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Review

Sol-gel metal oxide dielectrics for all-solution-processed electronics

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ABSTRACT

Metal oxide (MOx) dielectric materials are considered to be a key element for diverse thin-film electronic systems owing to their superior electrical and mechanical properties. While the vast majority of conventional methods for processing these materials rely on vacuum-based deposition, which seriously limits their potential for practical applications, the solution-based deposition of sol-gel MOx materials can ideally reduce both material and processing costs by introducing large-area printing methods. Nonetheless, the fundamental understanding of their film-forming mechanisms and optimum device architectures is still immature, and significant efforts should be devoted to reducing both temperature and duration for MOx polycondensation and film densification. This article reviews recent advances in solution-based MOx dielectric materials, with a specific focus on the extensive categorization of their structures/compositions and on advanced approaches for realizing ultimate material properties and next-generation device platforms. We expect that this review will manifest the strong potentials of sol-gel MOx dielectric materials toward all-solution-processed low-voltage transparent electronics with freedom in mechanical form factors along with unrivaled performance.

Contents

Abbreviations: AlPOx, aluminum phosphate; AlOx:Na, sodium-doped aluminum oxide; ZircSOx, zirconium sulfate; HafSOx, hafnium sulfate; AlOx, aluminum oxide; ZrOx, zirconium oxide; HfOx, hafnium oxide; YOx, yttrium oxide; GdOx, gadolinium oxide; Zr/Al/ZrOx, zirconium-aluminum-zirconium oxide trilayer; HfAlOx, hafnium aluminum oxide; HfLaOx, hafnium lanthanum oxide; ZrAlOx, zirconium aluminum oxide; TiAlOx, titanium aluminum oxide; LaAlOx, lanthanum aluminum oxide; PA, phosphonic acid molecule; M-SAND, MOx stacked with "self-assembled nanodielectrics"; PS, polystyrene; PI, polyimide; PVP, poly(4-vinylphenol); GPTS, 3-glycidoxypropylane; AlOOH, aluminum oxyhydroxide; PαMS, poly(α-methylstyrene); PMMA, poly(methyl methacrylate); P(MMA-co-MAA), poly(methyl methacrylate-co-methacrylic acid); PVAlA, poly (vinyl alcohol)-co-poly(vinyl acetate)-co-poly(itaconic acid); P(VDF-HFP), poly(vinylidenefluoride-co-hexafluoropropylene); PC, bisphenol-A-type polycarbonate; P(VDF-TrFE), poly(vinylidene fluoride-trifluoroethylene); TEOS-PU, triethoxysilane-capped polyurethane; CYELP, cyanoethyl pullulan; PVP-co-PMMA, poly(4-vinylphenol-co-methylmethacrylate); BTMH, 1,6-bis(trimethoxysilyl)hexane; SBA, sodium-beta-alumina; AlTiOx, aluminum titanate nanoparticles (AT); BTO, barium titanate nanoparticles ($BaTiO_3$); STO, strontium titanate nanoparticles ($SrTiO_3$); BST, barium strontium titanate nanoparticles; BZO, barium zirconia nanoparticles; ZrSiOx, zirconium silicate (zircon); BTH, 1,6-bis(triethoxysilyl)octane; CdS, cadmium sulfide; InO, indium oxide; ZnO, zinc oxide; SnO, tin oxide; ZTO, zinc-tin oxide; ZnGaO, zinc-gallium oxide; IGZO, indium-gallium-zinc oxide; IZO, indium-zinc oxide; ZnO:Li, Li-doped zinc oxide; GZTO, gallium-zinc-tin oxide; ITZO(ZITO), indium-titanium-zinc oxide; PTzQT-14, poly(2,5-bis(3-tetradecylthiophen-2-yl)thiophen-2-yl)thiophen-2-ylthiazolo[5,4-d]thiazole; PBTTT-C14, poly(2,5-bis(3-tetradecylthiophen-2-yl)thieno[3,2-b]thiophene); IYO, indium-yttrium oxide; SWCNT, single-walled carbon nanotubes C10-BTBT: 2,7-didecyl[1]benzothieno[3,2-b][1]benzothiophene; P3HT, poly(3-hexylthiophene); TIPS-Pentacene, bis(tri-isopropylsilylethynyl) pentacene; VOPc, vanadyl-phthalocyanine.

