Electronic Supplementary Material (ESI) for Journal of Materials Chemistry

Supporting Informations

Visible light-sensitive mesoporous N-doped Ta_2O_5 spheres: Synthesis and photocatalytic activity for hydrogen evolution and CO_2 reduction

Tomiko M. Suzuki,* Tadashi Nakamura, Shu Saeki, Yoriko Matsuoka, Hiromitsu Tanaka,

Kazuhisa Yano, Tsutomu Kajino and Takeshi Morikawa

Toyota Central R & D Labs, Inc., 41-1Nagakute, Aichi 480-1192, Japan

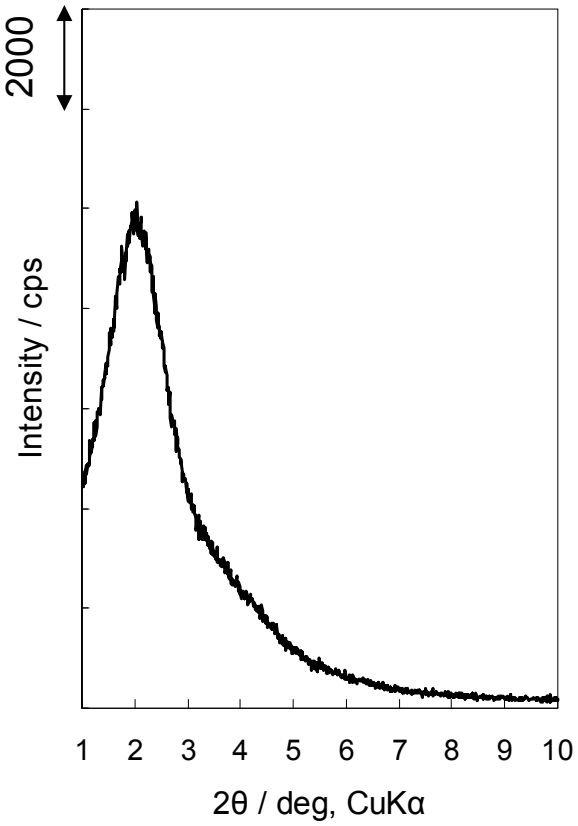

Fig. S1 XRD pattern of Ta_2O_5 sphere synthesized with water/methanol/ethylene glycol (30/50/20).

