



Faculty Details proforma for College Web-site



Title	Dr.	First Name	Bhawna	Last Name	Kaushik	Photograph
Designation	Assistant Professor					
Address	Department of Chemistry, Acharya Narendra Dev College, University of Delhi, Govindpuri, Kalkaji, New Delhi-110019					
Phone No Office	011-26294541					
Email/ Web-Page	bhawnakaushik@andc.du.ac.in					
Educational Qualifications						
Degree	Institution				Year	
Ph.D. Chemistry	Department of Chemistry, University of Delhi				2022	
M.Sc Chemistry	Department of Chemistry, University of Delhi				2016	
B.Sc (H) Chemistry	Miranda House, University of Delhi				2013	
Career Profile						
Teaching in Acharya Narendra Dev College since January 2022 till present.						
Administrative Assignments						
(i) Member of Placement Cell Committee, Acharya Narendra Dev College						
(ii) Member of Magazine Editorial Committee, Acharya Narendra Dev College						
(iii) Member of Dhvani (Theater Society), Acharya Narendra Dev College						
(iv) Member of NAAC Team, Acharya Narendra Dev College						
(v) Member of Organizing Committee for 60 Hours value added course in Research Methodology conducted by Prime Minister's Research Fellow Scholars from IIT Delhi at Acharya Narendra Dev College.						
Areas of Interest / Specialization						
Research Area : Photocatalysis						
Specialization : Inorganic Chemistry						
Subjects Taught						
<ul style="list-style-type: none">• B.Sc(H) VI Sem. – Bioinorganic and Organometallic Chemistry (Theory and Practical)• B.Sc (Prog.) Life Sciences – II Sem. – Chemical Bonding and Role of Metals in Biological Systems• GE – I Sem. – Atomic Structure and Chemical Bonding• GE – II Sem. – States of Matter and Chemical Kinetics• GE - II Sem.- Energy and Environment						

- B.Sc (Prog.) Life Science – III Sem – Functional Group Organic Chemistry – I

Research Guidance

NA

Publications Profile

S.NO.	Title of the Paper	Impact Factor
1	<i>“Precisely Engineered Type II ZnO-CuS based Heterostructure: A Visible Light Driven Photocatalyst for Efficient Mineralization of Organic Dyes.”</i> Bhawna Kaushik , Sneha Yadav, Pooja Rana, Pooja Rana, Kanika Solanki, Deepti Rawat and Rakesh Kumar Sharma; <i>Appl. Surf. Sci.</i> , 2022 , 590, 153053.	7.39
2	<i>“Ingeniously Designed Silica Nanostructures as an Exceptional Support: Opportunities, Potential Challenges and Future Prospects for Viable Degradation of Pesticides”</i> Rakesh Kumar Sharma, Bhawna Kaushik , Sneha Yadav, Pooja Rana, Pooja Rana, Kanika Solanki, Deepti Rawat; <i>J. Environ. Manage.</i> , 2022 , 301, 113821.	8.91
3	<i>“Magnetically Separable Type-II Semiconductor based ZnO/MoO₃ Photocatalyst: A Proficient System for Heteroarenes Arylation and Rhodamine B Degradation under Visible Light.”</i> Bhawna Kaushik , Pooja Rana, Deepti Rawat, Kanika Solanki, Sneha Yadav, Pooja Rana, and R. K. Sharma. <i>New J. Chem.</i> , 2022 , 46, 8478-8488.	3.92
4	<i>“Earth-Abundant Cobalt based photocatalyst: Visible light induced direct (Het)Arene C-H arylation and CO₂ capture”</i> Pooja Rana, Bhawna Kaushik , Rashmi Gaur, Sriparna Dutta, Sneha Yadav, Pooja Rana, Kanika Solanki, Bhavya Arora, Ankush Biradar, Manoj B. Gawande and Rakesh Kumar Sharma; <i>Dalton Trans.</i> , 2022 , 51, 2452-2463.	4.56
5	<i>“Tailoring the catalytic activity of cobalt decorated magnetic boron nitride nanosheets in the one-pot synthesis of 3,4-dihydropyrimidin-2(1H)-ones”</i> Pooja Rana, Ranjana Dixit, Shivani Sharma, Sriparna Dutta, Sneha Yadav, Bhavya Arora, Bhawna Kaushik , Pooja Rana and Rakesh K. Sharma. <i>ACS Appl. Nanomater.</i> , 2022 , 5, 4875-4886.	6.04
6	<i>“A magnetic CoFe₂O₄/Cu-ABDC MOF heterostructured composite unleashing enhanced catalytic performance towards the sustainable synthesis of 2H-indazole motifs”</i> Sneha Yadav, Ranjana Dixit, Shivani	3.92

	Sharma, Sriparna Dutta, Bhavya Arora, Pooja Rana, <i>Bhawna Kaushik</i> , Pooja Rana, Kanika Solanki and Rakesh K. Sharma. <i>New J. Chem.</i> , 2022 , <i>46</i> , 10829-10843.	
7	“Surface engineered iridium-based magnetic photocatalyst paving a path towards visible light driven C-H arylation and cyanation reaction.” Pooja Rana, Rashmi Gaur, <i>Bhawna Kaushik</i> , Pooja Rana, Sneha Yadav, Priya Yadav, Priti Sharma, Manoj B. Gawande and Rakesh K. Sharma; <i>J. Catal.</i> , 2021 , <i>401</i> , 297-308.	8.04
8	“Enhanced Catalysis through Structurally Modified Hybrid 2-D Boron Nitride Nanosheets Comprising of Complexed 2-hydroxy-4-methoxybenzophenone Motif.” Pooja Rana, Ranjana Dixit, Shivani Sharma, Sriparna Dutta, Sneha Yadav, Aditi Sharma, <i>Bhawna Kaushik</i> , Pooja Rana, Alok Adholeya and Rakesh K. Sharma. <i>Sci. Rep.</i> , 2021 , <i>11</i> , 24429.	4.99
9	“Unlocking the catalytic potency of a magnetic responsive CoFe ₂ O ₄ /Ni-BTC MOF composite for the sustainable synthesis of tri- and tetra-substituted imidazoles” Sneha Yadav, Ranjana Dixit, Shivani Sharma, Sriparna Dutta, Bhavya Arora, Pooja Rana, <i>Bhawna Kaushik</i> , Pooja Rana, Alok Adholeya, Manoj B. Gawande and Rakesh Kumar Sharma; <i>Mater. Chem. Front.</i> , 2021 , <i>5</i> , 7343-7355.	8.68
10	“In-situ synthesis of 3-D Hierarchical ZnFe ₂ O ₄ modified Cu ₂ S snowflakes: Exploring their bifunctionality in Selective Photocatalytic Reduction of Nitroarenes and Methyl Orange Degradation” <i>Bhawna Kaushik</i> , Pooja Rana, Deepti Rawat, Kanika Solanki, Sneha Yadav, Pooja Rana, and R. K. Sharma. <i>J. Photochem. Photobiol. A: Chem.</i> , 2022 , <i>433</i> , 114165.	5.14
11	“Synergetic Effect of Magnetic ZnO/BiVO ₄ for Enhanced Visible Light Mediated Degradation of Bisphenol A and Methyl Violet” <i>Bhawna Kaushik</i> , Pooja Rana, Deepti Rawat, Kanika Solanki, Sneha Yadav, Pooja Rana, and R. K. Sharma. <i>Appl. Organomet. Chem.</i> , 2023 , <i>37</i> (1), e6936.	4.07
12	“Preparation and characterization of the h-BN/Fe ₃ O ₄ /APTES-AMF/Cu II nanocomposite as a new and efficient catalyst for the one-pot three-component synthesis of 2-amino-4-aryl (or heteroaryl)-7, 7-dimethyl-5-oxo-5, 6, 7, 8-tetrahydro-4 H-chromene-3-carbonitriles” Pooja Rana, Ranjana Dixit, Shivani Sharma, Sriparna Dutta, Sneha Yadav, Bhavya Arora, “ <i>Bhawna Kaushik</i> , Manoj B. Gawande and Rakesh K. Sharma. <i>Nanoscale</i> , 2023 , <i>15</i> (7), 3482-95.	8.30

13	<i>“Insights into the catalytic potential of rationally designed magnetic boron nitride nanosheets supported nickel catalyst for the efficient synthesis of 1,4-dihydropyridines”</i> Pooja Rana, Ranjana Dixit, Shivani Sharma, Sriparna Dutta, Sneha Yadav, Bhavya Arora, Priyanka, <i>Bhawna Kaushik</i> and Rakesh K. Sharma. <i>React. Chem. Eng.</i> , 2023 , 8(1), 244-253.	5.20
14	<i>“Hierarchical 3D Flower-like Metal Oxides Micro/Nanostructures: Fabrication, Surface Modification, Their Crucial Role in Environmental Decontamination, Mechanistic Insights, and Future Perspectives”</i> Kanika Solanki, Shivani Sharma, Sneha Yadav, <i>Bhawna Kaushik</i> , Pooja Rana, Ranjana Dixit, and R. K. Sharma . <i>Small</i> , 2023 , 2300394.	15.15
15	<i>“Development of heterogeneous photocatalysts via the covalent grafting of metal complexes on various solid supports.”</i> Pooja Rana, <i>Bhawna Kaushik</i> , Kanika Solanki, Kapil Mohan Saini, and R. K. Sharma. <i>Chem. Commun.</i> , 2022 , 58(81), 11354-11377.	6.06

S.NO.	Book Chapters
1	Nanostructured inorganic–organic silica as green material for sustainable development of catalysts. Deepti Rawat, <i>Bhawna Kaushik</i> and Rahul Singhal. <i>Applications of Advanced Green Materials</i> , 2021, Woodhead Publishing (ScienceDirect), 151-167.

