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Unraveling the effects of substrate interaction on the chemical properties of atomic layer deposited ultra-thin ceria layers

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Atomic layer deposition (ALD) is well known to lead to amorphous and defective, non-stoichiometric films, potentially resulting in modified material properties that can also be affected by film/substrate interaction in the case of ultra-thin growths. For example, the formation, diffusion, and recovery of oxygen vacancies can be favored in disordered, reducible metal oxides compared to more ordered deposits, whereas interdiffusion processes can critically affect the film/substrate interface region. These effects have extensively been studied for thin thermal-ALD ceria films (below 15 nm) by combining in-situ and ex-situ characterization techniques in our lab and at synchrotron radiation facilities. While using alumina or silica substrates modifies the initial growth rate, Ce^{3+}/Ce^{4+} ratio, and ceria morphology, the formation of different species at the interface affects its reactivity. Interestingly, the experiments have shown high reducibility of ALD-ceria ultrathin films on silica for very low hydrogen concentrations, even at room temperature, whereas for alumina substrates the formation of aluminates at the interface prevents further oxidation. Moreover, the comparison with more ordered films indicates a key role of the defective structure of ALD films in Ce^{3+}/Ce^{4+} conversion.

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