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Titanium dioxide (TiO₂) represents a perfect example of a multifunctional metal-oxide semiconductor with applications ranging from microelectronics to photo catalysis or medical device materials [1]. Atomic layer deposition (ALD) is widely regarded as one of the most promising techniques for the growth of thin TiO₂ films due to its simplicity, reproducibility, the high conformity of the obtained films and an excellent control of the layer thickness at the angstrom level [2].

One of the factors that often dictates the properties of ALD films, and therefore their possible applications, is the crystallinity of the final film. While amorphous films of TiO₂ are preferable if diffusion barriers are required for a particular application, crystalline films with a specific phase are often desired for their specific chemical or electrical properties. In addition, the formation of crystallites in TiO₂ films is of great technological interest. For example, TiO₂ has been regarded as one of the most promising photo-catalytic materials for environment-protective coatings due to its high photo-catalytic activity, high chemical stability and low toxicity [3].

In the present work, the photocatalytic activities of TiO₂ thin films, grown by ALD, were investigated as a function of the grain size and crystal structure. The samples were characterized by scanning electron microscopy, grazing incidence X-ray diffraction, secondary ion mass spectrometry, X-ray photoelectron spectroscopy, atomic force microscopy, and near-edge X-ray absorption fine structure spectroscopy. We show that the crystallinity and the size of crystallites can be controlled over a large range of diameters, from around 70 nm up to 1 μ m with five parameters: the type of substrate, the type of Ti precursor, the deposition temperature, the number of ALD cycles (i.e. the film thickness) and the nanometric Al₂O₃ buffer layers deposited on substrates in the same ALD sequences prior to TiO₂ films. The most dramatic increase in size of the plate-like anatase grains, to more than 1 μ m in diameter, was obtained on films grown at 250 °C on Si substrate covered with a 10 nm Al₂O₃ layer.

The photocatalytic activity, determined for each TiO₂ film from the degradation of methylene blue under UV irradiation, is more efficient for the anatase phase of TiO₂ than for the rutile phase, and increases with the grain size of crystallites. The high photocatalytic activity, combined with the low processing temperatures used in the present study, open a wide range of applications for different substrates coated with ALD TiO₂ films, such as polymers or cellulose-based substrates, ranging from packaging materials for food to water or air purification systems.

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