Supporting Information

Construction of strontium tantalate homo-semiconductor composite photocatalysts with tunable type II junction structure for overall water splitting

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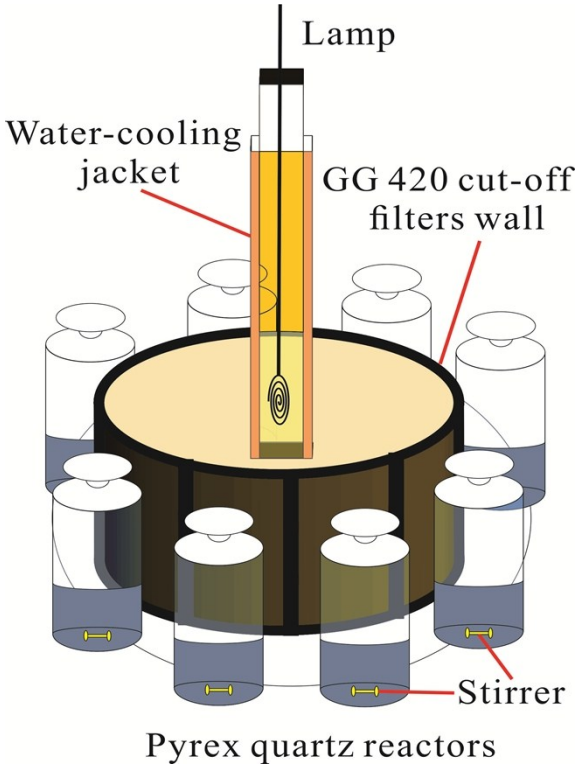
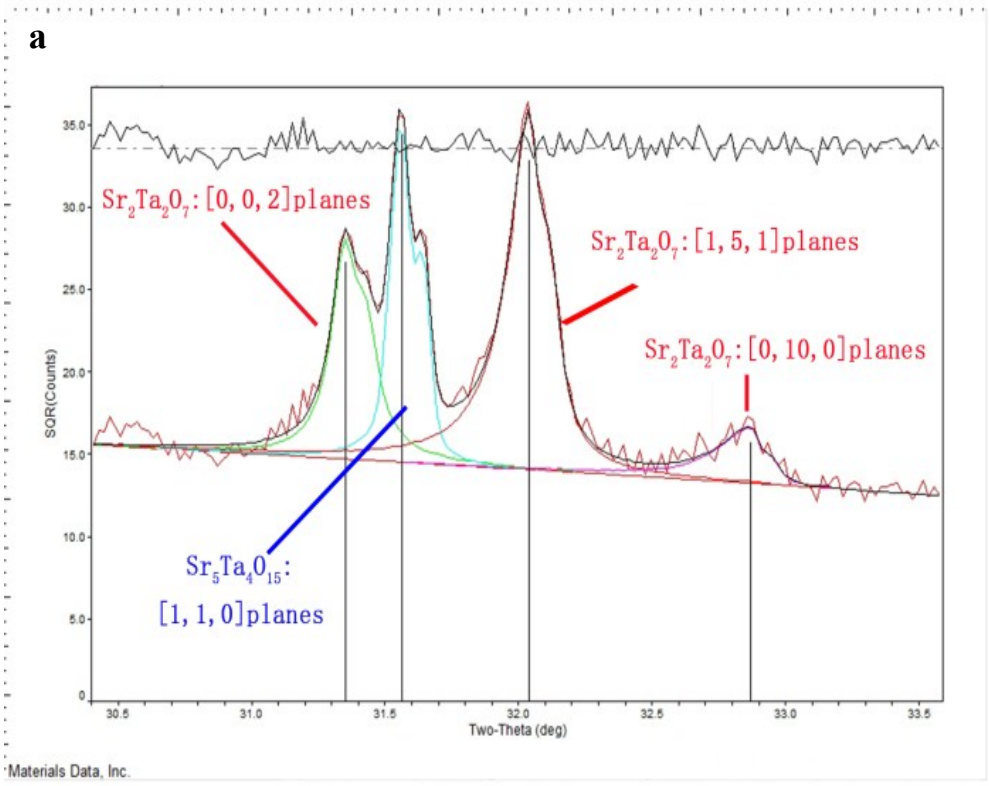
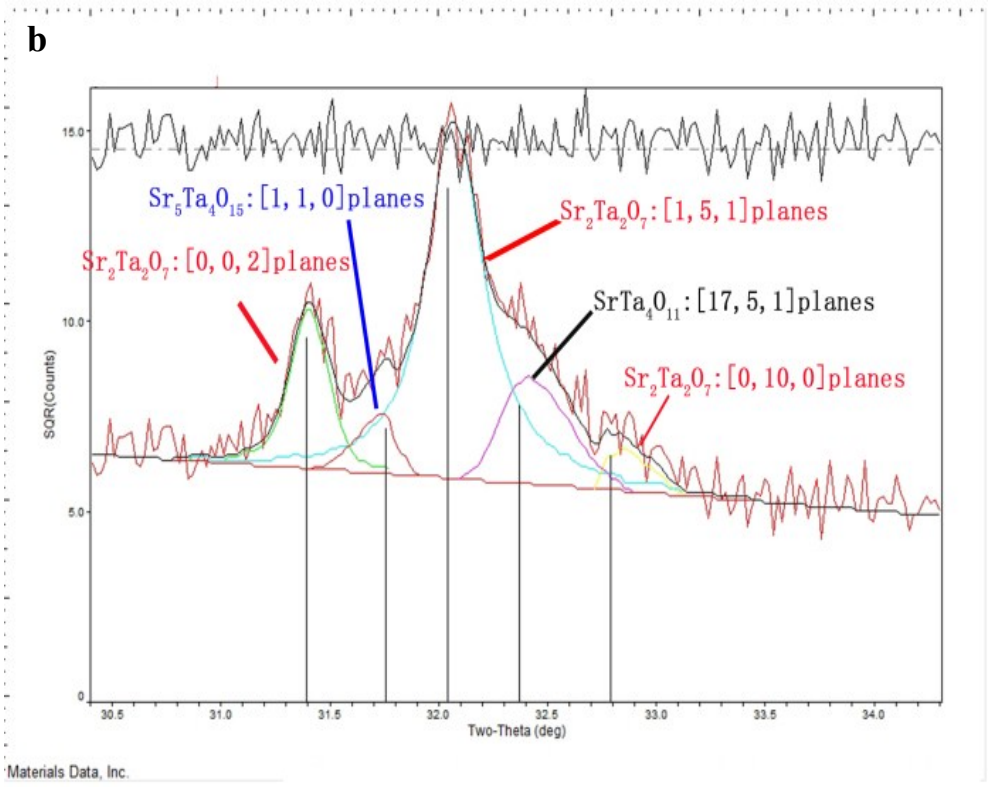
Reaction setup