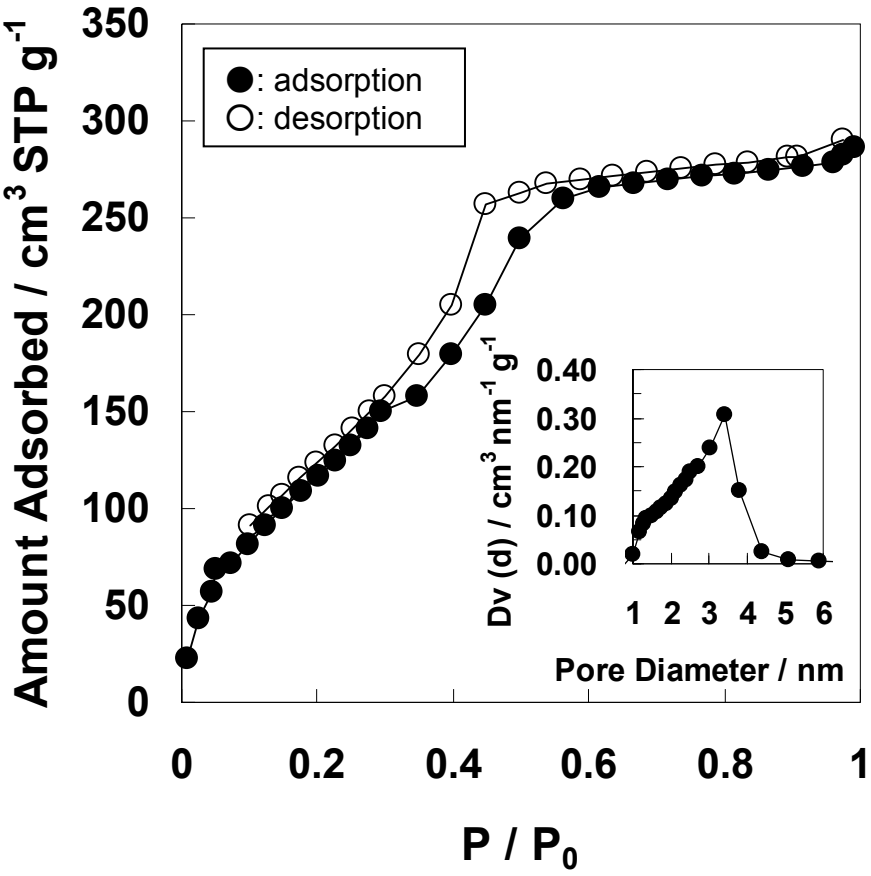

Fig. S2 The nitrogen adsorption-desorption for the MTS obtained with water/methanol/ethylene glycol (30/50/20). Inset shows pore size distribution curve for the MTS evaluated by the BJH method from adsorption branch.

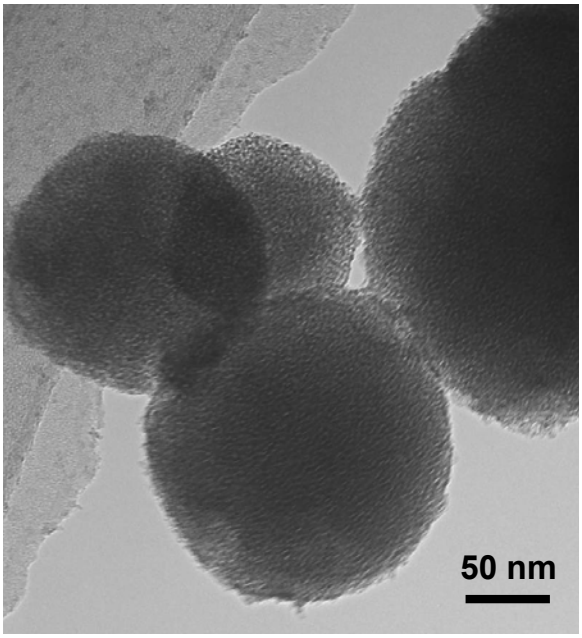

Fig. S3 TEM image of MTS.

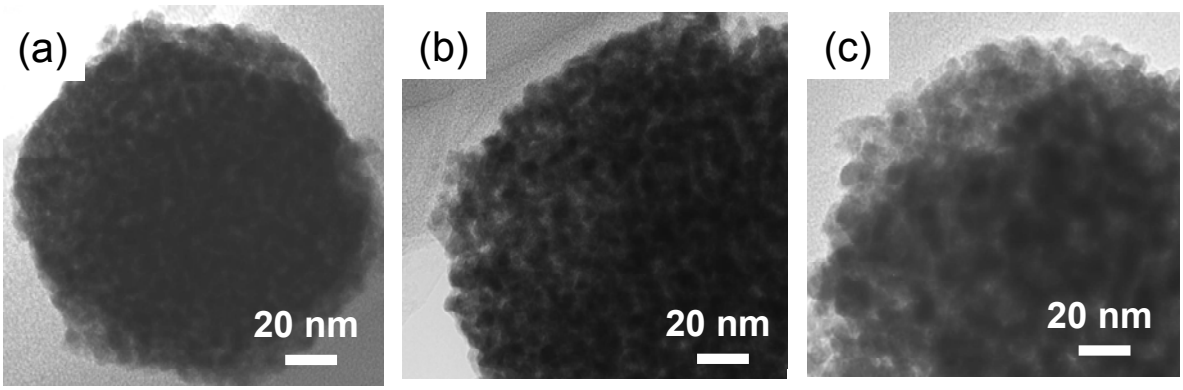

Fig. S4 TEM image of resultant CMTS heat-treated at (a) 1073 K (CMTS-1073K), (b) 1123 K (CMTS-1123K), and (c) 1173 K (CMTS-1173K).

