

# 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 ZnIn<sub>2</sub>S<sub>4</sub> 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 O<sub>2</sub> 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,<sup>[1–4]</sup> 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,<sup>[5–7]</sup> 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.<sup>[8–9]</sup> 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.<sup>[10–12]</sup> 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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The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adfm.202311214>

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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

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- Solar Power Applications Alcohols eBook Subscription Services
- Solar Power Applications Alcohols Budget-Friendly Options
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  - 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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