行政院原子能委員會

委託研究計畫研究報告

水分解用奈米結構金屬氧化物光觸媒之製造方法研究

Preparation of Nano-Structural Metal Oxide Photo-Catalyst using for Water Splitting

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中文摘要

本研究利用兩微乳膠技術直接合成奈米$TiO_{2-x}N_x$、$Pt/TiO_{2-x}N_x$ 微粉，水相是以檸檬酸配位異丙氧基鈦，以控制烷氧鈦化合物的水解及聚縮合反應，在鹼性溶液中，生成逆微胞，再蒸餾除去乳化系統中的油相(正己烷)製得乳凝膠，並利用不同煅燒溫度來控制$TiO_{2-x}N_x$結晶型態及結晶度。藉由FTIR、XRD、DTA/TGA、TEM、ED、XPS等分析儀器,探討前導物轉變成$TiO_{2-x}N_x$ 粉末的化學反應機構和微結構與粉末性質。觀察其$TiO_{2-x}N_x$ 粉末在不同製程參數下的粉末性質差異。結果顯示：當檸檬酸/異丙氧基鈦莫耳比3：1、氨水濃度為22%、反應溫度為50℃，煅燒溫度為500℃時為銳鈦礦結構，顆粒大小約為4~6 nm，由XPS證實粉末中含Ti、O、N等元素，顯示出此粉末為$TiO_{2-x}N_x$ 微粉。經由UV-Vis計算出$TiO_{2-x}N_x$ 的能隙值為2.78 eV。

由兩微乳膠製程技術中之最佳條件煅燒得 0.6 wt% $Pt/TiO_{2-x}N_x$ 微粉來進行光分解水產氫之研究。研究顯示：當 25 毫克 $Pt/TiO_{2-x}N_x$ 微粉懸浮在甲醇水溶液中時，在鹵素燈照光下，可得到最高的產氫速率，為 2 小時產生 580 μmol。

關鍵字：兩微乳膠技術、逆微胞、乳凝膠、微粉。

Keywords: nanometric-sized, nitrogen-doped titanium oxide,
two emulsion technology, .

Abstract

Nanometric-sized nitrogen-doped titanium oxide ($TiO_{2-x}N_x$) powders were synthesized through two emulsions method. In which two solutions of reverse emulsion, one containing Ti^{4+} chelated with citric acid aqueous droplets and the other aqueous ammonia droplets, with the same water/oil (w/o) ratio are mixed together to form a slurry of titania precursor. Then the precursors are recovered and calcined to form $TiO_{2-x}N_x$ at various temperatures.

The precursors were characterized by FTIR, XRD, DTA/TGA, TEM, ED and XPS, and those mechanisms for the evolution of crystalline powders in two emulsions method was proposed and discussed in the context of the microstructure. It reveals that the preparatory conditions were the molar ratio of citric acid/titanium isopropoxide = 3, ammonia concentration = 22 %, reaction temperature = 50 ^{o}C, calcination temperature = 500 ^{o}C, a nanometric-sized $TiO_{2-x}N_x$ with average particle size at 4-6 nm was synthesized. Furthermore, the obtained powder was also examined by XPS, exhibits the existence of Ti, O and N, in evidence of $TiO_{2-x}N_x$ produced. The powder demonstrates a binding energy at 2.78 eV measured by UV-visible spectrum.

Finally, photocatalytic experiments for producing hydrogen was examined in methanol solution by utilizing 25 mg nanometric-sized Pt (6 wt%) /$TiO_{2-x}N_x$ powder, and 580 μmol hydrogen was produced in 2 h remarkably.