行政院原子能委員會
委託研究計畫研究報告

光化學反應器之概念規劃與輸送現象理論分析

Concept Design of Photochemical Reactor and Its Transport Phenomenon

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中文摘要

本計畫依合約書完成三項工作，(1)分析水分解光觸媒與使用之現有技術文獻報告，(2)完成新型低能隙光觸媒之測試評估，(3)完成並提升光纖導光化學反應器之光催化效率。在水分解光觸媒文獻報告共有 4 篇美國專利和兩篇期刊論文，涵蓋最新的技術整理。新型低能隙光觸媒，以 $InTaO_4$ 光觸媒在可見光下的水分解反應為主，傳統 $InTaO_4$ 主要是以 In_2O_3 和 Ta_2O_5 當作初始材料，使用固態反應法製備而成，此方法不易均勻，本研究是利用溶凝膠法和熱水解法製備 In、Ta 覆膜液，使 In、Ta 在溶劑中能均勻混合，高溫鍛燒得到效果更好的 $InTaO_4$ 光觸媒。光纖導光化學反應器內容納 216 根光纖之光反應器內，以 100W 高壓汞燈、波長 365nm、光反應器溫度 348K、流動式穩定狀態氣相系統中進行二氧化碳光催化還原反應。利用改良式熱水解法製備一系列 TiO_2、Cu/TiO_2、Ag/TiO_2、Pt/TiO_2 覆膜液，藉由浸漬覆膜法於直徑為 0.1mm 之光纖表面。由 SEM 之觀測，光纖表面的觸媒顆粒略呈圓形，粒徑約為 10-20nm，觸媒層厚約 26-33nm，並具有許多奈米級孔隙。經 XRD 繞射圖譜分析觸媒皆為 anatase 晶相。由 XPS 表面分析結果顯示，負載於二氧化鈦的 Cu、Ag 及 Pt 等金屬，分別以 Cu^{+}、Ag^{0} 及 Pt^{2+} 或 Pt^{4+} 存在於二氧化鈦表面。實驗結果發現，當光強度增強時，觸媒活性也隨之增加。銅、銀、鉑等過渡金屬的添加皆有效地提高甲醇的產率。綜上所述，具

體成果包括水分解光觸媒與使用之現有技術資料彙整，初步開發新型低能隙光觸媒，改良先前光纖導光化學反應器，提升 CO_2 的光催化效率。

Abstract

The project carried out and accomplished three missions under this contract, (1) survey and report the literatures of photo catalysis in water splitting technology, (2) evaluate the low bandgap photo catalysts, (3) improve the catalytic performance of optical-fiber photo reactor. Four US patents and two journal papers were reviewed in the photo catalysis of water splitting which cover the current technology in this field. $InTaO_4$ is one of the most active water-splitting catalyst under visible light irradiation. Traditional synthesis of $InTaO_4$ was prepared by solid fusion method. However, such method cannot uniformly synthesized the catalyst. Our study used sol-gel and hydrolysis technique to prepare In an Ta sols separately so that two sols can be mixed homogeneously. Improved InTaO4 catalyst thus obtained after calcined at high temperature. The optical-fiber photo reactor was consisted of 216 optical fibers. The photo reduction of CO_2 was performed in steady-state flow under 365 nm UV irradiation using 100W Hg lamp at 348K. The catalysts are TiO_2, Cu/TiO_2, Ag/TiO_2 and Pt/TiO_2 which were prepared by modified thermal hydrolysis method and were dip-coated on the optical fibers. From the SEM micrographs, the nano porous catalyst films were 26-33 nm thick and composed of spherical TiO_2 particles

with 10-20 nm in diameters. The crystalline phase of catalyst was anatase revealed by XRD. The chemical status of metal loaded on TiO2 were Cu^{+}, Ag^{0} and Pt^{4+} from the XPS measurement. The photo activity increased with increasing light intensity in the experiment. The methanol yields were enhanced by doping Cu, Ag and Pt compared with that in TiO_2. In summary, this project presented the current technology survey in water splitting by photo catalysts. The low bandgap photo catalysts were preliminarily studied. The photo reduction of CO2 was enhanced by improving the optical-fiber photo reactor.