

A water-induced high- k yttrium oxide dielectric for fully-solution-processed oxide thin-film transistors



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ABSTRACT

In this work, we develop a simple and eco-friendly water-inducement method for high- k yttrium oxide (YO_x) dielectric. To prepare YO_x thin films at low temperature, yttrium nitrate and deionized water were used as the source materials. No toxic organic materials were required in the YO_x coating process. The YO_x thin film annealed at 350 °C showed a low leakage current density of $2 \times 10^{-9} \text{ A/cm}^2$ at 5 MV/cm and a large areal-capacitance of 448 nF/cm² at 1 kHz. On the basis of its implementation as the gate dielectric, the fully-water-induced In_2O_3 TFT based on YO_x exhibited a high field-effect mobility of 15.98 cm²/Vs, excellent subthreshold swing of 75 mV/dec, an on/off current ratio of 6×10^6 , and a negligible hysteresis of 50 mV. The as-fabricated TFT operated at a low voltage ($\sim 1.5 \text{ V}$) and showed high drain current drive capability, enabling oxide TFT with a water-induced high- k dielectric for use in backplane electronics for low-power mobile display applications.

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1. Introduction

As potential alternatives to thin-film transistors (TFTs) based on conventional silicon technologies, amorphous metal-oxide TFTs received considerable attention due to the high electron mobility, high transparency, excellent large-area uniformity, and solution processability in electronic applications for displays and sensor arrays [1–4]. In particular, low power consumption is one of the key issues for the mobile applications, such as smart phones and tablet personal computers, due to the limit capacity of the rechargeable lithium-ion battery [5]. The use of high- k materials as the dielectric is an effective approach to enhance the capacitive coupling and reduce the power consumption. Many studies have examined the adoption of binary high- k dielectric materials, such as Y_2O_3 [6], HfO_2 [7], ZrO_2 [8], Ta_2O_5 [9], and Al_2O_3 [10] in the metal-oxide TFTs. Since Nomura et al. firstly reported IGZO TFTs based on a Y_2O_3 dielectric in 2004 [1], there are few reports discussing the preparation of Y_2O_3 dielectric and its application in TFT devices.

Considering its desirable electrical properties (wide band gap $\sim 6 \text{ eV}$, relatively high dielectric constant ~ 18) and chemical properties (solubility in common solvents and stable, complete stoichiometry), Y_2O_3 dielectric material is regarded as a good choice in the future display industry.

To fabricate the dielectric thin films with low cost using a simple method, solution processes such as inkjet printing, dip-coating, and spin-coating techniques are proved to be promising. In addition, in order to reduce the annealing temperature for solution-processed dielectrics, several research groups have proposed several novel approaches, including chemical energetic combustion process through an oxidizer and fuel [11], UV/ozone photo-annealing method [12], O_2 plasma-assisted treatment [13], and impurity doping method [14,15]. However, the inert atmosphere is needed to handle precursor solutions, which makes the process complicated. Furthermore, the toxic organic solvents, e.g. 2-methoxyethanol (2-ME) and dimethyl formamide (DMF), are usually used. Additional additives or follow-up processing undoubtedly increase the environmental damage and the fabrication cost. In our previous reports, solution-processed high- k ZrO_x dielectrics were achieved under various annealing conditions and the TFT devices based on the high- k ZrO_x dielectrics exhibited excellent electrical performance

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[16–19]. However in these reports, the ZrO_x dielectrics were fabricated using toxic organic-based solution, which is harmful to the environment and the sustainable development.

Considering the sustainable benefits and the strong potential applications of the solution-based process, the eco-friendly and organic-species-free precursors are needed in the future large-area, flexible display industry. As a solvent, water meets the current environmental awareness restricting the use of ecologically harmful substances and process. Instead of the often used organic-related solvents, the water-inducement synthesis is considered to be healthier, safer and environmentally friendlier. Meanwhile, unlike conventional precursors, the water-induced solutions are insensitive to ambient moisture. Therefore, an inert atmosphere to store and handle the precursor solutions is not necessary [20]. More importantly, as the coordinating bond between the metal cation and the neighboring aquo ion is relatively weak (electrostatic interaction), it is easily broken compared with the covalent bonds in conventional precursors [21]. Therefore, water-inducement process is considered to be a promising technique to fabricate high-quality high- k dielectrics for eco-friendly and low-temperature oxide TFTs.

In this work, by using a water-inducement method, we established a nontoxic, low-temperature process for high- k YO_x thin films and applied the dielectrics in TFT devices. The effect of annealing temperature on the properties of YO_x thin films was systematically investigated. To verify the possibility of the water-induced YO_x thin film as gate dielectric, fully-water-induced TFT combined with In_2O_3 channel layer was also integrated and examined. By excluding toxic organic solvents, we demonstrate that this 'green' process can contribute to environmental safety and cost minimization simultaneously.