Scheme S1. An illustrative diagram of home-bulit multizone photocatalytic reaction system consisting of a 500 W mid-pressure Hg lamp and eight air-tight quartz reactors. The performance measurements of samples with the same Sr/Ta ratios prepared in different batches were reproducibly conducted at least three times.

XRD Rietveld refinement





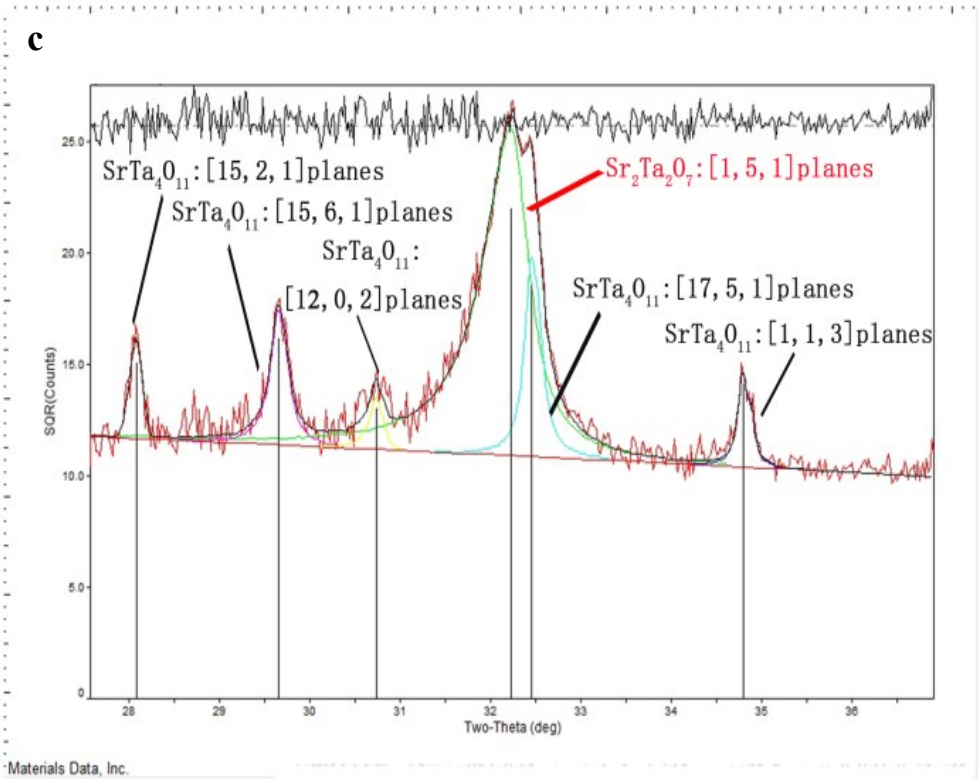


Figure S1. XRD Rietveld refinement of the typical SrTa0.77 (a), SrTa1.2 (b), and SrTa1.8 (c) samples in the low-angle 2θ range.