Conference / Presentations /Workshops

International

- Member of Organizing Committee** in International e-conference on “Mitigating contemporary environmental issues by sustainable approaches” held in February 2022, Delhi.
- Participated** in ACS Workshop on “Greening our education system: Initiatives for propagating and Preaching Beyond Benign Concepts in Classrooms and Laboratories” in December 2019 at Conference Centre, University of Delhi, Delhi.
- Participated** at “6th World Congress on Nanomedical Sciences” held in January 2019 at Vigyan Bhawan, Delhi.

National

1. **Member of Organizing Committee** in Going Global workshop on “Green chemistry education today for a sustainable tomorrow” on 27th- 29th May, 2022.
2. **Member of Organizing Committee** in RSC Symposium Series on “Frontiers in Green Chemistry & Sustainable Development: Special Emphasis on Food, Water & Energy” on 25th November 2021.
3. **Participated** in ACS Symposium Series on “Green Chemistry & Sustainable Development” on 24-25th November 2021.
4. **Participated** in Chemistry Festival for Students & Teachers: Green Chemistry, Education, Research & Practice on 1st October 2021.
5. **Participated** in National Seminar on Women Empowerment through Science and Technology: Power to Transform the World on 25th September 2021.
6. **Member of Organizing Committee** in National Conference on “Water Crisis, Public Health and Sustainable Solutions and First Annual Conference of Save The Environment” in October 2019 at INSA, Delhi.
7. **Best Poster Award** in the National Conference on “Recent Trends in Chemical Sciences” and RSC Workshop on “Periodic Table: Boon for Mankind” held in August-September 2019 at Hotel Maidens, New Delhi.
8. **Participated** in National Conference on “Green Chemistry for Clean Environment” held in July 2019 at J. R. N. Rajasthan Vidyapeeth, Udaipur.
9. **Member of Organizing Committee** in National Conference on “Chemistry for Human Health and Environment” held in December 2018 at University of Delhi.

Research Projects (Major Grants/Research Collaboration)

NA

Awards and Distinctions

NA

Association With Professional Bodies

Life Member of National Environmental Science Academy (NESA)

Other Activities

Completed 60 Hour Add-On certificate course entitled 'GREEN CHEMISTRY AND ENVIRONMENT' organized by Department of Chemistry, Miranda House.



दिल्ली विश्वविद्यालय



University of Delhi

विद्या-वाचस्पति * Doctor of Philosophy (Ph.D.)

प्रमाणित किया जाता है कि दिसम्बर, 2022 में

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This is to certify that after approval of the thesis on

Design and Fabrication of Visible Light Responsive Zinc Based Magnetic Photocatalysts for

Effective Heteroarylation Nitrobenzene Reduction and Degradation of Organic Pollutants

in December, 2022

Bhawna Kaushik

qualified for the degree of Doctor of Philosophy (Ph.D.) of this University.

The said degree was conferred upon him/her at the convocation held in 2023.

Department: Chemistry

Yash Gupta

कुलसचिव

दिल्ली विश्वविद्यालय

Registrar

University of Delhi



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Delhi, Dated the 25th February, 2023

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D/O SURENDER KAUSHIK
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GURGAON HARYANA



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The fellowship is tenable w.e.f. **01.01.2017**. It will be governed by the terms and conditions of the **UGC Junior Research Fellowship**. In order to accept this offer you may contact UGC at the address given overleaf. Your dossier is being forwarded to UGC.

While considering for fellowship /appointment, the Supervisor/Guide/Fellowship awarding Authority/ Lectureship appointing Authority **must verify the original record/certificates of the candidate** regarding eligibility criterion & caste /PH/VH certificate (if applicable) for this test. The details regarding the eligibility criterion for this test is given overleaf.

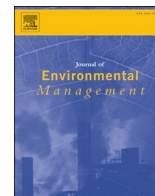
This letter may be treated as a certificate.

Kindly acknowledge the receipt.

Yours sincerely,

(Signature)
10/04/17
Deputy Secretary (Exam)

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उप सचिव (परीक्षा) / Deputy Secretary (Exam)
वैज्ञानिक तथा औद्योगिक अनुसंधान परिषद्
Council of Scientific & Industrial Research
सी. एस. आई. आर. कॉम्प्लेक्स, पूसा, नई दिल्ली-110012
CSIR Complex, Pusa, New Delhi-110012



Ingeniously designed Silica nanostructures as an exceptional support: Opportunities, potential challenges and future prospects for viable degradation of pesticides

Rakesh Kumar Sharma^{a,*}, Bhawna Kaushik^a, Sneha Yadav^a, Pooja Rana^a, Pooja Rana^a, Kanika Solanki^a, Deepti Rawat^b

^a Green Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi, 110007, India

^b Department of Chemistry, Miranda House College, University of Delhi, New Delhi, 110007, India

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ABSTRACT

Despite significant advancements in modern agricultural practices, efficient handling of pesticides is a must as they are continuously defiling our terrestrial as well as aquatic life. During the last couple of decades, substantial efforts by various research groups have been devoted to find innovative solutions to remove pesticides from our environment in a greener way. In this regard, functionalized silica nanoparticles (NPs) have gained considerable attention of scientific community due to their notable properties such as amenable design, large surface area as well as fine-tunable and uniform pore structures which make them an ideal material for pesticides removal. The present review aims to proffer current scientific progress attained by silica-based nanostructures as an excellent material for effective removal of noxious agrochemicals. Further, a brief discussion on the synthetic strategies as well as intrinsic benefits associated with different morphologies of silica have also been highlighted in this article. It also summarizes the recent reports on silica assisted degradation of pesticides *via* enzymatic, chemical as well as advanced oxidation protocols. Additionally, it presents a critical analysis of different support materials for decontamination of our ecosystem. The review concludes with potential challenges, their possible solutions along with key knowledge gaps and future research directions for successful deployment of silica supported materials in degradation of pesticides at commercial scale.

1. Introduction

For many decades, population spurt has been a topic of deliberation and preoccupation calling for an immediate action to intensify the global agricultural production to ensure food security. According to the “World Population Prospects 2019”, a United Nations (UN) report, world population is further expected to reach 9.7 billion by 2050 (DESA, 2019). Thus, the ever-expanding number of inhabitants stresses on the requirement to boost the crop production without increasing farmland footprint. To enhance crop productivity and make agriculture profitable in face of rising valuation, use of pesticides, herbicides and fertilizers have proliferated exponentially. Pesticides refer to a group of chemical compounds used to kill undesirable organisms in farmlands and have been playing a pivotal role in agronomics since the dawn of agricultural revolution (Rani et al., 2020). According to their usage, pesticides can be classified broadly in four categories namely, herbicides, insecticides,

fungicides and rodenticides. Synthetic organic pesticides generally contain carbamates, organophosphates, coumarins, organochlorines, organonitrogens, pyrethroids along with arsenic and mercury derivatives (Rani et al., 2017).

Nevertheless, with the rapid development and proliferation of chemical industries, their indiscriminate and sporadic use has led to 200,000 human deaths annually along with the contamination of natural resources (Mew et al., 2017). Most of the chemical pesticides are retained within the body of organism because of their lipophilicity and exhibit detrimental effects on human health due to their carcinogenic nature and ability to damage central nervous system (Sabarwal et al., 2018). Starting from flora to fauna, pesticides owing to their longer half-lives not only extensively accumulate but also remain persistent in water and soil, thereby posing serious threat to the environment (Fernández-Pérez et al., 2011; Liu et al., 2016a). Another major limitation associated with the use of conventional pesticides is that their

* Corresponding author.

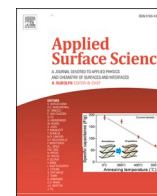
E-mail address: rksharmagreenchem@hotmail.com (R.K. Sharma).

