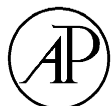
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Rare-earth oxide thin films as gate oxides in MOSFET transistors

Markku Leskelä* and Mikko Ritala

Department of Chemistry, University of Helsinki, P.O. Box 55, FIN-00014 Helsinki, Finland

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Abstract

In this paper, the possibilities of rare-earth oxides as gate dielectrics are discussed. The thin films have mostly been fabricated by physical vapor deposition methods. The rare earths most often studied are yttrium, lanthanum and gadolinium. The deposition of the gate oxide should be carried out under mild conditions, and therefore chemical deposition techniques are preferred. Atomic layer deposition of rare-earth oxides is introduced and special attention is given to the volatile precursors and deposition processes. The electrical properties of rare-earth oxide gate oxides will be highlighted. The results obtained are encouraging and the use of rare-earth oxides in gate stacks is possible. Especially, they may be important in connection to III–V compounds.
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Keywords: Rare-earth oxide; Gate oxide; MOSFET; ALD; CVD

1. Introduction

The scaling of gate dielectric thickness represents a major issue in the development of metal oxide semiconductor field effect transistors (MOSFET) of next generation. In sub-100 nm MOSFET devices, the so-far dominant SiO_2 should be made thinner than 15 Å but at these thicknesses tunneling leakage current through SiO_2 becomes unacceptably high [1]. Therefore, new dielectric materials with a higher permittivity are needed so that the gate oxide can be made thicker and yet obtain the same capacitance as the thinner SiO_2 would give. Thus, the equivalent thickness ($d_{eq} = (\varepsilon_{,SiO_2}/\varepsilon,\varepsilon)d = (3.9/\varepsilon,\varepsilon)d$) of the high-$k$ replacement should be less than 15 Å. On the other hand, silicon oxidizes easily from its surface, which increases the overall equivalent thickness ($d_{eq,tot} = d_{SiO_2} + d_{eq}$). To achieve the equivalent thicknesses required, the interfacial silicon oxide layer between silicon and the high-k material should be eliminated or controlled at a monolayer level. In fact, an ultra-thin SiO_2 layer would be beneficial as it ensures good interface characteristics.

The requirements of the new dielectrics can be divided into two categories: fundamental material properties and device processing, integration and performance issues [2]. The material properties include: high dielectric constant (>10), stability on silicon, low-interface trap densities, and smooth film morphology. Device processing includes deposition so that silicon does not oxidize and has good interface quality, gate and process compatibility and reliability. A number of materials are currently studied to replace SiO_2. Most promising of them are zirconium, hafnium and rare-earth oxides and silicates.

Compound semiconductors like GaAs have suffered from the lack of passivating and dielectric native oxide, and any other oxide has not worked successfully in compound semiconductor MOSFETs. However, recently it was shown that an epitaxial layer of Gd_2O_3 on GaAs surface could fulfill the MOSFET dielectric requirements [3]. This finding may start a new era for GaAs MOSFETs.

In this paper, the possibilities of rare-earth oxides as gate dielectrics are discussed. First, the oxide materials obtained by physical vapor deposition and their electrical properties as gate oxides are described. Then, atomic layer deposition (ALD) of rare-earth oxides is introduced. ALD technique is believed to become an important method for growing very thin films in microelectronics.

*Corresponding author. Fax: +358-9191-50198.
E-mail address: markku.leskela@helsinki.fi (M. Leskelä).

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2. PVD of rare-earth oxides and silicates

Rare-earth oxides are interesting candidates for gate oxides because according to the thermodynamic calculations they are stable on silicon [4]. The other advantages are good general thermal stability, relative high dielectric constants from 28 to 30 of La_2O_3, 18 Y_2O_3, and high conduction band offset with silicon [5,6]. Yet, another attractive feature of rare-earth oxides is the close lattice match with silicon. This offers the possibility for epitaxial growth, which in turn eliminates problems related to grain boundaries in polycrystalline films. The most often studied oxides are La_2O_3, Gd_2O_3, and Y_2O_3 [7] but Pr_2O_3 has been studied as well [8]. Because of the highest dielectric constant, La_2O_3 is a very attractive material but, on the other hand, it is the most unstable rare-earth oxide against ambient atmosphere.

