Title	溶液プロセスにより形成したイットリウムドープ酸化フニウム-ジルコニウムを用いた酸化物チャネル強誘電体ゲートトランジスタの研究
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Investigation of Oxide-Channel Ferroelectric-Gate Transistor using Yttrium Doped Hafnium- Zirconium Oxide Fabricated by Solution Process

Doctoral Degree
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1. Research Content

1.1. Background

Discovery of ferroelectricity in HfO_2 has gain great interest to develop future high density ferroelectric random-access memory (FeRAM) devices. Ferroelectricity in HfO_2 is due to the formation of orthorhombic phase. This phase is considered as metastable phase and can be stabilized by doping. The most widely used doping elements are Al, Si, Y and Zr. In particular, Zr doped HfO_2 or Hafnium-Zirconium Oxide (HZO) has gained most significant attention because of high remnant polarization with wide doping range. Atomic layer deposition (ALD) and sputtering are usually used to fabricate HfO_2. On the other hand, inexpensive devices can be fabricated by CSD method for various applications at low cost. However, there are only a few reports on CSD for ferroelectric HZO films and doping effect in HZO films by CSD has not been investigated.

In this work, ferroelectric yttrium-doped hafnium-zirconium oxide (Y-HZO) has been realized by the chemical solution deposition (CSD) and applied to gate insulator of ferroelectric gate thin film transistor (FGT). Oxide semiconductor or conductor, In_2O_3 or In-Sn-O (ITO) was used as a channel, which was fabricated by CSD, too

1.2. Results and discussion

At first, ferroelectric properties of Y-HZO films fabricated by CSD with various annealing temperature and environment was studied in **chapter 3**. Samples annealed at 600–800 °C in a vacuum, N_2 and O_2 environment showed ferroelectricity, which was confirmed by the polarization–electric field (shown in figure 1) and capacitance–voltage measurements. It was found that the ferroelectric properties were dramatically improved when the samples were annealed in a vacuum at 800 °C and the leakage current was decreased, compared to the Y-HZO films annealed at 800 °C in oxygen and nitrogen.

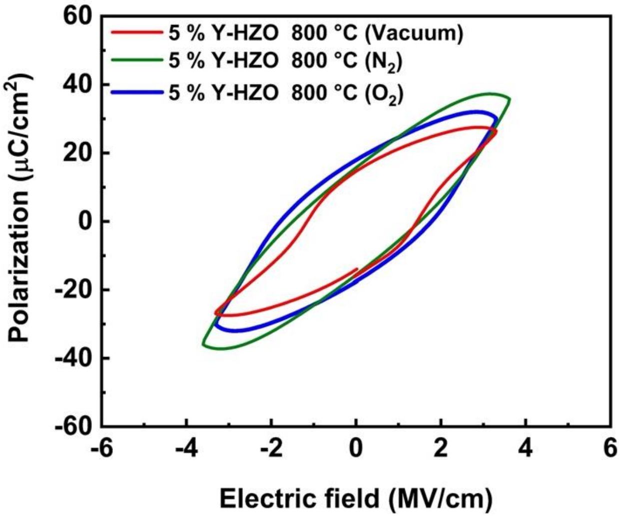

Figure 1: Electrical properties of Y-HZO annealed in various annealing environment

Next, stability of ferroelectricity in HZO films deposited by sputtering and CSD has been investigated for oxide channel ferroelectric-gate thin film transistor application **in chapter 4**. Since it is known that the ferroelectric orthorhombic phase is metastable in HfO_2-baseed ferroelectric materials and we need to deposit oxide channel layer on the ferroelectric gate insulator followed by the annealing process, the robustness of the ferroelectricity is important to fabricate FGTs. After confirming the ferroelectricity of both sputtered HZO and CSD yttrium-doped HZO (Y-HZO) films, indium-tin-oxide (ITO) was deposited by sputtering on sputtered HZO or CSD Y-HZO layer to fabricate metal-ferroelectric-semiconductor (MFS) structure. Then, the MFS structures were re-annealed in N_2 environment for 15 min to examine the robustness of ferroelectricity and electrical properties were measured as shown in figure 2 (a) and (b). Interestingly, it was found that the sputtered HZO films became paraelectric which was confirmed by both X-ray diffraction (XRD) pattern and electrical measurements. On the other hand, the CSD Y-HZO films showed ferroelectric nature even after the re-annealing with a negligible monoclinic phase.