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Full Length Article

Precisely engineered type II ZnO-CuS based heterostructure: A visible light driven photocatalyst for efficient mineralization of organic dyes

Bhawna Kaushik^a, Sneha Yadav^a, Pooja Rana^a, Pooja Rana^a, Kanika Solanki^a, Deepti Rawat^b, R.K. Sharma^{a,*}

^a Green Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi 110007, India

^b Department of Chemistry, Miranda House College, University of Delhi, New Delhi 110007, India



ARTICLE INFO

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Visible light
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Photocatalytic
Heterogeneous catalyst
Environmental remediation
Core-shell architectures

ABSTRACT

Herein, type-II band alignment of magnetic ZnO/CuS has been achieved by assembling p-type CuS nanoparticles on n-type ZnO heterostructures to accomplish the photocatalytic degradation of two colored cationic dyes, namely, methylene blue (MB) and toluidine blue (TB). The material exhibited excellent photocatalytic efficiency towards MB and TB with 93% and 87.5% degradation in just 16 and 18 min respectively. The efficacy of doped photocatalyst (FSZCS) was found to be 6 times higher than the undoped material ($\text{Fe}_3\text{O}_4@/\text{SiO}_2@/\text{ZnO}$; FSZ) whereas pristine CuS degraded only 50% of the dye sample under identical conditions. Therefore, the dramatic enhancement of the photocatalytic degradation performance could be attributed to the synergetic effect created by doping CuS over magnetic ZnO nanocomposites which extended the photoresponse of ZnO by driving the entire degradation process under visible light irradiation and also reducing the charge recombination rate. The plausible mechanistic pathway and identification of degradation products was discussed in detail on the basis of scavenger studies as well as GC-MS analysis. Furthermore, the designed catalyst could be recycled and reused up to 5 runs without any significant decrease in its photocatalytic activity. The reported procedure exhibited multiple advances as it proceeded by utilizing renewable household LEDs as power source at room temperature without the use of any additional oxidant under neutral pH conditions thus, paving a strong path towards sustainable, green and responsible chemistry.

1. Introduction

Photocatalytic degradation has surfaced as a burgeoning technique contributing immensely towards environmental remediation *via* the use of renewable, clean and abundant solar energy [1]. As a green and sustainable approach, it has found numerous applications in the arena of treatment of perilous and toxic effluents such as dyes, pesticides, pharmaceutical by-products, industrial waste etc. A wide range of photo-responsive materials like metal oxides, metal nitrides and oxy-nitrides, alkali metal base, metal sulfides and carbon-based components have been employed for carrying out the degradation of harmful organic pollutants into environmentally benign products [2–6]. Amongst them, metal oxide and sulfide based semiconductors have occupied a central place in the field of environmental reformation owing to their wide band gap due to deep 2p oxygen orbital in their valence band [7,8]. The process is set off by the generation of electrons when light with wavelength lesser than or equal to the band gap of the semiconductor is

illuminated in turn separating the charge carriers (electrons and holes). Semiconductors with wide band gap such as TiO_2 and ZnO have invariably proved their worth as dynamic photocatalysts [9–11]. ZnO specifically with a band gap of 3.37 eV has the potential of detoxifying water owing to its prominent reaction and mineralization rates, lower toxicity, economic nature, higher number of active sites and the fact that it can be moulded to obtain various morphologies [12]. Unfortunately, it works best in the UV region, which constitutes less than 5% of the solar energy thus conspicuously confining its photocatalytic utility. Another factor limiting its photocatalytic efficiency is the rapid charge recombination. Therefore, it is indispensable to shift the optical absorption band of ZnO towards the visible range and simultaneously circumvent the issue of charge recombination [13]. One of the simplest yet appealing pathway to overcome this roadblock is by doping such single-component semiconductor with another narrow band gap semiconductor which can synergistically bring the band gap in visible region and ensure prolonged separation of charge carriers [14]. Copious

* Corresponding author.

E-mail addresses: pooja.rana.kmc@gmail.com (P. Rana), ranapooja185@gmail.com (P. Rana), rksharmagreenchem@hotmail.com (R.K. Sharma).

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Magnetically separable type-II semiconductor based ZnO/MoO₃ photocatalyst: a proficient system for heteroarenes arylation and rhodamine B degradation under visible light†

 Bhawna Kaushik,^a Pooja Rana,^a Deepti Rawat,^b Kanika Solanki,^a Sneha Yadav,^a Pooja Rana^a and R. K. Sharma^{id}*^a

Herein, a new strategy is used to systematically tailor the band arrangement in a ZnO-based heterostructure to realize its bifunctionality towards the synthesis of heteroarylated products and degradation of Rhodamine B under visible light. A Type-II band alignment transition is carried out via introducing MoO₃ rods which subsequently enhance its light-harvesting properties, prolonged charge separation and abundant active sites. The designed heterostructure displayed broad functional group compatibility for both electron-donating and electron-withdrawing group substituted substrates with good product yield. Comparably, their visible light driven heteroarylation process is significantly enhanced to more than 4 times that of pristine ZnO and is superior to most photocatalysts reported previously. ZnO/MoO₃ also displayed remarkable degradation ability of 96.8% towards RhB into innocuous products under neutral conditions without utilizing any oxidant. More impressively, the developed catalyst demonstrated long-term stability and excellent reusability up to five runs without any significant change in efficiency. Hence, this work not only for the first time exhibits the untapped potential of using a highly active, economically efficient and magnetically separable ZnO/MoO₃ core-shell heterostructure to substitute for expensive metals in photocatalysis but also presents a new opening for synthesising hybrid materials with excellent performance in environmental remediation.

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Introduction

In the era of rapid industrialisation and growing pollution concerns, utilization of a renewable and green strategy for organic transformations is indispensable. Visible light photocatalysis constitutes a right step in paving a way towards a sustainable economy. Traditional organic synthetic protocols are accompanied by certain drawbacks, such as harsh reaction conditions (high temperature and pressure, use of toxic solvents) as well as energy-intensive processes.^{1,2} Interestingly, photocatalytic systems have the potential to synthesise various industrially important compounds with high efficiency along with good atom economy, lower energy consumption and innocuous solvents using an environmentally benign, non-conventional source of energy.^{3–12} Additionally, a photo-assisted system has the

tendency to alter the thermodynamic equilibrium of the reaction and facilitate the transformation at room temperature, thus averting the generation of undesirable products due to elevated temperatures. Therefore, the fusion of renewable light energy and transition metal assisted organic transformations represents an effective building block in developing useful synthetic protocols.¹³

The formation of a C–C bond represents an important class of core structural constituents of biomedical significance in the agrochemical and pharmaceutical industries, peptide mimetics, dyes *etc.*¹⁴ In particular, heteroarylation of arenes exemplifies an important class of reaction as it opens up avenues to various heterobiaryl motifs with applications in technological fields since they possess fascinating biological and optical properties.^{15–18} Until now, scientists have utilized various renowned reactions such as Suzuki, Stille, and Kumada coupling for synthesising the important class of biaryl compounds. However, these protocols come with many drawbacks, such as the employment of expensive and tediously separable homogeneous catalysts along with noxious organic halides such as organic bromides and less reactive organic chlorides.^{19–22} In order to make the process commercially beneficial, activation of these reagents *via* attaching various

^a Green Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi-110007, India. E-mail: rksharmagreenchem@hotmail.com;

Tel: +91-011-276666250

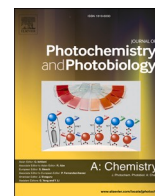
^b Department of Chemistry, Miranda House College, University of Delhi, New Delhi-110007, India

† Electronic supplementary information (ESI) available. See DOI: <https://doi.org/10.1039/d2nj00906d>



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In-situ synthesis of 3-D hierarchical ZnFe₂O₄ modified Cu₂S snowflakes: Exploring their bifunctionality in selective photocatalytic reduction of nitroarenes and methyl orange degradation

Bhawna Kaushik^a, Pooja Rana^a, Kanika Solanki^a, Deepti Rawat^b, Sneha Yadav^a, Pooja Rana^a, Dhanaji R. Naikwadi^c, Ankush V. Biradar^c, R.K. Sharma^{a,*}

^a Green Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi 110007, India

^b Department of Chemistry, Miranda House College, University of Delhi, New Delhi 110007, India

^c Inorganic Materials and Catalysis Division, CSIR-Central Salt and Marine Chemicals Research Institute, Bhavnagar 364002, Gujarat, India

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ABSTRACT

Designing hierarchical photocatalysts holds a great promise in the future due to their potential to solve energy and environmental problems. Herein, we report the design of intriguing Cu₂S snowflakes doped with ZnFe₂O₄ nanospheres through the *in-situ* self-assembly method. This effective heterojunction is constructed by controlling the amount of Cu₂S dopant during the formation of the photocatalyst which leads to efficient electron-hole separation and fast photo-response. Besides, the multidimensional structure of the catalyst enhances its light harvesting properties, provides high surface area and abundant active sites that accelerate the reaction kinetics. The obtained ZnFe₂O₄/Cu₂S composite not only exhibited an excellent photocatalytic reduction of nitrobenzene with 98 % yield (~35 % more than pristine Cu₂S and ZnFe₂O₄) but also degraded methyl orange and methylene blue dyes proficiently (94.3 % and 86 %) upon visible light illumination. Further, GC-MS studies were performed to identify the intermediates formed and deduce the mechanistic pathway for both processes. Another key feature that made the fabricated photocatalyst economical and sustainable is its easier magnetic retrievability from the reaction mixture along with a good reusability for several runs without any significant loss in its activity.

