



Plasmonic enhancement of light-harvesting efficiency in tandem dye-sensitized solar cells using multiplexed gold core/silica shell nanorods

Yan-Zhen Zheng^{a,b}, Xia Tao^{a,*}, Jin-Wen Zhang^a, Xue-Sen Lai^a, Nan Li^a

^a State Key Laboratory of Organic-Inorganic Composites, Beijing University of Chemical Technology, 15 Beisanhuan East Road, Beijing, 100029, China

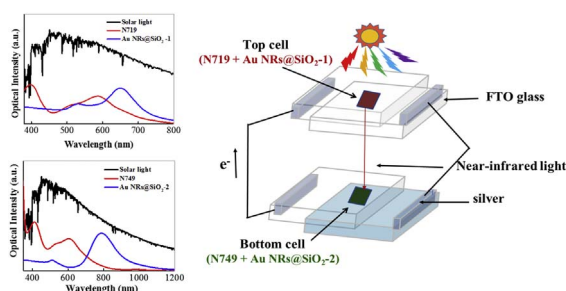
^b Research Center of the Ministry of Education for High Gravity Engineering & Technology, Beijing University of Chemical Technology, Beijing 100029, China



HIGHLIGHTS

- Multiplexed Au nanorods are used to tune the photoresponse of tandem solar cells.
- Plasmonic Au nanorods increase the sunlight harvesting from visible to NIR.
- Conversion efficiency achieves 10.73% for the plasmonic tandem DSSCs.

GRAPHICAL ABSTRACT



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ABSTRACT

Incorporation of plasmonic metal nanocrystals is a promising approach for broadening and enhancing the light harvesting of dye-sensitized solar cells (DSSCs). In this work, we report a facile and versatile route to tune the photoresponse of tandem DSSCs via incorporating Au nanorods with multiplexed length-to-diameter aspect ratios in the two sub-cells. Plasmonic Au nanorods with length-to-diameter aspect ratio of 2.5 (Au NRs-1) and 3.9 (Au NRs-2) are prepared, exhibiting their plasmon band at 500–700 nm and 500–900 nm, respectively. Au NRs-1 core/SiO₂ shell (Au NRs@SiO₂-1) and Au NRs-2 core/SiO₂ shell (Au NRs@SiO₂-2) are separately incorporated in TiO₂ photoanodes and then coupled with commercial dye N719 and N749 for the top and bottom sub-cells of a tandem DSSC, achieving a power conversion efficiency (PCE) of 10.73% for relative to 9.02% of reference (TiO₂ only) devices. By virtue of morphological, spectral and electrochemical characterizations and analysis, we find that the integration of Au NRs within dye-sensitized TiO₂ photoanode film enables to increase the sunlight harvesting from visible to near infrared region by plasmonic enhancement effect, reduce the charge recombination probability and facilitate charge transport via Au NRs, leading to enhancement of PCE.

1. Introduction

TiO₂-based dye sensitized solar cells (DSSCs) have been attracting much attention due to their easy manufacturing process, relative high power conversion efficiency (PCE) and low cost [1–6]. DSSCs are typically assembled into a sandwiched configuration, which consists of a dye adsorbed-mesoporous TiO₂ film, an iodide/triiodide electrode as

well as a platinum counter electrode. For full utilization of sunlight, exploitation of new efficient and stable dyes, design of photoanode architecture with high specific surface area, fast electron transport and prominent light-scattering capability are widely performed in the past two decades [7–12]. It is estimated that if a PCE attains over 15% using iodide/triiodide electrode, a DSSC should harness 80% of sunlight from 350 to 900 nm [13]. To date, the most efficient commercial sensitizers

* Corresponding author.

E-mail address: taoxia@yahoo.com (X. Tao).

are ruthenium complex dyes such as N3 and N719 with a strong absorption peak at around 530 nm and a much lower absorption coefficient at high longer wavelength [14]. Panchromatic sensitizers such as porphyrins were synthesized to enhance the light harvesting in the wavelength range of 600–900 nm [5,7,8]. However, the overall PCE of such panchromatic sensitizers is often lower than the ruthenium complex dyes owing to their low absorption coefficient in the extended spectral region and loss of the strong absorption at 500–600 nm [8,13]. Obviously, enhancing the photocurrent generation at longer wavelength and simultaneously retention of the original high absorption by conventional N3 or N719 dyes are highly desired.

Plasmonic metal nanostructures, which have large absorption/scattering cross-sections and are able to strongly focus sunlight on the metal surface, can couple incident photons to conduction band electrons under photoexcitation to generate surface plasmon resonances (SPR) [15–19]. Incorporation of plasmonic metal nanocrystals with photo-semiconductor nanostructures can therefore effectively broaden and enhance the light harvesting of sensitizing molecules through scattering, absorption enhancement, together with hot-electron injection [18,20–23]. Because of these unique optical merits, plasmonic metal nanocrystals (e.g. nanoparticles, nanorods) have been introduced to TiO₂ or C₃N₄ matrix for improving the photocatalytic and photo-electrochemical performance in energy conversion and environmental remediation such as photocatalytic H₂O splitting, CO₂ reduction, N₂ fixation, organic degradation, and DSSCs [23–33]. It is worthy of noting that the absorption enhancement and scattering efficiency are governed by both the metal nanostructure shape and size. In general, nanostructures larger than 100 nm in size predominantly scatter light, whereas smaller nanostructures primarily absorb sunlight. Unlike spherical nanoparticles, nanorod plasmonic nanostructures with its broadly tunable length-to-diameter aspect ratio-dependent longitudinal surface plasmon resonance (LSPR), exhibit a variable plasmon wavelengths ranging from visible, near-infrared and to infrared region, and larger local electric field enhancements [34]. For example, Au nanorods with the sizes of 87 ± 6 nm by 38 ± 3 nm, 94 ± 6 nm by 41 ± 3 nm, 60 ± 7 nm by 16 ± 1 nm, and 68 ± 5 nm by 15 ± 1 nm, exhibit the varied longitudinal plasmon wavelengths of 650 nm, 700 nm, 838 nm, and 890 nm, respectively. Coupling such length-to-diameter aspect-controllable plasmonic Au nanorods with dye in a DSSC, will bring three distinct advantages: i) Au nanorods can both capture solar energy in the long wavelength and scatter light, which can be re-absorbed by the dye molecules, increasing the total light harvested by the dye; ii) Au nanorods can serve as antennas, coupling the plasmonic near field with the optical absorption of the dye and thereby increasing the total absorption cross-section; and iii) Au nanorods can convert incident photons to “hot” electrons, which are rapidly injected into the conduction band of TiO₂, boosting the number of carriers available for photocurrent generation [35,36].