The rare-earth oxide films studied have mainly been deposited by different physical vapor deposition methods. Sputtering from metal targets and subsequent oxidation has been employed for yttrium oxide [9]; evaporation of metal has been employed for lanthanum [10] and yttrium oxides [11,12]; molecular beam epitaxy (MBE) with metal and rf discharge excited O_2 sources for lanthanum and yttrium oxides [13]. Electron beam evaporation from oxide source has been, however, the most frequently used method to deposit Gd_2O_3 and Y_2O_3 films for gate dielectric studies [1,5,14–17].

Careful oxidation is needed in the cases where the rare-earth metal is deposited (sputtered or evaporated) on silicon. The dielectric constant of films on silicon was lower than expected and this was explained by the formation of silicate. In experiments where Y metal was grown on SiO_2, a thin silicate layer was also formed but SiO_2 layer served as a diffusion barrier and a pure Y_2O_3 film was obtained [9]. If a yttrium film on silicon is heated in vacuum at 600°C, a silicide is formed and oxidation of this silicide results in yttrium silicate [18]. Silicates are thermally stable and may be interesting as gate dielectric materials. However, they have lower dielectric constant than the corresponding binary oxides. 26 nm thick Y_2O_3 films made by oxidation of metal have shown good electrical properties: low leakage current ($<10^{-10}$ Å/cm^2 at a 5 V bias), high breakdown field ($\sim$4 MV/cm), very little hysteresis in capacitance–voltage (C–V) curves, sum of fixed charge and interface trap charge of 6×10^{11} cm^{-2} and interface trap density near the middle of the silicon band gap of $\sim10^{11}$ cm^{-2} eV^{-1} [9]. The results from La_2O_3 oxidized from metal are very encouraging. Thirty-three-angstrom thick films had a d_{eq} of only 4.8 Å, leakage current of 10^{-1} Å/cm^2 at 1 V bias, interface trap density of 3×10^{10} cm^{-2} eV^{-1}, and flat band voltage shift of +0.7 V [10]. The La_2O_3 and Y_2O_3 films made by Guha et al. [13] showed that interfacial SiO_2 is formed much more extensively in the case of yttrium (15 Å) than lanthanum (<5 Å). The La_2O_3 films with physical thickness of 30–50 Å showed a d_{eq} of 10–14 Å while the leakage current values varied between 10^{-4} and 10^{-7} A/cm^2. Flat band voltage shift was rather large, −1.4 V. Y_2O_3 films made by Guha et al. [13] with MBE showed different electrical properties than La_2O_3 films: lower leakage current density (10^{-8} A/cm^2) and no flat band shift. The results of Y_2O_3 are certainly influenced by the thicker interfacial SiO_2 layer.

Cho et al. [12,19] have reported that a very thin SiO_2 layer helps to get epitaxial Y_2O_3 films. They e-beam evaporated yttrium metal on SiO_2 surface and noticed that oxide surface provided favorable interfacial reaction sites for Y_2O_3, and still maintained the structural registry with Si. Yttrium oxide was a product from the reaction between Y and SiO_2 and direct interaction between Y and Si was hindered by the nucleation of Y_2O_3 (Fig. 1).

In e-beam evaporation of rare-earth oxide films from oxide powder (rod) sources, the substrate temperature is an important process parameter. Kwo et al. [1,5] have obtained amorphous Gd_2O_3 and Y_2O_3 films at substrate

