Supplementary Information

Shell-in-Shell TiO_2 Hollow Spheres Synthesized by One-Pot Hydrothermal Method for Dye-Sensitized Solar Cell Application

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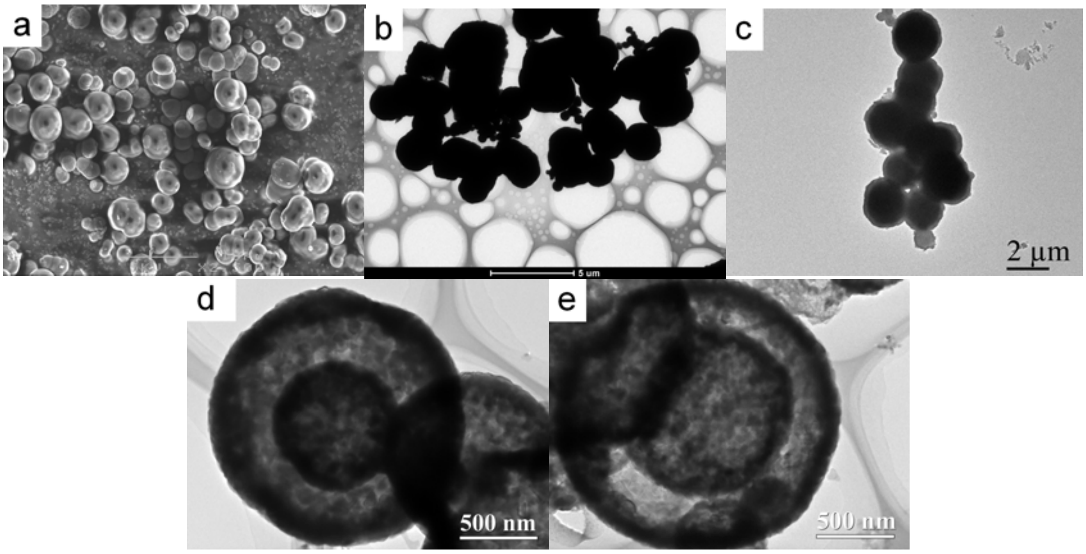
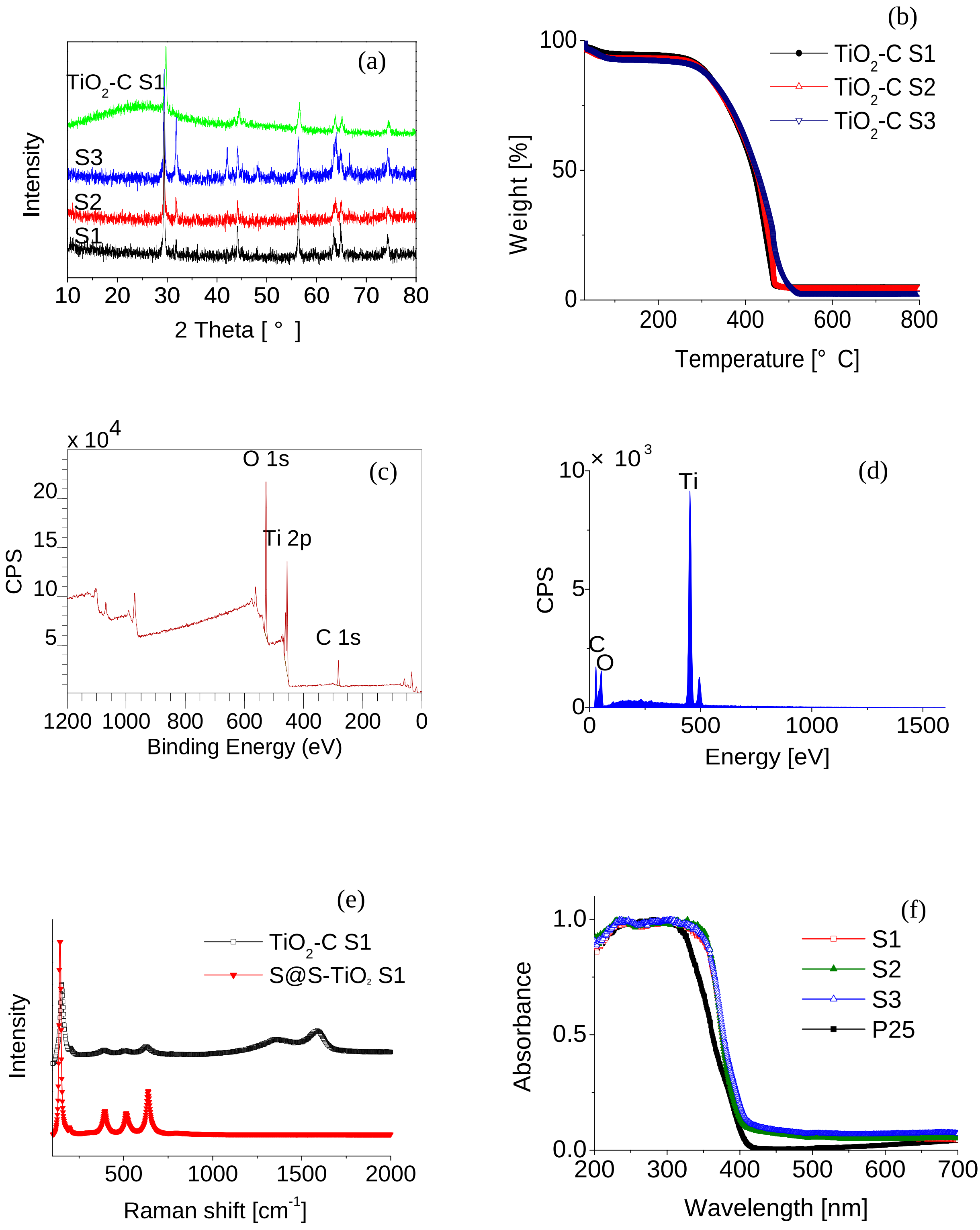
S-Table 1. Physical properties of the S@S-TiO_2 samples.

