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Induced reduction by H2 exposure at room temperature of ceria ultrathin films grown by atomic layer deposition

Content

Moving towards green energy vectors such as hydrogen needs direct energy production and storage systems, but also the development of secondary components like highly sensitive hydrogen gas sensors integrated into mass devices that operate at ambient conditions. A compound with great potential is ceria (CeOx), in which the easy exchange between the Ce3+ and Ce4+ oxidation states allows the design of resistive sensors.

In this context, atomic layer deposition (ALD) presents a high prospective for integration as a scalable process in microelectronics, allowing well-controlled layer-by-layer deposition and conformal growth on 3D structures. Yet, the ALD technique is also well known to lead to amorphous and defective, non-stoichiometric films, potentially resulting in modified materials properties that can also be affected by film/substrate interaction in the case of ultra-thin deposits. Interestingly, initial in situ XPS measurements of ceria ALD-deposits on Al2O3/Si, sapphire and SiO2 substrates confirm a Ce3+/Ce4+ mixture dependent on the substrate interaction and deposit thickness and morphology. Using near-ambient pressure X-ray photoemission spectroscopy, we have significantly reduced ultrathin (< 10 nm) ceria films grown by ALD by exposing them to different O2/H2 partial pressures at moderated temperatures (RT - 250 °C). Notably, the total reduction to Ce3+ depends on the deposit thickness and initial ceria/substrate interaction, being this process reversible. The initial film defects related to the atomic layer deposition method seem to play a critical role in the reversible reduction at low temperatures.

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