



Contents lists available at ScienceDirect

Materials Science in Semiconductor Processing

journal homepage: www.elsevier.com/locate/mssp

Photoreduction of carbon dioxide by graphene–titania and zeolite–titania composites under low-intensity irradiation

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ARTICLE INFO

Available online 25 October 2014

Keywords:

Graphene–titania
zeolite–titania
carbon dioxide reduction
Mechanism

ABSTRACT

As it is the most important of the greenhouse gases, the utilization and reduction of carbon dioxide have attracted a great attention. As compared to the technological demands for carbon capture and storage (CCS), carbon dioxide reduction is a safe and effective way to convert carbon dioxide to fuel. In this research, two different catalysts, graphene–titania and zeolite–titania, are used to achieve the carbon dioxide reduction. The characteristics of these materials are analyzed by Brunnauer–Emmett–Teller, X-ray diffraction, Fourier transform infrared spectroscopy, ultraviolet–visible, scanning electron microscope and transmission electron microscopy. Because of the different features of the catalysts, various products can be generated through different pathways. Formic acid and methanol are the final products when graphene is used as the catalyst, but only methanol can be generated when zeolite–titania is used as the catalyst. The reaction mechanisms and pathways are discussed.

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1. Introduction

The challenges of climate change have commanded considerable attention in recent years, and greenhouse gas (GHG) emissions are acknowledged to be the main cause of climate change. Carbon dioxide is the most important of the greenhouse gases as a result of the dependence of the world's economies on fossil fuels [1]. The concentration of carbon dioxide in the atmosphere rose from 200 to 300 ppm in the pre-industrial era to 370 ppm in 2001. More recently, the atmospheric level of carbon dioxide increased to 395 ppm in 2012 and 399.5 ppm in 2013 (Intergovernmental Panel on Climate Change, IPCC). The traditional method used to control the concentration of carbon dioxide in the environment is

carbon capture and storage (CCS), by which carbon dioxide is captured from a large point source and transported to a storage site. In the carbon capture and storage process, the carbon dioxide has always been injected into geological positions where there is the risk of the emissions escaping back into the atmosphere. Submarine storage would increase the possibility of ocean acidification. As compared with carbon dioxide storage, utilizing the abundant solar energy to convert carbon dioxide into fuels would not only generate clean energy, but it would also avoid the risk of secondary pollution.

For the light-harvesting reaction, titania is the most attractive material because of its low price, low toxicity and high stability. When this material is irradiated by light whose intensity matches or is larger than the band gap, the electron will be excited from the valance band to the conductive band. The photo-generated electron–hole will recombine in two ways (as shown in Fig. 1) in order to reduce the efficiency of the material [2].

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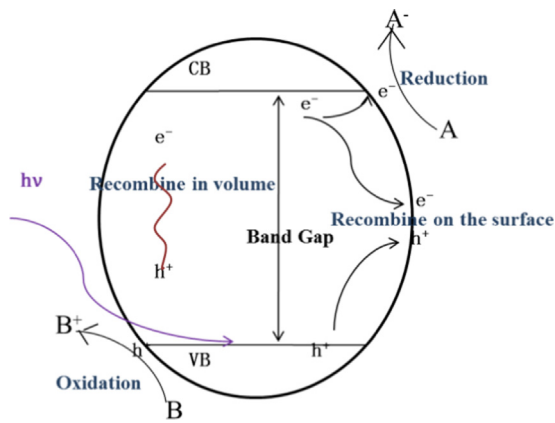


Fig. 1. The possible decay pathway of photo-generated electron and hole.

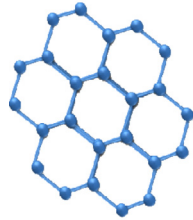
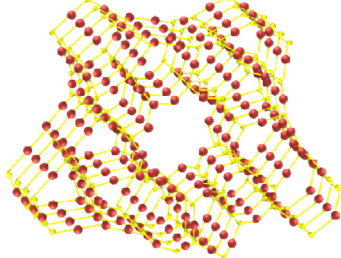
To achieve the high efficiency purpose, a lot of materials have been used and all the materials mentioned can successfully reduce the carbon dioxide to energy. Even all the materials can convert the carbon dioxide to fuels, but the efficiency of the reaction is totally different. The yield of fuels by carbon dioxide reduction reaction with different catalysts under various light sources was summarized in Table 1 [3–14].

To enhance the efficiency of the material, a second material has to be introduced to inhibit the recombination of the electron–hole pairs. In this research, two totally different materials with large surface areas (graphene–titania and zeolite–titania) were used as the titania substrate separately. Unlike the two-dimensional structure of graphene, zeolite has a three-dimensional structure and is nanoporous. For the two materials, the differences in the chemical and physical characteristics between graphene

Table 1
Summary of CO₂ reduction efficiency with different catalysts.

Catalysts	Reductant	Light source	Products	Yield	Ref.
Ti-oxide/Y-zeolite	H ₂ O	UV	CH ₄ CH ₃ OH	~40 μmol/g ~28 μmol/g	[3]
Ti-MCM-41 and Ti-MCM-48	H ₂ O	High pressure Hg light ($\lambda > 280$ nm)	CH ₄ CH ₃ OH	~8 μmol/g/h ~3 μmol/g/h	[4]
Cu/TiO ₂	H ₂ O	75-W high-pressure Hg lamp color filters ($\lambda > 290$ nm)	CH ₄ CH ₃ OH	~0.7 μmol/g ~8 μmol/g	[5]
TiO ₂	2-propanol	350 nm	CH ₄	~2 μmol	[6]
Cu/TiO ₂	H ₂ O	1 to 16 W/cm ² UV irradiation	CH ₃ OH	~0.45 μmol/g	[7]
Ag/TiO ₂	H ₂ O and methanol	150 W solar simulator	H ₂ CO CH ₄	~2300 μmol/g/h ~130 μmol/g/h ~25 μmol/g/h	[8]
Mesoporous In(OH) ₃	H ₂ O	300 W Xe arc lamp	CH ₄	~0.8 μmol/g/h	[9]
g-C ₃ N ₄ -N-TiO ₂	H ₂ O	300 W Xenon arc lamp	CO	~14.73 μmol	[10]
TiO ₂ -Mg	H ₂ O	550 W Xe-lamp	CH ₄ H ₂ CO	~1 μmol/g/h ~0.42 μmol/g/h ~0.15 μmol/g/h	[11]
Copper-decorated TiO ₂ nanorod	H ₂ O	8 W UVA lamps (365 nm)	CH ₄	~14 μmol/g	[12]
β-Ga ₂ O ₃	H ₂	200 W Hg-Xe lamp	CO	~4 μmol	[13]
Silica supported Cu/TiO ₂	H ₂ O	250–400 nm	CO CH ₄	~60 μmol/g/h ~40 μmol/g/h	[14]

Table 2
The character of graphene and zeolite.

	Graphene	ZSM-5
Thermal electron mobility	250,000 cm ² V ⁻¹ s ⁻¹	–
Charge carrier mobility	200,000 cm ² V ⁻¹ s ⁻¹	–
Surface area	Theoretical value of 2630 m ² g ⁻¹	300–600 m ² g ⁻¹
Structure		

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