## 行政院原子能委員會 委託研究計畫研究報告

## (計畫名稱【廢水處理新型光觸媒之研究】)

(計畫名稱【Novel Photocatalysts for Wastewater Treatment 】)

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## 中文摘要

本研究計畫透過使用三區塊共聚物 P123 此種中性界面活性劑作為有機模版合成奈米尺寸二氧化鈦中孔分子篩。由於界面活性劑可透過靜電、氫鍵、共價鍵和凡得瓦爾力作用將矽酸鹽單體組合形成多樣化的中孔洞材料。本研究計畫進一步的將此特性延伸應用並在溫和的反應條件下來合成奈米尺寸二氧化鈦中孔分子篩,透過在不同溫度下鍛燒(400 與 450°C)可得到同時具有銳鈦礦與晶紅石兩相共存且比例各異的產物。本計畫所合成出之奈米尺寸二氧化鈦中孔分子篩透過氮氣等溫吸脫附實驗得知在不同鍛燒溫度下其表面積約在 143 與71m²/g之間,並由 SEM 與 TEM 證明其顆粒大小在 40-80nm 且具有孔洞直徑約在 5 至 15nm 左右之蟲狀中孔洞。FT-IR 則提供了在不同溫度鍛燒下奈米尺寸二氧化鈦中孔分子篩其 titanol(Ti-OH)官能基縮合的證據,而從紫外光/可見光光譜(UV/Vis spectrameter)與高角度 X 光粉末繞射(wide angle XRD)則可觀察到奈米尺寸二氧化鈦中孔分子篩的晶相組成、兩晶相間的比率以及隨鍛燒溫度提高而產生之相變化 (phase transition)。

相較於奈米尺寸二氧化鈦中孔分子篩以表面修飾方式所合成之矽酸鈦中孔分子篩(Ti-SBA15)擁有相當高之表面積(>500m²/g),然而其光催化活性卻是相當的低落,可見表面積並非主宰光催化活性之主要因素。本計畫中,利用亞甲基藍(Methylene blue, MB)裝置於石英管中以UVUV光照射所產生的降解脫色反應(decolorization)來鑑定所合成之產物其光催化活性,而溶液的酸鹼值、通入不同性質之氣體(空氣、純氮)、不同光波長的選擇(306nm、365nm)與不同的亞甲基藍、光觸媒濃度對光催化效果的影響亦將一併探討。透過實驗證明,奈米尺寸二氧化鈦中孔分子篩由於其具有銳鈦礦與晶紅石兩相共存且表

面上擁有豐富的 Ti-OH 官能基,故不論在氮氣或是空氣的環境下對 MB 的光催化轉化率均較商業上常用的光觸媒 P-25 來得佳。本計畫 得到之結論,在 pH 值約為 3 之環境下使用本計畫內合成之奈米尺寸 二氧化鈦中孔分子篩,結合紫外光照射預期可有效的應用在廢水處理上。

## **Abstract**

Surfactants have been shown to organize silica into a variety of mesoporous forms, through the mediation of electrostatic, hydrogen -bonding, covalent and van der waals interactions. This approach to mesostructured materials has been extended, to titanium dioxide. Nanoscaled mesoporous titania with mixed phases of anatase and rutile has been synthesized using P123 block copolymer surfactant under mild condition and further calcinations at 400 and 450°C. The resulting materials were characterized by XRD, nitrogen adsorption, TEM, SEM analysis, UV-vis spectroscopy and FT-IR spectroscopy.

The surface areas for most samples are between 143 and 71 m<sup>2</sup>/g. The nanoporous titania has nano-size between 40-80 nm and worm-like pore with wide pore size distribution between 5 and 15 nm. The calcination of resulting materials at different temperature will promote titanol group condensation, crystallization with phase transformations and enlarge pore at beginning. A reasonable mechanism for this phenomenon is proposed as well.

Titanosilicate mesoporous SBA-15 materials owned pretty high surface area ( $>500 \text{ m}^2/\text{g}$ ) but they show lower photocatalytical activity than that of nanoscaled mesoporous titinia.

Parameters such as pH, gaseous environment, light wavelength and concentration of catalyst and MB(methylene blue) were studied using the photocatalytic decolorization of MB in a photoreactor. All experiments were performed in a quartz tube with UV light, a wavelength at 365 nm and 306 nm, in combination with a mesoporous TiO<sub>2</sub> powder as the catalyst. Resulting material shows slightly higher conversion of MB than the P25 effective commercial photocatalyst under saturated aqueous solution with air or nitrogen. The results can be illustrated due to mixed phases and abundant titanol group of catalyst.

The optimal tentative conditions were developed in a solution at pH of 3.0. These results suggest that nano-scaled nanoporous  $TiO_2/UV$  photocatalysis may be envisaged as a method for treatment of diluted waste water.