Supporting information

Anatase $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$: A novel photocatalyst for solar hydrogen production

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Experimental Section

*Synthesis of $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$ and TaON:*The $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$ powders investigated in this study was prepared by two-step process as previous report.[1] First, amorphous ternary phases in the system $Mg_{0.05}Ta_{0.95}O_{2.425}$ were prepared using a modified Pecchini method. In detailed, 0.1538 g $MgCl_2{\cdot}6H_2O$ was dissolved in 60 mL ethanol which contains 2 g citric acid. 5.15 g $TaCl_5$ was solved in 150 ml ethanol containing 36.5 g citric acid. Then both above solutions were mixed together and 16 ml ethylene glycol was added. After that, the solvent and HCl were evaporated and citrated complexes together with ethylene glycol have been polymerized at 150 °C. The organic residues were burnt off at 600 °C for 16 h to give white amorphous $Mg_{0.05}Ta_{0.95}O_{2.425}$ powders. Anatase $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$ and TaON were respectively prepared by annealing the amorphous $Mg_{0.05}Ta_{0.95}O_{2.425}$ and the pre-making amorphous Ta_2O_5 in moist ammonia (bubbled through saturated ammonia solution) at a constant flow rate of 150mL/min at 800 °C for 16 h.

Characterization: The XRD patterns were collected in the 2θ range of 10-82° on a Bruker D8 Advance Diffractometer, using Cu Kα radiation (λ=1.5406 Å). Diffuse reflection spectra (DRS) were taken on a UV-Vis spectrophotometer (Shimadzu UV-2550) by using $BaSO_4$ powders as a standard reference and converted from reflection to absorbance by the Kubelka-Munk method. The microstructure morphology was investigated using scanning electron microscopy (Nova NanoSEM230) and transmission electron microscopy (JEM-200CX).X-Ray photoelectron spectroscopy (XPS) data and valenceband X-ray photoelectron spectroscopy (VB XPS) spectrum were obtained on PHI5000 Veras Probe instrument with a 200 W monochromated Al Kaline source. XPS data were analyzed with the MultiPaksoftware. The visible light induced catalytic H_2 evolution reaction was carried out in a quartz cell connected to a closed glass gas circulation system. The H_2 was analyzed with an online TCD gas chromatograph (GC-8A, Shimadzu, argon carrier). Typically, 0.15 g of catalyst powder was suspended in 270 mL of methanol solution ($V_{water}/V_{methanol}$= 4:1). 0.1 wt% Pt (or 0.1 wt% Ru) cocatalyst was photodeposited on the catalyst by using H_2PtCl_6 (or

$(NH_4)_2RuCl_6$) dissolved in reaction solution. A cutoff filter (which can intercept the light of wavelength shorter than 420nm, 450nm, or 500 nm) was used for visible light and a water filter was used to eliminate the temperature effect.

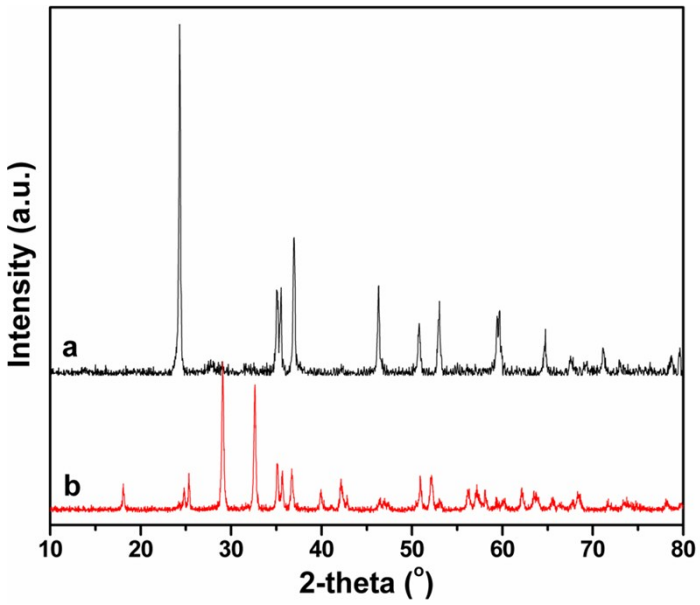

Figure S1 X-ray diffraction patterns of the as-prepared samples of $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$ (a) and TaON (b).