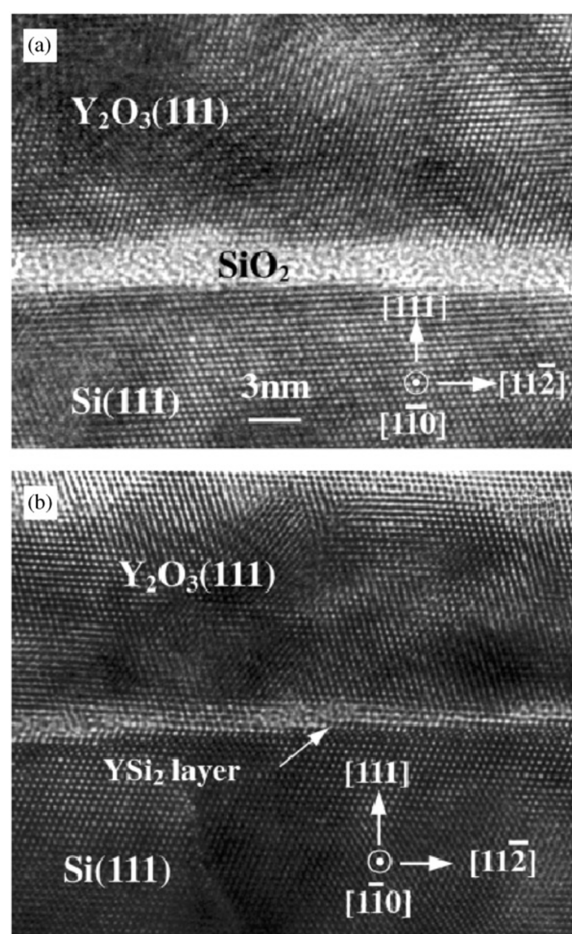

Fig. 1. HRTEM images of Y_2O_3(111) films grown on (a) oxidized and (b) clean Si by evaporating 6 Å Y and subsequently treated with O [12]. (Reproduced from Ref. [12], with permission.)

temperatures of 10–25°C and polycrystalline or even epitaxial films at 550°C on (100) Si. Dimoulas et al. [16], on the other hand, reported that optimum temperature for epitaxial Y_2O_3 films on (001)Si is 450°C. At 610°C, they obtained interfacial YSi_2 which damaged epitaxy. Kwo et al. [1,5] reported that there is no formation of interfacial SiO_2 layer during deposition while Landheer et al. [15] and Dimoulas et al. [16] had in their samples deposited above 400°C a thin SiO_2 layer which got thicker if the films were annealed in oxygen. The amorphous films showed lower leakage current densities (10^{-6} A/cm^2) than the crystalline films (10^{-3} A/cm^2) and the equivalent oxide thicknesses were 10 and 15 Å, respectively. The interfacial state density at the midgap has been reported to be $\sim 10^{11}$ [5] and 1.5×10^{12} cm^{-2} eV^{-1} [15]. The $C-V$ curves of e-beam evaporated films have been reported to be frequency dependent.

Ono and Katsumata [20] studied the reaction between silicon and 20–30 nm thick rare-earth oxide (La–Lu, Y) films made by spray pyrolysis at 400°C. The formation of SiO_2 interface layer is obvious but silicates were formed at higher annealing temperatures. Formation of the Si–O–RE bond strongly depended on the ionic radius of the rare earth being strongest in the case of lanthanum. Rare-earth silicates may be useful gate dielectrics because they have moderate dielectric constants (10–14) and they show superior leakage properties (4–5 orders of magnitude reduction) as compared to SiO_2 with the same equivalent thickness [21]. Yttrium-based silicate can be scaled down to the equivalent thickness regime of 10 Å [18]. Rare-earth silicates have been prepared by three methods: through oxidation of silicides obtained by deposition of metal [18,22], e-beam evaporation of mixtures of SiO_2 and rare-earth oxide [23,24], and deposition of rare-earth oxide on SiO_2 and subsequent heating [21]. In e-beam evaporation of gadolinium oxide, a $(SiO_2)_x(Gd_2O_3)_{1-x}$ layer can unintentionally be formed [14]. The silicates formed are non-stoichometric amorphous mixtures and crystallinity can observed only after annealing at 1050°C. The high crystallization temperature is beneficial in an attempt to avoid increased leakage currents along grain boundaries. Silicate films are stable against Si diffusion but oxygen may diffuse through them [24].

3. Passivation of III–V compounds

Extensive efforts have been devoted to realize viable GaAs MOSFETs. The fabrication of an insulating film with insignificant bulk trap charges and a low density of interfacial states on GaAs has been difficult [25]. The big invention was the discovery of Gd_2O_3 gate oxide [3]. Gallium oxide would have been the natural choice but it is too leaky. The addition of gadolinium to gallium oxide improves the situation dramatically. The effect of gadolinium in the $(Ga_2O_3)_{1-x}(Gd_2O_3)_x$ system has thoroughly been studied and it seems that x must be $>14\%$ to passivate GaAs surface and to achieve electrically insulating films with low leakage current, high breakdown strength and low density of interfacial states (below 10^{11} cm^{-2} eV^{-1}) (Fig. 2) [26–28].