1. Introduction

Design and development of functionalized heterostructured materials with intriguing morphologies has garnered the immense interest of the scientific community owing to their inherent physical and chemical properties such as large surface area, highly accessible active sites, modifiable surface and good chemical stability. [1,2]. These materials are not only highly desirable for their light-harvesting properties but also for effective charge transportation. Therefore, they find numerous applications in fundamental scientific areas of clean energy production, catalysis, photocatalysis, adsorption, gas sensing and biomedicine [3–8]. Semiconductor based heterogeneous photocatalysis has been in the limelight to accomplish aforementioned applications due to its economic efficiency, environmentally benign and sustainable attributes that also supports the “zero” waste scheme of the wastewater industry. The technology stands apart due to the direct utilization of solar energy

as the driving force to accomplish the desired transformations [9–12]. An ideal semiconductor photocatalyst should possess high activity, enhanced charge carrier separation, smaller bandgap for maximum utilization of visible light, relatively lower toxicity, economic viability, appreciable operational stability and reusability [13–16]. Unfortunately, these requisites are barely met by many contemporary photocatalysts such as TiO₂ and ZnO which hinder their practical utility [17,18]. To overcome these difficulties, metal ferrites have been an interesting choice as they possess smaller band gaps due to presence of the Fe-d orbitals above the valence band of metal [19]. Further, their physicochemical properties can be fine-tuned to suit the desired applications as these characteristics are highly dependent on their crystalline nature and size [20,21]. In this context, transition metal-based zinc ferrite (ZF) spinel has emerged as a potential photoactive semiconductor material since this smart nanocomposite possesses magnificent photocatalytic properties owing to its narrow band gap that lies in the visible

* Corresponding author.

E-mail address: rksharmagreenchem@hotmail.com (R.K. Sharma).

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RESEARCH ARTICLE

Synergic effect of Type II ZnO/BiVO₄ magnetic heterostructures for visible light-driven degradation of bisphenol A and methyl violet

Bhawna Kaushik¹ | Pooja Rana¹ | Deepti Rawat² | Kanika Solanki¹ |
Pooja Rana¹ | Shallu Sachdeva³ | Dhanaji R. Naikwadi⁴ |
Ankush V. Biradar⁴ | Rakesh K. Sharma¹ 

¹Green Chemistry Network Centre, Department of Chemistry, University of Delhi, 110007, New Delhi, India

²Department of Chemistry, Miranda House College, University of Delhi, 110007, New Delhi, India

³Department of Chemistry, Acharya Narendra Dev College, University of Delhi, 110019, New Delhi, India

⁴Inorganic Materials and Catalysis Division, CSIR-Central Salt and Marine Chemicals Research Institute, Bhavnagar, Gujarat, 364002, India

Correspondence

Rakesh K. Sharma, Green Chemistry Network Centre, Department of Chemistry, University of Delhi, 110007, New Delhi, India.
Email: rksharmagreenchem@hotmail.com

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Construction of an effective heterojunction for unimpeded flow of photogenerated charges and their prolonged separation is imperative for environmental photocatalysis. Herein, we have designed an efficient magnetic ZnO/BiVO₄ type-II heterostructure, which was employed for proficient degradation of persistent methyl violet dye with an efficiency of 97.6% in 90 min and a hazardous organic pollutant, namely, bisphenol A. UV-DRS and photoluminescence studies demonstrated that the fabricated nanocomposite exhibited effective light absorption and prolonged charge separation, thereby resulting in high photocatalytic efficacy under visible light irradiation. The efficacy of developed magnetic ZnO/BiVO₄ was also compared with pristine BiVO₄ and undoped magnetic ZnO, which indicated that the constructed heterostructure displayed approximately threefold and sixfold activity in contrast with bare BiVO₄ and undoped magnetic ZnO nanoparticles, respectively. Radical trapping studies, ESR analysis along with GC-MS analysis were conducted to elucidate the mechanistic pathway during the photodegradation process. This work provides a rational technical approach and research ideas for photocatalytic degradation of harmful organic pollutants in an environment-friendly manner by employing energy-efficient LEDs. Besides, good recyclability of catalyst makes it a promising candidate for large-scale applications.

KEYWORDS

environment, magnetic catalyst, nanostructures, pollutants, visible light mediated

1 | INTRODUCTION

Ever-increasing environmental pollution and rapid depletion of conventional energy resources are the two major challenges that plague development as well as the survival of human beings. Therefore, it is of utmost importance to employ renewable energy sources that are economical, green and sustainable.^[1] Amongst different

non-conventional resources, abundant and sustainable solar energy is a clean choice, and solar light-driven photocatalysis has drawn substantial attention worldwide in environmental remediation in the past few decades. In this regard, the development of semiconductor photocatalysis has ushered a new era in the field of global decontamination as it utilises pollution-free sunlight and cheaper transition metals.^[2] A typical photocatalyst

Magnetic Boron Nitride Nanosheets Decorated with Cobalt Nanoparticles as Catalyst for the Synthesis of 3,4-Dihydropyrimidin-2(1H)-ones/thiones

Pooja Rana, Ranjana Dixit, Shivani Sharma, Sriparna Dutta, Sneha Yadav, Bhavya Arora, Bhawna Kaushik, Pooja Rana, and Rakesh K. Sharma*



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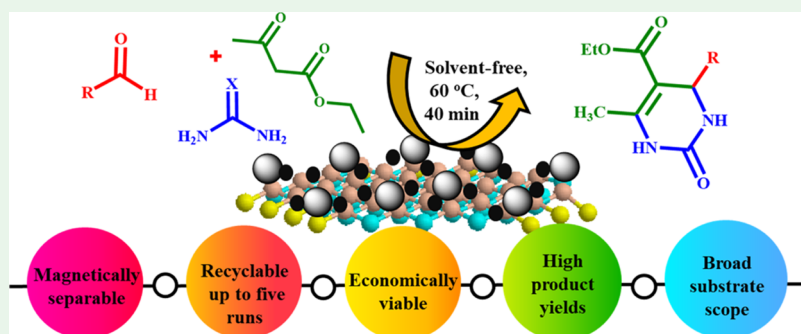
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Supporting Information



ABSTRACT: Atomically thin two-dimensional boron nitride nanosheets have spawned futuristic advancements in the arena of nanocatalysis research through their intriguing capability to act as exceptional support matrixes. Motivated by their phenomenal attributes, we have fabricated a magnetic boron nitride nanosheet-based cobalt catalytic system wherein boron nitride nanosheets are initially integrated with magnetic Fe_3O_4 nanoparticles (NPs), and the resulting nanostructure is further surface-engineered with cobalt NPs to yield an *h*-BN/ Fe_3O_4 /Co hybrid. For gaining an insight into their structural and morphological features, reliable spectroscopic and microscopic characterization techniques including TEM, SEM, XRD, FT-IR, VSM, ED-XRF, XPS, BET, TGA, and AAS were employed. The developed nanohybrid material was then utilized to provide ready access to a library of highly bioactive 3,4-dihydropyrimidin-2(1H)-ones/thiones under ambient conditions. A plausible mechanistic route for furnishing 3,4-dihydropyrimidin-2(1H)-ones catalyzed by *h*-BN/ Fe_3O_4 /Co has also been delineated. Ambient reaction conditions, solvent-free conditions, high product yield, and excellent thermal and mechanical stability of the catalyst along with facile magnetic retrievability and efficient recyclability are some of the phenomenal characteristics of this methodology. The present protocol besides exhibiting a wider functional group tolerance and a high turnover number was devoid of any additive, thus making it superior to literature precedents reported to date. In consideration of the striking catalytic activity of the *h*-BN/ Fe_3O_4 /Co nanomaterial, it can be anticipated that the present catalyst can not only possess a stupendous potential to expedite substantial manufacturing of other industrially demanding organic motifs but may also unlock insights for designing next-generation 2D catalytic materials.

