A number of challenges related to the development of new organic-inorganic photovoltaic systems exist, including the ability to enhance the materials interface and improve the control required in development of nanoscale materials. Layer-by-layer (LbL) assembly allows for the incorporation of a wide range of functional materials into structured thin films based on the alternate adsorption of cationic and anionic species. Biomolecules, and in particular viruses, show great potential as components of functional materials due to their capacity for molecular recognition and self-assembly. Here we report that by substituting a negatively charged variant of M13 bacteriophage for the negatively charged polymer during the dip LbL assembly process, phage can be incorporated into a hybrid material with characteristics of both its biological and polymeric components. The resulting mesoporous polymer films can be used as a template for the construction of the titania photoanode of dye sensitized solar cells (DSSCs) with a novel nanowire architecture to enhance electron transport. The biotemplated nanowires are shown to significantly increase device electron diffusion length and increase device efficiency as compared to LbL-templated titania photoanodes made without bacteriophage. Spray LbL is also investigated as an assembly method for the construction porous templates for titania photoanodes. The necessary porous transition is shown to occur on flat substrates, like those normally utilized for DSSCs, and on porous metal meshes, substrates that have been proposed as lower-cost DSSC current collectors. Spray LbL is demonstrated to coat metal to different degrees of conformality as a function of mesh pore size. The conformality of the coating, in turn, determines which functions it could assume within a LbL-based DSSC.