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Spatial Distribution of Light Scattering and Absorption Interactions with TiO_2-Nanoparticles from Monte Carlo and Generalized-Multiparticle-Mie based Simulations for Dye-Sensitized Solar Cell Analysis and Optimization

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TiO_2-based dye-sensitized solar cells (DSSCs) are a promising light harvesting technology that attracts tremendous research interest. The bulk of research however by far focuses on the light absorber and other components of the cell configuration through mostly experimental investigations [1,2]. The structure of DSSCs' photosensitive component, the photoelectrode, is based on a randomly organized network of TiO_2-nanoparticles (NPs). This highly complex structure has made the theoretical studies difficult. Currently, optical models proposed in the literature have been limited to an oversimplified study of isolated spherical nanoparticles [3]. The underlying theory relating the photoelectrode structure with its light harvesting capacity remains unclear. In this research, a computer simulation program is presented for modeling the optical response of DSSCs' photoelectrode, and developing fundamental understanding of light interactions with TiO_2-NPs. The light harvesting capacity of the photoelectrode is examined through simulated transmittance, reflectance, and absorptance spectra for different configurations. Unlike currently used models for similar applications, this C++ program is based on generalized-multiparticle-Mie (GMM) rigorous electromagnetic scattering solutions for aggregated spherical nanoparticles (SNPs) [4]. A Monte Carlo algorithm was coupled to GMM calculations to simulate light propagation within the photoelectrode. Using scanning electron microscope (SEM) micrograph to have realistic TiO_2-NP configurations, the simulation results were compared to experimental spectra.

The simulating program allowed study of a variety of DSSCs' photoelectrode configurations with different thicknesses, porosity, NP sizes, and aggregate geometries (e.g. cubes and cylinders) composed of randomly positioned SNPs (as shown in Figure 1). From the simulated results, three levels of analysis were possible: the distribution of cross-sections from individual particles in an aggregate (Figure 1), the variation of the total aggregate cross-sections (Figure 2, left), and the normalized intensity spectra (Figure 2, right) from the UV and near IR range of wavelengths. The geometrical properties of TiO_2 samples, such as NP morphology and size (Figure 3, left), and the sample porosity and thickness, were extracted from SEM micrographs then used to define the nanostructure configuration in the simulation. Simulated transmittance, reflectance and absorptance results were in agreement with experimental observation as seen in Figure 2 (right) and Figure 3 (right).

The individual-particle cross sections, as visualized in Figure 1, provided for the first time insight into the spatial distribution of interaction probabilities within an aggregate of TiO_2-NPs. This is of great importance for the design and optimization of the photoelectrode structure.

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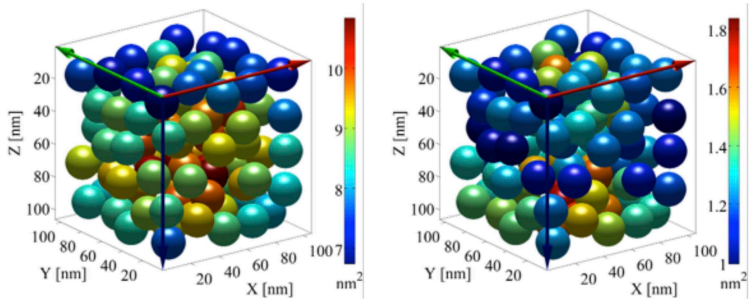

Figure 1. Simulation of individual-particle cross-sections (left, scattering; right, absorption) in an aggregate for an incident light of 503 nm wavelength. The aggregate consisted of 100 TiO_2-nanoparticles (NPs) randomly positioned in the volume. The probabilities (as given by the corresponding cross-sections) of scattering interactions and absorption, i.e., electron generation, increased with a deeper and centered position of the NP in the aggregate.

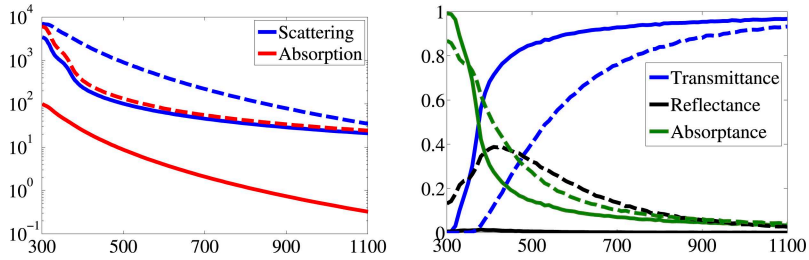

Figure 2. Variation of the total cross-sections (left) and the normalized intensity spectra (right) with the light scattering model used: Mie, for isolated nanoparticles (NPs), (full lines) or GMM, for aggregated NPs, (dashed lines). A volume of 1 cm x 1 cm x 2 μm with TiO_2-NP aggregates was specified for the Monte Carlo simulations. Each aggregate consisted of 100 nanoparticles (20 nm-size each).

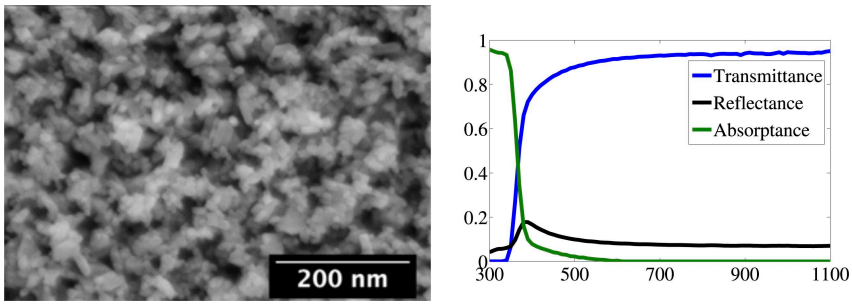

Figure 3. Characterization of a DSSC TiO_2-photoelectrode sample. (left) The SEM micrograph revealed an agglomeration of spherical and ellipsoidal nanoparticles (NPs) with an estimated size of 20 nm. This value was used to set the nanoparticles size in the Monte Carlo simulations (right).

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