

Biomimetic Solar Photocatalytic Reactor for Selective Oxidation of Aromatic Alcohols with Enhanced Solar-Energy Utilization

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Low utilization of solar energy remains a challenge that limits photocatalytic efficiency. To address this issue, this work proposes a bionic solar photocatalytic reactor (BSPR) for efficient selective oxidation of aromatic alcohols. A biomimetic phototropic hydrogel is prepared by coupling chlorine-doped polypyrrole (Cl-PPy) and poly(*N*-isopropyl acrylamide) (PNIPAm) to maximize light-harvesting efficiency automatically, allowing the BSPR to maintain high catalytic levels throughout the day. Molecular dynamics simulations are used to unveil the understanding of the fast photoresponsive behavior of Cl-PPy/PNIPAm from a molecular level, while COMSOL simulations are conducted to follow the macroscopically phototropic mechanism of BSPR. Attributing to the existence of PdS/S vacancies riched ZnIn₂S₄ nanocomposite in the top flower-shaped hydrogel, the BSPR displays a special function for efficiently photocatalytic oxidation of aromatic alcohols under solar illumination (yield of 4-methoxybenzaldehyde: 479.5 $\mu\text{mol g}^{-1} \text{h}^{-1}$; selectivity: 68.8%). Two possible reaction pathways are identified as follows: photogenerated holes can attract aromatic alcohols directly and generate aromatic aldehydes; photoexcited electrons oxidize O₂ to $\cdot\text{O}_2^-$ can also react with the adsorbed aromatic alcohol. This study presents a promising paradigm that explores opportunities for enhanced utilization of light energy, offering a novel approach to maximize its efficiency in practical applications.

1. Introduction

The self-adaption abilities of biological systems in nature (e.g., mimosa pedica, sunflower, flytrap, etc.) are promoting the

emergence of various smart materials,^[1–4] that can change their structures, properties, and functions autonomously to respond to the innocuous stimuli (e.g., heat, pH, light, electricity, etc.) in their surrounding environments. In particular, light-responsive smart materials allow flexible manipulations at remote distances,^[5–7] which are mainly driven by the intelligent composites of photothermal absorber and temperature-sensitive material for efficient photothermal conversion and thermally induced phase transition, respectively.^[8–9] For the fabrication of these intelligent composites, the main task is to find suitable temperature-sensitive materials that can satisfy the requirements of diverse temperature ranges. Up to now, the most commonly used temperature-responsive materials include poly(*N*-isopropyl acrylamide) (PNIPAm), 2(dimethylamino)ethyl methacrylate, 4-(3-Acryloyloxypropyloxy)-benzoicure 2methyl-1,4-phenylester), and some other composites.^[10–12] Among them, PNIPAm-based hydrogel can undergo a heat-induced conformational transition from a water-soluble coiled structure to a water-insoluble globular structure above its lower critical solubility temperature (LCST, $\approx 32^\circ\text{C}$), resulting in the massive loss of confined water in the network of PNIPAm-based hydrogel with its notable volume reduction and

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