

Pr-silicate Ultrathin Films for High-k Gate Dielectrics Prepared by Metal-Organic Chemical Vapor Deposition:

Yoshishige Tsuchiya¹; Hirotugu Fujita²; Hiroshi Mizuta³; Hiroshi Nohira²; Takeo Hattori²; Shunri Oda¹

¹Tokyo Institute of Technology, Research Centre for Quantum Effect Electronics, 2-12-1, O-Okayama, Meguro-ku, Tokyo 152-8552 Japan

²Musashi Institute of Technology, Department of Electrical and Electronic Engineering, 1-28-1, Tamazutsumi, Setagaya-ku, Tokyo 158-8557 Japan

³Tokyo Institute of Technology, Department of Physical Electronics, 2-12-1, O-okayama, Meguro-ku, Tokyo 152-8552 Japan

Technological need of overcoming an issue of the leakage current with ultrathin gate oxide films has driven the research on alternative high-k materials. Exploring new gate insulator materials with a higher dielectric constant is exceedingly important for further scaling down of device dimensions. Lanthanide oxides are attractive candidates for the post HfO₂ era. In Ref. [1], a high dielectric constant of ~31 was reported for the Pr₂O₃ film grown on the Si substrate by electron-beam evaporation. Their results showed Pr-based composites are potentially useful, but, for the practical purpose, the chemical vapor deposition (CVD) technique is more important, and there are few reports on the Pr-base composites grown by the CVD. In this paper, we report on material and electrical properties of Pr-silicate ultrathin films grown by using the metal-organic chemical vapor deposition (MOCVD) technique. A p-type Si (100) wafer was used as substrates and placed on a susceptor in a lateral flow-type deposition chamber. Pr(DPM)₃ was adopted as a source for praseodymium and introduced into the chamber by the Ar carrier gas. Since oxygen atoms are contained in the precursor, we did not use any oxidizing agent, and the source was continuously supplied to the chamber. Total pressure in the reaction chamber was maintained at 1.5 Torr during deposition. Films were grown at various substrate temperatures of 460, 600, and 770 °C. Al K α excited O 1s photoelectron spectra of the film grown at 770 °C showed that a Pr-silicate thin layer was formed inside the film. The band gap, the conduction and valence band offsets at the interface were estimated as 6.3 eV, 3.2 eV and 2.0 eV, respectively, from the measurement of O 1s and angle-resolved valence band spectra. These values are larger than those reported for Pr₂O₃[2]. With decreasing a growth

temperature, both the band gap and the band offsets decreased. This result suggests that the Pr/Si ratio in the Pr-silicate varies with deposition temperature. Even in the film deposited at lowest temperature, 460 °C, both conduction and valence band offsets are larger than 1 eV, which is a general criterion to judge if the material is appropriate for reducing the leakage current when it is used as a gate dielectric. The capacitance equivalent thickness (CET) and the leakage current density (J_g) were estimated from the C-V and J-V measurements for the Au/Pr-silicate/Si MIS diode structure. The chemical oxide prepared on the Si substrates prior to the deposition improved electrical properties. The CET of 1.5 nm and J_g of 9.6×10^{-6} A/cm² at V_{fb} -1 V were obtained for the film deposited at 770 °C. [1] H. J. Osten et al., IEDM Tech. Dig., 653 (2000). [2] H. J. Osten et al., Appl. Phys. Lett. 80, 297 (2002).