A composite Ta_3N_5 photoanode with ultrahigh photocurrent and stability for solar water oxidation

Yongle Zhao,[a,b] **Jingying Shi,**[a] **Can Li**[a]*
[a]*State key Laboratory of Catalysis, Dalian Institute of Chemical Physics, Chinese Academy of Sciences; Dalian National Laboratory for Clean Energy, Dalian, 116023, China*
[b]*University of Chinese Academy of Sciences, Beijing, 100049, China*
**Corresponding author: canli@dicp.ac.cn*

Abstract: Tantalum nitride (Ta_3N_5) is a very promising photoanode material with a narrow band gap (2.1eV) and suitable band alignment for solar water splitting. However, it suffers from severe photocorrosion during water oxidation. Here, we report functional bilayer modified Ta_3N_5 photoanode can generate a photocurrent approaching the theoretical limit of 12.9 mA cm^{-2} at 1.23 V versus the reversible hydrogen electrode (RHE) and be sustainably operated for 24 hours, which undoubtedly sets a new record in photocurrent and stability of Ta_3N_5 based photoanode. This work may inspire a new avenue for the design of highly stable PEC systems for practical applications.
Keywords: water splitting, tantalum nitride, highly stable.

1. Introduction (11-point boldface)

Photoelectrochemical (PEC) water splitting is an ideal approach for renewable solar fuel production. One of the major hurdles that impede the practical application of this strategy is the lack of efficient and stable photoanodes.[1] Tantalum nitride (Ta_3N_5) has been studied extensively as a photoanode in recent years due to its optimal band alignment together with a maximum solar energy conversion efficiency of 15.9 %.[2] The integrated Ta_3N_5 photoanode based on hole storage layer (HSL) shows a record photocurrent of 12.1 mA cm^{-2} at 1.23 V vs. RHE. [3] However, the duration against photocorrosion is less than 30 minutes. Herein, we introduce a composite Ta_3N_5 photoanode based on HSL can generate a photocurrent approaching the theoretical limit of 12.9 mA cm^{-2} at 1.23 V vs. RHE and be sustainably operated for 24 hours.

2. Experimental

The pristine Ta_3N_5 photoanodes were fabricated on metal tantalum foil by way of anodization-hydrothermal-nitridation. HSL was loaded by solution deposition. The decoration of cocatalyst nanoparticles was achieved in hydrothermal method.

All the photoelectrochemical tests were conducted in Ar saturated NaOH aqueous (1 M, pH=13.6) under simulated light (AM 1.5 G, 100 mW cm^{-2}) unless otherwise mentioned.

3. Results and discussion

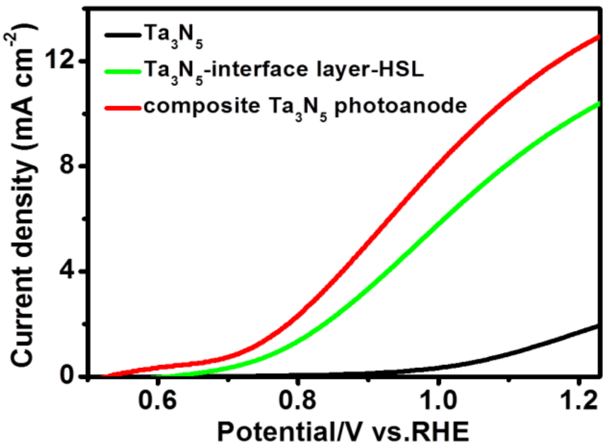

Figure 1. Current-potential curves of Ta_3N_5, Ta_3N_5-interface layer-HSL, composite Ta_3N_5 photoanodes.

In contrast to the pristine Ta_3N_5 photoanode, the combination of interface layer and HSL can extremely enhance the activity of Ta_3N_5 photoanode, and the photocurrent reaches 10 mA cm^{-2} at 1.23 V vs. RHE. To further ameliorate water oxidation kinetics, cocatalyst was loaded on the Ta_3N_5-interface layer-HSL. The obtained composite photoanode exhibits an incredible photocurrent of 12.9 mA cm^{-2} at 1.23 V, which is the theoretical maximum photocurrent of a Ta_3N_5 photoanode.

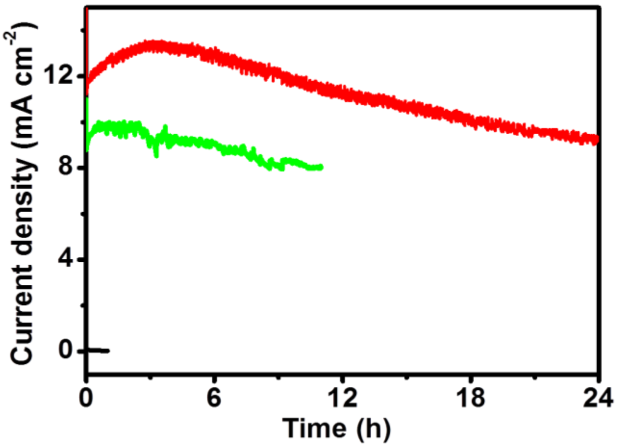

Figure 2. Chronoamperometry messurement.

We further confirmed the stability of the composite photoanode at 1.23 V under AM 1.5G simulated sunlight. The photocurrent can maintain about 90% of the initial value after 24 h, which outperforms all of the previous reports on Ta_3N_5 photoanodes for water oxidation.

4. Conclusions

In summary, we demonstrate the highly stable composite Ta_3N_5 photoanode delivers a theoretical limit photocurrent of 12.9mA cm^{-2} at 1.23 V vs. RHE and simultaneously shows excellent stability on the timescale of one day. The combination of HSL and other functional layers can contribute to hole extraction from Ta_3N_5, which suggests that the HSL strategy can open up a new avenue for developing efficient and stable photoelectrodes in the field of solar fuel production.

References

1. M. G. Walter, E. L.Warren, J. R. McKone, S. W. Boettcher, Q. Mi, E. A. Santori, N. S. Lewis, Chem. Rev 110 (2010) 6446.
2. A. Murphy, P. Barnes, L. Randeniya, I. Plumb, I. Grey, M. Horne, J. Glasscock, Int. J. Hydrogen Energy 31 (2006) 1999.
3. G. Liu, S. Ye, P. Yan, F. Xiong, P. Fu, Z. Wang, Z. Chen, J. Shi, C. Li, Energy Environ. Sci., 9 (2016) 1327.