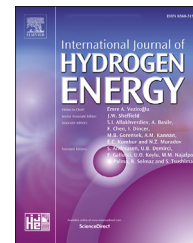




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# The enhanced photocatalytic hydrogen production of the fusiform $g\text{-C}_3\text{N}_4$ modification $\text{CaTiO}_3$ nano-heterojunction

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## ABSTRACT

The fusiform  $g\text{-C}_3\text{N}_4/\text{CaTiO}_3$  nano-heterojunctions is synthesized via simple preparation using the hydrothermal co-deposition method. The results of morphology and structure imply that the  $g\text{-C}_3\text{N}_4$  has deposited on the surface of fusiform  $\text{CaTiO}_3$  successfully, and the photocatalytic activity of the  $g\text{-C}_3\text{N}_4/\text{CaTiO}_3$  nano-heterojunctions exhibits a remarkably enhancement of 18 times than that of the unmodified sample. Further, proved by the transient photocurrent, PL, EIS and Mott-Schottky plots, the photocatalytic hydrogen production enhancement could be ascribed to the nano-heterojunction at the interface.

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## Introduction

Currently, the energy shortage and the environmental pollution are recognized as the global issues for restricting the further development of industry, thus, the exploration of new energy would be a burning issue. Compared with the wind energy, hydraulic energy or nuclear energy, the solar energy, with enormous reserves and renewability, would be regarded as the most potential choice [1–4]. Up to now, series of inspiring achievements have been reported by different methods, such as solar cells, photothermal technique or photochemical technique [5–9], etc. Especially the

photocatalytic hydrogen production [10,11], with the high energy density and green combustion product of the hydrogen [12,13], has become a focused research at present.

So far, series of materials have been reported to achieve the photocatalytic hydrogen production performance, such as  $\text{ZnO}$ ,  $\text{SnO}_2$ ,  $\text{MoS}_2$ ,  $\text{WO}_3$  or  $\text{Fe}_2\text{O}_3$ , etc [14–19]. Among these materials, the perovskites ( $\text{ABO}_3$ ) structures [20], with the appropriate conduction band to redox potential of  $\text{H}^+/\text{H}_2$ , have been reported as the promising materials. In particular the  $\text{CaTiO}_3$ , with the easy preparation and physical-chemical stability [21], has attracted lots of attentions. However, restricted by the wider band gap and higher photon-generated carriers recombination, the hydrogen production

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performance would be expected to improve further, and lots of attempts have been tried, such as Lozano-Sanchez's group, who have reported the  $\text{Er}^{3+}$  doped  $\text{CaTiO}_3$  with long-lived photoinduced charge-carriers for enhancing photocatalytic  $\text{H}_2$  production [22], Liu's group, who have prepared the Z-scheme three-dimensionally ordered macroporous  $\text{SrTiO}_3/\text{Ag}/\text{Ag}_3\text{PO}_4$  photocatalysts with remarkable contaminant degradation [23], Do's group, who have fabricated the Pt-loaded  $g\text{-C}_3\text{N}_4$  nanolayers with enhanced hydrogen production [24], etc. It's obvious that, most of the researches focus on increasing the photon-generated carriers efficiency, light utilization or surface reaction sites [25,26]. Integrated above factors, including the preparation cost and efficiency, the surface heterojunction modification would be perceived as a desired method involves above advantages, such as Cui et al., who have synthesized the  $\text{Cu}_2\text{O}/\text{BiVO}_4$  heterojunction catalysts with remarkable visible light photocatalytic activity [27], Liu groups, who have prepared the  $\text{WO}_3/\text{Sb}_2\text{S}_3$  heterojunction photocatalyst for enhancing the efficiency in water splitting [28], Shi et al., who have reported the  $\text{In}_2\text{S}_3/\text{CdIn}_2\text{S}_4/\text{In}_2\text{O}_3$  ternary heterostructure for enhancing the photocatalytic hydrogen production [29], Salavati-Niasari groups, who have fabricated the  $\text{Cu}_2\text{CdI}_4/\text{CuI}$  heterojunction for improving the photocatalytic performance [30], Liu et al., who have fabricated the Z-scheme  $\text{SnS}_2/\text{Bi}_2\text{MoO}_6$  heterojunction photocatalysts with excellent photocatalytic activity [31], and so on [32,33]. However, in view of the electric potential of the  $\text{H}_2\text{O}$ , the process of photocatalytic hydrogen production demands that the conduction band should be negative (vs NHE, Normal Hydrogen Electrode) [34–36]. As known, for such heterojunction, the photon-generated electrons would transfer to the lower conduction band, which means that the performance of photocatalytic hydrogen production depends on the lower conduction band [37,38] and restricts the optional material for this modification immensely. Therefore, the suitable materials are significant for improving the photocatalytic hydrogen production.

