## **Electronic Supplementary Information**

Organophotocatalysis system of p/n bilayers for wide visible-light-induced molecular hydrogen evolution

Toshiyuki Abe, \*<sup>a</sup> Junpei Chiba,<sup>a</sup> Misaki Ishidoya<sup>a</sup> and Keiji Nagai<sup>b</sup>

a: Department of Frontier Materials Chemistry, Graduate School of Science and Technology, Hirosaki University, 3 Bunkyo-cho, Hirosaki 036-8561, JAPAN

b: Chemical Resources Laboratory, Tokyo Institute of Technology, Suzukake-dai, Midori-ku, Yokohama 226-8503, JAPAN

\* Correspondence to be addressed

Absorption spectra of  $H_2Pc/C_{60}$  and PTCBI/ $H_2Pc$  bilayers



Fig. S1 Absorption spectra of  $H_2Pc/C_{60}$  and PTCBI/ $H_2Pc$  bilayers. Film thickness of  $H_2Pc/C_{60}$ ,  $H_2Pc = 75$  nm and  $C_{60} = 125$ nm. Film thickness of PTCBI/ $H_2Pc$ , PTCBI = 300 nm and  $H_2Pc = 60$  nm.

**Table S1** Comparison of photoenergy conversion efficiency ( $\eta$ ) and EQE for other visible-light photocatalysts with those values for the present system. For reference, the efficiencies for the photovoltaic cells of C<sub>60</sub>/H<sub>2</sub>Pc are also listed.

System	$\eta$ / %		EQE / % <sup>a</sup>	Reference
Organic photovoltaic cell				
$C_{60}/H_2Pc$	0.03	1.0	(600 nm)	1
$C_{60}/H_2Pc$	0.25		× /	2
$C_{60}/H_2Pc$ including co-evaporant	2.5			2
p-i-n-structured $C_{60}/H_2Pc^b$	2.5	65	(650 nm)	3
Photocatalyst for H <sub>2</sub> evolution				
(co-catalyst)				
TiO <sub>2</sub> <sup>c</sup>	0.4			4
$SrTiO_3$ :Rh (Ru) <sup>d</sup>	0.12	1.7	(420 nm)	5
		0.2	(500 nm)	
dye-modified KTa(Zr)O <sub>3</sub> (Pt)	0.013			6
$In_{0.9}Ni_{0.1}TaO_4$ (NiO <sub>y</sub> )		0.66	(402 nm)	7
$(CuAg)_{0.15}In_{0.3}Zn_{1.4}S_2$ (Ru)		7.4	(520 nm)	8
		1.0	(600 nm)	
TaON $(Pt)^e$		0.4	(420 nm)	9
$[(Ga_{1-x}Zn_x)(N_{1-x}O_x)](Rh_{2-v}Cr_vO_3)$		5.9	(420-440 nm)	10
CdS (Pt-PdS)		93	(420 nm)	11
$g-C_3N_4$ (Pt)		0.1	(420-460 nm)	12
$ZrO_2/TaON(Pt)^e$		6.3	(420.5 nm)	13
$CuGa_2In_3S_8$ (Rh)		15	(560 nm)	14
		12	(600 nm)	
		5	(640 nm)	
$ZnIn_2S_4$ (Pt)		34	(420 nm)	15
		24	(550 nm)	
A wired PEC cell of a commercial	4.7			16
triple-junction amorphous silicon				
photoanode <sup>f</sup>				
A wireless cell of a commercial	2.5			16
triple-junction amorphous silicon				
(NiMoZn)				
$H_2Pc/C_{60}$ (Pt)		4.0	(600 nm)	this work
		1.2	(700 nm)	

<sup>*a*</sup> The value in the parenthesis represents the wavelength employed for measuring EQE.

<sup>*b*</sup> The term "i" indicates a co-deposition layer of  $C_{60}$  and  $H_2Pc$ . The EQE value was estimated from the reported internal quantum efficiency and the absorption ratio of the film employed. <sup>*c*</sup> A typical UV-responsive photocatalyst.

<sup>*d*</sup> In the Z-scheme photocatalysis system, the photocatalyst was concurrently used in combination with BiVO<sub>4</sub> photocatalyst for  $O_2$  evolution.

<sup>*e*</sup> In the Z-scheme photocatalysis system, the photocatalyst was used in combination with Pt-loaded WO<sub>3</sub> photocatalyst for O<sub>2</sub> evolution in the presence of  $IO_3^-/I^-$ .

<sup>f</sup>H<sub>2</sub> occurred at a counter electrode of NiMoZn loaded on Ni mesh.

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