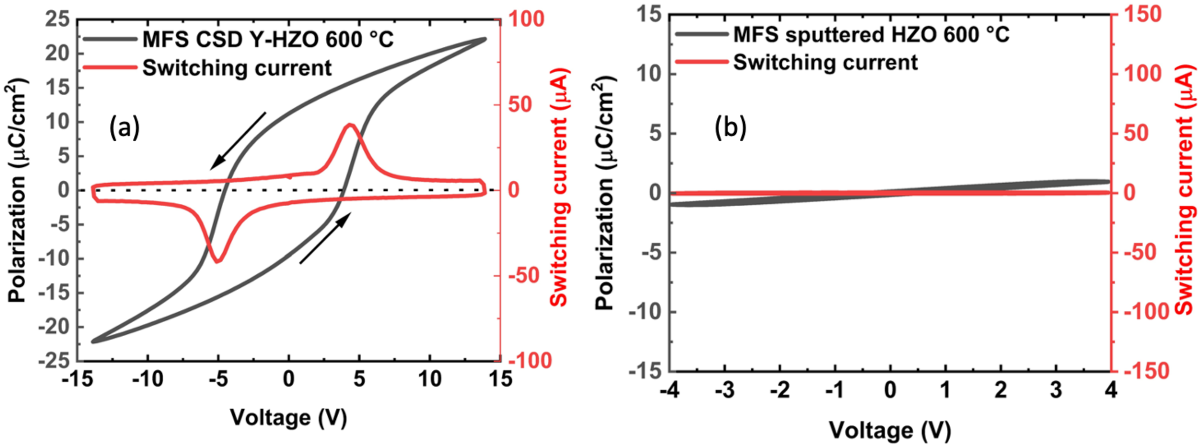

Figure 2: Electrical properties of HZO after re-annealed at 600 ^{o}C (a) CSD Y-HZO (b) sputtered HZO.

Next, ferroelectric gate transistor (FGT) with Y-HZO gate insulator and oxide channel with various thickness of In_2O_3 and ITO were fabricated and characterized in **chapter 5**. Figure 3(a) and (b) shows schematic illustration and photograph of FGT, where both Y-HZO and channel layers were fabricated by CSD. First, ferroelectric properties of vacuum and oxygen Y-HZO in the metal–ferroelectric–semiconductor (MFS) structure with 5-22 nm thick In_2O_3 and 6-24 nm thick ITO, have been confirmed by polarization-voltage (P-V) and capacitance-voltage (C-V) characteristics. The C–V curves showed clear butterfly loops showing the depletion of In_2O_3 and ITO layer. Vacuum annealed samples show good ferroelectric properties as compared to oxygen annealed samples. Secondly, the device performance of FGTs for both vacuum and oxygen annealed samples has been evaluated with various thickness of In_2O_3 and ITO channel layer as shown in figures 3(c) and (d). The fabricated FGTs exhibited typical n-channel transistor operation with a counterclockwise hysteresis loop in their transfer curves due to the ferroelectric nature of the Y-HZO-gate insulator. It was found that FGT shows low sub-threshold voltage swing (SS), high on/off drain current ratio of 10^6, large on current, and memory window for vacuum annealed sample. In contrast, FGTs with Y-HZO gate insulator annealed in oxygen show degraded device performance due to high leakage current. In this thesis, good electrical properties have been demonstrated for oxide channel FGTs with ferroelectric Y-HZO gate insulator fabricated by CSD with vacuum annealing process, which can be a suitable candidate for future low-cost ferroelectric device and memory applications.

2. Research Purpose

The objective of this study is to develop the chemical solution process for HfO_2-based ferroelectric films and to apply them to ferroelectric-gate transistors (FGTs). To realize FGTs, oxide semiconductor or conductor, In_2O_3 or In-Sn-O (ITO) was used as a channel and yttrium doped HZO was used as gate insulator material. To best of my knowledge, this is the first report of thin film transistors (TFTs) using solution processed HZO ferroelectric gate insulator with solution processed In_2O_3 or ITO oxide channel.

