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Battery-like semiconductors enable dark photocatalysis

Chunhua Wang[1], Amir Varamesh[1], Yang Ding[2,*], Jinguang Hu[1,*]

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Driving sustainable chemical transformations using solar energy remains a central challenge in modern chemistry, particularly in achieving temporal decoupling between energy capture and chemical conversion. Nature provides an elegant solution to this challenge through photosynthesis. In this process, the Calvin cycle produces carbohydrates not through direct illumination but by consuming energy carriers such as nicotinamide adenine dinucleotide phosphate (NADPH) and adenosine 5'-triphosphate (ATP) generated during the light-dependent reactions[1]. Inspired by this concept, writing in *Nature Chemistry*, Peidong Yang and colleagues at the University of California, Berkeley recently reported a photochargeable semiconductor capable of storing photogenerated electrons and releasing them on demand[2]. This capability allows chemical reactions to proceed even in the absence of continuous light irradiation, enabling highly efficient and scalable dehydrogenative coupling of amines into diamines with concurrent hydrogen evolution.

An artificial system that mimics segregated energy utilization was developed using zinc indium sulfide nanocrystals (ZIS NCs) as a model photochargeable material. These nanocrystals are capable of storing photogenerated charges in long-lived trap states [Figure 1A][2], thereby decoupling light absorption from catalytic turnover. Functioning similarly to solar batteries, they capture electrons generated under illumination and retain them beyond the irradiation period. As a result, reactive carriers are preserved over extended timescales and recombination losses are suppressed, enabling transformations that are difficult to achieve with conventional photocatalysts to be feasible[3]. Notably, diamines are important building blocks for polymers such as polyamides, epoxy resins and polyurethanes. Traditional synthetic methods for diamines often involve harsh conditions, toxic reagents and generate substantial waste, raising environmental concerns[4]. In contrast, photocatalytic dehydrogenative coupling using ZIS provides an efficient route in which two amine molecules are coupled[2,3], releasing hydrogen gas as the sole byproduct where hydrogen serves as a clean energy carrier, offering a more sustainable alternative.

[1]Department of Chemical and Petroleum Engineering, University of Calgary, Calgary, AB T2N 1N4, Canada.
[2]College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, Zhejiang, China.

***Correspondence to:** Prof. Jinguang Hu, Department of Chemical and Petroleum Engineering, University of Calgary, Calgary, AB T2N 1N4, Canada. E-mail: jinguang.hu@ucalgary.ca; Prof. Yang Ding, College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, Zhejiang, China. E-mail: dingyang@hdu.edu.cn

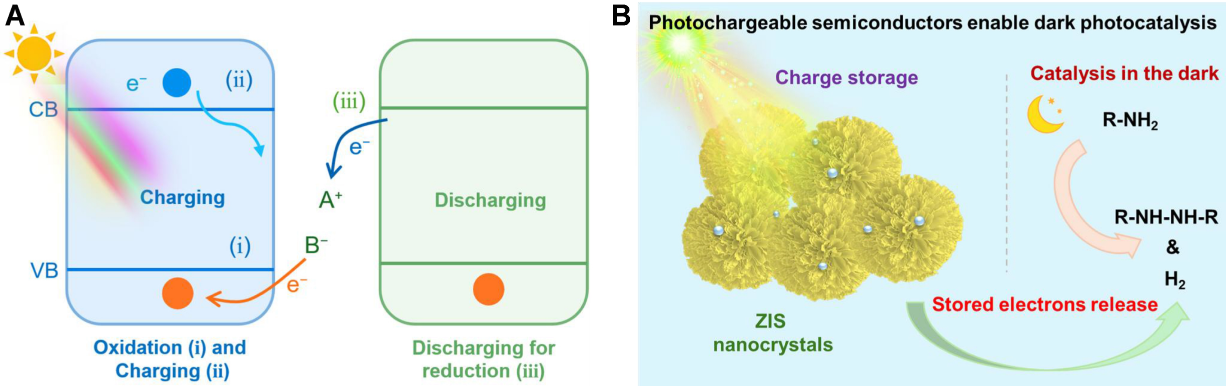

Figure 1. (A) Schematic illustration of photochargeable semiconductors enabling temporally decoupled photocatalysis; (B) Photocatalytic dehydrogenative coupling of amines over ZIS-based photochargeable catalysts. CB: Conduction band; VB: valence band; ZIS: zinc indium sulfide.

Incorporation of nickel as a hydrogen evolution cocatalyst, deposited *in situ* on the ZIS NCs via photoreduction of Ni^{2+}, enhances catalytic performance. Note that while ZIS NCs serve as photochargeable electron reservoirs, the nickel cocatalyst provides catalytic sites for proton reduction, enabling the stored electrons to be efficiently utilized for hydrogen evolution during the dark phase [Figure 1B]. Diamine production rates exceed 120 $mmol \cdot g^{-1} \cdot h^{-1}$, with selectivity surpassing 95% and an apparent quantum efficiency (AQE) of 39.4% under ambient conditions. Notably, the production rate substantially surpasses previously reported semiconductor photocatalysts in amine dehydrogenative coupling reactions[2], underscoring the remarkable efficiency enabled by the photochargeable ZIS NCs platform. Interestingly, hydrogen evolution persists for over 15 min after light irradiation is stopped [Figure 1B]. The presence of residual photogenerated electrons in the ZIS nanocrystals was further confirmed by introducing ferricyanide $[Fe(CN)_6]^{3-}$ as an electron scavenger after light-off. The system also shows excellent scalability, maintaining high efficiency on the tens-of-gram scales, along with robust recyclability over multiple cycles.