Table S1 Atomic structure parameters of $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$

Atom	x	y	z	B_{iso}	sof	Wyck.
Ta	0.000	0.250	0.375	0.340	0.950	4b
Mg	0.000	0.250	0.375	0.340	0.050	4b
O	0.000	0.250	0.584	1.800	0.575	8e
N	0.000	0.250	0.584	1.800	0.425	8e

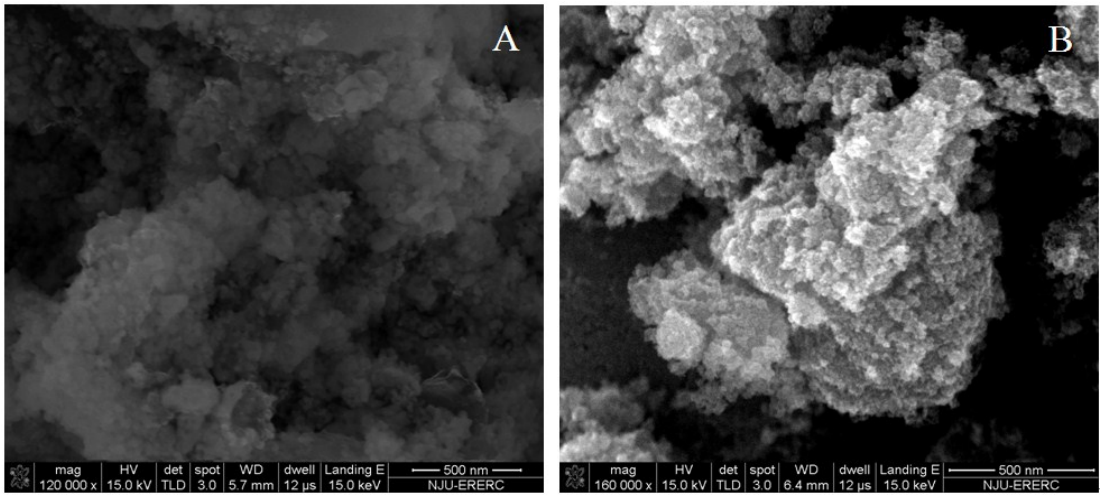

Figure S2 Scanning electron microscopy (SEM) images of the as-prepared $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$ (A) and TaON (B) powders.

