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C. R. Chimie 9 (2006) 275–281



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Full paper / Mémoire

Efficient solar to chemical conversion by a new-type n-Si electrode with metal nano-contact and surface methylation

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Received 28 June 2004; received in revised form 10 May 2005; accepted 18 May 2005

Available online 08 September 2005

Abstract

We have succeeded in developing a new-type Si electrode with metal nano-contact and surface methylation, which shows excellent and stable photovoltaic characteristics in aqueous redox electrolytes. The electrode has achieved solar decomposition of hydrogen iodide into hydrogen and iodine under no external bias, with a high solar-to-chemical conversion efficiency $\phi_{\text{chem}}^{\text{s}}$ of 7.4%. The present success is of much interest in that it has opened a new way to realize high-efficiency and low-cost solar energy conversion. **To cite this article:** S. Takabayashi et al., *C. R. Chimie* 9 (2006).

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Résumé

Un nouveau type d'électrode de silicium de type n, comportant des nanocontacts métalliques et une surface alkylée, a été développé avec succès. L'électrode présente une bonne stabilité et d'excellentes propriétés photovoltaïques dans les électrolytes redox aqueux. Cette électrode permet, sous exposition solaire, de décomposer l'acide iodhydrique en hydrogène moléculaire et iode à potentiel ouvert, avec un rendement de conversion énergie solaire–produit chimique $\phi_{\text{chem}}^{\text{s}}$ de 7,4 %, ce qui est la valeur la plus élevée publiée, à l'exception des électrodes à base de semiconducteurs multicouches de très haute qualité et de coût élevé. Ce succès est important, car il ouvre une nouvelle voie pour abaisser le coût de la conversion de l'énergie solaire. **Pour citer cet article :** S. Takabayashi et al., *C. R. Chimie* 9 (2006).

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Keywords: Solar energy; Hydrogen; Silicon; Nano-particles; Surface modification

Mots clés : Énergie solaire ; Hydrogène ; Silicium ; Nanoparticules ; Modification de surface

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1. Introduction

The main target in recent solar-cell studies lies in lowering the fabrication cost without reducing the conversion efficiency. Much attention has thus been paid to thin-film solar cells [1,2], fabricated with inexpensive thin-film semiconductor materials such as amorphous silicon (Si), polycrystalline Si, and dye-sensitized TiO₂. However, this type of solar cells has faced a serious deadlock that the use of expensive transparent conductive oxide (TCO) such as indium tin oxide is inevitably necessary for efficient current collection. In addition, the TCO has no enough electrical conductivity and thus we are compelled to adopt an elaborate solar-cell structure of series connection of a large number of tiny solar cells to avoid an increase in the ohmic loss [2], which leads to a significant cost increase. Furthermore, this series connection induces another cost increase because high-quality fabrication of tiny solar cells is required, for in this structure damage of one tiny cell ruins all the performance of tiny cells connected in series with it.

Solar to chemical conversion by use of a semiconductor/solution junction has attracted considerable attention as another new technology for solar energy conversion since the pioneering report of water photolysis with n-TiO₂ in 1972 by Fujishima and Honda [3]. This method has great advantages in cost lowering over the thin-film solar cells, in that it does not need to use any expensive TCO and is easily adaptable to inexpensive thin-film semiconductor materials. The main difficulty in this method is that most of semiconductors with band-gaps suitable for solar energy conversion are chemically unstable in aqueous electrolytes [4,5]. Much effort has been done to overcome the difficulty, and several interesting solar to chemical conversion systems with considerably high efficiencies have been reported such as photodecomposition of hydrogen iodide (HI) with a Rh-loaded p-InP photocathode [6] or an n-WS₂ photoanode [7], and photodecomposition of HBr and HI with a combination of a Rh-loaded p-InP photocathode and an n-WSe₂ or n-MoSe₂ photoanode [8]. Photodecomposition of water or other energy-storing reactions are also successfully achieved with stacked multilayer semiconductor electrodes [9–14].

Silicon (Si) is one of the most important semiconductors for solar energy conversion in view of an appropriate band-gap of 1.1 eV, abundance in natural re-

sources, and non-toxicity, but easily oxidized and passivated in aqueous electrolytes. We reported fairly long ago [15–17] that single crystal n-Si electrodes loaded with metal nano-particles, immersed in a Br⁻/Br₂ redox electrolyte, generated very high open-circuit photovoltages (V_{oc}) of 0.62–0.64 V, considerably higher than those of conventional solid-state p–n junction Si solar cells of a similar simple structure. This finding provided a new way to utilize Si for solar to chemical conversion [18], but unfortunately the n-Si electrode showed no enough stability for long-term operation. Theoretically, it was expected [16,17] that the Si surface was stabilized by metal particles at metal-coated parts and passivating Si oxide layers at naked parts, but actually gradual Si oxidation occurred even beneath the metal particles, which led to the electrode degradation.

Recently, a number of studies have been reported on modification of Si surfaces with organic alkyl groups [19–26]. A large merit of surface alkylation is the improvement of Si stability against the surface oxidation in air [27] and in an aqueous redox electrolyte [28], without any increase in the surface carrier recombination rate. However, it is also reported [29,30] that the surface alkylation tends to retard interfacial electron transfer at the Si/redox electrolyte contacts. We reported in a previous paper [31] that this dilemma could be overcome by metal nano-dot coating because the metal nano-dots acted as an effective catalyst for interfacial electron transfer reactions.

The above finding is of great importance because it finally provides an effective way to utilize Si for solar to chemical conversion. In the previous work [31], we studied the behavior of the methylated and Pt nano-dotted n-Si electrode in 8.6 M HBr + 0.05 M Br₂ with a highly positive redox potential to get high V_{oc} 's. In the present work, we have studied the behavior of the methylated and Pt nano-dotted n-Si in 7.6 M HI (or 7.6 M HI + 0.05 M I₂) to examine a possibility of efficient solar to chemical conversion via photodecomposition of HI into H₂ and I₂ (or I₃⁻) with no external bias.

2. Experimental

Single crystal n-Si (111) wafers of the resistivity of 1 ~ 5 Ω cm and the thickness of 825 ± 25 μm, donated by New Win Go Co. Ltd., were used. The Si surfaces were cleaned by immersing in a boiling mixture of conc.

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