Table S1. Photocatalytic activities for hydrogen evolution using various Ta_2O_5 samples under UV irradiation.[a]

run	sample	mesoporous structure	crystalline structure	Amount of H_2 evolved [b] (μ mol)
1	MTS	◯	–	19.3
2	MTP [c]	◯	–	14.2
3	CMTS-1073K	◯	◯	47.5
4	CMTS-1123K	◯	◯	41.6
5	CMTS-1173K	◯	◯	40.1
6	non-CMTS	–	◯	13.0
7	Ta_2O_5-fine [d]	–	◯	29.8

[a] Reaction condition: 4 mg catalyst in 4 ml Ar-saturated 20 vol% methanol/H_2O solution at ambient temperature. Solutions ware irradiated using a 500 W Xe lamp for 24 h. Hydrogen concentration was determined by gas chromatography.
[b] Total amount of evolved H_2 after 24 h reaction.
[c] prepared according to ref 26 (SEM image is shown in Fig. S5a).
[d] prepared according to ref 9 (SEM image is shown in Fig. S5b).

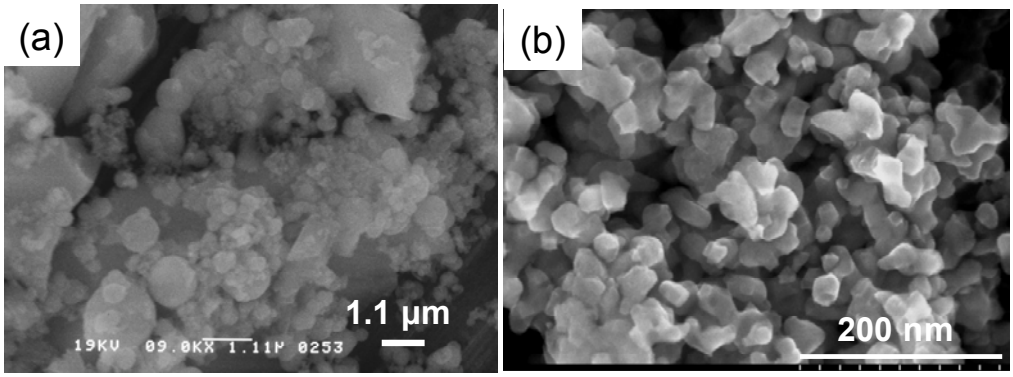

Fig. S5 SEM image of (a) MTP[a] and (b) Ta_2O_5-fine[b].

[a]prepared according to ref 26.

[b] prepared according to ref 9 .