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1. Introduction

The progress toward practical applications of large-area flexible electronics such as information displays [1], wearable [2,3] and ultra-light [4] devices, and low-cost disposable circuits, [5,6] requires the development of high-performance electronic materials whose properties are well suited to each target application. Of equal importance are the corresponding film fabrication methods, and a variety of state-of-the-art printing techniques [7–9] such as inkjet [10,11], screening [12,13], gravure [14,15], and bar coating methods [16,17] have shown great promise for cost-effective film deposition on large-scale substrates. In this technological context, solution-processable high-performance electronic materials are at the forefront of materials science and have been extensively researched for use in a versatile device building block, i.e., thin-film transistor (TFT). Among major components in a TFT, a gate dielectric plays an important role because it manipulates the conductance of the semiconducting channel by accumulating charge carriers while its electrical insulation to minimize a leakage current is another critical requirement for minimal static dissipation. Nonetheless, a strong emphasis on high-performance semiconducting materials [18–26] has been major driving force within the TFT community; hence, it is now increasingly important to discover how to exploit dielectric material properties to realize the well-balanced ultimate TFT performance.

Two most critical metrics of desirable dielectrics are high areal capacitance and ideal electrical insulation, which can be essentially achieved by adopting a very thin layer of robust material with a high dielectric constant (k). However, most conventional methods for creating solid-state high-k inorganic dielectrics rely on vacuum-based deposition techniques (e.g., magnetron sputtering, chemical vapor deposition, atomic layer deposition, and etc.). These methods typically require the long process time not only for film deposition but also high-vacuum stabilization, and/or the post-deposition treatment at relatively high temperatures for film densification; thus, they have limited applicability for futuristic large-area flexible mass-production-oriented electronics on plastic substrates. Despite solution processability, polymer-based dielectrics typically exhibit relatively low dielectric constant and may suffer from substantial leakage current, particularly, at relatively low film thickness without dense cross-linking. Furthermore, typical polymeric insulators are not compatible with chemical treatments due to dissolution in harsh solvents or etching against reactive gases, and/or high-temperature annealing due to the low glass-transition temperatures; because of these facts, complicated additional steps are often called for to achieve all-solution-processed high-performance organic electronics.

Since the first report on metal oxide (MOx) film formation in 1962 [27], solution-processed MOx dielectric materials have been widely studied due to their high-k values [20,23,24,28], excellent optical transparency [29–31], and chemical/environmental stability [32–43]. Until now, however, major challenges in solution-based MOx have been associated with the post-deposition treatment for film densification, which generally necessitates high-temperature annealing (ca. 400 °C) and/or hour-long processing time that are unsuitable for the industry-scale commercialization. Consequently, recent research efforts have been directed to reduce the temperature and time for post-deposition processes by introducing various wet-chemical [32–37] and/or smart low-temperature treatments [38–45]. However, the abovementioned techniques have been primarily designed and employed almost exclusively for MOx semiconductors, while their broader applicability to MOx dielectric materials still remains unclear.

In this article, we present a comprehensive review of recent progress in the development of sol-gel-processed MOx dielectric films and their application to high-performance electronic devices, in particular, TFTs. Special emphasis is placed on the classification of diverse structural variations of sol-gel oxide dielectrics and the recently-unveiled physicochemical mechanism on low-temperature oxide layer formation without electrical performance compromised.

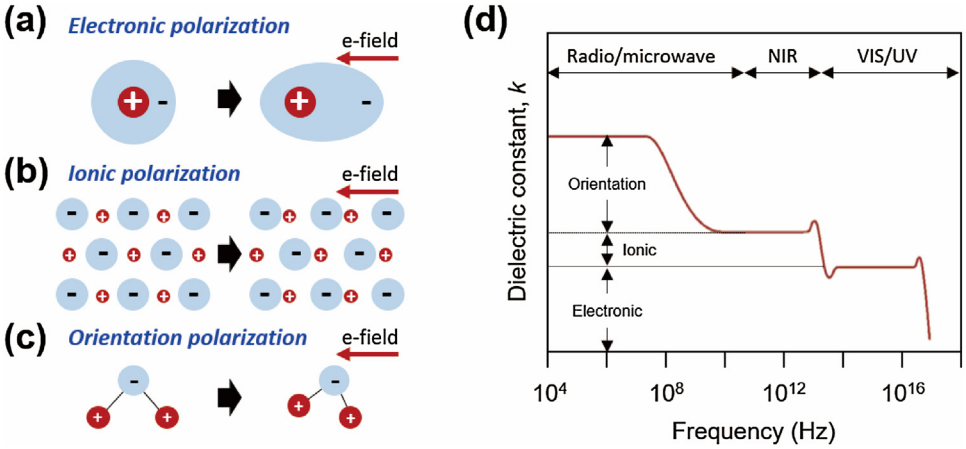

Fig. 1. The basic mechanisms of dielectric polarization. Schematics of (a) electronic polarization, (b) ionic polarization, (c) orientation polarization. (d) The contribution of electronic, ionic, and orientation polarization to the overall dielectric constant at the different frequency applied.