

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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Table of Contents Solar Power Applications Alcohols

1. Understanding the eBook Solar Power Applications Alcohols
 - The Rise of Digital Reading Solar Power Applications Alcohols
 - Advantages of eBooks Over Traditional Books
2. Identifying Solar Power Applications Alcohols
 - Exploring Different Genres
 - Considering Fiction vs. Non-Fiction
 - Determining Your Reading Goals
3. Choosing the Right eBook Platform
 - Popular eBook Platforms
 - Features to Look for in an Solar Power Applications Alcohols
 - User-Friendly Interface
4. Exploring eBook Recommendations from Solar Power Applications Alcohols
 - Personalized Recommendations
 - Solar Power Applications Alcohols User Reviews and Ratings
 - Solar Power Applications Alcohols and Bestseller Lists
5. Accessing Solar Power Applications Alcohols Free and Paid eBooks
 - Solar Power Applications Alcohols Public Domain eBooks
 - Solar Power Applications Alcohols eBook Subscription Services
 - Solar Power Applications Alcohols Budget-Friendly Options

6. Navigating Solar Power Applications Alcohols eBook Formats
 - ePub, PDF, MOBI, and More
 - Solar Power Applications Alcohols Compatibility with Devices
 - Solar Power Applications Alcohols Enhanced eBook Features
7. Enhancing Your Reading Experience
 - Adjustable Fonts and Text Sizes of Solar Power Applications Alcohols
 - Highlighting and Note-Taking Solar Power Applications Alcohols
 - Interactive Elements Solar Power Applications Alcohols
8. Staying Engaged with Solar Power Applications Alcohols
 - Joining Online Reading Communities
 - Participating in Virtual Book Clubs
 - Following Authors and Publishers Solar Power Applications Alcohols
9. Balancing eBooks and Physical Books Solar Power Applications Alcohols
 - Benefits of a Digital Library
 - Creating a Diverse Reading Collection Solar Power Applications Alcohols
10. Overcoming Reading Challenges
 - Dealing with Digital Eye Strain
 - Minimizing Distractions
 - Managing Screen Time
11. Cultivating a Reading Routine Solar Power Applications Alcohols
 - Setting Reading Goals Solar Power Applications Alcohols
 - Carving Out Dedicated Reading Time
12. Sourcing Reliable Information of Solar Power Applications Alcohols
 - Fact-Checking eBook Content of Solar Power Applications Alcohols
 - Distinguishing Credible Sources
13. Promoting Lifelong Learning
 - Utilizing eBooks for Skill Development
 - Exploring Educational eBooks
14. Embracing eBook Trends
 - Integration of Multimedia Elements

- Interactive and Gamified eBooks

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