Because the surfaces of small structures can dominate their properties, implementing functional nanoscale materials in applications such as energy conversion and electronics depends to a large extent upon developing control of their surfaces. This talk will discuss our research into modifying the surfaces of nanoscale semiconductors by organic and inorganic layers to change their chemical or electronic properties for applications in solar energy conversion and nanoelectronics. The work on solar energy conversion focuses on dye-sensitized solar cells (DSSCs) and the related quantum dot-sensitized solar cells (QDSSCs), both of which are being explored as next-generation photovoltaic technologies. We will describe the use of atomic layer deposition (ALD) both for fabrication of a barrier layer at interfaces within the solar cell to reduce electron recombination and for synthesis of quantum dots to absorb light in QDSSCs. ALD is a layer-by-layer deposition method that provides for precise thickness control and excellent conformality and can be used for a wide variety of materials. We will also describe a related deposition method, molecular layer deposition (MLD), which is used to grow precisely controlled, nanoscale organic thin films. An example from our laboratory will be presented in which MLD is applied to development of ultrathin photoresist materials for extreme ultraviolet lithography for nanoelectronics.