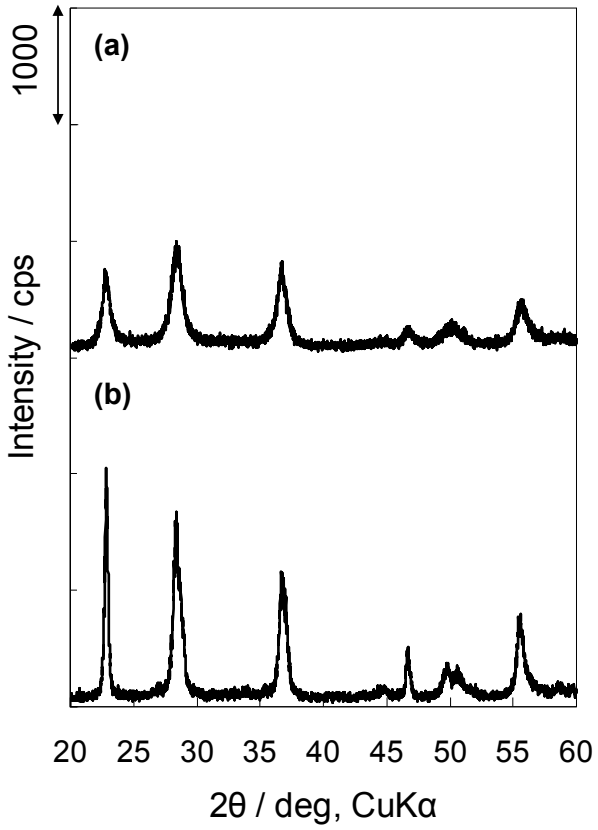

Fig. S6 XRD patterns of (a) N-CMTS and (b) N-Ta_2O_5-fine.

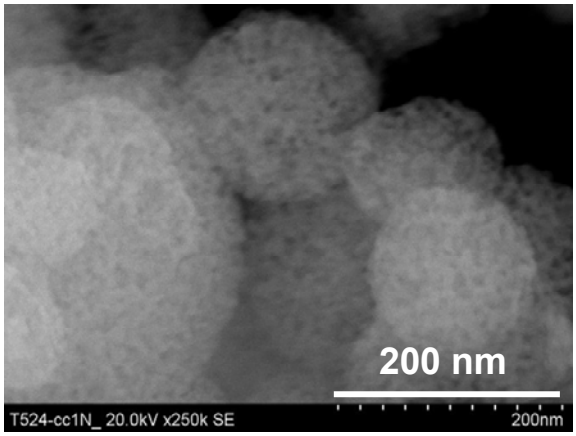

Fig. S7 FE-SEM image of N-CMTS.

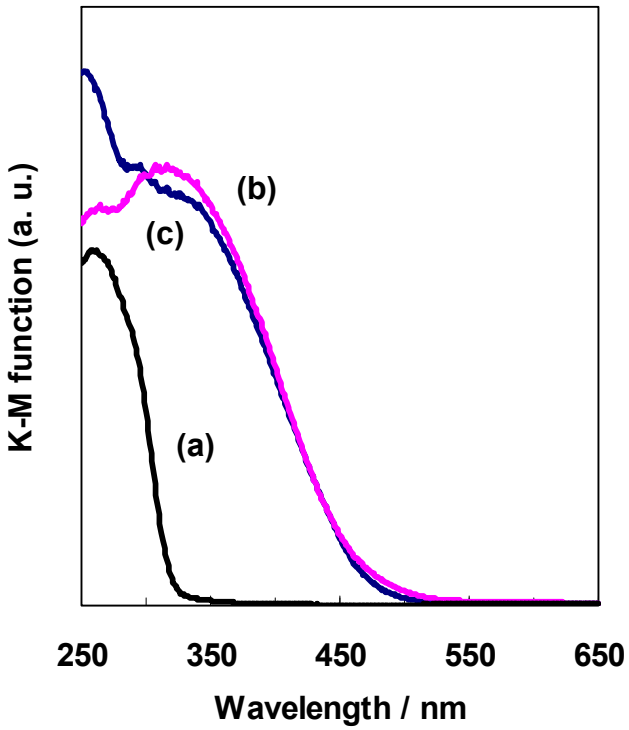

Fig. S8 UV-vis diffuse reflection spectra of (a) CMTS, (b) N-CTMS, and (c) N-Ta2O5-fine**.**

