UNITED Scientific Group
A non-profit organization

IV International Conference on

Catalysis and Chemical Engineering

February 24-26, 2020

Venue

Four Points by Sheraton
Los Angeles International Airport
9750 Airport Boulevard
Los Angeles, CA 90045, United States

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Photoredox Catalysts for Direct Phenol Production from Benzene

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Abstract

Photocatalytic oxygenation of benzene occurs in an oxygen-saturated acetonitrile solution containing 3-cyano-1-methylquinolinium ion ($CNQuH^+$), benzene and water under UV light irradiation to yield phenol and hydrogen peroxide selectively. The benzene radical cation, which is formed by photoinduced one-electron oxidation of benzene with $CNQuH^+$, reacts with H_2O to yield phenol.

Photocatalytic oxygenation of benzene with oxygen occurs under photoirradiation of an acetonitrile solution of 3-cyano-1-methylquinolinium perchlorate ($QuCN^+ClO_4^-$; l_{max} = 330 nm, 5.0 mM) in an oxygen-saturated acetonitrile (MeCN) containing benzene (30 mM) and H_2O (3.0 M) with a xenon lamp (500 W) attached with a color-cut glass filter (l = 290-600 nm). Phenol and hydrogen peroxide were selectively produced after photoirradiation for 5 h, which were identified by ^1H NMR and iodometry. The selectivity of formation of phenol was 98% with 16% quantum yield after 1 h irradiation, and then 51% of phenol yield after 5 h irradiation. The photocatalytic turnover number (TON) was 7.5. This is the first example of photocatalytic oxygenation of benzene to phenol in a homogeneous system. A preparative gram-scale photocatalytic reaction with benzene (2.3 g, 29 mmol) and $QuCN^+$ (210 mg, 0.8 mmol) in MeCN (200 mL) for 48 h was also examined to afford phenol (1.1 g, 12 mmol) as 41% yield.

Biography

Dr. Kei Ohkubo, Chemistry Doctor (Ph.D. Engineering), now is a Professor of Chemistry of Institute for Advanced Co-Creation Studies and Institute for Open and Transdisciplinary Research Initiatives, Osaka University, Japan. He earned his Ph.D. degree from Graduate School of Engineering, Osaka University in 2001. He was working as a JSPS fellow and a JST research fellow at Osaka University (2001-2005), a designated associate professor in Osaka University (2005-2014) and a specially appointed professor at Osaka University (2014-2017). He has been a full professor at Osaka University since 2017.

Hydrogen Evolution by Photocatalytic Splitting of Water Using $SrTiO_3$/PAN Based Fibers

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Abstract

When developing new sources of energy, hydrogen is one of the most attractive fuels for the 21st century. Hydrogen has significant potential as an alternative fuel, taking into account that it can be produced from such raw materials as water.

Investigation of the parameters of the process of electroforming nano-sized fibers that can be used for photocatalytic coatings consisted in their refinement and optimization in order to obtain fibers of the required sizes. In particular, the following parameters were tested: rate of precursor feed, distance between the needle and the collector, value of high voltage, position of collector. Polyacrylonitrile (PAN) was chosen as a standard polymer for investigation of parameters of the electroforming process of nanofibers, since this polymer is used to produce durable thermally stable fibers. PAN was dissolved in ethanol at 50 °C with stirring in a magnetic stirrer for 20 minutes.

A component material for the production of photocatalytic fibers is strontium titanate. Strontium titanate was obtained by chemical precipitation from a solution of titanium oxide and strontium nitrate, followed by annealing at a temperature of 1100 °C for 1 hour. The precipitate from a solution of titanium oxide and strontium nitrate after calcination in air for 1 hour at a temperature of 1100 °C contains 97% strontium titanate.

Nano-sized fibers based on strontium titanate/PAN were obtained using electrospinning method. The high voltage applied to the needle and the collector was 1.5 kV per centimeter. The rate of the syringe pump was 1.5 ml/hour. The obtained nano-sized fibers were calcined at 700 °C for 1 hour to improve their photocatalytic characteristics.

The study of the physicochemical properties of obtained fibers based on strontium titanate/PAN showed that obtained films are presented by network of fibers with particles of strontium titanate of a size of up to 100 nm, uniformly distributed over their surface. The presence of particles of strontium titanate on the surface of the fibers proved by X-ray phase analysis. It has been established that the size of nanofibers based on strontium titanate/PAN is 200 nm before calcination, after calcination it decreases to 80-100 nm, which in turn improves their photocatalytic activity. The results of thermogravimetric analysis showed that the optimum temperature for calcination of fibers based on strontium titanate/PAN is 700 °C.

It was found that films based on nano-sized strontium titanate/PAN can be used as photocatalysts for the production of hydrogen by splitting "water - organic alcohol" mixture with hydrogen evolution rate 305 μmol h^{-1} g^{-1}.

Biography

Gulmira Sh Yar-Mukhamedova is a Full Professor, Doctor of Science in Physics of Condensed Matter with over 25 years in the field. She is an academic with a research interests in Nanotechnology & Material Science. Gulmira graduate from Donetsk National University (Ukrain) with a Bachelor of Physics. She enrolled in graduate school in University, successfully defended her thesis in three years. After that she continued her research work in Kazakhstan and received a Doctor of Science degree in 2001. Now more than 20 of her inventions are introduced into production.

Synthesis and Photocatalytic Performance of Core/Shell Structure Rutile@Anatase TiO2 Nanofibers

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Abstract

TiO_2-based photocatalyst has raised a lot of interests due to the contribution to renewable energy production and organic pollutant decomposition. TiO_2 with mixed phases shows the higher activity compared to single phase TiO_2. The different potential between two phases acted as the necessary driving force for charged carrier. For this work, two types of core-shell structure TiO_2 nanofibers (noted as core@shell TiO_2 NFs) were synthesized by sequential hydrothermal, calcination, and impregnation processes. Rutile TiO_2 nanofibers (R TiO_2 NFs) core with anatase TiO_2 nanoparticles (A TiO_2 NPs) shell is denoted as R@A TiO_2 NFs, and the reverse structure is denoted as A@R TiO_2 NFs. The photodegradation of organic dyes and Kelvin probe force microscopy (KPFM) analysis were utilized to peer into the mechanism of the photoexcited carrier transport. From the photodegradation test, the A@R TiO_2 NFs has the highest activity both under UV-B and UV-A irradiation, being nearly 3-fold higher as compared to AEROXIDE® TiO_2 P25. The results in conjunction with KPFM measurements indicated that in the heterostructure, electron-hole pairs are efficiently separated and the excited electrons stay in the anatase phase, and holes are injected to the rutile phase. Our study not only explains the role of anatase-rutile junctions in photocarrier separation, but also projects the development of other efficient photocatalytic heterostructures for green energy production and conversion.

Biography

Prof. Ming-Chung Wu received his Ph.D. degrees of materials science and engineering from National Taiwan University in 2008. From 2009 to 2010, he was postdoctoral researcher at University of Oulu, Finland. In 2012, he joined Department of Chemical and Materials Engineering at Chang Gung University, Taiwan. Prof. Wu's group develops innovative nanomaterials for various applications including photocatalysis, VOCs sensor, and sustainable energy. Until now, he has published ~80 SCI journal papers in a number of leading materials journals including ACS Nano, JMCA, Nanoscale, etc. His publications were cited over 1400 times with an H index of 23.

Photoassisted Degradation of Rhodamine B and Caffeine by Synthesis of ZnO/Hydroxyapatite Nanoparticles

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