UV-Vis absorption spectra

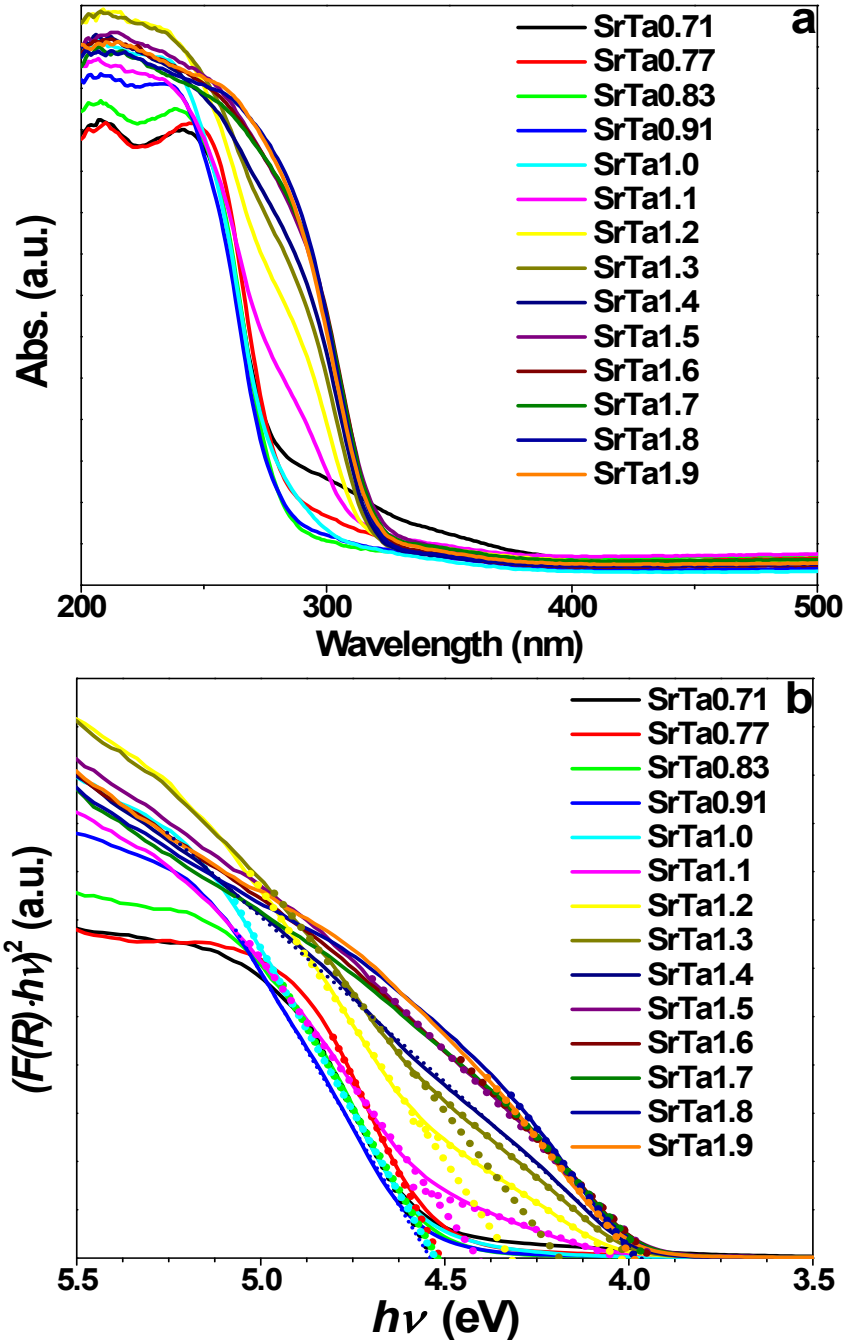


Figure S2. UV-Vis absorption spectra of the as-synthesized strontium tantalate samples with different Ta/Sr ratio (a) and Tauc plots (b).

X-ray photoelectron spectroscopy (XPS)

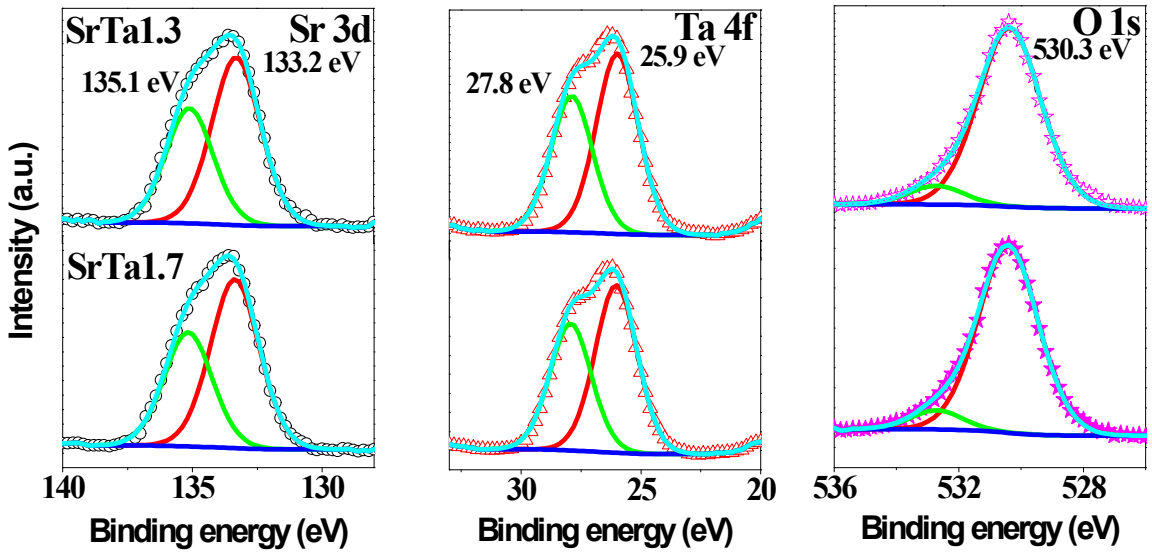


Figure S3. X-ray photoelectron spectra of narrow scans of Sr 3d, Ta 4f and O 1s regions for SrTa1.3 and SrTa1.7 samples.

