



Photocatalytic reduction of CO₂ to methane over HNb₃O₈ nanobelts

Xiukai Li^{a,b,c,*}, Huiqi Pan^{a,b,c}, Wei Li^{a,b,c}, Zongjin Zhuang^{a,b,c}

^a China–Australia Joint Research Center for Functional Molecular Materials, Jiangsu University, Zhenjiang 212013, PR China

^b Scientific Research Academy, Jiangsu University, Zhenjiang 212013, PR China

^c School of Chemistry and Chemical Engineering, Jiangsu University, Zhenjiang 212013, PR China

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ABSTRACT

KNb₃O₈ and HNb₃O₈ nanobelts were prepared by hydrothermal synthesis. The characteristics of samples were investigated by XRD, SEM, and UV–vis diffuse reflectance spectroscopy. The KNb₃O₈ and HNb₃O₈ nanobelts exhibited much higher activities for CO₂ photoreduction to methane than commercial TiO₂ (Degussa P25), and the KNb₃O₈ and HNb₃O₈ particles prepared by conventional solid state reaction. It was also found that either the HNb₃O₈ nanobelts or the HNb₃O₈ particles performed better than the corresponding KNb₃O₈ counterpart. It is proposed that the nanobelt-like morphology and the protonic acidity contribute to the higher photocatalytic activity of the HNb₃O₈ nanobelts.

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1. Introduction

Carbon dioxide (CO₂) is the primary green house gas that causes global warming. However, the atmospheric concentration of CO₂ is continuously increasing owing to human activities. For the consideration of sustainable development of human society, it is of great importance and interest to reduce the emission level of CO₂ as well as to convert CO₂ to useful substances. Among various approaches for the transformation of CO₂, the photocatalytic reduction of CO₂ to reusable hydrocarbons using water as the reductant is very attractive. It has been demonstrated that CO₂ could be photocatalytically reduced by water to methane, methanol, or ethanol at room temperature [1–4].

Up to date, the research on the photocatalytic reduction of CO₂ had been mostly focused on TiO₂-based materials [5–8]. It is urgent to explore other efficient photocatalysts for CO₂ photoreduction. Lamellar titanates and niobates (e.g., K₂Ti₄O₉, and HNb₃O₈) are one type of materials constructed by stacked thin slices built up from Ti–O or Nb–O polyhedron units. From the view point of photocatalysis, such layered configuration is favorable for the separation and transportation of photo-excited carriers (i.e., electrons and holes). Actually, some lamellar titanates and niobates are efficient photocatalysts for water splitting or organic compounds degradation

[9–12]. Previous research also revealed that lamellar solid acids usually show much higher photocatalytic activities than their corresponding salt phases [13–16].

In view of the unique photocatalytic properties of lamellar solid acids, it is very intriguing to investigate CO₂ photoreduction over this kind of materials. However, there has been no such report up to date. Moreover, lamellar solid acids have hygroscopic characters and could adsorb even the atmospheric water through hydrogen bonding at room temperature [17,18], this surely should exert certain positive impact on CO₂ photoreduction by water. In the present study, KNb₃O₈ and HNb₃O₈ nanobelts were therefore prepared by the template assisted hydrothermal synthesis, and were evaluated for CO₂ photoreduction. The influences of sample morphology and protonic acidity on the photocatalytic activity were investigated; the reaction mechanism was also proposed. It is envisaged that the present study might provide a feasible method to fabricate materials with high activity for CO₂ photoreduction and enable a more in depth understanding about the related photocatalytic reaction.

2. Experimental

2.1. Catalyst preparation

The KNb₃O₈ samples were prepared by the solid state reaction (SSR) and the hydrothermal (HT) synthesis, respectively. In the solid state reaction case, stoichiometric amounts of Nb₂O₅ and K₂CO₃ were well milled and then heated at 900 °C for 10 h; the obtained product was denoted as KNb₃O₈(SSR). In the case of the hydrothermal synthesis, Nb₂O₅ (0.5 g) powder was first dispersed

* Corresponding author at: Scientific Research Academy, Jiangsu University, No. 301 Xuefu Rd., Zhenjiang 212013, PR China. Tel.: +86 511 88797815; fax: +86 511 88797815.

E-mail address: li.xiukai@gmail.com (X. Li).