KEYWORDS: *nanocomposites, heterocyclic motifs, heterogeneous catalytic system, catalytic efficacy, recoverability, recyclability*

1. INTRODUCTION

Nanostructured materials have emerged at the forefront of catalysis science research as they play a significant role in the furtherance of the key goals of green chemistry.¹ In recent years, astonishing advancements have been witnessed in the rational design and fabrication of well-defined, highly active, and selective nanocatalysts.² The prospects of tuning the catalytic properties of nanomaterials by controlling the structural aspects of nanoparticles and manipulating the interaction between catalytically active species and its support matrix have aided in the engineering of new types of hybrid multifunctional materials that can be deployed for diverse industrially significant organic transformations.^{3–7} Indeed, the

prominence of nanocatalysts having precisely controlled surface morphology can be realized from the Green Chemistry Presidential Challenge Awards that have been conferred for innovations pertaining to the development of promising nanocatalysts. Among a plethora of support materials utilized

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An Earth-abundant cobalt based photocatalyst: visible light induced direct (het)arene C–H arylation and CO₂ capture†

Pooja Rana,^a Bhawna Kaushik,^a Rashmi Gaur,^a Sriparna Dutta,^a Sneha Yadav,^a
Pooja Rana,^a Kanika Solanki,^a Bhavya Arora,^a Ankush V. Biradar,^b
Manoj B. Gawande^c and R. K. Sharma^{a*}

In this work, we have reported a noble metal free heterogeneous photocatalyst to carry out direct (het) arene C–H arylation and solvent-free CO₂ capture *via* single-electron transfer processes at room temperature and under pressure. The catalytic system comprises a cobalt(III) complex grafted over the silica coated magnetic support for the efficient recovery of the photocatalytic moiety without hampering its light-harvesting capability. The novel Earth-abundant cobalt(III) based photocatalyst possesses various fascinating properties such as high surface area to volume ratios, large pore volume, crystalline behaviour, high metal loading, excellent stability and reusability. The general efficacy of the highly abundant and low-cost cobalt based heterogeneous nanocatalyst was checked for the selective conversion of aryl diazonium salts into synthetically and pharmaceutically significant biaryl motifs under ambient conditions upon irradiation with visible light. The highly efficient photocatalytic conversion of carbon dioxide (CO₂) to a value-added chemical was accomplished under mild reaction conditions with high selectivity, showing the added benefit of operational simplicity.

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Introduction

Visible light mediated photocatalysis, owing to its notable capacity to harvest light energy and convert it into chemical energy, has acquired tremendous significance in the research community showing promising applications in diverse fields such as water splitting, solar energy storage, photovoltaics, pollutant degradation and organic transformations.^{1–4} Generally, an efficient photosensitizer is required for harvesting free, clean, abundant, non-hazardous and environment friendly visible light energy that can meet the current and future energy demands.^{5,6} To date, many highly active and stable Ru^{II}, Re^I, Os^{II} and Ir^{III} based d⁶ complexes have been widely recognized and extensively exploited in photoredox reactions due to their prominent properties such as (i) strong absor-

bance in the visible light region, (ii) long-lived electronically excited states, (iii) ease of attaining highly oxidised and reduced species either in the ground or excited state in contrast to organic moieties, and (iv) highly reversible electrochemical nature and photostability of the photoactivated, oxidized and reduced species.^{7,8} Along with the above mentioned captivating properties, photoredox catalysis has clear benefits for sustainability as it adheres to the key principles of Green Chemistry including atom economy, design for energy efficiency, catalysis and use of renewable feedstock.⁹ In addition, the fascinating properties of photons aid in delivering sufficient energy to achieve the desired reactivity without using high temperatures or harsh conditions, contrary to thermally induced reactions.⁹ Despite the appealing properties of photocatalysts enabling bond formation processes, unfortunately the low abundance, toxicity and exorbitant cost associated with them, limit their practical utility.¹⁰ Recently, scientists have shown keen interest in eliminating the use of precious and toxic metals in photoactive coordination complexes by employing more Earth abundant elements for the development of sustainable photocatalysts.¹¹ Thus, various synthetic and material chemists are reorienting their efforts toward the employment of base metal photocatalysts as clean, energy efficient and versatile materials to process visible light driven organic transformations and CO₂ capture.¹² Nevertheless,

^aGreen Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi-110007, India. Tel: +011-276666250;

E-mail: rksharmagreenchem@hotmail.com

^bInorganic Materials and Catalysis Division, CSIR-Central Salt and Marine Chemicals Research Institute, Bhavnagar 364002, Gujarat, India

^cInstitute of Chemical Technology Mumbai-Marathwada Campus, Jalna, 431213 Maharashtra, India

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Surface engineered Iridium-based magnetic photocatalyst paving a path towards visible light driven C-H arylation and cyanation reaction



Pooja Rana^a, Rashmi Gaur^a, Bhawna Kaushik^a, Pooja Rana^a, Sneha Yadav^a, Priya Yadav^a, Priti Sharma^c, Manoj B. Gawande^{b,*}, Rakesh K. Sharma^{a,*}

^aGreen Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi 110007, India

^bDepartment of Chemistry and Engineering, Institute of Chemical Technology Mumbai-Marathwada Campus, Jalna 431213, Maharashtra, India

^cRegional Centre of Advanced Technologies and Materials, Czech Advanced Technology and Research Institute, Palacký University, 779 00 Olomouc, Czech Republic

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ABSTRACT

The report presents the fabrication and application of a highly versatile, magnetic and robust iridium based photoredox nanocatalyst. Herein, Ir(ppy)₃ based photocatalyst sites have been chemically engineered over the magnetic nanoparticles to encompass the captivating features of homogeneous iridium photocatalyst with the magnetically recyclable core. A household photoreactor was designed and fabricated to achieve highly selective visible light driven oxidative C-H arylation and C-H cyanation under sustainable and ambient reaction conditions utilizing the Ir@PyBz@ASMNPs photoredox nanocatalyst. The environment friendly Ir@PyBz@ASMNPs shows excellent photocatalytic activity, broad substrate adaptability and outstanding recyclability compared to the analogous homogeneous catalysts. Indeed, the Ir@PyBz@ASMNPs possess some key features including high surface area, high iridium metal loading and excellent stability. This work is expected to enlighten and provide new insights in the rational design of high performance and recoverable photoredox nanocatalyst through surface engineering strategy.

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1. Introduction

Photoredox catalysis has gained colossal attention for engineering environmentally sustainable processes due to its ability to exploit inexpensive, clean and endless renewable solar energy to engage single electron transfer (SET) for synthetic manipulation or catalytic activation of organic molecules [1]. Over the past few years, numerous research groups have widely employed the power of light for various applications such as photocatalysis, photodegradation, singlet oxygen generation, water splitting, reduction of carbon dioxide, etc. [2–5]. In fact, several transition metals based homogeneous catalysts (such as ruthenium, iridium and copper polypyridyl complexes) and organic moieties (like eosin Y, rose bengal etc.) have garnered particular appreciation in the field of photocatalysis [3,6–9]. These photocatalysts possess various attention grabbing properties such as energy efficiency, stability, absorbance in visible range, long-lived photoexcited states, versatile nature and readily tunable photophysical properties by modification of the ligand scaffold [6]. Unlike the thermal

and UV light catalyzed reactions, the visible light catalyzed reactions reduce the possibility of side reactions due to lack of visible light absorbance by organic compounds [9,10].

Over the years, the chemistry of tris-cyclometalated coordinatively saturated, 18-electron d⁶ complexes of iridium, such as Ir(ppy)₃, has been the focus of great attention due to the ease of synthesis and diversification, stability at room temperature, and excellent photoredox properties [11,12]. Additionally, the iridium complexes unveil a wide variety of applications in the field of solar cells, light-emitting diodes (LEDs), sensing, biology and as initiators in free radical polymerization [12]. Despite these above-mentioned advantages, progress must be made toward ease of separation, recyclability, stability, and environmental neutrality of these photoredox catalysts in order to move the field toward practical utility in academia and industry [13,14]. Consequently, the development of a heterogeneously immobilized photosensitizer system that maintains photoredox capability upon immobilization is a significant step toward this goal. Henceforth, coordination chemistry delivers a new pathway for engineering heterogeneous supports through immobilizing photocatalytically active sites having high activity and selectivity. To date, a limited number of covalently or physically immobilized photoredox catalyst over cores (such as graphene, TiO₂, silica, polymer, ZnO, NPs etc.) have been

* Corresponding authors.


E-mail addresses: mb.gawande@marj.ictmumbai.edu.in (M.B. Gawande), rksharmagreenchem@hotmail.com (R.K. Sharma).

RESEARCH ARTICLE

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Unlocking the catalytic potency of a magnetic responsive CoFe₂O₄/Ni-BTC MOF composite for the sustainable synthesis of tri- and tetra-substituted imidazoles†

 Sneha Yadav,^a Ranjana Dixit,^a Shivani Sharma,^a Sriparna Dutta,^a Bhavya Arora,^a Pooja Rana,^a Bhawna Kaushik,^a Pooja Rana,^a Alok Adholeya,^b Manoj B. Gawande^c and Rakesh K. Sharma *^a

With the advancements in materials engineering, unprecedented newer materials based on magnetic MOFs have emerged as one of the promising candidates in the strategic field of catalysis. Within this perspective, the present report unveils the fabrication of an intricately designed magnetic CoFe₂O₄/Ni-BTC based MOF composite via a one-pot solvothermal approach. The synthesized composite was comprehensively characterized using various analytical tools such as FTIR, XRD, XPS, VSM, TGA, EDS, ED-XRF and AAS. Advanced microscopic tools like SEM and TEM provided deep insights into the structural aspects of the synthesized material. The catalytic performance of the hybrid composite was further explored in one-pot multicomponent reactions to obtain a combinatorial library of biologically and pharmacologically significant 2,4,5-trisubstituted (up to 98% yield) and 1,2,4,5-tetrasubstituted (up to 97% yield) imidazoles in good to excellent yields under solvent free conditions. Wider functional group tolerance, excellent turnover numbers, high atom economy, ambient reaction conditions, magnetic retrievability and good recyclability are some of the striking attributes of this protocol. We envisage that the present methodology employing magnetic MOFs as catalytic reactors is highly desirable for designing cleaner and greener processes in modern organic chemistry.