High-angle annular dark-field scanning TEM (HAADF-STEM) with energy dispersive X-ray (EDX) elemental mapping analyses

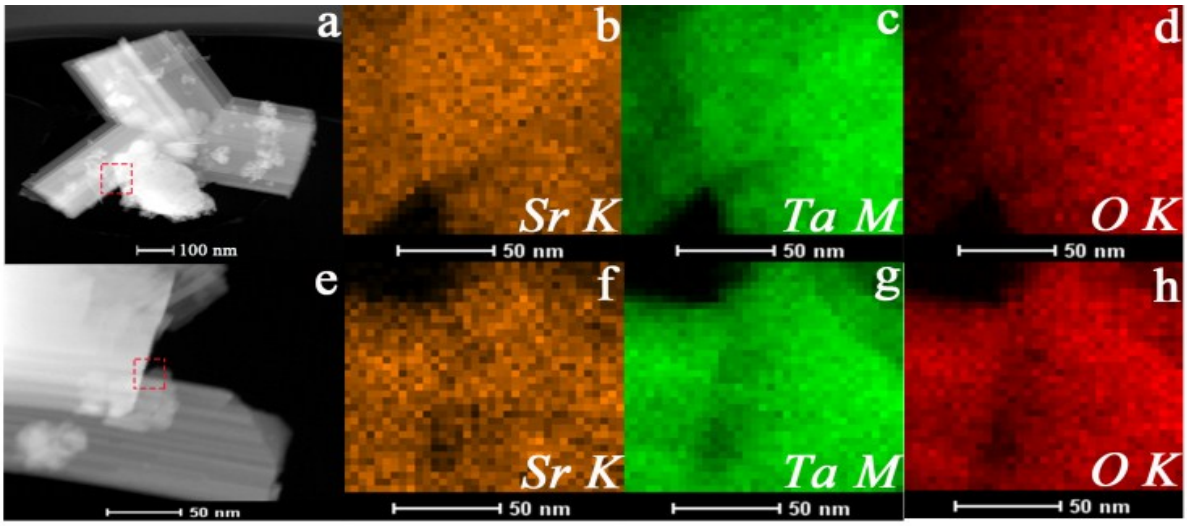


Figure S4. HAADF-STEM images of the as-synthesized samples: SrTa1.3 (a) and SrTa1.7 (b) and the corresponding elemental mapping images of Sr K (b and f), Ta M (c and g) and O K (d and h) on the selected area as indicated by the red dashed frame in a and e, respectively