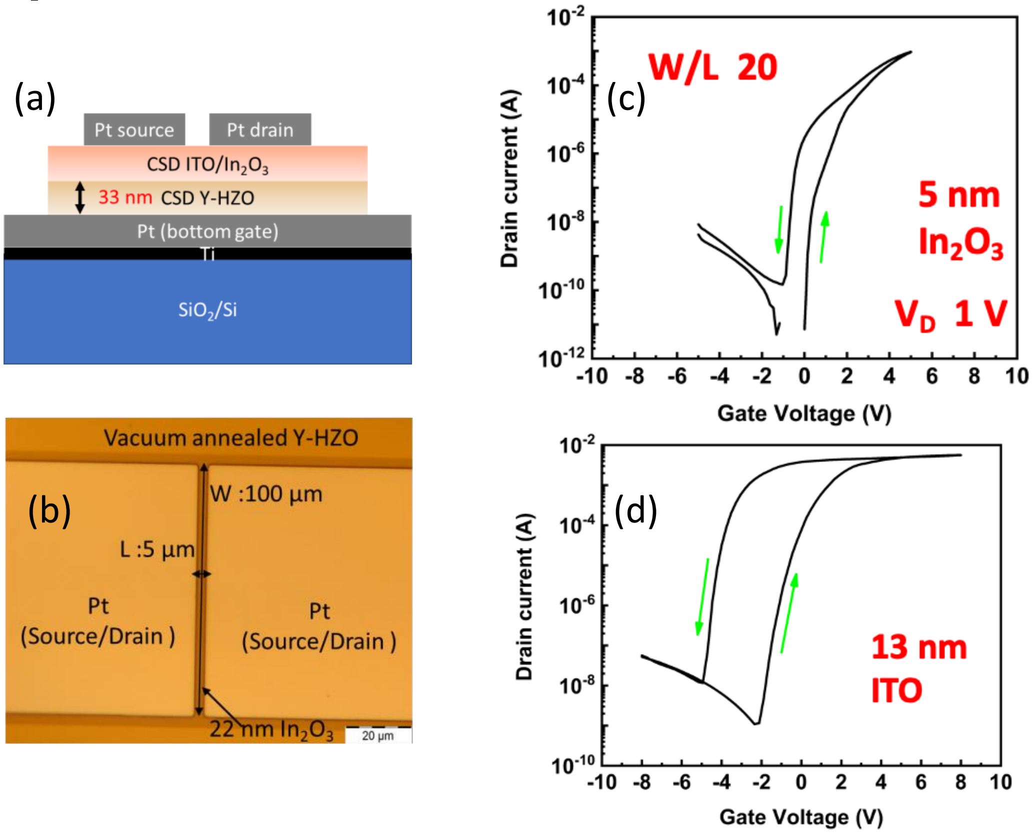

Figure 3: (a) schematic illustration (cross section), (b) photograph of FGT (top view), I_d-V_g characteristics of FGT with (c) 5-nm-thick In_2O_3 channel and (d) 13-nm-thick ITO channel.

3. Research Achievements

A) <u>Publications in peer-reviewed Scientific journals</u>

1. Mohit, K. Haga, E. Tokumitsu, "Electrical properties of yttrium-doped hafnium-zirconium dioxide thin films prepared by solution process for ferroelectric gate insulator TFT application", *Jpn. J. Appl. Phys.*, 59, pp. SMMB02-1-9, 2020.
2. Mohit, T. Murakami, K. Haga and E. Tokumitsu, "Impact of annealing environment on electrical properties of yttrium-doped hafnium-zirconium dioxide thin films prepared by the solution process" *Jpn. J. Appl. Phys.* 59 SPPB03, 2020.
3. Mohit, Takaaki Miyasako and Eisuke Tokumitsu, "Indium oxide and indium-tin-oxide channel ferroelectric gate thin film transistors with yttrium doped hafnium-zirconium dioxide gate insulator prepared by chemical solution process", *Jpn. J. Appl. Phys* 60 SBBM02, 2021.
4. Mohit, Shinji Migita, Hiroyuki Ota, Yukinori Morita and Eisuke Tokumitsu, "Thermal stability of ferroelectricity in hafnium-zirconium dioxide films deposited by sputtering

and chemical solution deposition for oxide-channel ferroelectric-gate transistor applications” 14 041006, 2021.