2. Experiments

The YO_x precursor solution (0.15 M) was prepared by dissolving yttrium nitrate hydrate $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ in 10 mL deionized water. The solution was stirred vigorously for 5.5 h under ambient condition before spin-coating. The precursor solution was spun on heavily-doped silicon ($\text{p}^+\text{-Si}$) substrates at 5000 r.p.m. for 20 s and baked at 130°C for 10 min. This procedure was repeated two times to obtain an appropriate thickness. The thin films were then annealed by a sequential process using UV/ozone treatment for 40 min and thermal annealing in the temperature range from 150 to 450°C for 1 h in air. For convenience, the YO_x thin films annealed at 150, 250, 350, and 450°C will be called as $\text{YO}_x\text{-150}$, $\text{YO}_x\text{-250}$, $\text{YO}_x\text{-350}$, and $\text{YO}_x\text{-450}$, respectively. The average thicknesses of $\text{YO}_x\text{-150}$, $\text{YO}_x\text{-250}$, $\text{YO}_x\text{-350}$, and $\text{YO}_x\text{-450}$ were around 30, 22, 17, and 15 nm, respectively.

To fabricate the TFT devices based on the YO_x dielectrics, In_2O_3 thin film was fabricated as the semiconductor channel layer by a solution process. 0.1 M indium nitrate hydrate ($\text{In}(\text{NO}_3)_3 \cdot \text{H}_2\text{O}$) was dissolved in deionized water, and the solution was stirred vigorously for 5 h under ambient condition and filtered through $0.2\ \mu\text{m}$ syringe filter before spin-coating. The In_2O_3 precursor solution was spun on YO_x dielectrics at 4000 r.p.m. for 30 s, the laminated thin films were subsequently annealed at 250°C for 1 h. Finally, Al source and drain electrodes were deposited on In_2O_3 channel layer by thermal evaporation through a shadow mask. In this report, the channel length and width of the TFTs were 100 and $1000\ \mu\text{m}$, respectively. The schematic of the TFT structure used in this work is shown in Fig. 1. For comparison, the In_2O_3 TFT based on thermally-grown SiO_2 (100 nm) dielectric was also fabricated, which is referred as the reference device.

The thermal behavior of the YO_x xerogel was monitored under air ambient using a thermal-gravimetric analyzer (TGA, Pyris 1)

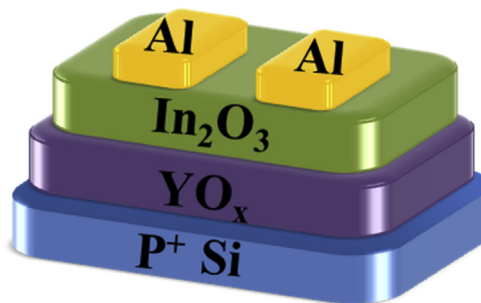


Fig. 1. Schematic structure of In_2O_3 TFT with YO_x gate dielectric.

with a heating rate of $10^\circ\text{C}/\text{min}$. The crystal structures of YO_x thin films were investigated by an X-ray diffractometer (XRD, X'Pert-PRO MPD and MRD, PANalytical, Holland) with a $\text{CuK}\alpha 1$ radiation. The surface morphologies of YO_x thin films were measured by using an atomic force microscope (AFM, SPA-400, Seiko). The chemical compositions of the YO_x thin films were analyzed by X-ray photoelectron spectroscopy (XPS, ESCALAB 250). In order to evaluate the dielectric properties of the YO_x thin films, the capacitors with a structure of $\text{Al}/\text{YO}_x/\text{p}^+\text{-Si}$ were fabricated and investigated by using an impedance analyzer (Agilent 4294A). The electrical properties of the YO_x capacitors and the integrated TFT devices were measured by using a semiconductor parameter analyzer (Keithley 2634B) in a dark box.

3. Results and discussion

The combination of metal nitrates and water as the precursors and solvent provides a simple and unique structure in a solution state. Water is a frequently-used solvent with a high static dielectric constant of ~ 80 at room temperature, which favors the dissociation of ionic specie and acts as the σ -donor molecule that reacts as a nucleophilic ligand [21]. The details of solvated cation and hydrolysis reaction were described in our previous report [19] and in Ref. [20].

To understand the formation of YO_x thin films from water-based precursor solution, TGA measurement was performed and the result is shown in Fig. 2. The initial decrease in weight of the YO_x xerogel below 230°C is related to the decomposition of the residual nitrate species originating from the metal nitrate salt [22]. The continuous decrease in weight is mainly attributed to the dehydration and the alloy reaction of yttrium hydroxide precursor

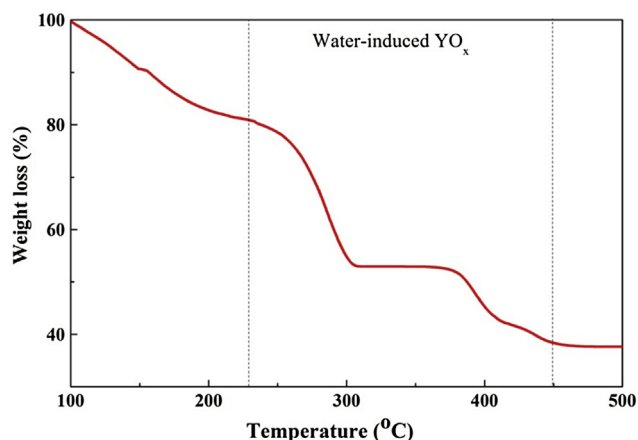


Fig. 2. Thermal behavior of YO_x xerogel.

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