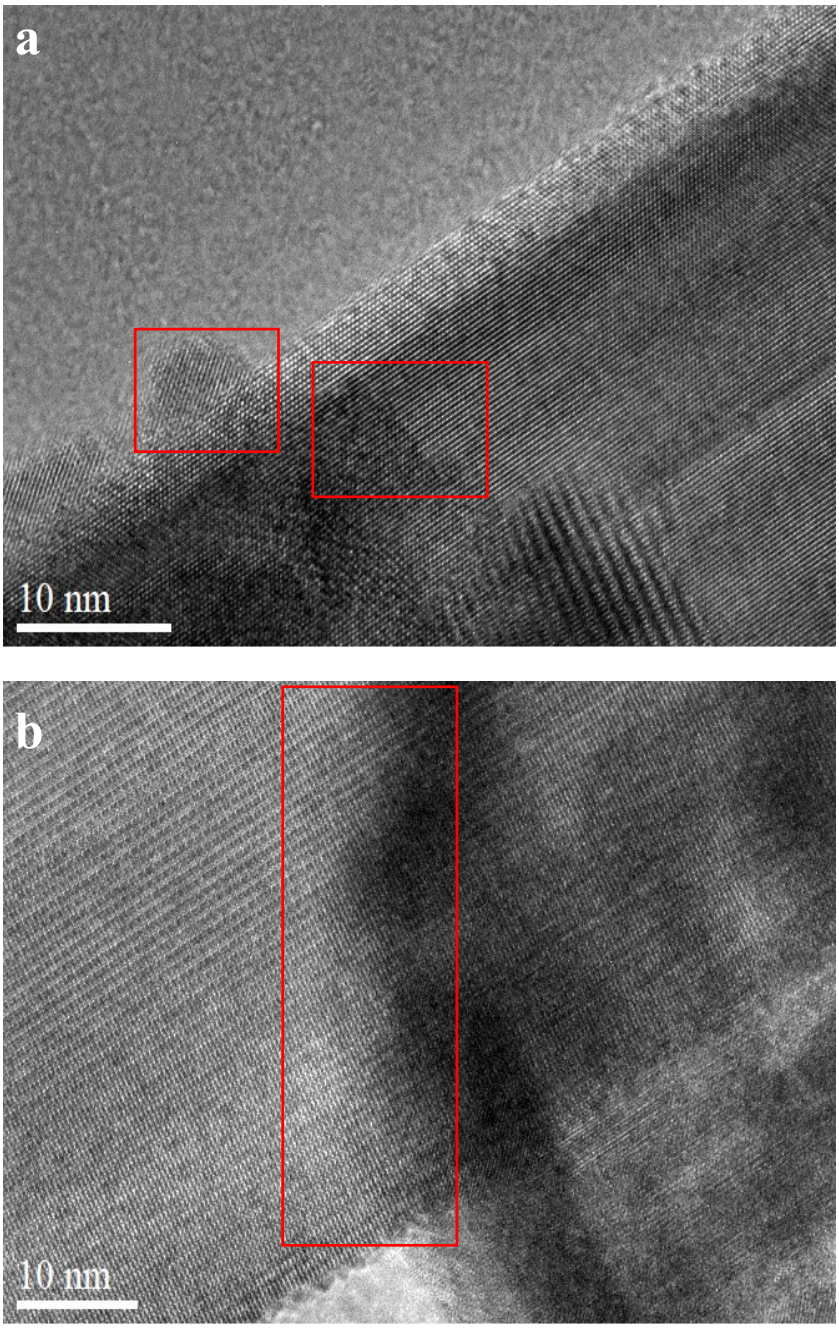


Figure S5. The HRTEM images of the typical SrTa1.3 (a) and SrTa1.7 (b) samples and the formation of the interface junctions among the phase components in an intimate manner (marked by red rectangles).

Theoretical predictions of the position of band edges

The position of band edges of $Sr_5Ta_4O_{15}$, $Sr_2Ta_2O_7$ and $SrTa_4O_{11}$ are calculated by Butler and Ginley method using the equation related to Mulliken electronegativity.[1] Thereby, the conduction band (E_{CB}) and valence band (E_{VB}) edge positions can be calculated according to the empirical equations:

$$E_{CB} = \chi - E^e - 0.5E_g \quad (1)$$

$$E_{CB} = E_{VB} - E_g \quad (2)$$

where χ is the electronegativity of the semiconductor, which is the geometric mean of the electronegativity of the constituent atoms, E^e is the energy of free electrons on the hydrogen scale (4.44 eV), and E_g is the band gap energy of the semiconductor. The χ values for $Sr_5Ta_4O_{15}$, $Sr_2Ta_2O_7$ and $SrTa_4O_{11}$ are ca.5.57 and 5.66 and 6.10 eV, respectively. The bottom conduction band level (E_{CB}) of $Sr_5Ta_4O_{15}$ (-1.12 eV) is much more negative than that of $Sr_2Ta_2O_7$ (-1.02 eV) and $SrTa_4O_{11}$ (-0.34 eV) and the top valence band level (VB) of $Sr_5Ta_4O_{15}$ (+3.38 eV) is more positive than that of $Sr_2Ta_2O_7$ (+3.48 eV) and $SrTa_4O_{11}$ (+3.66 eV). The photogenerated electrons on surface of $Sr_5Ta_4O_{15}$ particles can successively transfer to $Sr_2Ta_2O_7$ and $SrTa_4O_{11}$ via the formed interfaces, and simultaneously, the photogenerated holes on surface of $SrTa_4O_{11}$ can transfer to $Sr_2Ta_2O_7$ and $Sr_5Ta_4O_{15}$ in turn.

Table S1. Estimation of the band positions of strontium tantalates.

	χ	E_g	E_{CB}	E_{VB}
$Sr_5Ta_4O_{15}$	5.57	~4.5	-1.12	+3.38
$Sr_2Ta_2O_7$	5.66	~4.5	-1.02	+3.48
$SrTa_4O_{11}$	6.10	~4.0	-0.34	+3.66

References

1. M. A. Butler and D. S. Ginley, *J. Electrochem. Soc.*, 1978, **125**, 228-232.