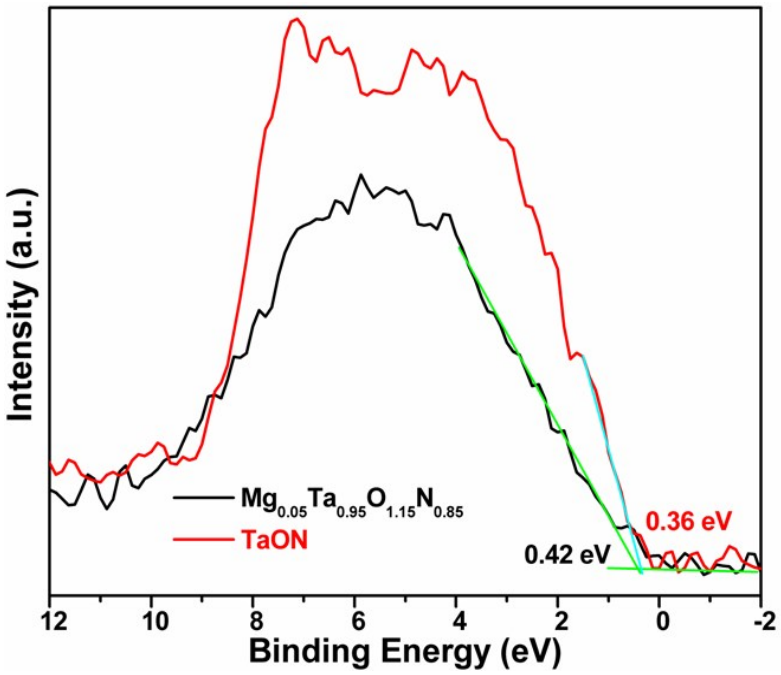

Figure S3 XPS valence band spectrum of anatase phase $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$ and TaON samples.

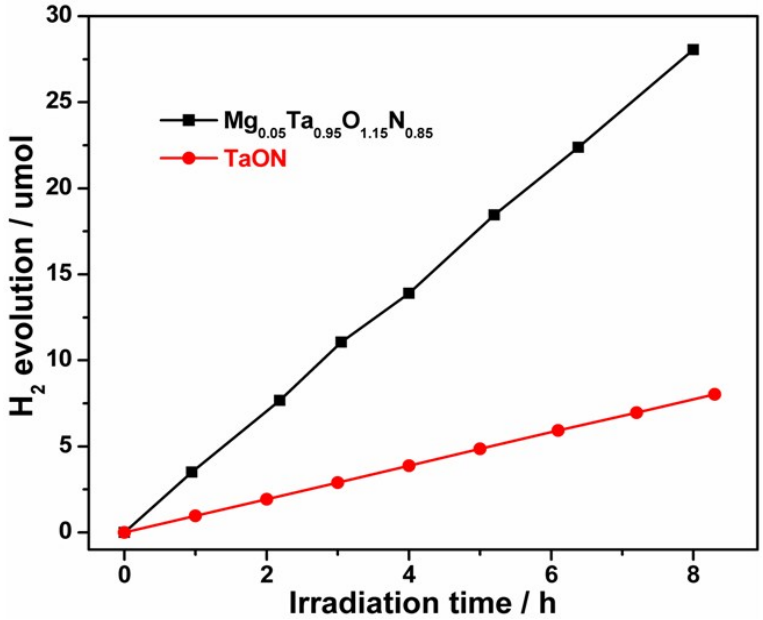

Figure S4 H_2 evolution from methanol solution under λ≥420 nm light irradiation by 0.1 wt% Pt-deposited $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$ and TaON.

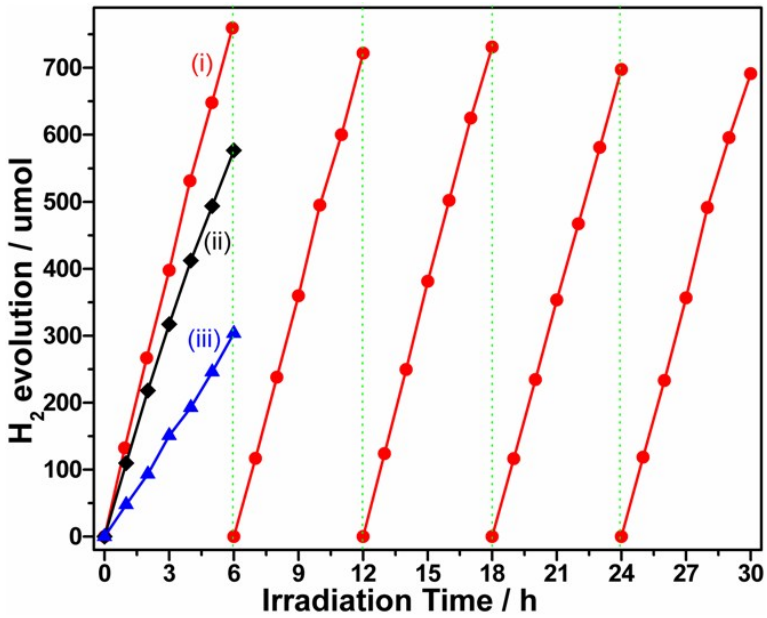

Figure S5 Hydrogen evolution from methanol solution over 0.1 wt% Ru-deposited $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$ under λ≥420 nm light irradiationduring five reaction cycles (i) and under λ≥450 nm(ii)and (iii) 500 nm light irradiation.