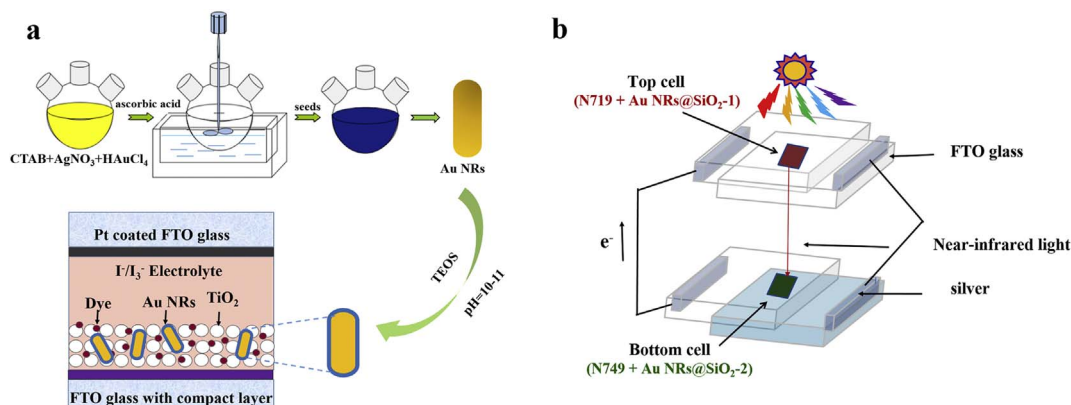
Alternatively, another effective approach to broadening the absorption region in DSSCs is to build a tandem-architecture, in which two or more sub-cells absorbed by complementary absorption spectral dyes are monolithically stacked together and connected in series [37,38]. Indeed, tandem architecture has been demonstrated to be valid to overcome the sun energy loss barrier originating from the narrow absorption of single dye. Moreover, the thermalization loss of the excess energy photons can be minimized by combining absorbers with different bandgaps in a tandem architecture. Most studies focused on the optimization of tandem structures and the match of two absorbers [37,38]. Considering that the longitudinal plasmon wavelength of Au nanorods is length-to-diameter aspect ratio-dependent, we are motivated to introduce various aspect-ratio Au nanorods into the tandem DSSCs for the purpose of tuning the absorption spectra and enhancing the PCE of the devices. However, such a strategy has not yet been reported for tandem DSSCs.

In this work, we report for the first time a facile and versatile route to tune the sunlight harvesting of tandem DSSCs via incorporating Au nanorods with multiplexed length-to-diameter aspect ratios in the two sub-cells. Au nanorods with aspect ratios of 2.5 and 3.9 are prepared and exhibit the longitudinal plasmon wavelengths centering at ~634 nm and ~784 nm, respectively. These two self-prepared Au nanorods are separately coupled with N719 and N749 dyes at top and bottom sub-cells to be capable of harnessing the wide visible and NIR light (400–900 nm) of tandem DSSC in union. The Au NR@SiO₂-incorporated plasmonic tandem DSSC exhibited an overall PCE of 10.73% under AM 1.5 G sunlight illumination, corresponding to 19% increment from that of its counterpart without Au NR@SiO₂. The enhanced optoelectronic performance is studied and discussed in terms of extended spectral absorption, effective charge transport and reduced charge recombination probability owing to the incorporation of Au NR@SiO₂ within DSSCs.

2. Experimental

2.1. Materials

Chloroauric acid (HAuCl₄), chloroplatinic acid (H₂PtCl₆), 4-*tert*-butylpyridine (TBP), silver nitrate (AgNO₃), Iodide (I₂), lithium iodide (LiI), and 1, 2-dimethyl-3-propylimidazolium iodide (DMPII) were purchased from Sigma-Aldrich Co. Ltd. Ascorbic acid (Vc), sodium borohyde (NaBH₄), cetyltrimethylammonium bromide (CTAB) were obtained from Sinopharm Chemical Reagent Company. The Ru dye *cis*-bis(isothiocyanato)bis(2,2'-bipyridyl-4,4''-dis-carboxylato)-ruthenium(II) (N719) and tri(isothiocyanato) (2,2'; 6',2''-terpyridyl-4,4',4''-tri-carboxylic acid) ruthenium(II) (N749) were acquired from Solaronix. All water used in this work was deionized (18.2 MΩ cm⁻¹).



Scheme 1. Schematic illustration of the synthesis process of Au NRs and Au NRs@SiO₂ for a DSSC, and device structure of Tandem DSSC (b).

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