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TITLE: The Effect of Atomic Layer Deposited TiO_2 Oxide Thickness on the Water Oxidation Performance of Metal-Insulator-Silicon Anodes

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ABSTRACT BODY: Overcoming the intermittency of solar radiation using energy storage methods is a major challenge for adoption of solar energy at very large scale. Synthesis of fuels from sunlight is one potential storage approach, providing the need for optimized photoelectrochemical devices and materials. We have recently reported on atomic layer deposition (ALD) of ultrathin TiO_2 tunnel oxides that can stabilize highly efficient silicon anodes for photocatalytic water splitting [1]. Pin-hole free films of ~ 2 nm thickness have been found to offer substantial protection of underlying n-Si anodes during water oxidation. This report probes the effects of varying the ALD-TiO_2 thickness on device performance. Uniform films of atomic layer deposited TiO_2 are prepared in the thickness range ~1-12 nm on degenerately-doped p^+-Si yielding water oxidation overpotentials at 1 mA/cm^2 of 300 mV to 600 mV in aqueous solution (pH 0 to 14). Additionally, the electron/hole transport through Schottky tunnel junction structures of varying TiO_2 thickness was studied using the reversible redox couple ferri/ferrocyanide. The results of these analyses reveal mechanistic insights into the conduction through the ALD-protection layer. A thorough understanding of the conduction mechanisms and inherent tradeoffs of using an ALD-oxide protection coating for water splitting devices is crucial for further development of this device technology.

[1] YW Chen, J.D. Prange, et al. Nat. Mater. 2011, 10, 539-44.