Electronic Supplementary Information

for

A composite photocatalyst of an organic electron donor-acceptor dyad and a Pt catalyst supported on semiconductor nanosheets for efficient hydrogen evolution from oxalic acid

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Fig. S1 Nitrogen adsorption-desorption isotherms of (a) $K_4\text{Nb}_6O_{17}$ and niobate-NS and (b) $K\text{TiNbO}_5$ and titanoniobate-NS at $-196 \, ^{\circ}\text{C}$. 
Fig. S2 Powder X-ray diffraction patterns of K₄Nb₆O₁₇ in the form of (a) nanoscroll and (b) layered oxide (as-prepared). (c) Peak positions and intensities indicated in literature.⁵¹ (d) Schematic drawings of crystal structure. NbO₆ units are indicated by green octahedra with red dots and K ions are indicated by purple circles.⁵²

Fig. S3 Powder X-ray diffraction patterns of KTiNbO$_5$ in the form of (a) nanoscroll and (b) layered oxide (as-prepared). (c) Peak positions and intensities indicated in literature.$^3$ (d) Schematic drawings of crystal structure. TiO$_6$ or NbO$_6$ units are indicated by blue octahedra with red dots and K ions are indicated by purple circles.$^2$

Fig. S4 TG/DTA curves of (a) niobate-NS and (b) titanoniobate-NS at temperature ranging from 20 °C to 600 °C.
Fig. S5 (a) Nitrogen adsorption-desorption isotherms at -196 °C and (b) powder X-ray diffraction of QuPh⁺–NA/niobate-NS (red) and niobate-NS (black).
Fig. S6  TG/DTA curves of (a) QuPh⁺–NA/niobate-NS and (b) QuPh⁻–NA/titanoniobate-NS at temperature ranging from 20 °C to 600 °C.
**Fig. S7** IR absorption spectra of (a) (QuPh$^+$–NA)(ClO$_4$), (b) QuPh$^+$–NA/niobate-NS and (c) QuPh$^+$–NA/titanoniobate-NS. The samples were pelletized under pressurized conditions after mixing with KBr. The IR spectra were taken on a Jasco FT/IR-6200 spectrometer with the resolution of 4 cm$^{-1}$. 
Fig. S8 TEM images of QuPh\textsuperscript{7−}–NA/niobate-NS after the photoirradiation for 19 h in the presence of oxalic acid and [Pt(NH\textsubscript{3})\textsubscript{4}]\textsuperscript{2+}.
**Fig. S9** Time course of absorbance of QuPh$^+$–NA/niobate-NS at 400 nm during the photocatalytic H$_2$ evolution.

**Fig. S10** H$_2$-evolution rates depending on the irradiance of a solar simulator (AM 1.5G). The photocatalytic H$_2$ evolution was performed by photoirradiation of an N$_2$-saturated aqueous suspension (2.0 mL) containing oxalic acid (50 mM), K$_2$PtCl$_4$ (10 μM) and QuPh$^+$–NA/niobate-NS (11.6 mg) using an HAL-320 solar simulator (Asahi spectra, AM 1.5G). The irradiance for each reaction was determined by a CS-20 irradiation meter (Asahi spectra).
Figure S11 Time courses of CO$_2$ evolution by photoirradiation ($\lambda > 340$ nm) of an O$_2$-saturated aqueous suspension (2.0 mL, pH 1.4) containing oxalic acid (50 mM) and niobate-NS (11.6 mg, black) and QuPh$^+$–NA/niobate-NS (11.6 mg, red).