  AFM images of 20 nm-thick pentacene films grown on (a) SiO2, (b) HfO2, and (d) HfON.