The exceptional photocatalytic performance originates from the unique photocharging behavior of the ZIS NCs. UV-vis spectroscopy reveals that illumination induces electron accumulation, leading to excitonic bleaching associated with conduction band filling, which is reversible upon oxygen exposure[5]. Quantitative analysis suggests that the stored electron density exceeds the conduction band capacity, indicating additional charge storage in deep trap states. Upon illumination, photogenerated holes oxidize surface-bound amines, while electrons are stabilized in sulfur-vacancy-associated trap states. Electron paramagnetic resonance and operando X-ray absorption spectroscopy confirm that these vacancies act as long-lived electron reservoirs, enabling charge persistence on the timescale of tens of minutes.

Such behavior fundamentally alters photocatalytic dynamics. Oxidation and reduction occur sequentially: oxidation takes place at the catalyst surface under illumination, whereas reduction proceeds after the light is turned off as trapped electrons are gradually released. The reaction proceeds via α-amino carbon-centered radical intermediates[6]. Importantly, the highly reducing environment created by trapped electrons suppresses overoxidation to imines by stabilizing radical intermediates, thereby ensuring high selectivity toward diamines. Hydrogen evolution is simultaneously facilitated by the nickel cocatalyst, highlighting efficient charge utilization between the semiconductor and cocatalyst.

Most importantly, the ZIS NCs photochargeable system exhibits broad substrate compatibility. A wide range of aryl amines undergo efficient coupling to yield diamines in high yield and selectivity. Amino acid esters are also readily converted, while secondary and tertiary amines, as well as dehydrogenative polymerization reactions, are also efficiently converted under similar conditions. Such versatility highlights the general applicability of the photochargeable concept in both small-molecule synthesis and polymer construction under mild conditions.

In summary, this work highlights the importance of storing and releasing photogenerated carriers in a controlled manner. Integrating energy storage functionality into photocatalysts bridges solar energy harvesting with chemical synthesis, allowing reactions to proceed independently of continuous illumination. Therefore, photochargeable semiconductors offer a promising strategy to overcome key limitations in conventional photocatalysis and open new avenues for sustainable chemical production.

Looking ahead, such systems provide a versatile platform for solar-driven chemical transformations ranging from organic synthesis to solar fuel production. Nevertheless, several challenges remain, including extending light absorption into the visible region, improving performance for non-aromatic substrates and integrating such systems into continuous-flow or industrial reactors. Despite these limitations, we still remain optimistic that the ability to separate light and chemistry opens new avenues for designing next-generation photocatalytic systems.

DECLARATIONS

Authors' contributions

Drafted the manuscript: Wang, C.
Revised the manuscript: Varamesh, A.; Ding, Y.; Hu J.

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Hu, J. is a Section Editor of the journal *Chemical Synthesis*. Hu, J. was not involved in any steps of the editorial processing, notably including reviewers' selection, manuscript handling, or decision-making. The other authors declare that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable

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Chunhua Wang

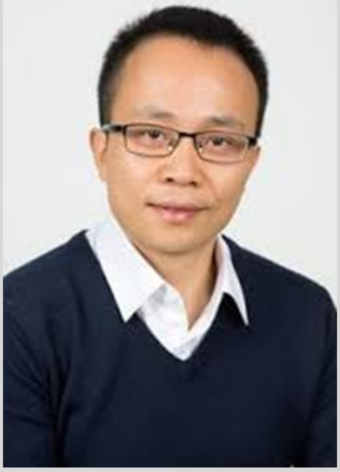
Dr. Chunhua Wang received his Ph.D. in Chemistry from KU Leuven (Belgium), in 2022 studying sustainable chemical conversion under the supervision of Prof. Johan Hofkens. After postdoctoral stays at the City University of Hong Kong and at the University of Michigan, Ann Arbor (United States), focusing on carbon neutrality and clean fuel generation, he joined Prof. Hu's group at the University of Calgary as a research fellow, continuing his research on solar-driven chemical conversion and sustainable energy materials.

Amir Varamesh

Dr. Amir Varamesh obtained his Ph.D. from the University of Calgary under the supervision of Dr. Jinguang Hu. His expertise spans advanced carbon materials for energy storage and bio-based foams and aerogels for water treatment, thermal insulation and environmental applications. He is currently the CEO and Co-Founder of NovaCarbon Technologies Inc., a University of Calgary spin-off company focused on commercializing advanced carbon materials derived from bitumen, biomass and plastic waste for next-generation energy storage applications.

Yang Ding

Dr. Yang Ding is currently an Associate Professor in the College of Materials and Environmental Engineering, Hangzhou Dianzi University, China. He received his Ph.D. degree from the Laboratory of Inorganic Materials Chemistry (CMI), University of Namur, Belgium, in 2022 under the supervision of Prof. Bao-Lian Su. His research interests include but are not limited to photocatalysis, porous material design, environmental pollutant degradation, and sustainable energy conversion.



Jinguang Hu

Dr. Jinguang Hu is currently an Associate Professor at the University of Calgary and a Member of the Royal Society of Canada's College of New Scholars, Artists and Scientists. He holds the Schulich Research Chair in Sustainable Energy and Materials. His research focuses on developing transformative technologies that convert biomass and renewable carbon resources into sustainable fuels, chemicals, and advanced materials. By integrating catalysis, biotechnology, photochemistry and materials engineering, his group aims to establish energy-efficient pathways for transforming agricultural residues, forestry by-products, and other renewable resources into high-value products that support a circular and low-carbon economy.