B) Manuscripts submitted or under preparation

1. Mohit, Diasuke Hirose, Yuzuru Takamura and Eisuke Tokumitsu,“Development of novel DNA biosensor using HfO_2 gate insulator and In_2O_3 channel BioTFTs prepared by solution process” (under preparation)

C) Conferences

International Conferences:

1. Mohit, Ken-Ichi Haga, Eisuke Tokumitsu, “Preparation of Ferroelectric Yttrium-doped Hafnium-Zirconium Dioxide Thin Films by Solution Process”, International Workshop on Dielectric Thin Films for Future Electron Devices: Science and Technology (IWDTF 2019), Tokyo, Japan, Nov 2019 (Poster)
2. Mohit, Takaaki Miyasako and Eisuke Tokumitsu, “Fabrication of Indium-Tin-Oxide Channel Ferroelectric-Gate Thin Film Transistors using Yttrium Doped Hafnium-Zirconium Dioxide by Chemical Solution Process”, SSDM, Toyama September 2020 (Oral).
3. Mohit, Shinji Migita, Hiroyuki Ota, Yukinori Morita and Eisuke Tokumitsu, “Robustness of ferroelectricity in hafnium-zirconium dioxide films deposited by sputtering and chemical solution deposition for ferroelectric transistor applications”, ECS PRiME, Hawaii, USA, October 2020. (Poster)
4. Mohit, Daisuke Hirose, Eisuke Tokumitsu and Yuzuru Takamura, “DNA biosensing using Indium Oxide thin film transistor with HfO_2 as gate insulator prepared by solution process”, E-MRS spring 2021, France, May 2021. (Oral-accepted)
5. Mohit and Eisuke Tokumitsu “Preparation of ferroelectric lanthanum doped hafnium-zirconium oxide thin films by solution process”, EM-Nano 2021, May 2021. (Oral-accepted)

Domestic Conferences:

1. Mohit, Yoshiaki Sumitomo, Ken-chi Haga, Keisuke Ohdaira, Eisuke Tokumitsu, “Sub-20 nm thick ferroelectric Hf-Zr-O films fabricated by solution process for ferroelectric-gate TFT applications”, Thin Film Materials & Devices Meeting (TFMD 2019), Kyoto, Japan, November 2019 (Poster).
2. Mohit, Jyotish Patidar, Ken-Ichi Haga, Eisuke Tokumitsu, “Fabrication of ferroelectric hafnium-zirconium dioxide thin films by solution process”, JSAP spring meeting 2020, Tokyo, March 2020 (Oral)
3. Mohit, Ken-Ichi Haga, Eisuke Tokumitsu, “Effect of Annealing Environment on Ferroelectric Properties of Hf-Zr-O (HZO) Thin Films Prepared by Solution Process”, Ferroelectric Meeting and Their Applications (FMA 37), Kyoto, Japan, May 2020 (Oral).
4. Mohit, Eisuke Tokumitsu, “Ferroelectric Properties of Hafnium-Zirconium-Dioxide Prepared by Chemical Solution Process for MFM and MFS Structures”, JSAP autumn meeting 2020, Kyoto, September 2020 (Oral)
5. Mohit, Shinji Migita, Hiroyuki Ota, Yukinori Morita and Eisuke Tokumitsu, Fabrication of ferroelectric gate thin film transistors using CSD Y-HZO and sputtered HZO with sputtered ITO channel JSAP spring meeting 2021, Tokyo, March 2020 (Oral-submitted)

6. Yuki Hara, Mohit, Shinji Migita, Hiroyuki Ota, Yukinori Morita and Eisuke Tokumitsu Improvement of stability of ferroelectricity in sputtered Hf-Zr-O films, FMA 38, Kyoto Japan, May 2021 (Oral).

D) Award and Research grant

Principal investigator for JAIST Research Grant (Fundamental Research) 2020 (on going).

Keywords: Ferroelectric Hafnium dioxide, oxide channel, Ferroelectric Gate Transistor, solution process, thin films.