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Platinum-loaded mesoporous titania by single-step sol–gel process with surfactant template: photocatalytic activity for hydrogen evolution

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Abstract

The utilization of mesoporous TiO_2 photocatalyst with platinum loading was investigated towards the photocatalytic H_2 evolution. A surfactant-assisted templating sol–gel process was used to obtain the mesoporous TiO_2 with uniform and narrow pore size distribution. Various platinum contents (0.1–0.9 wt%) were supported on the synthesized mesoporous TiO_2 by single-step sol–gel (SSSG), incipient wetness impregnation (IWI), and photochemical deposition (PCD) methods. The photocatalysts were methodically characterized by XRD, N_2 adsorption–desorption analysis, and TEM. The influence of different Pt loading methods on the photocatalytic performance was examined. The SSSG method, in which Pt precursor was introduced into the completely hydrolyzed TiO_2 sol prepared with a mesopore-directing surfactant template, was revealed to be a helpful candidate, exhibiting the highest photocatalytic activity (optimum loading of 0.6 wt%) as well as the broadest photocatalytic activity window. This method also proposes a great prospect to the development of highly active photocatalysts. ***To cite this article: T. Sreethawong et al., C. R. Chimie 9 (2006)***.

Résumé

L'activité catalytique d'un photocatalyseur de TiO_2 mésoporeux chargé en platine vis-à-vis de l'évolution photocatalytique de H_2 a été étudiée. Un procédé sol/gel assisté par une matrice de surfactant a été utilisé pour fabriquer TiO_2 avec une distribution uniforme de pores étroits. Différentes quantités de platine (0.1–0.9% en masse) ont été déposées sur du TiO_2 mésoporeux synthétisé par la méthode sol/gel en une seule étape (SSSG) par imprégnation sèche et dépôt photochimique. Les photocatalyseurs ont été caractérisés par diffraction des rayons X, analyse de désorption/adsorption et microscopie électronique à transmission. La méthode SSSG, dans laquelle le précurseur de Pt est introduit dans le sol de TiO_2 complètement hydrolysé, préparé avec une matrice mésoporeuse de surfactant, conduit à un matériau qui présente l'activité photocatalytique la plus grande (chargement maximum de 0.6% en masse) et la fenêtre d'activité photocatalytique la plus large. Cette méthode est très prometteuse pour le développement des photocatalyseurs à haute activité. ***Pour citer cet article : T. Sreethawong et al., C. R. Chimie 9 (2006)***.

Keywords: Pt/TiO_2; Single-step sol–gel; Incipient wetness impregnation; Photochemical deposition; Photocatalytic hydrogen evolution

Mots-clés : Pt/TiO_2 ; Sol/gel en une seule étape ; Imprégnation sèche ; Dépôt photochimique ; Évolution photocatalytique de l'hydrogène

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1. Introduction

The mesoporous materials with different compositions, new pore systems, and novel properties have attracted considerable attention because of their remarkably large surface areas and narrow pore size distributions, which make them ideal candidates for catalysis [1]. Recently, much attention has been directed to the synthesis of nanostructured mesoporous oxides using surfactant templating method. It has been developed for the synthesis of materials with a narrow mesopore size distribution and controlled pore structure [2–4]. It is generally anticipated that the use of a high surface area mesoporous oxide support, rather than a commercial, low surface area support, for noble metals or transition metals has some beneficial effects on the catalytic performance [5]. The mesoporous support would give rise to well dispersed and stable metal particles on the surface upon thermal treatments and as a consequence would also show an improved catalytic efficiency.

The interest in platinum-loaded titania (Pt/TiO_2), has rapidly increased due to their high activity in a multitude of important reactions [6–9]. Among them, photocatalytic H_2 generation from water splitting has recently been attracting rapidly growing interest due to clean, storable, and renewable energy development, aiming to replace exhausting fossil fuels by use of solar energy ever since Fujishima and Honda's report on the photoelectrochemical water splitting by TiO_2 electrode [10]. The photocatalytic activity and especially the stability of platinum strongly depend on both the state and structure of the support and the specific interaction between platinum and support. The type of the support is of crucial importance to obtain highly dispersed platinum particles with good performance [11]. To our knowledge, it has been suggested a good possibility to apply the mesoporous TiO_2 for the first time as a support for platinum towards the photocatalytic reaction of H_2 evolution.

In our previous articles, the mesoporous TiO_2 photocatalyst synthesized via a surfactant-assisted templating sol–gel process was reported [12,13] and used as a support for nickel oxide loaded by single-step sol–gel method [14]. In this work, we present a considerably high photocatalytic performance over platinum supported on such the mesoporous TiO_2. The methods of platinum loading namely single-step sol–gel (SSSG), incipient wetness impregnation (IWI), and photochemical deposition (PCD) and their characterizations by XRD, N_2 adsorption–desorption analysis, and TEM were described. The photocatalytic H_2 evolution activity of the photocatalysts with different platinum contents was tested systematically.

2. Experimental

2.1. Materials

Tetraisopropyl orthotitanate (TIPT, Tokyo Chemical Industry Co., Ltd.), hydrogen hexachloroplatinate(IV) hydrate (Nacalai Tesque, Inc.), laurylamine hydrochloride (LAHC, Tokyo Chemical Industry Co., Ltd), acetylacetone (ACA, Nacalai Tesque, Inc.) and methanol (Nacalai Tesque, Inc.) were used for this study. All chemicals were analytical grade and used without further purification. LAHC was used as a surfactant template behaving as a mesopore-directing agent. ACA serving as a modifying agent was applied to moderate the hydrolysis and condensation processes of titanium precursor.

2.2. Photocatalyst synthesis

Single-step sol–gel (SSSG) made Pt-loaded mesoporous TiO_2 photocatalyst was synthesized via a combined sol–gel with surfactant-assisted templating mechanism in a LAHC/TIPT modified with ACA system. In typical synthesis, a specified amount of analytical grade ACA was first introduced into TIPT with the same mole. The mixed solution was then gently shaken until intimate mixing. Afterwards, 0.1 M LAHC aqueous solution of pH 4.2 was added into the ACA-modified TIPT solution, in which the molar ratio of TIPT to LAHC was adjusted to a value of 4. The mixture was kept continuously stirring at 40 °C for 10 h to obtain transparent yellow sol. To the aged TiO_2 sol solution, a necessary amount of hydrogen hexachloroplatinate(IV) hydrate in methanol for desired Pt loading of 0.1–0.9 wt% was incorporated, and the final mixture was further aged at 40 °C for 5 days to acquire homogeneous solution. Then, the gel was formed by placing the sol-containing solution into an oven kept at 80 °C for a week. Subsequently, the gel was dried overnight at 80 °C to eliminate the solvents. The dried sample