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Introduction

Since the groundbreaking research by Yaghi, Robson and Kitagawa on functional polymers, metal organic frameworks (MOFs) have found a niche in modern materials science.^{1–5} The significant progress in MOFs can be attributed to their structural versatility driven by coordination chemistry that allows designing hierarchically advanced multifunctional architectures with novel topologies and superior properties.^{6–8} The additional enthusiasm for these materials in academia and industries arises because of their exceptionally good thermal and chemical stabilities, structural robustness, tunable pore size, incredibly high surface area and record breaking porosity.^{9–11} Besides, the extraordinary periodic network of metal

nodes and inorganic functional linkers in MOFs allows the incorporation of diverse functionalities within the framework for targeted applications.^{12,13} These outstanding and remarkable features of MOFs play a crucial role in diverse fields including gas storage and separation, heterogeneous catalysis, proton conduction, sensing, luminescence, drug delivery and water remediation.^{14–21} Moreover, the phenomenal and prodigious skeleton of MOFs renders spatially separated unsaturated metal centers and functional linkers which further obstruct catalyst deactivation and deterioration, thereby enhancing the overall catalytic efficiency.^{19–23} Furthermore, to expedite the separation process, the idea of magnetic retrievability has been successfully utilized by decorating the surface of MOFs with inverse spinel magnetic nanoparticles.^{22–31} Stimulated by these advancements, tremendous efforts of the research community are oriented towards finding neoteric magnetic MOF catalyzed routes for several unexplored transformations that generate countless heterocyclic motifs of fundamental interest. Amongst the numerous heterocycles, imidazoles are privileged structural moieties present in several biologically active molecules and natural products. Additional soaring interest stems from the bioactive nature presented by these moieties that renders them appealing in pharmaceutical and drug development fields, ligand chemistry and materials

^a Green Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi 110007, India. E-mail: rksharmagreenchem@hotmail.com;

Fax: +91-011-27666250; Tel: 011-276666250

^b TERI-Deakin Nanobiotechnology Centre, TERI Gram, The Energy and Resources Institute, Gurugram-122102, India

^c Department of Industrial and Engineering Chemistry, Institute of Chemical Technology Mumbai-Marathwada Campus, Jalna, 431213, Maharashtra, India. E-mail: mb.gawande@marj.ictmumbai.edu.in

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Unravelling the catalytic potential of a magnetic CoFe₂O₄/Cu–ABDC MOF composite in the sustainable synthesis of 2*H*-indazole motifs†

Sneha Yadav, Ranjana Dixit, Shivani Sharma, Sriparna Dutta, Bhavya Arora, Pooja Rana, Bhawna Kaushik, Kanika Solanki and Rakesh K. Sharma *

Incessant advancements made in catalytic processes during the past few decades prompted researchers to design prodigious sustainable materials that lie within the domain of green synthesis. From this perspective, metal organic frameworks (MOFs) with abundant chemical functionalities and ultra-high chemical as well as structural tunability have emerged as the most prolific heterogeneous catalysts in synthetic organic chemistry. Herein, we report the fabrication of a magnetic CoFe₂O₄/Cu–ABDC (ABDC = 2-aminoterephthalate) hybrid composite *via* a one-pot solvothermal strategy whose catalytic efficiency has been investigated in a three-component coupling reaction to obtain biologically active and pharmacologically significant 2*H*-indazole scaffolds (up to 98% yield). By integrating magnetic inverse spinel cobalt ferrite nanoparticles with the functional properties of the MOF, two most important parameters, *i.e.* environmental compatibility and recyclability, have been well established on a single platform. Aided by the advanced microscopic, spectroscopic and property characterization tools, the morphological as well as structural information of the developed hybrid composite has been deduced well. The aim of this study is to design a sustainably viable process that would provide highly demanding pharmacophores, *i.e.* 2*H*-indazoles, in surprisingly high yields from comparatively cheap benchmark substrates – “substituted anilines, 2-bromobenzaldehydes and sodium azide”. Furthermore, our protocol has successfully accomplished the remarkable task of replacing toxic volatile organic solvents with eco-friendly solvents such as water. A broad substrate scope, high atom economy, ambient and greener reaction conditions devoid of any ligands, additives or activators, a lower reaction time and temperature, a high turnover frequency and magnetic retrievability are some of the salient features of this methodology which render it highly promising in industry and academia. Besides, the present study is the first report employing a magnetic MOF as a heterogeneous catalyst for the preparation of 2*H*-indazole moieties.

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Introduction

The advent of metal organic frameworks (MOFs) as porous coordination polymers comprising a systematic array of metal ions linked with organic linkers has completely metamorphosed the landscape of materials chemistry across the globe through their notable attributes. Quite appreciably, the chemistry as well as synthesis of these transformative materials (MOFs) is not only expanding at an impressive pace but has rather garnered worldwide attention. Indeed, striking progress made in this field during the last two decades is apparent from

the exponential rise in the number of publications appearing each year in the literature.^{1,2} Their intriguing or unique properties such as high chemical and thermal stability, an extremely high surface area, numerous topologies, an unprecedented degree of chemical tunability and record breaking porosity have provided a great impetus to researchers.^{3–5} Owing to their magnificent features, these multimodal architectures are finding tantalizing applications in the fields of catalysis, medicinal chemistry, optics, sensing, magnetism, conductivity, gas storage and separation.^{6–9} Besides, the virtually infinite choice of spacers or high degree of structural tunability in MOFs provides viable opportunities to tune their activity as per requirement.^{10–12} Therefore, by judiciously selecting an appropriate metal ion and an organic linker possessing chemical functionalities, it is possible to rationally design MOF architectures for specific applications.^{13,14} In fact, certain MOF materials possess coordinatively unsaturated or exposed metal sites which play a



Green Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi – 110007, India. E-mail: rksharmagreenchem@hotmail.com;
Fax: +91-011-27666250; Tel: +91-011-27666250

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Insights into the catalytic potential of a rationally designed magnetic boron nitride nanosheet supported nickel catalyst for the efficient synthesis of 1,4-dihydropyridines†

Pooja Rana,^a Ranjana Dixit,^a Shivani Sharma,^a Sriparna Dutta,^a Sneha Yadav,^a Bhavya Arora,^a Priyanka,^a Bhawna Kaushik,^a Manoj B. Gawande ^b and Rakesh K. Sharma ^{*a}

Considering the unprecedented attributes of two-dimensional nanomaterials, the present report discloses the fabrication of a novel magnetic boron nitride nanosheet based nickel nanocatalyst (*h*-BN@Fe₃O₄@APTES@TPC@Ni) through a covalent bonding approach. In order to certify the successful synthesis of the catalyst, various advanced microscopic and spectroscopic techniques, including TEM, SEM, FT-IR, XRD, EDS, ED-XRF and VSM, were employed. The developed *h*-BN@Fe₃O₄@APTES@TPC@Ni catalytic agent acts as a promising candidate to deliver various pharmaceutically relevant 1,4-dihydropyridine scaffolds using aryl aldehydes, dimedone, ethyl acetoacetate and ammonium acetate as reacting species. The devised methodology is accompanied with noteworthy benefits, including mild reaction conditions, wide substrate scope, excellent products yield and short reaction time. Besides this, facile magnetic retrievability and remarkable reusability of the catalyst for six successive runs without any appreciable loss in catalytic efficacy are the additional factors that make this protocol worthwhile.