Samples	Phase content [%]	Crystalline size [nm]	S_{BET} [m^2 g^{-1}]	Dye uptake [$\times10^{-5}$ mol g^{-1}]
S@S-TiO_2 S1	A: 89.2 R: 10.8	A: 41.5 R: 42.6	25.5	3.0
S@S-TiO_2 S2	A: 74.8 R: 25.2	A: 33.9 R: 41.9	27.6	3.1
S@S-TiO_2 S3	A: 66.4 R: 33.6	A: 32.6 R: 33.6	29.6	3.3
P25	A:83.3 R:16.7	A:22.7 R:32.0	50	4.3

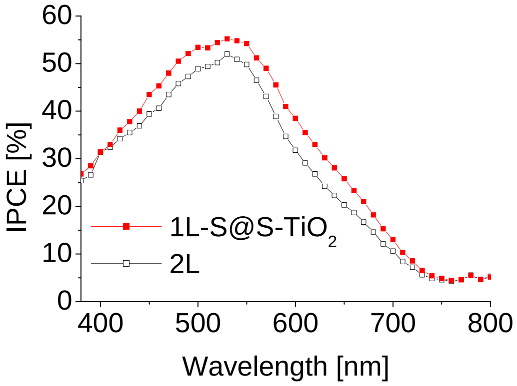
S-Figure 1 SEM image of (a) TiO_2-Carbon composite S1 with Sucrose: TiF_4=5:1; (b, c) TEM images of the samples prepared with Sucrose: TiF_4 molar ratio (b) 1:0.5; (c) 1:2, followed by calcination at 550^{o}C. (d, e) TEM images of the samples prepared with Sucrose: TiF_4 molar ratio (d) 10:1 and (e) 15:1 by decreasing the Ti concentration according to S1 (5:1), followed by calcination at 800^{o}C.



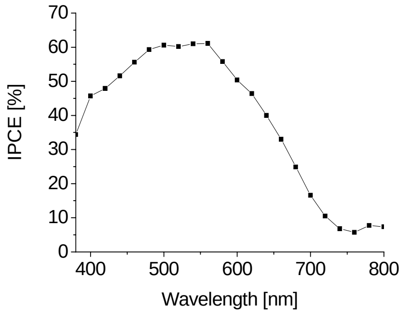
S-Figure 2 (a) XRD patterns of the samples S1, S2, S3 and the as-prepared TiO_2-Carbon composite (TiO_2-C S1); (b) TGA/DTG curves of the TiO_2-Carbon composites S1, S2 and S3; (c) XPS survey scan of S@S-TiO_2 S1; (d) EDS scan of S@S-TiO2 S1; (e) Raman spectra of TiO_2-Carbon composite S1 and S@S-TiO_2 S1. Similar results were observed for S2 and S3. (f) UV-visible spectra of the S@S-TiO_2 samples in comparison with that of P25.



S-Figure 3a: IPCE spectra of DSSCs based on 2L P25 film and the 1L-S@S-TiO2 bilayer structured film.



S-Figure 3b. IPCE curve of the DSSC composed of Dyesol paste.



S-Table 2. Photovoltaic properties of the DSSCs composed of Dyesol paste.

Sample	J_{sc} [mA cm^{-2}]	V_{oc} [mV]	FF [%]	η [%]	Area [cm^2]	Condition
Dyesol paste	19.7	774	56.9	8.71	0.20	AM1.5G one sun

The IPCE was measured in an incident light intensity from 0.009-0.036 mW/cm^2 (measured by the Si detector), therefore, the IPCE value could be underestimated in the red region (> 700nm) since the photocurrents are very small due to the low monochromatic output in this region, i.e., when the incident light intensity is below 0.2mW/cm^2.[53] The electrons are lost due to electron trapping and insufficient conductivity of the nanocrystalline TiO_2 film, and therefore the calculated short-current photocurrent might be underestimated.

We also investigated the photovoltaic performance of DSSC using commercially available Dyesol paste (TiO_2-coated test cell glass plate (Transparent), Dyesol Pty Ltd., Australia) and the results were listed above (S-Table 2 and S-Figure 3b). Again, the underestimation of the IPCE value would possibly due to the weaker incident light intensity of the system.

Note that although our IPCE system indicated the underestimated value due to the lower intensity of the incident light, it can still be a very useful tool to verify and compare the performance of different cells. On the other side, the I-V measurement results should be taken into account as priority.

S-Table 3 Photovoltaic properties of the DSSCs composed of S@S-TiO_2 S1.

Sample	J_{sc} [mA cm^{-2}]	V_{oc} [mV]	*FF* [%]	η [%]	Thickness [μm]	Area [cm^2]	Condition
S@S-TiO_2 S1	13.0	733	59.3	5.65	*ca.* 11	0.16	AM1.5G one sun