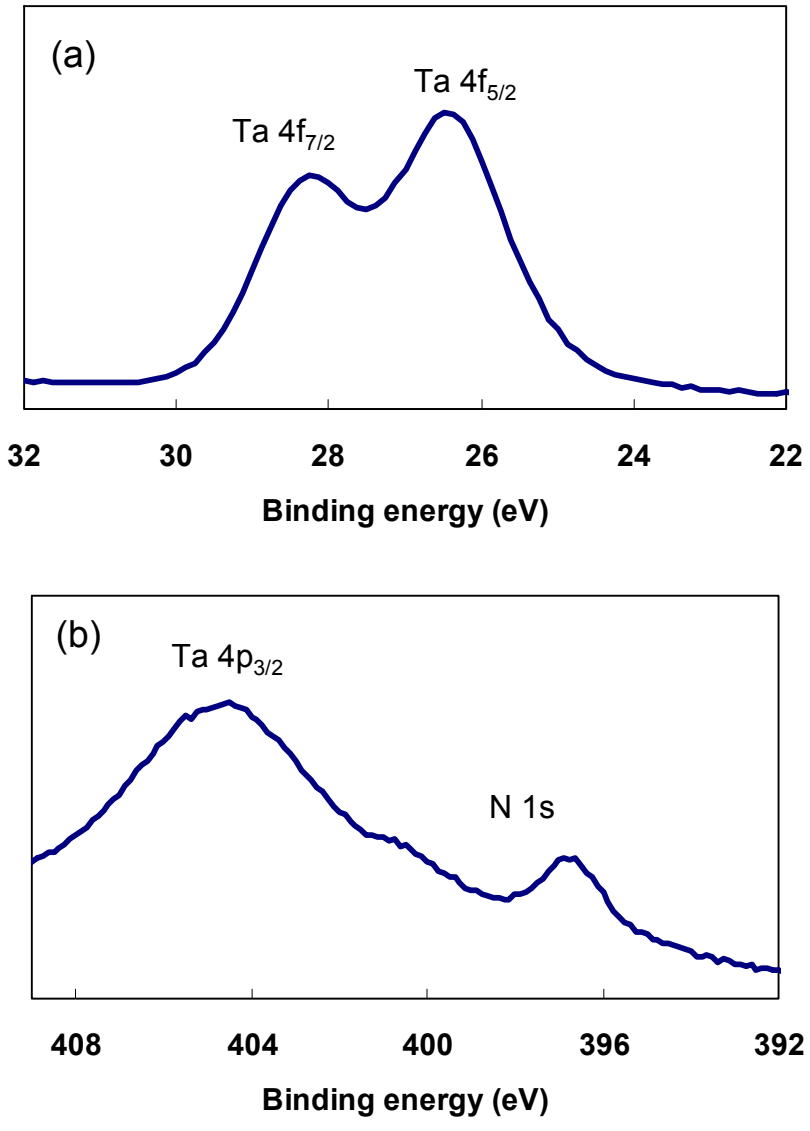

Fig. S9 XPS spectra of N-CMTS. (a) Ta $4f_{7/2}$ and Ta $4f_{5/2}$ spectra and (b) Ta $4p_{3/2}$ and N 1*s* spectra.

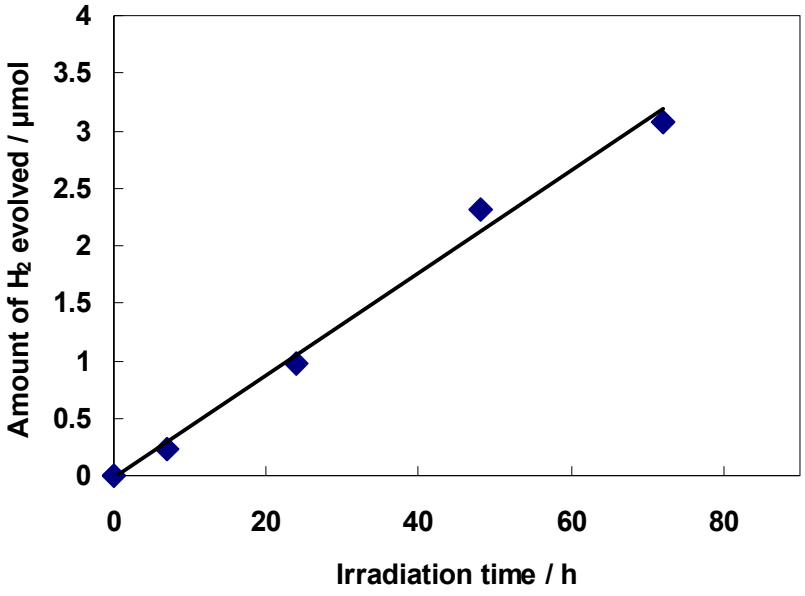

Fig. S10 Photocatalytic activities for hydrogen evolution using N-CMTS under visible-light irradiation.

