沈建华

Date: Sep 16, 2025
To: "Jianhua Shen" jianhuashen@ecust.edu.cn
From: "Applied Catalysis B: Environment and Energy" noreply_EMsupport@elsevier.com
Subject: Your Submission

Ms. Ref. No.: **APCATB-D-25-05672R1**
Title: Enhanced Lewis Acid-Base Synergistic Photocatalyst for Selective Benzimidazole Synthesis Coupled with Hydrogen Evolution
Applied Catalysis B: Environment and Energy

Dear Dr. Shen,

I am pleased to inform you that your manuscript, entitled "Enhanced Lewis Acid-Base Synergistic Photocatalyst for Selective Benzimidazole Synthesis Coupled with Hydrogen Evolution", is now accepted for publication in Applied Catalysis B. For your information, I append the reviewers' comments below.

Your accepted manuscript will now be transferred to our production department and work will begin on creation of the proof. If we need any additional information to create the proof, we will let you know. If not, you will be contacted again in the next few days with a request to approve the proof and to complete a number of online forms that are required for publication.

Congratulations! Thank you for submitting your work to this journal and I am looking forward to receiving further manuscripts from your group.

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Yours sincerely,

Junwang Tang
Editor
Applied Catalysis B: Environment and Energy

Comments from the Editors and Reviewers:

%ATTACH_FOR_REVIEWER_DEEP_LINK INSTRUCTIONS%

%EDITOR_DECISION_PHRASES%

Reviewer #1:

Reviewer #2: ok

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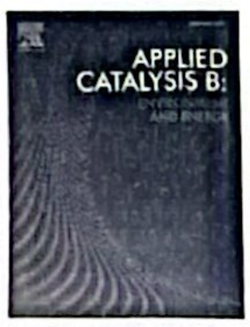

Enhanced Lewis acid-base synergistic photocatalyst for selective benzimidazole synthesis coupled with hydrogen evolution

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ABSTRACT

Developing photocatalytic systems capable of simultaneously synthesizing value-added heterocycles and producing green hydrogen is of great importance for solar-driven chemical transformations and sustainable manufacturing. In this work, we report a highly efficient bifunctional photocatalyst composed of Au_4Pd_5 alloy nanoparticles anchored on nitrogen-doped TiO_2 (N-TiO_{2-x}), wherein the reinforced Pd Lewis acidic (LA) sites and reinforced O Lewis basic (LB) sites synergistically activate o-phenylenediamine (OPD) and ethanol, respectively. This cooperative interaction facilitates a single-intermediate pathway (ethanol - ethoxy radical - acetaldehyde), leading to the selective formation of 2-methylbenzimidazole (2MBZ) and concurrent H_2 evolution. Under optimal conditions, the system achieves a 2MBZ yield of 3.80 $mmol \cdot g^{-1} \cdot h^{-1}$, H_2 generation rate of 14.17 $mmol \cdot g^{-1} \cdot h^{-1}$, and a selectivity of 94.43 %, outperforming all previously reported photocatalytic systems to date. Mechanistic insights derived from *in-situ* characterizations, and density functional theory (DFT) calculations reveal that electron redistribution at the Au-Pd interface and oxygen vacancy (O_v) induced charge modulation in N-TiO_{2-x} are responsible for the enhanced charge separation, reactant adsorption, and activation. The system maintains excellent performance under natural sunlight, with H_2 and 2MBZ production rates of 11.96 and 3.11 $mmol \cdot g^{-1} \cdot h^{-1}$, respectively, which will be more economical than the conventional thermal route and demonstrating strong scalability.

1. Introduction

Benzimidazoles are nitrogen-containing heterocyclic compounds formed by fusing benzene and imidazole rings. Their excellent biological activities and structural diversity make them significant in pharmaceuticals, agrochemicals, and materials science [1,2]. Benzimidazole scaffolds are widely present in clinically approved drugs, with their broad pharmacological activity driving applications in antitubercular, antiviral, and antitumor therapies [3–5]. However, conventional synthetic routes to benzimidazoles often require harsh conditions, such as high temperatures, strong acids, or toxic oxidants, which limit their environmental and practical applicability [6]. At the same time, hydrogen (H_2) has emerged as a clean energy carrier and an essential industrial feedstock with wide applications in food processing, petroleum refining, ammonia synthesis, and metal purification [7,8]. Under the guidance of sustainable chemistry principles, developing catalytic strategies that enable the simultaneous production of high-value organic compounds and green H_2 has become a promising research frontier. Recently, visible-light-driven semiconductor photocatalysis has been recognized as a green and efficient approach for such transformations under mild reaction conditions [9].

Most recently, the visible-light-driven cross-coupling of o-phenylenediamines (OPD) with alcohols catalyzed by photocatalysts has attracted growing attention as a sustainable approach for benzimidazole synthesis accompanied by H_2 evolution [10,11]. Despite notable progress, this system still faces two critical challenges: only a fraction of hydrogen atoms from alcohol substrates is utilized for H_2 production, resulting in low atom economy and limited energy conversion efficiency; the key intermediates (such as acetaldehyde) are prone to side reactions or rapid desorption upon formation, diminishing their availability for subsequent condensation steps and thereby reducing the selectivity of the target benzimidazole product. The previously reported

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