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Introduction

With the rapid advancements in modern materials science, substantial efforts by researchers have been devoted towards the rational design of highly tunable nanomaterials and their applicability in the realm of catalysis. In consideration of the dynamic aspects of heterogeneous catalytic systems, two-dimensional nanomaterials, especially hexagonal boron nitride (*h*-BN) layered nanostructures, have garnered extensive interest as solid supports owing to their exceptional attributes, such as high surface area to volume ratio; excellent thermal, chemical and mechanical strength; nanometer size; high oxidative resistance; coordinatively unsaturated active sites; and capability of dissipating substantial heat in exothermic reactions.^{1–11} Further, the catalytic characteristics of boron nitride nanosheets can be enhanced *via* immobilizing numerous active sites with organic–inorganic

functionalities on them through a covalent approach. In addition, integration of *h*-BN nanosheets with magnetite nanoparticles further enhances their separation from the reaction media using a readily available external magnet, which represents a better separating technique than its energy consuming counterparts.^{12–17} Taking into account the salient attributes offered by atomically thin 2D *h*-BN, we herein designed a magnetically responsive *h*-BN nanostructured based nickel nanocatalyst and examined its catalytic performance in the one-pot multicomponent synthesis of highly privileged heterocyclic compounds, namely 1,4 dihydropyrimidines. These Hantzsch derived synthons exhibit various pharmacological activities, such as anti-inflammatory, antimicrobial, antihypertensive, anti-tumor, anti-diabetic, and cyclooxygenase-2 inhibitor properties.^{18–21} For instance, nifedipine, amlodipine and nicardipine exhibit calcium channel blocking properties that are used to treat hypertension by relaxing cardiac muscle contraction.²² In relevance to aforementioned activities, some of the biologically active molecules bearing 1,4-dihydropyridine moieties are displayed in Fig. 1. To date, several synthetic approaches for the synthesis of polyhydroquinolines have been reported but due to considerable shortcomings manifested by these protocols, such as prolonged reaction times, acidic or basic conditions, tedious experimental procedures, use of expensive metal salts,

^a Green Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi 110007, India. E-mail: rksharmagreenchem@hotmail.com; Fax: +91 011 27666250; Tel: +91 011 27666250

^b Department of Industrial and Engineering Chemistry, Institute of Chemical Technology Mumbai-Marathwada Campus, Jalna, 431213, Maharashtra, India. E-mail: mb.gawande@marj.ictmumbai.edu.in

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OPEN

Enhanced catalysis through structurally modified hybrid 2-D boron nitride nanosheets comprising of complexed 2-hydroxy-4-methoxybenzophenone motif

Pooja Rana¹, Ranjana Dixit¹, Shivani Sharma¹, Sriparna Dutta¹, Sneha Yadav¹, Aditi Sharma¹, Bhawna Kaushik¹, Pooja Rana¹, Alok Adholeya²✉ & Rakesh K. Sharma¹✉

Tuning the structural architecture of the pristine two dimensional hexagonal boron nitride (*h*-BN) nanosheets through rational surface engineering have proven advantageous in the fabrication of competent catalytic materials. Inspired by the performance of *h*-BN based nanomaterials in expediting key organic transformations, we channelized our research efforts towards engineering the inherent surface properties of the exclusively stacked *h*-BN nanosheets through the incorporation of a novel competent copper complex of a bidentate chelating ligand 2-hydroxy-4-methoxybenzophenone (BP). Delightfully, this hybrid nanomaterial worked exceptionally well in boosting the [3 + 2] cycloaddition reaction of azide and nitriles, providing a facile access to a diverse variety of highly bioactive tetrazole motifs. A deep insight into the morphology of the covalently crafted *h*-BN signified the structural integrity of the exfoliated *h*-BN@OH nanosheets that exhibited lamellar like structures possessing smooth edges and flat surface. This interesting morphology could also be envisioned to augment the catalysis by allowing the desired surface area for the reactants and thus tailoring their activity. The work paves the way towards rational design of *h*-BN based nanomaterials and adjusting their catalytic potential by the use of suitable complexes for promoting sustainable catalysis, especially in view of the fact that till date only a very few *h*-BN nanosheets based catalysts have been devised.

Two-dimensional hexagonal boron nitride nanosheets based architectures with long-range ordered atomic arrangements have recently stimulated the exponential growth in the arena of materials chemistry. Indeed, it is the exclusive stacked structure of BN nanosheets due to electronegativity difference between B and N atom which imparts several fascinating features such as excellent mechanical strength, outstanding thermal and chemical stability, low dielectric constant, oxidative resistance, nanometre size, large surface area to volume ratio and high complex loading¹⁻³. Considering such intrinsic characteristics, research on structurally flexible *h*-BN based nanomaterials has been flourishing across the globe in myriad of diverse fields including sensing, electronics, sensors, hydrogen storage, gas separation, etc⁴⁻⁸. Very recently, these exotic materials have significantly garnered the attention of scientific community as a promising candidate to design new generation catalytic materials for cascade reactions due to their unique atomic structure. Notably, atomically thin *h*-BN nanosheets have received tremendous recognition as a solid matrix amongst various nanostructured materials to develop surface engineered catalysts as they are capable of dissipating considerable amount of heat in exothermic organic reactions^{9,10}. Besides, they not only prevent catalyst deactivation by driving off the moisture owing to hydrophobic surface but also prevent the issue of silicates or aluminates formation often encountered

¹Green Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi 110007, India. ²TERI-Deakin Nanobiotechnology Centre, TERI Gram, The Energy and Resources Institute, Gurugram 122102, India. ✉email: aloka@teri.res.in; rksharmagreenchem@hotmail.com

ARTICLE

Development of Heterogeneous photocatalysts by the covalent grafting of metal complexes onto various solid supports

Pooja Rana,^a Bhawna Kaushik,^a Kanika Solanki,^a Kapil Mohan Saini^b and R. K. Sharma*^aReceived 00th January 20xx,
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Till date, remarkable progress has been made in the development of photocatalysts owing to their high activity, selectivity and tunable light absorption in visible light range. Recently, the heterogeneous photocatalytic systems have emerged as potential candidates due to their beneficial attributes (like high surface area, ease of functionalization and facile separation etc). This article provides succinct overview of rational designing of heterogeneous photocatalysts by grafting photoactive complexes over heterogeneous support matrices via covalent grafting, their detailed characterization techniques which have been followed by landmark examples of their applications. Also, major challenges and opportunities in the forthcoming progress of these appealing areas are emphasised.

Introduction to photocatalyst

Sunlight is a cleaner, greener, plentiful, inexhaustible, and widely distributed sustainable energy source that has played an instrumental role since the evolution of life on Earth. Inspired from mother nature, artificial photosynthesis has been garnering immense attention of scientific fraternity as a promising candidate to tackle the current energy and environmental crisis by utilizing vivid and renewable energy of sunlight.¹⁻⁴ This untapped potential of sunlight has been very well exploited in photocatalytic processes wherein solar energy is used to drive thermodynamically uphill reactions to generate desired products. It is worth mentioning here that visible-light-driven photocatalysis proved its dominance and significance in various fields of chemistry such as organic transformations, dye degradation, H₂ generation *via* water splitting, CO₂ reduction etc. owing to its high proficiency, non-polluting nature and mild conditions for substrate activation to solve various synthetic problems.^{5, 6} The term “photocatalysis” is derived from Greek word, where “photo” stands for light and “catalysis” means a process to enhance the rate of reaction by the addition of a substance without getting consumed itself during the course of reaction. Thus, photocatalysis entails the concurrent arrangement of photochemistry and catalysis to accelerate a chemical reaction.

When a photocatalyst (PC) is irradiated with light of suitable wavelength, it gets photoexcited and transfers an electron from the ground state, or the highest occupied molecular orbital (HOMO) of a molecule to its excited electronic state, or the lowest unoccupied molecular orbital (LUMO). The photoexcited catalyst (PC*) accepts or donates a single electron through oxidative or reductive quenching

cycles depending on the substrates and reagents present in the reaction mixture.⁷ Here, the photoexcited species acts as the reactive catalytic species and help in achieving cleavage of challenging bonds, activation of organic species or generation of radicals to promote a chemical transformation which could not be realized by the classical organic chemistry techniques. Thus, “Photocatalysis” has emerged as an environment friendly synthetic route wherein without taking recourse to acids, bases, or water sensitive reagents such as Grignard reagents, the activation of innocuous starting reagent could be facilely achieved that drastically reduces or limits the production of hazardous by products during chemical synthesis. Lately, a major breakthrough has been achieved in this field with the discovery of energy efficient and economical irradiation systems for the visible light mediated organic transformations that garnered the significant attention of scientists in photoinduced reactions to activate substrates, reagents, or catalytic intermediates under mild conditions by employing cost effective, clean and low-consumption irradiation setups.

Early breakthroughs in the field of photoredox catalysis

Utilization of photocatalysts (PC) has gained widespread recognition as a powerful tool for achieving the activation of organic motifs by using low-energy photons and paved a path towards sustainable chemical synthesis by promoting the use of non-hazardous and environment-friendly reagents. Although, employment of solar energy to initiate organic transformations is not a novel concept. The idea was originally proposed by Ciamician as early as 1912.⁸ Nevertheless, the potential of photocatalysts was identified by Fujishima and Honda in 1972 on water splitting by opting titanium dioxide electrode and irradiating it with UV light and a weak external electric field of 0.5 V,⁹ which made photocatalysis to be one of the oldest explored techniques for transforming solar energy into chemical energy. In 1976, Carey and co-workers investigated the


^a Green Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi-110007, India; Tel: 011-27666250, E-mail: rksharma@greenchem@hotmail.com

^b Kalindi College, University of Delhi, New Delhi, Delhi 110008, India.