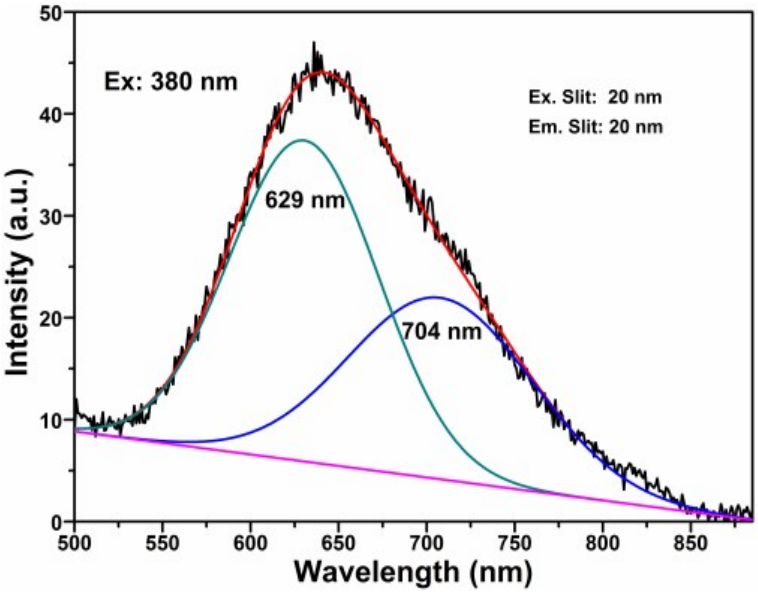

Figure S6 Room temperature photoluminescence spectra existed under 380 nm of TaON sample.

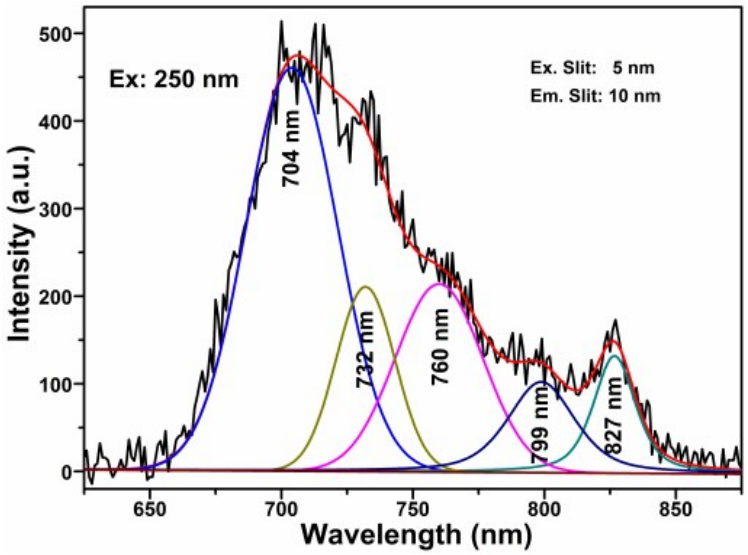

Figure S7 Room temperature photoluminescence spectra existed under 250 nm of anatase phase $Mg_{0.05}Ta_{0.95}O_{1.15}N_{0.85}$ sample.

Reference

1 H. Schilling, M. Lerch, A. Börger, K. D. Becker, H. Wolff, R. Dronskowski, T. Bredow, M. Tova and C. Baehtz,*J. Solid State Chem.*, 2006,**179**, 2416−2425