## Light driven water oxidation on silica supported NiO-TiO<sub>2</sub> heteronanocrystals yields hydrogen peroxide

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Figure S1. HRTEM image of NiO-TiO<sub>2</sub> heteronanocrystal.



**Figure S2.** A) UV-Visible absorption spectra of dilute dispersions of TiO<sub>2</sub>, TiO<sub>2</sub>-NiO, and B) Photoluminescence emission spectra ( $\lambda_{ex}$  = 300 nm) of *anatase* TiO<sub>2</sub> and TiO<sub>2</sub>-NiO HNCs.



**Figure S3**. Photograph of aqueous suspension of SiO<sub>2</sub>/NiO-TiO<sub>2</sub> (NiO-TiO<sub>2</sub> heteronanocrystals supported on fumed silica) after 24 hours illumination with unfiltered light from 150W Xe arc lamp.



**Figure S4**. Irradiation of SiO<sub>2</sub>/NiO-TiO<sub>2</sub> in dilute aqueous silver nitrate leads to formation of Ag nanoparticles. A) Photograph of the catalyst suspension after irradiation. B) TEM image of Ag nanoparticles.

Alternatively, we demonstrate a fast colorimetric test for hole scavenging upon irradiation of aqueous, anaerobic  $Fe^{2+}$  and thiocyanide ion that gives dark red colored solutions upon oxidation to  $Fe^{3+}$  (Figure S2). In these conditions, the absorbance at 450 nm abruptly rises, then decreases to a constant value that we presume is due to the back-reaction in which  $Fe^{3+}$  is reduced back to  $Fe^{2+}$  by photogenerated electrons on  $TiO_2$ .



**Figure S5**. a) Formation of an iron (III) thiocyanate complex ion  $Fe(SCN)^{2+}$  from  $Fe^{3+}$  and  $SCN^{-}$  in the reaction vessel b) absorbance maxima  $\lambda_{max}^{} = 450$  nm of  $Fe(SCN)^{2+}$  taken at desired time interval.

## Analysis of EDS data for NiO-TiO2 heteronanocrystal sample.

Z	Element Family Mass Error (%)		Atomic Fraction (%) Fit error (%)			Atomic Error (%)	Mass Fraction (%)	
6	С	К	72.91	5.19	57.99	2.48	0.09	
7	Ν	К	0.00	0.04	0.00	0.04	0.00	
8	0	К	18.47	3.92	19.57	4.00	0.15	
11	Na	К	1.59	0.33	2.42	0.49	0.24	
13	AI	К	2.02	0.42	3.60	0.73	0.23	
14	Si	К	0.84	0.17	1.56	0.31	0.46	
15	Ρ	К	0.21	0.04	0.43	0.08	2.18	
19	K	К	0.34	0.06	0.87	0.16	0.88	
22	Ti	К	0.75	0.11	2.38	0.33	0.38	
26	Fe	К	0.16	0.02	0.59	0.08	1.26	
28	Ni	К	2.73	0.41	10.60	1.48	0.12	

2023-07-07 11:49:39 Analysis of spectrum: Spectra from Area #1 (Brown-Powell general)

2023-07-07 11:52:58 Analysis of spectrum: Spectra from Area #1 (Schreiber-Wims oxide-optimized)

Z	Element Family Mass Error (%)		Atomic Fraction (%) Fit error (%)			Atomic Error (%)	Mass Fraction (%)	
6	С	K	69.25	5.28	52.23	2.38	0.09	
7	Ν	K	0.00	0.04	0.00	0.04	0.00	
8	0	K	19.87	4.25	19.97	4.10	0.15	
11	Na	K	1.83	0.39	2.64	0.54	0.24	
13	AI	K	2.35	0.50	3.99	0.81	0.23	
14	Si	K	0.99	0.20	1.74	0.34	0.46	

15	Ρ	К	0.25	0.05	0.48	0.09	2.18
19	К	К	0.41	80.0	1.00	0.18	0.88
22	Ti	К	0.94	0.14	2.82	0.40	0.38
26	Fe	К	0.21	0.03	0.75	0.11	1.26
28	Ni	К	3.90	0.60	14.39	2.03	0.12