PAPER

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Preparation and characterization of the *h*-BN/Fe₃O₄/APTES-AMF/Cu^{II} nanocomposite as a new and efficient catalyst for the one-pot three-component synthesis of 2-amino-4-aryl(or heteroaryl)-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitriles†

Pooja Rana,^a Ranjana Dixit,^b Shivani Sharma,^b Sriparna Dutta,^a Sneha Yadav,^a Bhavya Arora,^a Bhawna Kaushik,^a Manoj B. Gawande^c and Rakesh K. Sharma *^a

The intriguing features of surface-engineered hexagonal two-dimensional boron nitride (*h*-BN) nanostructures have captivated the immense interest of researchers working in the arena of materials science. Inspired by striking attributes exhibited by *h*-BN nanosheets as the support material, we devoted our efforts towards synthesizing a novel magnetically retrievable *h*-BN/Fe₃O₄/APTES-AMF/Cu^{II} catalytic system, which was then comprehensively characterized using various techniques including SEM, TEM, EDX, SEM-based elemental mapping, ED-XRF, AAS, XRD, FT-IR, VSM, XPS, TGA, and BET. Further, the catalytic potential of *h*-BN/Fe₃O₄/APTES-AMF/Cu^{II} nanocomposites was investigated in the one-pot multicomponent coupling reaction to gain access to a library of biologically active 2-amino-4-aryl(or heteroaryl)-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitriles under ambient conditions. In addition, the use of green solvent, facile magnetic recoverability, and reusability of up to six successive runs made this protocol environmentally benign and economical. This work throws light on the development of covalently functionalized 2D-BN nanostructure-based copper catalysts and establishes its significance in furnishing industrially demanding products that would pave the way towards sustainable chemistry.

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Introduction

Over the decades, nanomaterials have emerged as fundamental channels for diverse applications, attributed to their striking physical and chemical properties arising at nanodimensions. However, sometimes these nanoparticles themselves fail to deliver satisfactory results as they are associated with certain shortcomings including toxicity, hydrophobicity, and tendency to undergo agglomeration. Homogeneous catalytic systems in particular give rise to separation issues, which in turn limit their reusability to a maximum extent. Hence, many

research groups are heading towards the design and development of highly tunable hybrid nanomaterials that combine both characteristics of homogeneous as well as heterogeneous catalysts, which can be further utilized in the synthesis of organic compounds, especially heterocyclic compounds due to their countless applications in various sciences.¹ In view of the importance of hybrid nanomaterials, a plethora of solid-supported catalysts have been fabricated to date amongst which two-dimensional nanomaterials analogous to graphene-like nanostructures, especially boron nitride nanosheets, have proven their efficacy as a solid-support owing to their exceptional structural features, such as a large surface area to volume ratio, excellent mechanical, thermal, and chemical strength, resistance towards oxidation, high thermal conductivity, nanometer size, and electronegativity characteristics offer a number of active sites for introducing organic functionalities or metal complexes.^{2–12} In order to promote easy separation of the catalyst, the incorporation of magnetite nanoparticles has emerged as a greener alternative route, especially in comparison to the energy demanding techniques.^{13–25} Such advancements have fascinated a wide domain of researchers

^aGreen Chemistry Network Centre, Department of Chemistry, University of Delhi, New Delhi-110007, India. E-mail: rksharmagreenchem@hotmail.com;
Fax: +91-011-27666250; Tel: +011-27666250

^bDepartment of Chemistry, Ramjas College, University of Delhi, New Delhi-110007, India

^cDepartment of Industrial and Engineering Chemistry, Institute of Chemical Technology Mumbai-Marathwada Campus, Jalna, 431213 Maharashtra, India.
E-mail: mb.gawande@marj.ictmumbai.edu.in

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Hierarchical 3D Flower-like Metal Oxides Micro/Nanostructures: Fabrication, Surface Modification, Their Crucial Role in Environmental Decontamination, Mechanistic Insights, and Future Perspectives

Kanika Solanki, Shivani Sharma, Sneha Yadav, Bhawna Kaushik, Pooja Rana, Ranjana Dixit, and R. K. Sharma*

Hierarchical micro/nanostructures are constructed by micro-scaled objects with nanoarchitectures belonging to an interesting class of crystalline materials that has significant applications in diverse fields. Featured with a large surface-to-volume ratio, facile mass transportation, high stability against aggregation, structurally enhanced adsorption, and catalytical performances, three dimensional (3D) hierarchical metal oxides have been considered as versatile functional materials for waste-water treatment. Due to the ineffectiveness of traditional water purification protocols for reclamation of water, lately, the use of hierarchical metal oxides has emerged as an appealing platform for the remediation of water pollution owing to their fascinating and tailorable physiochemical properties. The present review highlights various approaches to the tunable synthesis of hierarchical structures along with their surface modification strategies to enhance their efficiencies for the removal of different noxious substances. Besides, their applications for the eradication of organic and inorganic contaminants have been discussed comprehensively with their plausible mechanistic pathways. Finally, overlooked aspects in this field as well as the major roadblocks to the implementation of these metal oxide architectures for large-scale treatment of wastewater are provided here. Moreover, the potential ways to tackle these issues are also presented which may be useful for the transformation of current water treatment technologies.

of our planet. Around 71% of the surface of this big blue marble is covered by water but only 1% of it is accessible to human beings which can be extracted and utilized for numerous purposes.^[1-3] Being central to all the facets of life and society, water is crucial for socio-economic development, health, education, and environmental protection. In today's era of rapid evolution, supply of clean and drinking water has transpired as a critical challenge for mankind due to climate change, population growth, and industrialization.^[4,5] Existence of more than 60 million organic and inorganic materials was reported by the Registry of the American Chemical Society across the world in its latest database of chemicals and surprisingly around 12 000 new chemical substances enter the global market every day. Most importantly, over and above 49 million chemicals are available in retail sector, amongst them only less than 1% are monitored or regulated, rest end up in our water bodies without any indication/idea of their potential toxicity.^[6] Indeed, influx of large amounts


1. Introduction

Water is prime requirement for the existence of life on earth and it is also referred to as one of the defining characteristics

of these dreadful pollutants into our ecosystem has drastically increased during the past century.^[7,8] Disposal of life-threatening wastes into our aquatic systems by industries as well as other anthropogenic activities has made this scenario graver. Uncontrolled release of waste streams into water bodies not only leads to the migration of contaminants to the sub-surface of soil but also to groundwater which exhibits severe consequences on human health, negative impact on economic activities, and inimical repercussions on mother nature. Higher concentrations of these noxious substances pose a serious threat to the environment due to their bioaccumulation and non-biodegradable properties.^[9,10] Their detrimental effects on Homo sapiens include cancer, typhoid, cholera, diarrhea, neurological, mental, reproductive, and endocrinal disorders. Further, their adverse ramifications on aquatic as well as terrestrial flora and fauna incorporate abnormal behavior, growth inhibition, physiological disorders, glandular damages, hormonal imbalances, and death.^[11] Therefore, to circumvent jeopardizing our natural

K. Solanki, S. Yadav, B. Kaushik, P. Rana, R. K. Sharma
Green Chemistry Network Center
Department of Chemistry
University of Delhi
New Delhi 110007, India
E-mail: rksharmagreenchem@hotmail.com

S. Sharma, R. Dixit
Department of Chemistry
Ramjas College
University of Delhi
New Delhi 110007, India

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Nanostructured inorganic–organic silica as green material for sustainable development of catalysts

6

Deepti Rawat¹, Bhawna Kaushik² and Rahul Singhal³

¹Department of Chemistry, Miranda House, University of Delhi, Delhi, India, ²Department of Chemistry, University of Delhi, Delhi, India, ³Department of Chemistry, Shivaji College, University of Delhi, Delhi, India

6.1 Introduction

Due to the rising environmental cognizance across the globe, there has been an ever-increasing demand for clean and sustainable technologies. With “green chemistry” coming into the limelight, this dream to move towards a cleaner and greener world has been transformed into reality. Green chemistry works toward efficient utilization of raw materials and eradicating waste at the very beginning rather than employing end of the pipe solutions. It also primarily focusses on circumventing or minimizing the utilization of hazardous chemicals as solvents and reagents to carry out organic transformations [1]. Green chemistry opens up the doors to sustainability by providing more credible solutions to the existing perilous chemical methodologies employed in the industries [2]. It provides cleaner avenues for designing new products. In fact, it has earned its position as a dazzling star in the chemical industries by making a wide array of manufacturing processes economically viable. Various operating costs are reduced tremendously by employment of greener and sustainable methods. Treatment and disposal also become obsolete with minimal or zero waste generation. Moreover, steering clear of using stoichiometric reagents and other toxic solvents leads to massive cut in material and energy costs of the manufacturing protocols [3]. Essence of green chemistry can be consolidated by a cohesive set of 12 principles based on intelligently transforming the existing chemical processes into environmentally benign ones [4]. Catalysis is one such overarching principle encompassing the soul of green chemistry as it incorporates many underlying factors—milder reaction conditions, lesser by-products generation, and significantly lesser energy input.

In the past few decades, green chemistry and catalysis have become an unstoppable juggernaut. Catalysis lies at the heart of chemistry due to its outstanding ability of altering the rate of chemical reactions. A catalyst has the magical ability to provide an alternate route to a reaction without getting spent itself. It can also change



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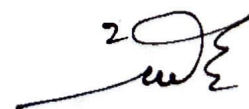
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