There, the graphitic carbon nitride ( $g\text{-C}_3\text{N}_4$ ) [39], with remarkable visible light response (2.7 eV), unique electronic band structure and low cost, has been reported as an promising visible light photocatalyst for the photocatalytic hydrogen production [40], such as Liu's group, who have obtained the  $\text{Au}/\text{C}_3\text{N}_4$  with plasmonic hot-electron injection for enhanced photocatalytic hydrogen evolution [41], Li's group, who have reported the 2D  $\text{MoS}_2/\text{O-g-C}_3\text{N}_4$  heterostructure with enhanced photocatalytic hydrogen evolution [42], Shi's group, who have prepared the  $g\text{-C}_3\text{N}_4/\text{InVO}_4$  nanocomposites and enhanced the photocatalytic hydrogen production activity [43], etc [44]. As revealed, in vast researches, the  $g\text{-C}_3\text{N}_4$  is always considered as the main photocatalyst. There, it's interesting that the  $g\text{-C}_3\text{N}_4$  with nanostructure could also act as the modification materials, which could provide sufficient specific surface areas and increase the separation of the photon-generated carriers for improving the photocatalytic hydrogen production [45].

In addition, the morphology is deemed as another important factor for the nano-photocatalyst [46], and lots of inspiring achievements have been reported, such as Xu et al., who have reported the hollow  $\text{CaTiO}_3$  cubes for efficient photocatalytic hydrogen production [47], Ghanbari et al., who

have prepared different morphologies of  $\text{RbPbI}_3$  nanostructures to investigate the photocatalytic performance [48], Hu et al., who have prepared the  $\text{SrTiO}_3$  nanofiber with enhanced photocatalytic activity via electrospun [49], etc. There, the  $\text{CaTiO}_3$ , with the fusiform nanostructure, could provide large reaction sites for the deposition of the  $g\text{-C}_3\text{N}_4$  nanosheets and photocatalysis, which is also considered as another advantage for this heterojunction.

In this work, we prepared the fusiform  $g\text{-C}_3\text{N}_4$  modification  $\text{CaTiO}_3$  nano-heterojunction via simple hydrothermal co-deposition method. Compared with the unmodified samples, the as-prepared  $g\text{-C}_3\text{N}_4/\text{CaTiO}_3$  nano-heterojunction possesses an obvious enhancement of photocatalytic hydrogen production performance. Further, the recycle stability and mechanism of the heterojunction were studied.

## Experiment

The details of the experiments are shown in ESI. The samples modified by  $g\text{-C}_3\text{N}_4$  with the different molar ratios ( $g\text{-C}_3\text{N}_4/\text{CaTiO}_3$ ) of 1/13.5, 2/13.5, and 3/13.5 were labeled as Ca-C-1, Ca-C-2 and Ca-C-3, respectively.

## Result and discussion

The crystal structures and the phase components of the as-prepared  $g\text{-C}_3\text{N}_4/\text{CaTiO}_3$  are characterized by XRD. As illustrated in Fig. 1, the obvious diffraction peaks at the curve of pure  $\text{CaTiO}_3$  could be ascribed to the (101), (110), (111), (200), (021), (120), (210) (103), (202), (004), (024), (312) and (224) planes of the  $\text{CaTiO}_3$  (PDF#78-1013). With the  $g\text{-C}_3\text{N}_4$  being introduced, the new diffraction peak appears at  $27.7^\circ$ , increases with the ratio of the modification obviously, could be ascribed to the layer stacking of conjugated aromatic rings on  $g\text{-C}_3\text{N}_4$  (PDF#87-1526) [50]. It's interesting that the diffraction peaks of the  $\text{CaTiO}_3$  in composites are similar to the intrinsic  $\text{CaTiO}_3$ , which indicates the hydrothermal process would not

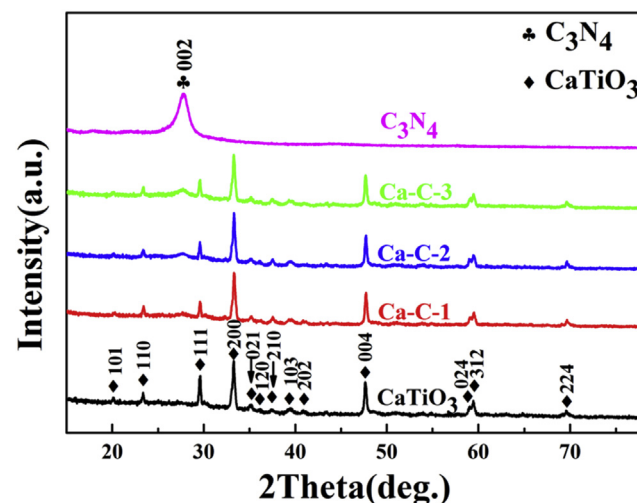


Fig. 1 – The XRD spectra of the fusiform  $\text{CaTiO}_3$  modified with different ratio of  $g\text{-C}_3\text{N}_4$ .

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