Reaction condition: 4 mg catalyst in 4 ml Ar-saturated acetonitrile/ triethanolamine solution at ambient temperature. Solutions ware irradiated using a 500 W Xe lamp with filters to produce light in the range $410 \leq \lambda \leq 750$ nm at room temperature. Hydrogen concentration was determined by gas chromatography.

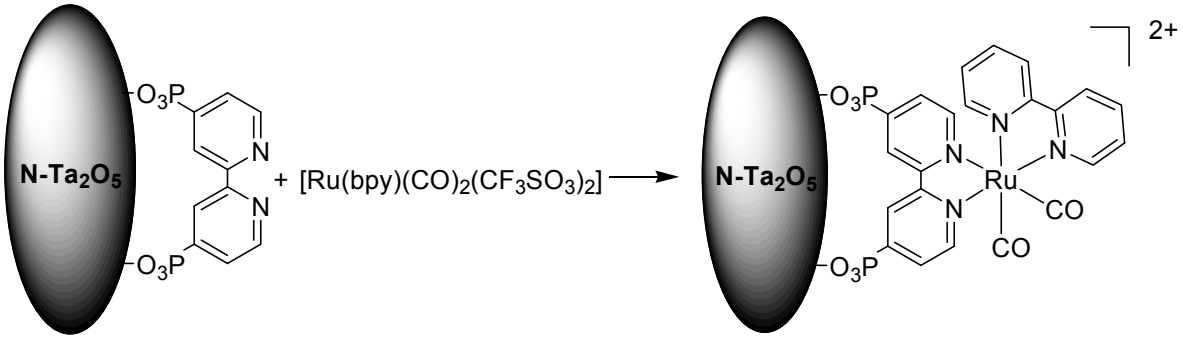

Fig. S11 Ru-complex assembled on bipyridine (bpy)-functionalized N-CMTS (direct assembly method).

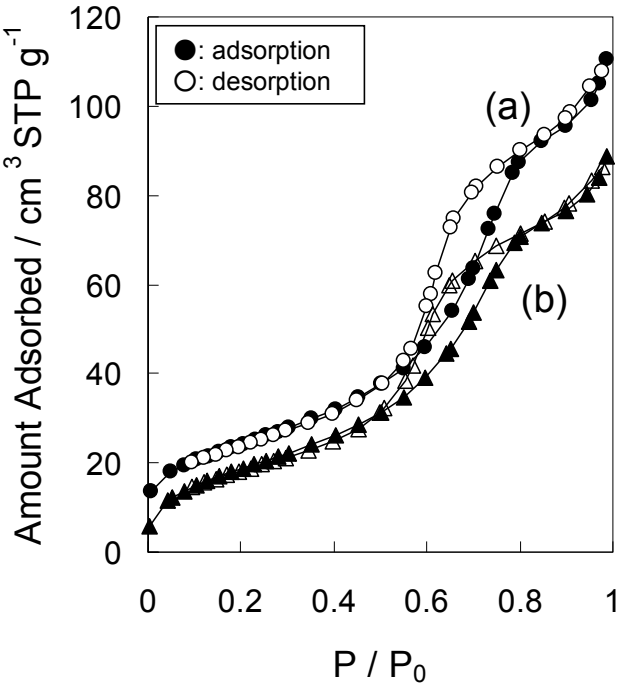

Fig. S12 Nitrogen adsorption-desorption for (a) N-CMTS and (b) [Ru-dpbpy]/N-CMTS (Ru content: 0.12 wt%).