

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

Jingjing Qin, Jiahao Li, Kaibin Chu, Guozheng Yang, Leiqian Zhang, Xuemeng Xia, Pengyang Xuan, Xin Chen, Bo Weng, Haowei Huang, Yujie Chen, Wei Fan, YinBo Zhu, HengAn Wu, Feili Lai,\* and Tianxi Liu\*

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)-benzocure 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

J. Qin, K. Chu, L. Zhang, X. Xia, P. Xuan, X. Chen, W. Fan, T. Liu  
The Key Laboratory of Synthetic and Biological Colloids,  
Ministry of Education  
School of Chemical and Material Engineering  
International Joint Research Laboratory for Nano-Energy Composites  
Jiangnan University  
Wuxi 214122, P. R. China  
E-mail: txliu@jiangnan.edu.cn

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University of Science and Technology of China  
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K. Chu, F. Lai  
Department of Chemistry  
KU Leuven  
Celestijnenlaan 200F, Leuven 3001, Belgium  
E-mail: feililai@kuleuven.edu.cn

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