$(Ga_2O_3)_{1-x}(Gd_2O_3)_x$ films have been deposited by e-beam evaporation from single-crystal $Ga_5Gd_3O_{12}$ and the amount of gadolinium depends on the substrate temperature [29]. A mixture of gallium and gadolinium oxides can also be used as evaporation source [26]. Pure Gd_2O_3 film grows epitaxially in (110) orientation on arsenic stabilized (100) GaAs surface at temperatures below 600°C [3]. A careful study of the interface GaAs/$(Ga_2O_3)_{1-x}(Gd_2O_3)_x$ showed that, also in this case, Gd_2O_3 locates closest to GaAs and it is epitaxially ordered [28]. This explains the good electrical results obtained. Recently it was found that a new tetragonal phase was epitaxially grown on GaAs(100) besides the normal cubic α-Gd_2O_3 [30]. This new phase was observed in epitaxially grown thin (18 Å) Gd_2O_3 films or in disordered and recrystallized cubic α-Gd_2O_3 films.

Ren et al. [31] have also demonstrated the first n-channel enhancement-mode $In_{0.53}Ga_{0.47}As$ MOSFETSs on an InP substrate using $(Ga_2O_3)_{1-x}(Gd_2O_3)_x$ gate dielectrics. The device exhibited transconductance, which was one order of magnitude better than previously reported values. $(Ga_2O_3)_{1-x}(Gd_2O_3)_x$ films have also proven to be the best gate dielectric material among the Gd_2O_3, Ga_2O_3 and Y_2O_3 films studied for strained $Si_{0.74}Ge_{0.26}$ MOS devices resulting in low fixed positive charge and interface state densities [32].

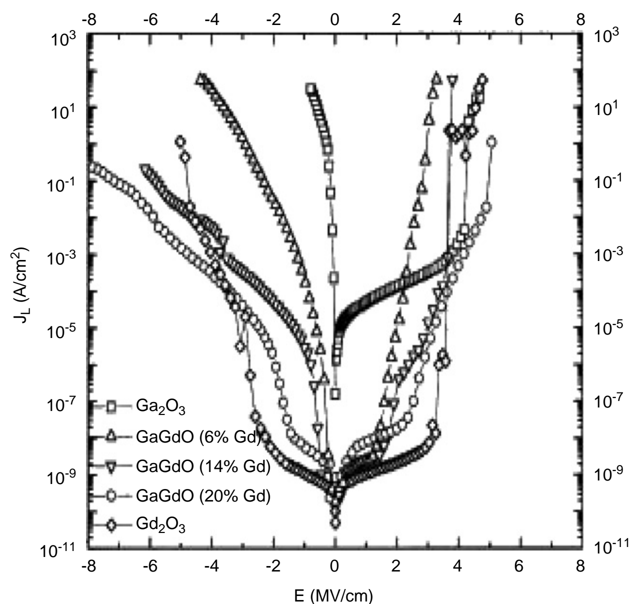

Fig. 2. Leakage current density vs. electric field for $(Ga_2O_3)_{1-x}(Gd_2O_3)_x$ films with varying x [26]. (Reproduced from Ref. [26], with permission.)

Single-crystal Gd_2O_3 films have also been grown on GaN by gas source MBE [33,34]. Gadolinium metal source was used and the growth proceeded in 10^{-4} Torr oxygen pressure. Substrate temperature was 650°C. The large difference in lattice spacing between Gd_2O_3 and GaN causes dislocations in the oxide film. These dislocations act as current leakage paths and the breakdown voltage remains low. The breakdown behavior can be improved by depositing amorphous SiO_2 film on Gd_2O_3.

La_2O_3 films have been demonstrated in metal/ferroelectric/insulator/semiconductor (MFIS) field effect transistor. The films were grown by MOCVD from thd-complex and O_2 at 350°C [35]. The as-deposited films were amorphous and had a dielectric constant of 28. The films crystallized upon annealing and the k value lowered to 18. The leakage current density and other electrical properties showed the potential of La_2O_3 films in MFIS structures.

