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Abstract (Doctor)

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 Nanoscale Interface Control for Advanced Electrochemical Devices

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Introduction: Nanoscience and nanotechnology are currently one of the most researched areas due to its wide-range areas of application which include the and engineering fields. Nanoscale materials have \mathbf{the} unique sciences characteristics of exhibiting different properties from their bulk materials. Thus, they induce novel functionalities in applied materials; have the prospects of creating new knowledge; enhance the functionalities of devices; and the feasibility of miniaturization of devices. In this thesis we have applied nanoscale fabrications under two research themes of: 1) using nanofilm coatings to obtain a robust superhydrophobic surface and an extremely low actuation electrowetting voltage (3 V); and 2) using various morphologies of Ag@TiO₂ core-shell nanostructures to study their plasmonic effects on the performance of dye-sensitized solar cells. Thus, this thesis is structured into four main topics from studies under these two themes:

1. Preparation of thermally and chemically robust superhydrophobic coating from and low voltage reversible electrowetting liquid phase deposition Superhydrophobic surfaces and electrowetting have many applications in various fields. Two of the most desired features of surfaces in many of these fields are robustness and low actuation voltage electrowetting. In this work a thermally and chemically robust nanoscale superhydrophobic surface was prepared using facile techniques of sol-gel and hot-water treatment to obtain rough pseudoboehmite structures; and then a subsequent layer-by-layer hydrophobizing coating using Nafion[®]. This coated surface exhibited a high superhydrophobicity with a contact angle value of 167.2 ± 0.8°; thermal stability up to 300 °C; stability to acid, base, organic solvents and UV irradiation. Subsequently replacing Nafion® with 1H, 1H, 2H, 2H-perfluorodecyltrimethoxysilane and an initial conductive Au layer on the soda-lime glass substrate, an extremely low actuation electrowetting voltage of 3 V and reversible electrowetting were achieved in air and dodecane media, respectively.

The second theme of this thesis was the systematic study of the effect of various $Ag@TiO_2$ core-shell nanostructures on the performance of plasmonic dye-sensitized solar cells – The demand and cost of energy are rising due to population growth. The current major source in the form of fossil fuel is limited and fast exhausting, non-renewable, and not environmentally friendly; especially

with regards to the release of CO₂ which causes global warming. Hence the search for alternative energy sources is imperative. These sources should be reliable, cost effective. sustainable and environmentally friendly. Among the current alternative sources solar energy seems to be the most viable alternative. Further, a 3rd generation solar cell, the dye-sensitized solar cell (DSSC) has emerged as a credible alternative to the conventional solid state p-n junction solar cells. It has the core advantages of facile and low-cost fabrication process, environmentally friendly and good performance under variable light conditions. However it has a competitively lower power conversion efficiency (PCE) of about 12 %, compared to about 25 % of the Si-based solar cells. One approach in the effort to increase the PCE is the exploitation of the localized surface plasmon resonance (LSPR) phenomenon effect of metal nanostructures to increase the light absorption of DSSCs, ultimately increasing their PCEs. However, these plasmonic nanostructures have secondary effects on the performance the cells, which are not yet well understood. Thus, the aim of this second part of the thesis was an effort to increase the understanding of the role of these plasmonic nanostructures on the performance of the DSSC using 3 different morphologies of Ag@TiO2 core-shell nanostructures:

2. Systematic characterization of uniformly sized and shaped Ag@TiO₂ nanoparticles – the results from this study showed that apart from enhancing light absorbance to increase short circuit current and ultimately increased PCE, the negative and positive effects of the plasmonic nanostructures on the various DSSC performance parameters can be tuned to optimally balance to produce at least one more optimal plasmonic NP loading concentration to achieve highly efficient DSSCs. I was also observed that one of the major negative effects of plasmonic nanostructures is the exponential increase in the recombination of photogenerated charges as a result of the abundant generation of these charges.

3. Effects of multi-sized and -shaped Ag@TiO₂ nanoparticles – similar effect trends were observed under this study as in the study with uniform NPs above. However, the complex composite of varying sizes and shapes caused destructive interference of their plasmonic effect, which possibly prevented the progressive enhancement effect of cell performance in the NIR region with increasing amount of plasmonic NP loadings as observed in the previous study.

4. Effect of $Ag@TiO_2$ nanowires – the results of this study also showed similar effect trends as observed in the two previous studies above. However, in this study the unique performance enhancing effect was the efficient transport of injected photogenerated electrons, attributed to the 1-D morphology of the nanowires with no, or very limited number of, grain boundaries that impede electron transport. Thus, these results will serve as very good guides to other researchers in these fields and also of high potential for possible industrial applications.