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Lifetime of hafnium oxide dielectric in thin-film devices fabricated on deformable softening polymer substrate



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ABSTRACT

In this work, we investigate the electrical behavior and reliability of thin-film devices using a high-k dielectric on top of softening polymer. Hafnium oxide (HfO₂) 50 nm thick was used for gate dielectric in both capacitors and thin-film transistors (TFTs) and is deposited by atomic layer deposition at 100 °C. A thermoset thiol-ene/acrylate shape memory polymer (SMP) is used as flexible substrate with softening properties. The SMP belongs to a class of mechanically active materials used to store a metastable shape and return to a globally stable shape upon activation by stimuli, such as temperature, which softens the polymer via a decrease in storage modulus. An average dielectric constant of 13.6 was obtained for the HfO₂ layer after an annealing treatment at 200 °C for two hours in forming gas. Here, a clear dependence between the electrical behavior and the device dimensions was observed. In the same experimental process, indium-gallium-zinc-oxide TFTs with different dimensions were fabricated showing mobility values of approximately 17 cm²/V-s, presenting similar dependence on channel dimensions. Finally, the lifetime projection of the HfO₂ film was estimated from a time-dependent-dielectric breakdown and leakage current analysis.

1. Introduction

The technology of low-temperature processing semiconductors has enabled the development of electronic devices on top of flexible substrates such as plastics and polymers [1–4]. This has received much attention in recent years due to the huge potential applications in medical devices, flexible displays and self-powered circuits among others [5–9]. However, the electrical instability of flexible electronic devices has become an issue for the development of reliable applications. There are different studies which focus on thin-film transistors (TFTs) as the basic unit for future complex circuits and on the improvement of its threshold voltage (V_{TH}) stability [10–12]. On the other hand, time-dependent dielectric breakdown (TDDB) technique is a wellknown characterization method to predict dielectric lifetime by applying different voltage ranges (or electric fields) to a dielectric layer (or capacitor). This characterization is also used to define the operation ranges for electronic circuits and it is commonly used for single-crystalline silicon (Si) technology, which is incompatible with flexible substrates [13–15].

In this work, we present the electrical stability and lifetime of hafnium oxide (HfO₂) dielectric on top of softening thiol-ene/acrylatebased shape memory polymer (SMP) as flexible substrate, which has the ability to remember a programmed shape after deformation and then to return to its original form when an external stimulus is applied [16–19]. As previously reported, this activation by stimulus, such as temperature, humidity, light or a combination of these can drop the polymer storage modulus from the order of GPa to MPa [20–22]. Because of the change in the storage modulus, this softening polymer is compatible with the paradigm "stiff for insertion and soft chronically," useful for a broad class of implantable bioelectronic applications. To our

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Fig. 1. a) 3D diagram of Indium-Gallium-Zinc-Oxide (IGZO) thin-film transistors (TFTs) and metal-insulator-metal (MIM) capacitors. b) Optical micrographs of the fabricated MIM capacitors (100×100 , 100×200 and $200 \times 200 \mu$ m) using 50 nm of hafnium oxide as insulator. c) Optical image of the thin-film devices on top of the shape memory polymer (SMP) based on thiol-ene/acrylate, as softening substrate, after being cut and released from the glass carrier.

knowledge, this is the first time an electrical reliability analysis of devices based on High-K dielectric, more specifically HfO_2 , have been reported on a softening polymer as substrate. Also, this SMP has shown resilience through photolithography and aggressive fabrication processes reaching temperatures up to 250 °C without significant mass loss [23]. The HfO_2 was deposited by atomic layer deposition (ALD) to fabricate metal-insulator-metal (MIM) capacitors and indium-gallium-zinc oxide (IGZO) TFTs on top of SMP as shown with the 3D structure in Fig. 1a. The lifetime of the HfO_2 was obtained by analyzing the TDDB of the fabricated capacitors with different dimensions (Fig. 1b).

2. Experimental

All the steps for SMP synthesis were performed in a fume hood using, as previously reported: 1, 3, 5 Triallyl 1, 3, 5-triazine-2, 4, 6(1 H, 3H, 5H)-trione (TATATO), Tricyclo [5.2.1.02, 6] decanedimethanoldiacrylate (TCMDA), Tris [2-(mercaptopropionyloxy) ethyl] isocyanurate (TMICN) and 2, 2-Dimethoxy-2-phenyl-acetophenone (DMPA) [22]. The obtained polymer on a glass slide (5 \times 7.5 cm) was 50 µm thick. The SMP roughness obtained by an atomic force microscopy (AFM) Veeco (Dimension 5000) was of $R_a = 0.19 \text{ nm}$ (Fig. 2a). Then, the MIM capacitors and IGZO TFTs were fabricated on top of the SMP substrate without any posterior treatment or passivation layer as previously reported [23]. The device fabrication was carried out using photolithographic processes, as well as wet and dry etch. A gold layer, deposited by thermal evaporator, of 100 nm was used for bottom contact (Gate) and another as top contact (Drain/Source). For both devices 50 nm of HfO₂ by ALD at 100 °C was deposited, sharing the same gate dielectric. A tetrakis(dimethylamido) precursor was used for hafnium and DIW for the oxidation. The HfO2 thickness was measured by ellipsometry and cross-section SEM (Fig. 2b). An RF sputtering and a In:Ga:Zn:O = 1:1:1:4 target were used for the semiconductor deposition at room temperature. During the entire fabrication process, the devices were exposed to a maximum temperature of 100 °C with a posterior annealing of 200 °C for one hour (in forming gas, 5% H₂) after fabrication. Posteriorly, the SMP substrate was cut and released as shown in Fig. 1c. A cascade probe-station with a Keithley 4200 and HP/Agilent 4284A Precision LCR Meter instrument were used for the electrical characterization of the capacitors and TFTs.

3. Results and discussion

Electrical characterization of the metal-insulator-metal (MIM)



Fig. 2. a) Atomic force microscopy (AFM) used to measure the SMP roughness ($R_a = 0.19$ nm). b) Cross-section scanning electron microscopy (SEM) of HfO₂ (50 nm) on Si/SiO₂ wafer. Analysis of the average capacitance density analysis using c) voltage sweep @1 MHz and d) frequency sweep at 5 V, using twelve MIM capacitors with 50 nm HfO₂ on top of SMP substrate.

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