4. ALD of rare-earth oxides

Compared to PVD methods, CVD has rarely been used in deposition of rare-earth oxide films. Only one recent paper on liquid injection MOCVD of La_2O_3 films and an older paper on $LaAlO_3$ films can be found [36,37]. Studies of ALD of rare-earth oxides are also at the early stage but a few papers have already appeared. ALD is a chemical vapor deposition method, which relies on alternate pulsing of the precursor gases and vapors onto the substrate surface and subsequent chemisorption or surface reaction of the precursors [38]. The reactor is purged with an inert gas between the precursor pulses. With a proper adjustment of the experimental conditions, the process proceeds via saturative steps. Under such conditions, the growth is stable and the thickness increase is constant in each deposition cycle. The self-limiting growth mechanism facilitates the growth of conformal thin films with accurately controlled thickness on a large area [39]. The growth of multilayer structures, called as nanolaminates, is also straightforward [40]. These advantages make the ALD method attractive for microelectronics [41]. In deposition of gate oxides, the accurate thickness control and possibility to grow nanolaminates are the most important ones [42]. In ALD, the layer-type growth and flexibility in chemistry allows the tailoring of the Si/gate oxide interface. It is, for example, possible to make the interface from one oxide and continue then by some other. With a smart chemistry, the oxidation of silicon can be minimized keeping the oxide formation reaction as weakly oxidizing as possible [42].

Niinistö et al. [43–47] have studied ALD of rare-earth oxides since the early 90s and La, Ce, Y and Sc oxides have been grown. Thd (2,2,6,6-tetramethyl-3,5-heptanedionate) complexes are the most common volatile rare-earth precursors. However, the thd-complexes do not react with water, the most common oxygen precursor in ALD oxides. Ozone is needed to decompose the thd-complexes. Growth rate in these $RE(thd)_3/O_3$ processes has been modest, around 0.25 Å/cycle and polycrystalline films are formed at 250–400°C [46]. Lanthanum shows a different behaviour and forms oxycarbonate below 275°C [44]. The instability of La_2O_3 against ambient atmosphere became also obvious in these experiments. Cyclopentadienyl (Cp) compounds form another group of volatile rare-earth compounds. The Cp compounds are more reactive than the thd-complexes and they react with water forming oxide. Cp-precursor has so far been used only with scandium. The Sc_2O_3/H_2O process has a growth rate of 0.75 Å/cycle at 200–500°C and the resulting films contain only minor amounts of C and H residues [47].

Y_2O_3 made with $Y(thd)_3/O_3$ process is so far the only ALD rare-earth oxide studied in gate structure. Due to the strongly oxidizing conditions in this ozone-based process, silicon became oxidized so that 11–14 Å interfacial layer was formed. In addition, during annealing, the yttrium oxide film reacted with the SiO_2 layer forming a silicate [48]. It is obvious that the ozone-based processes can hardly be used in achieving equivalent oxide thicknesses of 10 Å or below, and therefore the use of rare-earth Cp compounds has to be studied in more detail. There are good reasons to expect that a better silicon interface can be obtained by this process than the ozone process. On the other hand, as the thermal stability of the common Cp compounds is only modest, there is an obvious need for Cp compounds with improved stability.

Another group of volatile rare-earth metal compounds potentially useful in ALD are silylamide compounds. Gordon et al. [49] noted that $La[N(Si(CH_3)_3)_2]_3$ reacts with water forming La_2O_3 films and with $(^tBuO)_3SiOH$ forming amorphous lanthanum silicate. No details were given, however. Similarly to the cyclopentadienyls, also with the silylamides, thermal stability may call for an attention.

5. Conclusions

Rare-earth oxides are attractive candidates for alternative gate dielectrics because of the thermodynamics and their relative high dielectric constants. Due to their electropositive nature, the rare-earth oxides are reactive with water, lanthanum oxide being the utmost example. The use of a capping layer may remove this problem. Rare-earth oxides have not been studied as gate dielectrics as widely as, for example, Zr and Hf oxides. Thus, a lot more research is needed to make the final evaluation from their possibilities as gate oxides.

Rare-earth oxides can be deposited by ALD and the resulting films are polycrystalline and relatively pure. Very limited electrical characterization has so far been made on ALD rare-earth oxides. It is to be seen if ALD is the most suitable method to deposit rare-earth oxides.

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